Mass transfer approach and the designing of horizontal subsurface flow constructed wetland systems treating waste stabilisation pond effluent

Anita M. Rugaika, Damian Kajunguri, Rob Van Deun, Bart Van der Bruggen and Karoli N. Njau

ABSTRACT

Pilot-scale constructed wetlands (CWs) that allowed wastewater to flow with high interstitial velocities in a controlled environment were used to evaluate the possibility of using mass transfer approach to design horizontal subsurface flow constructed wetlands (HSSF-CWs) treating waste stabilisation ponds (WSPs) effluent. Since CW design considers temperature which is irrelevant in tropics, mass transfer approach could improve the design. HSSF-CWs were operated in batch recycle mode as continuous stirred tank reactors (CSTR) at different interstitial velocities. The overall removal rate constants of chemical oxygen demand (COD) at various interstitial velocities were evaluated in mesocosms that received pretreated domestic wastewater. The mean overall removal rate constants were 0.43, 0.69, 0.74 and 0.73 d^{-1} corresponding to interstitial velocities of 15.43, 36, 56.57 and 72 md^{-1}, respectively. Results showed that the interstitial velocities up to 36 md^{-1} represented a range where mass transfer effect was significant and above it insignificant to the COD removal process. Since WSPs effluent has high flowrates and low organic load, it is possible to induce high interstitial velocities in a HSSF-CW treating this effluent, without clogging and overflow. The performance of these HSSF for tertiary treatment in tropical areas could be improved by considering flow velocity when designing.

Key words | chemical oxygen demand, horizontal subsurface flow constructed wetland, interstitial velocity, mass transfer, removal rate constant, waste stabilisation pond

Anita M. Rugaika (corresponding author)
Bart Van der Bruggen
Department of Chemical Engineering,
KU Leuven,
Celestijnenlaan 200F, B-3001 Leuven,
Belgium
E-mail: *anita.rugaika@nm-aist.ac.tz*;
anitamukarugaika.rugaika@kuleuven.be

Anita M. Rugaika
Karoli N. Njau
The Nelson Mandela African Institution of Science and Technology,
P.O. Box 447, Arusha,
Tanzania

Damian Kajunguri
Department of Mathematics,
Kabale University,
P. O. Box 317, Kabale,
Uganda

Rob Van Deun
Thomas More University College Kleinhoefstraat 4,
2440 Geel,
Belgium

Bart Van der Bruggen
Faculty of Engineering and the Built Environment,
Tshwane University of Technology,
Pretoria,
South Africa

INTRODUCTION

Constructed wetland (CW) systems involve several physical, chemical and biological processes influencing each other when treating wastewater (Langergraber *et al.* 2009; Rizzo *et al.* 2014). This phenomenon has made it difficult to understand the functioning of CWs in detail. As a result, various numerical models were developed to describe the activities in a CW. Such models aim at a better understanding of the biochemical transformation and degradation processes occuring in CWs (Langergraber *et al.* 2009; Llorens *et al.* 2011).

The introduction of CW in Tanzania started in 1998. This was achieved through pilot scale CWs which were used for establishing design guidelines and criteria by using the first order plug flow model based on hydraulic retention time suggested by Reed *et al.* (1995). At that time it was the most used kinetic model. The pilot scale studies were later on transferred into full scale.

$$C_o = C_i\, e^{-kt} \tag{1}$$

where C_o is the outlet concentration (mg/L), C_i is the inlet concentration (mg/L), k is the overall removal rate constant (d^{-1}) and t is the hydraulic retention time (d).

Since temperature influences the reaction rate of biochemical reactions, the first order removal rate constant is commonly corrected using the modified Arrhenius equation. However, Njau *et al.* (2011) mentioned that in tropical climates where there is no significant temperature difference

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between hot and cold seasons the performance of horizontal subsurface flow constructed wetland (HSSF-CW) is more influenced by the system hydrodynamics than seasonal changes in ambient temperature. Njau *et al.* (2011) furthermore indicated that the performance of CW systems in a tropical climate is highly influenced by mass transfer of reacting pollutant in the system. Although the authors recommended consideration of mass transfer effects in CW design in order to enhance pollutant removal, an increase in velocity will result into a CW lay-out with a high length to width ratio. This lay-out might lead to overflow or flooding (Darcy's law) and clogging of the inlet zone due to the high organic loading of the cross sectional area in the inlet zone. Several studies have shown water velocity to influence the removal of some pollutants in CWs (Bounds *et al.* 1998; Kadlec & Wallace 2009; Njau *et al.* 2011; Lohay *et al.* 2012). García *et al.* (2004), Nivala *et al.* (2012), Marato *et al.* (2014) and Wu *et al.* (2015) also mentioned that HSSF systems with shallow depth (smaller cross sectional area hence higher velocity) tend to remove pollutants more efficiently than systems that are deeper, resulting in higher overall removal rate constants.

The removal of pollutants in CWs involves both transport and bio-reaction in the biofilm. Processes taking place in the biofilm contribute to the removal of biodegradable pollutants (Lohay *et al.* 2012). However, for removal of pollutants to take place diffusion of pollutants from the bulk liquid to the reaction site is necessary. The dissolved pollutants need to be transported from bulk water to the stagnant boundary layer surrounding the solid surface, diffuse through the stagnant boundary layer and penetrate the biofilm (Kadlec & Knight 1996; Njau *et al.* 2011). Thus, the overall removal rate constant of pollutants is determined by the combination of mass transfer coefficient and the reaction rate constant.

In systems involving processes in series, the overall rate depends on the rate of the slowest step (Zwart & Janssen 1998; Njau *et al.* 2011). When HSSF-CWs are used as tertiary treatment systems for polishing wastewater, the concentration of pollutants in such wastewater is generally low. With such low concentrations, processes in the HSSF-CW systems are likely to be influenced by mass transport limitation. The degree of influence of mass transport processes can be investigated by studying the influence of velocity to the overall reaction rate. A study by Njau *et al.* (2011) on HSSF-CWs treating domestic wastewater showed a good correlation between organic matter removal and interstitial velocity (ranging from 3.4 m/ to 46 m/d) with higher velocities bringing about higher pollutant removal rates. Lohay *et al.* (2012) focused on domestic wastewater and indicated the overall removal rate constant for the removal of faecal coliforms to also vary with interstitial flow velocity in the HSSF-CW. However, the results leading to this conclusion were obtained in a system that operated as a plug flow reactor under gravity flow where high velocities were difficult to reach.

The hydraulic flow pattern in HSSF-CWs is also extensively debated in literature (Ríos *et al.* 2009; von Sperling & de Paoli 2013). The authors documented actual wetlands not to behave as ideal plug flow reactors hence the model (Equation (1)) leads to misrepresentation of the real wetland units. As a result, the first order plug flow model was modified based on non-ideal reactor hydraulics (Equation (2)) with the aim of better describing the actual CW units (Ríos *et al.* 2009). Thus, the number of tank in series (TIS) model gives the intrinsic removal rate constants as the model gives a better representation of the actual wetland hydraulics. Since a single completely stirred tank reactor, CSTR ($N = 1$) behaves as a perfect complete mix tank, the CW perfomance has been documented to be best represented by the TIS model as it is an intermediate between the two extremes, idealised plug flow and CSTR hydraulic models (Laaffat *et al.* 2015; Dotro *et al.* 2017).

$$C_o = \frac{C_i}{(1 + (\mathrm{kt/N}))^{\mathrm{N}}} \tag{2}$$

where $\mathrm{C_o}$ is the outlet concentration (mg/L), $\mathrm{C_i}$ is the inlet concentration (mg/L), k is the overall removal rate constant ($\mathrm{d^{-1}}$), t is the hydraulic retention time (d) and N is the number of equivalent tanks in series which are presumed to be equal in size, dimensionless.

The challenge of this research was to design mesocosms that could be used to compare different flow velocities without creating different hydraulic retention times (HRTs) or different CW lay-out (length to width ratios). The use of a CSTR was the solution since this hydraulic pattern has been documented to show a better description of CWs than the plug flow pattern (Ríos *et al.* 2009). The interstitial velocity through these mesocosms can be calculated using Equation (3) while the Equation (4) is applied to calculate the rate constant, k.

$$\mathrm{u} = \frac{\mathrm{Q}}{\mathrm{W.h.\varepsilon}} \tag{3}$$

where u is the interstitial velocity (m/d), Q is the flow rate ($\mathrm{m^3/d}$), W is the width of the wetland cell (m), h is the depth of the water column in the wetland cell (m) and ϵ is the porosity as a decimal fraction.

$$C_o = \frac{C_i}{(1 + kt)} \tag{4}$$

where C_o is the outlet concentration in mg/L, C_i is the inlet concentration in mg/L, k is the overall removal rate constant (d^{-1}) and t is the hydraulic retention time (d).

Secondary wastewater treatment in waste stabilisation ponds (WSPs) results only into partial pollutant removal hence producing an effluent quality that does not meet the WHO discharge limits (Mbwele *et al.* 2004). The poor effluent quality of these systems brings the need to integrate WSP and HSSF-CW system in order to meet the discharge limits (Kihila *et al.* 2014). The authors reported the COD effluent levels from Moshi municipality WSP to range between 71.04 mgO_2/L to 221.76 mgO_2/L. Mbwele *et al.* (2004) also reported reduction of COD and BOD in WSPs found in Dar es Salaam to be only 46% and 27%, respectively. This implies that since the organics are reduced in WSPs then the possibility of clogging in the inlet zone of HSSF-CW is highly reduced. However, since WSP effluents are characterised by high flow rates which might result into overland flow (Darcy's law) the aspect of length to width ratio must be taken into account when designing the HSSF-CW system. High flow rates (high interstitial flow velocity) of WSP effluent in the HSSF-CW makes mass transfer approach an interesting approach for pollutant removal.

This study aims at evaluating the possibility of using mass transfer approach to improve the design of HSSF-CWs treating effluent of WSPs. This will be achieved by using pilot-scale HSSF-CWs that work as a CSTR applied with high interstitial velocities in a controlled environment. Moreover, the performance of planted and unplanted systems will be compared together with the influence of interstitial velocity on dissolved oxygen in such system. The system will provide the overall removal rates based on high interstitial velocities, which will determine an ideal flow velocity in HSSF-CW operating as a tertiary treatment unit.

METHODOLOGY

Determination of rate information

The experiments were performed in a batch recycle system (Zwart & Janssen 1998), in which the wastewater is continuously pumped from a receiving tank (500 L tank) through the pilot-scale CW and back. In modeling the behavior of this system the following assumptions were made:

(i) As the conversion per pass is low, the wetland cell can be approximated as a differential reactor behaving like an ideal CSTR.
(ii) The process is isothermal.
(iii) The reservoir is well mixed.
(iv) The porosity is uniform and constant with time.

With these conditions the overall system operates as a batch reactor and can be used to evaluate the rate information in a similar manner as batch reactors are used to determine the rate information of chemical reactions.

Study site

The study was conducted in a greenhouse located at the Nelson Mandela African Institution of Science and Technology (NM-AIST) premises in Arusha, Tanzania covering an area of about 120 m^2. The experimental set-up (Figure 1) erected in a ventilated greenhouse comprised of four (4) identical HSSF-CW cells of size 100 cm high, 150 cm long and 50 cm wide.

All wetland cells were filled with clean and graded gravel (12–20 mm) in size with porosity of 0.35 covering a depth of 80 cm (constant water level). Two cells were planted with *Typha latifolia* while the other two were unplanted and hence used as controls. Each wetland cell was connected to a receiving tank of 500 L. A rotameter (LZT Liquid- Sp. Gr.1.0 with range 1–7 L/min) and a pump (Pedrollo type, PKm 60 with HP 0.5) were also fitted after each wetland cell for setting different flow rates and pumping the wastewater from wetland cell back to the

Figure 1 | The experimental set up erected in a ventilated greenhouse.

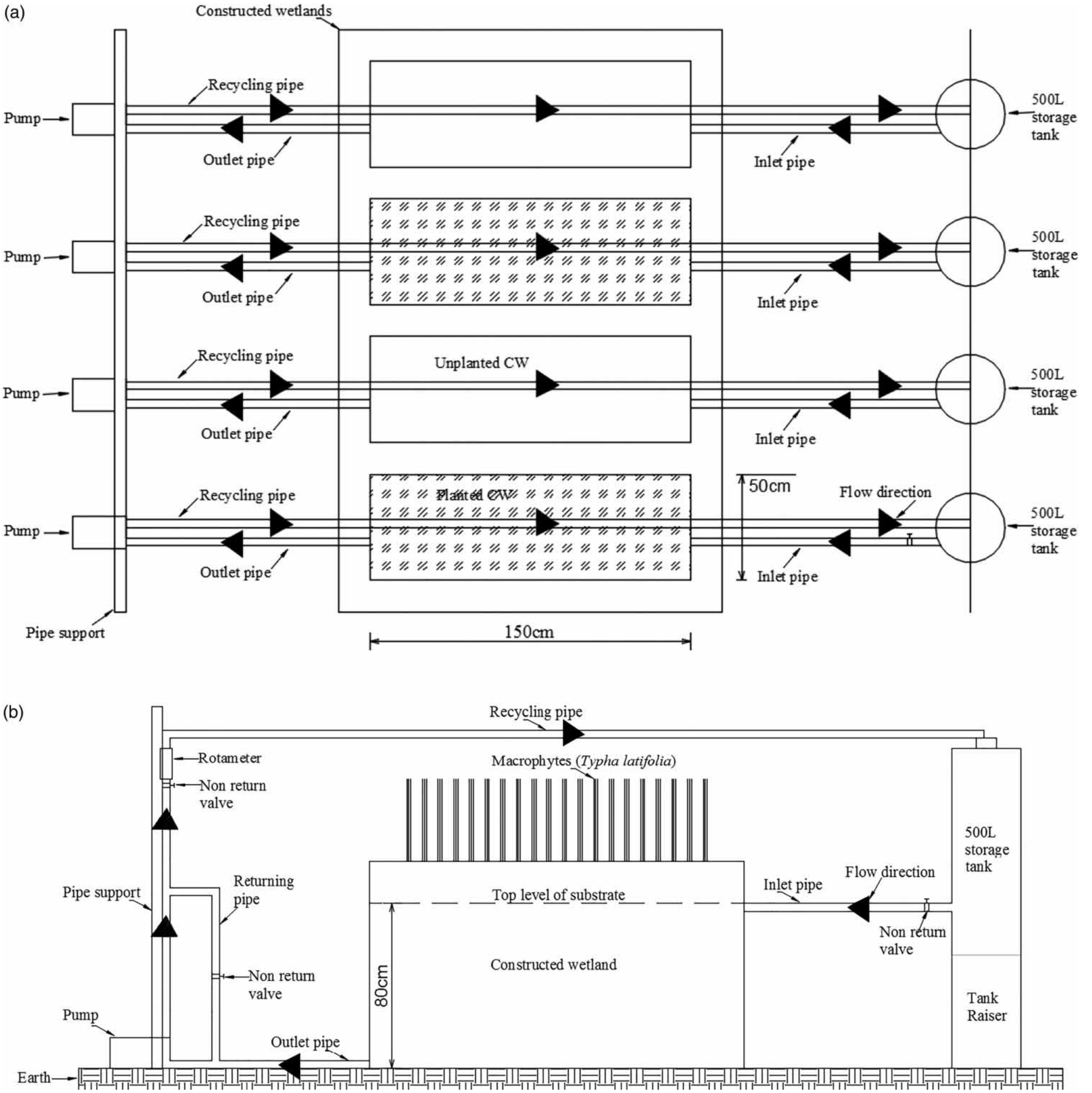

Figure 2 | (a) Plan of the experimental set- up. (b) Schematic diagram of the experimental set up.

receiving tank. Below is the plan of the experimental set- up (Figure 2(a)) and schematic diagram (Figure 2(b)).

Operational procedure

The objective of this experimental set up was applying different interstitial flow velocities to recycle wastewater with the same organic loading between the wetland cell and the receiving tank in batch mode at constant HRT. Pretreated domestic wastewater from NM-AIST offices was taken from septic tanks and filled into a 500 L receiving tank. Wastewater was continuously recycled between a 500 L receiving tank and the wetland cell by using a pump for the whole treatment period. The systems were operated at four different flow rates (1.5, 3.5, 5.5 and 7.0 L/min), which were controlled by rotameters of flowrate range

(1–7 L/min) mounted after each cell and regulated by gate valves (England-BS 5154–20/100 °C).

These flow rates correspond to the following interstitial velocities 15.43, 36, 56.57 and 72 md^{-1}, respectively. The random effect on the experiments was obtained by setting different flow rates for each pair (control and planted cell). This meant that for each run two different flow rates were set. At the beginning of each run (new flow rate set) a new feed of the same wastewater volume was used and at the end of each run the wastewater in all wetland cells was flushed out.

Sample collection and analysis

For each new flow rate set, daily sampling and analysis of wastewater was done from the receiving tank for seven consecutive days. Sampling took place at 11:00 each sampling day. After seven days a new flow rate was tested with fresh septic tank wastewater. This was done between January and February, 2015. Plastic bottles of 250 mL were used for sample collection. Samples were then taken to NM-AIST laboratory for COD analysis. DO data were measured *in situ* using a multi-parameter meter type Hanna HI 9829.

Wastewater samples taken to the laboratory were filtered using Whatman filter paper (0.45 μm pore size) to remove suspended solids. For COD analysis, 2 mL of filtered sample was added to the vial containing the reagent and 2 mL of deionized water was added to the vial containing the reagent as blank. The vials were then placed in the COD test tube heater HI 839,800 at a temperature 150 °C for 2 h. After the digestion period the heated vials were left at the test tube heater and after 20 minutes they were removed and put at the test tube rack to cool to room temperature. Then the vials were inserted to HI 83099 (COD and multi-parameter bench photometer) for COD analysis. The analysis was done according to the Standard Method for the Examination of Water and Wastewater (APHA/AWWA/WEF 1998).

Statistical data analysis

Statistical data analysis was done using Origin Pro 9.0 software (Origin Lab Corporation, Northampton, MA, USA). Before the analysis, data were subjected to a normality test using Shapiro- Wilk test. Since the data were normally distributed, the analysis was done using parametric test, one way ANOVA with Tukey's multiple comparison test. Significant differences were at 5% level of significance ($P < 0.05$).

RESULTS AND DISCUSSION

Chemical oxygen demand (COD)

The statistical summary of the mean removal rate constants of COD for planted and unplanted cells at different interstitial velocities are shown in Table 2. Table 2 also shows COD removal percentages to vary from 69.2% to 81.3% for planted cells and from 61.4% to 70.4% for unplanted cells at different interstitial velocities. From the results, planted cells performed better in COD removal compared to unplanted cells.

Soluble organic matter is degraded both aerobically and anaerobically by microorganisms attached in the plant roots and substrate media (Gagnon *et al.* 2007). Thus, planted cells reduce more COD levels compared to unplanted cells because of more surface area provided by the plant roots and a more complex microflora (Gagnon *et al.* 2007; Toscano *et al.* 2015) together with the additional oxygen leakage from macrophyte roots (Zhang *et al.* 2014). (Figure 3).

A significant difference ($P < 0.0001$) in COD removal rate constants was observed for different interstitial velocities in planted cells. Tukey's multiple comparison test showed a significant difference ($P < 0.05$) between different COD removal rate constants at different velocities, however at high interstitial velocities there was no significant difference. The results showed a significant difference ($P < 0.05$) when the interstitial velocity of 15.43 md^{-1} was compared with 36 md^{-1}, 56.57 md^{-1} and 72 md^{-1} when the initial COD concentration was ≤93 mg/L, thus a dilute system. However, no significant difference ($P > 0.05$) was observed when 36 md^{-1}, 56.57 md^{-1} and 72 md^{-1} were compared (Figure 3).

The interstitial velocity up to about 36 md^{-1} represented a range where mass transfer is limiting, while above 36 md^{-1} the influence of mass transfer was insignificant to the overall COD removal process. This implies that the removal rate

Table 1 | Randomized flow rate settings in control and planted cells

	Flow rate settings (L/min)			
Runs	**Control 1**	**Planted 1**	**Control 2**	**Planted 2**
1st run	1.5	1.5	7.0	7.0
2nd run	3.5	3.5	5.5	5.5
3rd run	7.0	7.0	1.5	1.5
4th run	5.5	5.5	3.5	3.5

Table 2 | Statistical summary of the overall removal rate constants of COD for planted and unplanted cells together with their percentage removal at different interstitial velocities

Interstitial velocities (md^{-1})	15.43		36		56.57		72	
HSSF-CW system	**Planted**	**Control**	**Planted**	**Control**	**Planted**	**Control**	**Planted**	**Control**
COD in (mgO$_2$/L)	78	79	85	81	84	82	80	81
COD out at HRT of 6days (mgO$_2$/L)	24	31	19	27	18	31	15	24
COD-removal (%)	69.2	61.4	78.2	67.3	79.2	62.8	81.3	70.4
Number of values	12	12	12	12	12	12	12	12
Mean removal rate constant (d^{-1})	0.429	0.361	0.692	0.337	0.745	0.462	0.733	0.428
Std. deviation	0.1002	0.1064	0.1195	0.0840	0.1945	0.2179	0.1083	0.1121
P value (unpaired T-test)	0.1224		<0.0001		0.0029		<0.0001	
P value summary	ns		****		**		****	

ns = $p > 0.05$, ** = $p < 0.01$, **** = $p < 0.0001$.

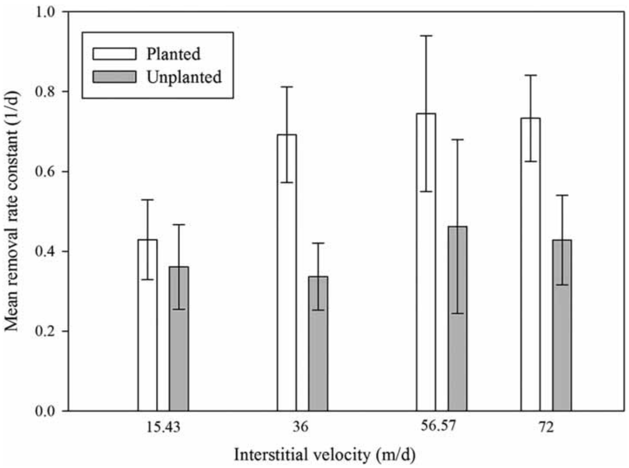

Figure 3 | The mean overall removal rate constants of COD for planted and unplanted systems at different interstitial velocities.

constants tend to increase with increase in interstitial velocities until a point where mass transfer effect is insignificant and the removal rate constants were practically constant (Figure 3). This suggested that the rates at which the organic pollutants were brought to the reaction site were higher than the rates of biochemical reactions taking place within the biofilm.

Dissolved oxygen (DO)

The mean DO levels obtained in the receiving tank at different velocities ranged from 2.66 to 3.32 mg/L in planted cells and from 1.94 to 2.42 mg/L in unplanted cells. These DO values showed significant difference (U = 868, *P* = 0.0000327) between planted and unplanted cells (Table 3).

The difference in DO levels between planted and unplanted systems confirms the reason for planted system to performed better in COD removal than unplanted system. In CWs, the major pathways that supply oxygen to the system are water inflows, direct diffusion from the atmosphere (Kayombo *et al.* 2005) and leakage from macrophyte roots (Zhang *et al.* 2014). In this study, no direct diffusion of oxygen took place since the water was not exposed to the atmosphere. The recycled stream was slowly aerated when it returned back to the tank. Higher

Table 3 | Statistical summary of DO levels for planted and unplanted cells at different interstitial velocities

Interstitial velocities (md^{-1})	15.43		36		56.57		72	
HSSF-CW system	**Planted**	**Control**	**Planted**	**Control**	**Planted**	**Control**	**Planted**	**Control**
Number of values	14	14	14	14	14	14	14	14
Mean DO levels (mg/L)	2.78	2.13	2.66	1.94	2.78	2.08	3.32	2.42
Std. Deviation	0.879	0.717	1.268	0.876	1.368	1.060	1.084	1.156
P value (paired t-test)	0.0032		0.0004		0.0003		0.0045	
P value summary	**		***		***		**	

** = $p < 0.01$, *** = $p < 0.001$.

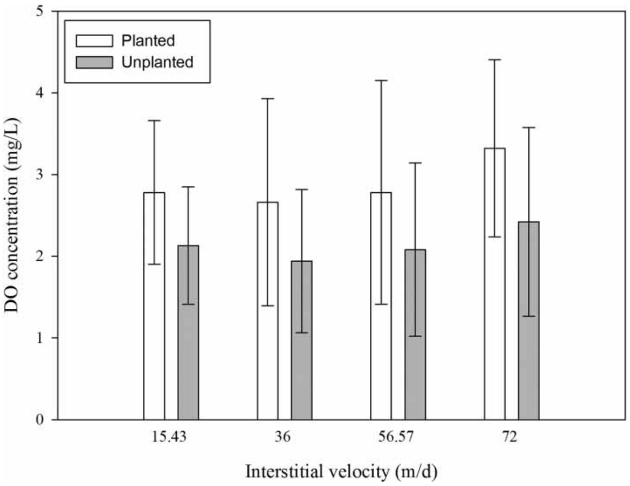

Figure 4 | Possible correlation of DO levels at different interstitial velocities in the planted and unplanted cells.

DO levels were observed in planted cells than unplanted cells due to macrophyte photosynthetic activities. During photosynthesis process, plants release oxygen through stomatal and root pores. Thus, leakage of oxygen from the root pores aerated the system and favored aerobes in biochemical reactions. A study by Colmer (2003) reported well development of aerenchyma in most wetland plants. The authors went further mentioning the importance of these spongy tissues in promoting emission of gases from the plant to the atmosphere and improving the movement of oxygen to submerged tissues. Various studies have shown plant roots to be capable of releasing 0.02–12 g/m^2d of oxygen (Armstrong & Armstrong 1990; Brix 1994). Although DO levels were observed to increase with increasing interstitial velocity, the data did not show any significant difference in DO levels at different interstitial velocities (Table 3 and Figure 4).

CONCLUSION AND RECOMMENDATIONS

It is clear from this study that at low interstitial velocities below 36 md^{-1}, the overall removal rate constant value for COD removal is a function of interstitial velocity hence mass transfer is the limiting process. However, this was not the case for higher interstitial velocities whereby the dependency of rate constant on interstitial velocity diminishes. Since WSP effluents normally flow at high flow rates, interstitial velocities close to 36 md^{-1} could be achieved when a HSSF-CW is used to treat such effluent. Moreover, since the organic loading of the WSP effluent is already lowered from previous treatments, the risk of HSSF-CW clogging in the inlet zone is minimal. Based on the results of this research, the lay-out of a HSSF-CW treating WSP effluent should be optimized to reach flow velocities of approximately 36 md^{-1}. However, consideration of length to width ratio is an important aspect when HSSF-CW is used as a tertiary treatment unit so as to prevent overflow (Darcy's law). If the overall removal rate constants from this research (0.43 and 0.74 d^{-1}) were to be used to design a HSSF-CW, optimizing the lay-out based on the interstitial velocity would result in a reduction of the necessary surface area with approximately 40%.

Since this study only considered the effects of high interstitial velocities on COD removal, it should be taken as the starting point for determining such effects on other pollutants such as nitrogen and fecal coliform as they were not taken into account. However, from wastewater reuse point of view, there is no need to remove nitrogen and phosphorus from wastewater as they are important components in agriculture.

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CONFLICT OF INTEREST

No conflict of interest was declared.

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