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Kinetic behavior and modeling the space distribution of basic pollutants in a subsurface flow constructed wetlands with *Pontederia cordata*

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Abstract: Water pollution is one of the most serious environmental problems nowadays, so it is relevant to study improvements in the wastewater treatment system. In this work, the kinetics of experimental degradation of basic pollutants were evaluated, as well as the behavior of their spatial distribution, within a subsurface flow constructed wetland (SSFCW) with *Pontederia cordata*. To achieve this, a factorial design was used with five treatments factors at different planting distances within the wetland (0, 0.42, 1.25, 2.08 and 2.4 m) with three replicates and nine sampling points. The experimental reactors showed a hydraulic retention time of 4.5 days, treating 185.1±58.6 L/day of wastewater, with river gravel as support medium with a volume of 0.66±0.05 m^3, porosity n=56±3.5%, density of 2670±19.3 kg/m^3, allowing a water volume of 0.1±0.05 m^3. In the evaluation phase, a k of -0.37 $days^{-1}$ was obtained, removing 81.8% of BOD_5 and 70% turbidity. With the modeling of the spatial distribution, it was demonstrated that the biomass (microorganisms) adhered to the support medium, as well as the BOD_5, have a significant decrease related to the distance within the SSFCW throughout the reactor. The contaminant removal efficiency is presented from the distance 2.08 to 2.4 m in length within the experimental wetland.

Keywords: BOD_5, Phytoremediation, Wastewater, Treatment system, Macrophytes.

Introduction

There is a growing need to improve and implement wastewater treatment systems. Systems that are economical, easy to operate, adaptable to the weather conditions in the study area and that are also environmentally sustainable. Constructed wetlands (CW) are sewage phytodepuration systems, that consists in the development of rooted macrophytes crop on a bed of gravel. The action of the macrophytes makes possible a series of complex physical, chemical and biological interactions through which the tributary wastewater is progressively and slowly purified (Delgadillo et al., 2010).

Even though, CW technology is not new worldwide as plant species that eliminate pathogens have been studied (Calheiros et al., 2017), domestic wastewater pollutants (Gallegos et al., 2018; Tran et al., 2019), nitrogen (Quintero et al., 2021) and even variants in construction designs (García, López y Torres 2019). Highlighting among these the importance of determining the kinetics in this type of systems to know a priori the concentration of organic matter in the effluent for situations of interest. Some CW design and operation criteria establish that the species require two or three growing seasons to reach maximum efficiency and the removal of contaminants depends on the support material and the hydraulic retention time (HRT), in addition the CW can be influenced by hydrometeorological factors (CONAGUA, 2019).

Romellón et al, (2022) evaluated the effect of macrophyte pruning on microorganisms adhered to the support medium and the removal of BOD_5 in three experimental subsurface flow artificial wetlands (SSFCW) with species of *Phragmites australis* (SSFCW -Carrizo), *Pontederia sagittata* (SSFCW -Tule) and gravel as control (SSFCW -Grava). The experimental units operated with 3.1 days of hydraulic retention, the best treatment was SSFCW -Tule: before pruning, the biomass of microorganisms presented median values (N = 12) of 42931.6 mg/kg (Q1 = 40259.7; Q3 = 54478.4) and after pruning 33444.6 mg/kg (Q1 = 31210.9; Q3 = 36581.8), the plant biomass removed in pruning was 40.85 ± 2.58 kg, 95.44% of BOD_5 was removed with a k = 1.004 $days^{-1}$ (27.6 ° C), which allowed compliance with NOM-001-SEMARNAT-1996.

Alasino et al., (2015), evaluated a horizontal SSFCW with *Cortadeira Selloana* to treat the wastewater generated by a department of the Universidad Nacional de Córdoba, Argentina, the removal kinetics of BOD_5 were estimated assuming a first order reaction, they used an average residence time of 11.7 days, from the average of the k calculated for the months 18, 21 and 23 they obtained a k=0.115 days^{-1} for the removal of BOD_5. Marín et al., (2016), conducted a study with SSFCW evaluating two plant species: *Pontederia cordata* and *Phragmites australis*, the SSFCW were fed with domestic wastewater, achieving HRT of seven days. The best treatment was the SSFCW with *Potenderia cordata* that obtained removal efficiencies for BOD_5 (95 %), COD (95 %), NT (94 %), PT (81 %) and OSH (94.09%). Marín-Muñiz, (2016) evaluated 12 microcosms with *Typha* spp, six with tepezil (ST) and six with river porous stone (RPS) of each one there were three without plants that were control, no significant effect was observed regarding the type of substrate (P>0.05), the average removals of N-NO_3, P-PO_4 and BOD_5 were (60.3, 55.4 and 80.1) % respectively.

Mexico has 230 systems of constructed wetlands (CW) documented throughout the country, eleven of them in the state of Tabasco (CONAGUA, 2021). There is a low coverage in water treatment in our state, as there is a marked predominance of importance of conventional technologies and primary treatment whose efficiency and costs have not yielded the expected results. The CW, built in Tabasco are combined free flow constructed wetland systems (FFCW) and subsurface flow constructed wetland (SSFCW) (CONAGUA, 2019). For this reason, it is necessary to look for alternative treatment systems that are economical, easy to operate and appropriate to the weather conditions and natural resources of the state. Hence, the purpose of this research was to evaluate the potential for phytoremediation of wastewater using the species *Pontederia cordata* in CW, analyzing its behavior in the removal of basic pollutants by through of a physical modeling of the spatial distribution and the determination of the kinetics of degradation in relation to BOD_5.

Materials and Methods

Experimental Unit

The treatment system consisted of a wastewater receiving tank with a capacity of 200 L, plus the treatment unit that is an SSFCW. The units were built in the Universidad Juárez Autónoma de Tabasco. The SSFCW were made of carbon steel sheet with a thickness of 0.5 cm, with dimensions of 2.5 m long x 1.2 m width x 1.0 m depth, operating with 0.5 m (water and gravel column) long. For this design, the minimum area necessary to remove BOD was considered, according to the design criteria of López et al., (2014). At the inlet and outlet, a flow valve was installed to control the flow rate and for the sampling of water throughout the experiment. The support medium consists of river gravel with grain size ~21 mm in diameter at a depth of 50 cm, porosity of n=56±3.5%, density of 2670±19.3 kg/m^3, with a volume of 0.66±0.05 m^3, allowing a water volume of 0.1±0.05 m^3. The plant species installed in the SSFCW was *Pontederia cordata*, 24 organisms were planted by experimental wetland, the stems were 10 cm high and the root was planted five cm under the surface of the support medium. The species were stabilized in the experimental units for a period of three months (From sowing to the first reproduction cycle). At the end of the treatment processes, the treated waters were discharged into a natural wetland adjacent to the site, ensuring that they comply the maximum permissible limits for wastewater discharges, In the authorization of discharge rights by CONAGUA, TAB-L-0080-12-03-13 (CONAGUA, 2013).

Collection, planting and stabilization of the plant species.

The plants were collected from a flood zone located at coordinates 17° 59.219´ N and 92° 57.588´O in Villahermosa, Tabasco, corresponding to complete young plants, including flowers and fruits. To avoid ~~the~~ stress in the collected they were washed with water from the natural environment for a period of 10 days, and then they were transplanted to the SSFCW. The stabilization period involved the planting of 15 individuals, keeping the water at 0.5 m. Valles and Alarcón (2014), points out that this period can last about three months after which a bacterial film is formed in the rhizosphere and support medium. The evaluation of the removal of contaminants is carried out after 300 days of operation, especially when it is desired to remove heavy metals. Several authors have reported that the best efficiency in wetlands is achieved between the first and second year (Amabilis et al., 2016; Torres et al., 2017). The experimentation period (stabilization and operation) of the SSFCW comprised two years from March 2016. Followed by an evaluation phase carried out in May 2018. For this reason, we recommend the analysis after approximately one year of operation.

The plants were measured and weighed at the beginning and at the end of the experiment, considering leaf, root and stem, this to check the correct stabilization and the absorption of nutrients by the plant.

Physico-chemical characterization

The water characterization phase (NMX-AA-003-1980) began by evaluating variables with its respective methodology such as temperatures SM 2550 B, turbidity SM 2130 B, EC SM 2510, pH SM 4500 H+, color SM 2120 B, BOD SM 5210 B. The characterization of the support medium was performed at the sampling points indicated in Figure 1. The biomass present on the rocks was determined by gravimetry (volatile matter), adapting the total volatile solids (TVS) method (NMX-AA-034-SCFI-2001) to a rock sample. The density of the particle and the porosity of the support medium were determined based on NOM-021-RECNAT-2000. The flow was estimated by the direct method volume over time (Briones and García, 2014), monitoring 10 days during the hours of operation (5:00 a.m. to 7:00 p.m.).

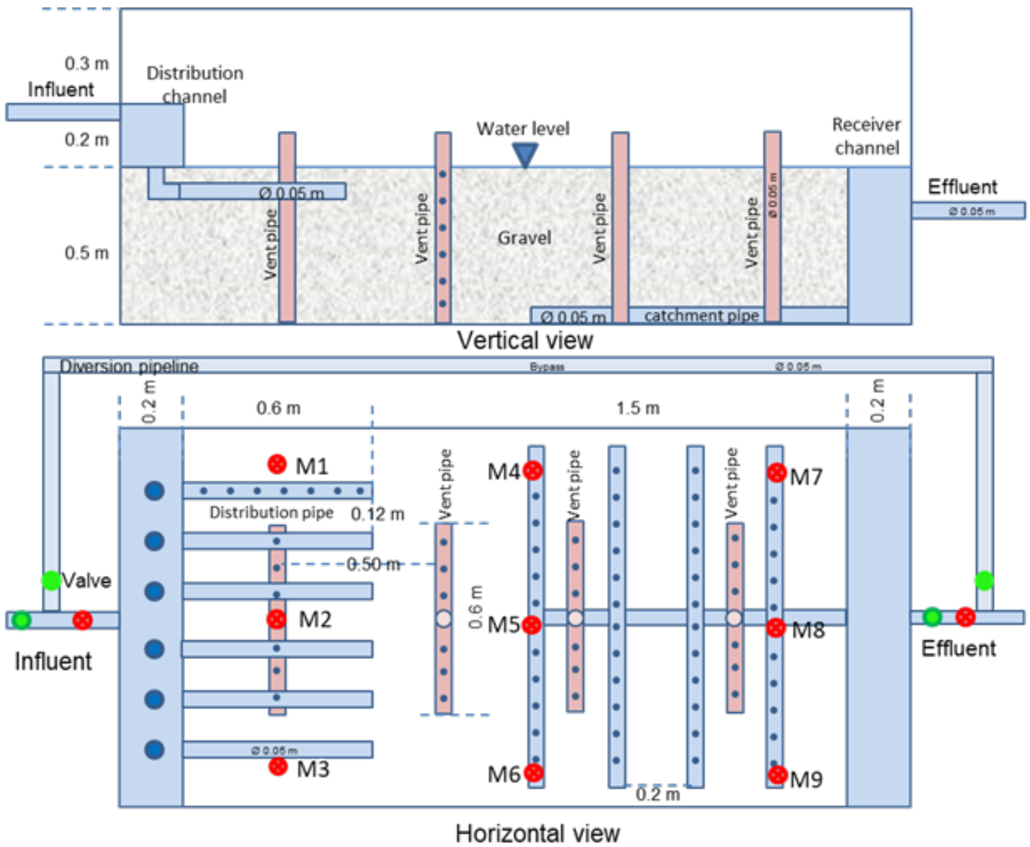

Figure 1. The red points from M1 to M9 indicate the sampling point in the Subsurface Flow Constructed Wetland (SSFCW). The effluent and effluent points were also measured. Design López et al., (2014).

Removal efficiency of basic pollutants

Based on the data obtained, the removal efficiency in the treatments, expressed as removal percentage, it was calculated for each variable observed (García et al., 2019), using Equation 1:

$$\eta = \left(\frac{C_i - C_o}{C_o}\right) 100 \quad \textbf{(1)}$$

where η is the removal (%); C_i is the inflows concentration of wastewater (mg/L); and C_o is the outflows concentration of treated wastewater (mg/L).

Experimental Design

In this work, an experimental design of one factor was performed for SSFCW was conducted with *Pontederia cordata*, with five treatments at different distances (0, 0.42, 1.25, 2.08 and 2.4) m with three replica units at each point as shown in Figure 1. A statistical analysis was performed to find differences between treatments, through a Kruskal wallis test followed by a Mann-Whitney U median contrast for the variables of pH, color, turbidity and vegetable biomass. For the parametric temperature and EC data, a simple ANOVA was performed followed by a multiple Tukey contrast. The data were analyzed in the STATGRAPHICS 7.0^{MR} statistical package.

Degradation kinetics

On the other hand, the criteria applied in the theoretical designs of constructed wetlands for the removal time of BOD_5, depends on the type of flow. The importance of calculating the HRT time of wastewater based on BOD_5 is that in this way the degradation time of organic matter is estimated. In this work, the degradation kinetics was calculated considering that there is a first order behavior (Crites and Tchobanoglous, 2000; Romellón et al., 2021), using Equation 2:

$$C_e = C_i e^{-kt} \quad \textbf{(2)}$$

where t is the retention time for the removal of BOD (days); Ce is the concentration of BOD_5 in the effluent, from the reactor n of the series (mg/L); C_i is the Concentration in the influent (mg/L); and k is the degradation constant (d^{-1}).

Results and Discussion

Stabilization of the plant species

The stabilization of the plants can be seen in Table 1, where the conditions before planting and at the end of the experimentation are presented (one year after the stabilization phase), the plants presented almost twice their size, from young plant to adult plant, presenting greater growth in the vegetative part than in the roots, however, the roots were the ones that gained the most mass. The initial mass per plant (15 individuals) was 1.82±0.38 kg and at the end of two years (47 individuals) presented 2.17±0.33 kg, which shows that its biomass increases four times once stabilized.

Table 1. *Pontederia cordata* growth data

	Parameter	Initial	Final
Leaf	Length (cm)	22.8±1.1	49.4±1.1
	Width(cm)	12.9±1.1	24.6±1.0
	Quantity	7.4±0.5	7.8±1.9
	Wet weight (gr)	0.02±0.0	0.15±0.0
	Dry weight (gr)	0.01±0.0	0.06±0.0
	Humidity (%)	56.9±1.3	56.0±1.4
Stem	Height (cm)	68±2.91	112.7±1.0
	Diameter (cm)	1.0±0.1	2.42±.0.1
	Quantity	9±0.7	10.8±2.7
	Wet weight (gr)	0.09±0.0	1.3±0.11
	Dry weight (gr)	0.03±0.00	0.5±0.0
	Humidity (%)	59.8±1.2	58.2±0.2
Root	Width(cm)	17.4±1.1	15.6±3.2
	Length (cm)	12.1±1.1	35.6±11.4
	Wet weight (gr)	1.9±0.07	1.6388±0.1
	Dry weight(gr)	0.6±0.0	0.5±0.06
	Humidity (%)	68.6±0.8	66.8±3.2

Flow rate and hydraulic retention time

The SSFCW was designed to operate with a flow of 200 L/day, however, when carrying out the corresponding measurements and its volumetry, it was possible to establish that the HRT is 4.2 days. The mean operating flow of CW is 185.1±58.6 L/day, the minimum 86±10 L/day and the maximum 260±26 L/day.

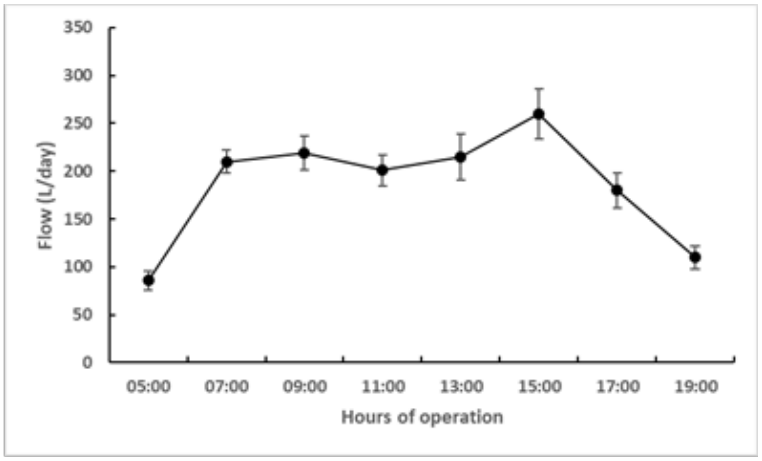

Figure 2. Hourly average flow (SD) during the day in the SSFCW.

Physicochemical characterization of water

The physicochemical characterization of the control tank and the SSFCW are shown in Table 2 respectively, where it can be observed that the treatment with *P. cordata* presented results below the maximum permissible limit according to NOM-001-SEMARNAT-2021 for the parameters temperature, and pH. These values were used to calculate the removal efficiencies corresponding to the sampling from May to June 2018 at the end of two years of operation, two simple input and output samples were taken per day, for a total of 40 simple samples. The monitored spatial distribution corresponds to 11 days from May to July 2017, taking one sample per sampling point per day, with a total of 121 samples. The hydraulic retention time in this phase was 4.14 days, one more day to the one used by Marín-Muñiz, (2016).

Removal efficiency of basic pollutants and statistical analysis of the SSFCW main parameters of the Table 2 presents the removal efficiencies of the *P. cordata* treatment based on the input values to the treatment system (distribution tank) and treatment effluent.

Table 2. Average removal efficiency of the SSFCW with *Pontederia cordata (N=11)*.

Parameter	**Units**	**Distribution tank**		**SSFCW *Pontederia cordata***		
		Value	**SD (±)**	**Value**	**SD (±)**	**RE %**
Temperature	° C	26.54	1.90	25.23	1.55	5.0
Turbidity	NTU	19.79	4.25	5.89	2.06	70.2
Color	CU	1038.67	208.17	189.20	25.34	81.0
pH	pHU	8.93	0.46	8.41	0.34	5.9
EC	µS/cm	1765.43	208.68	1044.51	649.40	40.8
BOD_5	mg/L	348.55	69.86	63.49	8.50	81.8

pH

For this variable, low neutralization efficiencies (RE) are recorded due to the pH range that does not allow to see a high efficiency, however 5.9% of RE shows the decrease in basicity approaching neutrality. The Kruskal-Wallis test evaluates the hypothesis that the medians pH (pHU) within each of the five levels of Distance (m) are equals. Since the P value is less than 0.05, there is a statistically significant difference between the medians with a 95.0% confidence level. The difference is shown between the distance 0 and 0.42 m, since from the distance 1.25 to 2.4 m there is no difference between the treatments. This means that the water stabilizes from 1.25 m (Figure 3). In the treatments that Marín-Muñiz evaluated, (2016) the pH remained between 6.5 and 7.4 value lower than the one registered by us but is within the permissible range of NOM-001-SEMARNAT-2021 that establishes from five to nine pH units.

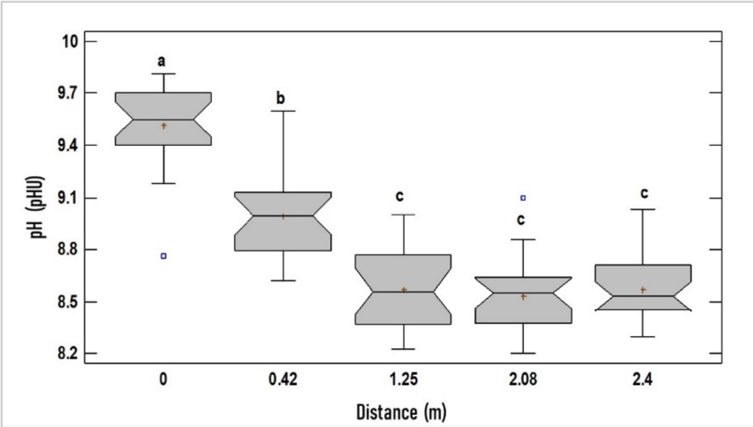

Figure 3. Median values (±SD) of the pH variable of the different treatments of the CW evaluated (*N=11*). Different letters indicate statistically significant differences between treatments.

Temperature

The decrease of the temperature shown with an efficiency of 5% shows how the vegetation cover helps to reduce this parameter. When performing the Multiple Ranges test for temperature (°C) by distance (m). This applies a multiple comparison procedure to determine which means are significantly different from others. There are no statistically significant differences between those levels from 0.42 to 2.4 m. Since the P value of the Fisher (F) test is less than 0.05, there is a statistically significant difference between the average temperature (°C) between one level of distance (m) and another, with a level of 95.0% confidence (Figure 4). The temperature at the end of treatment complies with what is established by NOM-001-SEMARNAT-2021, an appropriate value for the development of mesophilic microorganisms.

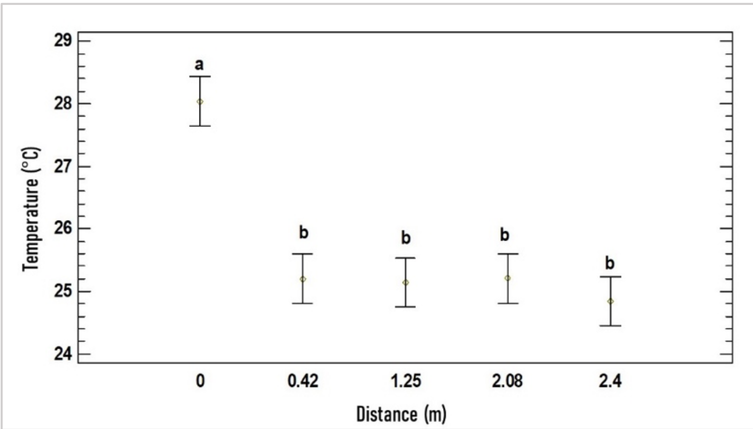

Figure 4. Average values (±SD) of the Temperature variable of the different treatments of the CW evaluated (*N=11*). Different letters indicate statistically significant differences between treatments.

Turbidity

The Kruskal-Wallis test evaluates the hypothesis that Turbidity medians (NTU) within each of the five distance levels (m) are equal. Since the P value is less than 0.05, there is a statistically significant difference between the medians with a 95.0% confidence level (Figure 5). Marín et al., (2016), show higher turbidity values (10.97 and 23.34 NTU) for the Tule and Carrizo species respectively, resulting in our most efficient treatment (5.89±2.06 NTU, 70.2% RE). Regarding them removal efficiency RE, they managed to decrease 91.65% with their best treatment, even though we obtained lower values in this parameter, this is due to the water quality of the tributary of them, it was found with more turbidity than ours.

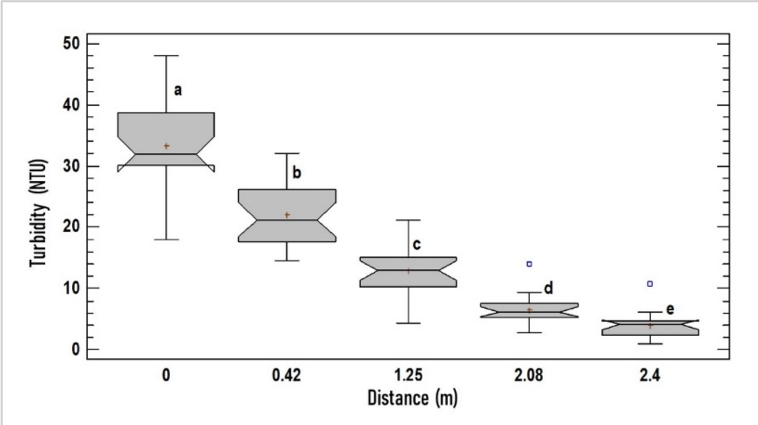

Figure 5. Median values (±SD) of the Turbidity variable of the different treatments of the CW evaluated (*N=11*). Different letters indicate statistically significant differences between treatments.

Electrical conductivity (EC)

The ANOVA analysis of ratio F equal to 15.51, is less than 0.05, there is a statistically significant difference between the mean EC (µS/cm) between a distance level (m) and another, with a confidence level of 95.0%. In Figure 6 the remarkable decrease of the EC between each distance can be observed, however ~~the~~ statistics shows that there is no significant difference between the stand and the distance of 0.42 m. Charris and Caselles (2016) evaluated a pilot scale SSFCW, planted with *Cyperus ligularis* and *Echinochloa colomun*, obtaining efficiencies of 34.8% and 24.4% respectively. They suggest that there may be a difference in this parameter due to the type of macrophyte and climatic conditions, given that high evapotranspiration concentrates dissolved salts. For our part, the efficiency in our CW was 40.8%, higher than previously presented by the aforementioned authors.

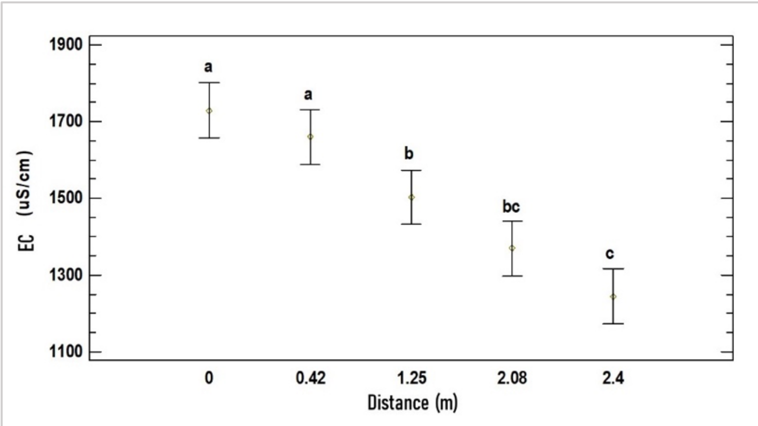

Figure 6. Average values (±SD) of the EC variable of the different treatments of the CW evaluated (*N=11*). Different letters indicate statistically significant differences between treatments.

Color

The Kruskal-Wallis test evaluates the hypothesis that the Color medians (CU) within each of the five distance levels (m). Since the P value is less than 0.05, there is a statistically significant difference between all medians with a 95.0% confidence level (Figure 7). Bedoya et al. (2014) evaluated laboratory scale CWs HRT of nine days, using *Typha latifolia* and *Cyperus papirus* where for this variable, they found efficiencies of 90.9% and 92.2% respectively. On our part, a lower efficiency (81.8%) was obtained in contrast the one evaluated by Bedoya et al. (2014). These differences may be based on the retention time of the system, since in this experiment the HRT was close to half (4.14 days) of the described in the experiment explained above (Arias-Martinez et al., 2010).

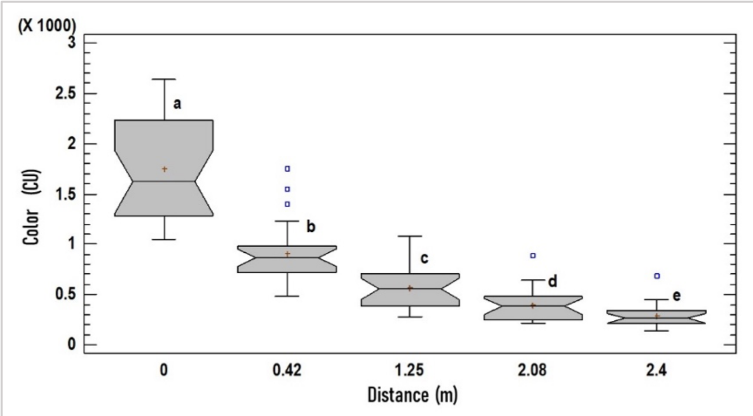

Figure 7. Median values (±SD) of the Color variable of the different treatments of the CW evaluated (*N=11*). Different letters indicate statistically significant differences between treatments.

BOD_5

Marín et al., (2016), obtained better efficiencies (95%) than those presented in this work with 81.8% for the BOD_5, as for the TP (81.8%) the efficiency they present is almost the same as that found with 81.1% within our CW, while for NT it found 95% efficiencies, exceeding those found in this work (81.88%). Marín-Muñiz, (2016), with *Typha* reports similar removals of BOD_5 (80.1%). These similarities are related to temperature and seasonal season. So, there is no difference with respect to the dry and rainy period, operating its CW at a temperature close to 26 °C, similar to the temperature presented in our monitoring period.

Zamora et al., (2018), reported the effects of *Pontederia sagittata* in CW for treating rural community wastewater, using tepezyl porous media and porous river gravels, in a period of 12 months, they present of BOD_5 removal of 82.6% in tepezyl and 83.9 in river gravels. Although this research was reported under different conditions (microscale), the results were similar to those found in our research.

Mathematical model adjustment and spatial distribution of basic pollutants in SSFCW

The normalized average values and the fit of the model to which the results are related are presented. For the pH (Figure 8), a decrease from alkaline to slightly alkaline is observed from the 2.08 m point to the exit point (2.4 m), the distance at which the pH stabilizes for its exit in the effluent (R^2=0.98. C). With respect to temperature (Figure 9), the behavior is similar to pH. Water that is closer to the surface has higher temperatures (>26°C) than the water sampled from the column. There is a decrease in temperature throughout the system, such behavior allows mesophilic bacteria to operate under an optimum range of 25-40°C, which promotes an improvement in the removal of contaminants.

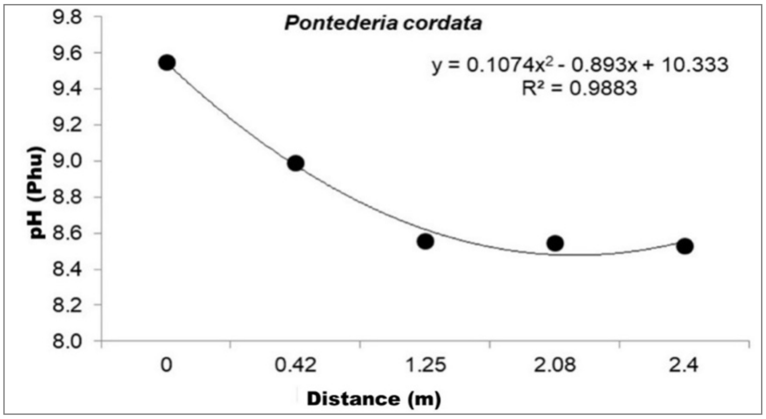

Figure 8. Model Neutralize to the pH parameter**(*N=11*)**.

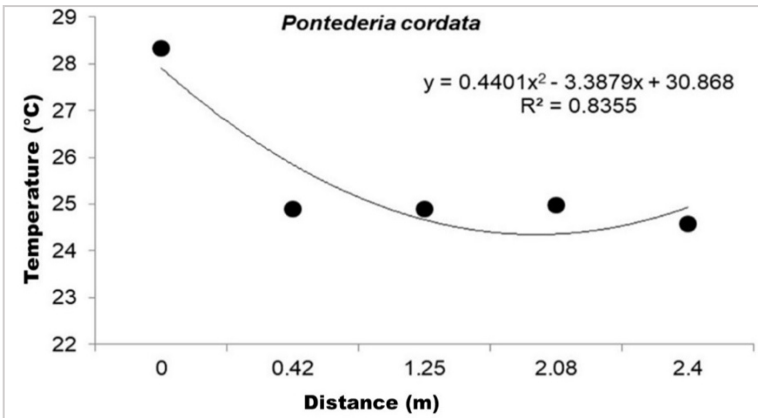

Figure 9. Model adjusted to the temperature parameter**(*N=11*).**

The EC (Figure 10) presents an adjustment R^2=0.95 and a decrease with a linear tendency in the concentration because it is linked to dissolved solids concentrations. The decrease in this property (EC) is related to the absence of solids. The presence of these solids is influenced by the action of the entire system that consists of the support medium, the roots of the plant and the bacteria attached to them. Since the EC reading is directly related to ions present in the solution, such as chloride, nitrate, sulfate, phosphate, sodium, magnesium, and calcium. Goyenola (2007). The interaction between plants, filtering media, microorganisms, and the atmosphere, help to remove solids by sedimentation, filtration and biodegradation

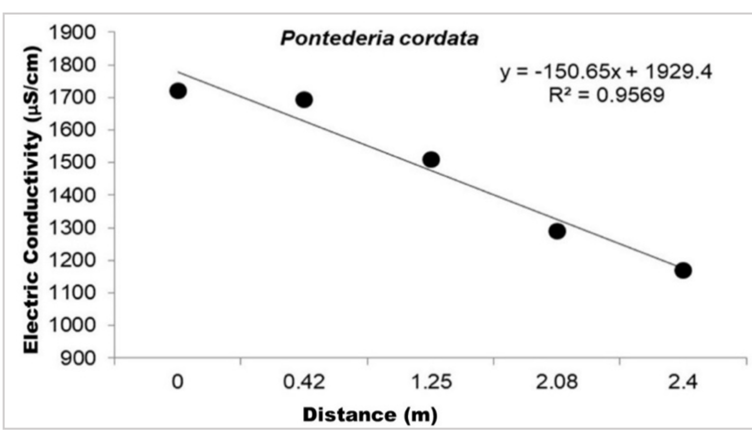

Figure 10. Model adjusted to the EC parameter**(*N=11*).**

The color (Figure 11) shows values of UC that are reduced along the wetland showing an exponential decrease from 1038.67 UC to 189.20 UC with a R^2=0.98. Turbidity showed the same trend (R^2=0.99) (Figure 12), showing the lowest values at the exit of the wetland with an RE of 70.2%.

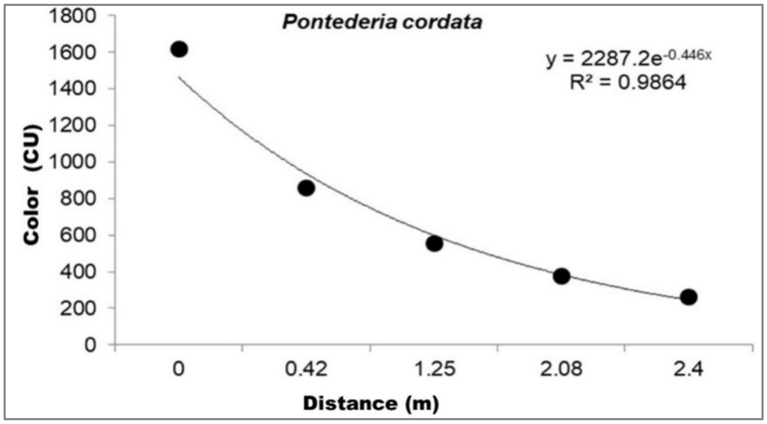

Figure 11. Model adjusted to the color parameter***(N=11)*.**

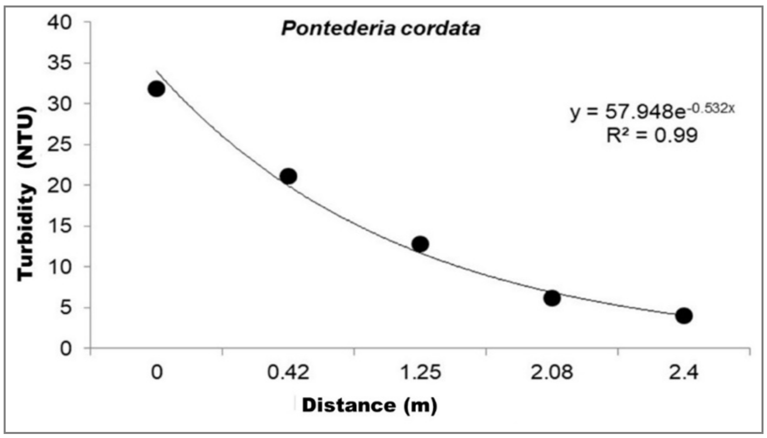

Figure 12. Model adjusted to the turbidity parameter***(N=11)*.**

BOD_5 (Figure 13) behaves as a decreasing function with a setting of R^2=0.98 where the highest rate of reduction is observed between the input and the third sampling point. However, the maximum reduction point is not appreciated, so the reactor could be designed for a longer retention time. The value of BOD_5 is related to the amount of biodegradable organic matter, as shown in Figure 12 at the exit of the reactor there is less organic matter since in its passage through the wetland the microorganisms of the system degrade it, with a decrease of 330 mg/L to 110 mg/L.

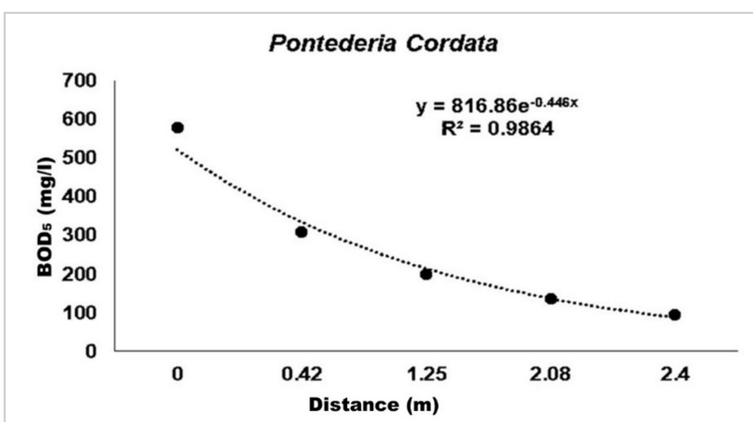

Figure 13. Model adjusted to the BOD_5 parameter(*N=11*).

The spatial distribution of microorganisms adhered to the support medium (Figure 14) presents a gradual decrease towards the effluent from 50,000 mg/kg to 4,000 mg/kg. This behavior is due to the fact that microorganisms depend on the amount of food, so that the greater the amount of food, the greater the presence of microorganisms. These results can be compared with the study conducted by López-Ocaña et al., (2019) that evaluated an SSFCW with the same dimensions using *Thalia geniculata*, where it managed to reduce from 33,000 to 2,000 mg/kg of bacterial biomass attached to the support medium.

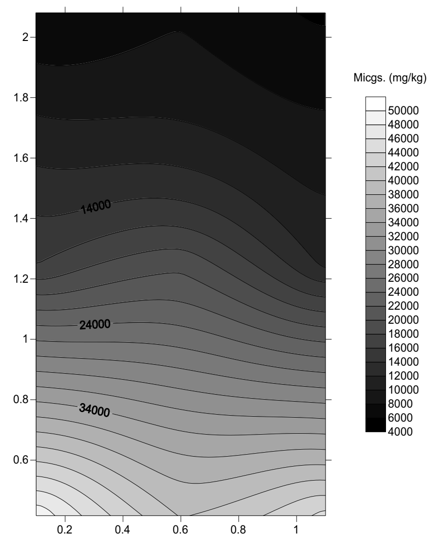

Figure 14. Spatial distribution of microorganisms**(*N=11*).**

Degradation kinetics in SSFCW

Figure 15 shows the kinetic behavior during the evaluation phase two years after the wetland starts, it is observed that the SSFCW has a k=-0.37 days^{-1}, which is a value greater than that obtained by Alasino et al., (2015) who reported average values of k at 18, 21 and 23 months of 0.115 days^{-1}. Compared to these results, the present work reports a better BOD5 removal efficiency with respect to retention time, since the value was reduced to 52.2 mg/L BOD_5 in 4.5 days, complying with discharge criteria established by CONAGUA, which is 70mg/L. Figure 15 shows the estimated time that would have to continue to comply with the maximum permissible limit for the protection of aquatic life (30 mg/L) as established by NOM-001-SEMARNAT-1996, which is seven days, clarifying that the current NOM-001-SEMARNAT-2021 standard does not include this parameter (BOD_5) within its basic pollutants.

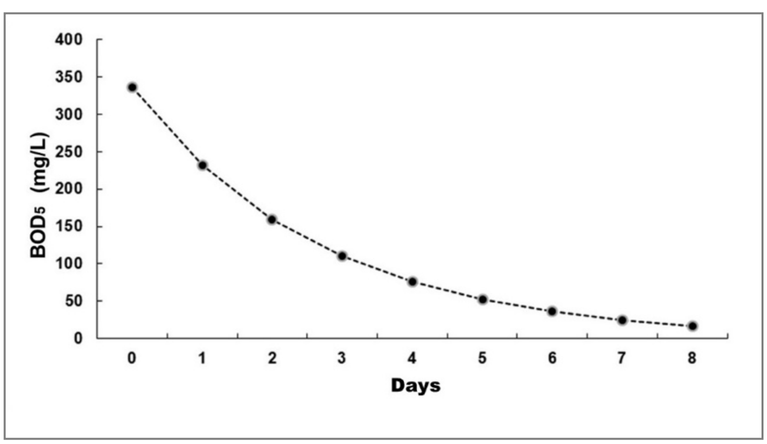

Figure 15. Kinetic behavior during the evaluation phase, which was evaluated after two years of operation (n=1, k=-0.37 d^{-1}).

Conclusions

The macrophyte *Pontederia cordata* has a good adaptation since the plants produces almost 4 times their biomass in their adaptation, they are easy to handle for their collection, planting and harvesting in the operation. After two years of operation, the SSFCW obtained a k=-0.37 days^{-1}, achieving a turbidity removal of 70.2% and for BOD of 81.8%, with a hydraulic retention time of 4.54 days, which allows it to comply with the permissible limits for these contaminants established in the discharge right. Longitudinal spatial distribution modeling shows that the parameters of turbidity (R^2=0.99) and BOD (R^2=0.98) at 2.08 m inside the reactor are significantly removed, the behavior of microorganisms being inverse, which are in greater concentration in the first sections of HA, due to the presence of organic load. *Pontederia cordata* can be a good alternative for wastewater phytoremediation in the humid tropics of Mexico.

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