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REMOVAL OF NITROGEN AND ORGANIC MATTER IN AN INTERMITTENTLY AERATED HORIZONTAL SUBSURFACE FLOW CONSTRUCTED WETLAND

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Abstract: The objective of this work was to evaluate the effect of intermittent aeration on the removal of ammonium (NH_4^+), total nitrogen (TN) and chemical oxygen demand (COD) in a horizontal subsurface flow constructed wetland (HFCW). Two HFCWs were studied, one non-aerated and another aerated intermittently, and both cultivated with *Typha latifolia*. Each system received 8.6 L d^{-1} of synthetic wastewater, resulting in 3 days of hydraulic retention time. The two systems displayed high efficiencies in the removal of COD (>90%); however, the intermittently aerated HFCW showed a higher rate of COD removal. Additionally, the removal of TN (48.8%) and NH_4^+ (57.7%) in the non-aerated system was limited, while in the aerated system, the efficiencies of TN (81.2%) and NH_4^+ (98.6%) removal were satisfactory. Thus, the results indicate that via intermittent aeration, nitrification and denitrification occur simultaneously, improving the performance of HFCW in the removal of TN and NH_4^+.

Keywords: Artificial aeration; constructed wetlands; nutrients removal; rural sanitation; wastewater treatment

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INTRODUCTION

The treatment of domestic wastewater in isolated communities is still a global issue, primarily in developing and/or low-income countries. The distance from urban centers and the lack of public policies often make it impossible to collect and treat effluents generated in these communities, resulting in damage to the environment and human health. Thus, in recent decades, several studies have been conducted on decentralized wastewater treatment systems, including subsurface flow constructed wetlands.

Subsurface flow constructed wetlands display high efficiency in the removal of pollutants, particularly solids, organic matter, and pathogens (Abou-Elela *et al.*, 2013; Weerakoon *et al.*, 2013). However, owing to the limited supply of dissolved oxygen (DO) the efficiency in the removal of total nitrogen (TN) in constructed wetlands is limited (Saeed & Sun, 2012; Wu *et al.*, 2014). Nitrification is the first limiting process in the removal of TN, since nitrifying bacteria depend upon the availability of DO (Fan *et al.*, 2013). Thus, to allow effective nitrification, continuous artificial aeration is implemented in constructed wetlands as an alternative to supplement oxygen (Butterworth *et al.*, 2013). Although this technique allows for nitrification, the high DO levels supplied to the system can alter the conditions of the anoxic/anaerobic medium to aerobic, inhibiting the denitrification process, which consequently limits the removal of TN (Maltais-Landry *et al.*, 2009).

On the other hand, some recent studies show that the use of intermittent artificial aeration is preferable, as it shifts the environmental conditions between aerobic and anoxic, allowing nitrification and denitrification to occur simultaneously, increasing the TN removal efficiency (Fan *et al.*, 2016; Hou *et al.*, 2017). However, most studies evaluate the effects of intermittent aeration in vertical subsurface flow constructed wetlands (VFCW) (Fan *et al.*, 2013; Wu *et al.*, 2016a; Liu *et al.*, 2019) and, therefore, there are few data in the world literature on the performance of horizontal subsurface flow constructed wetland (HFCW) with intermittent aeration. In this way, the objective of this study was to evaluate the effect of intermittent aeration on the removal of NH_4^+, TN and chemical oxygen demand (COD) in HFCW.

MATERIALS AND METHODS

Characteristics of the treatment system

The experiment was conducted in a protected environment (greenhouse) at the Center for Environmental Studies (CEA) - UNESP, Rio Claro, SP, Brazil. Two HFCWs were evaluated at a pilot scale (CW1: not aerated; and CW2: aerated intermittently). Each HFCW was comprised of a rectangular polypropylene water tank with an approximate capacity of 61 L (31.0 × 35.5 × 55.5 cm) filled with gravel #0 (Ø = 2.4–9.5 mm) with a porosity of 53%. The water tanks were filled with gravel up to a height of 30 cm and the level of the effluent was maintained at 25 cm, resulting in saturated volume of approximately 26 L. The systems were cultivated with *Typha latifolia*. CW2 was aerated intermittently for 3 h day^{-1} (1 h aeration/7 h without aeration) at an aeration rate of 10 L min^{-1}. Air was applied to the system using a tube diffuser installed at the bottom and along the longitudinal section of the polypropylene tank. The schematic diagram of the constructed wetlands is shown in **Fig. 1**.

System operation, origin of the influent and hydraulic retention time

The systems were operated continuously for 250 days from April 4, 2018 to December 7, 2018. The synthetic effluent was prepared using feed water with the addition of 387 mg L^{-1} sucrose, 188 mg L^{-1} $(NH_4)_2SO_4$, 18 mg L^{-1} of KH_2PO_4, 10 mg L^{-1} of $MgSO_4$, 10 mg L^{-1} of $FeSO_4$, and 10 mg L^{-1} of $CaCl_2$, as described in the works of Fan *et al.* (2016), Wu *et al.* (2015a) and Wu *et al.* (2016a). Using the peristaltic pumps, 8.6 L d^{-1} of synthetic effluent was applied to the HFCWs, resulting in 3 days of hydraulic retention time (HRT). Sampling points were distributed along the HFCW located at distances of 0, 18.5, 37 and 55.5 cm from the effluent entrance zone. The HRT of each of the sampling point would be proportional to its distance relative to the beginning of the HFCWs; therefore, the HRT adopted for each sampling point was 0, 1, 2, and 3 days, respectively.

Sample analysis

Five samples were collected at each sampling point to determine COD, NH_4^+, NO_3^-, NO_2^-, and pH.

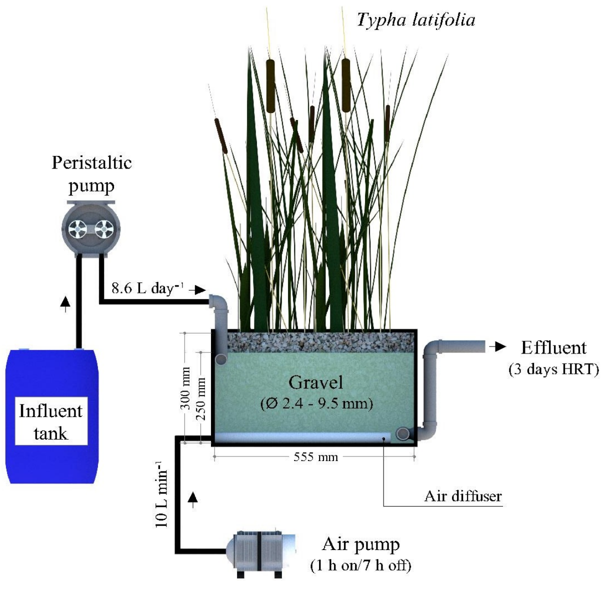

Fig. 1 Schematic diagram of the HFCWs used in the experiment.

Table 1. Characteristics of the influent and effluent of HFCWs and their removal efficiencies (mean ± SD, n = 5).

Parameters	Influent	System	Effluent	Removal (%)
COD (mg L^{-1})	388.8 ± 19.7	CW1	23.8 ± 6.6	93.9 ± 1.6
		CW2	9.4 ± 4.3	97.6 ± 1.1
TN (mg L^{-1})	40.5 ± 2.3	CW1	20.8 ± 2.0	48.8 ± 6.3
		CW2	7.6 ± 1.5	81.2 ± 4.4
NH_4^+ (mg L^{-1})	39.2 ± 2.3	CW1	16.5 ± 1.0	57.7 ± 4.5
		CW2	0.5 ± 0.5	98.6 ± 1.4
NO_3^- (mg L^{-1})	1.4 ± 0.3	CW1	4.3 ± 1.6	-
		CW2	7.0 ± 1.3	-
NO_2^- (mg L^{-1})	0.004 ± 0.002	CW1	0.02 ± 0.01	-
		CW2	0.05 ± 0.07	-
DO (mg L^{-1})	-	CW1	0.5 ± 0.2	-
		CW2	2.4 ± 1.6	-
Temperature (ºC)	23.1 ± 1.5	CW1	21.5 ± 1.7	-
		CW2	20.2 ± 1.8	-
pH	7.0 ± 0.3	CW1	7.1 ± 0.4	-
		CW2	6.3 ± 0.6	-

concentrations were estimated as the sum of the other forms of nitrogen. DO levels within the HFCWs were determined in situ using a portable Akso DO meter (model: DO Eco 1.00). All samples were collected, stored, and analyzed according to the methodologies described by APHA (2012).

Statistical analysis

The COD, NH4+, and TN data obtained in the HFCWs were compared via analysis of variance (ANOVA) in the statistical program Statgraphics (version 16. 2. 04).

RESULTS AND DISCUSSION

Table 1 shows the characteristics of the influent and effluent of HFCWs and their removal efficiencies.

DO levels

The average DO levels of the HFCWs over an aeration cycle are shown in **Fig. 2**. As expected and in agreement with published data (Fan *et al.*, 2013; Fan *et al.*, 2016; Uggeti *et al.*, 2016), CW1 presented an anaerobic condition with a mean DO level of 0.5 mg L^{-1} (range: 0.3-0.8). On the other hand, the system with intermittent aeration (CW2) had a higher average DO level: 2.4 mg L^{-1} (range: 0.9-5.4). An increase in DO levels in the CW2 system occurs during aeration, and the maximum level is obtained at the end of the aeration phase. After aeration, during the 7 h resting period, the DO level decreases until the observed minimum value is reached. Similar DO profiles in intermittently aerated constructed wetlands were also described by Fan *et al.* (2013) and Wu *et al.* (2016b).

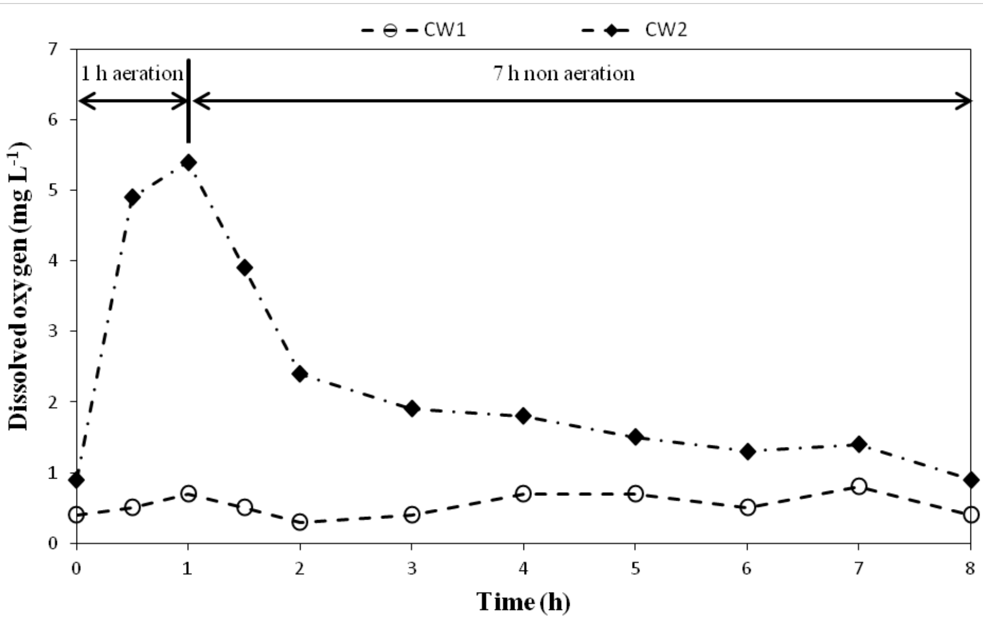

Fig. 2 Average levels of dissolved oxygen of HFCWs along an aeration cycle.

COD removal

CW1 and CW2 displayed a COD removal of 93.9 and 97.6% respectively. COD removal difference between the systems was small; the mean COD concentration in the CW1 and CW2 effluents was 23.8 mg L^{-1} 9.4 mg L^{-1}, respectively, that is, a removal difference of only 3.7% was observed. Non-aerated HFCWs may exhibit high COD removal performance, as described by Abou-Elela *et al.* (2013), Caselles-Osorio *et al.* (2017) and Liu *et al.* (2018) who obtained efficiencies of 91.5, 80-91, and >85%, respectively. Thus, depending on the operating conditions such as HRT and effluent characteristics, the efficiency of organic matter removal in non-aerated HFCWs might be similar to that of the aerated systems. For example, Uggeti *et al.* (2016) reported a difference of only 1% in COD removal efficiency between a non-aerated system and another intermittently aerated system.

Although the final COD levels were similar, there was a clear difference in efficiency of removal of COD along the systems (**Fig. 3**). With 1 day of HRT, the average COD level in CW2 decreased to 23 mg L^{-1} and in CW1 it decreases to 119 mg L^{-1}; these concentrations correspond to 94% and 69% removal efficiency, respectively. These data are similar to those reported by Wu *et al.* (2016a), in which the authors evaluated VFCWs and in only 12 hours obtained in intermittent aerated systems and in non-aerated systems COD removal efficiencies of, respectively >88% (<50 mg L^{-1}) and 76-82% (75-100 mg L^{-1}). These results indicate that the use of intermittent aeration in constructed wetlands increases the organic matter removal rate. The increase in the COD removal rate in intermittently aerated HFCWs may be associated with the stimuli in the direct biological removal pathways (respiration/fermentation) and also to the removal of organic carbon in the nitrate reduction (part of denitrification) process (Saeed & Sun, 2012).

Nitrogen removal

According to Saeed & Sun (2012), the transformation and removal of nitrogen in subsurface flow constructed wetlands occurs via both classical pathways (ammonification, nitrification/denitrification, plant absorption, biomass assimilation, and volatilization of ammonia) and newly discovered pathways (partial nitrification/denitrification, ANAMMOX, and Canon process). Maltais-Landry *et al.* (2009) observed that TN removal is mainly associated with four processes: 1) plant absorption; 2) storage in the sediment; 3) partial nitrification/denitrification; and 4) complete nitrification/denitrification. However, complete nitrification/denitrification is considered to be the main mechanism of TN removal, accounting for 47% to 96% of nitrogen removal (Lin *et al.*, 2002; Maltais-Landry *et al.*, 2009; Chen et al., 2014).

With regard to the plants, a few studies have demonstrated that macrophytes are responsible for up to 34.3% of the removal of TN (Maltais-Landry *et al.*, 2009; Wu *et al.*, 2013; Chen *et al.*, 2014; Liu *et al.*, 2019). The results obtained by Maltais-Landry *et al.* (2009) show that, in continuously aerated subsurface flow constructed wetlands, the species *Typha angustifolia* was responsible for the removal of approximately 135 mg N m^{-2} d^{-1}, which represents 7.6% of the rate of application of nitrogen in the present study (1768 mg N m^{-2} d^{-1}). Other authors (Wu *et al.*, 2013) estimated rates of removal of 40 mg N m^{-2} d^{-1} by *Typha orientalis,* representing only 2.3% of the nitrogen applied under our conditions. Therefore, in the analysis of the results obtained in the present study, we considered that complete nitrification/denitrification

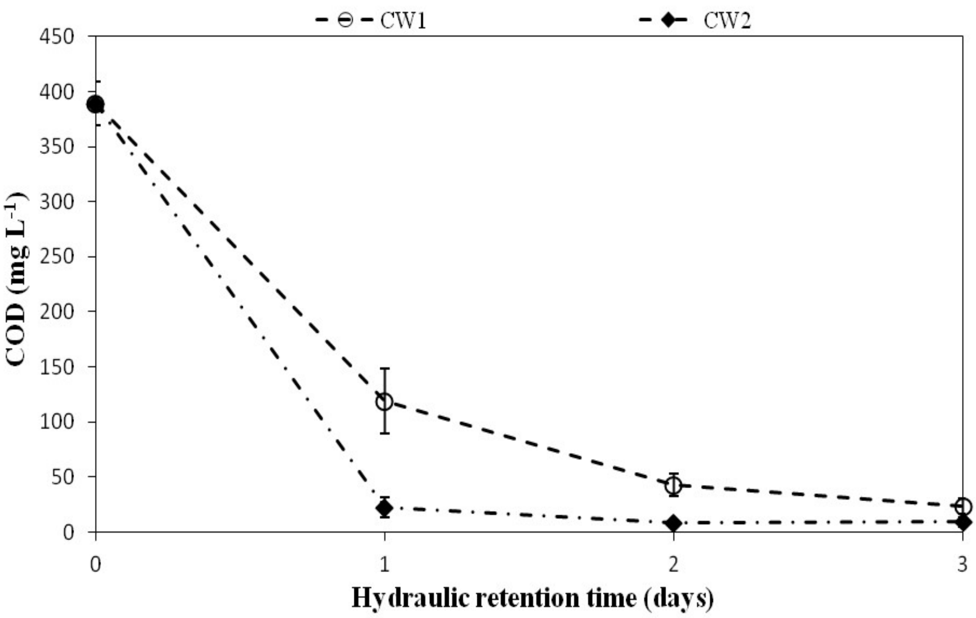

Fig. 3 Average concentrations of COD along the HFCWs.

was the main route of TN removal.

The average removal of NH_4^+ and TN in CW1 was 57.7% and 48.8%, respectively, confirming that conventional HFCWs (without aeration) have limitations in the removal of nitrogen. For example, Hua et al. (2017) reported the removal of NH_4^+ and TN in the range of 20.7%–66,9% and 38.1%–51.6%, respectively, and Wu *et al.* (2015b) reported values of 32%–61% for NH_4^+ removal and 13%–44% for TN removal (Wu *et al.*, 2015b). On the other hand, CW2 presented satisfactory efficiencies in the removal of NH_4^+ (98.6%) and TN (81.2%). **Fig. 4** presents the dynamics of transformation of nitrogen in the two HFCWs.

Note that the concentrations of NH_4^+ declined rapidly in CW2, reaching average values of 7.1, 1.2, and 0.5 mg L^{-1} with 1, 2, and 3 days of HRT, respectively. In contrast, CW1 had a lower efficiency in the removal of NH_4^+, with a final average concentration of 16.5 mg L^{-1}. These results were anticipated, as this system presented anaerobic conditions (DO = 0.3-0.8 mg L^{-1}) that consequently limited nitrification. The removal of NH_4^+ in CW2 exceeded 98.6%, corroborating the findings of Li *et al.* (2014), who also reported a near total removal of NH_4^+. Moreover, the removal of NH_4^+ in CW2 was 40.9% higher than that observed in CW1 (57.7% vs 98.6%). Likewise, increases in the NH_4^+ removal efficiency between intermittently aerated systems and non-aerated systems of 46%, 65% and 45%–88% have been reported by Uggetti *et al.* (2016), Fan *et al.* (2016), and Liu *et al.* (2019), respectively. Finally, these results show that artificial aeration creates adequate conditions for the effective occurrence of nitrification in HFCWs.

The CW2 wetland presented lower final concentrations of TN (7.6 mg L^{-1}) than the non-aerated system (20.8 mg L^{-1}), and the average removal efficiencies of TN were equal to 48.8% in CW1 and 81.2% in CW2. Therefore, these data show that the CW2 was 32.4% more efficient in the removal of TN than CW1. These results are higher than those obtained

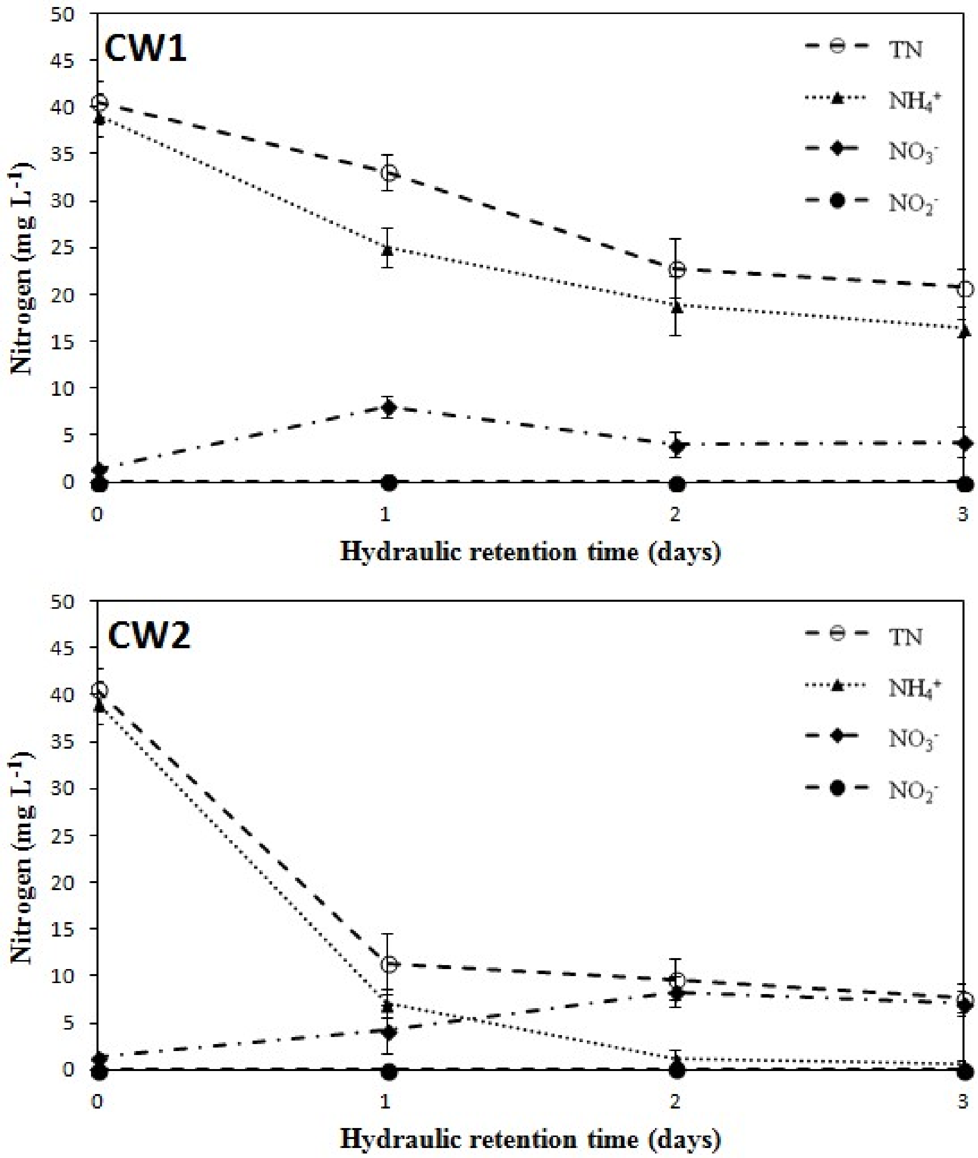

Fig. 4 Nitrogen transformation dynamics in the two HFCWs.

by Uggetti *et al.* (2016) (improvement of 23%) and lower than those reported by Liu *et al.* (2019) (improvements of 37% to 81%). These findings suggest that intermittent aeration permitted the occurrence of nitrification and denitrification simultaneously in HFCWs, thus increasing the efficiency of TN removal.

The average concentrations of NO_2^- registered during the experiment were very low (<0.3 mg L^{-1}), indicating that the conversion of NO_2^- to NO_3^- occurs rapidly. The CW1 effluent was composed mainly of NH_4^+ owing to limited nitrification. The effluent from CW2 in turn was composed almost exclusively of NO_3^-, indicating that nitrification was effective. However, although the data indicate that nitrification and denitrification occur simultaneously, the accumulation of NO_3^- in these effluents indicates that complete denitrification did not occur, probably owing to the lack of organic carbon. Wu *et al.* (2016a) also reported that the lack of carbon prevented the complete denitrification of effluent in an intermittently aerated VFCW.

Data statistical analysis

The ANOVA performed on COD, NH_4^+, and TN data indicated a statistically significant difference ($p<0.05$) between the CW1 and CW2 ($P<0.0000$), i.e., the use of intermittent aeration changed the removal efficiency of COD, NH_4^+, and TN in the HFCW.

CONCLUSIONS

Intermittent aeration allows the simultaneous occurrence of nitrification and denitrification, thus improving the performance of HFCWs in the removal of TN and NH_4^+. Additionally, intermittently aerated HFCWs also display a higher rate of removal of COD compared to non-aerated systems. However, the lack of organic carbon may limit the denitrification in systems with intermittent aeration. Finally, our results show that intermittently aerated HFCWs show high efficiency in the removal of COD (97.6%), TN (81.2%), and NH_4^+ (>97%) and, therefore, are a viable option for the effective treatment of wastewater in isolated communities.

However, some limitations of the present study should be highlighted: 1) Synthetic effluent: although allowing the evaluation of nitrogen removal dynamics in HFCWs, the synthetic effluent is composed of simple substances that can be easily biodegraded and, therefore, do not adequately represent the complexity of a real effluent; 2) DO concentrations: the values obtained were determined on the center of the HFCWs at 10 cm deep, that is, in a single location above the air diffuser and thus, the concentrations obtained may not represent adequately the general distribution of DO in the systems under study. Thus, to consolidate the understanding of the effects of intermittent aeration in HFCW, it is necessary to conduct studies with non-synthetic effluents and that also evaluate more consistently the spatial and temporal distributions of dissolved oxygen.

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