

Determination of Hydraulic Residence Times in Several UK Mine Water Treatment Systems and their Relationship to Iron Removal

Natalie A. S. Kruse · E. Gozzard · A. P. Jarvis

Received: 16 September 2008 / Accepted: 12 February 2009 / Published online: 5 March 2009
© Springer-Verlag 2009

Abstract In the UK, the Coal Authority has more than 40 mine water treatment systems, most of which are wetland systems with settlement lagoon pretreatment. The purpose of treatment in wetlands is the oxidation of ferrous to ferric iron and the subsequent hydrolysis and precipitation of ferric hydroxide within the wetland. It is generally accepted (Hedin et al., Passive treatment of coal mine drainage, 1994, p 35; Skousen and Ziemkiewicz, Acid mine drainage control and treatment, 1996, p 362; Younger et al., Mine water: hydrology, pollution, remediation, 2002, p 442) that this process proceeds by a first-order rate law, although most systems are designed based on an areal removal rate ($10 \text{ g/m}^2/\text{day}$) developed by the U.S. Bureau of Mines (Hedin et al., Passive treatment of coal mine drainage, 1994, p 35); this design guideline inherently assumes a constant removal rate. Given the actual kinetics of iron removal in wetlands, it follows that residence time will control iron removal; given the wide range of system geometries and aspects, it is logical to ascertain the actual hydraulic residence time of wetlands and settlement lagoons and determine the effect this has on iron removal. To make a preliminary assessment of this link, hydraulic residence time of two Coal Authority wetlands (Lambley and Whittle) and two Coal Authority settlement lagoons (Acomb East, Acomb West and Whittle) were measured using bromide tracer tests. Water samples for iron analysis and flow measurements were taken during each tracer test. The Lambley wetland performs well in terms of residence time, and, as reeds become established and adsorptive

processes increase, its iron removal performance (currently 58% removal) may improve, but the low influent iron concentration appears to be a significant impediment to meeting the original performance target. In contrast, the hydraulic performance of the Whittle wetland system is poor, which appears to be due to accumulation of dead plant material coupled with a high length to width ratio. However, performance in terms of iron removal is good (92% removal), which appears to be due to the higher influent iron concentration, and especially the fact that the iron enters the wetland largely in particulate form. The longer residence time of water within the Acomb lagoons ($\approx 12 \text{ h}$) resulted in far more effective iron removal (72% in the east lagoon and 85% in the west lagoon) than the shorter residence time at Whittle (24% iron removal, $\approx 5 \text{ h}$ residence time). Performance (in terms of iron removal) of the settlement lagoon systems appears to be far more closely related to the hydraulic residence time (albeit this conclusion must be tentative, given that only three systems have been investigated, and the Acomb system receives chemical addition). Based on this study, treatment system sizing using 100 m^2 lagoon area per 1 L/s flow appears to be a more appropriate basis for design rather than an areal iron removal rate.

Keywords Hydraulic retention time · Mine water · Passive treatment · Wetland

Introduction

The U.K. Coal Authority has established more than 40 mine water treatment systems that remediate contaminated discharges from abandoned deep coal mines. In the majority of instances in the UK, the discharges treated are

N. A. S. Kruse (✉) · E. Gozzard · A. P. Jarvis
Hydrogeochemical Engineering Research and Outreach Group,
Sir Joseph Swan Institution, Newcastle University,
Newcastle Upon Tyne NE1 7RU, UK
e-mail: natalie.kruse@ncl.ac.uk

net-alkaline (i.e. alkalinity > acidity), due to neutralisation processes occurring as mine water rises through carbonaceous strata en route to the surface (Younger et al. 2002). The primary objective for treatment of these net alkaline, ferruginous, waters is the oxidation of ferrous iron to ferric iron, and then hydrolysis and precipitation of ferric hydroxide within the confines of the treatment system (Skousen et al. 1996). By far the most common treatment train for these mine waters is aeration, followed by settlement lagoons, followed by aerobic wetland(s). In cases where the iron concentration of the mine water is comparatively low, aerobic wetland treatment alone may be sufficient for effective remediation (aeration being assumed to occur within the shallow waters of the wetland) (Skousen and Ziemkiewicz 1996; Younger et al. 2002).

The design of settlement lagoons and aerobic wetlands is based on empirical formulae derived from measurements of performance at pre-existing systems. For aerobic wetlands, by far the most common approach has been to use the simple design formula recommended by the former U.S. Bureau of Mines (Hedin et al. 1994), which is to assume an iron removal rate of 10 g/m²/day. This has been taken as a standard design criteria for wetlands, although the initial publication intended this to be sizing criteria for a system of settlement ponds and wetlands. For settlement lagoons, a number of approaches have been proposed, based on the time to fill the lagoon (usually 48 h) and/or the rate of removal of iron per unit area (as above) (PIRAMID Consortium 2003).

The Coal Authority typically employs settlement lagoons as the preliminary treatment unit for the removal of iron from net-alkaline mine waters, aiming to remove 50–80% of the iron from the mine water being treated. Settlement lagoons are typically preceded by aeration systems, such that the sole purpose of the lagoons is to allow

hydrolysis of ferric iron to ferric hydroxide and settlement of this precipitate (Younger et al. 2002).

A number of simplified approaches have been adopted for the design of settlement lagoons. These sizing formulae can be summarised as follows (from PIRAMID Consortium 2003):

- a standard, nominal hydraulic retention time of 48 h (in fact, the time to fill, as defined in the notes to Table 1)
- 100 m² of lagoon area per 1 L/s of mine water to be treated
- Application of the aerobic wetland sizing criterion outlined above (i.e. assuming an iron removal rate of 10 g/m²/day).

The length to width ratios of such units varies widely, and is as much determined by the availability and topography of land at a treatment site as it is by any other consideration.

An underlying assumption of all of these design formulae is that the reactions of iron oxidation, hydrolysis, precipitation, and settling of particulate iron will occur at a certain (constant) rate. Certainly under laboratory conditions, iron oxidation and hydrolysis is known not to occur at a constant rate; rather, it approximates a first-order reaction (i.e. at higher initial iron concentrations, it will occur faster) (Hedin et al. 1994). However, no investigation has ever assessed the actual hydraulic residence time of the Coal Authority's mine water treatment systems, although some guidance can be drawn from studies conducted in the United States (Keefe et al. 2004; Lin et al. 2003), in U.K. RAPS systems (Wolkersdorfer et al. 2005), and modelling studies (Goebes and Younger 2004). Much could be gained from such an investigation, since this would allow quantification of the actual rate of iron attenuation, and provide an indication of the

Table 1 Summary of design and operations characteristics of field sites

	Lambley	Whittle wetland	Acomb East	Acomb West	Whittle lagoon
Age of system (years)	1	5	5	5	5
System area (m ²)	4 388	2 400	375	375	900
Nominal water depth (m)	0.300	0.300	3	3	1.65
Nominal design volume (m ³)	2409	721	1082	1082	1320
Length (m)	65	200	7.5	7.5	75
Width (m)	23	25	5	5	25
Ratio length/width	2.8	8	1.5	1.5	3
Inflow rate (L/s)	84	25	6.25	5.85	25
Influent Fe (mg/L)	3.8	20.8	34.1	34.1	28.6
Influent Fe loading (kg/day)	27.6	44.9	18.4	17.2	61.8
Effluent Fe (mg/L)	1.6	1.7	9.65	5.05	21.6
Effluent Fe loading (kg/day)	11.6	3.67	5.21	2.55	46.7

hydraulic efficiency of these systems. Since the effectiveness of iron removal is time-dependent (irrespective of any geochemical differences), it is logical that a more hydraulically efficient system will achieve better iron removal per unit area. By measuring hydraulic retention time in systems, we may begin to understand what type of system geometries facilitate the greatest hydraulic efficiency, and thereby improve engineering design to optimise the overall performance of such mine water treatment units.

The objective of this work was to make a preliminary assessment of precisely these issues. Hydraulic residence time of the lagoons at the Coal Authority's Whittle and Acomb mine water treatment systems were therefore determined, together with residence times of the wetlands at Lambley and Whittle mine water treatment systems, all in northeast England.

Methods

System characteristics of each field site are summarised in Table 1.

Lambley Wetland

The Lambley wetland is a system of four ponds in series that became operational in 2006; they were planted during construction, although at the time of the tests, the plants had not become fully established. They were designed to treat a low influent iron concentration, 6 mg/L. The wetlands are lined with a geosynthetic clay liner. The system was designed based on the area-adjusted removal rate for iron of 10 g/m²/day (Hedin et al. 1994), giving a total treatment area (with a margin for safety) of 4,388 m². Mine water is pumped from a wet well at a mean rate of 88 L/s; over the period of tracer tests at Lambley, the flow was in fact just under 84 L/s (measured using a CTD diver situated behind the outlet weir and a corresponding baro-diver for atmospheric correction). Variation in flow is mostly due to the pump switching on and off to maintain a near-constant water level in the pumping shaft. Turbulence above the weir is a source of error for the flow data since it adds noise to the data recorded by the diver. Based on water levels recorded during the period of the tracer tests, the system volume is approximately 2,047 m³, giving a time to fill (not hydraulic residence time) of about 7 h.

Whittle Wetland and Lagoons

The Whittle system comprises an aeration cascade and two separate elongate lagoons of similar size followed by

three reedbeds in series; each unit in the system is lined with a geosynthetic clay liner. The system became operational in 2002. The design influent iron concentration is 40 mg/L with a design flow rate of 46 L/s. The lagoons were designed to remove 50% of the influent iron while the wetlands were designed using the area-adjusted removal rate of 10 g/m²/day (Hedin et al. 1994) for the remaining iron (20 mg/L after lagoon treatment). The lagoons are separated by a central bund and have a nominal design depth of 1.65 m. The total surface area of the lagoons is 900 m² with a total volume of 1,320 m³. The lagoons have a high length to width ratio of 10:1.

The total treatment area of the three wetlands is 7,210 m² (approximately 2,400 m² per reedbed). The total volume is approximately 2,160 m³. The design assumes a nominal depth of 300 mm, although this is now reduced due to ochre and reed debris build up. The flow rate of the mine water discharge is approximately 25 L/s, so the time to fill the lagoons is slightly under 15 h and the time to fill the wetlands is about 30 h.

Only the first of the three reedbeds was studied during this investigation since the three nearly identical wetlands operate in series. The Whittle reedbeds are a good contrast to the Lambley wetlands because the wetlands at Whittle are mature, with thick stands of *Typha latifolia* and *Phragmites australis* in all three wetlands.

Acomb Lagoons

The Acomb treatment system includes two lagoons that operate in parallel, followed by wetland treatment. This study focused only on the lagoons. The two lagoons are identical in size and layout; the total surface area of the lagoons is 750 m². Each lagoon is a 3 m deep basin with a trapezoidal cross-section; they have a combined volume of 2,164 m³. The design flow of the system was 15 L/s. Originally, the system was designed to ensure a 48 h nominal residence time, but limitations of land availability restricted the final size. The calculated design time-to-fill of the system is 40 h. The flow during the tracer tests was 12.1 L/s; this gives an actual time-to-fill of 50 h. Initially, the performance of the lagoons for Fe removal was poor, presumably due to incomplete oxidation of ferrous iron; hence, hydrogen peroxide was added to the treatment scheme after the aeration cascade to aid rapid conversion of ferrous iron to ferric iron. During February 2007, field observations showed that the East lagoon had greater build-up of ochre, reaching the water surface in places. Approximately 60 tonnes of wet sludge were removed from the East lagoon in September 2006; subsequently, sludge was again removed from the East lagoon in July 2008.

Hydraulic and Chemical Testing

Estimates of hydraulic residence time for the units selected were initially made by a combination of the nominal time to fill (defined as the time to completely fill the unit with water from empty) calculations and visual observations. Based on the volume of the system (and assuming complete mixing), a suitable mass of sodium bromide (NaBr) was then accurately weighed, and dissolved in 10–20 L of water. A suitable mass was that which would ensure that, even if complete mixing within the wetland or lagoon occurred, a detectable concentration of bromide would still be evident in the effluent from the treatment unit being investigated. Fisher Scientific Laboratory reagent grade NaBr (>99%) was used for all tests.

Aquamatic Auto Cell P2 autosamplers were used to collect influent and effluent samples in 24 l L HDPE bottles over the test period. All bottles were cleaned with 10% HNO₃, rinsed three times with tap water, and then rinsed three times with deionised water prior to deployment; only total iron was determined in the samples since samples were taken over the course of 24 h and could not be filtered or preserved immediately. Only unfiltered total iron was measured; the auto-sampler bottles were acidified immediately after the conclusion of the tracer test. Influent bromide concentrations were measured every 4 h for 24 h from the start of the tracer tests and were consistently below 0.30 mg/L for all sites. Specific field methods for each system are summarised in Table 2.

Samples were subsequently analysed. Bromide concentration was used to determine hydraulic residence time and iron concentration was used to determine system performance. Fe and Br were analysed using a calibrated Varian Vista MPX ICP-OES, and anions were determined using a Dionex IC 25 Ion Chromatograph; the limit of detection of bromide was 0.1 mg/L.

Residence time was nominally defined in this study as the time it took for 70% of the recovered NaBr tracer to leave the wetland (rather than the total added to the wetland). This was used rather than the time to the peak of the tracer test to decrease the influence of preferential flow paths on the results. Despite this decision, the peak residence time has been determined and recorded in the results section of this paper.

The percent recovery was calculated for each test. This was done with mass balance calculations using a piecewise integration of the bromide breakthrough curve to determine mass of bromide per litre of water that left the system during a unit of time (taken as the time between samples); this value has the units of mass volume⁻¹ time. The flow rate was then used to convert this into a mass of bromide that left the treatment system during that unit time. These were summed over the duration of the tracer test and compared to the mass of bromide added to the system (calculated from the chemical formula for NaBr) to determine the percent recovery.

Results

Lambley Wetland

Of the tracer added to the system, 84.4% was recovered in the outflow samples. This is a very high proportion for tracer tests of this sort. For example, Whitmer et al. (2000) recovered an average of 48% of bromide tracer from 12 tracer tests. Losses may be due to a combination of retention of the tracer in quiescent zones of the wetland and some adsorption of the bromide to organics in the wetland substrate.

Figure 1 shows the results of the tracer test. The peak bromide concentration in the effluent from the wetland was

Table 2 Summary of tracer test methods

	Lambley wetland	Whittle wetland	Acomb lagoons	Whittle lagoon
Test date	26 February 2007	28 February 2007	26 February 2007	1 March 2007
Mass of NaBr (kg)	1.90	2.16	2.17	1.320
Volume of tracer solution (L)	12	10	13	13
Duration of inflow sampling (h)	24	24	24	24
Inflow sample frequency (min)	60	60	60	60
Duration of outflow sampling (h)	16	16	24	16
Outflow sample frequency (min)	20	20	1	20
Flow measurement method	CTD diver	Historic avg ^a	Historic avg ^a	Historic avg
Duration of flow measurement (h)	24	n/a	n/a	n/a
Flow measurement frequency (min)	15	n/a	n/a	n/a

^a Average inflows supplied by Weirs Engineering

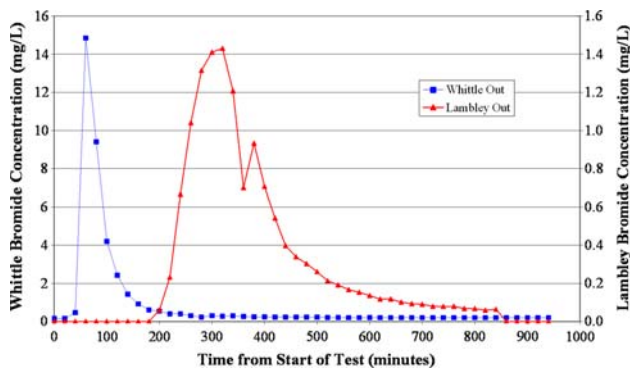


Fig. 1 Bromide concentrations in effluent from Lambley and Whittle wetlands during tracer test (commenced 26/02/2007 at 11:45 and 28/02/2007 at 11:30, respectively)

reached 5 h and 20 min (320 min) after the tracer was added to the inlet. During the test period, the bromide concentration measured at the inlet was below the limit of detection. The earliest detectable bromide in the outflow was found 3 h and 20 min (200 min) after the tracer was added. The last detectable tracer in the effluent was recorded 14 h after commencement of the test. The hydraulic residence time (which here we define as the time it took for 70% of the recovered tracer mass to leave the wetland) of the system was determined to be 6 h, 25 min (385 min). The implications of these findings, in terms of hydraulic efficiency of the system, are discussed below.

The Lambley wetland system was designed to reduce an influent iron concentration of 6 mg/L to an effluent concentration of 1 mg/L. To achieve this level of treatment, the system must act at a removal rate of 8.25 g Fe/m²/day. During the test period, the average influent iron was 3.84 mg/L and the average effluent iron was 1.63 mg/L. Based on a total treatment area of 4388 m² and an average flow of 83.8 L/s, the actual area-adjusted removal rate during the test period was 3.66 g/m²/day, nearly one-third of the design removal rate and less than half the rate required to reach design iron removal performance.

Whittle Wetland

Of the tracer added to the system, 60.3% was recovered in the outflow samples. The lower total recovery of tracer than at Lambley may be due to (a) leakage from the system via a breach mid-way along the southern embankment of the wetland and (b) greater adsorption of the bromide tracer by the mature plants in this system.

The peak of the tracer test was reached 90 min after the tracer was added to the inlet. The bromide was detected in a single peak (Fig. 1). The bromide concentration in the influent water was 0.216 mg/L on average over the sample period. The earliest detectable bromide in the outflow was

found 70 min after the tracer was added. The last of the tracer left the system after 8 h and 50 min. The residence time was 1 h, 43 min (103 min).

The Whittle mine water treatment wetland system was designed to treat water with 20 mg/L iron at a flow rate of 46 L/s. To achieve this level of treatment, the system would have to act at an area-adjusted removal rate of 11 g/m²/day. During the test period, the average influent iron concentration to reedbed 1 was 20.8 mg/L and the average effluent iron was 1.69 mg/L. Based on the treatment area of one wetland (2,400 m²) and an average flow-rate of 25 L/s, the actual area-adjusted removal rate was 17.2 g/m²/day. Thus, the system was performing better than the design removal rate, despite the comparatively poor hydraulic performance of the system (illustrated in Fig. 1). Possible reasons for this result are discussed below.

Acomb Lagoons

It was not possible to inject tracer into each of the East and West lagoons at Acomb separately. Therefore, a single injection was made upstream of the bifurcation of the channel into each lagoon. Over the test period, 83.2% of the NaBr tracer was recovered in the outflow samples from both lagoons.

The relative flow of the two lagoons was determined based on the mass of bromide detected from each lagoon. The east pond received about 51.7% of the flow and the west pond received about 48.3% of the flow. Hence, the estimated flow was 6.25 L/s through the east pond and 5.85 L/s through the west pond.

In the East Lagoon, the peak bromide concentration in the outflow samples occurred after only 80 min. The bromide came in two peaks; the second was detected at 8 h 20 min (500 min) (Fig. 2). This suggests three primary flow paths through the lagoon. The residence time was 12 h, 12 min (732 min). The influent iron concentration

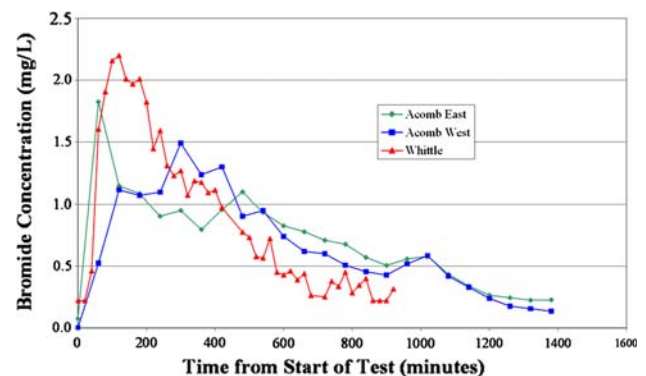


Fig. 2 Bromide concentration in effluent from Acomb East, Acomb West and Whittle lagoons during tracer test (test commenced on 26/02/2007 at 14:10 at Acomb and 01/03/2007 at 11:55 at Whittle)

was 34.1 mg/L. The average effluent iron concentration over the test period was 9.65 mg/L. Based on the calculated flow distribution and half of the total system area, the removal rate of the East lagoon was determined to be 35.2 g/m²/day.

The West lagoon tracer test showed a similar residence time to the East pond, although the time to peak was significantly longer. The peak bromide concentration, as shown in Fig. 2, was detected 5 h, 20 min (620 min) after addition of the tracer. The residence time of the west pond was 11 h, 38 min (698 min). The West lagoon receives slightly less water than the East lagoon, and also has much less build-up of ochre. The mean effluent iron concentration of the West lagoon over the test period was 5.05 mg/L. Based on the calculated flow and half of the total area, the removal rate of the West lagoon was 39.2 g/m²/day. The areal removal rate of the west lagoon was therefore more than 10% better than that of the East lagoon.

Whittle Lagoons

During the tracer test, 82.4% of the NaBr tracer was recovered in the outflow samples (All calculations refer to both lagoons, treated as a single unit.). The peak bromide

concentration was detected in the effluent 125 min after the tracer was added to the influent mine water. The first detectable tracer in the outflow was 45 min after the tracer was added to the influent. The last detectable tracer in the effluent was 14 h and 5 min (845 min) after injection. The residence time of the lagoon was 5 h, 13 min (613 min). The long tail after the peak in bromide concentration (Fig. 2), illustrates that a portion of the water was retained within the lagoon and released slowly over time (approximately between 2.5 and 10 h after injection of the tracer).

During the test period, the mean influent iron concentration was 28.6 mg/L. The average effluent iron concentration over the same time period was 21.6 mg/L. The effluent iron concentration was highly variable, but this variation appears to be systematic rather than chaotic. Based on the mean effluent iron concentration, the lagoon had an area-adjusted removal rate of 16.8 g/m²/day during the test, but was only operating at a Fe removal efficiency of approximately 25% (efficiency in this context is defined in the footnotes to Table 3). Given that the lagoon was designed to remove 50% of the iron at a flow rate of 46 L/s (*cf* actual flow rate of 25 L/s), it is clear that the unit is performing well below expectations.

Table 3 Summary performance data and metrics for the Lambley and Whittle wetland treatment systems and the Acomb and Whittle lagoon treatment systems

	Lambley	Whittle wetland	Acomb East	Acomb West	Whittle lagoon
Area-adjusted iron loading (g/m ² /day)	6.3	18.7	49	45.9	68.7
Mean iron removal efficiency (%) ^a	58	92	72	85	24
Mass of iron removed (kg/day)	16	41.2	13.2	14.7	15.1
Area-adjusted removal rate (g/m ² /day)	3.6	17.2	35.2	39.2	16.8
Peak residence time (h) ^b	5.6	1.5	1.3	5.3	2.1
% mass passed at time of peak (%)	49.7	57.7	49.9	28.7	27.5
70% residence time (h) ^c	6.4	1.7	12.2	11.6	5.2
Calculated time to fill (h) ^d day	6.9	12	48.1	51.3	14.7
Residence time adjusted removal rate (g/m ²) ^e	0.96	1.32	17.9	18.9	3.6
Hydraulic loading rate (m/day) ^f	1.65	0.90	1.4	1.3	2.4
NaBr Tracer mass added (kg)	1.901	2.163	2.165	2.165	1.320
Tracer recovered (%)	84.4	60.3	83.2	83.2	82.4
Volumetric removal rate (g/m ³ /day)	9.43	305.6	451.4	131.5	80.0
Aeration prior to unit? (yes or no)	No	Yes	Yes	Yes	Yes
Chemical dosing prior to unit?	None	None	35% peroxide	35% peroxide	None

^a Influent iron concentration minus effluent iron concentration, divided by influent iron concentration and multiplied by 100 i.e. $((C_{\text{inf}} - C_{\text{eff}})/C_{\text{inf}}) \times 100$

^b Defined as the time at which the highest concentration of tracer is recorded in the wetland effluent stream

^c Defined as the time taken for 70% of the tracer to leave the wetland

^d Defined as the time it would take to fill the system from empty, and calculated from system volume data provided by the Coal Authority and its consultants

^e Calculated by dividing 70% residence time by 24, and then multiplying by area-adjusted removal rate

^f Flow-rate in units of m³/day divided by wetland area (in m²)

Discussion

The bromide tracer tests conducted were highly effective in determining the hydraulic residence times of the systems investigated. Mass balance calculations showed that typically over 80% of the tracer was transported through the treatment units, i.e. Br is a suitable conservative tracer for these systems. This is an especially good recovery of bromide; Whitmer et al. (2000) had a recovery of 48% and Keefe et al. (2004) recovered as little as 16% from their wetland tracer experiments. It has been shown (Xu et al. 2004) that both *Pragmites australis* and *Typha latifolia* can uptake significant amounts of bromide within leaf and root structures; this is likely the main cause of bromide loss in these tracer tests.

Tables 1 and 3 summarise the key statistics for iron removal of the two wetland systems evaluated, and also interprets these data using a number of performance metrics. The most commonly applied design criteria for aerobic wetlands for the treatment of net-alkaline, iron-rich, mine water were proposed by Hedin et al. (1994). Both the Lambley and Whittle systems were designed using this guidance, specifically by calculating the wetland area using the area-adjusted iron removal rate of 10 g/m²/day.

What is immediately clear from Table 3 is that actual performance of the wetlands is markedly different in terms of area-adjusted removal rate; 3.6 g/m²/day at Lambley compared to 17.2 g/m²/day for the Whittle wetlands. Thus, the obvious conclusion is that the wetlands at Whittle are considerably more effective—a conclusion clearly borne out by the mean influent and effluent iron concentrations given in Table 3 (and, in this case, also the iron removal efficiency of the system), although this may be misleading due to the effects of influent iron concentration (Whittle receives a significantly higher load, see Table 1).

It is therefore surprising that the residence times of the two wetland systems show that mine water is retained within the Lambley system for nearly 4 times as long it is in the Whittle system. Notwithstanding the influence of any differences in mine water geochemistry (discussed below), the extent of oxidation of ferrous iron and precipitation of ferric hydroxide is clearly time-dependent. Based on the hydraulic residence time alone, the Lambley system would be expected to out-perform the Whittle system. Therefore, the calculated values for area-adjusted removal rate and hydraulic residence time give contradictory indications in terms of the performance of the two wetland systems. There are a number of factors that may help explain these apparently paradoxical results:

- (1) The kinetics of oxidation of ferrous iron and hydrolysis of ferric hydroxide approximates a first-order

reaction; the reaction rate is slower at lower initial iron concentrations (Hedin et al. 1994; Younger et al. 2002). Therefore, the lower area-adjusted removal rate at Lambley may be a function of the lower initial iron concentration (only 3.8 mg/L) rather than reflecting the influence of residence time.

- (2) The mine water at Whittle has passed over an aeration cascade and through a settlement lagoon prior to entering the wetland; there is therefore opportunity for oxidation and precipitation of ferric iron. The effect of this is evident from visual inspection; mine water entering the Lambley wetland is very clear, whilst that entering the Whittle wetland is notably turbid due to iron hydroxide solids.
- (3) The Lambley unit was commissioned in 2006, and therefore, at the time of the tracer test, the reeds were very sparsely populated in the wetland and were only 200–300 mm in height. The absolute instantaneous volume of water within the wetland per unit area may therefore be relatively high in the absence of the dead plant material that builds up in such systems in the medium- to long-term, resulting in a longer residence times, although this relationship is complex since dead plant matter will also act as a carbon source, aiding treatment. The smooth shape of the Lambley curve with a single peak shown in Fig. 1 also implies that the flow is reasonably well distributed through the system (*cf* the ‘spikey’ curve seen for Whittle; see below). In contrast, the Whittle wetland is a mature system, with well-established reeds and a substantial build up of dead plant material. This can lead to the development of channels through a wetland, resulting in poor hydraulic performance, as evidenced by the short (1.5 h) peak residence time and very rapid transit of the tracer through the system (less than 4 h, as shown by the very sharp peak on Fig. 1). However, the build up of plant material (living and dead) may also have beneficial consequences for Fe removal. Adsorption of Fe to plant material, as iron plaques, is well documented (e.g. Batty and Younger 2002), and physical filtering of precipitated ferric iron may be a particularly important attenuation process. This process is expected to be important at Whittle, given the turbid nature of the water entering the Whittle wetland, as noted above.

Several conclusions can be drawn from these findings, despite being based on observations at only two systems. There does not appear to be a relationship between area-adjusted removal rate and hydraulic residence time. Other factors, in particular the initial iron concentration and the maturity of the plants within the wetland, appear to mask any relationship. Notwithstanding this, intuitively there

seems little doubt that, where all other factors are equal, iron removal would be more efficient in a wetland with longer hydraulic residence time. The system at Lambley appears more effective in ensuring long retention times given the flow rate and wetland area. Two reasons that may be responsible for the longer retention time are the distribution of water across the full width of the wetland and the lower length to width ratio. Additionally, the influence of plants on the hydraulic residence time appears to be complex. The main cause of the short hydraulic residence time in the Whittle wetland may be the maturity of the reeds and accumulation of dead plant material, resulting in channelization within the wetland and short residence time as a consequence. Conversely, adsorption processes (on live and dead plant material) may assist in the effective immobilisation of iron within the Whittle wetland. It would be an interesting exercise to evaluate iron removal rates and hydraulic residence time over a period of 5–10 years, from initial colonisation of reeds through to complete maturation and accumulation of dead plant material, in order to establish whether there is an optimum period for long retention time and high iron adsorption capacity. The results of such an investigation could certainly influence the frequency of maintenance of such systems and the possible need for thinning of reeds.

Summary results for the Acomb and Whittle lagoon systems are given in Table 3. The two lagoons at Acomb are almost identical in terms of their size and layout and estimated flow to each is also similar. The notable difference is that the West lagoon has a significantly lower mean effluent iron concentration (5.05 mg/L compared to 9.65 mg/L). Given the otherwise similar nature of the systems, we can only conclude that this is due to the accumulation of ochre sludge in the East lagoon. This may cause short-circuiting of flow across the East lagoon due to the reduction in water depth, resulting in a shorter peak residence time (1.3 h compared to 5.3 h in the West lagoon) and ‘carry over’ of solids in the effluent, although ochre may provide surface area for further iron precipitation. The clear conclusion from this is that regular removal of sludge from settlement lagoon systems is very important if efficient performance is to be maintained, since pond volume will decrease with time due to build up of both ochre and debris.

The Acomb mine water is dosed with hydrogen peroxide prior to discharge to the settlement lagoons, whereas the Whittle mine water is not. Direct comparison of performance between the two systems must therefore be tentative. Notwithstanding this *caveat*, it is clear from Table 3 that the residence time in the Whittle system is less than half that of the Acomb lagoons; this is reflected in far less effective Fe removal (see iron removal efficiency and area-adjusted removal rates). The high hydraulic loading

rate (2.4 m/day) combined with the high length to width ratio of the Whittle system, appears to result in streaming across the system, and consequently poor Fe treatment performance. Again, regular removal of iron hydroxide sludge from the lagoon may improve performance, particularly given that this lagoon is only 1.65 m deep.

Like the wetlands tested, iron removal rate (and volumetric removal rate) had an inverse relationship with hydraulic loading. This points to the need for increased hydraulic efficiency (residence time per unit wetland area), both for better land use and for more efficient iron removal. Additionally, removal rates were higher in the systems (both wetlands and lagoons) with higher influent iron concentrations; this is consistent with the assumption of first-order removal of iron by wetland and lagoon systems.

Conclusions

The Lambley wetland performs well in terms of hydraulic efficiency (i.e. long residence time), and as reeds become established, performance may improve as adsorptive processes increase, but the low influent iron concentration appears to be a significant impediment to meeting the original performance target. In contrast, the hydraulic performance of the Whittle system is poor, which appears to be due to accumulation of dead plant material coupled with a high length to width ratio. However, performance in terms of iron removal is good, which appears to be due to the higher influent iron concentration, and the fact that the iron enters the wetland largely in particulate form. Consequently, the wetland actually exceeds the design area-adjusted removal rate of 10 g/m²/day but, given the very short residence time, it is questionable whether this could have been anticipated at the design stage.

Performance of the lagoon systems appears to be far more closely related to the hydraulic residence time (albeit this conclusion must be tentative given that only three systems have been investigated, and the Acomb system receives chemical addition). In short, the longer residence time of water within the Acomb lagoons (approximately 12 h) results in far more effective Fe removal performance than the shorter residence time at Whittle (approximately 5 h). Some more recommendations for design and maintenance are as follows:

- Accumulation of ochre in lagoons would appear to reduce hydraulic residence time, and consequently reduce performance in terms of iron removal. Regular sludge removal operations are therefore recommended if consistent performance is to be maintained.
- Irrespective of the hydraulic efficiency of the systems investigated, use of the area-adjusted removal rate for

iron of 10 g/m²/day would appear to be a highly conservative approach to design of lagoons; even the Whittle lagoon, with a hydraulic residence time of just 5 h, has an area-adjusted removal rate of ca. 17 g Fe/m²/day. From the evidence of this investigation, lagoons designed on this basis would be substantially over-sized.

- Sizing using 100 m² lagoon area per 1 L/s flow appears to be the most appropriate basis for design. This is equivalent to a hydraulic loading rate of 0.864 m/day. The lagoons at Acomb perform well, and have a hydraulic loading rate of 1.3–1.4 m/day. Given that performance is enhanced by the addition of hydrogen peroxide, design based on a slightly more conservative hydraulic loading rate of 0.864 m/day would seem to be appropriate for systems where no chemical addition is used. One *caveat* to this is that hydraulic loading rate does not account for lagoon depth. Shallower depths may result in streaming across lagoons, shortening hydraulic residence time. Since this problem may be exacerbated by accumulation of ochre sludge, where feasible, greater lagoon depth will help ensure robust performance.

Acknowledgment This work was funded by the Coal Authority; the authors relied greatly on assistance from Dr. Ian Watson.

References

- Batty LC, Younger PL (2002) Critical role of macrophytes in achieving low iron concentrations in mine water treatment wetlands. *Environ Sci Technol* 36:3997–4002
- Goebes M, Younger PL (2004) A simple analytical model for interpretation of tracer tests in two-domain subsurface flow systems. *Mine Water Environ* 23:138–143
- Hedin RS, Nairn RW, Kleinmann RLP (1994) Passive treatment of coal mine drainage. US Bureau of Mines IC9389, Pittsburgh, PA, USA, p 35
- Keefe SH, Barber LB, Runkel RL, Ryan JN, McKnight DM, Wass RD (2004) Conservative and reactive solute transport in constructed wetlands. *Water Resour Res* 40 (article W01201)
- Lin A, Debroux J, Cunningham J, Reinhard M (2003) Comparison of rhodamine WT and bromide in the determination of hydraulic characteristics of constructed wetlands. *Ecol Eng* 20:75–88
- PIRAMID Consortium (2003) Engineering guidelines for the passive remediation of acidic and/or metalliferous mine drainage and similar wastewaters. University of Newcastle Upon Tyne, Newcastle, UK, 166 pp
- Skousen JG, Ziemkiewicz PF (1996) Acid mine drainage control and treatment. West Virginia University, Morgantown, WV, USA, p 362
- Skousen JG, Sexstone A, Garbutt K, Sencindiver J (1996) Wetlands for treating acid mine drainage. In: Skousen JG, Ziemkiewicz PF (eds) *Acid Mine Drainage Control and Treatment*. West Virginia Univ, Morgantown, WV, USA, pp 199–210
- Whitmer S, Baker L, Wass R (2000) Loss of bromide in a wetland tracer experiment. *J Environ Qual* 29:2043–2045
- Wolkersdorfer C, Hasche A, Göbel A, Younger PL (2005) Tracer test in the Bowden Close passive treatment system (UK)—preliminary results. *Wissenschaftliche* 28:87–92
- Xu S, Leri A, Myneni S, Jaffe P (2004) Uptake of bromide by two wetland plants (*Typha latifolia* L. and *Phragmites australis* (Cav.) Trin. ex Steud). *Environ Sci Technol* 38:5642–5648
- Younger PL, Banwart S, Hedin RS (2002) *Mine water: hydrology, pollution, remediation*. Kluwer Academic Publisher, London, UK, p 442