A comparative study of hydraulic residence time using a multi-tracer approach at a coal mine water treatment wetland, Lambley, Northumberland, UK[1]

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Abstract. The design approach most commonly applied for coal mine treatment wetlands in the UK by the Coal Authority has been to use the area-adjusted iron removal formula proposed by the US Bureau of Mines. Because the efficiency of iron removal is dependent on concentration-time distribution, a more hydraulically efficient system will result in better removal of iron. Thus, in this study the efficiency of iron removal was studied in relation to hydraulic residence time in an aerobic wetland system receiving net-alkaline, iron-rich mine water at Lambley, Northumberland. Two tracer tests were completed, the first was conducted four months after site commissioning (February 2007), the second was conducted 21 months after construction (July 2008). Significantly greater removal of iron (85% Fe removal efficiency) was observed during the more recent experiment as a consequence of considerably longer residence time compared to the initial tracer study during the first year of wetland operation (58% Fe removal efficiency). The results of multi-tracer tests (July 2008) using sodium bromide, Na-Fluorescein and sodium chloride demonstrated an essentially identical retention time between 9.95-10.7 hours, with tracer recovery between 64 and 120%. The relative merits of using these different tracers are discussed. Initial tracer tests using sodium bromide at the site (February 2007) yielded a residence time of 5.6 hours. Factors attributed to these differences over time include the maturity of reeds since site commissioning, seasonal effects and dispersive behaviour of flow within the wetland. Despite the improved removal of iron observed during the more recent summer tracer test, the wetland system is still performing at a rate below the designed 10 $g/m^2/d$ of iron removal by 30%. Implications for design guidance for coal mine drainage surface flow wetlands are discussed.

Additional Key Words: tracer test, aerobic wetland, iron removal

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INTRODUCTION

Tracers can be used to determine the transport and fate of contaminants in surface and ground waters. Multi-tracer tests consisting of simultaneous slug injection of sodium bromide (NaBr) and sodium fluorescein (Na-Fluorescein) and a separate slug injection of sodium chloride (NaCl) were carried out at Lambley mine water treatment wetland, Northumberland, to determine the hydraulic residence time of the system. In addition, iron removal was determined so that the hydraulic efficiency within the treatment wetland could be more precisely evaluated. Optimisation of existing wetland operations requires a better understanding of the hydraulic and geochemical factors that govern contaminant behaviour (Lin et al., 2003).

The Lambley wetland consists of four ponds in series with a total treatment area of 4388 m^2 (Kruse et al., 2007). The treatment system was designed using an area-adjusted removal rate of 10 $g/m^2/d$ (after Hedin et al., 1994). The design flow-rate was 88 L/s and design influent and effluent iron concentrations were 6 mg/L and 1 mg/L respectively. With a volume of 2409 m^3 and mean flow rate of 84 L/s, a previous tracer experiment conducted in the wetland (February 2007) indicated a peak residence time of 5.6 hours, 58% iron removal efficiency, and an area-adjusted removal rate of 3.6 $g/m^2/d$ (Kruse et al., 2007). The purpose of the investigation described here was twofold:

(1) Compare the results of the tracer test conducted in February 2007 with those of a more recent test conducted in July 2008, to assess the influence of maturing wetland vegetation on hydraulic performance
(2) Evaluate and compare the use of sodium chloride, sodium bromide, and sodium fluorescein as tracers for determining hydraulic residence time

In selecting a tracer to be used in the recent experiment, factors such as conservative behaviour, and sorption and degradation effects of the tracer, were primary considerations so that tracer movements in the wetland would represent the actual retention time within the system. Bromide is generally considered as one of the best hydrologic tracers due to its nonreactive (conservative) behaviour in most environments, its low background concentration levels, ease of measurement, low cost and relatively low toxicity (Bowman, 1984; Davis et al., 1980; Whitmer et al., 2000). Chloride is commonly used because it is simple to detect using conductivity probes, is highly soluble in water, relatively inexpensive and does not adsorb to negatively charged soil minerals (Wood and Dykes, 2002; Flury and Papritz, 1993). Fluorescein is used because it is visibly detected in low concentrations and considered resistant to adsorption on organic and inorganic materials (Smart and Laidlaw, 1977).

METHODS

Sampling and field measurements

In-situ field measurements were conducted using Myron 6P Ultrameter for pH, redox potential (Eh), electrical conductivity and temperature. Dissolved oxygen was measured using Sonde DO meter, while alkalinity was measured using a Hach digital titrator with 1.6 N sulphuric acid and Bromocresol Green Methyl-Red indicator powder to give results in units of mg/L as $CaCO_3$. Apart from the samples collected by the autosamplers, grab samples were also taken in the inlet and outlet point of the wetland to observe the background parameters within the treatment system. Unacidified samples were collected at both inlet and outlet for analysis of bromide and major anions in the water. Unfiltered samples, acidified with 1% by volume concentrated nitric acid, were taken at the inlet and outlet for total metal concentrations analyses. Dissolved metal concentrations were also determined by undertaking on site filtering through 0.45 μm cellulose nitrate membrane filters.

Flow rates throughout the experiment were ascertained using water levels data recorded by a Eijelkamp CTD Diver. The Diver semi-continuously measured the water and atmospheric pressure, temperature and conductivity at a point behind a 90º V-notch weir at the outlet of the wetland. The Diver was then adapted to a LoggerDataManager (LDM) software for transfer and reading of the measurements. The actual water levels were taken as the difference between the atmospheric pressure recorded by a BaroDiver and the levels recorded by the CTD Diver. The equation used for determination of flow from the V-notch weir is as follows (Shaw1994):

$$Q = K \tan(\theta/2)\, H^{2.5}$$

where Q is in units of m^3/s, H (in metres) is the height of water above the apex of the 'V', θ is the angle of the 'V', and K is a coefficient typically equal to ~ 0.6.

Simultaneous sodium bromide and Na-fluorescein injection

The simultaneous injection of NaBr and Na-flourecsein was initiated on 29/07/2008 at 13:30 pm. The amounts of tracer added were initially designed so that the peak tracer concentrations could be detected even if complete mixing with the mine water occurred (Kruse et al., 2007). Based on the assumptions that 1 mg/L of bromide (Kruse et al., 2007) and 50μg/L of Na-fluorescein (Wolkersdorfer et al., 2005) peaks could be expected during the tracer test, a mass of 3.102 kg of bromide and 120.45 g of Na-fluorescein had been calculated for the tracer injection. The 3.102 kg of NaBr (2.41 kg of bromide) were dissolved in 17 litres of water in the field prior to injection. The 120.45 g of NaFluorescein were dissolved in 2 litres of deionised water in the laboratory to facilitate the dye mixing with the mine water.

After injection of the tracers, inlet samples were subsequently collected at hourly intervals for 24 hours duration and outlet samples at every 20 minute intervals for 16 hours. The basis for the assigned interval times was the previous calculated time to fill of the wetland of 6.9 hours (Kruse et al., 2007). The samples were automatically collected by Aquamatic Auto Cell P2 Autosamplers equipped with 24 x 1L HDPE bottlers. Both the inlet and outlet autosamplers were logged at the same time at 13:30 pm on 29/07/2008 however, the sample sequence terminated at 13:30 pm on 30/07/2008 for inlet autosampler and at 05:10 am on 30/07/2008 for outlet autosampler. Approximately 250 mL of samples were collected in each of the HDPE bottler to be taken back to laboratory for analysis of bromide, anions and selected cations. In the laboratory, all samples from each bottler were transferred into two 125 mL polypropylene bottles (one acidified with 1% by volume concentrated nitric acid and one non-acidified) for subsequent analysis of bromide, anions and cations.

The analyses of bromide and major anions (Cl^- and SO_4^{2-}) were conducted in the laboratory using a calibrated Dionex IC 25 Ion Chromatography. The acidified sample was analysed for cations (Ca, Mg, Na, K, Fe, Mn, Al, Zn and Si) using a Varian Vista MPX ICP-OES. Fluorescein was continuously measured in the field using a Seapoint fluorimeter which was set up to take the concentration readings at 5 minute intervals. The fluorimeter commenced logging at 12:44 pm on 29/07/08 and lasted for 25.5 hours before being taken back to the laboratory for download onto a Dataron Data Bank data logger for data transfer.

Sodium chloride injection

In addition to NaBr and Na-fluorescein, NaCl was also used in the tracer test for comparison of the residence time of the wetland. The injection of NaCl was conducted a day after the simultaneous tracer injection to avoid possible interference on conductivity due to other salt addition (i.e. bromide). When conducting multiple tracing tests, it can be advantageous to plan delayed injections of different tracers with respect to the sampling times and the possible overlapping of the tracer reappearance (Kass, 1998). 20 kg of NaCl was injected and conductivity was recorded for 1 week to ensure all NaCl was carried through the system. The conductivity was measured using a Eijelkamp CTD Diver. Background conductivity was measured a day before NaCl injection at the inlet to the system, and during the tracer test, for comparison with concentrations recorded by the CTD Diver at the outlet.

RESULTS AND DISCUSSION

Bromide

During the test, the flow rate varied between 55.66-68.42 L/s with a mean flow of 61.182 L/s. Based on the mean flow and the volume of the system, the theoretical residence time was approximately 9.33 hours; greater than the previous approximation of retention time of 6.9 hours (Kruse et al., 2007) simply due to the higher flow rate (84 L/s) during the previous test. The breakthrough curve of bromide and flow rates for 16 hours duration is shown in Figure 1. Bromide was first detected after 3.95 hours of NaBr injection, and the breakthrough curve shows that bromide peaks at approximately 10 hours (at concentration of 1.014 mg/L), a slightly longer retention than the calculated residence time (i.e. time to fill). Residence time must be greater or equal to the reaction time needed to achieve desired effluent concentration in order to achieve effective treatment within a wetland (Kadlec and Knight, 1996). Obviously, this indicated a more hydraulically efficient performance of the treatment wetland since it last measured within a year ago. Factors attributable to the longer retention time were principally the lower flow-rate during the second tracer test, but also likely due to the establishment of vegetation n in the system and the absence of significant preferential flows which were prevalent in the early test soon after site commissioning.

The recovery of bromide over the 16 hour test was 64.83% (1.56 of 2.41 kg of bromide). The incomplete recovery of tracer is concluded to be due to the fact that bromide concentration was still decreasing when the test finished. Thus, a significant mass of bromide was still held within the wetland.

Na-fluorescein

A 120 g of Na-fluorescein was injected into the wetland which was then distributed by the 3 inlet channels that were designed to uniformly spread the incoming water through the wetland. However, field observations showed that tracer dispersal was biased towards one of the three inlet channels suggesting some preferential flow in the distribution structure The concentration of the tracer was measured by the fluorimeter which was logged to start capturing the concentration readings for a 24 hour duration. The flow during the NaFluorescein tracer test was in the range of 55.24-68.42 L/s, with a mean flow rate of 61.232 L/s.

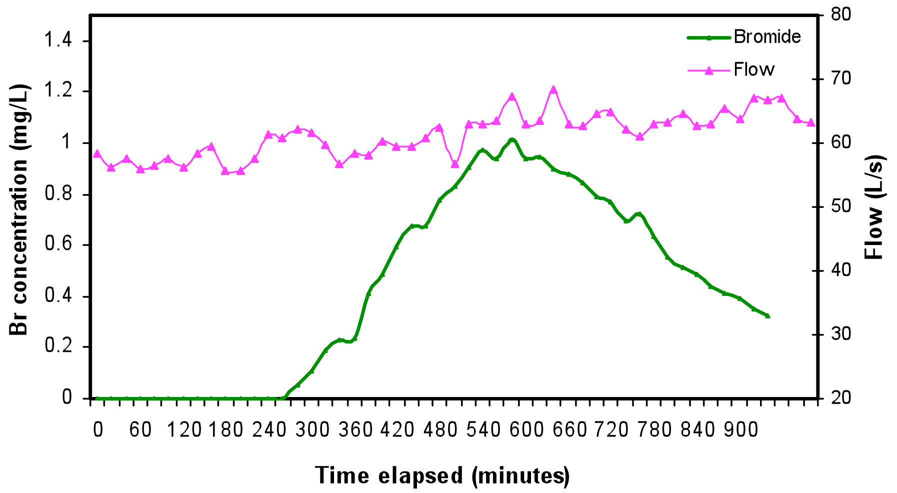

Figure 1. Bromide breakthrough curve of Lambley wetland for 16 hours duration.

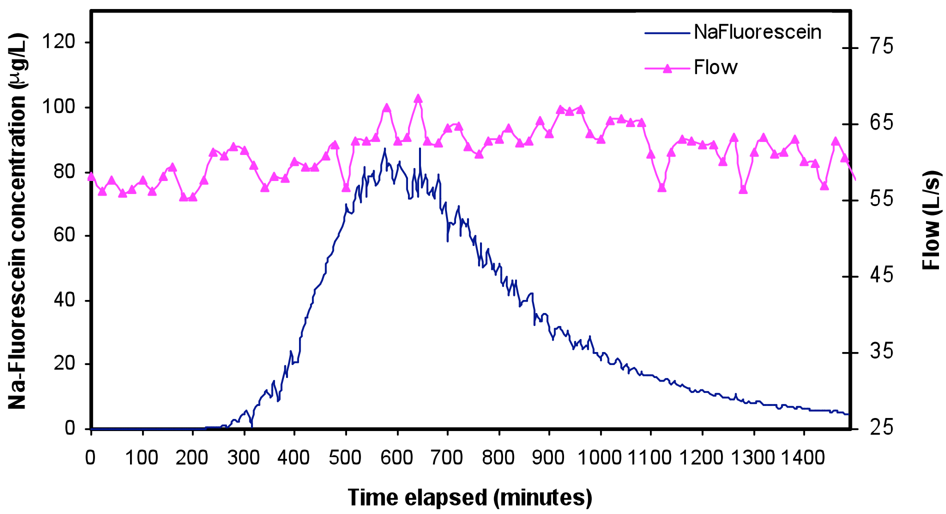

Figure 2. Na-fluorescein breakthrough curve for the Lambley wetland.

The breakthrough curve of the Na-fluorescein obtained from the experiment (Figure 2) can be considered as an ideal tracer plot: the leading edge (the point at which the tracer starts to rise over the background level (Kilpatrick and Wilson, 1989)) is observed 3 hours after injection and declines to 4.9 µg/L, less than 10% of the tracer peak (87.25 µg/L).

The first Na-fluorescein detection in the outlet point was observed 3.6 hours after the injection of tracer. As shown in Figure 2 the peak concentration was observed after 10.7 hours, which corresponds to the retention time determined by the bromide test (Figure 1). Again, this is indicative of an improvement in hydraulic efficiency within the wetland compared to the previous tracer tests undertaken by Kruse et al. (2007). The retention time was also greater than the theoretical residence time of the wetland which is 9.33 hours.

Of the total amount of Na-fluorescein injected, a mass recovery of 119.97% was obtained in the wetland. This unusual tracer recovery is principally thought to be a function of potential inaccuracies in flow rate measurements. The very noisy stage and flow records appear to be a feature of turbulence in the stilling well where the CTD-Diver was located and may impart large error (>10%) margins on derived flows. Spot measurements of flow rate (e.g. bucket/stopwatch or impeller measurements) would improve flow records in future studies, as would a longer approach channel to the V-notch weir, which would minimise turbulence. Furthermore, the presence of background fluorescence (suspended sediment and / or natural fluorescence background) may also cause the tracer recovery to be apparently in excess of 100% ((Smart and Laidlaw, 1977).

The bromide and Na-fluorescein test indicate very similar tracer peaks: 9.95 and 10.7 hours respectively (Figures 2 and 3). It can also be clearly seen from the plots that the first detection of the tracers occurs at almost the same time interval: 3.95 and 3.62 hours respectively. It is therefore concluded that both tracers behave similarly in terms of their dispersive characteristics within the wetland. Furthermore, the shape of the tracer curves indicate that water is well-mixed within the system, and improved by the establishment of vegetation which limits the potential for hydraulic chort-circuiting.

Sodium Chloride

Figure 3 indicates that an increase in conductivity could be observed some 10 hours after NaCl injection, albeit the background conductivity is noisier than either bromide or Na fluorescein concentrations. Because of the ease of recording conductivity, readings were logged at 5 minute intervals for a period of 1 week. Of the 1 week conductivity measurement, the possible portion of conductivity plot that corresponds to the NaCl injection is as shown in Figure 3 (small diagram). The inset diagram in Figure 3 in particular illustrates the elevation of conductivity in the effluent water during the tracer test. The peak detection of conductivity was observed after 10 hours of NaCl injection. Therefore the NaCl tracer test indicated an approximately identical time of retention (10 hours) as the bromide and the Na-fluorescein tests: 9.95 and 10.7 hours respectively.

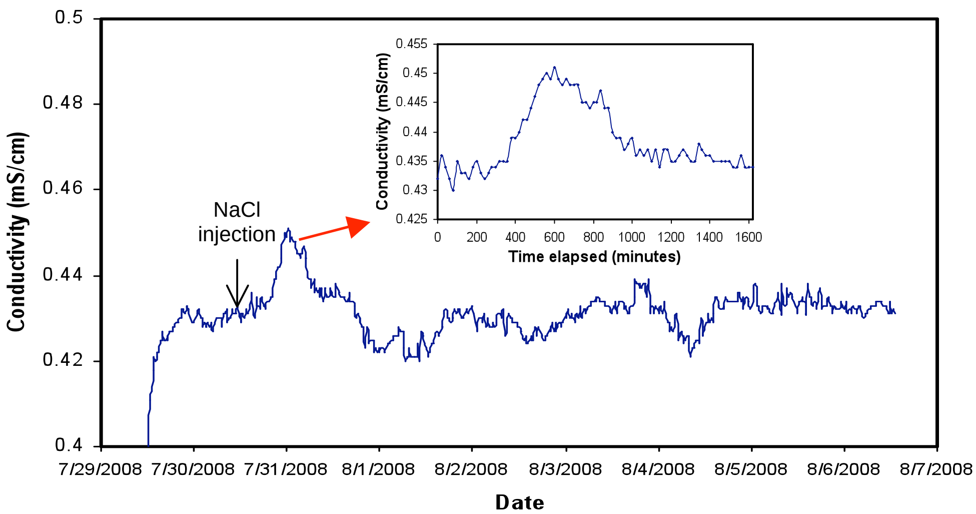

Figure 3. Conductivity plot of Lambley wetland outlet from 12:00 pm 29/07/2008 to 13:00 pm 06/08/2008. Sodium chloride injection was conducted at 14:15 pm on 30/07/2008. Peak conductivity was detected after 10 hours of injection (insert plot).

Comparison of tracer performance

Because flow-rate remained consistent throughout, the performance of the three tracers used can be compared directly even though the NaCl tracer was not synchronous with the other two tracers. Summary performance data for the three tracers are shown in Table 1, while Figure 4 shows a normalised tracer mass plot to aid comparative interpretation of tracer performance (i.e. tracer concentrations, flow rates and duration of tests).

Table 1. Comparison of different tracer performance in Lambley wetland treatment system

Tracer	Bromide	NaFluorescein	NaCl
Mean Flow rate (L/s)	61.182	61.232	62.283
Nominal retention time (hr)	9.33[a]	9.32[a]	9.16[a]
Duration of tracer injection (hr)	16	24	168
First detection (hr)	3.95	3.62	-
Peak residence time (hr)	9.95	10.7	10
Mass recovered at peak time (%)[b]	45.31	44.07	34.56
50% breakthrough (hr)[c]	10	11.17	11.08
10% breakthrough (hr)[c]	6.67	7.67	7.68
90% breakthrough (hr)[c]	13.67	17.67	17.67
Tracer mass added	3.102 kg	120.45 g	20 kg
Tracer recovery (%)[d]	64.83	119.97	70.51

[a] Calculated by dividing the nominal wetland volume of the system by the mean flow of 61.182 L/s, 61.232 L/s and 62.283 L/s for bromide, Na-fluorescein and NaCl, respectively (Kadlec and Knight, 2009)

[b] Calculated as the percentage of mass recovered (area underneath the curve) at peak time

[c] Time required for 10%, 50% and 90% breakthrough of the total mass recovered (Lin et al., 2003) (i.e. 10% breakthrough of recovered 64.83% bromide mass is at 6.67 hour)

[d] Calculated as the percentage of total mass recovered from the amount of tracer added

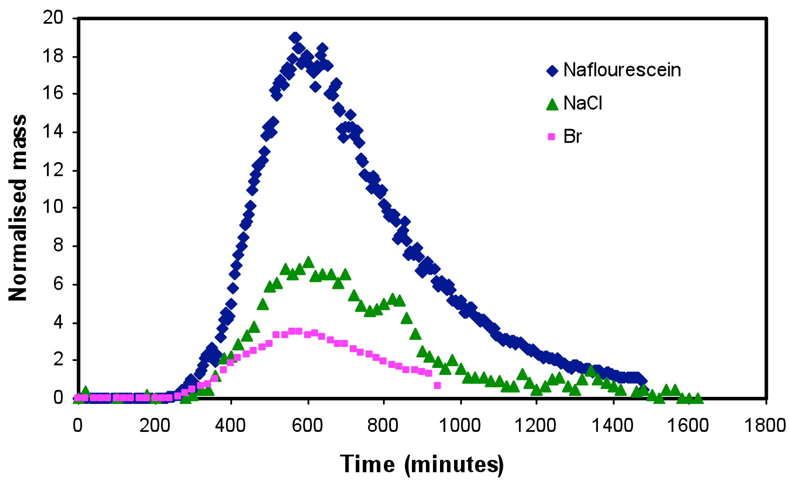

Figure 4. Normalised curve for bromide, NaFluorescein and NaCl to indicate the performance of the different tracers. All tracers show an essentially identical time of retention.

The bromide, Na-fluorescein and NaCl curve are normalised (Figure 4) so that areas underneath the curves represent fractional mass recovery (Lin et al., 2003). The normalisation curve produced was essentially to remove the effects of different tracer concentration and flow rate so that comparison could be made upon the tracer recoveries and the breakthrough times. The recoveries of tracer were obtained by the zeroth moment analysis to yield the total mass of tracer that has been recovered at the exit of the wetland (Martinez and Wise, 2003; Werner and Kadlec, 1996). As presented in Table 1, bromide indicates 64.83% of mass recovery, NaCl of 70.51% and Na-fluorescein of 119.97%. The anomolous recovery of Na-fluorescein was probably due to the flow rate monitoring issues described earlier. The lower recovery of bromide is thought simply to be due to the test being terminated before bromide concentrations had returned to background, though sorption onto wetland sediment and uptake by wetland vegetation cannot be ruled out (Lin et al., 2003).

Approximately identical results were found for the peak residence time of the three tracers which is 9.95, 10 and 10.7 hours for bromide, Na-fluorescein and NaCl, respectively. The 10% breakthrough of the recovered bromide occurred at 6.67 hours as compared to 7.67 and 7.68 hours for Na-fluorescein and NaCl, respectively. It can be seen that there was a slight difference of the bromide breakthrough to that of Na-fluorescein and NaCl, which could be attributed to a shorter experimental duration of bromide since the measurement was terminated after 16 hours of injection. Similar results were found for 90% breakthrough in which bromide indicated 13.67 hours of residence time, whereas Na-fluorescein and NaCl occurred at 17.67 hours both. Again, ahd the tail of the bromide trace been captured, the 90% breakthrough time would have been longer, and therefore closer to those of the NaCL and Na-fluorescein.

Iron Removal

During the test, the influent iron concentration was in the range of 4.50-7.53 mg/L (mean of 6.81 mg/L). The average of effluent iron concentration was 1.01 mg/L, with a range of 0.85-1.31 mg/L. The system was initially designed to treat an influent iron concentration of 6 mg/L to achieve an effluent concentration of 1 mg/L, and to receive 88 L/s of flow (Kruse et al., 2007). Based on the average influent and effluent iron concentrations, iron removal efficiency was calculated to be of 85.2%. This was considerably higher removal of iron in the treatment system compared to the 57.67% reported by Kruse et al. (2007). This significant increase in iron removal efficiency was likely due to increased residence time and comparatively higher influent iron concentration (6.810 mg/L compared to 3.841 mg/L previously) which reflects the first-order kinetics that the removal of iron is concentration-dependent (Tarutis et al., 1999). Nevertheless, this did not directly give the higher first-order removal rate as the recent experiment indicated only 0.63 m/d compared to 1.92 m/d during the previous tracer test. The improved removal of iron may also be ascribable to the well–established reeds within the wetland which in turn improve the settlement of iron solids and provide a relatively larger specific surface area for precipitation / adsorption of iron onto the plant materials.

The area-adjusted removal rate of iron during the 2008 tracer tests was 6.99 $g/m^2/d$ based on the wetland area of 4388 m^2 (Kruse et al., 2007) and mean flow rate of 61.182 L/s during the test. The performance of the treatment wetland was in fact lower than the design removal rate of 10 $g/m^2/d$ by 30%. However, it performed better than the expected removal (6.02 $g/m^2/d$) under current condition of the wetland. Overall, the performance of the Lambley wetland is as summarised in Table 2.

Table 2. Performance data of Lambley wetland from 29/07-6/08/2008 in comparison to data obtained on 28/02-1/03/2007.

Performance parameters	28/02-1/03/2007	29/07-6/8/2008
Mean flow rate (L/s)[b]	84.830	61.232
Velocity (m/hr)[c]	44.26	31.95
Hydraulic loading rate (m/hr)[d]	0.07	0.05
Influent Fe concentration (mg/L)	3.841	6.810
Effluent Fe concentration (mg/L)	1.626	1.014
Mean iron removal efficiency (%)[e]	57.67	85.17
Area adjusted removal ($g/m^2/d$)[f]	3.69	6.99
First-order removal (m/d)[g]	1.92	0.63

[b] Flow rate reported in the table is the mean flow of the 1 week data

[c] Calculated as Q/A; where A=open area perpendicular to flow (Kadlec and Wallace, 2009)

[d] Calculated as Q/A; where A=wetland area (wetted land area) (Kadlec and Wallace, 2009)

[e] Calculated as Cin-Cout/Cin x 100; where C=Fe concentration (Tarutis et al., 1999)

[f] Calculated as Q[Cin-Ctargeted]/A; where Q=flow rate , A=treatment area (Hedin et al., 1994; Tarutis et al., 1999)

[g] Calculated as Q/A ln[Cin/Cout]; where Q=flow rate , A= treatment area (Tarutis et al., 1999)

CONCLUSIONS

In general, the three tracers gave essentially identical results in terms of hydraulic residence time and considerably high tracer mass recoveries. The peak residence times were between 9.95 and 10.7 hours, which were greater than the theoretical residence time of 9.33, 9.32 and 9.16 hours for bromide, Na-fluorescein and NaCl, respectively. The recoveries of the tracers were between 64.83 and 119.97%.

When comparing the performance of the three tracers, it is difficult to simply identify the most reliable tracer due to their nearly equal results. Bromide, although only 64.83% of the tracer mass added was recovered, had the advantage that bromide background within the wetland was negligible and, therefore, significantly facilitated the interpretation of the tracer recovery. The cause of the greater than 100% mass recovery of Na-fluorescein is unknown and the likely contributor was probably the presence of very high concentrations of Na-fluorescein above the expected peak for quite a long duration. This could lead to inaccurate interpretation of the residence time determined based on the recovered tracer. For the NaCl tracer, the fluctuation in the baseline conductivity measured throughout the test affected the calculated recovery of the tracer. It was found that during the NaCl injection that the conductivity dropped significantly below the baseline making it difficult to identify the exact conductivity portion attributable to the salt addition and what portion was due to the increase in conductivity caused by other ions present in the water. Despite the higher tracer recoveries of Na-fluorescein and NaCl, bromide was still considered the most conservative tracer in water, although due to the consistent performance of the tracers, the selection of tracer should be made based on site specific restrictions.

Apart from the evaluation of the three tracer performances, it was found that there was a connection between the residence time and the rate of iron removal. The higher residence time (~ 10 hours compared to previous tracer test of 5.6 hours) had resulted in a significantly higher removal of iron (85.18% efficiency compared to 57.67% previously and 6.99 $g/m^2/d$ compared to 3.69 $g/m^2/d$ previously) within the treatment system. The increase is probably attributable to a lower flow rate and maturation of the wetland. Despite the better removal observed during the experiment, the wetland is still functioning at a rate below (30%) than the design 10 g/m^2/d of iron removal.

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