PERFORMANCE EVALUATION OF TWO REDUCING AND ALKALINITY PRODUCING SYSTEMS FOR COAL MINE DRAINAGE REMEDIATION AFTER 4 YEARS OF OPERATION

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ABSTRACT

The Bowden Close passive mine water treatment system (PTS), in the north east of England, was commissioned in 2003 to treat net-acidic coal mine drainage. The monitoring data for iron, aluminium and acidity in its influents and effluent for the first four years of the treatment are presented here. The PTS consists of two Reducing and Alkalinity Producing Systems (RAPS) operating in parallel, followed by a single polishing aerobic wetland. The main purpose of the treatment system is to raise pH and alkalinity of the mine water and to lower iron and aluminium concentrations. The two influent waters derive from an abandoned underground coal mine (influent 1, discharged into RAPS 1) and from a waste rock pile located on the former colliery site (influent 2, discharged into RAPS 2). The influents are moderate to strongly acidic, and iron- and aluminium-rich (pH_{mean} = 4.9-5.6, $alkalinity_{mean}$ = 10-24 mg L^{-1} $CaCO_3$ eq, acidity = 12-863 mg L^{-1} $CaCO_3$ eq, Fe_{mean} = 16-68 mg L^{-1}, and Al_{mean} = 5-32 mg L^{-1}. Average flow rates are 36 L min^{-1} (influent 1) and 99 L min^{-1} (influent 2). The pH of the final effluent is raised to 7.0 (median) and always exceeds 5.7. With few exceptions, the effluent is net alkaline (i.e. alkalinity>acidity) and iron and aluminium concentrations are lowered to 3.4 mg L^{-1} and 1.5 mg L^{-1} (median), respectively. The treatment system appears to be load limited and has a mean area-adjusted acidity removal rate of 9.4 g d^{-1} m^{-2} (range: 2.3-96 g d^{-1} m^{-2}, as $CaCO_3$ eq). The monitoring data collected indicate the system serves the operational purpose for which it was designed. However, further work is ongoing: (1) to quantify the comparative importance of bacterial sulphate reduction and calcite dissolution in generating alkalinity, and (2) to establish what changes in the biogeochemical and physical characteristics of the system are likely to limit the overall lifetime of the treatment system.

Key Words: reducing and alkalinity producing systems, RAPS, SAPS, Bowden Close, passive treatment, mine water, coal mine drainage, net acidic mine water, iron, aluminium, acidity

1. INTDROCUTION

Passive Treatment Systems for Net-Acidic Coal Mine Drainage

Several options exist for the passive treatment of net-acidic coal mine drainage where it emerges at the surface. One option is Reducing and Alkalinity-Producing Systems (RAPS) (Watzlaf *et al.* 2002), also referred to as Successive Alkalinity Producing Systems (SAPS) (Kepler and Mc Cleary 1994) or vertical flow ponds (Riefler *et al.* 2008)). A RAPS is essentially a combination of anoxic limestone drains and compost wetlands (Kepler and Mc Cleary 1994). The key difference from the alternative compost wetlands, which are horizontal flow systems, is that RAPS units operate as vertical flow units. The distinct advantage of the arrangement of RAPS is that they occupy significantly less land than equivalent compost wetland systems, albeit greater hydraulic head is required if such units are to operate without energy inputs. The principal treatment processes are discussed elsewhere, and are therefore not repeated here (e.g. Younger *et al.* 2002, Kepler and Mc Cleary 1994).

RAPS can be combined with other anaerobic (Demchak *et al.* 2001, Norton *et al.* 1998)) or aerobic treatment systems (Bhattacharya *et al.* 2007, Jage *et al.* 2001, Nairn and Mercer 2000). In some instances they have been combined with active treatment units in *hybrid* systems (Hilton *et al.* 2003).
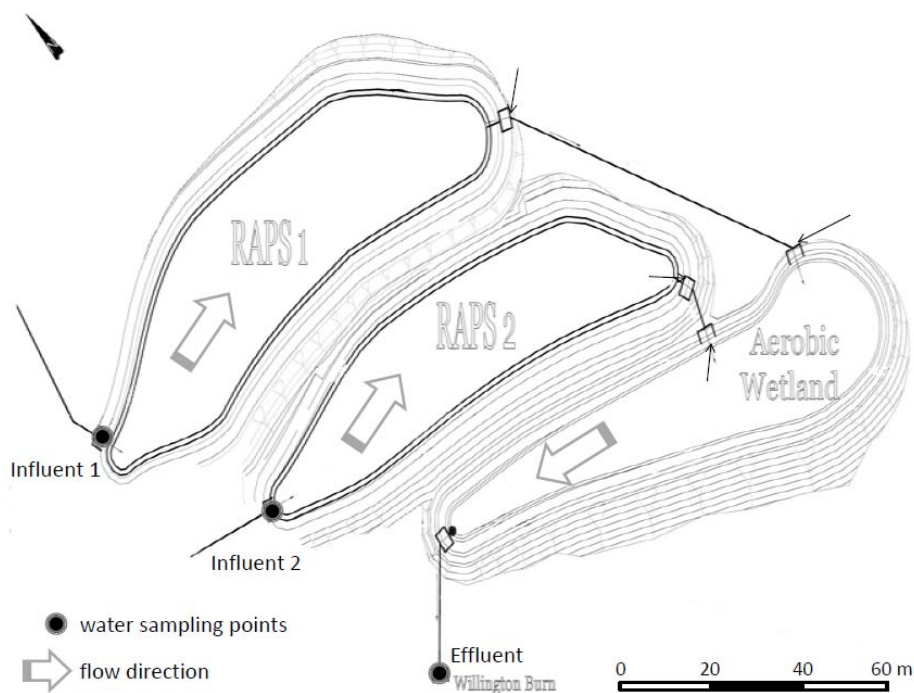
The Bowden Close Treatment Scheme

The Bowden Close treatment scheme was commissioned in December 2003 and has been operating continually ever since. The treatment system is located 12 km southwest of Durham, England, and is composed of two RAPS operating in parallel followed by an aerobic reed wetland that is planted with *Typha latifolia* and *Juncus effusus* (Figure 1). The treated water is discharged into a local burn (Willington Burn), a tributary of the River Wear.

The reactive substrate of the RAPS (limestone gravel, horse manure and straw compost) has an approximate thickness of 0.8 m. The aerobic wetland consists of 1 m deep reed support mineral soil (Durham County Council Design Services 2003).

There are three distinct coal mine water discharges at Bowden Close (Younger *et al.* 2004). Discharges 1 and 2 (directed into RAPS 1 as influent 1) arise from underground coal workings of the Durham coalfield (Younger 1998). The coal seams are characterized by total sulphur concentrations of 1-5% (Younger 1998). Discharge 3 (directed into RAPS 2 as influent 2) emerges from a waste rock pile of the former Bowden Close colliery (Younger *et al.* 2004, Younger *et al.* 2003). The colliery was closed in the 1960s but no measures had been applied to prevent acid mine drainage generation or control its discharge. As a consequence there was a significant impact on the aquatic environment of local streams (Jarvis and Younger 1997). It was this environmental impact that triggered the construction of the treatment system.

The aim of this study was to assess acidity, iron and aluminium removal in the Bowden Close system during its first four years of treatment, with a view to evaluating the likely long-term performance of such passive treatment units.

$A_{RAPS\ 1} = 1511\ m^2$, $A_{RAPS2} = 1124\ m^2$, $A_{AerobicWetland} = 990\ m^2$

Figure 1. The Bowden Close treatment scheme is composed of two Reducing and Alkalinity-Reducing Systems (RAPS) operating in parallel, and one aerobic wetland

2. METHODOLOGY

Since commissioning of the system, in December 2003, influent and effluent water quality monitoring has been undertaken on a monthly basis.

Field Parameters

During each sampling event pH, redox potential (Eh), electrical conductivity (EC), temperature (T), alkalinity and flow rates were determined. In addition, two water samples (60 mL acid washed polyethylene bottles) for the analyses of total concentrations of cations (Ca, Mg, K, Na, Al, Fe, Mn, Si and Zn) and anions (SO_4^{2-} and Cl^-) were taken. Samples for cation determination were acidified (pH<2, conc. HNO_3). Occasionally, filtered (0.45 μm Cellulose nitrate filter) and acidified samples were taken from all sampling points. All samples were stored at 4°C and analysed within four weeks of collection.

The field parameters (pH, EC, Eh, T) were determined with a calibrated Myron 6P Ultrameter II (calibration solution for EC: 1214 μS cm^{-1} solution, calibration solution for pH: 4, 7, and 10). The Ultrameter measures with an accuracy of ±0.01 for pH (data range: 0-14), ±1 mV for the redox potential (data range ±999 mV), ±1% for the electrical conductivity (0-9999 μS cm^{-1}, and 10-200 mS cm^{-1}) and ±0.1°C for the temperature (data range: 0-71°C). The electrodes were cleaned with deionised water and rinsed three times with the mine water prior to each measurement. The alkalinity was measured by colorimetric titration with 1.6 N sulphuric acid to an endpoint of pH = 4.5 (bromcresol green-methyl red indicator, Hach AL-DT test kit). The physicochemical parameters were measured in triplicate every three months as part of the quality assurance / quality control procedure (R^2 >0.98 ($p<0.05$)). Flow rates were determined in triplicate by bucket and stop watch method.

Laboratory Methods

An Inductively Coupled Plasma-Optical Emission Spectrometer (CCP simultaneous ICP-OES, type Vista MPX, Varion) and an Ion Chromatograph (IC, type IC25 Dionex) were applied for the quantification of cations and anions, respectively. The instruments were calibrated by two point and one point calibration. Standard checks were included every ten samples and the instruments were recalibrated as necessary in light of these checks. Pearson coefficients and Pearson residuals of duplicates and triplicates from anion and total and filtered cation analyses indicated good precision above 0.98 (R^2; $p<0.05$) for all parameters. Ion balances (IA, IA = $(c-a)/(c+a)*100\%$; $c = \Sigma$cations, $a = \Sigma$anions in meq l^{-1}) were within $\pm6\%$. Pearson correlation coefficients showed good reproducibility for EC and sum of cations and anions, respectively ($r_{cations\ vs.\ EC}$ 0.983; $r_{anions\ vs.\ EC}$ 0.985, both $p<0.05$).

3. RESULTS

Influent Water Quality

Summary water quality data for the influent water quality of the two RAPS units are shown in Table 1 and Figure 2. A piper plot outlines major geochemical facies (Figure 3).

Both influents were of moderate to strong ionic strength. Influent 1 (from underground workings) was moderately net acidic whilst influent 2 (waste rock) was strongly acidic. Influent 1 showed twice as much bicarbonate buffering capacity than influent 2. These data compared well with the historic data obtained by Younger *et al.* (2003) who assessed pre-treatment mine water conditions. Overall, influent 2 was net acidic (acidity > alkalinity) with pronounced seasonal variations (Figure 2). Influent 1 was largely net-acidic with an approximately five-fold lower acidity concentration than influent 2. Influent 1 was characterised by calcium-magnesium-sulphate water type (Figure 3) whilst influent 2 was Ca-Mg-Fe-Al-SO_4. Total concentrations of iron and aluminium were enriched in influent 2 by a factor of 4 and 6 respectively, compared to influent 1 (Figure 2). Coupled with the significantly higher flow-rates of influent 2, RAPS 2 consequently receives approximately 9 to 10 times higher metal loads than RAPS 1.

Compared to average world stream concentrations (Reimann and de Caritat 1998), iron is enriched by a factor of 395 (influent 1) to 1690 (influent 2) and aluminium is enriched by a factor of 17 and 105 in influents 1 and 2, respectively. Iron, manganese, zinc, sulphate and pH were identified as main treatment targets in both influents. There is no guideline limit defined for aluminium in the UK. However, at elevated concentration aluminium can have suffocating effects on aquatic biota (Nordstrom 1982). Therefore the latter was included in the list of treatment targets based on its enrichment compared to world stream concentrations (Reimann and de Caritat 1998).

Table 1. Average, median, minimum and maximum values for influent water chemistry for the period 2003 to 2007 (underlined values exceed UK guideline values where applicable)

Parameter*	Influent 1	Influent 2
Q	36 / 33 (1-132)52	99 / 72 (15->400)
pH	5.65 / 5.99 (3.2-6.9)	4.93 / 4.87 (4.2-6.2)
EC	836 / 829 (380-1040)	1553 / 1435 (622-2599)
Eh	350 / 320 (150-640)	370 / 373 (180-500)
T	9.7 / 9.6 (5.3-14.1)	9.4 / 9.6 (6.4-18.5)
Alkalinity	24 / 19.5 (0-110)	10 / 1 (0-75)
Acidity	75 / 70 (15-170)	373/302 (46-1189)
Fe	15.8 / 16.2 (1.0-36.4)	67.5 / 57.5 (12.1-355.0)
Al	5.1 / 4.5 (1.2-13.3)	31.6 / 26.3 (1.1-66.0)

Q= flow L min^{-1}, concentrations = mg L^{-1} except pH, Eh in mV (raw data corrected for the standard hydrogen electrode), EC = µS cm^{-1}, T = °C, alkalinity and acidity = mg CaCO$_3$ eq L^{-1}; mean / median (min-max)^{n}, n = sample size: 68 unless otherwise indicated; underlined values indicate exceedances of guideline limits for aquatic life protection, acidities were calculated after Hedin et al. (Hedin et al. 1994)
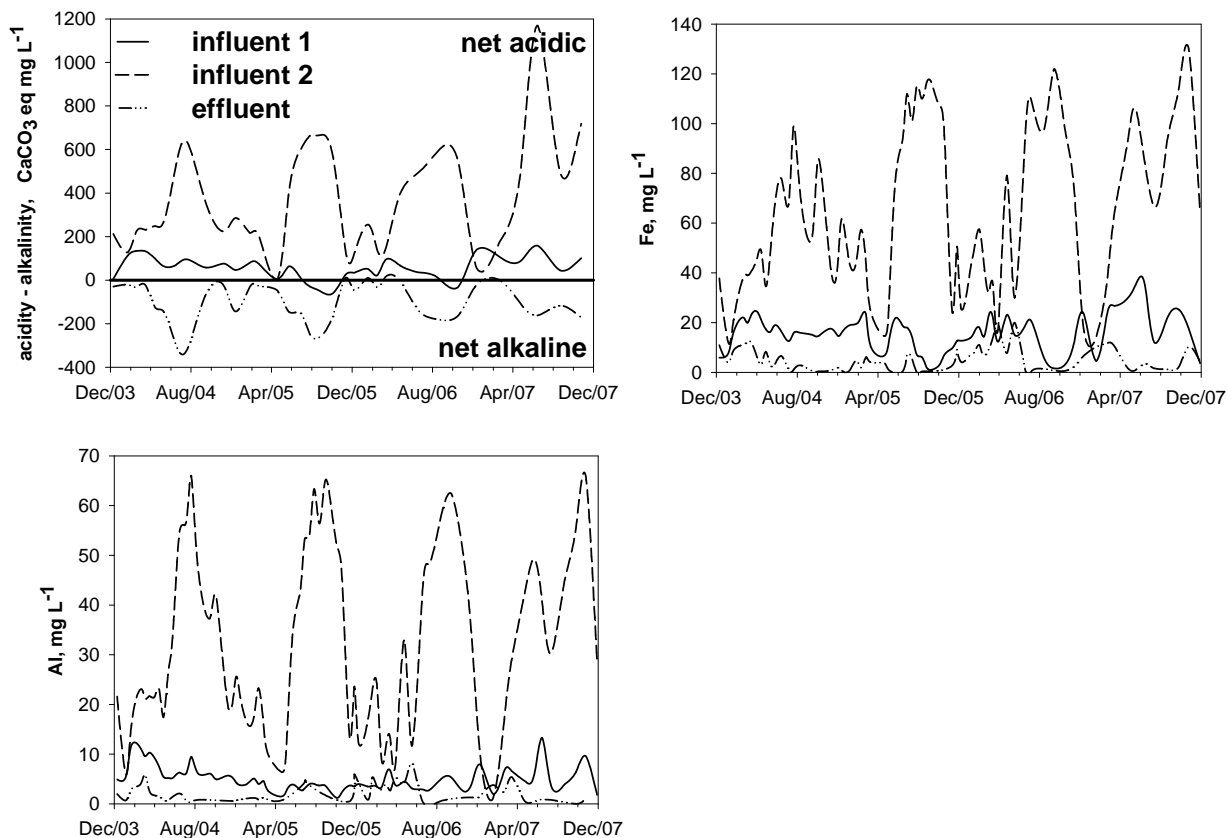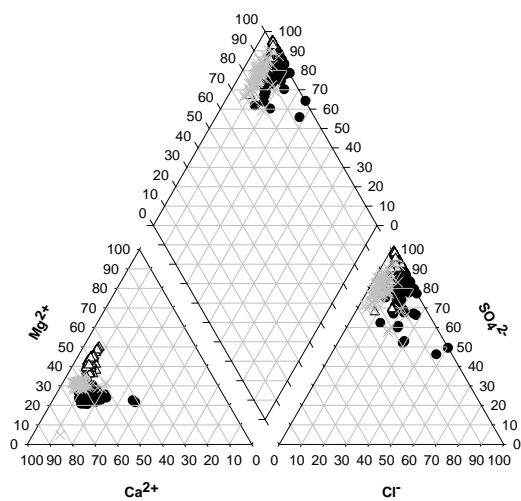
Figure 2 Net acidity (acidity-alkalinity), iron and aluminium concentrations in influents 1 and 2 and effluent

Figure 3. Piper plot for major cations (calcium, magnesium, sodium and potassium) and major anions (sulphate, bicarbonate and chloride) in influents 1 (filled circles) and 2 (open triangles) and the effluent (thin X)

EFFLUENT WATER QUALITY

Water chemistry results and dominant geochemical facies for the effluent are summarised in Table 2 and Figures 2 and 3. The effluent was characterized by circum-neutral pH. Over the four years of monitoring the pH remained above 5.7 (Table 2). Net-acidities were mostly negative (i.e. net-alkaline) with few exceptions during 2006 and 2007 (Figure 2). The water was characterised by a Ca-Mg-SO$_4$–HCO$_3$ water type (Figure 3) with no significant (p<0.05) seasonal percentage variation of major cations, and only minor variations of sulphate and bicarbonate ratios between 0-0.39 (HCO$_3$: SO$_4^{2-}$ in meq L^{-1}). Highest values of pH, alkalinity, electrical conductivity, sulphate, calcium, magnesium, potassium, sodium and manganese were observed during the summer months while maxima of flow, aluminium and iron were determined during the winter season. Acidity, iron and aluminium concentrations were all lower in the effluent than the influent. Despite the improvement of the mine water quality iron (Table 2) and zinc, and especially sulphate and manganese, still exceeded guideline limits. It is worth noting, however, that the UK regulatory authority have not imposed strict requirements on effluent concentrations for the discharge from the system. Rather, the regulator accepts a 'reasonable improvement' in quality given the orphan nature of the site.

Table 2 Average, median, minimum and maximum of physicochemical parameters and major ion concentrations in the effluent of the aerobic wetland for 2003 - 2007 (underlined values breach UK guideline limits)

Parameter	Effluent	Parameter	Effluent
pH	6.95 / 7.0 (5.7-7.9)	Alkalinity	123 / 103 (12-322)
EC	1395 / 1258 (579-2421)	Acidity	30 / 20 (4-105)
Eh	230 / 210 (100-380)	Fe	5.1 / 3.4 (0.2-20)[64]
T	10.1 / 10.5 (1.7-23.6)	Al	2.3 / 1.5 (bdl-8.2)[45]

Concentrations = mg L^{-1} except pH, Eh in mV (raw data corrected for the standard hydrogen electrode), EC = µS cm^{-1}, T = °C, alkalinity and acidity = mg $CaCO_3$ eq L^{-1}; mean / median (min-max)^n, n = sample size: 68 unless otherwise indicated; underlined values indicate exceedances of guideline limits for aquatic life protection, n.m. = not measured, acidities were calculated after Hedin et al. (Hedin et al. 1994)

Treatment Performance

Concentration Efficiencies and Load Removal

Concentration efficiencies are calculated based on concentration differences between two sampling points (Wieder 1993). This approach has the disadvantage that flow rates, loads and the area of the treatment system are not considered. Hence, outcomes are relative and cannot be compared directly to other treatment facilities. Therefore, concentrations have been substituted for loads (Equation 1). The average load removal (ΔL, g d^{-1}) was determined by subtracting effluent loads from influent loads.

(1) $$\Delta L\% = (L_{inf}-L_{eff})/L_{inf} *100\%$$

$\Delta L_{\%}$ = percentage load removal, L_{inf} = influent load including influent 1 and influent 2 (g d^{-1}), L_{eff} =effluent load (g d^{-1})

Overall, the target contaminants acidity, iron and aluminium were removed by 93%, 92% and 96% (median), respectively. Per day, on average 34.1 kg acidity (range: 8.3-347 kg), 6.9 kg iron (range: 0.8-103.8 kg) and 2.7 kg aluminium (range: -0.39-12.4 kg) were removed.

Area Adjusted Removal Rates

For the assessment of the treatment performance area-adjusted removal rates are most commonly calculated (Hedin et al. 1994), which enables the load removal to be adjusted to account for the surface area of the system (Equation 2). Acidity ($\bar{x}$ = 9.4 g d^{-1} m^{-2}, range: 2.3-96 g d^{-1} m^{-2}), iron ($\bar{x}$ = 1.9 g d^{-1} m^{-2}, range: 0.2-29 g d^{-1} m^{-2}) and aluminium ($\bar{x}$ = 0.7 g d^{-1} m^{-2}, range: -0.1-3.4 g d^{-1} m^{-2}) were removed in the substrate.

(2) $$AR = (L_{inf}-L_{eff})/ A$$

AR = area adjusted removal rate in g d^{-1} m^{-2}; L_{inf} = sum of loads of influent 1 and 2 in g d^{-1}, L_{eff} = element load of the effluent in g d^{-1}, A = surface area in m^2 of the Bowden Close treatment site

Total Removal

The total amount of target contaminants removed in the Bowden Close treatment scheme over the first four years of treatment was calculated by multiplying the median monthly removal rates by 48 months. Between 2003 and 2007 about 7.3 t of iron and 3.4 t of aluminium were removed.

Seasonal Treatment Variations

To assess seasonal variability of the treatment performance the data were separated into seasons: 1) November to January, 2) February to April, 3) May to July and 4) August to October and the above performance parameters calculated (Table 3).

Table 3. Seasonal load removal and area adjusted removal rates of major contaminants in Bowden Close (2003-07)

Parameter	Nov-Jan	Feb-Apr	May-Jul	Aug-Oct
<u>Fe</u>	6.3 / 1.7	5.6 / 1.7	4.8 / 1.6	2.9 / 1.3
<u>Al</u>	2.5 / 0.7	2.9 / 0.8	2.2 / 0.6	1.6 / 0.4
<u>Acidity</u>	30.0 / 8.3	31.9 / 8.8	25.4 / 7.0	17.0 / 4.7

Load removal = kg d^{-1} / area adjusted removal rates g d^{-1} m^{-2}, underscores indicate seasonal significant (p<0.05) differences of the parameter

Parametric and non-parametric one way ANOVA indicate that area adjusted removal rates of acidity, iron and aluminium differed significantly (p<0.05) between the seasons. Highest daily removal rates were observed between February and April with exception of iron. For the latter, highest removal rates were recorded for November to January. Occasionally the area-adjusted removal rate was negative i.e. there is a remobilisation of contaminants from the substrate.

Annual Variability

Annual trends of treatment performance were inspected to assess variations during the ageing of the system, with the specific aim of beginning to evaluate the potential lifetime of the system. Parametric and non-parametric one way ANOVA tests indicate that aluminium and net alkalinity (=Δalkalinity +Δacidity) were significantly different (p<0.05) over the course of the four year treatment. However, there was no trend obvious (Table 4). During year 1, about 16.8 t (12.7 g d^{-1} m^{-2}) of net alkalinity has been generated in Bowden Close, compared to 15.5 t (11.7 g d^{-1} m^{-2}), 11.3 t (8.56 g d^{-1} m^{-2}), and 13.3 t (10.1 g d^{-1} m^{-2}) in year 2,3 and 4. Highest iron and aluminium removal was observed in year 1.

Table 4. Annual total removals of major contaminants in Bowden Close and average daily area adjusted removal rates

Parameter	Year 1	Year 2	Year 3	Year 4
Fe	1.69 / 1.28	2.08 / 1.57	1.75 / 1.32	2.39 / 1.81
<u>Al</u>	1038 / 0.78	833 / 0.63	659 / 0.50	988 / 0.75
Acidity	10.1 / 7.7	10.8 / 8.1	8.7 / 6.6	10.6 / 8.0

Units in kg (acidity, iron in t) / area adjusted removal rates in g d^{-1} m^{-2} (calculated from median), underscores indicate annual significant (p<0.05) differences of the parameter

4. DISCUSSION

A considerable improvement of mine water chemical qualities has been observed as a result of the Bowden Close passive treatment system, with particularly marked removal of iron (1.9 g d^{-1} m^{-2}) and aluminium (0.7 g d^{-1} m^{-2}). The treatment scheme removes acidity by an average 9.4 g d^{-1} m^{-2}. This removal rate is higher than the *compliance* and *abandoned mine land* criteria (3.5 and 7 g d^{-1} m^{-2}, respectively) that were suggested as sizing criteria for compost wetlands treating net acidic water (Hedin *et al.* 1994). However, the removal rates in the Bowden Close scheme are substantially lower than the suggested sizing criteria for treatment schemes including RAPS of 20-30 g d^{-1} m^{-2} (Riefler *et al.* 2008, PIRAMID Consortium 2003, Ziemkiewicz *et al.* 2003, Watzlaf *et al.* 2002, Demchak *et al.* 2001, Nairn and Mercer 2000). Danehy *et al.* (2001) reported acidity removal rates for RAPS alone ranging between 6-44 g d^{-1} m^{-2} with averages of 27 g d^{-1} m^{-2}, whilst Nairn and Mercer (2000) stated mean acidity removal rates of 51 g d^{-1} m^{-2} for the first RAPS and iron removal of 17 g d^{-1} m^{-2}. Demchak *et al.* (2001) reported acidity removal rates of 11-52 g d^{-1} m^{-2} for four RAPS. In a review undertaken by Ziemkiewicz *et al.* (2003) the authors included 16 RAPS across several US American states of the age of 1-5 years. Average removal rates of 62 g d^{-1} m^{-2} and ranges of 0-293 g d^{-1} m^{-2} were reported.

In general, removal trends observed in Bowden Close contradict observations at similar treatment systems elsewhere (Woulds and Ngwenya 2004, Hedin *et al.* 1994, Wieder 1993). Specifically, lowest acidity removal rates occur during winter months whereas highest removal rates occur in summer. Bowden Close data clearly show that highest removal rates are obtained during those months in which highest influent loads were determined.

Pearson Correlation Coefficient of area-adjusted removal rates in Bowden Close compared to total influent loads showed positive correlation for target contaminants ($r_{acidity} = 0.99$, $r_{Fe} = 0.99$, $r_{Al} = 0.96$). The latter are removed effectively within the system, but area-adjusted removal rates are low compared to other systems around the world.

This appears to be largely attributable to load limitation rather than underperformance and indicates that the system is operating below its maximum capacity, but it is anticipated that this will extend the lifetime of the system. Certainly, the assessment of data for the period 2003 to 2007 does not indicate that there is any substantive deterioration in performance.

5. CONCLUSIONS AND OUTLOOK

Compared to similar treatment systems, low area adjusted removal rates of iron, aluminium and acidity were observed for the Bowden Close treatment scheme. This, and the elevated percentage removal rates (exceeding 90%) for all three parameters, suggest that the system is load limited. Basic statistical analyses indicate that iron and acidity removal in year four of the treatment was not significantly different compared to year 1. Hence, no decrease in treatment performance was observed. There was a significant difference in element removal with highest removal rates observed during the high flow – high load seasons.

For future data assessment it is critical to include more sophisticated time series analysis methods (e.g. ARIMA) for which at least 5 years of monthly monitoring data are required. Furthermore, in order to allow assessment of the main removal processes, the sampling of filtered surface water and the geochemical and mineralogical analysis of the reactive sediments are required. Only then can long-term predictions of the treatment performance be undertaken.

6. ACKNOWLEDGEMENTS

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