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Horizontal subsurface flow constructed wetlands for tertiary treatment of dairy wastewater

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ABSTRACT

The aim of this work was to evaluate the efficiency of horizontal subsurface flow constructed wetlands (HSFCWs) planted with Typha domingensis and Phragmites australis in the final treatment of dairy wastewater. Ten microcosms-scale reactors simulating HSFCWs were arranged outdoors under a semi-transparent plastic roof. Five replicates were planted with T. domingensis and five with P. australis. In both cases, light expanded clay aggregate (LECA) 10/20 was used as a substrate. Real effluent with previous treatment was used. In order to evaluate contaminant removal efficiencies in each reactor, pH, electrical conductivity, suspended solids, ammonium, nitrate, nitrite, total phosphorus, and chemical oxygen demand (COD) were analyzed before and after treatment. HSFCWs planted with T. domingensis and P. australis were efficient for the final treatment of dairy wastewater. Removal efficiencies obtained in microcosms planted with both macrophytes were over 96% for ammonium and nitrite. Nitrate removal efficiency was 39%. COD decreased along the experiment near 75% for both treatments. High removal percentages for suspended solids (78.4–81.1%) were also achieved. However, systems planted with T. domingensis were significantly more efficient for total phosphorus removal (88.5%) than those planted with P. australis (71.6%).

KEYWORDS
wetlands; dairy wastewater; macrophytes

Introduction

There are several dairy basins in Argentina. Such areas are located mainly in the Pampa Region, where there are numerous dairy farms. In most cases, milk is processed by medium-scale or large-scale factories located in such areas. These industries manufacture dairy products for subsequent commercialization, for exports and the internal market as well.

As a result of their operations, dairy companies generate a large volume of effluents of varying quality. Although wastewater is treated through different methods, dumping limits established by current regulations are often exceeded. Therefore, after biological treatment, a polishing wastewater treatment is required.

Generally, dairy effluents contain high concentration of nutrients (nitrogen and phosphorus), high chemical oxygen demand (COD), and suspended solids as well as pathogenic microorganisms. Nitrogen is usually present in the form of ammonium, which is one of the pollutants controlled by current legislation. There are numerous polishing techniques that can be applied to achieve adequate values that allow wastewater discharge, namely: membrane processes, adsorption, oxidation ponds, filtration, etc. Constructed wetlands (CWs) could be an alternative to conventional methods. These systems have been deployed worldwide, but they have not been studied in our country for dairy wastewater treatment under local conditions (Maine *et al.* 2017).

In Argentina, even though the environmental conditions are favorable, surrounding lands at a low cost are easily available in the vicinity of industries and macrophytes are adapted to the climate, CWs are not widely implemented. However, there is a growing interest in the potential of alternative treatment methods like CWs.

CWs are man-made systems that have been designed to emphasize specific characteristics of wetland ecosystems for improved treatment capacity (Kadlec and Wallace 2009; Vymazal 2011). Their most relevant benefits are operation simplicity, low or zero energy consumption, low waste production, low sound environmental impact, and good integration to the environment (Kadlec and Wallace 2009; Maine *et al.* 2009; Vymazal and Kröpfelová 2011).

For the present research, based on previous experiences, horizontal subsurface flow constructed wetlands (HSFCWs) were used. They have been successfully used for dairy and milking parlor wastewater treatment worldwide (Kern and Brettar 2002; Mantovi *et al.* 2002; Mantovi *et al.* 2003; Hill *et al.* 2003) and specifically, in cheese producing industries (Wallace 2002; Khalil *et al.* 2005; Gorra *et al.* 2007). However, climatic conditions and effluent composition in our region are different from those already studied.

According to Vymazal and Kröpfelová (2008) average BOD_5 loadings for agricultural wastewater are the highest in comparison with municipal, industrial, and landfill leachate wastewater.

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These effluents generally presented an organic loading rate of 541 Kg BOD_5.Ha^{-1}·d^{-1}. Knight *et al.* (2000) compiled the Livestock Wastewater Treatment Wetland Database and showed average BOD_5 and TSS reductions of 68% and 47%, respectively. Case studies of dairy treatment wetlands worldwide reported nitrogen and phosphorus removals between 48–98% and 35–96% respectively, depending on nutrient loading conditions and wetland age (Hammer *et al.* 1991; Hunt and Poach 2000; Newman *et al.* 2000; Schaafsma *et al.* 2000). Drizo *et al.* (2006) used horizontal wetlands to treat dairy wastewaters in Vermont, USA. Results indicated that constructed wetlands have a good potential for dairy farms wastewater management under cold climate conditions. The use of HSFCWs to treat dairy farm effluents was also reported by Gray *et al.* (1990) in United Kingdom, Chen *et al.* (1995) in USA, and Tanner (1992) in New Zealand.

Macrophyte selection is an important design parameter in these systems because they play a significant role in pollutant removal. Several authors pointed out that there could be a difference in treatment efficiency regarding plant species (Vymazal 2013). Plants must also survive to the high variability and toxic effects of wastewater. *Typha domingensis* (Cattail) was chosen for this study due to its high productivity and nutrient removal efficiency (Maine *et al.* 2007, 2009). *Phragmites australis* (Common reed) was also evaluated since it is one of the plants most frequently used worldwide in constructed wetlands with horizontal subsurface flow (Vymazal 2013).

The aim of this work was to evaluate the efficiency of horizontal subsurface-flow constructed wetlands (HSFCWs) planted with *T. domingensis* and *P. australis* in the final treatment of dairy wastewater.

Materials and methods

– *Experimental design*

Ten microcosm-scale reactors (0.35 × 0.25 × 0.30 m; length × width × depth) simulating HSFCWs were set outdoors under a semi-transparent plastic roof. Temperature ranged from 2.6 to 28.4°C during the experimental period. Five replicates were planted with *T. domingensis* and the other five with *P. australis*. In both cases, 2 macrophytes were planted in each reactor and Light Expanded Clay Aggregate (LECA) 10/20 was used as a substrate. LECA consists of small, lightweight, bloated particles of burnt clay and it has been widely used as an adsorbent for removal of pollutants due to its low cost and high porosity. Particles are composed primarily of quartz. It is an environment-friendly and entirely natural product.

Reactors were operated with horizontal subsurface flow. The hydraulic load applied was 1000 mm.d^{-1}, corresponding to a nominal hydraulic residence time (HRT) of about 7 days. Organic load applied was 0.7 g.m^{-2}.day^{-1}.

Macrophytes were collected from a natural environment, pruned to 30 cm, set in the reactors, and acclimatized before the experiments. The acclimatization period lasted 30 days. The first 15 days, tap water was added to the reactors. Then, diluted dairy effluent was added during 15 days. Wastewater was taken from a local dairy manufacturer. It had undergone previous treatment which consisted of the following sequence: Equalization + Dissolved Air Flotation (DAF) + aerated lagoons.

Three successive loading experiences were carried out, each one with a HRT of 7 days. The experiment lasted 21 days.

– *Plant growth monitoring*

Growth monitoring was carried out by measuring plant height and performing visual inspection in each reactor during the acclimatization stage (September 2016) and the experimental stage (October 2016). The external appearance of plants was observed daily to detect possible senescence.

– *Experiment and analytical determinations*

In order to evaluate contaminant removal efficiencies in each reactor, pH, electrical conductivity, suspended solids (SS), ammonium (NH_4^+), nitrate (NO_3^-), nitrite (NO_2^-), total phosphorus (TP), and COD were analyzed. Chemical analyses were performed following the American Public Health Association (APHA 2012) guidelines. Conductivity was measured with an YSI 33 conductivity meter and pH with an Orion pH-meter. $NO_2^-$was determined by coupling diazotation followed by a colorimetric technique. NO_3^- and NH_4^+ were determined by potentiometry (Orion ion selective electrodes, sensitivity: 0.01 mg.$l^{-1}$of N, reproducibility: ±2%). In the case of TP, non-filtered water samples were digested with sulfuric acid-nitric acid. Soluble reactive phosphorus was determined in the digested samples (Murphy and Riley 1962). COD was determined by the open reflux method. All these measurements were carried out before and after the treatment in each reactor. Mean concentrations of total inorganic nitrogen were also calculated, by considering the addition of ammonium, nitrate, and nitrite expressed as nitrogen. Evapotranspiration was estimated and compensated with distilled water.

– *Statistical analysis*

Analysis of variance was used to determine significant differences in contaminant removal efficiencies between treatments, considering each successive loading experience such as a completely randomized block. The normality of residuals was analyzed graphically and homogeneity of variances was checked using Bartlett's test. Duncan's test was applied to differentiate means where appropriate. A level of $p < 0.05$ was used for all comparisons. Calculations were performed using the Statgraphics Plus 5.0 software.

Results and discussion

All reactors presented good biomass development growth and plant height increased significantly, indicating positive growth. In all treatments, it could be verified that plant growth increased considerably when the reactors were filled with wastewater. This was due to the nutrient supply from the dairy effluent which allowed macrophytes to increase their height 3 times at the end of experiment. Figure 1 shows macrophytes height and biomass evolution at the beginning and at the end of the experiment.

Figure 1. Plants growth evolution.

Contaminant concentrations in the inlet did not present variability throughout the experiment. pH did not show significant differences before and after treatment, varying between 7.95 and 8.30. Electrical conductivity did not present significant differences before and after the treatment, ranging between 4.82 and 5.07 mS.cm^{-1} throughout the experiment. Mean concentrations of different parameters measured at the inlet and outlet wastewater are shown in Figures 2–8.

The concentration of suspended solids in wastewater decreased around 80%, not presenting statistically significant differences between the HSFCW planted with the different macrophytes under study (Figure. 2). Manios *et al.* (2003) reported that the presence of cattails did not produce a significant difference between planted and unplanted beds. For this reason, it could be inferred that suspended solids are mainly removed by physical processes like sedimentation and filtration (Kadlec and Knight 1996). Not only removal efficiencies were high but also sample color decreased remarkably after treatments. This is a point of interest for industries that discharge their effluents into open channels with low flow, such as those located in the study area.

Ammonium was removed from wastewater efficiently, and there were no significant differences between treatments.

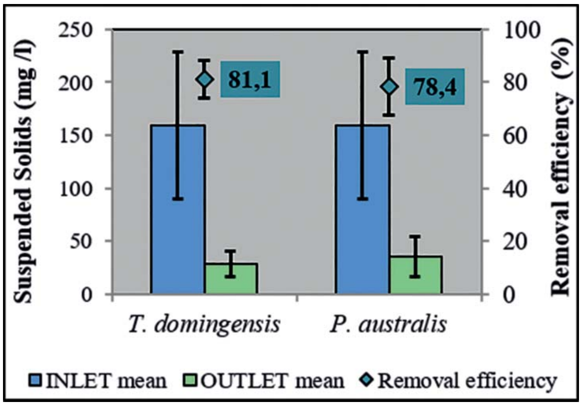

Figure 2. Suspended solids: removal efficiencies, inlet, and outlet concentrations.

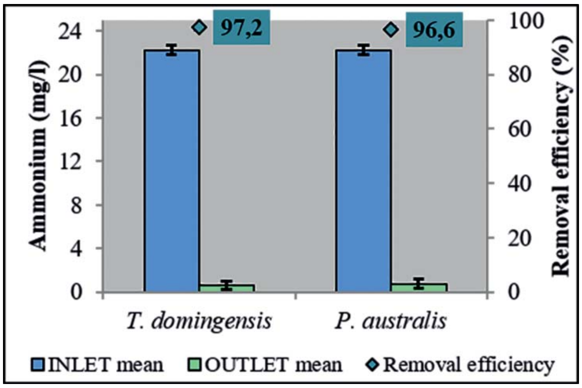

Figure 3. Ammonium: removal efficiencies, inlet, and outlet concentrations.

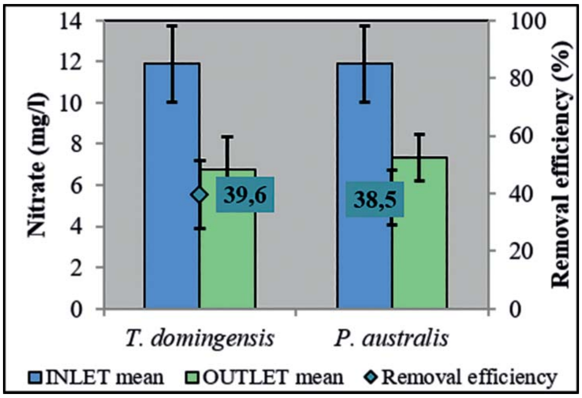

Figure 4. Nitrate: removal efficiencies, inlet, and outlet concentrations.

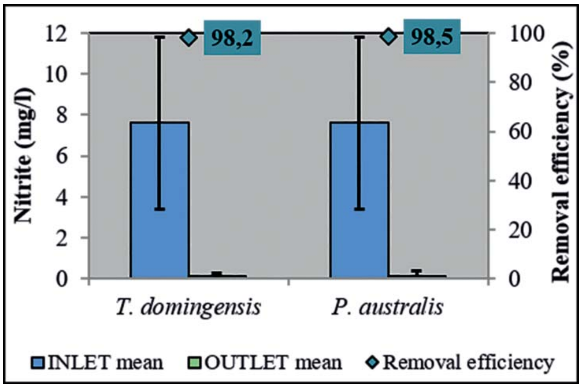

Figure 5. Nitrite: removal efficiencies, inlet, and outlet concentrations.

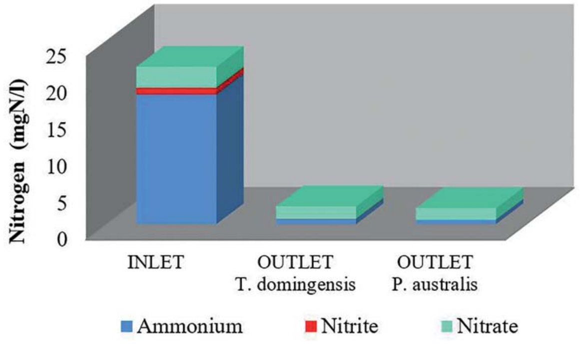

Figure 6. Nitrogen species: inlet and outlet concentrations in each macrophyte.

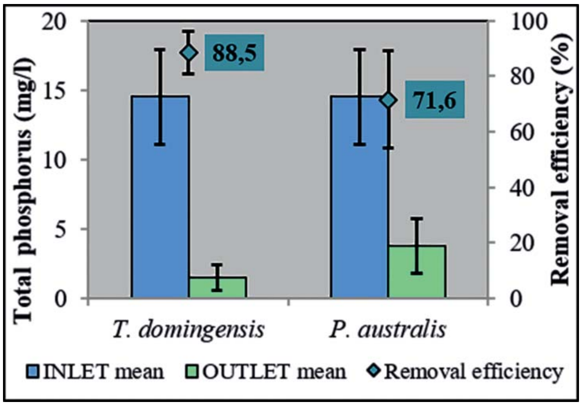

Figure 7. Total phosphorus: removal efficiencies, inlet, and outlet concentrations.

Removal percentages were higher than 95% (Figure. 3). Ammonium removal was associated with the following mechanisms: (1) absorption by macrophytes; (2) absorption by bacteria biomass; (3) adsorption in filter media; (4) nitrification in aerobic microzones near roots; and (5) volatilization as NH_3, favored by high pH (Kadlec and Knight 1996; Vymazal 2007; Paul and Clark 1996). Another mechanism that is currently under study and can also justify ammonium decrease is ANAMMOX (anaerobic ammonium oxidation), which consists of nitrite and ammonium conversion into gaseous nitrogen (Vymazal 2007; Hunt *et al.* 2005; Strous and Jetten 2004). Oxygen concentration is limited in HSFCWs. Therefore, ANAMMOX could be an important variable for nitrogen elimination since it requires less oxygen than the nitrification/denitrification process. Furthermore, ammonia-nitrogen loss through volatilization was negligible since it generally requires a pH of 9.3.

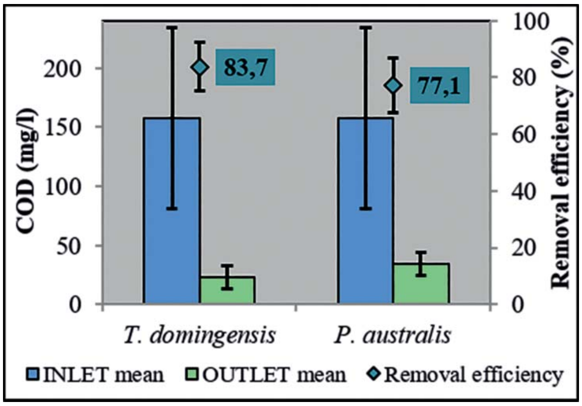

Figure 8. COD: removal efficiencies, inlet, and outlet concentrations.

Inlet and outlet nitrate concentrations for both treatments can be observed in Figure 4. Removal efficiencies achieved were less than 40%, without statistically significant differences between treatments. Since nitrate removal efficiencies were low, it could also be inferred that denitrification was not enough to remove all available nitrate. This contaminant came from both inlet effluent and from nitrification. Nitrite removal efficiencies, by contrast, were higher than 98% for both species and did not differ significantly from one treatment to another, as it can be seen in Figure 5.

Figure 6 shows inlet and outlet concentrations of different nitrogen species under analysis, expressed as nitrogen (ammonium, nitrate, and nitrite). It could be verified that in the initial effluent, most of the nitrogen was found in the form of ammonium, which was almost 100% removed by both systems. As it was previously mentioned, nitrate presented the lowest removal efficiency while nitrite outlet concentrations were not detectable.

Phosphorus removal in HSFCWs is limited due to the fact that the media used for horizontal flow wetlands (gravel, crushed stones, etc.) do not usually contain great quantities of iron, aluminum, or calcium to facilitate precipitation and sorption of phosphorus. In order to achieve better TP removal efficiencies, light weight clay aggregates have been used recently (Kröpfelová 2008). One of the aggregates mostly used is LECA and for this reason it was chosen as a substrate for the present research. The use of LECA allowed high phosphorus removal efficiencies, ranging from 72 to 88%.

Phosphorus removal was significantly different between treatments, being 88.5% in reactors planted with *T. domingensis* and 71.6% for those planted with *P. australis* (Figure. 7). Since *T. domingensis* had a better performance than *P. australis*, it could be suggested that plant uptake is another important mechanism for phosphorus removal. *T. domingensis* presents a higher aboveground biomass than *P. australis*, which could contribute to a higher accumulation of phosphorus in tissues.

Figure 8 shows inlet and outlet values of COD for both treatments. Mean removal efficiency obtained in microcosms planted with *P. australis* was 77% and 84% for reactors with *T. domingensis*. However, there were no significant differences between treatments. In constructed wetlands COD removal is mainly related to microbiological degradation attached to the matrix and plants roots (Sawaittayothin and Polprasert 2007).

The results reported in studies focusing on plant influence on organic matter removal in HSFCWs are controversial, however, most studies agree on the positive effect of macrophytes (Vymazal and Kröpfelová 2009).

Due to the increase in plant biomass achieved along the experiment, a high microbial development might be feasible on roots. Therefore, microbiological degradation might be favored. Substrate could also contribute to bacterial biofilm development that could influence COD removal.

Table 1 shows wastewater mean concentrations before and after treatments. The central column shows the discharge limits established by the state regulations (Resolution N° 1089/82. Title C. Regulatory law for the control of wastewater discharge.

Table 1. Comparison between mean concentrations (inlet and outlet) and regulatory discharge limits (Santa Fe Province, Argentina).

Parameter	INLET concentration	Law regulatory limits	OUTLET *T. domingensis* ($N = 15$)	OUTLET *P. australis* ($N = 15$)
pH	8.09	5.5–10.0	7.31–7.84	7.31–7.85
Suspended solids (mg.L^{-1})	159	30	28.4	35.2
COD (mg.L^{-1})	157	75	22.9	34.0
Ammonium nitrogen (mg N.L^{-1})	17.3	25	0.48	0.58
Total phosphorus (mg P.L^{-1})	14.3	2	1.53	3.81

Santa Fe Province, Argentina. Dumping point: open stormwater pipeline). As it was mentioned before, inlet wastewater came from a biological treatment. However, 4 out of 5 parameters under analysis did not comply with discharge limits (suspended solids, COD, ammonium, and TP).

After *T. domingensis* treatment, concentrations of all parameters decreased, meeting regulatory discharge limits. In this regard, *P. australis* treatments had a low performance. They also showed satisfactory removal efficiencies, but in the case of suspended solids and TP, the final concentrations exceeded the limits established by regulations. COD and ammonium mean concentrations complied with regulations. The cells highlighted in Table 1 show outlet concentrations that met the discharge limits.

Conclusions

HSFCWs planted with *T. domingensis* and *P. australis* were efficient for the tertiary treatment of dairy wastewater. Removal efficiencies obtained in the microcosms planted with both macrophytes were over 96% for ammonium and nitrite. Nitrate removal efficiency was 39%. As ammonium was the dominant nitrogen species at the inlet effluent, total inorganic nitrogen removal efficiency was very satisfactory. COD decreased along the experiment by more than 75% for both treatments. High removal percentages for suspended solids and TP were also achieved.

However, HSFCWs planted with *T. domingensis* were more efficient than those planted with *P. australis*, making the outlet effluent comply with regulations and suitable for being discharged into surface water bodies.

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