



Vertical flow wetlands and hybrid systems for the treatment of landfill leachate

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Abstract

Landfill leachates contain a variety of toxic compounds, which makes them one of the most difficult types of wastewater to be treated. An alternative “green” technology for leachate treatment is the use of constructed wetlands (CWs). The aims of this study were to select macrophytes and substrates to be used in vertical flow wetlands (VFWs) and to evaluate the performance of hybrid systems composed by a VFW and a horizontal subsurface flow (HSSW) or a free water surface flow (FWSW) wetlands for the treatment of a high ammonium concentration landfill leachate. In microcosms scale experiments, *Typha domingensis*, *Scirpus californicus*, and *Iris pseudacorus* were studied to assess their tolerance to raw and diluted leachate. Substrate selection for VFWs was evaluated using different layers of light expanded clay aggregate (LECA), coarse sand, fine sand, and gravel. Contaminant removals were higher in planted than in unplanted wetlands. Plants did not tolerate the raw effluent but showed a positive effect on plant growth when exposed to the diluted leachate. *T. domingensis* and *I. pseudacorus* showed higher contaminant removal ability and tolerance to landfill leachate than *S. californicus*. VFW with LECA + coarse sand showed the best performance in removal efficiencies. Hybrid system composed by VFW-FWSW planted with *T. domingensis* presented the best performance for the treatment of landfill leachate with high concentrations of ammonium.

Keywords Macrophytes · Substrates · Ammonium · Constructed wetlands

Introduction

Landfill leachate treatment is one of the major environmental concerns since the volume of waste is growing significantly (Renou et al. 2008). Moreover, Landfill leachate treatment is complex because of its varying chemical composition that depends on age, waste origin, climatic condition, and degradation rate of solid waste. The expected volume and the chemical quality of a leachate are unique on each site and change over time. A young leachate has a low pH and a high BOD₅/

COD ratio, while an old leachate has a low BOD₅/COD ratio and a high ammonium concentration (Kjeldsen et al. 2002).

An alternative “green” technology for leachate treatment is the use of constructed wetlands (CWs), where plants, microorganisms, and media play an important role in pollutant removal (Cooper 1999). In Argentina, CWs are of special interest due to their low cost, easy operation and maintenance, and the usual large availability of land around landfills. Macrophytes are important constituents of the treatment system contributing to the optimization of the wetland performance (Brix 1997; Guo et al. 2017; Kizito et al. 2017). For example, aerial tissues store nutrients, provide insulation to the system during winter, and add esthetic values. Submersed plant tissues act as a filter medium, release oxygen, and reduce water velocity enhancing sedimentation and contact time with the wastewater. The choice of the plant species to be used is a key design issue for CWs. To participate in the removal of contaminants, macrophytes must withstand the harsh environmental conditions and the possible toxic effects of the effluent to be treated (Tanner 1996). High ammonium concentrations in wastewater can limit the macrophyte species to be used in CWs (Clarke and Baldwin 2002).

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CWs of different types have been used to treat landfill leachate, such as sub-surface flow (Akinbile et al. 2012), downflow reed beds (Connolly et al. 2004), and aerated horizontal sub-surface flow (Nivala et al. 2007). Vertical flow wetlands (VFW) possess a high capacity to oxidize ammonium (Kadlec and Wallace 2009; Kadlec and Zmarthie 2010), which is one of the major contaminants of old landfill leachate (Kjeldsen et al. 2002). Dissolved oxygen (DO) in VFW converts ammonium to nitrate under aerobic conditions (nitrification). To achieve a complete nitrogen removal, an anaerobic stage is necessary. Free water surface flow wetlands (FWSW) and horizontal sub-surface flow wetlands (HSSW) may provide the anaerobic conditions to convert nitrate in nitrogen gas (denitrification). Thus, when a second stage of treatment, such as FWSW or HSSW, is added after the VFW, total nitrogen removal can be achieved (Vymazal 2005; Politeo 2013; Vymazal and Kröpfelová 2015; Wojciechowska 2017).

CWs are a promising option for sustainable landfill leachate treatment systems in developing tropical regions (Ogata et al. 2018). In Argentina, this technology is not still widely used for effluent treatment (Maine et al. 2009, 2017). Therefore, the use of CWs for the treatment of landfill leachate is a novel issue in our country. The aims of this study were to select macrophytes and substrates to be used in VFWs and to compare the performance of two configurations of hybrid systems (VFW-FWSW and VFW-HSSW) for the treatment of landfill leachate.

Materials and methods

Macrophyte selection experiments in VFWs

Three different macrophytes were studied: *Typha domingensis*, *Scirpus californicus*, and *Iris pseudacorus*. VFWs were built at microcosms scale using plastic reactors (25 × 25 cm and 35 cm depth) filled with washed light expanded clay aggregates (LECA) (Fig. 1). Plants were collected from natural wetlands belonging to the Middle Paraná River floodplain, Argentina. The chemical composition of the water from the sampling site employed in the study was (mean ± standard deviation) pH = 7.8; conductivity = 223 ± 1 μS cm⁻¹; dissolved oxygen (DO) = 6.71 ± 0.10 mg L⁻¹; soluble reactive phosphorus (SRP) = 0.023 ± 0.002 mg L⁻¹; NH₄⁺ = 0.990 ± 0.005 mg L⁻¹; NO₃⁻ = 0.410 ± 0.005 mg L⁻¹; NO₂⁻ = non-detected (detection limit = 5 μg L⁻¹); Ca²⁺ = 9.8 ± 0.1 mg L⁻¹; Mg²⁺ = 2.2 ± 0.2 mg L⁻¹; Na⁺ = 36.8 ± 0.5 mg L⁻¹; K⁺ = 16.1 ± 0.5 mg L⁻¹; Fe = 0.291 ± 0.005 mg L⁻¹; Cl⁻ = 14.6 ± 1.0 mg L⁻¹; SO₄²⁻ = 10.5 ± 1.0 mg L⁻¹; total alkalinity = 104.2 ± 1.2 mg L⁻¹. Only healthy plants of a uniform size were selected. All plants were washed. *T. domingensis* and *S. californicus* were pruned at a 20 cm high before the experiments, and four plants were planted per microcosms. In order to assess the role of macrophytes in contaminant removal, planted

and unplanted reactors (controls) were used. Experiments were carried out outdoors under a semi-transparent plastic roof.

The landfill is located in Villa Domínico, Buenos Aires (Argentina). It covers 450 ha and it is currently closed. It produces 600 m³ of raw leachate a day. This leachate was used in all experiments. Diluted leachate was used for the acclimation period. This solution was prepared by mixing raw leachate with tap water. After the acclimation period of 1 month, the removal efficiency of the microcosms wetlands planted with the different macrophytes was evaluated in two different experiments. In the first experiment, raw landfill leachate was used. This experiment lasted 1 week due to plant senescence. In a second experiment, diluted landfill leachate (1:10) was treated for 4 weeks. In both experiments, 8 L per day of landfill leachate was loaded in the VFWs, with a hydraulic loading rate (HLR) of 0.1 m day⁻¹. Samples were collected before and after treatment on a weekly basis.

Substrate selection experiment in VFWs

The substrate selection experiment was carried out using VFW microcosms (same methodology as macrophyte selection experiments). VFWs were planted with *I. pseudacorus*. In this experiment, diluted leachate (1:10) was used.

The studied substrates were LECA, fine sand, coarse sand, and gravel. Different substrate layer configurations were used according to Fig. 2. The gravel used in this experiment consisted of broken granite stone. The particle size of the substrates ranged from 1 to 2 cm for LECA, 2 to 3 cm for gravel, 0.3 to 0.6 cm for coarse sand, and 0.1 to 0.2 for fine sand.

In this experiment, 8 L per day of landfill leachate was loaded in the VFWs, with a hydraulic loading rate (HLR) of 0.1 m day⁻¹. Sampling was performed before and after treatment from January to March. Samples were collected on a weekly basis.

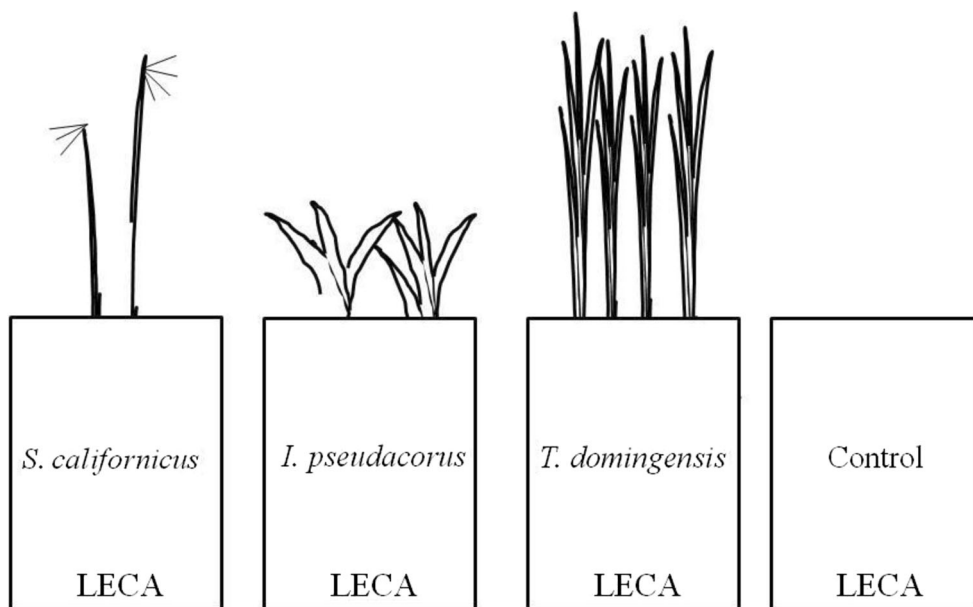
Hybrid constructed wetlands

Considering that a second anaerobic stage would enhance the denitrification processes and chemical oxygen demand (COD) removal, an experiment was carried out studying hybrid constructed wetlands (HWs). The wetlands were planted with *T. domingensis* and *I. pseudacorus*. Four HWs were compared (Fig. 3):

- HW1: VFW (*T. domingensis*)-FWSW (*T. domingensis*)
- HW2: VFW (*T. domingensis*)-HSSW (*T. domingensis*)
- HW3: VFW (*I. pseudacorus*)-FWSW (*T. domingensis*)
- HW4: VFW (*I. pseudacorus*)-HSSW (*I. pseudacorus*)

According to the results obtained in the previous experiment, the substrate used for VFWs was LECA and coarse sand.

Fig. 1 Schematic representation of VFWs with the studied macrophytes for raw and diluted experiments



I. pseudacorus did not develop in FWSW. Therefore, FWSW with *T. domingensis* were used in HW3. To enhance aeration, PVC pipes were installed in the HSSWs. The FWSW microcosms were built using plastic reactors (20 × 40 × 30 cm depth) filled with 12 kg of soil. The HSSW microcosms were constructed using similar plastic reactors as FWSWs. The substrate used was LECA. To enhance aeration, PVC pipes were installed in the HSSWs. After passing through VFWs, the treated leachate was loaded into second stage wetlands. Hydraulic retention time (HRT) was 7 days. The experiment lasted 5 weeks. Effluent samples were collected before and after treatment in each stage.

Analytical methods

Conductivity was measured with a YSI 33 conductimeter, and pH with an Orion pH-meter. Chemical analyses were performed following APHA (2012). NO₂⁻ was determined by coupling diazotation followed by a colorimetric technique.

NH₄⁺ and NO₃⁻ by potentiometry (Orion ion-selective electrodes, sensitivity: 0.01 mg L⁻¹ of N, reproducibility: 2%). Inorganic total nitrogen (Inorg. TN) was estimated as the sum of NH₄⁺, NO₃⁻, and NO₂⁻. COD was determined by the open reflux method and biochemical oxygen demand (BOD₅) by the 5-day BOD test (APHA 2012).

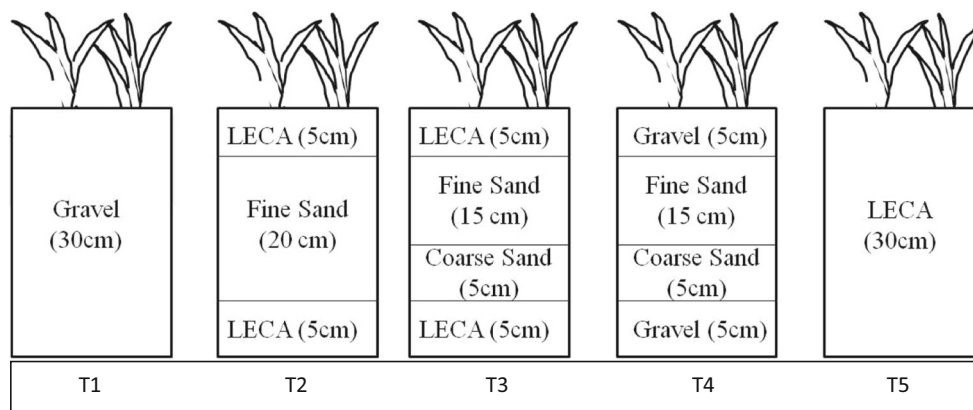
Calculation

Mass removal efficiency calculations were estimated considering not only concentrations but also volumes. Evapotranspiration (ET) was measured in each CW. Removal was calculated according to eq. (1):

$$\text{Removal (\%)} = [(C_{in} V_i - C_{out} V_{out}) / C_{in} V_{in}]^* 100 \quad (1)$$

where C_{in} is the inlet concentration (mg L⁻¹), V_{in} the inlet volume (L day⁻¹), C_{out} and V_{out} are the outlet concentration and outlet volume, respectively.

Fig. 2 Schematic representation of VFWs according to the different substrate (depth of each substrate is indicated in each case)



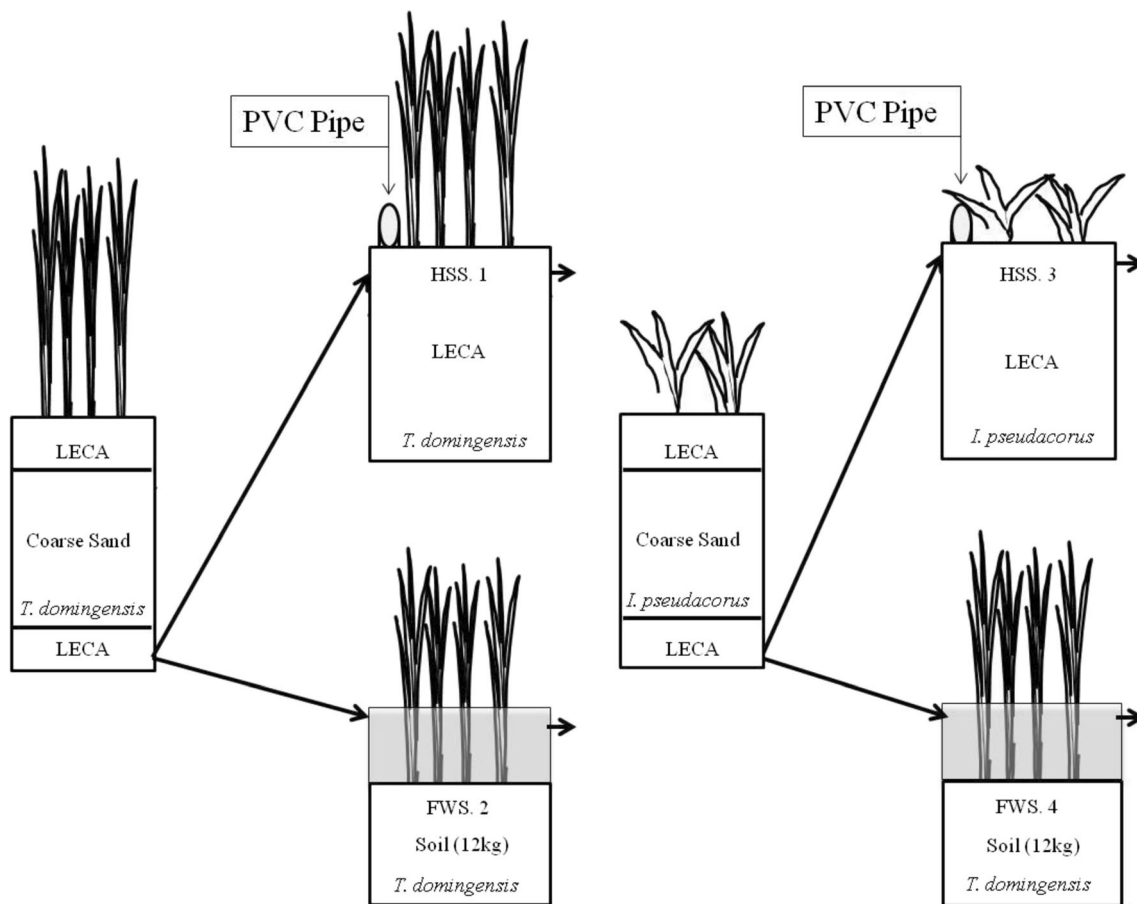


Fig. 3 Schematic representation of HWs. Number indicates which HW it represents

Statistical analysis

All CWs were arranged in triplicate. One-way analysis of variance (ANOVA) was used to determine whether significant differences existed in contaminant concentrations among different treatments. Duncan's test was used to differentiate means where appropriate. In all comparisons, a level of $p < 0.05$ was used.

Results and discussion

Macrophyte selection experiments

Raw leachate

Raw landfill leachate used in the experiments presented high conductivity, a low BOD_5/COD ratio (0.28) and high ammonium concentrations (Table 1). According to Renou et al. (2008), those are characteristics of a leachate from an old landfill. Nitrogen was mainly in the form of ammonium (2484 mg L^{-1}) while mean nitrate concentration was 12.5 mg L^{-1} . Mean ammonium concentrations were

remarkably higher than those reported by other authors in landfill leachates: 238 mg L^{-1} (Akinbile et al. 2012), 642 mg L^{-1} (Bulc 2006), and 264 mg L^{-1} (Reddy et al. 2013). Nitrite concentration was below the detection limit of the method.

After treatment, a significant decrease in ammonium concentration was registered. Removals were 69, 66, and 66% for VFWs planted with *S. californicus*, *I. pseudacorus*, and *T. domingensis*, respectively, and 45% for the control. Nitrate removal was 43, 52, and 36% for *S. californicus*, *I. pseudacorus*, and *T. domingensis*, respectively, and 12% for the control (Table 1). There were no significant differences among macrophyte species regarding ammonium and nitrate removal. However, the control showed significantly lower ammonium removal if compared with the VFWs with macrophytes.

COD removal was 50, 44, and 44% for VFWs planted with *T. domingensis*, *S. californicus*, and *I. pseudacorus*, respectively, and 50% for the control. No significant differences were found among treatments and the control. Low removal in COD is explained due to the recalcitrant characteristic of an old landfill leachate (Renou et al. 2008). BOD_5 presented removal of 80, 79, and 74% for *T. domingensis*,

Table 1 Chemical composition of raw landfill leachate before and after treatment with different macrophytes, and percent removal (%) of each parameter (1-week experiment with two samplings, three VFWs for eachtreatment). Different letters represent statistically significant differences among treatments. Conductivity ($\mu\text{mho cm}^{-1}$, 25 °C), concentrations (mg L^{-1})

Treatments	pH	Conductivity	COD	BOD ₅	NH ₄ -N	NO ₃ -N	NO ₂ -N
Initial	8.4 ± 0.3a	23,800 ± 1329a	3023 ± 380a	837 ± 133a	2484 ± 203a	12.5 ± 2.2a	2497 ± 318a
<i>T. domingensis</i>	8.6 ± 0.3a	15,300 ± 976b	1495 ± 278b	170 ± 54.3b	845 ± 71.4b	8 ± 1.3b	1123 ± 93.2b
Removal (%)	–	–	50	80	66	36	55
<i>S. californicus</i>	8.4 ± 0.4a	11,420 ± 841b	1705 ± 304b	220 ± 55b	770 ± 71.5b	7.1 ± 2.2b	1021 ± 80.2b
Removal (%)	–	–	44	74	69	43	59
<i>I. pseudacorus</i>	8.3 ± 0.3a	13,500 ± 890b	1692 ± 290b	172 ± 55.2b	842 ± 77.7b	6 ± 1.1b	1123 ± 91.1b
Removal (%)	–	–	44	79	66	52	55
Control	8.7 ± 0.4a	15,655 ± 742b	1344 ± 268b	331 ± 60.1c	1351 ± 77.9c	7.9 ± 0.8b	1774 ± 143.3c
Removal (%)	–	–	55	60	45	12	29

I. pseudacorus, and *S. californicus*, respectively. The control showed 60% removal and significantly lower BOD₅ removal if compared with the VFWs with macrophytes.

Despite the satisfactory contaminant removal, plants did not tolerate raw landfill leachate showing senescence after 1 week. Therefore, these results are not valid because the system is not sustainable for long-term treatment of the raw leachate. The experiment was ended, leachate was drained, and tap water was added to the wetland microcosms. After 7 days, macrophytes showed new shoots demonstrating resilience capacity. As a consequence, VFWs were loaded with diluted landfill leachate.

Diluted leachate

Macrophytes tolerated 1:10 diluted landfill leachate. VFWs with *I. pseudacorus* and *T. domingensis* showed significantly higher contaminant removals than VFWs with *S. californicus* (Table 2). Ammonium removal was 53, 59, and 38% for VFWs with *T. domingensis*, *I. pseudacorus*, and *S. californicus*, respectively, and 26% for the control. A et al. (2017) studied at laboratory-scale a VFW for the treatment of a synthetic landfill leachate and found ammonium removals of 44–73% in systems planted with *Juncus effusus* and 46–76% in systems planted with *Phragmites australis*. Nitrite removals were 17, 37, and 10% for VFWs planted with *T. domingensis*, *I. pseudacorus*, and *S. californicus*, respectively, and 6% for the control. As expected in a VFW, an increase in mean nitrate concentration in leachate from 7.2 to 34.3 mg L⁻¹ was observed due to the nitrification process (Vymazal 2007).

The control showed significantly lower COD removals than planted VFWs (16%). COD removals showed significant differences among VFWs with *T. domingensis*, *I. pseudacorus*, and *S. californicus* (50, 48, and 39%, respectively). Lavrova (2016) studied the treatment efficiency of a landfill leachate using laboratory-scale VFWs. Significant

removal efficiency of COD (95%) and BOD (96%) was achieved. According to Yalcuk and Ugurlu (2009), low COD removal can be explained due to poor active microorganisms present in the media of the VFW during the first months of experimentation. Such findings agree with our results: low removals were observed due to the fact that plants were not fully developed and presented a poor biofilm attachment in roots. BOD₅ removal was of 51% for VFWs with *T. domingensis* and *S. californicus*, 36% in *I. pseudacorus*, and 20% for the control. Low removal of COD and BOD₅ is explained due to low HRT in VFWs. The BOD₅/COD ratio is a factor that explains age and biodegradability of leachate. When the ratio is lower than 0.3, it is considered an old leachate with low biodegradability. In the experiment using diluted leachate, the BOD₅/COD ratio was 0.18, while in raw wastewater, it was 0.13, which explained poor COD removal (Wojciechowska et al. 2016).

Song et al. (2018) determine the most suitable macrophyte for the treatment of a landfill leachate. These authors found that in comparison with *Phragmites australis*, *Typha angustifolia* showed the most promising potential for remediation, reaching the highest aboveground biomass and demonstrating maximum N concentrations in tissues when grown in leachate filled tank for 6 months. In our work, *T. domingensis* and *I. pseudacorus* tolerated wastewater conditions, while *S. californicus* showed senescence symptoms and the worst performance in contaminant removal.

Substrate selection experiments

pH did not show significant differences among treatments (Table 3). Except for pH, all parameters showed significant differences between the initial and final values of each treatment. After the treatment with LECA (T5), the leachate showed the lowest conductivity. Ammonium removal did not present significant differences among treatments. Although initial

Table 2 Chemical composition of diluted landfill leachate before and after treatment with different macrophytes and percent removal (%) of each parameter (five samplings, three VFWs, $n = 15$ for each treatment).Different letters represent statistically significant differences among treatments. Conductivity ($\mu\text{mho cm}^{-1}$, 25 °C), concentrations (mg L^{-1})

Treatments	pH	Conductivity	COD	BOD ₅	NH ₄ -N	NO ₃ -N	NO ₂ -N
Initial	8.5 ± 0.4a	3500 ± 300a	353 ± 89a	62.9 ± 13.4a	178 ± 61.1a	7.2 ± 5.2a	33.4 ± 29.5a
<i>T. domingensis</i>	8.3 ± 0.3a	2100 ± 200b	176 ± 54b	30.7 ± 9.3b	83.1 ± 40.8b	40.1 ± 38b	27.6 ± 24.1a
Removal (%)	–	–	50	51	53	–	17
<i>S. californicus</i>	8.1 ± 0.2a	2100 ± 150b	214 ± 60c	30.7 ± 9.4b	110 ± 24c	45 ± 39b	30 ± 28.1a
Removal (%)	–	–	39	51	38	–	10
<i>I. pseudacorus</i>	7.9 ± 0.3a	2200 ± 200b	183 ± 55b	40.3 ± 9.9b	73.5 ± 32.6b	17.8 ± 19.4b	20.9 ± 20.8a
Removal (%)	–	–	48	36	59	–	37
Control	8.1 ± 0.3a	3000 ± 300c	294 ± 44.2c	50.3 ± 10.1c	132 ± 45.1c	10.1 ± 6a	31.2 ± 18.1a
Removal (%)	–	–	16	20	26	–	6

concentrations were higher than the prior experiment, showing the high chemical variability of leachate, ammonium removals remained in the range of 47–54%. According to Lee et al. (2009), the development of the root system affects ammonium removal due to the fact that O₂ availability is higher in a mature system. Ammonium removal may be expected to increase with system maturity. Regarding nitrate and nitrite, removal efficiencies were negative, as expected in a VFW, in agreement with the results reported by Butterworth et al. (2013), Vymazal (2007), and Molle et al. (2015).

COD and BOD₅ did not show significant differences among substrates, except T1 that showed the lowest removal (19 and 18%, for COD and BOD₅, respectively). BOD₅/COD ratio ranged between 0.14 and 0.16, indicating a low biodegradability in wastewater (Wojciechowska et al. 2016).

In this experiment, VFWs with LECA showed a higher removal of COD, ammonium, and conductivity than those of gravel, which have a low capacity for adsorption. The key

role of substrates that present large specific surfaces, large micropores, and high cation exchange capacities in nitrogen transformations in CWs was reported by Liu et al. (2014). Regarding gravel, another disadvantage observed in this experiment was a decrease in macrophyte growth. Gravel has a pointed shape that tears plant root tissues when they penetrate the substrate. This produces a significant stress causing a lack of growth. LECA has the advantage to be round-shaped and light. When VFWs were dismantled, the VFWs with gravel showed a more poorly developed root system than the observed in the other substrates. When building a real CW, these problems can be magnified and system operation may become compromised.

Our results showed that LECA, coarse, and fine sand are appropriate substrates for treating landfill leachate. However, in our VFWs, clogging was a concern when using fine sand. For this reason, coarse sand and LECA were selected.

Table 3 Chemical composition of diluted landfill leachate before and after treatment with different substrates and percent removal (%) of each parameter (five samplings, three VFWs for each treatment). Differentletters represent statistically significant differences among treatments. Conductivity ($\mu\text{mho cm}^{-1}$, 25 °C), concentrations (mg L^{-1})

Treatments	pH	Conductivity	COD	BOD ₅	NH ₄ -N	NO ₃ -N	NO ₂ -N
Initial	7.9 ± 0.3a	5030 ± 205a	357 ± 45a	48.5 ± 8.8a	284 ± 33a	16 ± 7a	< 0.005a
T1	7.7 ± 0.2a	4210 ± 150b	288 ± 38b	39.8 ± 6.3b	151 ± 40b	27 ± 2.3b	3.0 ± 1.1b
Removal (%)	–	–	19	18	47	–	–
T2	7.5 ± 0.1a	4000 ± 200b	213 ± 40c	34.6 ± 5.4c	134 ± 42b	34 ± 2.4c	0.8 ± 0.7b
Removal (%)	–	–	40	29	53	–	–
T3	7.5 ± 0.1a	4030 ± 175b	213 ± 39c	34.1 ± 5.4c	131 ± 43b	36 ± 2.8c	1.4 ± 1.1b
Removal (%)	–	–	40	30	54	–	–
T4	7.4 ± 0.2a	3900 ± 200b	229 ± 40c	35.1 ± 5.5c	149 ± 40b	29 ± 2.6b	2.5 ± 1.3b
Removal (%)	–	–	36	27	47	–	–
T5	7.7 ± 0.2a	3550 ± 175c	228 ± 39c	35.3 ± 5.5c	135 ± 42b	35 ± 2.1c	3.4 ± 1.5b
Removal (%)	–	–	38	27	52	–	–

Hybrid constructed wetlands

First stage

All parameters showed significant differences before and after VFW treatments (Tables 4 and 5). In both treatments, pH tended to neutrality. COD initial values (519 ± 16.5) were significantly higher than those registered in previous experiments. COD removal was lower than in previous experiments, with 16 and 18% in VFWs planted with *T. domingensis* and *I. pseudacorus*, respectively. High COD concentrations combined with its recalcitrant form could explain the poor removal of this parameter.

Ammonium removal was 55% in both VFWs. Landfill leachate used in this experiment presented higher ammonium concentration (478 mg L^{-1}) than that used in previous experiments (178 mg L^{-1} , Table 2, and 284 mg L^{-1} , Table 3). However, plants did not show toxicity symptoms. There were no significant differences for ammonium and COD removal between the VFWs planted with *T. domingensis* and *I. pseudacorus*. However, VFWs planted with *T. domingensis* presented higher Inorg. TN removal than VFWs planted with *I. pseudacorus* (59 and 48%, respectively). Nitrate and nitrite concentrations increased due to the nitrification process in both cases, indicating that a second stage for denitrification is necessary. A second anaerobic stage could improve COD and nitrogen removal in landfill leachate (Wu et al. 2016).

Comparison of HWS

Comparing the different wetlands of the second stage, high ammonium removal was achieved after all treatments (Tables 4 and 5). Significantly higher ammonium and Inorg TN removals were registered in the FWSW planted with

T. domingensis of HW1 than in the other wetlands, while the lowest ammonium removal was registered in HSSW planted with *I. pseudacorus* of HW4. Ammonium removal achieved under anaerobic conditions is probably due to nitrifying bacteria from VFWs. According to Molle et al. (2008), influent coming from a VFW can inoculate the HSSWs with nitrifying bacteria, improving nitrification rate.

Regarding final efficiencies, all HWS showed high ammonium and Inorg TN removals. Significantly higher ammonium and Inorg TN removals were registered in HW1 than in the other HWS (94 and 91%, respectively). This fact is due to the higher removals of ammonium, nitrate, and Inorg. TN registered in the second stage of this system (87, 67, and 83%, respectively) than in the other second stages.

The nitrate concentration decreased in FWSs, probably due to anoxic conditions and N_2 volatilization enhanced by high temperatures. High nitrate concentration registered in HW2 and HW4 was probably due to an enhanced nitrification process owing to PVC pipes that aerated the system (Butterworth et al. 2013). According to Wu et al. (2016), during the treatment of a pig manure effluent using an aerated HSSW, aeration favored ammonium removal, while nitrate and nitrite were produced in the effluent because of the nitrification process.

There were no significant differences in COD decrease among HW1, HW3, and HW4. The lowest COD removal was registered in HW2. After HWS, COD decreased between 58 and 66%. Final COD meet Argentinean law regulatory limits for this effluent ($350 \text{ mg L}^{-1} \text{ O}_2$), except in the case of HW2. Wojciechowska (2017) evaluated the performance of a multistage HSSW treating municipal landfill leachate during 3 years of operation. The average COD removal efficiency varied from 47.8 to 86.6%, and the average total nitrogen removal efficiencies were 98.5%, 68.9%, and 79.6% in subsequent research periods.

Table 4 Measured parameters (mean \pm standard deviation) at the inlet and outlet of each wetland planted with *T. domingensis* (five samplings, three CWs for each treatment). Different letters represent statistically

significant differences among treatments. Conductivity ($\mu\text{mho cm}^{-1}$, 25°C), concentrations (mg L^{-1}). Final removal (%) of HW1 and HW2

Parameters	Inlet	Outlet			Removal		
		First stage		Second stage	First stage		Final
		VFW	HW1 FWSW	HW2 HSSW	HW1 V F W - FWSW	HW2 V F W - HSSW	
pH	8 ± 0.1	7.3 ± 0.1	7.7 ± 0.2	7.3 ± 0.1			
Conductivity	5203 ± 130	4280 ± 500	2580 ± 880	3192 ± 1058			
COD	519 ± 16.5	438 ± 80.5	258 ± 68	314 ± 41	16%a	65%b	58%c
N-NH ₄ ⁻	478 ± 133	214 ± 72.7	40.8 ± 39.0	57.6 ± 53.8	55%a	94%b	92%c
N-NO ₃ ⁺	11.5 ± 7.5	23.9 ± 13.8	11.1 ± 8.7	51.3 ± 31.8	–	32%	–
N-NO ₂ ⁺	0.7 ± 0.1	13.1 ± 12	10.7 ± 13.3	10.7 ± 7.7	–	–	–
Inorg TN	491	251	62.6	119	59%a	91%b	83%c

Table 5 Measured parameters (mean \pm standard deviation) at the inlet and outlet of each wetland planted with *I. pseudacorus*, with exception of FWSW which were planted with *T. domingensis* (five samplings, three CWs for each treatment). Different letters represent statistically significant differences among treatments. Conductivity ($\mu\text{mho cm}^{-1}$, 25 °C), concentrations (mg L^{-1}). Final removal (%) of HW3 and HW4

Parameters	Inlet	Outlet			Removal		
		First stage	Second stage		First stage	Final	
		VFW	HW3 FWSW	HW4 HSSW		HW3 V F W - FWSW	HW4 V F W - HSSW
pH	8 \pm 0.1	7.4 \pm 0.2	7.8 \pm 0.1	7.4 \pm 0.2			
Conductivity	5203 \pm 130	4366 \pm 505	2676 \pm 762	3130 \pm 677			
COD	519 \pm 16.5	427 \pm 67.5	259 \pm 63	251 \pm 66	18%a	65%b	66%b
N-NH ₄ ⁻	478 \pm 133	213 \pm 56.1	62.5 \pm 34.3	77.5 \pm 34.4	55%a	91%b	89%b
N-NO ₃ ⁺	11.5 \pm 7.5	29.3 \pm 18.1	15.8 \pm 11.8	36.8 \pm 36.3	–	4%	–
N-NO ₂ ⁺	0.7 \pm 0.1	11 \pm 9.2	10.3 \pm 11.9	5.5 \pm 5.3	–	–	–
Inorg TN	491	253	88.6	120	48%a	87%b	83%c

HWs planted with *T. domingensis* are suitable to treat high strength landfill leachates. Biomass and transpiration rate of the plant species should be considered for the selection of the macrophytes to be used in CWs (Milani and Toscano 2013). *T. domingensis* showed higher ET and developed higher biomass than *I. pseudacorus* in all studied wetlands. These experiments were carried out during summer with high temperatures. FWSWs presented higher ET than HSSWs, due to the direct contact of the water column with the atmospheric air. High temperatures also favored N₂ volatilization. In further experiments, winter conditions need to be tested to better understand N removals in HWs for the treatment of this leachate.

HWs have demonstrated to be efficient for ammonium removal (Adyel et al. 2017). The most commonly used hybrid system configuration for ammonium removal is VFW-HSSFW, which has been used for the treatment of both sewage and industrial wastewaters (Kadlec and Wallace 2009; Vymazal 2011; Vymazal and Kröpfelová 2015). In our work, HWs composed by VFW-FWSW presented the best performance in the treatment of landfill leachate with high concentrations of ammonium. Vymazal (2013) compared different configurations of hybrid systems operating all over the world. He concluded that all types of HWs are more efficient in TN removal than single CWs and that the most used VFW-HSSW hybrid systems did not show significant differences in ammonia removal with other hybrid system configurations.

CWs are used to treat municipal sewage, as well as agricultural and mine drainage, industrial effluents, landfill leachate, or stormwater runoff (Guo et al. 2017; Kizito et al. 2017; Vymazal 2018). According to Ogata et al. (2018), CWs were designed to reduce the leachate amount and contaminant removal by 83–100% and 92–99%, respectively. However, there is a lack of knowledge on the evaluation of the capacities of CWs to treat landfill leachate in Argentina. The first studies about this topic have been carried out by our research group (Camaño Silvestrini et al. 2019).

Conclusion

The studied macrophytes did not tolerate raw leachate. However, plants showed resilience ability. In experiments using diluted landfill leachate, *T. domingensis* and *I. pseudacorus* tolerated wastewater conditions, while *S. californicus* showed senescence symptoms.

HWs composed by VF-FWSW presented the best performance in the treatment of landfill leachate with high concentrations of ammonium. *T. domingensis* is a suitable species to be used in this hybrid system. This configuration is not commonly used in hybrid systems for the treatment of wastewater with high ammonium concentrations.

In further studies, a pilot scale VFW-FWSW hybrid system will be constructed in the landfill facility. Water for the dilution of raw leachate would be collected by means of a pump from a river near the landfill. The treated leachate would be reused for irrigation of nearby crops.

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References

- A D, Oka M, Fujii Y, Soda S, Ishigaki T, Machimura T, Ike M (2017) Removal of heavy metals from synthetic landfill leachate in lab-scale vertical flow constructed wetlands. *Sci Total Environ* 584:585:742–750
- Adyel TM, Oldham CE, Hipsey MR (2017) Storm event-scale nutrient attenuation in constructed wetlands experiencing a Mediterranean climate: A comparison of a surface flow and hybrid surface-subsurface flow system. *Sci Total Environ* 598:1001–1014

- Akinbile CO, Yussof MS, Zuki Ahmad AZ (2012) Landfill leachate treatment using sub-surface flow constructed wetlands by *Cyperus haspan*. *Waste Manag* 32:1387–1393
- APHA (2012) Standard methods for the examination of water and wastewater. Amer. Publ. Health Assoc, New York
- Brix H (1997) Do macrophytes play a role in constructed treatment wetlands? *Water Sci Technol* 35(5):11–17
- Bulc T (2006) Long term performance of a constructed wetland for landfill leachate treatment. *Ecol Eng* 26:365–374
- Butterworth E, Dotro G, Jones M, Richards A, Onunkwo P, Narroway Y, Jefferson B (2013) Effect of artificial aeration on tertiary nitrification in a full-scale subsurface horizontal flow constructed wetland. *Ecol Eng* 54:236–244
- Camaño Silvestrini NE, Maine MA, Hadad HR, Nocetti E, Campagnoli MA (2019) Effect of feeding strategy on the performance of a pilot scale vertical flow wetland for the treatment of landfill leachate. *Sci Total Environ* 648:542–549
- Clarke E, Baldwin A (2002) Response of wetlands plants to ammonia and water level. *Ecol Eng* 18:257–264
- Connolly R, Zhao Y, Sun G, Allen S (2004) Removal of ammoniacal-nitrogen from an artificial landfill leachate in downflow reed beds. *Process Biochem* 39(12):1971–1976
- Cooper P (1999) A review of the design and performance of vertical-flow and hybrid reed bed treatment systems. *Water Sci Technol* 40(3):1–9
- Guo L, Lv T, He K, Wu S, Dong X, Dong R (2017) Removal of organic matter, nitrogen and faecal indicators from diluted anaerobically digested slurry using tidal flow constructed wetlands. *Environ Sci Pollut Res* 24:5486–5496
- Kadlec RH, Wallace SD (2009) *Treatment wetlands*, second edition. Boca Raton. CRC Press, Florida
- Kadlec RH, Zmarthie LA (2010) Wetland treatment of leachate from a closed landfill. *Ecol Eng* 36:946–957
- Kizito S, Lv T, Wu S, Ajmal Z, Luo H, Dong R (2017) Treatment of anaerobic digested effluent in biochar-packed vertical flow constructed wetland columns: role of media and tidal operation. *Sci Total Environ* 592:197–205
- Kjeldsen P, Barlaz MA, Rooker AP, Baum A, Ledin A, Christensen T (2002) Present and long-term composition of MSW landfill leachate: a review. *Environ Sci Technol* 32:297–336
- Lavrova S (2016) Treatment of landfill leachate in two stage vertical-flow wetland system with/without addition of carbon source. *J Chem Technol Metall* 51(2):223–228
- Lee CG, Fletcher TD, Sun G (2009) Nitrogen removal in constructed wetland systems. *Eng Life Sci* 9(1):11–22
- Liu M, Wu S, Chen L, Dong R (2014) How substrate influences nitrogen transformations in tidal flow constructed wetlands treating high ammonium wastewater? *Ecol Eng* 73:478–486
- Maine MA, Suñé N, Hadad HR, Sanchez G, Bonetto C (2009) Influence of vegetation on the removal of heavy metals and nutrients in a constructed wetland. *J Environ Manag* 90(1):355–363
- Maine MA, Hadad HR, Sánchez GC, Di Luca GA, Mufarrege MM, Caffaratti SE, Pedro MC (2017) Long-term performance of two free-water surface wetlands for metallurgical effluent treatment. *Ecol Eng* 98:372–377
- Milani M, Toscano A (2013) Evapotranspiration from pilot-scale constructed wetlands planted with *Phragmites australis* in a Mediterranean environment. *J Environ Sci Health Part A: Tox Hazard Subst Environ Eng* 48(5):568–580
- Molle P, Prost-Boucle S, Lienard A (2008) Potential for total nitrogen removal by combining vertical flow and horizontal flow constructed wetlands: A full-scale experiment study. *Ecol Eng* 34(1):23–29
- Molle P, Lombard Latune R, Riegel C, Lacombe G, Esser D, Mangeot L (2015) French vertical-flow constructed wetland design: adaptations for tropical climates. *Water Sci Technol* 71(10):1516–1523
- Nivala J, Hoos MB, Cross C, Wallace S, Parkin G (2007) Treatment of landfill leachate using an aerated, horizontal subsurface-flow constructed wetland. *Sci Total Environ* 380(1–3):19–27
- Ogata Y, Ishigaki T, Ebie Y, Sutthasil N, Witthayaphirom C, Chiemchaisri C (2018) Design considerations of constructed wetlands to reduce landfill leachate contamination in tropical regions. *J Mater Cycles Waste Manag* 20(4):1961–1968
- Politeo M (2013) Performance of hybrid constructed wetland for piggery wastewater treatment. *Ecol Eng* 51:229–236
- Reddy GB, Forbes DA, Phillips R, Cyrus JS, Porter J (2013) Demonstration of technology to treat swine waste using geotextile bag, zeolite bed and constructed wetland. *Ecol Eng* 57:353–360
- Renou S, Givaudan JG, Poulain S, Dirassouyan F, Moulin P (2008) Landfill leachate treatment: review and opportunity. *J Hazard Mater* 150(3):468–493
- Song U, Waldman B, Park JS, Lee K, Park JS, Lee K, Park SJ, Lee EJ (2018) Improving the remediation capacity of a landfill leachate channel by selecting suitable macrophytes. *J Hydro Environ Res* 20:31–37
- Tanner CC (1996) Plants for constructed wetland treatment systems - A comparison of the growth and nutrient uptake of eight emergent species. *Ecol Eng* 7:59–93
- Vymazal J (2005) Horizontal sub-surface flow and hybrid constructed wetlands systems for wastewater treatment. *Ecol Eng* 25(5):478–490
- Vymazal J (2007) Removal of nutrient in various types of constructed wetlands. *Sci Total Environ* 31(380):48–65
- Vymazal J (2011) Plants used in constructed wetlands with horizontal subsurface flow: A review. *Hydrobiologia* 674(1):133–156
- Vymazal J (2013) The use of hybrid constructed wetlands for wastewater treatment with special attention to nitrogen removal: A review of a recent development. *Water Res* 47(14):4795–4811
- Vymazal J (2018) Constructed wetlands for water quality regulation. In: Finlayson C et al (eds) *The Wetland Book*. Springer, Dordrecht 1313–1320 pp
- Vymazal J, Kröpfelová L (2015) Multistage hybrid constructed wetland for enhanced removal of nitrogen. *Ecol Eng* 84:202–208
- Wojciechowska E (2017) Potential and limits of landfill leachate treatment in a multi-stage subsurface flow constructed wetland – evaluation of organics and nitrogen removal. *Bioresour Technol* 236:146–154
- Wojciechowska E, Gajewska M, Ostojki A (2016) Reliability of nitrogen removal processes in multistage treatment wetlands receiving high-strength wastewater. *Ecol Eng* 98:365–371
- Wu S, Lei M, Lu Q, Guo L, Dong R (2016) Treatment of pig manure liquid digestate in horizontal flow constructed wetlands: effect of aeration. *Eng Life Sci* 16(3):263–271
- Yalcuk A, Ugurlu A (2009) Comparison of horizontal and vertical constructed wetland systems for landfill leachate treatment. *Bioresour Technol* 18(100):2521–2526