Int. J. Environ. Sci. Technol. (2014) 11:1189–1196
DOI 10.1007/s13762-013-0336-7

ORIGINAL PAPER

Monitoring accelerated clogging of a model horizontal sub-surface flow constructed wetland using magnetic resonance transverse relaxation times

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Received: 19 June 2012 / Revised: 23 April 2013 / Accepted: 6 May 2013 / Published online: 29 May 2013

Abstract Horizontal sub-surface flow wetlands are essentially a bed of porous material in which suitable plants are grown to facilitate the removal of organic matter and particulates from wastewater. The aim of this study is to assess the reliability and accuracy of magnetic resonance transverse relaxation time for monitoring clogging development in a constructed wetland. In this study, three different horizontal sub-surface flow constructed wetland models have been produced using tubes packed with different sizes of glass beads with diameter 3, 8 and 14 mm. Accelerated clogging has been achieved by pumping sludge extracted from a real clogged wetland through the bead pack. A desktop MRI tomography system has been used to monitor the transverse relaxation rate as a function of position along the tube and hydraulic conductivity. To corroborate the clogging with magnetic resonance measurements, the head loss was monitored to determine the hydraulic conductivity. Using a bi-exponential fit to the spin echo train data, the slow relaxation rate contribution shows good correlation with the changing hydraulic conductivity. Both fast and slow contributions map well to the expected clog patterns for a constructed wetland. We have demonstrated that there is a linear correlation between the hydraulic conductivity and both parameters of a bi-exponential fit to R_2^{eff}, but particularly for the case of the short T_2 component.

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Keywords T_2 · MR relaxation time · Constructed wetland · Clogging

Introduction

Constructed wetlands are an important tool for sustainable development both for their role in wastewater treatment and as support for biodiversity and conservation (Kadlec and Wallace 2009). Horizontal sub-surface flow (HSSF) wetlands are essentially a bed of porous material, usually gravel, in which suitable plants, such as the common reed or iris, are grown and through which contaminated water slowly flows enabling the removal of organic matter and particulates (Vymazal 2010). They are a highly effective way of removing pollutants and increasing the quality of water before it is released back into a water course. There are numerous differing recommendations for the optimum size of gravel to use in a HSSF constructed wetland (Kadlec and Wallace 2009), however, most use gravel in the size range 3–15 mm for at least the top layer.

Excessive clogging can have a severe impact on the performance, and subsequently, a detrimental effect on the surrounding environment if flooding occurs (Knowles et al. 2011). Once fully clogged, a reed bed must be refurbished if it is to continue to be used, although if detection and assessment of the extent of this clogging could be achieved, early localized replacement of gravel substrates could be performed. Alternatively, an active intervention and management strategy could be adopted, such as excavation and replacement or excavation and washing (Nivala et al. 2012). The current state of the art for the assessment of clogging in constructed wetlands is in situ permeameter-based hydraulic conductivity (HC) tests (Pedescoll et al. 2009). These represent a significant major advance in comparison with initial estimations of clogging which were based on water level surveys (Kadlec and Wallace 2009). However, the in situ tests (e.g. hydraulic conductivity, clog matter characterization and hydrodynamic visualization) are time consuming, limited to providing a snapshot in time and subjected to scrutiny over whether or not the media disturbance caused by the method renders results unrepresentative (Knowles et al. 2010).

Magnetic resonance (MR) is a technique which has found application in sub-surface logging, particularly as a tool for oil industry site viability surveys (Coates et al. 1999). More recently, low-cost MR sensors have been used to study the drying process of cement (Cano-Barrita et al. 2009). Small sensors were developed and embedded in Portland cement mixture as they dried causing a decrease in MR signal intensity. Magnetic resonance imaging (MRI) has been demonstrated to be a quantitative technique to image the porosity of packed gravel (Kleinhans et al. 2008). In addition to sensitivity to the water content of a sample, MR can be used to determine the properties of the water, such as its local association. A system comprising of both free and bound water will exhibit a continuous superimposition of MR relaxation times which may be analysed using a Laplace inversion (Moody and Xia 2004). By approximating this relaxation time distribution to a bi-exponential decay, a good indicator may be obtained for the ratio of the two contributions of water (Jaeger et al. 2008). Since it is the ratio of free to associated (bound) interstitial water that will determine the HC through clog matter (Suliman et al. 2006), bi-exponential fits to data may be used to characterize clogging in constructed wetlands. Whilst tracer tests can deduce the relative active and stagnant wetland volumes (Knowles et al. 2010), this is a bulk parameter and unlike MR, it does not provide spatially localized data. The ability to extract two superimposed relaxation parameters using a small permanent magnet-based MR probe should allow in situ HC measurements at various locations in a saturated gravel bed. A recent study (Morris et al. 2011) demonstrated that it is possible to monitor the extent of clogging in a constructed wetland model consisting of a coral sand packed tube by using an MR sensor. The quantity of restricted and unrestricted water was determined using bi-exponential fits to the longitudinal relaxation time (T_1) which was shown to correlate well with the hydraulic conductivity. The aim of this study is to determine the sensitivity of the NMR parameter T_2 (transverse relaxation time) to accelerated clogging by performing experiments with sterilized sludge to achieve accelerated clogging in a tube packed with glass beads. In this article, we report a set of experiments using the inverse of the effective transverse relaxation time R_2^{eff} to characterize HC in three model HSSF wetlands made using mono-disperse glass beads of three different sizes undergoing accelerated clogging with material extracted from a clogged wetland. A comparison of the average values of the two fitted R_2^{eff} values is presented as a function of HC. Spatial changes of R_2^{eff} along the model system as a function of HC are also reported. These results suggest that in future, a field deployable NMR probe capable of providing reliable bulk relaxation measurements could be used in situ that would enable a better understanding and insight into the clogging occurring in constructed wetlands.

This work was carried out in 2011 in Nottingham, UK.

Materials and methods

Laboratory models of HSSF constructed wetlands were made using acrylic tubes of 1 m in length, packed with mono-disperse glass beads. The bole hole diameter of the MRI tomography system which was used in this study is 40 mm, and therefore, the tubes having 38 mm outer diameter and 32 mm inner diameter were used. Three different tubes were used and each was filled with glass beads of one specific size from 3, 8 or 14 mm; this covers most of the officially recommended range at least for the top layer of the constructed wetlands (Kadlec and Wallace 2009). For the measurement of hydraulic conductivity, two take-off manometer tubes, each 1 m long, one at the inlet and one at the outlet, were used to measure head loss. The acrylic tube passed through a 20 MHz ACT (Aachen) desktop MRI tomography system (Danieli et al. 2009) which provides the bulk R_2^{eff} values for a 40 mm long by 32 mm diameter volume in the middle of the scanner. T_2^{eff} was measured using the Carr–Purcell–Meiboom–Gill sequence (Meiboom and Gill 1958) with 4-ms echo time, 512 echoes, 2.1-s repetition time and 6 repeats. This produced a train of spin echoes, to the envelope of which a bi-exponential decay curve was fitted. The standard deviation of the data to the curve was computed for each measurement, and the fitting procedure repeated 500 times on an artificially generated data set with identical bi-exponential decay parameters with numerically generated thermal noise allowing the error to be estimated. By further calculating the standard deviation of the 500 fitted values of short and long T_2 values, their error was obtained. The tube was moved through the magnet bore, to make measurements at six equally spaced points between inlet and outlet. Figure 1 shows a schematic diagram of the arrangement.

The model clogging material was provided by ARM Ltd. (Rugeley, UK) from a tertiary treatment wetland which was used to treat domestic wastewater. The sample

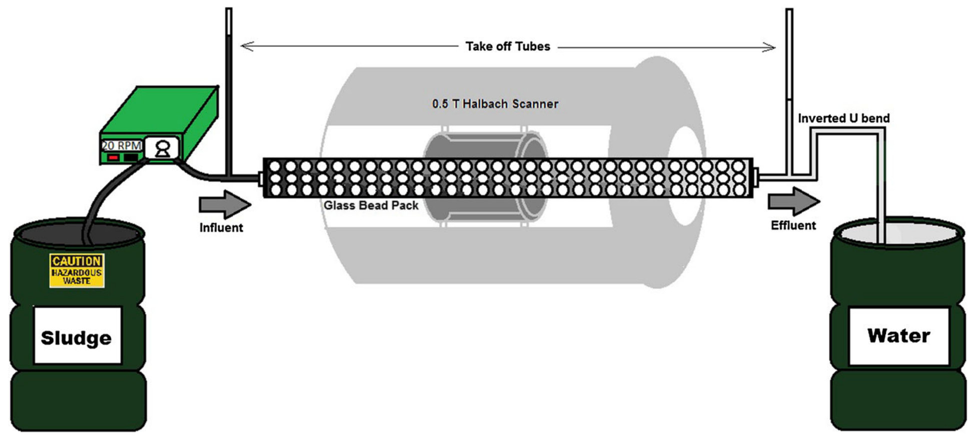

Fig. 1 The *schematic diagram* shows the model HSSF wetland running through the bore of the MRI tomography system with the take-off tubes to measure the pressure difference between *inlet* and *outlet*

consists of 20 kg of gravel bed from the inlet zone of a constructed wetland which was clogged and was undergoing refurbishment. The gravel was washed carefully to isolate the sludge. A minimal amount of water was used to ensure the concentration remained high. The gravel was removed, and only the clog matter was extracted and used. Once isolated from the gravel, the concentration of solids in the sludge was made to 1/5. Due to microbial activity in constructed wetlands, biofilms are formed which contribute to the clogging. However, these biofilms take months to develop (Ragusa et al. 2004), and it was not part of this investigation; therefore, to perform the accelerated clogging experiments, we sterilized the sludge sample. The sludge was sterilized to kill bacterial growth by stirring for one day and then standing for 3 days with an immersed ultraviolet light before performing the experiments. In order to determine the sensitivity of NMR parameter T_2 to the accelerated clogging, sludge sample with very high concentration of solid particles was used. To avoid the particulate settling, we keep stirring the sample prior to use and throughout the experiments and to avoid the particulate settling occurring inside the tubing high flow rate was used. The sludge was pumped into the bead packed tube using a Watson Marlow peristaltic pump set at a flow rate of 130 ml/min. The flow rate was checked at the outlet and remained constant throughout all the experiments. An inverted U bend was also attached at the outlet after the manometer tube as shown in Fig. 1, to ensure that the water remained inside the tube whilst the pump was turned off. Initially, six equally spaced locations were marked along the length of the tube, and the tubes were filled with water. Whilst the water was being pumped through the tube, the head loss was measured. The measured head loss was then used to calculate the hydraulic conductivity using the equation

$$K = QL/Ah$$

where K is the hydraulic conductivity, Q is the flow rate, A is the cross-sectional area of the tube and h is the head loss. To prevent flow artefacts, the pump was then switched off, and the relaxation time measurements at the marked six locations on the tube were made. The sludge was then pumped into the tube for 3 min, and the head loss was measured again before the pump was switched off which was followed by the relaxation time measurements at the exact same six locations; this process was repeated until the maximum height was reached in the inlet manometer tube for the 3-mm bead tube, and the height where the level of the water at the inlet manometer tube reaches equilibrium for 8- and 14-mm bead tube. After a set of measurements were made, the tubes were emptied and the glass beads cleaned and repacked to undertake two further repeats for each of the bead diameters.

Figure 2 shows typical MR data for a set of echo amplitudes as a function of time for a Carr–Purcell–Meiboom–Gill sequence with one plot for the clogged and one for the unclogged tube. The echo amplitudes were fitted using the sum of two exponentials plus a constant

$$M = A\exp\left(-t/T_2^{\mathrm{eff}(S)}\right) + B\exp\left(-t/T_2^{\mathrm{eff}(L)}\right) + C$$

where A and B are the amplitudes of the two exponentials, $T_2^{\mathrm{eff}(S)}$ and $T_2^{\mathrm{eff}(L)}$ are the short and long exponential time constants and C is a constant that represents the thermal noise. Whilst performing the fitting in MATLAB, thermal noise parameter was also fitted and then the fitted parameter was averaged to get the average thermal noise which was subtracted from the data before performing a second pass fitting of the exponentials providing an enhanced signal to noise. Data are presented in the form of relaxation

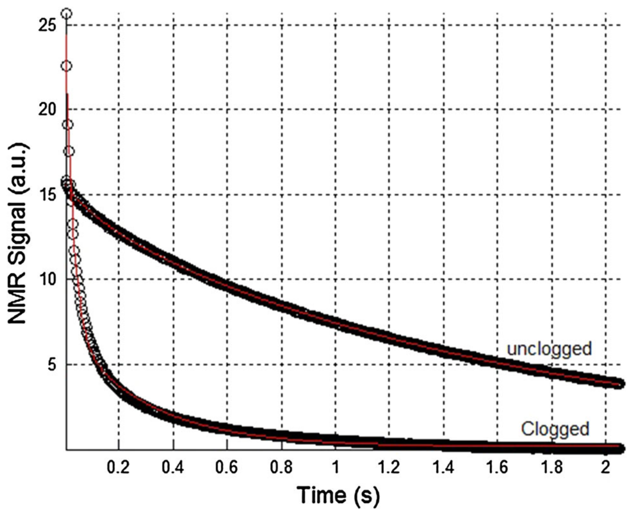

Fig. 2 The CPMG echo sequence produces a set of 512 echo's whose integrals are shown as a function of time (*circles*); this is fitted with a bi-exponential decay shown as the *red line*

rates $R_2^{\mathrm{eff}(S)} = 1/T_2^{\mathrm{eff}(S)}$ that will predominantly originate from the bound water and $R_2^{\mathrm{eff}(L)} = 1/T_2^{\mathrm{eff}(L)}$ which will predominantly originate from the free water.

Results and discussion

Figure 3 shows typical data for the hydraulic conductivity, derived from the head loss measurements, as a function of clogging time (the time for which the sludge was pumped through the tube); in the early stages of clogging, the hydraulic conductivity is higher with larger bead size (38,813 m/day for 14 mm glass beads whilst 4,312 m/day for 3 mm glass beads), as would be expected with any filter with monodisperse pore size. The 3-mm beads are also seen to clog faster (15 min), whilst the 14-mm beads clog the slowest (30 min) as would be expected given the reduced inter-pore spacing. After 15 min of accelerated clogging, the values are significantly reduced with 3 mm reaching the value of 352 m/day and 14-mm beads reaching the value of 6,653 m/day.

In Fig. 4, we show the value of $R_2^{\mathrm{eff}(L)}$ as a function of HC for the 3-mm beads (blue diamonds), 8-mm (orange squares) beads and 14-mm beads (green triangles). At the highest HC values, irrespective of bead size, the relaxation rate is near that of bulk water, around 0.5 s^{-1}. Whilst there is a trend in the data towards a lower $R_2^{\mathrm{eff}(L)}$ at higher hydraulic conductivities, there is significant scatter in the data: whether there is free water or not in the system is irrelevant to the stage of clogging. However, in Fig. 5, we

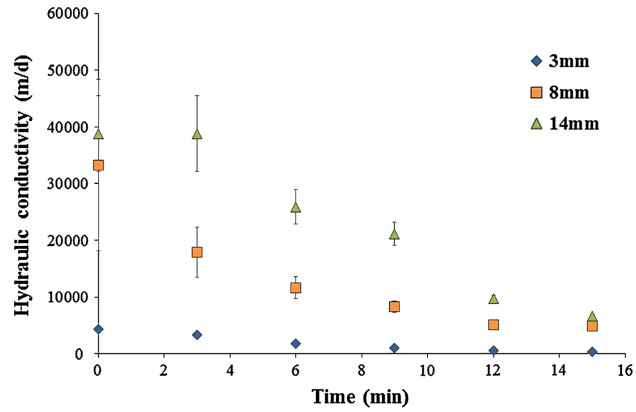

Fig. 3 The hydraulic conductivity as a function of clogging time for the 3-mm (*diamonds*), 8-mm (*squares*) and 14-mm (*triangles*) beads

show the same graph for $R_2^{\mathrm{eff}(S)}$ which is associated with bound water, and whilst there is some scatter for the 14-mm beads, the 3 and 8 mm give a linear trend up to a HC of 25,000 m/day. The coefficient of determination was calculated for 14-, 8- and 3-mm glass beads in MS-EXCEL and was found to be 0.8878, 0.9403 and 0.8811, respectively. This appears to be in agreement with the arguments of Suliman et al. (2006) who suggested that it is the ratio of free to bound interstitial water that will determine the hydraulic conductivity, although it is possible that in a functional constructed wetland, bound water will also include biofilm growth (not accounted for in our study) which is responsible for a large contribution to overall clogging (Zhao et al. 2009). Therefore, biofilm growth should be considered in future studies in order to develop an understanding of effect of clogging caused by the presence of biofilms in constructed wetlands. A comparison with the data of Morris et al. (2011) shows that the rate of change of $R_2^{\mathrm{eff}(S)}$ with HC is 1.7 times greater for the 8-mm and 5.6 times greater for the 3-mm beads than the equivalent plot using the previously reported T_1 data. Whilst the trend line slopes, calculated using regression analysis, are approximately similar for the 8- and 14-mm beads (95 ± 5.8 and 107 ± 7.1 m^{-1}, respectively), those of the 3-mm bead data are three times larger (307 ± 28.2 m^{-1}), resulting in enhanced MR sensitivity to HC changes. In smaller pore sizes, the particulate clogging is likely to affect pores more homogeneously than in larger ones, in which gravity-driven settling is more prone to occur. In this latter case, changes in MR relaxation will still occur with clogging in a given volume, even though fairly high conductivity will be retained towards the top of the system. This possible explanation, which remains to be investigated by means of MRI resolved in the transverse

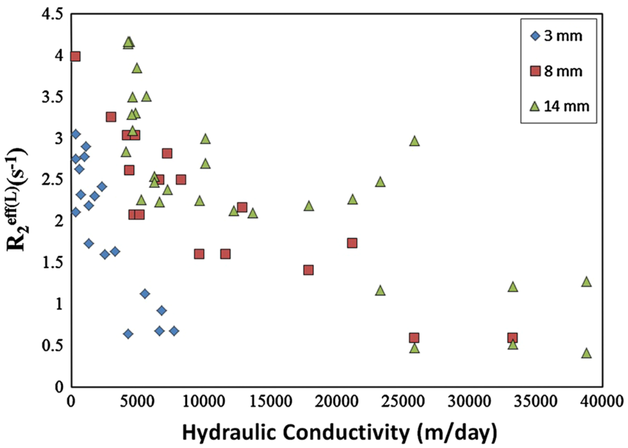

Fig. 4 The average value of the relaxation rate $R_2^{\mathrm{eff}(L)}$ along the tube is shown as a function of hydraulic conductivity for the 3-mm (*diamonds*), 8-mm (*squares*) and 14-mm (*triangles*) diameter beads. Data are included for the three repeats at each bead diameter

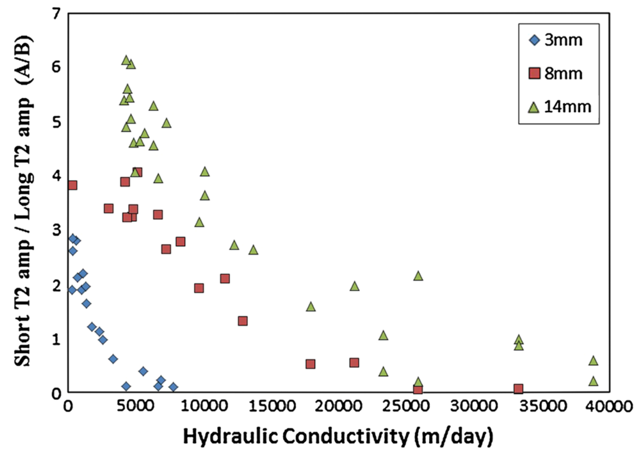

Fig. 6 The average value (along the tube) of the ratio of short-lived MR amplitude to long-lived MR amplitude is shown as a function of hydraulic conductivity for the 3-mm (*diamonds*), 8-mm (*squares*) and 14-mm (*triangles*) diameter beads. Data are included for the three repeats at each bead diameter

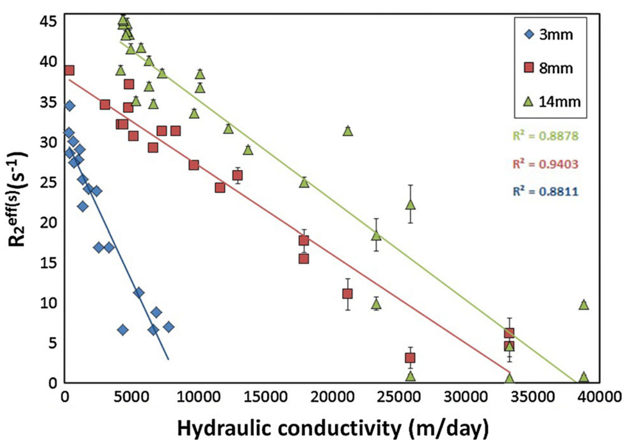

Fig. 5 The average value of the relaxation rate $R_2^{\mathrm{eff}(S)}$ along the tube is shown as a function of hydraulic conductivity for the 3-mm (*diamonds*), 8-mm (*squares*) and 14-mm (*triangles*) diameter beads. Data are included for the three repeats at each bead diameter

plane, is also further backed up by the fact that the three lines do not merge towards the same point for the highest values of HC: a given amount of bound water in a given volume clearly has a more severe effect on the smallest bead size system.

The fluid we drove in our system carries polydisperse particulates. This will result in a heterogeneous, random spatial distribution of the pores that will gradually undergo particulate clogging during our experiments, and a specific value of $R_2^{\mathrm{eff}(S)}$, corresponding to a specific value of particulate clogging, will not always correspond to exactly the same value of local HC. This is the most likely reason for the scatter of our data and also for the larger scatter in the larger bead size.

In Fig. 6, we also show the average (along the tube) of the ratio of the amplitudes of the short-lived MR signal (A) to the long-lived one (B), for all bead sizes, as a function of the HC; the correlation here is nonlinear. This result further substantiates the ability of MR to quantitatively probe the porous medium's hydraulic conductivity by means of estimating the ratio of bound water to free water. This quantitation is perhaps less promising in future applications, firstly because it is nonlinear, secondly because the amplitude of any measured MR signal is affected by multiple parameters such as probe matching/tuning, RF acquisition chain stability, temperature, B_1 field homogeneity. As the quantity displayed is a ratio of amplitudes coming from the same train of spin echoes, it should, however, remain fairly insensitive to these effects.

Whilst $R_2^{\mathrm{eff}(S)}$ gives a reliable measure of the HC, this is not the only consideration for the health of a HSSF wetland, and knowledge of the positions of the bound interstitial water will contribute to a better understanding of where clogging is occurring. In Figs. 7 and 8, we show the values of relaxation rate $R_2^{\mathrm{eff}(L)}$ and $R_2^{\mathrm{eff}(S)}$, respectively, as the variation in colour (right-hand scale bar) at the six different positions along the tube and as a function of the changing HC for the 3-mm (top), 8-mm (middle) and 14-mm (bottom) beads. For the highest HC, the values at both the inlet and the outlet are equivalent since the system contains only water. As the HC reduces, moving from the top to the bottom of the graph, the clogging can be seen to

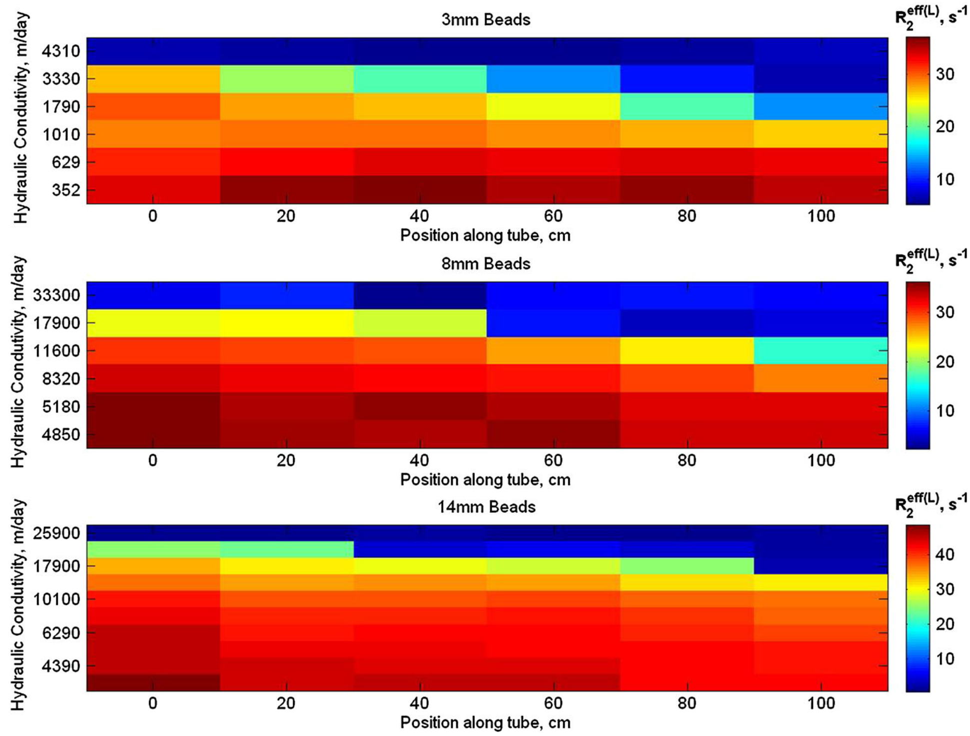

Fig. 7 The values of relaxation rate $R_2^{\mathrm{eff}(L)}$ are shown as the variation in colour (*right-hand scale bar*) at the six different positions along the tube and as a function of the changing hydraulic conductivity for the 3-mm (*top*), 8-mm (*middle*) and 14-mm (*bottom*) beads

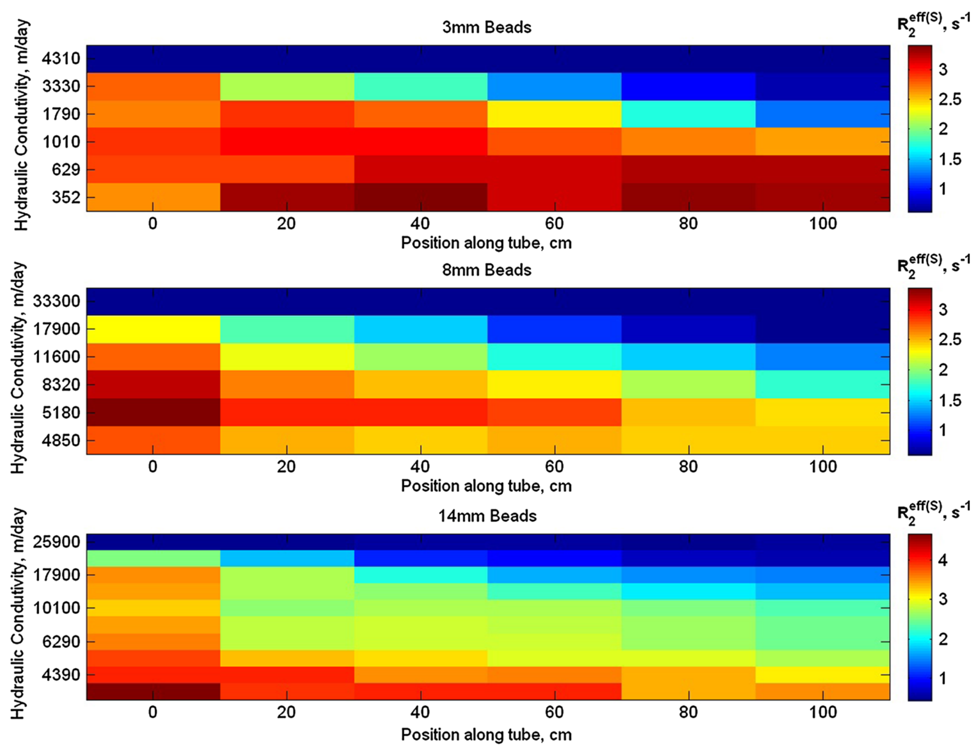

Fig. 8 The values of relaxation rate $R_2^{\mathrm{eff}(S)}$ are shown as the variation in colour (*right-hand scale bar*) at the six different positions along the tube and as a function of the changing hydraulic conductivity for the 3-mm (*top*), 8-mm (*middle*) and 14-mm (*bottom*) beads

change the relaxation rates much faster near the inlet which is consistent with previously published maps of clogging in HSSF constructed wetlands (Knowles et al. 2010). As the sludge used in this work had been sterilized, our experiments do not account for biofilm growth, which would result in an additional MR signal from this component in an active system (Neu et al. 2010). It is likely that this would give an even shorter relaxation time as previous studies have shown water associated with biofilms in bead packs to give T_2 values between 50 and 110 ms (Hoskins et al. 1999). The results from such an active system would require a tri-exponential fit to the CPMG data to account for the free, nonbiologically bound and biological bound water.

Conclusion

The aim of this study was to assess the reliability and accuracy of magnetic resonance transverse relaxation time for monitoring clogging development in a constructed wetland. We have demonstrated that there is a linear correlation between the hydraulic conductivity and both parameters of a bi-exponential fit to R_2^{eff}, but particularly for the case of the short T_2 component. Using the same fit, a strong, nonlinear correlation is also seen between the hydraulic conductivity and the ratio of amplitudes of the short-lived to the long-lived MR signals.

We have also shown a greater sensitivity to R_2^{eff} than was seen in previous publications using T_1 with an average value of 0.02 % signal change per m/day. The measurement of the MR relaxation time is absolute and does not necessitate calibration as in spin density assessments by means of signal strength. The absolute measurement of the hydraulic conductivity by means of MR does, however, require knowledge of the gravel bed mean pore size. Results also suggest that in future, a field deployable NMR probe capable of providing reliable bulk relaxation measurements could be used in situ that would enable a better understanding and insight into the clogging occurring in constructed wetlands; however, further study would be required to include the effect of biofilms. These results demonstrate that magnetic resonance relaxation measurements are a viable alternative to current technology for measuring the clog state of constructed wetland models and could be used to form part of a remediation system.

Acknowledgments The authors acknowledge ARM Ltd. (Rugeley UK) for providing material from a clogged constructed wetland. M. F. S. acknowledges Nottingham Trent University for bursary funding.

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