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Kinetic of on Nutrient Removal in Low-Strength Domestic Wastewater under Continuous Operation of Pilot Scale of Hybrid Reed Bed System (SF-VF-HF)

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Abstract: Hybrid Reed Bed Constructed Wetland system (HRBCWs) for domestic wastewater treatment has been proven to be effective and as an alternative for conventional wastewater treatment technologies. Combination of constructed wetlands enhances pollutant removal efficiency as hybrid CWs could cover the limitation of each single (CWs). In our study, we employed a HRBCWs in three-stages, consisting of a Surface-Flow (SF) system followed by a Vertical-Flow (VF) system and finally by a Horizontal-Flow (HF) system. The system was planted with *Scirpus grossus* plant that could survive and reproduce with a continuous feed of 86.5 L/day of the raw wastewater. The results indicate that, the system with a 3 days HRT and different sizes of gravels can achieve average removal of 85% NH_4-N and 71% PO_4-P. Also, the kinetic removal were investigated as absorbents in laboratory scale experimental, the results showed that the q_e, k_1 were found to be 132.68 mg NH_4-N/g biomass and 0.46 $d^{\bullet 1}$ for the surface flow system and 152.76 mg NH_4-N/g biomass and 0.53 $d^{\bullet 1}$ for the vertical flow system, respectively and 34.99 mg NH_4-N/g biomass and 0.35 $d^{\bullet 1}$ for the horizontal flow system with a pseudo-first-order. The NH_4-N biosorption by SF, VF and HF were well fitted to the pseudo first-order plot with R^2 of 0.99, 0.98 and 0.98 for SF, VF and HF, respectively. The planted HRBCW system shows a better performance than the unplanted system. These results indicate that the HRBCW system using *Scirpus grossus* plant has a high effectiveness for treating domestic wastewater.

Key words: Hybrid Reed Bed Constructed Wetland (HRBCW) system, domestic wastewater, kinetic removal, phytoremediation, Surface-Flow (SF), Vertical-Flow (VF)

INTRODUCTION

Constructed wetlands are artificial design for wastewater treatment systems which have been planted with aquatic plants and which rely upon natural microbial, biological, physical and chemical processes to treat wastewater use rooted wetland plants with courage media to provide the treatment process (Kadlec and Knight, 1996). As well as the green technology, contaminant treatment systems such as Constructed Wetland systems (CWs) have the competitive advantage of producing greater quality without having the input of fossil energy power thereby impair the treatment operation charge (Sim *et al*., 2007; Lee *et al*., 2010 and Bruch *et al*., 2011).

Actually, there are two basic types of constructed wetlands; Surface Flow (SF) and Sub-Surface Flow (SSF) systems (Kadlec and Knight, 1996). SF systems are similar to natural wetlands with shallow flow of wastewater (usually <60 cm deep) over saturated media substrate. SSF systems mostly employ gravel as the main media to support the growth of plants, wastewater flows vertically or horizontally through the substrate where it comes into contact with microorganisms, living on the surfaces of

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plant roots and substrate (Cooper 1996; Kadlec and Knight, 1996) allowing pollutant removal from the bulk liquid. SSF constructed wetlands are further divided into two types: Vertical Flow (VF) and Horizontal Flow (HF) systems, both are typically more effective than the Surface systems (SF) in terms of mass pollutant removal per m^2 of surface area system (Luederitz *et al.*, 2001; Ruan *et al.*, 2006; Vymazal *et al.*, 2006 and Vymazal, 2007). There are many types of contaminants in the human water use (like: phosphorus, nitrogen, heavy metals, organics, suspended solids and cadmium), removal can be achieved by using combined constructed wetlands, through a complicated system of plants, media, microbial activity and bio-mass population (Fountoulakis *et al.* 2009). Nitrogen and organics contaminants removal from domestic wastewater in such designed human ecosystems is globally important because there was an uncontrolled discharge of nutrients wastewater into natural water bodies like river, ponds and ocean (Xinshan *et al.*, 2010; Chen *et al.*, 2011). The literature affirmed that effective removal of organics contaminants in SSF system compared to SF system, nonetheless as a result, producers from these systems for reducing nitrogen rates are often weak (Vymazal *et al.*, 2006).

Recently, a combination of VF and HF systems, known as hybrid system can be also, employed for the treatment of domestic wastewater. Such combinations often optimize nitrogen and organics removal due to presence of aerobic, anaerobic and anoxic phases (Vymazal, 2005; Kadlec and Wallace, 2009). Organic compounds can be degraded aerobically and anaerobically in SSF systems. Oxygen for aerobic treatment can be supplied via. atmospheric oxygen diffusion, convection (wind effect) and/or macrophyte root transfer into the plant rhizosphere (Cooper, 1996). Anaerobic (lacking oxygen) organics removal can proceed inside the media pores.

Nowadays, a human design of household removal have become a very popular treatment technologies (Obarska-Pempkowiak and Klimkowska, 1999). These systems considered as low-cost alternative for human water use treatment, especially, suitable for developing countries. They also have low operation and maintenance requirements (Wittgren and Maehlum, 1997). They have been proved to be efficient in reducing different undesired constituents such as nitrogen and organics contaminants from household wastewaters. Biochemical transformations, adsorptions, precipitations, volatilization and plant uptake of pollutants are the main pollutant removal mechanism in a constructed wetlands system (Inamori *et al.*, 2007). Constructed wetland is principally using the same natural degradation processes and nutrient uptake.

Scirpus grossus was used in this study due to its potential as a tolerance plant (Tangahu *et al.*, 2013). This plant has fibrous roots in white to brown color, triangular and solid stems, more than 2 m long leaves with bisexual flowers grouped together. It is a perennial tropical aquatic plant, the common names are *Giant bulrush*, *Greater club rush* and *Rumput menderong* in Malaysia. It is an aquatic perennial plant which is widely used to treat domestic wastewater in phytoremediation or wetland treatment (Jinadasa *et al.*, 2008).

A large number of physical, chemical and biological processes are involved in these systems influencing each other (Langergraber *et al.*, 2009) which are not fully understood to date due to lack of appropriate models. As far as first order kinetics is considered, Lagergren equation seems to be the most widely applied equation. This equation was the first rate equation for the sorption of liquid-solid systems based on solid capacity (Wang *et al.*, 2010). A study on the characteristic curves of this equation showed that Lagergren equation can yield superior correlation with the sorption data over the pseudo second order kinetics for a number of sorption systems (Tseng *et al.*, 2010). Another important observation regarding Lagergren equation is that it correlates better data in the adsorption systems that are not far from equilibrium (Rudzinski and Plazinski, 2007). The aims of this study were to evaluate the kinetic of ammonia-nitrogen (NH_4-N) removal in low-strength domestic wastewater under continuous operation of pilot scale of combination Hybrid Reed Bed Constructed Wetland systems (HRBCWs) and find an appropriate model for the kinetics of removal.

MATERIALS AND METHODS

Design of Hybrid Reed Beds Constructed Wetland systems (HRBCWs): Hybrid Reed Beds Constructed Wetland (HRBCW) system is an engineering design that can be used for single family houses or for small communities. In current study, a HRBCW system Fig. 1 containing of three similar tanks that are combined to each other's and all tanks material were constructed from fiber glass (1 m, W*2 m, L*1 m, D) with a PVC pipe. Gravels-sand-gravel were used as media of these systems and arranged with 10-15, 3-5 mm (river sand) and 30-35 mm diameter of media, respectively, from the top of each thank to down layer to draw nutrients from domestic

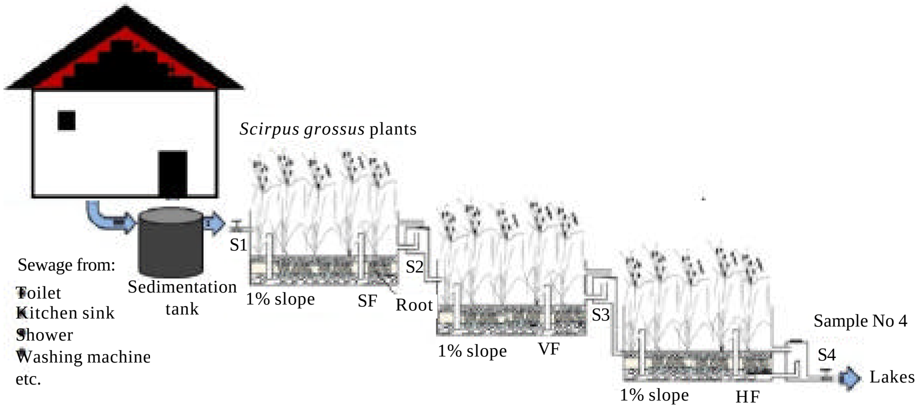

Fig. 1: Schematic layout of HRBCW systems: a) Side view and b) Plan view

wastewater. All tanks were filled with the mixture media to depth around 40 cm and provided Surface Flow (SF) and Vertical Flow (VF) systems with oxygen by pumping pipe. Domestic wastewater level in the sub-surface flow system was 10 cm below the surface of the media and >15 cm for the surface flow system. All tanks were planted with *Scripus grossus* by installing HRBCW systems entailing putting the operating system in continuous process to find out the feasibility of this combined systems to eliminate nutrients from household wastewater.

The domestic wastewater of the Bukit Putri effluent of primary was pumped into the sedimentation tank and then distributed to two HRBCW systems. *Scirpus grossus* was planted in the first three series systems (HRBCWs). In the second three series systems, there were control HRBCW system with the gravels-sand-gravels as a media and without vegetation plant. The influent flow rate of each HRBCW system was 86.5 L/day with continuous feed of the household sewage and 3 days HRT. During the course of the experiments (4 months), sewage samples were collected every week from the influent and effluents of HRBCW systems and were measured in the laboratory of the Engineering Faculty of UKM campus.

Sampling analysis: Samples of the sewage wastewater treatment were collected every week depending on the sheet time of study and basis from outlet zone as well as at distances of 2 m from inlet to the outlet in each tank at a depth of 0.15 m below the surface for sub-surface flow system and up to 0.15 m for surface flow system. Samples were taken from the end of each tank, then, the samples were immediately analyzed for the parameters. The test of ammonia-nitrogen, nitrate-nitrogen and phosphorus are carried, according to standard method of water and wastewater which is approved by EPA. The test was conducted using DR3900 HACH machine.

Statistical analysis: Statistical analysis using the two-way ANOVA (Analysis of Variance) on the performance of HRBCW systems in terms of nutrients removal was carried out using Statistical Package for the Social Sciences (SPSS), Statistics 21.

Ammonia-nitrogen uptake: The NH_4-N uptake by the plant was calculated using the following Eq. 1 (Vieira and Volesky, 2000):

$$q_e = \frac{V(C_i - C_e)}{X} \qquad (1)$$

Where:

q_e = The NH_4-N uptake (mg NH_4-N/g biomass)
V = The Volume of the sewage wastewater (L)
C_i = The influent Concentration of NH_4-N in the solution (mg NH_4-N /L)
C_e = The effluent Concentration of NH_4-N in the sewage wastewater (mg NH_4-N/L)
X = The dry weight of the biomass (g)

Kinetics removal: In this study, kinetics removal was described using the first order equation of Lagergren and the pseudo second-order equation (Ho and McKay, 1999). The sorption kinetic provides valuable insights into the reaction pathways and the mechanism of a sorption reaction. The pseudo-first order model is used to describe the sorption kinetics (Ho and McKay, 1999). The model is based on the assumption that the adsorption follows first order chemisorption's and predicts the behavior over the whole range of concentration and is in agreement with an adsorption mechanism being the controlling step rate. This kinetic model is represented as Eq. 2 (Yuan *et al*., 2009):

$$\log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \qquad (2)$$

where, q_t and q_e are the NH_4-N uptake on the biosorbent at any time and at equilibrium (mg NH_4-N/g biomass), k_1 is the rate constant of Lagergren first-order biosorption (d^{-1}). The straight-line plot of $\log(q_e-q_t)$ against t gives $\log(q_e)$ as slope and intercept equal to $k_1/2.303$. Hence, the amount of solute sorbed per gram of sorbent at equilibrium (q_e) and the first-order sorption rate constant (k_1) can be evaluated from the slope and the intercept.

RESULTS AND DISCUSSION

Nutrients removal: There are three types of nutrient analyzed in this study that is NH_4-N, NO_3-N and PO_4-P. The high NH_4-N content was mainly due to the fact that after NH_4-N was formed by ammonification (Ehrig and Stegmann, 1992). Phosphorus is the most important nutrient enhancing eutrophication in lakes and coastal waters (Klapper, 1992). Therefore, phosphorus must be removed largely by HRBCW systems of treatment.

Ammonia-nitrogen (NH_4-N) removal: Figure 2 shows the overall performance of NH_4-N removal for both the HRBCW systems (planted and control). Influent concentration for HRBCW system (SF-VF-HF) ranges from 5.60-8.15 mg/L while the effluent concentration ranges from 0.23-2.96 mg/L in the planted systems and 0.97-2.80 mg/L in control systems. The removal efficiency ranges from 36-81% for planted systems and 40-62% for control systems. The results indicated that control HRBCW system have lower removal efficiency as compared to planted HRBCW system. Table 1 illustrates the correlation between removal efficiency with time of treatment with correlation coefficient (R^2) that ranged from 0.58-0.88. This shows that removal efficiency percentages are increases with increase time of treatment. Generally, it shows that better NH_4-N removal are obtained in planted systems when compared to the control systems.

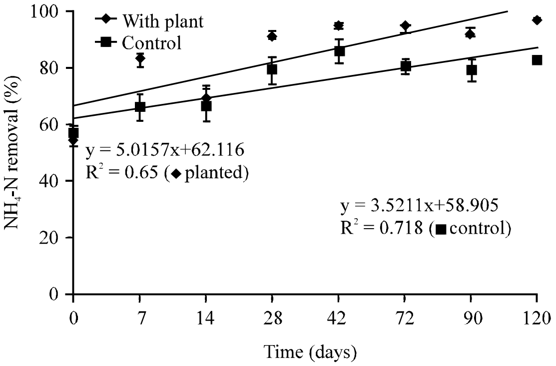

Fig. 2: Removal efficiency of NH_4-N in planted and control HRBCW systems (SF-VF-HF)

The NH_4-N mean removal values are showed in VF and HF flow systems were more reliable than the SF flow system which was also, noted by the USEPA because in sub-surface flow system (VF and HF), water must remain below the media surface to minimize human contact.

Decomposition and mineralization processes in the wetlands will convert a significant part of organic-N to ammonia. The source of ammonia is believed to be from the anaerobic decomposition of the organic-N trapped in the bed as particulate matter. Biological nitrification followed by denitrification is believed to be the major contribution towards ammonia removal in constructed wetlands (Reed, 1993). Vertical and horizontal flow system which is planted by *Scirpus grossus* shows a better removal as compared to the control system. This is because *Scirpus grossus* has long penetrating roots to enhance removal efficiency of NH_4-N.

Based on statistical analysis at 5% level of significance, it was found that effluent ammonia concentrations ranged from 3.57-5.37 mg/L for the surface flow system, the ammonia removal efficiency of the planted system was generally, higher than the control system. Maximum ammonia removal efficiency for the planted system was 45.9% while maximum efficiency for the control system was 38.9%. For the vertical flow in the HRBCW system, it was found that effluent ammonia concentrations ranged from 0.72-3.1 mg/L, ammonia removal efficiency for planted system was higher than of the control system. Maximum ammonia removal efficiency for planted system was 80.8% while that for the control

Table 1: Correlation between removal efficiency with time of treatment for NH_4-N

Flow types/Systems	Equations	Correlation coefficient (R^2)	Removal (%)
Surface Flow (SF)			
With plant	y = 2.4833x+28.025	0.67	39.20
Control	y = 2.7524x+18.105	0.74	30.49
Vertical Flow (VF)			
With plant	y = 5.0868x+43.324	0.58	66.21
Control	y = 2.5927x+40.5	0.69	52.17
Horizontal Flow (HF)			
With plant	y = 8.985x+0.3948	0.88	40.83
Control	y = 2.9222x+14.176	0.61	27.33
HRBCW system			
With plant	y = 5.0157x+62.116	0.65	84.69
(SF-VF-HF)			
Control	y = 3.5211x+58.905	0.72	74.75

Table 2: The pseudo first-order kinetic constants of Lagergren for HRBCW systems

	Experimental results	Pseudo first-order of Lagergren		
Systems	q_e (mg NH_4-N/g biomass)	q_e (mg NH_4-N/g biomass)	k_1 (d^{-1})	Error (%)
SF	134.98	132.68	0.46	1.71
VF	159.59	152.75	0.53	4.28
HF	36.35	34.99	0.35	3.73

k_1; Pseudo-first-order kinetic constant of NH_4-N; q_e Adsorption amount from pseudo-first-order equation; q_e; Experimental: adsorption amount from experiment data

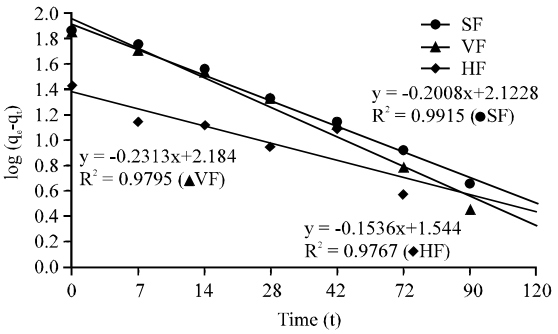

Fig. 3: The first-order of Lagergren plot for NH_4-N biosorption

system was only 61.4%. For the horizontal flow system, it was found that effluent ammonia concentrations ranged 0.23-2.96 mg/L, ammonia removal efficiency for the planted system was higher than that of the control system. Maximum ammonia removal efficiency for planted system was 70.9% while the maximum efficiency for control was 40.1% throughout the study period. The p-value was statistically significant between removal with (planted and control) systems of exposure and also, used two-way ANOVA test to analyze the percentage of NH_4-N removal in the HRBCW system and to explain the interaction between parameters involved. Percentage of NH_4-N removal was statistically significant with systems of exposure with $p<0.05$. The two-way ANOVA test also, showed insignificant interactions with $p<0.05$ between (day and removal) (hybrid system and removal) and (day*hybrid system*removal). The results showed that the HRBCW system in this study has a high efficiency in removing NH_4-N from the domestic wastewater.

NH_4-N kinetics removal: Figure 3 showed a pseudo-first-order of Lagergren plot for NH_4-N biosorption on HRBCW system. The q_e and k_1 obtained from the respective intercept and slope of Eq. 2 were found to be 132.68 mg NH_4-N/g biomass and 0.46 d^{-1} for the surface flow system and 152.76 mg NH_4-N/g biomass and 0.53 d^{-1} for the vertical flow system, respectively, also, 34.99 mg NH_4-N/g biomass and 0.35 d^{-1} for the horizontal flow system with a pseudo-first-order. As shown in Table 2, the calculated q_e values for SF, VF and HF demonstrated lower errors (<5%) when compared with the experimental q_e. The obtained q_e was 134.98 mg NH_4-N/g biomass, 159.59 mg NH_4-N/g biomass for SF and VF systems, respectively. Also, getting 36.35 mg NH_4-N/g biomass for HF system.

However, the NH_4-N biosorption by HRBCW systems were well fitted to the pseudo first-order plot with R^2 of 0.99, 0.98 and 0.98 for SF, VF and HF, respectively, suggesting that the pseudo first-order was favourable to describe the biosorption mechanism of NH_4-N by SF, VF and HF. Moreover, higher k_1 values for VF than those of SF and HF proved that vertical flow system is much better for NH_4-N biosorption.

Nitrate (NO_3-N) removal: The overall results show inlet and outlet of NO_3-N concentration in HRBCW systems. Influent concentration ranges from 0.1-0.5 mg/L while the effluent concentration for surface flow system ranges from 0.72-1.69 mg/L in planted systems and 0.61-1.11 mg/L in HRBCW control system. Effluent concentrations from the vertical and horizontal constructed wetland system ranges from 2.10-3.49 mg/L, 1.67-2.79 in planted systems and 1.39-1.63, 1.14-1.58 mg/L in control systems, respectively.

The major removal mechanism of NO_3-N is due to the processes such as ammonification, nitrification and denitrification. Ammonia is oxidized to nitrate by nitrifying bacteria in aerobic zones. Nitrates are converted to nitrogen gas (N_2) and Nitrous Oxide (N_2O) by denitrifying bacteria in anoxic zones. To do nitrification process, oxygen is needed and it is supplied by diffusion from the air pump and also, from the macrophytes roots. NO_3-N is not only removed through these processes, it is also, taken up by plants and turned to biomass and released back as organic nitrogen after decomposition of the plants.

Phosphorus (PO_4-P) removal: Figure 4 shows the overall performance of phosphorus removal for both the HRBCW systems (planted and control). Influent concentration ranges from 2.54-3.22 mg/L. Effluent concentrations in the surface flow system ranges from 1.69-2.78 mg/L for planted system and 2.14-3.08 mg/L in control system. Also, for sub-surface flow systems, ranges from 0.76- 2.38, 0.34-2.10 mg/L in planted systems and 1.19-1.71, 0.58-2.42 mg/L in control systems for vertical and

Table 3: Correlation between removal efficiency with time of treatment for PO_4-P

Flow type/Systems	Equations	Correlation coefficient (R^2)	Removal (%)
Surface Flow (SF)			
With plants	y = 3.3288x+14.411	0.93	29.39
Control	y = 1.9006x+7.9217	0.73	16.47
Vertical Flow (VF)			
With plants	y = 5.5374x+14.05	0.95	38.97
Control	y = 4.0778x+14.612	0.90	32.96
Horizontal Flow (HF)			
With plants	y = 6.2698x+10.846	0.93	39.06
Control	y = 5.8711x+6.3186	0.96	32.74
HRBCW systems			
With plants (SF-VF-HF)	y = 6.8155x+40.308	0.87	70.98
Control	y = 6.8102x+29.795	0.87	60.44

Table 4: A comparison of the removal efficiency of the different flow systems in HRBCWs with other studies in different countries

Flow system/ Location of pilot system	NH_4-N (%) Rem	NO_3-N (%) Rem	PO_4-N (%) Rem	HRT days	Plant type in the pilot system	References
SF						
Taihu, China	22.8	34.2	35.1	-	*Typha angustifolia*	Li *et al.* (2008)
Putrajaya, Malaysia	-	70.7	84.3	-	*Lepironia articulata*	Sim *et al.* (2007)
Petchaburi, Tailand	75.4	-	44.9	5	*Typha angustifolia*	Klomjek and Nitisoravut (2005)
Nyanza, Kenya	36	-	29	-	*Cyperus papyrus*	Bojcevska and Tonderski (2007)
Bangi, Malaysia	39.2	-	29.39	3	*Scirpus grossus*	This study in 2018
VF						
Longdao, Beijing	10.5	-	30.6	-	*Phragmites australis*	Chen *et al.* (2008)
Wuxi, China	61.7	-	48.9	-	*Phragmites typhia*	He *et al.* in 2006
Ankara, Turkey	57.86	-	4.19	-	*Phragmites australis*	Korkusuz *et al.* (2004)
Chalkidiki, North Greece	49.6	-	17.9	2	*Phragmites australis*	Gikas *et al.* (2007)
Bangi, Malaysia	66.21	-	38.97	3	*Scirpus grossus*	This study in 2018
HF						
Pingtung County, Taiwan	22.16	54	32.14	8.5	*Eichhornia crassipes*	Lee *et al.* (2004)
Barcelona, North-East Spain	30.1	-	58.33	5.5	*(Phragmites australis*	Garcia *et al.* (2004)
Juja, Nairobi city, Kenya	17.13	22	57.14	-	*Cyperus papyrus*	Mburu *et al.* (2012)
Dar es Salaam, Tanzania	23.01	44.3	-	-	*Typha latifolia*	Kaseva (2004)
Bangi, Malaysia	40.83	-	39.06	3	*Scirpus grossus*	This study in 2018

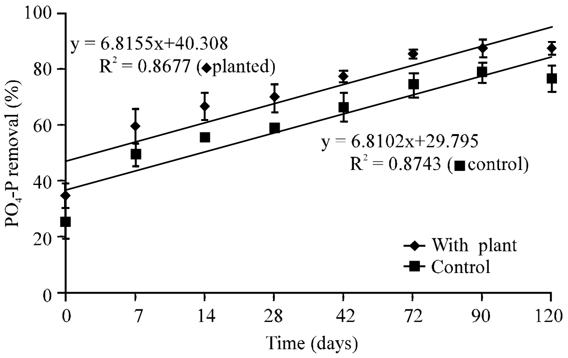

Fig. 4: Removal efficiency of PO_4-P-N in (planted and control) HRBCW systems (SF-VF-HF)

horizontal flow systems, respectively, resulting in high removal rates that were largely dependent on inlet concentration. The phosphorus removal efficiency of surface flow system is ranged from 13.7-38 % for planted systems and 4.3-21 % for control system. For vertical and horizontal flow systems, the removal ranges from 14.4-55 and 11.8-55.4% for planted systems and 12.7-44.4%, 10-51.3% for control systems, respectively. The results showed that the control system has lower removal efficiency as compared to planted system. Removal efficiency in planted systems for HRBCW systems reaches up to 87% where else removal efficiency for HRBCW control system is only 78.6%. The efficiency of the process decreases slowly towards the end of the experimental period (Fig. 4).

Table 3 illustrates the correlation between removal efficiency with time of treatment with correlation coefficient (R^2) from 0.73-0.96. This shows that removal efficiency percentages are increases with increase time of treatment. A comparison of the removal efficiency of the different flow systems in HRBCWs with other studies in different countries was carried out in Table 4. As illustrated in Table 3, the removal efficiencies of NH_4.N are better compared to those of the other CW systems.

Based on statistical Analysis of Variance (ANOVA) at 5% level of significance, the p-value was statistically significant between removal with (planted and control) HRBCW systems of exposure and also used two-way ANOVA test to analyze the percentage of PO_4-P removal in the HRBCW systems and to explain the interaction between parameters involved. Percentage of PO_4-P removal was statistically significant with $p<0.05$. The two-way ANOVA test also showed the interactions with $p<0.05$ between (day*removal) (hybrid system*removal) and (day*hybrid system*removal). The results showed that the HRBCW system in this study has a high efficiency in removing PO_4-P from the domestic wastewater.

CONCLUSION

The HRBCW systems for different flow direction beds had significantly higher performance during the wetlands operational period for nutrients pollutants treatment. Removal efficiency of this study did show an effective performance as predicted. Presence of *Scirpus grossus* did increase the ability of the HRBCW systems to decrease the level of nutrients as showed in the results.

Kinetic data for NH_4-N adsorption onto the various adsorbents were fitted to pseudo first-order Lagergren and pseudo second-order to investigate the mechanism of adsorption. The Lagergren second-order (calculations not shown) was ruled out because its regression was not significant. Hence, NH_4-N removal by the adsorbents did not follow the Lagergren second order kinetics. The first-order fitted the experimental data well with a correlation coefficient for the HRBCWs ($R^2>0.95$).

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