



Effect of hydraulic retention time on chemical oxygen demand and total nitrogen removal in intermittently aerated constructed wetlands

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ABSTRACT

This study evaluated the effect of hydraulic retention time on chemical oxygen demand (COD) and total nitrogen (TN) removal in an intermittently aerated constructed wetlands. Two horizontal subsurface-flow constructed wetlands were used: one without aeration and the other aerated intermittently (1 hour with aeration/7 hours without aeration). Both systems were evaluated treating domestic wastewater produced synthetically. The flow rate into the two CWs was 8.6 L day⁻¹ having a hydraulic retention time of 3 days. The results show that the intermittently aerated constructed wetland were highly efficient in removing COD (98.25%), TN (83.60%) and total phosphorus (78.10%), while the non-aerated constructed wetland showed lower efficiencies in the removal of COD (93.89%), TN (48.60%) and total phosphorus (58.66). These results indicate, therefore, that intermittent aeration allows the simultaneous occurrence of nitrification and denitrification processes, improving the removal of TN in horizontal subsurface-flow constructed wetlands. In addition, the use of intermittent aeration also improves the performance of constructed wetlands in removing COD and total phosphorus.

Keywords: ecotechnology, effluent treatment, nutrients removal.

Remoção de DQO e Nitrogênio Total em alagados construídos com aeração intermitente em relação ao tempo de detenção hidráulica

RESUMO

O objetivo do presente trabalho foi avaliar o efeito do tempo de detenção hidráulica na remoção da demanda química de oxigênio (DQO) e nitrogênio total (NT) em alagados construídos aerados intermitente. Foram utilizados dois alagados construídos de fluxo subsuperficial horizontal: um sem aeração e outro aerado intermitentemente (1 hora com aeração/7 hora sem aeração). Ambos os sistemas foram avaliados tratando água residuária doméstica sintética. Cada sistema recebeu uma vazão de 8,6 L dia⁻¹, resultando em tempo de detenção de 3 dias. Os resultados mostram que o alagado construído aerado intermitentemente apresentou elevada eficiência na remoção de DQO (98,25%), NT (83,60%) e fósforo total



(78,10%), enquanto que o sistema sem aeração apresentou menor eficiência na remoção de DQO (93,89%), NT (48,60%) e fósforo total (58,66%). Esses resultados indicam, portanto, que a aeração intermitente permite a ocorrência simultânea dos processos de nitrificação e desnitrificação, melhorando a remoção de NT em alagados construídos de fluxo subsuperficial horizontal. Além disso, o uso de aeração intermitente também melhora o desempenho de alagados construídos na remoção de DQO e fósforo total.

Palavras-chave: ecotecnologia, remoção de nutrientes, tratamento de efluente.

1. INTRODUCTION

Constructed wetland (CW) is a simple technology which not only mimics the functions of a natural wetland but is easy to operate and maintain and cost-effective (Al-Isawi *et al.*, 2015; 2017). In recent decades, it has garnered increasing interest in the treatment of several types of wastewater rich in biodegradable organic materials (Matos *et al.*, 2010a). Its main component is the support medium, which can be composed of soil, sand, gravel, plant species characteristic of wetland environments, and microorganisms associated with these elements. Its design is generally aimed at maximizing the wastewater treatment efficiencies (Fia, 2009).

Macrophytes play an important treatment role in CWs, especially in systems with large amounts of organic matter and ammonia (Ciria *et al.*, 2005). The most commonly used plants are cattail (*Typha* sp.), common reed (*Phragmites* sp.), and bulrush (*Scirpus* sp.) (Matos and Lo Monaco, 2003). However, plant characteristics such as local climate adaptability, photosynthetic rate, oxygen transport capacity, pollutant absorption capacity, resistance to pests and diseases, and root system development should be considered.

According to Suliman *et al.* (2004), the main variables considered while designing CWs are the hydraulic retention time (HRT), width, height and length, and hydraulic application rate. The hydraulic load and HRT significantly influence the efficiency in CWs (Wu *et al.*, 2015). Low HRT can be associated with inefficient denitrification, with a longer retention time required for nitrogen removal compared to that required for organic matter removal (Lee *et al.*, 2009).

The present study therefore evaluated the dynamics of chemical oxygen demand (COD) and total nitrogen (TN) removal in relation to the HRT in intermittently aerated horizontal subsurface-flow constructed wetlands.

2. MATERIALS AND METHODS

The experiment was conducted in a green-house environment located at the Center for Environmental Studies (Centro de Estudos Ambientais - CEA) of the Institute of Geosciences and Exact Sciences - UNESP, Rio Claro, São Paulo, Brazil. The climate of the region is defined as Cwa (Köppen): warm temperate climate (mesothermal) with rainy summer and dry winter, with the temperature of the hottest month averaging over 22°C. The average annual precipitation and average annual temperatures are 1366.8 mm and 21.6°C, respectively.

Two horizontal subsurface-flow constructed wetlands were evaluated: one without aeration (CW1) and the other aerated intermittently (CW2). Each treatment system was composed of a rectangular polypropylene water tank with a capacity of ~61 L and dimensions of 310 × 355 × 555 mm. The media used was gravel ($2 \leq \emptyset \leq 9$ mm) with a porosity of 53%, having a height of 30 cm and saturated by the effluent up to a height of 25 cm with a useful volume of 26 L. Two 32-mm PVC perforated tubes were installed in the longitudinal section of each constructed wetland with 18.5-cm spacing between them so that wastewater samples could be collected in the system.

CW2 was aerated intermittently for 3 h day⁻¹ (1 h aeration/7 h non aeration) at an average aeration rate of 20 L min⁻¹, resulting in an oxygen surface application rate of ~101.5 L m⁻² min⁻¹. The air was applied to the system using an aquarium air compressor and a tubular air diffuser installed inside the constructed wetland support material.

The aquatic macrophyte species used in the CWs was cattail (*Typha latifolia*), which was collected from a natural swamp and pruned at half height. The replanting of macrophytes in the CWs was carried out on March 07, 2018, and the planting density was approximately 15 plants per m², that is, each system was cultivated with three plants. During the period of macrophyte fixation, the constructed wetlands were saturated with water and only on April 4, 2018, after the fixation of the plant species, was the application of wastewater started.

Figure 1 shows the schematic diagram of the CWs.

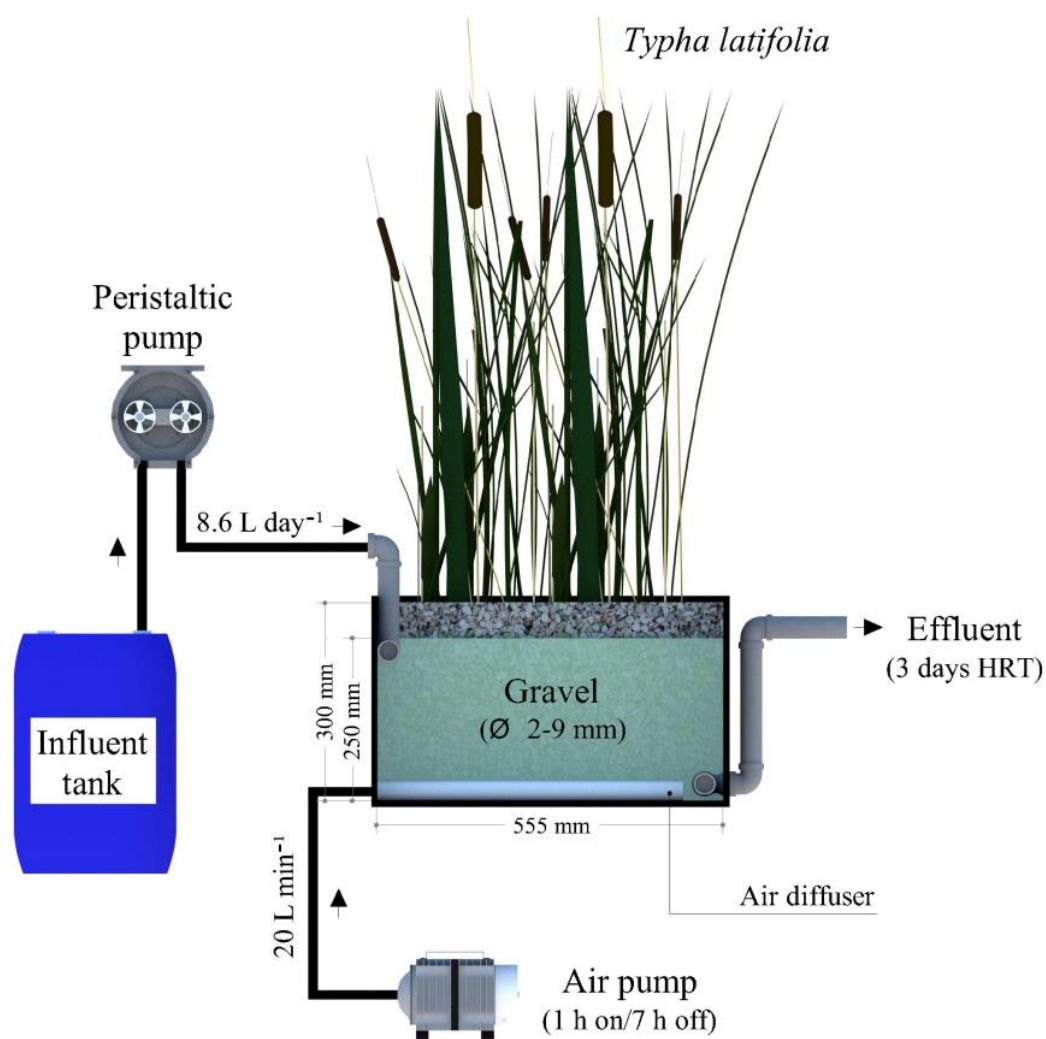


Figure 1. Schematic diagram of the CWs used in the experiment.

The CWs were operated from April 4, 2018 to December 7, 2018, that is, the systems were kept in operation for 250 consecutive days. The experiment was conducted using synthetic wastewater prepared with tap water with the addition of C₁₂H₂₂O₁₁ (sucrose), (NH₄)₂SO₄ (ammonium sulfate), KH₂PO₄ (monopotassium phosphate), MgSO₄ (magnesium sulfate), FeSO₄ (iron(II) sulfate), and CaCl₂ (calcium chloride), equal to the synthetic effluent used in Wu *et al.* (2015), Fan *et al.* (2016), and Wu *et al.* (2016) works. Table 1 shows the compounds and their respective concentrations used in the synthetic effluent.

Table 1. Composition of the synthetic primary effluent.

Chemical compound*	Concentration (mg L ⁻¹)
C ₁₂ H ₂₂ O ₁₁	386
(NH ₄) ₂ SO ₄	188
KH ₂ PO ₄	18
MgSO ₄	10
FeSO ₄	10
CaCl ₂	10

* Chemical compounds were diluted in tap water to prepare the synthetic effluent.

Source: Wu *et al.* (2015), Fan *et al.* (2016) and Wu *et al.* (2016).

The effluent was applied to the CWs using peristaltic pumps, with a hydraulic loading rate of 0.044 m³ m⁻² d⁻¹ (8,6 L d⁻¹). Therefore, considering the saturated volume of the CWs and the hydraulic loading rate, it was estimated that the HRT was approximately 3 days. However, as perforated tubes were installed along the longitudinal section in each water tank, it was possible to evaluate the concentration of pollutants according to the distance from the entry point of the influent and, consequently, according to different HRT: 0, 1, 2, and 3 days.

In the period from November 1, 2018 to December 7, 2018, five effluent samples were collected from each sampling point, and following parameters were analyzed: COD, TN, ammonium (NH₄⁺), nitrite (NO₂⁻), nitrate (NO₃⁻), total phosphorus (TP), potential of hydrogen (pH), and temperature. The analyses were performed at CEA-UNESP, Rio Claro Campus, according to the methodologies described in *Standard Methods* (APHA *et al.*, 2012).

3. RESULTS AND DISCUSSION

The characteristics of the influent and effluent throughout the CWs and their respective removal efficiencies are shown in Table 2.

Table 2. Physico-chemical characteristics of the influent and effluent along the CWs and their respective removal efficiencies (ε).

Parameter	Effluent	HRT (d) CW1			HRT (d) CW2		
		1	2	3	1	2	3
COD (mg L ⁻¹)	388.±19.7	119±29.9	43±10.4	23.8±6.6	42.2±15	9.6±5.6	6.8±2.9
ε (%)	-	69.37±8.0	88.98±2.5	93.89±1.6	89.24±3.6	97.51±1.5	98.25±0.8
NH ₄ ⁺ (mg L ⁻¹)	39.16±6.2	25.08±5.6	18.86±8	16.48±2.3	6.52±3.3	0.82±1.9	0.58±0.6
ε (%)	-	35.64±7.9	51.54±9.7	57.72±4.5	83.23±3.6	97.89±2.1	98.52±0.7
NO ₂ ⁻ (mg L ⁻¹)	0.0042	0.043±0.04	0.03±0.05	0.019±0.01	0.059±0.04	0.027±0.02	0.048±0.08
NO ₃ ⁻ (mg L ⁻¹)	1.36±0.3	8.02±1.2	3.96±1.3	4.26±1.6	6.98±2.6	7.9±1.4	5.98±1.1
TN (mg L ⁻¹)	40.52±2.1	33.14±3.3	22.85±5.0	20.76±3.1	13.56±3.2	8.75±1.8	6.61±1.16
ε (%)	-	17.81±8.8	43.40±9.3	48.60±6.3	66.42±7.2	78.23±4.7	83.60±3.2
TP (mg L ⁻¹)	3.88±0.48	-	-	1.58±0.56	-	-	0.89±0.69
ε (%)	-	-	-	58.66±17.8	-	-	78.1±16.36
pH	6.96±0.25	-	-	7.09±0.45	-	-	6.68±0.49
S (μS cm ⁻¹)	562±19.3	-	-	397±15.6	-	-	578.5±72.8
T (°C)	23.0±1.5	-	-	21.5±1.7	-	-	20.3±1.8

All samples were collected and analyzed between November 1, 2018 to December 7, 2018 (Mean ± SD, n = 5).

3.1. Chemical Oxygen Demand

Figure 2 shows the average COD concentrations along the CWs. The removal efficiencies of both systems within 3 days of HRT was high, with similar removal efficiencies of 93.89% for CW1 and 98.25% for CW2. Similar values were reported in the literature, for example, Matos *et al.* (2010b) obtained, with HRT of ~4.8 days, COD removal efficiencies ranging from 71% to 96% in non-aerated horizontal subsurface-flow constructed wetlands treating hog wastewater; Brasil *et al.* (2005) obtained, in non-aerated horizontal subsurface-flow constructed wetlands treating domestic wastewater, average COD removal efficiencies of 84% and 93% for HRTs of 1.9 and 3.8 days, respectively; and Wu *et al.* (2016) obtained, in intermittent-aerated vertical subsurface-flow constructed wetlands treating synthetic wastewater, COD removal efficiencies >95% for HRT of 3 days.

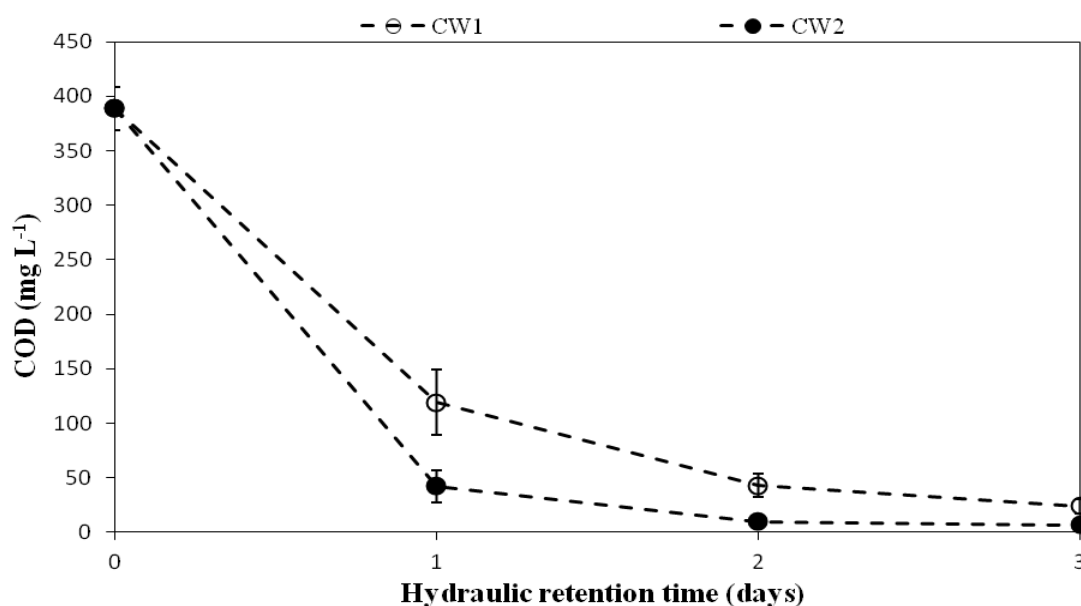


Figure 2. Average COD concentrations along both CWs.

However, the removal efficiency of CW2 for HRT of 1 day was $89.24 \pm 3.57\%$, while the value obtained for CW1 was $69.37 \pm 8.04\%$ (Table 2). The results of the present work are similar to the data reported by Wu *et al.* (2016), in which the authors evaluated vertical subsurface-flow constructed wetlands and with a HRT of 12 hours obtained in intermittent aerated systems and in non-aerated systems COD removal efficiencies of >88% and 76-82%, respectively. The higher efficiency of CW2 compared to that of CW1 after 1 day HRT can be attributed to the intermittent artificial aeration, which stimulates the increase of biological activity, thereby contributing to a high efficiency of COD removal, nitrification, and denitrification. This is because during aeration periods aerobic heterotrophic microorganisms use organic matter as a source of energy, leading to COD removal. Moreover, during non-aeration periods (anoxic), denitrifying heterotrophic microorganisms remove COD (Wendling, 2017), indicating the occurrence of denitrification.

Our results indicate, therefore, that the use of intermittent aeration improves the COD removal performance of horizontal subsurface-flow constructed wetlands, corroborating the results reported by Wu *et al.* (2015), in which the authors achieved COD removal efficiencies ranging from 96.42 to 98.49% in intermittently aerated CWs, and ranging from 62.68 to 85.49% in non-aerated systems.

3.2. Nitrogen removal

The dynamics of nitrogen removal along the CWs can be seen in Figure 3. The results obtained in the present study show that the final average concentration in CW1 was

16.48 mg L⁻¹ and 20.76 mg L⁻¹ for NH₄⁺ and TN, respectively. Thus, the mean final removal efficiency (Table 2) of CW1 is 57.7 ± 4.5% and 48.6 ± 6.3% while in CW2, it is 98.5 ± 0.7% and 83.6 ± 3.2% for NH₄⁺ and TN, respectively. Although the removal efficiencies of CW1 for NH₄⁺ and TN with only 1 day of HRT are 35.64 ± 7.9% and 17.8 ± 8.8%, respectively, CW2 has a removal efficiency of 83.23 ± 3.6% and 66.4 ± 7.17% with the same HRT. That is, the efficiency levels of the aerated system increase by ~47.6% for NH₄⁺ and by 48.6% for TN, thereby surpassing the final removal efficiency rates of CW1 in the first 24 hours of treatment.

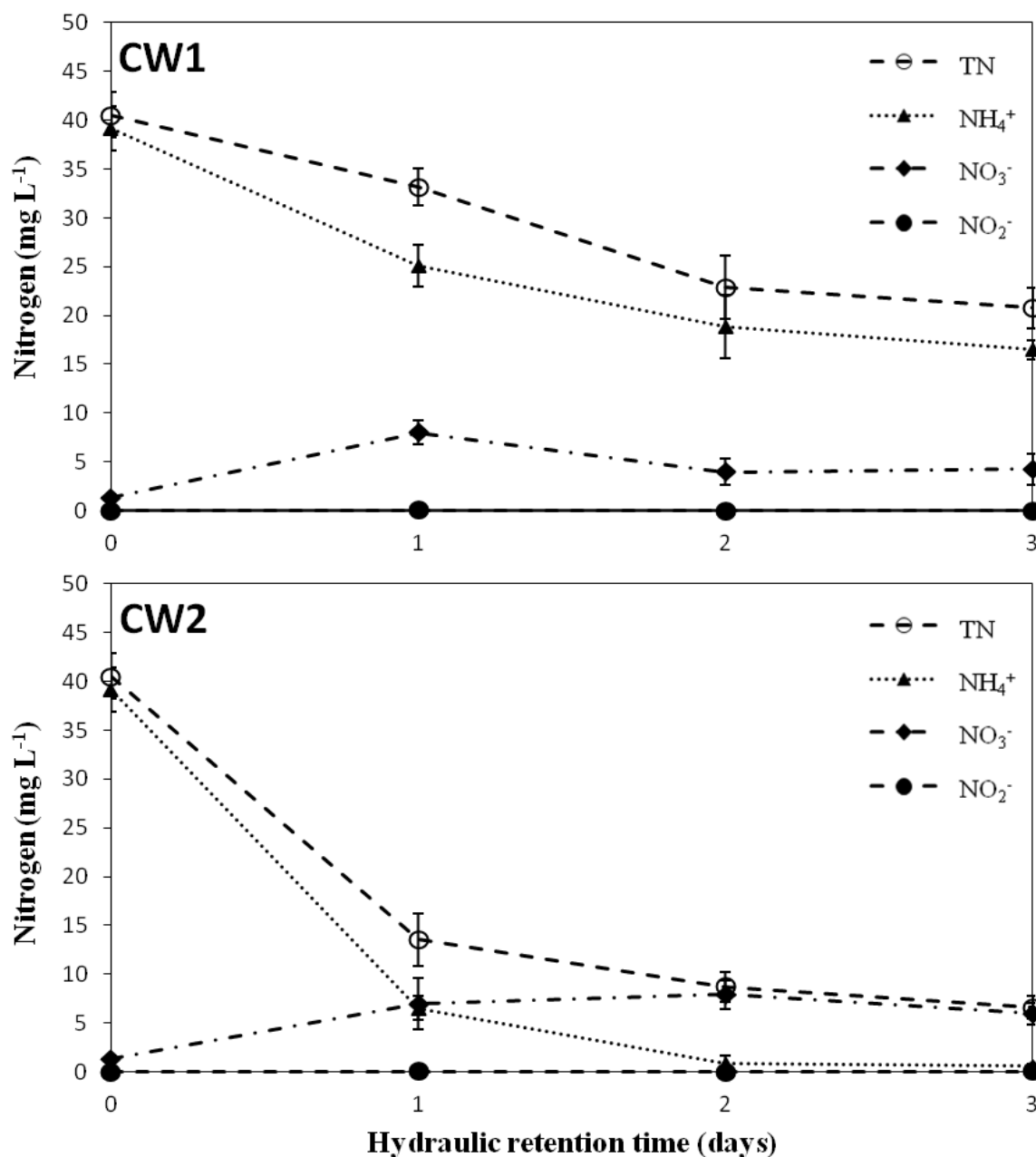


Figure 3. Dynamics of nitrogen removal along the CWs.

The TN and NH₄⁺ removal efficiencies in the CW2 can be considered satisfactory, since the average values obtained are higher than those reported in the literature for non-aerated constructed wetlands. Ramos *et al.* (2017) reported average TN removal efficiencies of 38-48% in systems planted with *Polygonum punctatum* and *Chrysopogon zizanioides* with HRT of 3.2 days. It should be noted that these removal efficiencies were exceeded in only 24 hours in our

CW2 system. The results obtained by Wang *et al.* (2016) showed a mean removal efficiency of 46% for NH_4^+ and 43% for TN in CWs planted with *Canna indica* L. with 5-day HRT. The removal efficiencies obtained in CW2 for both parameters in 1 day of HRT were significantly higher than the values obtained by those authors. Therefore, our results indicate that intermittent aeration has enhanced the aerobic environment for nitrifiers and, in addition, the cattails have aerenchyma tissues that transport oxygen from above-ground parts into the below-ground parts, that is, the macrophyte roots released oxygen in the soil media, also increasing aerobic microbial activity. Both these mechanisms improved the NH_4^+ and TN removal through nitrification and also plant uptake.

Due to the predominantly anaerobic conditions non-aerated subsurface-flow CWs have limitations in nitrogen removal, since the low availability of oxygen in the substrate compromises the nitrification process (Wu *et al.*, 2015). The results obtained in the present study confirm these limitations, as can be seen in the NH_4^+ and TN average removal efficiencies achieved in the CW1 system, which were 57.7% and 48.8%, respectively. Similar results were also reported in the study developed by Hua *et al.* (2017), in which the authors obtained NH_4^+ and TN removal efficiencies in the ranges of, respectively, 20.7-66.9% and 38.1-51.6%.

Moreover, as stated above, the intermittently aerated system (CW2) showed high efficiency in the removal of NH_4^+ and TN, with values of, respectively, 98.5 and 83.6%. These results indicate, therefore, that the use of intermittent aeration allows the simultaneous occurrence of nitrification and denitrification, improving nitrogen removal performance of CWs. These results are similar to those obtained by Fan *et al.* (2016) and Wu *et al.* (2016). However, it is observed that the remaining fraction of nitrogen in the CW2 effluent is mainly composed of NO_3^- . This fact indicates that complete denitrification has not occurred, probably due to the lack of organic matter (carbon). The occurrence of partial denitrification of the effluent due to the absence of organic carbon was also observed by Wu *et al.* (2016).

Vymazal and Kröpfelová (2008) concluded that, in most cases, the removal of TN in non-aerated horizontal subsurface-flow constructed wetlands is small (40-50%), as these systems do not provide simultaneous aerobic and anaerobic conditions for the occurrence of nitrification and denitrification, respectively. On the other hand, the results of the present study show that the use of intermittent aeration allows nitrification and denitrification to occur simultaneously, increasing CWs performance in the removal of TN (CW2 was 35% more efficient than CW1).

3.3. Total Phosphorus

The results of the phosphorus analysis can be observed in Figure 4. Although they were considered satisfactory, since constructed wetlands are efficient in removing biochemical oxygen demand (BOD) and total suspended solids (TSS), phosphorus removal is still considered a challenge (De Rozari *et al.*, 2015). Moreover, the results partly contradict the results of Ayaz *et al.* (2003) and Vymazal (2007); the former study stated that the phosphorus removal efficiency varies between 20 and 70%, depending on the plant used and the feeding regime, while according to the latter, total phosphorus removal varies between 40 and 60% among all wetland systems.

The final phosphorus concentration in the CW1 was 1.58 mg L^{-1} and in the CW2 was 0.89 mg L^{-1} , representing average removal efficiencies of, respectively, 58.66% and 78.1%. Although these results are much higher than those obtained by Mendonça *et al.* (2012), who obtained average removal efficiencies of 33.6% and 34.3% in CWs planted with cattails and with 2 days of HRT, they are similar to the results found by Ramos *et al.* (2017) who obtained, with 3.2 days of HRT, removal efficiencies of $51 \pm 24\%$, $69 \pm 22\%$ and $45 \pm 19\%$ in CWs without vegetation, planted with *P. punctatum*, and planted with and *C. zizanioides*, respectively. Fia *et al.* (2017) also reported high phosphorus removal efficiencies (73-78%) in the treatment of swine wastewater in CWSs planted with cattails with HRT of 11.8-12 days.

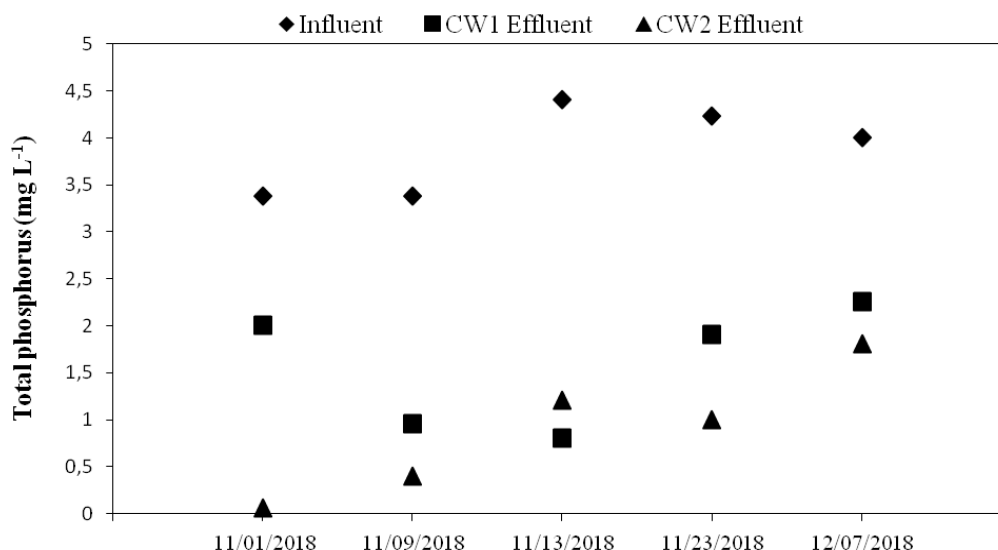


Figure 4. Total phosphorus input and output concentrations for both CWs.

It is noteworthy that the CW2 system was on average ~20% more efficient in removing TP than the CW1 system. This difference can be attributed to the use of intermittent aeration, as it promotes an increase in biological activity, including polyphosphate-accumulating organisms (PAOs). In addition, intermittent aeration also promotes the mixing of the effluent, a fact that intensifies the contact between phosphorus and the substrate and, consequently, improves the absorption of this nutrient by the support material. Shi *et al.* (2017) also reported significant improvements (18-46%) in the removal of phosphorus in intermittently aerated vertical subsurface-flow constructed wetlands compared to non-aerated systems, both filled with gravel. The results obtained by these authors indicate that intermittent aeration increased TP accumulated by support material storage and also by microbial uptake.

According to Vymazal (2007), phosphorus removal can be mainly associated with physical processes such as adsorption, precipitation, accumulation of organic matter in the substrate, microbial activity, and plant absorption. According to Greenway and Woolley (2001) plants play a significant role in removing phosphorus, being responsible for the absorption of up to 80% of this nutrient in CWs. However, other studies suggest that plants are responsible for only 4.81–22.33% of phosphorus uptake (Wu *et al.*, 2013; Dzakpasu *et al.*, 2015) and therefore have no significant role in TP removal.

It is also important to note that the age of CWs has an impact on the phosphorus removal process. According to Fia *et al.* (2017) and Sousa *et al.* (2004), newly constructed wetlands have greater capacity to adsorb and precipitate phosphorus-based compounds and, over time, the system ends up saturated with these compounds, reducing their removal efficiency.

4. CONCLUSIONS

Based on the results obtained, it can be concluded that intermittent aeration increased the efficiency of organic matter removal (represented by COD) and nutrient removal mainly in the nitrification process. The best average values of COD (98.25%), NH₄⁺ (98.52%), TN (83.6%), and TP (78.1%) removal were obtained in the system using intermittent aeration, which demonstrates the importance of aeration in nitrogen removal by creating excellent aerobic and anaerobic conditions alternately; this also contributes to the removal of organic matter that is consumed in the denitrification process. It was observed that although intermittent aeration in a HRT greater than 3 days does not considerably improve the performance of COD removal as opposed to the results observed in the removal of the nitrogen species, it is an alternative that

significantly increases the efficiency in the treatment of organic matter and nutrients in a HRT of less than 3 days. This suggests that it is an appropriate alternative for obtaining high performance in constructed wetland systems.

5. ACKNOWLEDGMENTS

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