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Multistage Horizontal Subsurface Flow vs. Hybrid Constructed Wetlands for the Treatment of Raw Urban Wastewater

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Received: 1 May 2020; Accepted: 19 June 2020; Published: 23 June 2020

Abstract: In this study, pilot-scale hybrid constructed wetlands (CWs) and multistage horizontal subsurface flow CWs (HF CWs) have been studied and compared for the treatment of raw urban wastewater. In the hybrid CWs, the first stage was a mulch-based horizontal subsurface flow CW and the second stage was a vertical subsurface flow CW (VF CW). The VF CWs were used to determine if sand could improve the performance of the hybrid CW with respect to the mulch. In the multistage HFs, mulch, gravel and sand were used as substrates. The effect of water height (HF10: 10 cm vs. HF40: 40 cm) and surface loading rate (SLR: 12 vs. 24 g Chemical Oxygen Demand (COD)/m^2d) has been studied. The results show that the use of sand in the vertical flow stage of the hybrid CW did not improve the average performance. Additionally, the sand became clogged, while the mulch did not. The effect of water height on average pollutant removal was not determined but HF10 performed better regarding compliance with legal regulations. With a SLR of 12 g COD/m^2d, removals of HF10 were: 79% for COD, 75% for NH_4^+-N, 53% for dissolved molybdate-reactive phosphate-P (DRP), 99% for turbidity and 99.998% for *E. coli* and total coliforms. When SLR was doubled, removals decreased for NH_4^+-N: 49%, DRP: −20%, E coli and total coliforms: 99.5–99.9%, but not for COD (85%) and turbidity (99%). Considering the obtained results and the simplicity of the construction and operation of HFs, HF10 would be the most suitable choice for the treatment of raw urban wastewater without clogging problems.

Keywords: wastewater; shallow constructed wetland; horizontal flow; substrate; organic mulch; gravel; sand

1. Introduction

Constructed wetlands (CWs) are natural, low-cost reactors, particularly adequate for wastewater treatment and reuse in small communities. Molinos-Senante et al. [1] collected the opinions of 29 international experts from the academic, research and industrial fields about the most suitable wastewater treatment technology for small communities. CWs (24%) and ponds (23%) were the most preferred alternatives, while extended aeration and membrane bioreactors were the least preferred ones. Considering the water flow, CWs can be classified as horizontal flow or vertical flow and surface flow or subsurface flow CWs. Horizontal subsurface flow CWs (HF CWs) are easily designed and constructed. Additionally, their operation and maintenance is simple and inexpensive. However, their efficacy regarding nitrification and total N removal is usually low. Consequently, vertical subsurface flow CWs (VF CWs) and hybrid CWs (combinations of HF CWs and VF CWs) have been successfully tested [2]. Nevertheless, two important drawbacks of VF CWs when compared with HF CWs are achieving a homogeneous distribution of the influent over the bed, and the higher clogging risk because of the finer substrate material used.

The porous media is a basic CW component since it supports biofilm and plants. Gravel and sand are the conventional components of porous media, but these materials are being extracted faster from the environment than they can be replaced. Sand and gravel are the most extracted group of materials worldwide, exceeding both fossil fuels and biomass [3]. The environmental impact of sand extraction includes coastal and river erosion, coral reef degradation, biodiversity loss and the spread of invasive species. These environmental impacts have a profound effect on the local human population in terms of seawater intrusion in coastal aquifers, thus affecting domestic water supply and increasing salinization of cultivated land, the increase in potential breeding sites for malaria-transmitting mosquitoes, etc. All these effects can result in social and political conflicts [4], particularly in developing countries [5]. Thus, more sustainable materials for CW beds should be found.

In recent decades, many different alternative materials such as industrial wastes and agricultural and forest byproducts have been tested in an attempt to improve CW performance and/or environmental sustainability [6,7]. Organic substrates obtained from agro-forest wastes can be more environmentally friendly than gravel and sand since local material would be used and the impacts of mining and transport would be minimized. In this regard, Wang et al. [8] obtained good results with an organic substrate composed of peat/crushed pine bark. Additionally, rice husk and organic mulch have been successfully employed in the removal of total nitrogen [9–11]. Moreover, organic substrates can increase the removal of micro-pollutants by sorption [12,13]. Denitrifying bioreactors are a particular case of organic-based CWS that have been used to reduce the nitrate concentration of different effluents. The carbon-rich media promotes nitrate reduction to N_2 gas under anaerobic conditions. However, recent studies have shown their long term performance regarding the removal of fecal indicators [14].

The typical substrate depth of subsurface flow CWs is 0.5 m, but several authors have found that shallow constructed wetlands (SCWs) can improve the removal of nitrogen, organic matter [15,16] and estrogenic disruptors by a more efficient dissolved oxygen transfer to the media [17]. Additionally, a SCWs require fewer building materials, and consequently a lower cost of construction, operation and maintenance [18]. Holland et al. [19] studied the effect of wetland depth and flow rate on residence time distribution (RTD) characteristics. They used two different average depth settings: 16.6 cm for low water level and 39.8 cm for high water level. The obtained results indicated that flow rates did not have a significant effect on RTD characteristics, but all the parameters indicating hydraulic efficiency exhibited lower efficiency for the higher water levels.

In the literature, there are studies on the comparison of horizontal and hybrid CWs, but few consider the use of agro-forest residues as substrates of CWs, and particularly the possibility of substituting gravel and sand by agro-forest wastes in VF CWs. In our opinion, using locally abundant and available wastes as CW substrates can greatly improve their economic and environmental sustainability. Thus, the main goal of this work was to compare a multistage HF CW with hybrid CWs for the treatment of raw wastewater without clogging. Thus, the hybrid CWs were used to test the effect of the age of the mulch-based substrate in the HF stage and to compare sand with mulch substrates for VF CWs. With the multistage HF CWs, the effects of water height, surface loading rate (SLR), and hydraulic loading rate (HLR) were studied.

2. Experimental

The pilot-scale CWs were constructed in September 2015. Monitoring started in November 2015 and ended in July 2016. The CWs were located outdoors and the influent was wastewater from the

Campus of Tafira of the University of Las Palmas de Gran Canaria (Canary Islands, Spain). The inflow was collected in a 17-m^3 pit and controlled with a timer-controlled triturating pump located at the bottom of the pit. The pump was programmed to work for 3 min every 2 h during the whole day, seven days a week, during all the experimental period. No sampling was performed during rainy days and during the university vacation periods of Christmas, summer and Easter. Figure 1 illustrates the CWs employed. The multistage HF CWs and the HF stage of the hybrid CW were built with plastic recipients, each with a volume of 265 L, a height of 50 cm, width of 55 cm, length of 125 cm, and a surface area of 0.7 m^2. As can be seen in Figure 1, four treatment lines were designed to study:

- The effect of water depth and SLR on the multistage HF CWs. For this, two lines composed of four recipients each were built. The first two units had a mulch substrate, the third one had gravel and the last one had sand. The plastic recipients were perforated at the bottom to be connected in series with plastic tubes of 5 cm in diameter. The holes were covered with a 0.02-m gravel layer to avoid the clogging of the tubes by the mulch. The purpose of this substrate sequence (mulch–gravel–sand) was to minimize the clogging of the porous media, which is usually stronger near the inlet [20]. Additionally, the first unit was located above the other ones to favor the aerobic degradation of the retained solids and to minimize clogging and ponding [21]. The palm mulch allows for significant loads of Total Suspended Solids (TSS) and Chemical Oxygen Demand (COD), without clogging [22]. The mulch was obtained by trituration of dry branches of the Canarian palm tree (*Phoenix canariensis*) which is very abundant in the zone. The mulch had a porosity of 54% and a hygroscopicity of about 10%. The gravel was basaltic with 49% porosity and average diameter of 6.5 mm. The last unit of the HF was built with sand (45.5% carbonate, porosity: 42%, average diameter: 0.32 mm) in an attempt to provide a final polishing of the water, for the improved removal of pathogens [23], and to reduce the water flow speed. These lines were used to determine the efficiency of a HF in which gravel has been partially replaced by mulch, as well as the effect of water height. Thus, a line was built with a water height of about 10 cm (HF10), as well as a second line with a 40 cm height (HF40). The CWs were planted with Phragmites, Cyperus and Canna, with the exception of the final sand-based units that were not unplanted. In addition to the influent and effluent, an intermediate sampling point was included in the HF CW systems to determine the contribution of the first (HF40-1, HF10-1) and second stages (HF40-2, HF10-2) of the removals.
- The hybrid CWs were built with an organic-based HF CW followed by a VF CW. The HF stage had mulch and the VF stages were plastic cylindrical recipients with a surface area of 0.1 m^2 and depth of 70 cm. The hybrid CWs were used to: (i) compare them with the multistage HF CW, and (ii) compare sand with mulch in the vertical flow stage. A 10-cm-high layer of gravel was used at the bottom of the VF stages to avoid the outlet clogging. The effluents of the HF stages were directed by gravity to the VF stages. The hybrid CWs were not planted in order to minimize the number of variables to be considered.

2.1. Water Analysis

All the parameters were analyzed according to standard methods [24] in un-filtered samples. N and P species have been determined photometrically with a Zuzi Uv-vis spectrophotometer 4201/50 (Auxilab, Spain). Total N (TN) and P (TP) were determined after alkaline peroxydisulfate digestion at 120 °C for 90 min. After digestion, TN was determined as nitrites and TP as molybdate-reactive phosphates. Dissolved molybdate-reactive phosphate-P (DRP) was measured directly without digestion. Nitrite ions were measured photometrically after the formation of a pink diazo dye with sulfanilamide and N N-(1-naphthyl)-ethylene-diamine. Nitrate ions were analyzed as nitrites after reduction with hydrazine-Cu-Zn solution. *E. coli* and total coliforms were enumerated with membrane filtration and cultivation at 37 °C for 24 h with a selective chromogenic agar (Panreac, Spain). Chemical Oxygen Demand (COD) was determined by open reflux digestion with dichromate and titration and a ferrous standard. Biochemical Oxygen Demand (BOD_5) was measured with the manometric method with nitrification inhibition (Velp, Italy). TSS was determined gravimetrically. Turbidity was determined with a portable nephelometer (Velp, Italy). Sulfate ions were determined nephelometrically after precipitation with $BaCl_2$. Ammonium-N ($NH_4{}^+$-N) was determined with a selective electrode (Metrohm, Switzerland). COD, turbidity, $NH_4{}^+$-N, DRP, *E. coli* and total coliforms have been routinely measured to determine the effect of the selected design parameters.

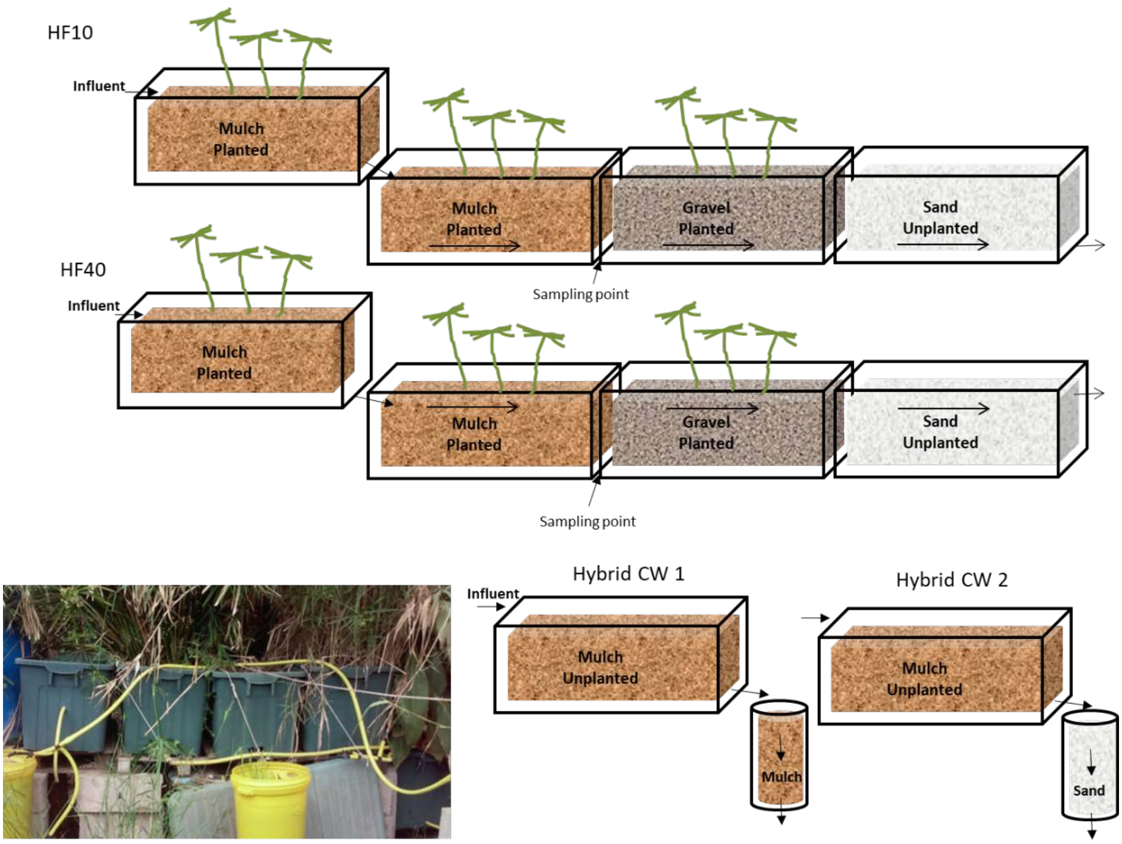


Figure 1. Scheme of the constructed wetland (CW) mesocosms employed in the study: multistage horizontal subsurface flow CWs (HF CWs) with water heights of 10 cm (HF10) and 40 cm (HF40); hybrid CWs with mulch-based HF CW followed by vertical subsurface flow (VF) with mulch (Hybrid CW 1) and sand (Hybrid CW 2). The photograph shows the hybrid CWs and the first unit of the multistage HF CWs.

2.2. Statistics

Removals, surface loading rates and removal rates have been determined according to the following expressions:

- Removal: $100 \times (C_{inf} - C_{eff})/C_{eff}$
- SLR: $C_{inf} \times Q \times 10^{-3}/S$
- Removal rate: $(C_{inf} - C_{eff}) \times Q \times 10^{-3}/S$

where C_{inf}: concentration (mg/L) in the influent; C_{eff}: concentration (mg/L) in the effluent; Q: inflow (L/d), S: surface area of the reactor (m^2).

A simple ANOVA test was used for statistical analysis to determine significant statistical differences in the performance of wastewater treatments. The analyses were carried out with the R-Commander program from the free R software package. When data were not homoscedastic (Bartlett test) or normally distributed (Shapiro–Wilks test), the non-parametric Kruskal–Wallis test was employed. A significance level of $p = 0.05$ was used.

3. Results and Discussion

3.1. Characteristics of the Influent and Surface Loading Rates

The influent can be considered as medium strength [25] for almost all the parameters, with the exception of TN and NH_4^+-N, which were closer to high strength (Table 1). As expected, the concentrations of NO_x^- (nitrates + nitrites) in the influent were always lower than 1 ppm, being undetectable in most samples. The average concentration of TN (70 mg/L) was dominated by that of NH_4^+-N (57 mg/L). The high NH_4^+-N/TN ratio (81%) may be due to the particular characteristics of the campus, in which the urine proportion in wastewater can be higher than those of other N sources. Similar concentrations and NH_4^+-N/TN ratio were found for a schoolhouse in Estonia [26].

Table 1. Average concentrations (±standard deviation) and number of data of the measured parameter. The units are mg/L except turbidity (NTU) and pathogen indicators (CFU/100 mL).

	Average ± Std. Dev., Number of Data
COD	497 ± 197, 32
BOD	259 ± 57, 13
TSS	170 ± 121, 14
Turbidity	180 ± 105, 23
pH	7.5 ± 0.22, 14
Sulfate	62 ± 37, 10
NH_4^+-N	57 ± 20, 37
(NO_2^- + NO_3^-)-N	0.4 ± 0.82, 24
TN	70 ± 22, 11
DRP	7.4 ± 3.4, 25
TP	10 ± 4, 12
E. coli	5.4 ± 8.2 (× 10^6), 16
Total coliforms	8.1 ± 10 (× 10^6), 15

Similarly, the average concentration of DRP was about 74% of that of TP.

The HF CWs and the hybrid CWs were fed with 144 L/d and 44 L/d, respectively, of raw wastewater from the campus. Table 2 shows the average SLRs for the multistage HF CWs and hybrid CWs. The surface loadings of this study (HLR: 41–54 L/m^2d and COD-LR: 20–25 g/m^2d, Table 2) were not statistically different enough to be able to compare their removals and were above those reported by other authors. For example, Vymazal found an average value of 13.9 g COD/m^2d in the Czech Republic, where the unsatisfactory removal of NH_4^+-N or P was achieved [2].

Table 2. Average surface loading rate (SLR) (±standard deviation) of the HF CWs and the hybrid CWs.

	HF CWs	Hybrid CWs
HLR, L/m^2d	41 ± 14	HF: 61 ± 14, VF: 426 ± 102, HF-VF: 54 ± 13
COD-LR, g/m^2d	20 ± 11	HF: 30 ± 14, VF: 60 ± 49, HF-VF: 25 ± 11
TSS-LR, g/m^2d	6 ± 3	HF: 10 ± 6, VF: 6 ± 4, HF-VF: 9 ± 5

3.2. Multistage HF CWs

3.2.1. Effect of the Water Level Height

The conventional depth of HF CWs is 50–60 cm. However, different authors have observed that shallow HFs (<27 cm deep) achieved better removals of COD, BOD_5, NH_4^+-N and DRP. The better performance of shallower HF CWs could be explained by considering that: (i) water can be forced to go through the rooting zone of the plants, and (ii) shallower substrates displayed higher redox potential and slightly higher dissolved oxygen concentrations [15,27]. Thus, the effect of water height was considered a key parameter in this study. According to the results for the entire experimental period (Table 3), HF10 was, in general, more efficient than HF40, and was significantly better for turbidity.

Table 3. Average removal (%) (±standard deviation) of HF40 and HF10 for entire experimental period.

	HF40	**HF10**
COD	77 ± 10	82 ± 9
BOD	96 ± 4	96 ± 5
TSS	92 ± 7	99 ± 1
Turbidity	95 ± 4	99 ± 1
TN	63 ± 25	53 ± 26
NH_4^+-N	57 ± 26	61 ± 29
TP	20 ± 31	37 ± 25
DRP	18 ± 55	14 ± 64
E. coli	99.7 ± 0.6	99.9 ± 0.2
Total coliforms	99.6 ± 0.6	99.95 ± 0.1

As can be observed, both treatment lines were very efficient at the removal of organic matter (BOD_5 and COD), turbidity and TSS. The average removals of (HF40: 63%, HF10: 53%) and NH_4^+-N (HF40: 57%, HF10: 61%) for all the experimental period fall in the upper limit of the range provided for CWs (40–60%) by Vymazal [28]. Those of TP (20–37%) and DRP (18–14%) were clearly lower. However, the removals of NH_4^+-N and DRP were strongly affected by HLR, as will be discussed in the following section.

The effect of a design parameter on the performance of CWs is usually measured by comparing the obtained average removals [29], but the high dispersion of the data can obscure the results and lead to ambiguous conclusions [27]. Another way to compare the efficiencies of HF40 and HF10 is to determine their capability to meet the legislation by measuring the percentage of samples with concentrations below the legal limits. Table 4 shows the degree of compliance with the Spanish National Regulation as the percentage of samples with values below those established by the Royal Decrees that regulate the limits for: (i) COD regarding the discharge of treated wastewater into the environment [30] and (ii) TSS, turbidity and *E. coli*, considering the possible reuse (urban, agricultural, industrial, recreational and environmental) of treated wastewater [31].

Table 4. Degree of compliance with the Spanish National Regulation measured as percentage of samples (%) with values below the legal limits for discharging treated wastewater into the environment [30] and re-use [31]. Units of Chemical Oxygen Demand (COD), TSS, turbidity and *E. coli* are mg COD/L, mg/L, NTU and CFU/100 mL, respectively.

	HF40 (%)	**HF10 (%)**	**Legal Limit**	**Legal Regulation**
COD	73	90	125	R.D. 509/1996
TSS	54	100	10	
Turbidity	25 83	61 100	2 10	R.D. 1620/2007
E. coli	33 40 80	33 53 93	102 103 104	R.D. 1620/2007

As can be seen, the degree of compliance of HF10 was higher than that of HF40 for COD (90% vs. 73%), TSS (54% vs. 100%), turbidity (2 NTU: 61% vs. 25%, 10 NTU: 100% vs. 83%).

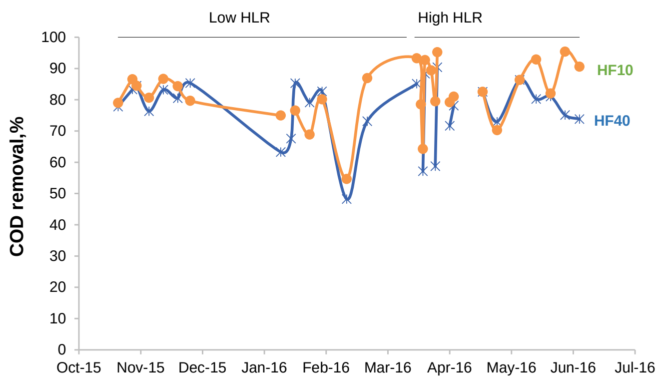
3.2.2. Effect of HLR

To determine the effect of SLR on the efficiency of the HF CWs, the influent pump working time was doubled on 30 March 2016. Thus, two different periods can be regarded. Table 3 summarizes the SLRs of HF10 and HF40 for the periods of high and low SLRs.

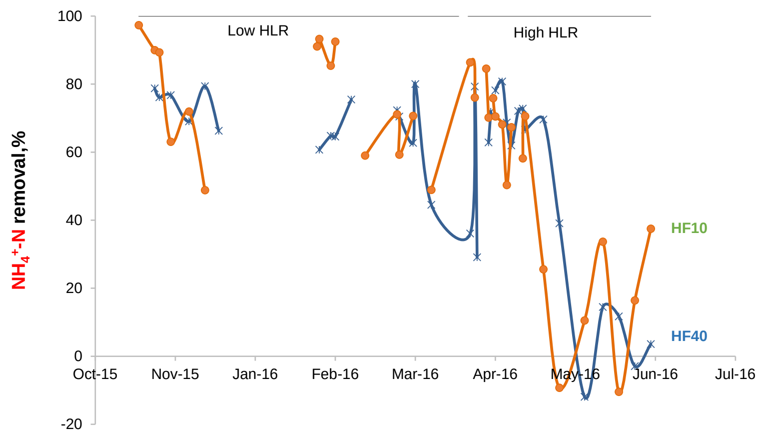
As can be observed in Table 5, the SLRs of both treatment lines were quite similar ($p > 0.05$) for each period and the removals can be compared. Figure 2 illustrates the removals of COD, NH_4^+-N, DRP, turbidity, *E. coli* and total coliforms in HF40 and HF10 and Table 6 summarizes the average SLRs and the removals achieved.

Table 5. Average surface loadings rates (±standard deviation) of HF40 and HF10 in the periods of high and low loading rates.

	HF40		HF10	
	Low	**High**	**Low**	**High**
HLR, L/m^{2}d	25 ± 5	51 ± 8	26 ± 4	51 ± 7
COD-LR, g/m^{2}d	12 ± 5	24 ± 10	12 ± 5	25 ± 11
TSS-LR, g/m^{2}d	4 ± 2	6.5 ± 4	5 ± 3	7 ± 4

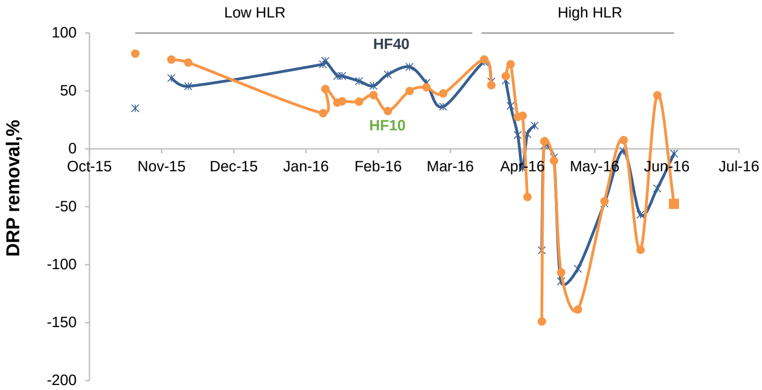


(**a**)



(**b**)

Figure 2. *Cont.*



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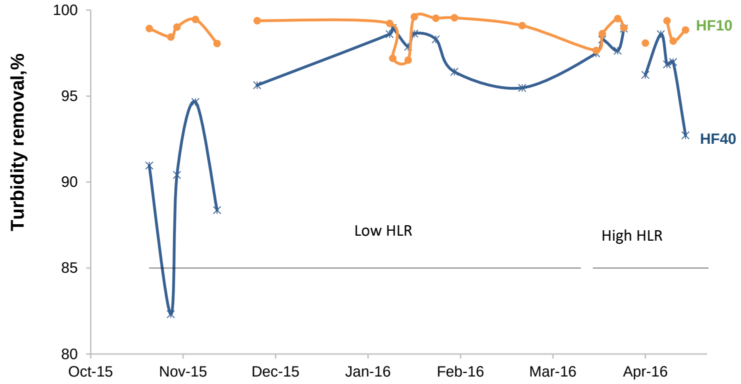

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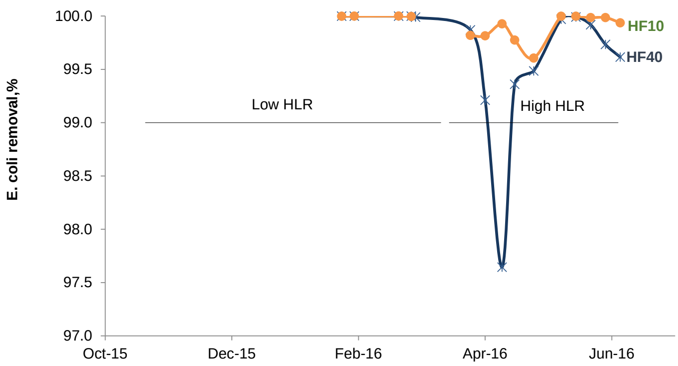

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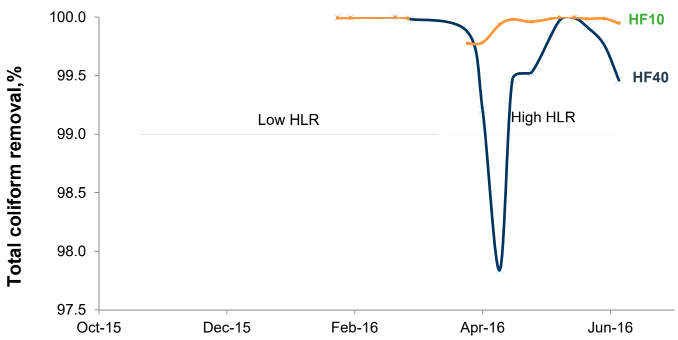


(**f**)

Figure 2. Removal (%) of: (**a**) COD, (**b**) NH_4^+-N, (**c**) DRP, (**d**) turbidity, (**e**) *E. coli* and (**f**) total coliforms in HF40 and HF10 during the periods of high and low HLR.

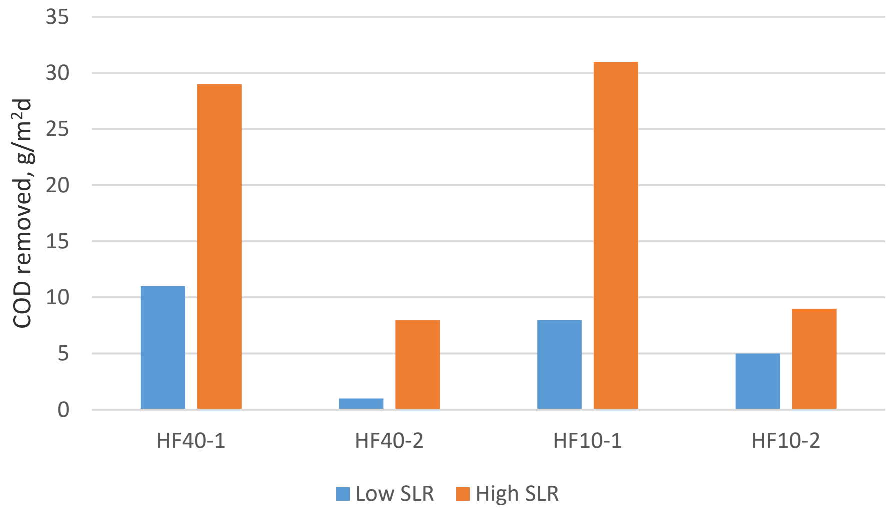
Table 6. Average removals (%, ±standard deviation) for HF40 and HF10 in the periods of low and high hydraulic loading rate (HLR). The different letters next to the same raw values denote significant differences for the considered parameter.

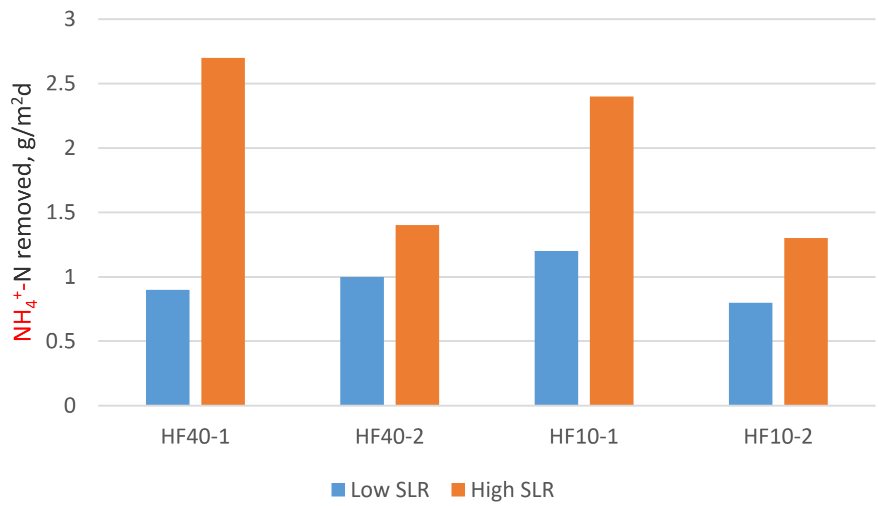
	HF40		HF10	
	Low HLR	**High HLR**	**Low HLR**	**High HLR**
COD	76 ± 11	78 ± 11	79 ± 9	85 ± 9
NH_4^+-N	67 ± 12	48 ± 31	75 ± 10	49 ± 31
DRP	60 ± 13	−16 ± 52	53 ± 17	−20 ± 61
Turbidity	94 ± 5 ^{a}	97 ± 2 ^{b}	99 ± 1 ^{b}	99 ± 1 ^{b}
E. coli	99.997 ± 0.004 ^{a}	99.5 ± 0.7 ^{b}	99.998 ± 0.1 ^{a}	99.9 ± 0.1 ^{b}
Total coliforms	99.994 ± 0.01 ^{a}	99.5 ± 0.6 ^{b}	99.994 ± 0.0 ^{a}	99.9 ± 0.08 ^{b}

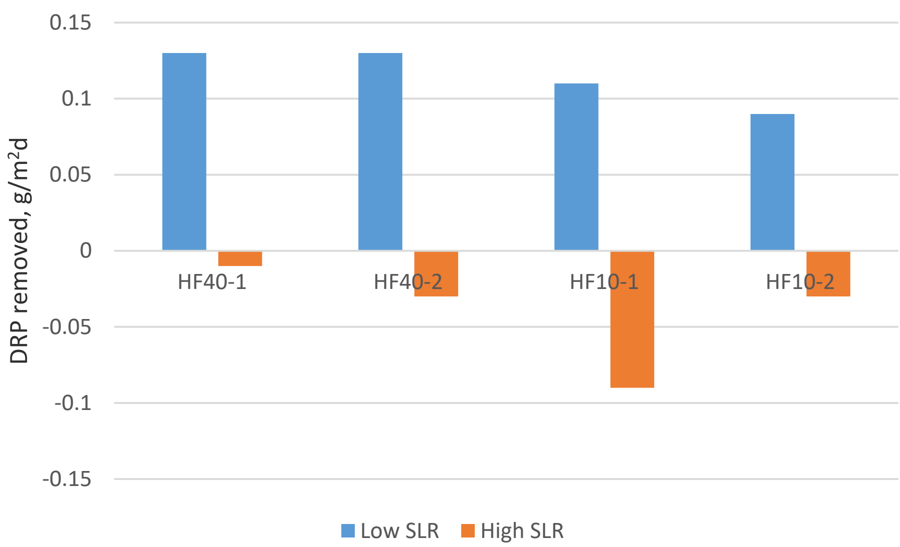
The results from Figure 2 and Table 6 indicate that augmenting the SLR reduced the removals of NH_4^+-N, DRP and fecal indicators but those of COD and turbidity remained high. Figure 3 shows the mass removal rates (g/m^{2}d) of COD, NH_4^+-N and DRP and *E. coli* in the first stages (HF40-and HF10-1) and second stages of the HF CWs (HF40-2 and HF10-2) during the periods of high and low HLR. In the case of COD, the first stages achieved the highest COD mass removal rates in both periods.

In the period of low HLR, the average removals of NH_4^+-N were high for HF CWs (HF40: 67%, HF10: 75%, Table 6), suggesting the availability of enough dissolved oxygen to support nitrification. This could be partially due to the fact that the first unit of the CWs was placed above the other ones and the influent was intermittently dosed. Thus, a fill and drain effect was obtained that would favor the aeration of the substrate. Unfortunately, the role of the plants in terms of N removal was not considered in this study. When the HLR was increased, the removal of NH_4^+-N decreased (HF40: 48%, HF10: 49%) and became more unstable, as shown by the higher standard deviations observed (Table 6 and Figure 3). Unlike the results of this study, Nivala et al. [27] found negligible NH_4^+-N removal in shallow HF CWs, though the shallowest systems achieved the lowest mean effluent NH_4^+-N concentrations. In this study, in the low HLR period, similar surface removal rates were observed in both parts of the HFs. However, when the HLR increased, the surface removal rates also increased in both stages, being slightly better in the first stage, as observed for COD (Figure 2).

The average DRP removals in the period of low HLR were statistically similar (HF40: 60%, HF10: 53%, $p > 0.05$) and were high for HF CWs. Nonetheless, the high HLR had a notoriously negative effect, since DRP removals became negative (HF40: −16%, HF10: −20%). Dierberg et al. [32] found that TP retention efficiency decreased at high HLR due to lower HRT, as it favored preferential flow and affected the P diffusion and sorption processes. It is widely accepted that the main DRP removal mechanism in CWs is the adsorption/precipitation on the substrate, as plant uptake and microbial activity are less important [33]. It was expected that the sand used in the final unit of HF10 and HF40, with a high carbonate content, and thus in Ca^{2+} and Mg^{2+} ions, could help in the removal of DRP by precipitation [34]. However, the porous media used in these experiments (palm mulch, gravel and sand) did not have any removal phosphorus removal capacity, as observed in lab experiments. This can explain why there was no difference in the DRP surface removal rate removal in both stages of HF10 and HF40 (Figure 3). Thus, the results of this study indicate that plant uptake and microbial activity had a more relevant role than expected. In fact, plant uptake is considered to be the second most important mechanism of P removal [35]. Additionally, microbial P removal by means of polyphosphate-accumulating organisms can be an important mechanism [36]. These authors found that an intermediate HRT provided the best removal of TP in VFs, as it provided a balance between P adsorption on the substrate and release under anaerobic conditions. This effect can explain the negative DRP removals observed in the high HLR period of the present study.







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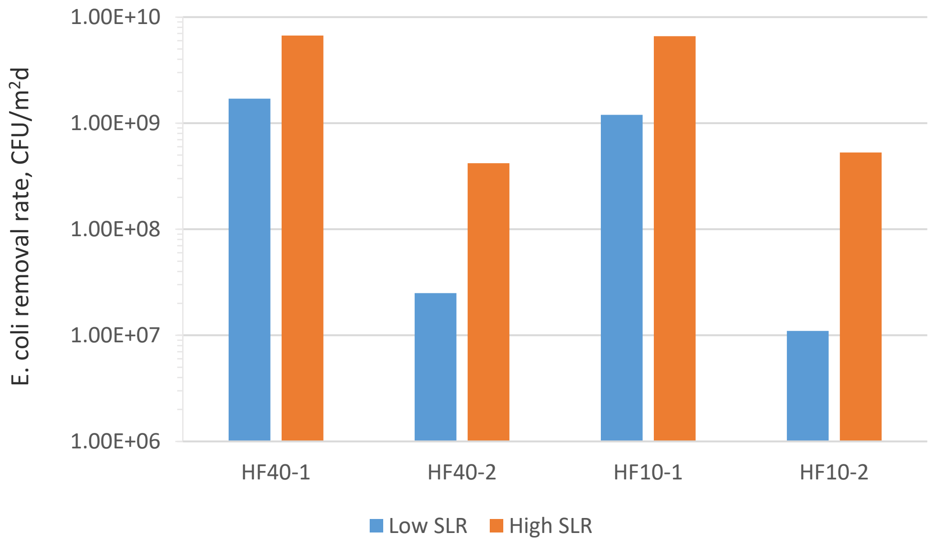

Figure 3. Average surface loading removal rates (g/m^{2}d) of COD, NH_4^+-N, DRP, and *E. coli* (CFU/m^{2}d) in the first stage (HF40-1 and HF10-1) and second stage (HF40-2 and HF10-2) of the HF CWs for the periods of low and high SLR.

The main mechanisms of pathogen removal in CWs are to kill them off by starvation or predation, sedimentation, filtration and adsorption [37]. Many different variables play a role in the removal of pathogens in CWs: temperature, influent composition, HRT and HLR, water flow type (surface, sub-surface, vertical or horizontal), the presence and type of macrophytes or the substrate [38]. Additionally, Morató et al. [23] observed that water depth had also an important effect on disinfection. The results from the present study do not reveal a significant role of water depth regarding *E. coli* and total coliform elimination (Table 6). During the low SLR period, *E. coli* and total coliform removals of almost five orders of magnitude were registered for both treatment lines (Table 6). Other authors have found similar *E. coli* and pathogen removals in CWs using fine soil [39] and sand [40] as substrates, but such high removals are not usually found in CWs [38].

In the case of *E. coli* and total coliforms, a clear removal reduction was observed after increasing the HLR for HF40 with respect to HF10 (Figure 2). This fact suggests the higher robustness of the latter. The analysis of surface removal rates for the first and second stages of HF40 and HF 10 (Figure 3) reveals a similar behavior of that of COD: a higher average SLR removal of both parameters at higher SLR and a stronger retention in the first part of the system (H40-1 and HF10-1). The results of this study suggest that the presence of the sand in the last treatment units of HF10 and HF40 did not play a key role in the removal of pathogens, since SLR removals were not better in the second stages of the treatment lines. This fact suggests that the presence of the plants and the good aeration of the substrate (as shown by the good NH_4^+-N removals observed) are the main pathogen removal mechanisms. Macrophytes can improve pathogen removal by increasing HRT with their root system [41] and providing a larger surface area for microbe attachment, favoring the formation of biofilms, oxygen seepage through roots, the secretion of plant exudates, etc. [38].

3.2.3. Hybrid CWs: Sand vs. Mulch Substrates for the VF Stage

The hybrid CWs achieved similar average removals without significant differences, as can be seen in Table 7. However, the preliminary results of the first 3 months of operation of Hybrid CW 1 with sand, were especially good and better than those of Hybrid CW 2, with removals of NH_4^+-N and *E. coli* greater than 90% and 4–5 log units, respectively. The *E. coli* concentrations in the effluent were 40–540 CFU/100 mL, although HLRs as high as 426 L/m2d were used in the VF stage (Table 2). However, after about 3 months of operation, the first symptoms of clogging began to be observed, i.e., ponding on the surface of the sand-based VF. In consequence, performance was dramatically reduced. Initially, maintenance strategies such as resting periods of several days and scraping the surface were enough to unclog the filter, but after about 9 months of operation it became irreversibly clogged. It was necessary to remove a 0.10 m layer on its surface to recover an acceptable hydraulic conductivity, but the initial high performances were not observed again. Clogging is the main cause of malfunction in vertical sand filters and is the cause of the oxygen transfer reduction in vertical flow filters [42]. This can explain the obtained results, since nitrification is very sensitive to oxygenation and *E. coli* removal is faster in aerobic conditions. The results of the present study partially agree with those of de Oliveira Cruz [43] who found clogging problems after only 4 months of operation in a sand filter. However, the full restoration of its function was recovered by scraping off a shallow layer on its surface.

Table 7. Average removal (%) (±standard deviation) of Hybrid CW 1 (HF: mulch, VF: sand) and Hybrid CW 2 (HF: mulch, VF: mulch).

	Hybrid CW 1	**Hybrid CW 2**
COD	73 ± 18	79 ± 10
BOD	96 ± 4	96 ± 4
TSS	98 ± 1	96 ± 5
Turbidity	98 ± 1	98 ± 2
TN	43 ± 28	55 ± 20
NH_4^+-N	52 ± 77	64 ± 28
TP	23 ± 26	24 ± 32
DRP	9 ± 36	14 ± 40
E. coli	99.0 ± 1.5	99.5 ± 0.7
Total coliforms	99.0 ± 2	99.5 ± 1

Nevertheless, Hybrid CW 2 (only mulch) remained unclogged throughout the whole experimental period. The only maintenance required was the addition of mulch because of the reduction in the height of the substrate, which occurred more rapidly at the beginning of the experimental period, falling by 20% in the first months. The substrate was added, and the subsequent reductions were minimal. Regarding clogging, woodchips were also preferred to sand as substrates for filters treating soiled dairy water [44].

3.3. Comparison of the General Performance of the Multistage HF CWs and Hybrid CWs

The selection of a treatment system is made based on its efficiency, but also by taking into account the economic cost and ease of construction and maintenance. Table 8 shows the concentrations of the different parameters analyzed in the effluent of the hybrid and multistage HF CWs. In the case of the latter, only those of the higher SRL period were included, with the goal of comparing results with similar SLRs. As can be observed, the effluent concentrations of HF10 are lower than those of the other systems for BOD, COD, TSS, turbidity, *E. coli* and total coliforms, but not significantly. On the other hand, the construction and maintenance of a HF CW is easier than that of a hybrid CW. Thus, the recommended system for the treatment of raw urban wastewater without clogging problems would be HF10.

Table 8. Concentrations in the effluents of the CWs studied.

	Hybrid CW 1	Hybrid CW 2	HF40	HF10
COD	120 ± 82	102 ± 48	110 ± 47	93 ± 53
BOD	11 ± 11	12 ± 15	10 ± 13	9 ± 2
TSS	6 ± 6	7 ± 9	6 ± 5	2 ± 1
Turbidity	2 ± 1	3 ± 2	8 ± 8	2 ± 1
TN	37 ± 14	28 ± 9	28 ± 15	30 ± 14
NH_4^+-N	20 ± 15	25 ± 17	24 ± 14	23 ± 12
TP	8 ± 4	9 ± 5	9 ± 5	8 ± 5
DRP	7 ± 3	7 ± 3	8 ± 4	8 ± 4
E. coli	2.9 (±4.3) × 10^4	1.8 (±2.4) × 10^4	2.4 (±5.8) × 10^4	9.1 (±1.8) × 10^3
Total coliforms	2.7 (±4.9) × 10^4	1.3 (±1.3) × 10^4	3.4 (±6.5) × 10^4	3.8 (±4.8) × 10^3

4. Conclusions

CWs are regarded as a sustainable wastewater treatment technology. However, the extraction and transport of gravel and sand used to build CWs have a non-negligible environmental impact that can be reduced by using residual materials such as agro-forest wastes. To improve our knowledge on the use of this alternative material, pilot scale multistage HF CWs and hybrid CWs, in which gravel and sand have been partially or totally replaced by palm mulch, have been compared.

In the multistage HF CWs, the effect of water level height and HLR was studied. The best results were obtained with the shallowest system and an SLR of 12 g COD/m^2d. Using sand in the VF stage of the hybrid CW provided remarkably good NH_4^+-N and *E. coli* removals, but only during the first months of operation. Then, clogging problems considerably reduced the performance, which was initially similar to that of the mulch-based hybrid CW, which, on the other hand, did not clog.

Considering the removals obtained and the fact that there were no clogging incidents in this study, in addition to the easier construction and operation of HF CWs, a mulch-based multistage HF CW would be a better option than a mulch-based hybrid CW for the economic and sustainable treatment of raw urban wastewater in small communities.

Author Contributions: The idea for this article was developed by J.A.H.-M. Sampling and analyses were performed by M.M.-A. The paper was written by J.A.H.-M., R.G.-A., P.G.-J., M.C.-A. and E.R. All authors have read and agree to the published version of the manuscript.

Funding: This research was partially funded by the Departments of Chemistry and Biology of the University of Las Palmas de Gran Canaria (Spain).

Acknowledgments: The authors acknowledge the financial and technical support of the University of Las Palmas de Gran Canaria (Spain).

Conflicts of Interest: The authors declare no conflict of interest.

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