Effect of different bypass rates in hybrid vertical-horizontal flow constructed wetlands treating synthetic and real municipal wastewater.

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ABSTRACT

This work reports the performance of a hybrid constructed wetland ($Bp(VF+HF)_{2:1}$) system which consists of a vertical flow (VF) unit followed by a horizontal flow (HF) unit with 2:1 HF/VF area ratio and influent bypass to the HF unit. Treating synthetic wastewater simulating municipal wastewater, optimum total nitrogen (TN) removal (57%) was reached at 39% bypass and 33 g $BOD_5/m^2\cdot d$ (overall system). On the other hand, treating actual municipal wastewater, the system reached 63% TN removal at 30% bypass and 18 g $BOD_5/m^2\cdot d$. Surface removal rates reached 5.5 and 3.0 g $TN/m^2\cdot d$ for synthetic and municipal wastewater. Surface nitrification rate in the VF unit was in the range of 5.0-7.4 and 3.6-3.8 g $N/m^2\cdot d$ for synthetic and municipal wastewater, respectively, indicating a large effect of wastewater characteristics on the nitrification process. Infiltration rate in the VF unit remained high and far from clogging risk. Overall greenhouse gas emissions were 30 (CO_2), 0.11 (N_2O) and 0.41 (CH_4) $g/m^2\cdot d$ which corresponded to emissions factors (relative to total organic carbon and TN influent) of 94% (CO_2), 0.7% (N_2O) and 3.6% (CH_4). Compared to a similar system with 1:2 HF/VF area ratio, TN removal efficiency was similar but surface removal rates were about 3 times higher.

Keywords
Hybrid constructed wetlands, Vertical flow, Horizontal flow, Nitrogen removal, Influent bypass.

INTRODUCTION

Constructed wetlands (CWs) have been established as an alternative to technical wastewater treatment systems for the sanitation of small communities, but single stage CWs are not able to get the more stringent discharge limits for nitrogen. N removal efficiency reported for constructed wetlands is variable, ranging from high removals of over 90% to removals as low as 11%, and even the consideration than N removal efficiency could not exceed 50% existed, even with optimal design (Xu et al., 2013). A main pathway for N removal in a constructed wetland is ammonification (conversion of dissolved organic N to NH_4^+), followed by coupled nitrification (NH_4^+ to NO_2^- to NO_3^-) and canonical denitrification (NO_3^- to NO_2^- to NO to N_2O to N_2) (Saeed and Sun, 2012; Meng et al., 2014). Nitrification is an aerobic chemoautotrophic microbial process and oxygen availability to the nitrifying bacteria is the most rate limiting step observed in N removal in constructed wetlands. Denitrification is an anoxic microbial process and was generally not considered the limiting step in N removal in constructed wetlands, but denitrification and then N removal could be limited by lack of labile carbon or by excessive oxygenation (Xu et al., 2013).

The necessity of maintaining alkalinity of wastewater, sequential aerobic-anaerobic conditions, and availability of organic carbon are the major problems for optimizing classical nitrification-denitrification routes in conventional treatment systems as well as in CWs. It was found that 2.86 g BOD is needed for entire denitrification of 1 g NO_3-N to N_2, but effective removal of nitrogen in Surface CWs usually requires a COD/N of 5 or higher (Meng et al., 2014). Hu et al. (2014) found limited denitrification (TN removal was limited to 60%) in tidal CWs due to carbon deficiency at influent COD/TN ratio of 3.3-3.7, while at COD/TN ratio of 7-9 the system achieved effective TN elimination of about 86%. Foladori et al. (2013) reported partial denitrification at influent COD/TN ratio of 6.0.

Hybrid CWs constituted for a VF unit followed by and HF unit are the simplest configuration in order to obtain sequential aerobic and anaerobic conditions. Among others, the use of this configuration has been recently reviewed by Vymazal (2013) who found that this was the most commonly used hybrid CW system for treatment of both sewage and industrial wastewaters. Reported removal rates ($g/m^2 \cdot d$) were 10.8 (BOD_5), 24.9 (COD), 11.4 (TSS), 0.27 (TP), 2.5 (NH_3-N) and 2.3 (TN) on average. TN removal ranged from 43 to 78% reaching 65% on average. Vymazal (2013) reported that VF+HF hybrid CWs are slightly more efficient in ammonia removal than other hybrid configurations, as well as all types of hybrid constructed wetlands are more efficient in total nitrogen removal than single HF or VF constructed wetlands.

Besides the type of wastewater, environmental conditions, influent concentration and loading rates, the surface area ratio of saturated HF unit to unsaturated VF unit varied largely in the range of 0.5 to 7.6 (2.7 on average), as can be calculated from Vymazal (2013). However, any of the systems reviewed by Vymazal (2013) adopted influent bypass to the second HF unit. In other types of CWs, multi-feeding or bypass of untreated influent to the second unit has been reported (Stefanakis et al., 2011; Hu et al., 2012; Wang et al., 2014; Li et al., 2014). Ávila et al. (2013) reported that a hybrid VF+HF+FWS system treating domestic wastewater (SLR of 3.8 g $BOD_5/m^2 \cdot d$ and 0.8 g $TN/m^2 \cdot d$ on average) reached limited TN removal (46%) and indicated that nitric nitrogen remained almost invariable along the HF and FWS beds. The authors suggested partial bypass from the Imhoff tank to the HF unit or applying higher loading rates in order to promote denitrification.

Among other benefits, influent bypass to the second unit treating nitrified wastewater aim to increase the COD/TN ratio and increase reduction conditions which favour denitrification. Recently, Torrijos et al. (2016a) reported the operation of a hybrid VF+HF CW treating synthetic wastewater with different bypass ratios of untreated influent to the HF unit. The system of Torrijos et al. (2016a) had an HF/VF area ratio of 2.0, being named the $Bp(VF:HF)_{1:2}$ system. This unplanted system reached 50% TN removal at a bypass rate of 50% with maximum nitrification and denitrification rates of 2.2 and 1.6 g $N/m^2 \cdot d$, respectively, for the overall system. However, ammonia and mainly nitrate accumulated in the effluent, and the authors concluded that, even at 50% bypass, operational conditions in HF unit (dissolved oxygen, redox, COD/TN ratio) were not suitable enough for denitrification.

The objective of this work is to check the effect of bypass and HF/VF area ratio on TN removal in a hybrid VF+HF CW. We hypothesised that a lower HF/VF area ratio would require a lower bypass ratio in order to obtain reducing conditions in the second HF unit, thus improving removal and increasing the overall surface removal rate. Thus, a hybrid VF+HF system with HF/VF area ratio of 0.5, so-called the $Bp(VF+HF)_{2:1}$ system, was used for the present study. The second hypothesis was that the kind of wastewater could affect the results and thus the study was carried with both synthetic and real municipal wastewater. Attention was also paid to organic matter removal, as it affect TN removal, clogging risk and gas emissions.

MATERIAL AND METHODS

Synthetic and real wastewater

A synthetic wastewater simulating domestic wastewater (SW) was used during the first part of the study. A concentrate of SW was prepared with the following composition (mg/L): urea (1600), Na-acetate$\cdot 3H_2O$ (2250), NH_4Cl (200), peptone (300), $MgHPO_4 \cdot 3H_2O$ (500), $K_2HPO_4 \cdot 3H_2O$ (400), $FeSO_4 \cdot 7H_2O$ (100), $CaCl_2$ (100), starch (1000), milk powder (2000), dried yeast (900), soy oil (500), $Cr(NO_3)_3 \cdot 9H_2O$ (15), $CuCl_2 \cdot 2H_2O$ (10), $MnSO_4 \cdot H_2O$ (2), $NiSO_4 \cdot 6H_2O$ (5), $PbCl_2$ (2) and $ZnCl_2$ (5). This concentrate was maintained at 4 ºC until the moment of use, when it was diluted with tap water by a factor of 13 in order to prepare the influent SW. The pH of diluted SW was regulated to 7.1 ± 0.3. Real municipal wastewater (MW) was obtained from the municipal treatment plant of A Coruña (serving about 600000 equivalent people) after the pre-treatment operations. MW batches were further left to settle for about 2 h prior to recover the supernatant and remove the settled solids. The supernatant was analysed and then kept at 4 ºC until the moment of use. Mean characteristics of both SW and MW are given in Table 1. $NaHCO_3$ (500mg/L) was added to the influent wastewater during some operational periods as indicated in Section 3.

Table 1. Characteristics of influent wastewater.

Influent	N[a]	pH	TSS	VSS	COD	BOD_5	TN	NH_3-N	NO_3^--N	PO_4^{3-}-P
SW[b]	7	7.0 ± 0.2	120 ± 32	106 ± 10	539 ± 48	260 ± 49	78 ± 8	8 ± 1	3 ± 1	11 ± 2
MW[c]	8	7.2	81 ± 26	73 ± 27	405 ±49	225 ± 44	57 ± 3	45 ± 7	2 ± 1	5.4 ± 1

[a] Number of samples (for PO_4^{3-}-P, N=2). [b] SW: synthetic domestic wastewater. [c] MW: real municipal wastewater. Concentration in mg/L.

Configuration of the hybrid $Bp(VF+HF)_{2:1}$ system

The hybrid system consisted of two methacrylate columns connected in series and operated the first one as unsaturated VF unit and the second one as saturated unit simulating an HF unit. The internal diameter of the columns was 13.9 cm (VF) and 10.2 cm (HF) whilst both had the same active height of 47 cm plus a head height of 12.5 cm. The cross-sectional area of the VF column was twice that of the HF columns, then the system was named as the $Bp(VF+HF)_{2:1}$, being the HF/VF area ratio of 0.5. The VF column medium consisted of a drainage layer (gravel of 6-12 mm particle size), a main filtering medium (FM1, 32 cm height) of 1-3 mm sand (d_{60} 2.5 mm, d_{10} 1 mm), and an upper layer (MF2, 5cm height) of 0-2 mm sand (d_{60} 0.9 mm). The HF column consisted of a drainage layer (gravel of 20 mm particle size) and a unique filtering medium (FM1, 40 cm height) of 6-12 mm gravel. All bed media were crushed granitic gravel and sand.

The influent entered the VF unit over the filtering medium MF2 by means of a peristaltic pump (Dinko Instruments D-21V) in an intermittent mode (12 pulses per day uniformly distributed each 2 hours) and drained by gravity to the bottom of the column and from there drained free to a receiving tank. Resting was applied to the VF unit that operated with a week regime of 3 days feeding and 4 days resting. From the tank receiving the VF effluent, a second peristaltic pump fed the HF column throughout small and frequent pulses (at least 16 pulses a day). The influent to the HF column (composed by VF effluent and raw wastewater at different rates along the study) entered the unit at 3 cm below the gravel surface. This disposition created continuous saturated conditions and simulated a continuous feeding regime in the HF column. A third peristaltic pump created a bypass of raw influent to the HF column. The columns were kept at 20ºC in a thermostatic chamber and the tanks containing the daily influent and effluent wastewater in a fridge at 10 ºC. The units were not planted because of the short operational time of the study carried out and to exclude the variability due to vegetation development.

Sampling and analysis

On a week basis, effluent composite samples were obtained by integrating daily samples and storing them at 4ºC. Obtained samples were analysed in the laboratory for total and volatile suspended solids (TSS, VSS), COD, BOD_5 (only for the final effluent), ammonia, nitrate and TN. pH, oxidation-reduction potential (ORP) and dissolved oxygen (DO) were determined in situ on the effluent stream. Allylthiourea was used as nitrification inhibitor in BOD_5 assays. An integrated pH & redox 26 Crison electrode was used for pH and ORP determination, a selective electrode (Crison 9663) for ammonia and a YSI ProODO electrode for DO. The determination of NO_3^--N was made in a Biochrom Libra S6 spectrophotometer. To analyse TN, samples were first digested with potassium persulfate to oxidize all the nitrogenous compounds to NO_3^- and after this the nitrates were determined by spectrophotometry.

The infiltration rate was periodically determined every two weeks in both columns. Gas emissions (CH_4, CO_2 e N_2O) from the columns were measured at the end of the system operation.

Calculations

The following equations were used to obtain the hydraulic loading rate (HLR), the surface loading rate (SLR), the surface removal rate (SRR) and the percentage removal efficiency (RE):

$$HLR = Q / A$$
$$SLR = HLR \cdot S$$
$$SRR = HLR \cdot (S_{Influent} - S_{Effluent})$$
$$\%RE = SRR \cdot 100 / SLR$$

where Q is the volumetric flow, S the considered substrate concentration and A the cross sectional area of the column.

The influent flow (Q_{INHF}) and concentration (S_{INHF}) to the HF column were obtained by the following equations:

$$Q_{INHF} = Q_{VF} + Q_{Bp}$$
$$S_{INHF} = (Q_{VF} \cdot S_{VF} + Q_{Bp} \cdot S_{WW}) / Q_{INHF}$$
$$Bp\ (\%) = (Q_{Bp} / Q_{VFIN}) \cdot 100$$

Where Q_{VFIN} is the influent flow to the VF unit, Q_{VF} is the VF effluent flow pumped to the HF column, Q_{Bp} is the bypass flow to HF unit, Bp is the bypass flow expressed as percentage of influent flow to VF, S_{VF} is the VF effluent concentration and S_{WW} is the raw influent wastewater concentration.

The surface emission rate (SER) of each gas (in $mg/m^2 \cdot d$) was obtained from the evolution of the percentage of that gas in the confined head space of each column, following the methods described by de la Varga et al., (2015) and Torrijos et al. (2016a).

3 RESULTS

System performance and organic matter removal

The operational characteristics of both VF and HF units are indicated in Table 2. The first part of the study corresponds to the operation with synthetic wastewater and was divided in four operational periods which are differentiated mainly by the amount of bypass applied. During this part of the study, the amount of SW directly fed to the HF unit (%Bp) ranged from 0% to 40% of the influent flow to the VF unit. During the second part of the study, the synthetic influent was changed to real municipal wastewater, the operational variables were again adjusted in order to optimize TN removal and the %Bp ranged from 18% to 34%.

The evolution of several operational parameters is shown in Fig. 1. During Period I, the VF effluent pH showed a low value of 5.7±0.6 on average. In order to avoid a negative effect of low pH on the nitrification and denitrification process, 500 mg/L of $NaHCO_3$ were added from period I towards. In subsequent operational periods, VF effluent pH was in the range of 6.3 to 6.9 on average. The pH in the effluent of HF unit showed always higher values in the range of 6.9 to 7.5. Thus, after period I, a noticeable effect of pH on the performance of the system was discarded. DO and POR clearly varied from period to period and will be analysed in the corresponding Sections.

System performance treating synthetic wastewater

The HLR to the VF unit was maintained practically constant at 119 mm/d (70 g $COD/m^2 \cdot d$) during periods I-III and then was increased to 143 mm/d during period IV (84 g $COD/m^2 \cdot d$). However, the applied HLR in the HF unit progressively increased from 40 to 60 mm/d due to the increase in bypass, while the SLR increased from 12 to 85 g $COD/m^2 \cdot d$. In this way, the overall HLR increased from 76 to 129 mm/d and the overall SLR from 45 to 76 g $COD/m^2 \cdot d$. Initial SLRs were in the usual ranges reported in literature for VF and HF units. However, the bypass clearly increased the SLR in HF unit over the usual values. On the other hand, compared to the previous study of Torrijos et al. (2016a) in a similar system with an HF/VF area ratio of 2.0 ($Bp(VF+HF)_{1:2}$), the VF unit was operated at a HLR and SLR of about 50% higher but the HF unit reached 3 to 6 higher values. As a consequence, the $Bp(VF+HF)_{2:1}$ system operated at overall HLRs and SLRs 3 times higher than the $Bp(VF+HF)_{1:2}$.

Organic matter removal efficiency was very high in the overall system (Fig. 2), in the range of 94% to 99% for TSS, COD and BOD_5 removal. During these periods, removal of organic matter was high in both units, although the VF unit accused the increase in HLR and SLR from period III to IV (Table 2) showing a reduction in the percentage of TSS and COD removal (Fig. 2) derived from the increase in effluent concentrations (Fig. 1). The influent concentration to HF unit clearly increased as the %Bp increased (Periods I to III, Fig. 2) or due to the increase in VF effluent concentration (Period IV). In spite of this, the HF unit showed a high removal percentage during this part of the study treating SW. Fig. 2 also shows the relative effect on COD and TN concentration in HF influent: whilst the TN concentration remained constant at about 62 mg TN/L, the influent COD increased from 63 mg/L (I, 0% Bp) to 200 mg/L (III, 40% Bp) and even to 229 mg/L during period IV. This clearly increased the COD/TN ratio from 0.9 to 3.9 in the influent to the HF unit (Table 2).

Table 2. Operational characteristics for the $Bp(VF+HF)_{2:1}$ system.

PERIOD (days)	I (0-49)	II (50-75)	III (76-104)	IV (105-125)	V (126-153)	VI (154-165)	VII (166-180)
Wastewater	SW	SW	SW	SW	MW	MW	MW
$NaHCO_3$ (mg/L) [a]	0	500	500	500	500 [a]	500	500
Bypass to HF (% Inf. VF)	0	26.0	39.7	38.6	34.4	18.1	30.3
Overall HLR (mm/d)	76.5	96.8	109.3	128.7	124.2	72.6	79.5
VF HLR (mm/d)	117.8	118.3	120.3	142.8	142.2	94.6	93.9
HF HLR (mm/d)	217.8	222.3	295.7	370.6	350.5	206.2	230.2
Overall SLR ($g/m^2 \cdot d$)							
TSS	9.3	11.7	13.2	15.6	9.9	6.0	6.5
COD	45.1	57.0	64.4	75.8	53.0	27.9	30.6
BOD_5	19.4	24.5	27.6	32.6	28.9	16.0	17.5
TN	5.8	7.3	8.2	9.7	7.0	4.3	4.7
VF SLR ($g/m^2 \cdot d$)							
TSS	14.2	14.3	14.6	17.3	11.4	7.8	7.7
COD	69.4	69.7	70.9	84.1	60.6	36.4	36.1
BOD_5	29.8	29.9	30.4	36.1	33.1	20.8	20.7
TN	8.9	8.9	9.1	10.8	8.0	5.6	5.5
HF SLR ($g/m^2 \cdot d$)							
TSS	4.8	8.0	13.7	24.2	22.7	7.5	12.6
COD	11.8	31.9	59.0	85.0	80.0	24.5	32.3
TN	13.7	13.8	19.2	22.0	14.1	12.6	11.3
COD/TN Influent HF	0.86	2.31	3.07	3.86	5.68	1.94	2.84

[a] Bicarbonate was added to the influent from day50 to 125 and again from day 138 to the end of operation.

System performance treating real municipal wastewater

The influent wastewater was changed from synthetic to real municipal wastewater at the beginning of period V. Main differences between SW and MW (Table 1) were related to a slightly lower concentration of MW (15% to 33% lower depending on the considered parameter) and to the predominant form of nitrogen, as the MW was highly ammonified (80% of TN as ammonia in MW against only 10% in SW). However, the last could be of low importance because ammonification in wastewater is usually considered a fast process. However, the biodegradability of some COD fractions and TSS in MW may be slow, at least in anaerobic conditions (Álvarez et al., 2004) that prevail in HF systems. Although the relative biodegradability of SW and MW is not studied in the present work, it is of great interest to compare the performance and operation of the Bp(VF+HF) system working on both substrates.

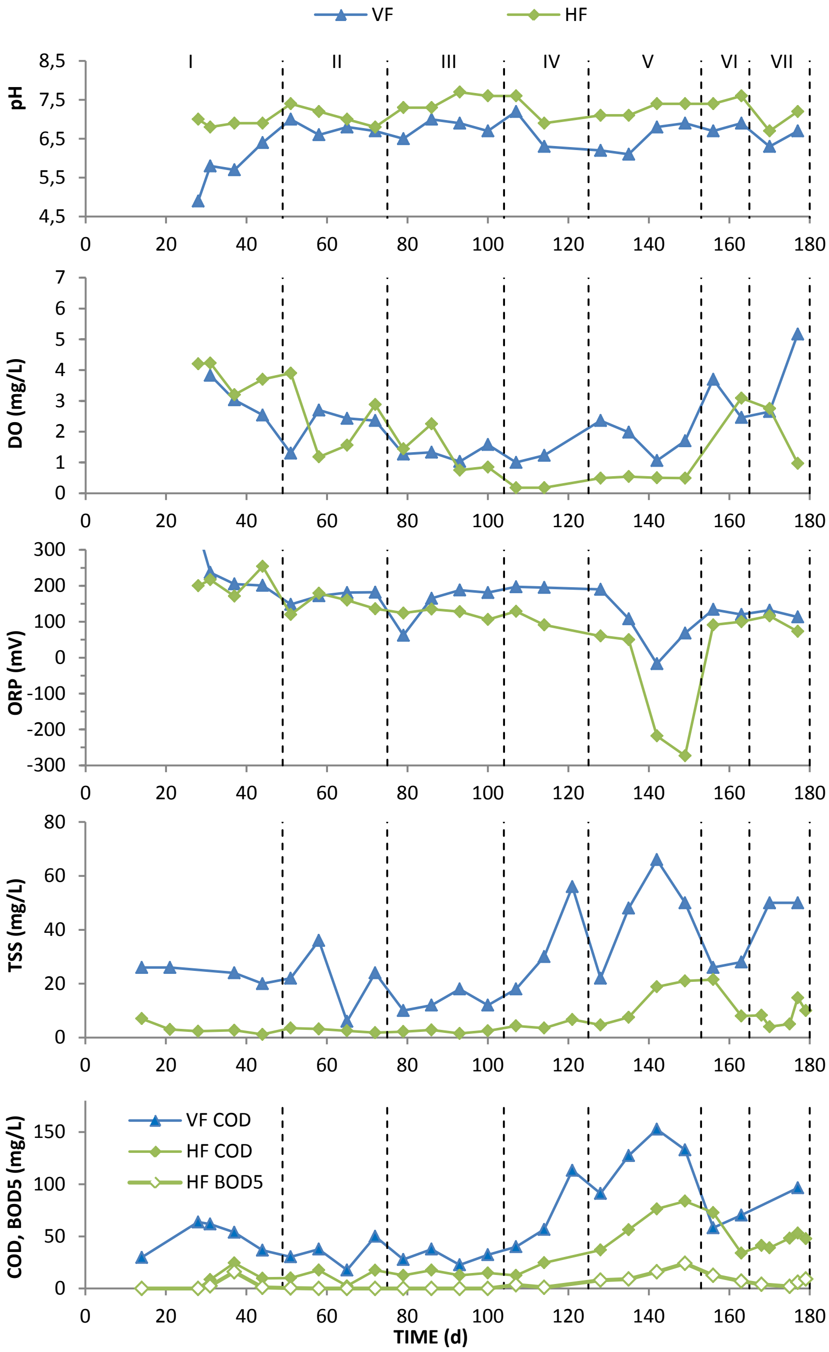

Fig. 1. Evolution of several operational parameters in the effluent of Bp(VF+HF) units.

Fig. 1 shows a clear increased in the final effluent concentration of TSS, COD and BOD_5, which changed from average values of 3, 15 and 2 mg/L for SW (I-IV) to 15, 61 and 12 mg/L for MW (V-VII), respectively. This increase in effluent concentration led to lower percentage removals during periods V to VII (Fig. 2), that averaged 82% TSS, 85% COD and 95% BOD_5. However, both effluent concentration and percentage removals during the treatment of MW continued to be in the typical range of field-scale CWs treating domestic wastewater.

Effluent concentrations of TSS, COD and BOD_5 (as well as ammonia but not TN, as presented in Section 3.2) were highest during period V. In fact, working in similar conditions of HLR during both periods IV and V, and even slightly lower SLR during period V (Table 2), the system increased final effluent concentrations from 5 mg TSS/L, 25 mg COD/L and 1 mg BOD_5/L at period IV to 20 mg TSS/L, 80 mg COD/L and 20 mg BOD_5/L at period V. Considering that final effluent concentrations during period V were too high, and the simultaneous increase in ammonia concentration, the HLR and SLR was decreased during periods VI and VI in order to optimize the system efficiency. This led to a reduction in final effluent concentration and higher removal efficiencies as indicated in Fig. 1 and 2.

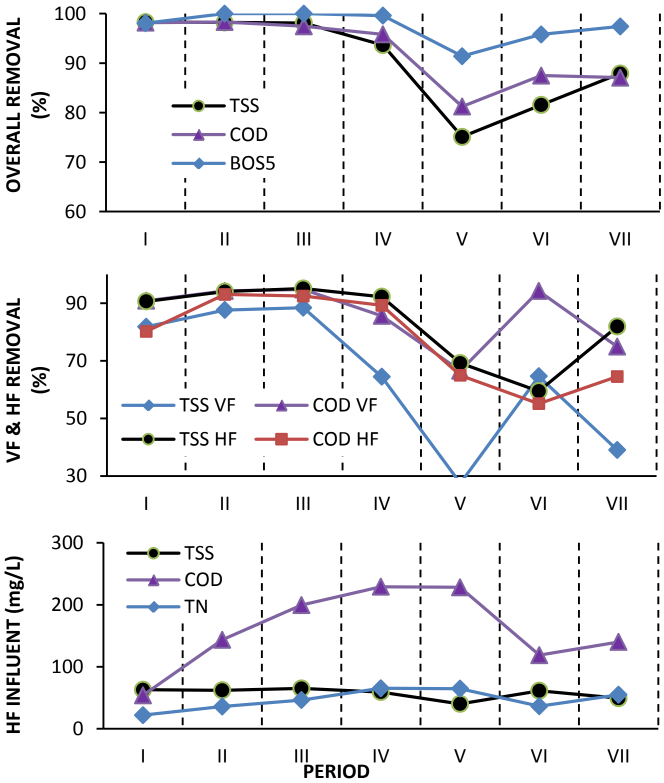

Fig. 2. Percentage removal of TSS, COD and BOD_5 in the overal system (A) and individual units (B), and influent concentration to HF (C).

Nitrogen conversion and TN removal

Treatment of synthetic wastewater

The evolution of ammonia, nitrate and TN in the VF and HF effluents is shown in Fig. 3. The VF effluent maintained a TN concentration nearly constant during the period of SW treatment, with an average value of 57.1 ± 8.4 mg N/L. On the contrary, the TN concentration in HF effluent clearly decreased from periods I and II to period III and IV. This evolution was mainly the result of the denitrification capability of the system, as we will discuss in detail.

Ammonia concentration in VF effluent was high during period I and clearly decreased during Period II because of pH correction (addition of bicarbonate). This ammonia decrease was accompanied by the continuous increase in nitrate concentration (Fig. 3). Thus, the optimum nitrification efficiency in the VF unit was achieved during period III which showed the highest nitrate concentration and the lowest ammonia concentration. This optimum corresponded to SLRs of 71 g COD/m^2·d, 30 g BOD_5/m^2·d and 8 g TN/m^2·d in which the VF unit reached an ammonia concentration of 9 mg N/L. This can be considered the higher SLR to be applied in the VF unit, because the operation with SLRs about 20% higher (Period IV, Table 2) led to an increase of ammonia to 17 mg N/L.

Ammonia concentration in HF effluent was slightly lower than ammonia concentration in the VF effluent during periods I and II (no or low bypass) and slightly higher for periods III and IV, due in part to the effect of increasing the bypass (Fig. 3). Nitrate concentration in HF effluent progressively increased until period II when it reached the maximum value of approximately 50 mg N/L. This suggests a reduced denitrification activity and a reduced effect of bypass ratios below 26% Bp. The high DO and ORP values and the low amount of readily biodegradable organic matter are considered the reasons for the accumulation of a high amount of nitrate in the final effluent. In fact, DO and ORP was very similar in both VF and HF units during periods I to II, the HF unit reaching lower values only at the end of Period III and during Period IV (Fig. 1). Increasing the bypass to about 40% Bp, periods III and IV, significantly decreased the HF effluent concentration of nitrate to approximately 20 mg N/L.

TN removal in VF unit was nearly constant in the range of 16% to 23% during periods I to III, and increased to 30% during period IV, due to the increase in SLR and the more reducing conditions in this unit during period IV. On the other hand, TN removal in HF unit was very low during periods I and II but increased up to 40-45% during periods III and IV. TN removal in the overall system followed the evolution of TN removal in HF system. The highest TN removal for the overall system was obtained during period IV at 39% Bp and COD/TN ratio of 3.9 in the influent to the HF unit. In these conditions, ammonia and nitrate concentration in the final effluent were 19 and 21 mg N/L, respectively. During period IV, the overall system removed 5.5 g TN/m^2·d (57% TN), at SLRs of 76 g COD/m^2·d, 33 g BOD_5/m^2·d and 9.7 g TN/m^2·d. Similar results were obtained during period III at 40% Bp while TN removal was clearly lower at %Bp of 26 (period II).

This results agreed with those of Torrijos et al. (2016a) who reported limited TN removal of 31-41% TN for the $Bp(VF+HF)_{1:2}$ system at %Bp below 25%, whilst at %Bp of 50% the system removed 47-50% TN. However, the system of Torrijos et al. (2016a) operated at overall SLR of 10 g BOD_5/m^2·d and 3.1 g TN/m^2·d which was clearly lower than that applied in the present study. In both cases a high bypass ratio clearly improved the efficiency of TN removal, the $Bp(VF+HF)_{2:1}$ system achieving a higher SRR than the $Bp(VF+HF)_{1:2}$ system.

Treatment of real municipal wastewater

The introduction of real municipal wastewater instead of synthetic wastewater (beginning of Period V) clearly increased the ammonia concentration in both the VF effluent and the final effluent, whilst decreased the nitrate concentration in both effluents to near zero (Fig. 3). This occurred in spite of the lower SLR during period V as compared to period IV (Table 2), and was accompanied by a clear reduction in ORP, mainly in HF unit (Fig. 1). These results indicated that the nitrification process was impaired by the change of SW to MW.

The influent flow in the subsequent Periods VI and VII has been reduced in order to reduce the SLR and improved nitrification. This reduction of the SLR in the VF unit from 61 g COD/m^2·d (Period V) to 36 g COD/m^2·d during periods VI and VII led to the reduction of ammonia, which reached about 18 mg N/L in the final effluent. The bypass rate has been decreased to 18% during period VI, leading to an increase in HF nitrate concentration. The bypass increased again to 30% during period VII, which allowed the system to reduce nitrate concentration in the final effluent and increased TN removal up to 63% (Figure 3).

Optimal conditions for the Bp(VF+HF) system treating real municipal wastewater were those of Period VII, that correspond to SLR of 36 g COD/m^2·d (21 g BOD_5/m^2·d) for the VF unit, %Bp of 30% and COD/TN rate of 2.8 in the influent to the HF unit. In these conditions, ammonia and nitrate concentration in the final effluent were 18 and 9 mg N/L, respectively. SLRs for the overall system during period VII were 31 g COD/m^2·d, 18 g BOD_5/m^2·d and 4.7 g TN/m^2·d. The overall system removed 3.0 g TN/m^2·d (63% TN). Increasing TN removal would require a lower SLR in the VF unit while maintaining the bypass rate to the HF unit.

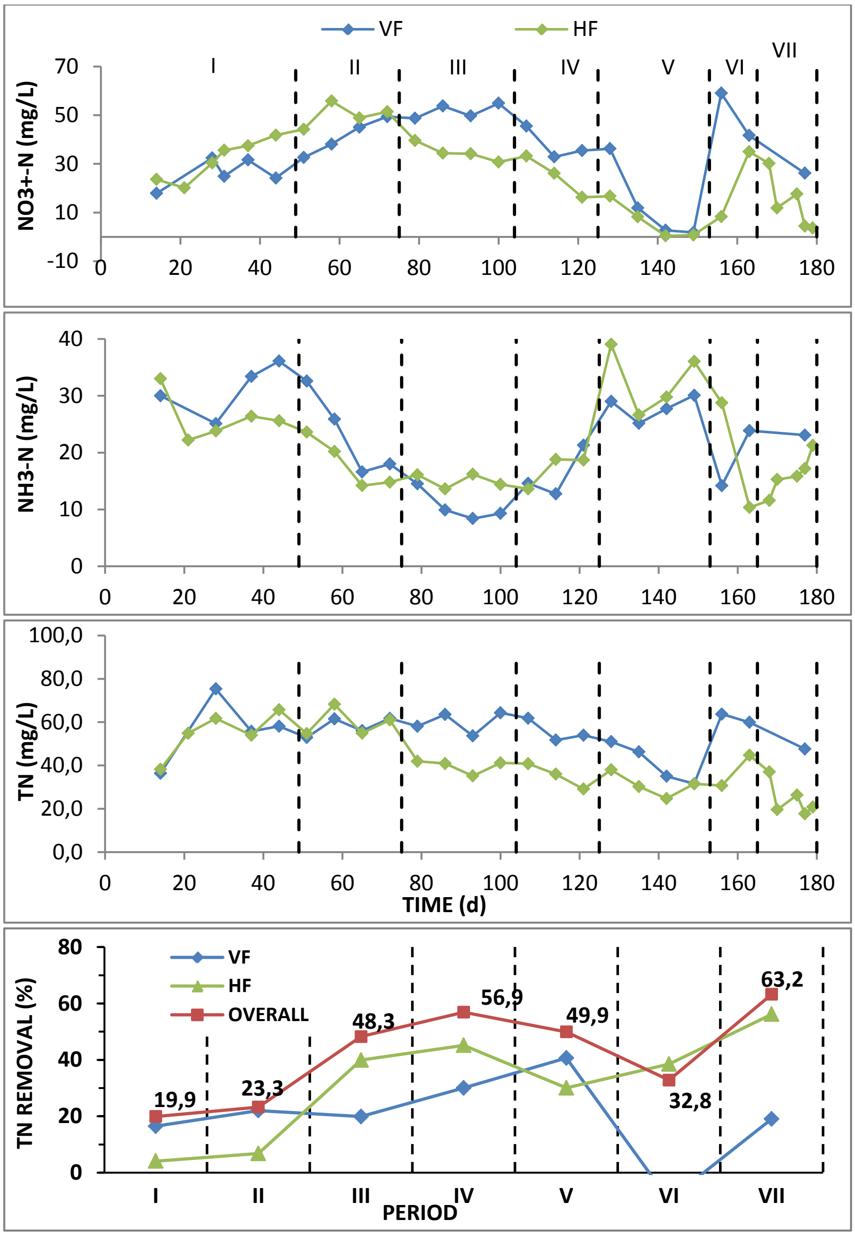

Fig. 3. Evolution of ammonia, nitrate and TN in the VF and HF effluents (A) and evolution of TN removal (B).

Flow profiles in VF unit

Maximum flow ranged from 63 to 130 mL/min. Retard time ranged from 1.4 to 3.0 min whilst mean time ranged from 3.7 to 9.1 min. These parameters indicate a progressive reduction of the retention time with the operation time but also a complete absence of clogging risk. This is a matter of interest because both the VF and the HF units received high SLR of suspended solids which reached 17 (VF) and 24 (HF) g TS/m^2·d during SW treatment. On the other hand, drainage flow from HF column was nearly constant at 1242 ± 196 mL/min on average for all the operation time. This high infiltration flow indicates a total absence of clogging.

Greenhouse gas emissions

Gas emissions (CH_4, CO_2 e N_2O) were measured at the end of operation (day 177). emission factors (EF) are defined as percentage ratio of CO_2-C or CH_4-C emitted to influent total organic carbon (TOC) and as percentage ratio of N_2O-N emitted to influent TN. Conversions factors of 0.5 g TOC/g BOD_5 (Mander et al., 2014) and 0.5 g BOD_5/g COD for HF influent were used in order to obtain EF. Obtained gas emission rates and EF are given in Table 3.

CO_2 emissions averaged 109% (VF) and 38.3% (HF) of influent carbon, being in the range of reported values (Mander et al., 2014; de la Varga et al., 2015; Carballeira et al., 2016). Lower SLR and higher pH in HF unit when compared to VF unit could explain the lower CO_2 EF in HF unit, in which most of the generated CO_2 could leave the system solubilized in the water stream. N_2O emissions reached 160 mg mg $N_2O/m^2 \cdot d$ in VF while they were below the detection limit (14 mg $N_2O/m^2 \cdot d$, EF of 0.04% TN) in HF, thus being in the range of mean emission factors reported by Mander et al. (2014). CH_4 emissions were high in HF than in VF as we could expect due to the respective DO and ORP levels. CH_4 EFs were 1.3% (VF) and 6.3% (HF). Comparatively, Mander et al. (2014) reported mean CH_4 EF of 1.2% (VF) and 4.5% (HF). However, both N_2O and CH_4 EF were higher than those reported by Torrijos et al. (2016a) for the $Bp(VF+HF)_{1:2}$ system receiving lower SLR. Torrijos et al. (2016a) found that methane generation was not observed and N_2O emissions were below 1% TN in VF unit. Higher emissions could be explained by higher SLR in the present study, particularly in the HF unit.

Table 3. Surface emission rate and emission factor for CO_2, CH_4 e N_2O in VF and HF units.

	VF			HF			Overall		
	CO_2	N_2O	CH_4	CO_2	N_2O	CH_4	CO_2	N_2O	CH_4
Emission rate ($mg/m^2 \cdot d$)	38578	160	164	14669	0	873	30021	109	414
Emission factor (%)[a]	108.5	1.0	1.3	38.3	0	6.3	94.2	0.7	3.6

[a] Expressed as %TOC for CO_2 and CH_4 and %TN for N_2O.

Final remarks on the effect of HF/VF area ratio and comparison of municipal and synthetic wastewater

Table 4 summarizes data on selected parameters for the two systems $Bp(VF+HF)_{1:2}$ (Torrijos et al., 2016a) and $Bp(VF+HF)_{2:1}$ (this study) relative to conditions for optimal TN removal. A similar COD/TN of 3.1-3.2 was reached for both systems treating SW but at different bypass ratios of 50% and 40%, respectively. Compared to the reviewed literature, this COD/TN ratio was below the optimum for advanced denitrification which ranged from 5 to 9 (Foladori et al. 2013; Hu et al. 2014; Zhu et al. 2014). This explains the similar but limited TN removal efficiency of about 48-50% in both systems. However, increasing the bypass ratio would not be a good solution because it probably lead to an increase in ammonia concentration in the final effluent. Furthermore, the COD SLR on the second HF step was very high in the case of the $Bp(VF+HF)_{2:1}$ system, which reached 59.0 g $COD/m^2 \cdot d$ at 40% Bp. This high COD SLR is considered the reason for the high methane emissions that reached 6% of the carbon feed. Thus, a limitation of the bypass strategy in order to achieved complete TN removal in this type of CW systems treating domestic or municipal wastewater arises, although somewhat higher TN removal can be expected in planted CWs due to the role of plants in capturing nitrogen as well as in providing additional carbon sources to denitrification.

In spite of the similar TN removal efficiency, the system $Bp(VF+HF)_{1:2}$ showed a higher SRR of both organic matter and TN than the system $Bp(VF+HF)_{2:1}$ (Table 4). Treating SW, the system $Bp(VF+HF)_{2:1}$ with the lower HF/VF ratio accommodated two to three times higher HLR and SLR and removed 4 g $TN/m^2 \cdot d$ whilst the system $Bp(VF+HF)_{1:2}$ removed only 1.6 g $TN/m^2 \cdot d$.

The change of synthetic to real municipal wastewater led to different figures in SLR and SRR of the $Bp(VF+HF)_{2:1}$ system (Table 4). While the TN removal increased from 48% (SW) to 63% (MW), the SLR has to be lowered in order to facilitate nitrification. SNR in the VF unit decreased from 5.8-7.6 g $N/m^2 \cdot d$ (SW) to 2.3-3.4 g $N/m^2 \cdot d$ (MW) while TN SRR in the overall system decreased from 4 to 3.0 g $N/m^2 \cdot d$, respectively. Differences in performance using either SW or MW are of main interest, because most of the studies about new configurations and intensified systems were carried out using synthetic wastewater (Stefanakis et al., 2011; Fan et al., 2013; Foladori et al. 2013).

However, organic matter and TN SLR for the Bp(VF+HF)$_{2:1}$ system treating MW were still higher than those of the Bp(VF+HF)$_{1:2}$ system treating SW (Table 4). SRR of 27 g COD/m^2·d, 17 g BOD$_5$/m^2·d and 3.0 g TN/m^2·d were also higher than the mean SRR reported by Vymazal (2013) for hybrid VF+HF systems (25, 11 and 2.3 g/m^2·d, respectively), in spite of the absence of plants and the lack of system maturation. In general, better results could be expected from plants presence and longer operation times because of the maturation effects (Torrijos et al., 2016b).

Table 4. Selected operation and nitrogen removal efficiency parameters for the Bp(VF+HF)$_{1:2}$ and Bp(VF+HF)$_{2:1}$ systems.

System	Bp(VF+HF)$_{1:2}$	Bp(VF+HF)$_{2:1}$	Bp(VF+HF)$_{2:1}$
Wastewater	SW	SW	MW
HF/VF area ratio	2.0	0.5	0.5
Bypass to HF (% Inf. VF)	50	39.7	30.3
Overall HLR (mm/d)	40.4	109.3	79.5
Overall SLR (g COD/m^2·d)	23.8	64.4	30.6
Overall SLR (g BOD$_5$/m^2·d)	10.2	27.6	17.5
Overall SLR (g TN /m^2·d)	3.1	8.2	4.7
VF SLR (g TSS /m^2·d)	9.3	14.6	7.7
VF SLR (g COD / m^2·d)	45.3	70.9	36.1
HF SLR (g COD /m^2·d)	13.1	59.0	32.3
COD/TN Influent HF	3.2	3.1	2.8
Overall TN removal (%)	50.0	48.3	63.2
Overall SRR (g TN/m^2·d)	1.6	4.0	3.0
VF SNR (g N/m^2·d)	3.8-4.6	5.8-7.6	2.3-3.4
Final (HF) effluent concentration			
NO_3^--N	19.6	33.1	8.6
NH_4^+-N	17.2	14.7	18.1
N Total	37.7	39.1	21.6
Reference	Torrijos et al., 2016a	This study	This study

CONCLUSIONS

The hypothesis that a lower HF/VF area rate would require a lower bypass ratio in order to obtain reducing conditions in the second HF unit and would improve TN removal rate has been confirmed. Treating synthetic wastewater, the Bp(VF+HF)$_{2:1}$ system reached maximum TN removal of 48-57% at 39% Bp, 33 g BOD$_5$/m^2·d and 9.7 g TN/m^2·d of SLR. By comparison, the Bp(VF+HF)$_{1:2}$ system reached maximum TN removal of 50% at 50% Bp, 10 g BOD$_5$/m^2·d and 3.1 g TN/m^2·d of SLR. On the other hand, treating actual municipal wastewater, the Bp(VF+HF)$_{2:1}$ system reached 63% TN removal at 30% Bp, 18 g BOD$_5$/m^2·d and 4.7 g TN/m^2·d of S.

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