Effects of design and operational parameters on ammonium removal by single-stage French vertical flow filters treating raw domestic wastewater

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Abstract

Four pilot-scale single-stage vertical flow filters (of 2.25m² each), treating raw domestic sewage, were studied over 20 months in order to assess the impact of different designs and operational conditions on treatment efficiency. One of them was designed and operated as a standard 1st stage "French" vertical flow constructed wetland unit. The other 3 pilots differed from the standard pilot with respect to the filtration depth, the loading rate or the partial replacement of gravel by zeolite (chabazite), respectively. The pilots were monitored by analysing 24-hour flow-weighted composite samples for TSS, COD_{tot}, COD_d, ammonium, nitrate and carbonate. All pilots showed a high ability to remove TSS and COD_{tot}, with average removal of 81% and 75%, respectively. Increasing the depth of the filtration layer from 40 to 100 cm allowed to significantly improve ammonium removal (81%), whereas the simultaneous increase in hydraulic and organic loads resulted in a deterioration of ammonium and COD_d removals (44% for both parameters). Using zeolite did not induce any observable improvements in ammonium removal under the conditions of the study.

Keywords : Ammonium, Vertical flow constructed wetland, Domestic wastewater, Design

I) Introduction

Constructed wetlands (CWs) for wastewater treatment met an increasing worldwide interest during the past three decades because of their performances, low investment and operational costs and their environmental friendly image. Moreover, this technique is efficient to treat various kinds of effluents such as domestic wastewater, industrial wastewater or combined sewer overflows, etc. (Ávila *et al.*, 2013; Wu *et al.*, 2015; Meyer *et al.*, 2013).

The classical design of "French CW systems" treating raw domestic wastewater (Molle *et al.*, 2005) consists of two vertical flow constructed wetland (VFCWs) stages operating in a sequential mode of feeding and rest periods (3.5 days and 7 days, respectively). The first stage (1.2 m²/population equivalent), composed of three parallel filters filled with gravel, is fed by batches of raw screened wastewater. Most of the suspended solids and a part of the dissolved pollution (organic matter and ammonium) are removed at this stage. The second stage (0.8 m²/pe divided in 2 parallel units) filled with sand ensures a further treatment of dissolved pollution under aerobic conditions. This configuration allows high removal performances on COD_{tot}, TSS and TKN, namely over 90%, 95% and 85%, respectively (Morvannou *et al.*, 2015) and also easier sludge management than other conventional processes. Besides, "French systems" have a high tolerance to variation of hydraulic and organic loads (Molle *et al.*, 2006; Arias *et al.*, 2014).

TKN removal is dependent on various parameters such as wastewater composition, design considerations (media characteristics, design loads…) or external parameters (maintenance, climate). Proper design and optimal operation are needed in order to provide favourable conditions for nitrification. Molle *et al.* (2005) reported that a minimum surface area of 2 m²/p.e. was required in order to achieve full nitrification for a two-stage VFCW configuration. This may be a problem for larger units or when land availability is limited. Recirculation has been reported to improve TKN removal performance (Prigent *et al.*, 2011). Nevertheless, recirculation increases hydraulic loads and can thus negatively affect oxygen transfers. Prost-Boucle and Molle (2012) proposed to limit the hydraulic load to 0.7m/d on the filter in operation in order not to affect nitrification. Oxygen transfer can be increased by implementing passive or active aeration systems (e.g. tidal flow (Sun *et al.*, 2005) or forced bed aeration (Boog *et al.*, 2014; Foladori *et al.*, 2013; Nivala *et al.*, 2013)). However, such intensifications lead to additional operating costs (Austin and Nivala, 2009).

Current methods for design improvement appear to favour more complex and more intensified systems. The objective of the present study was to assess the extent of removal performance improvement by adapting design parameters without increasing energy consumption. Since nitrification is known to be highly sensitive to several operational conditions such as oxygen transfer into the filter, hydraulic and organic loads or the feeding strategies, it was used as an indicator for design optimisation. Four pilot-scale French VFCWs were monitored over 20 months for this purpose. One of them was designed and operated as a standard 1st stage filter according to the French guidelines (Molle *et al.*, 2005) in order to serve as a reference. The design parameters tested were the filter depth (0.4 to 1.0 m), the use of zeolite (chabazite) as filter media and the hydraulic and organic loading rates.

II) Materials and Methods

Experimental setup

Four vertical flow pilot filters of 2.25 m² each were monitored for 20 months, from March 2014 to October 2015. One of them, denoted as Vertical Flow Standard (VFSt), was designed and operated as a standard 1st stage "French" VFCW unit. The other 3 pilots differed from the standard pilot with respect to the filtration depth (Vertical Flow Gravel$^+$, VFG$^+$), loading rate (Vertical Flow High Load, VFHL) or a partial replacement of gravel by zeolite (Vertical Flow Zeolite, VFZ), respectively.

The pilots were all composed, from bottom to the top, of a 15-cm-deep drainage layer made of 16/22 mm grain size cobbles and a filtration layer whose characteristics are given in Table 1. To avoid particulate migration from the filtration layer to the drainage layer in the VFHL pilot, a 10-cm-deep transition layer (grain size 16/22mm) was implemented above the 15-cm-deep drainage layer which was composed of 20-50 mm cobbles as shown in Figure 1.

The pilots were operated outdoors on an experimental site located at the site of a domestic wastewater treatment plant (Jonquerettes, south east of France). This facility allowed us to assess the performance of VFCWs for the treatment of real raw domestic wastewater screened at 20 mm under Mediterranean climate.

A sludge deposit layer was progressively formed at the surface of the filters by accumulation of filtered particles (up to a thickness of 3cm at the end of the monitoring period). The pilots were planted in September 2013 with one year old plantlets of *Phragmites australis* at a density of 6 plants.m^{-2}. According to French guidelines, the pilots were fed for 3.5 days and rested for 7 days. During the feeding periods, 18 batches of 2 cm were applied daily (2 $m^3.h^{-1}$), except for the high load pilot VFHL where 32 batches a day were applied which was considered as the highest acceptable hydraulic load based on full-scale observations. The monitoring started after a commissioning period of five months which was meant to allow for the establishment of microorganisms and reeds.

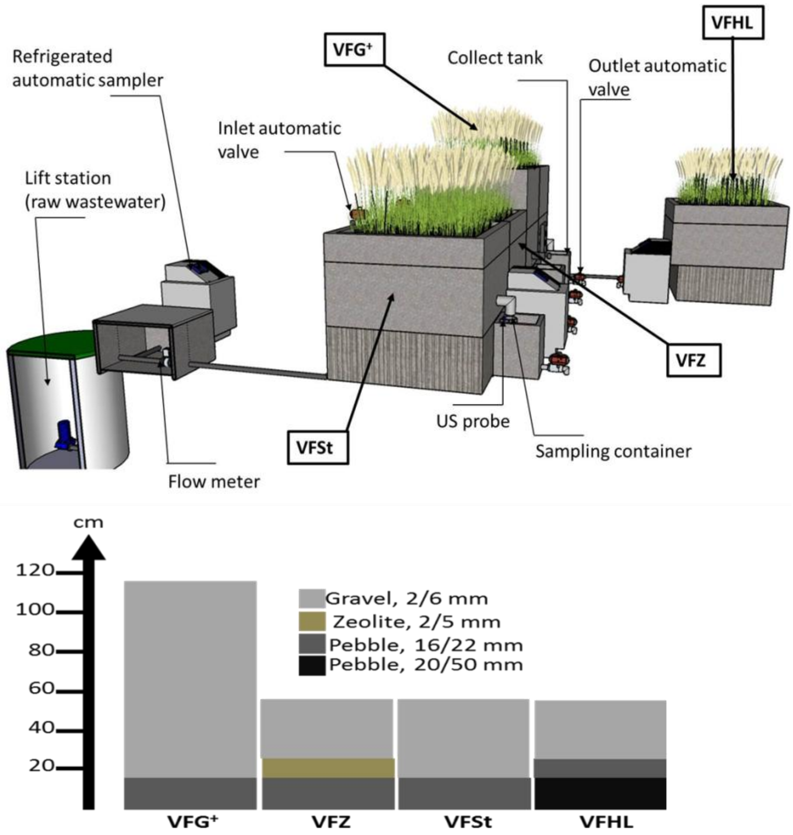

Figure 1 Experimental setup and design characteristics

Table 1 Characteristics of the pilot design

Pilot units	Studied parameters	Filtration layer			Passive aeration location (cm)(1)/(3)	Hydraulic load $(m^3.m^{-2}.d^{-1})^{(2)}$	Organic load $(gCOD.m^{-2}.d^{-1})^{(2)}$
		Material	Depth (cm)	Sampling systems (cm)(1)			
Standard (VFSt)	Unit of reference	Gravel 2/6 mm	40	10 and 30	Bottom	0.36	234 (62)

Deep Filtration (VFG+)	Effect of filtration depth	Gravel 2/6 mm	100	10, 20, 40, 60, 80	Bottom, 30 and 60	0.36	240 (80)
High Load (VFHL)	Effect of hydraulic and organic loads	Gravel 2/6 mm	30	10	Bottom and 30	0.64	536 (276)
Zeolite Chabazite (VFZ)	Effect of sorbent materials	Gravel 2/6 mm Zeolite 2/5 mm	30 + 10	30 and 40	Bottom	0.36	237 (71)

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[1] Depth from the filter surface

[2] Loads are calculated for the filter in operation

[3]All drains are connected to the atmosphere, resulting in a passive aeration from the bottom on the length of the filter. The intermediate passive aeration systems consist of drilled pipes, with connection to the atmosphere, which are crosswise implemented in the filtration layer of the pilot.

Preliminary validation of reference pilot

A preliminary step of validation of the reference unit was required to verify whether the treatment performance of the VFSt was in the range of those usually observed at full-scale 1st stages of a classical French VFCW. For that purpose, inlet and outlet concentrations of the VFSt were compared with a set of data collected from three full-scale treatment plants with the same design (part of data from Morvannou *et al.*, 2015). The inflow of each pilot was also compared in order to confirm that they received the same wastewater during the study so that their performance could be compared.

Experimental monitoring of the pilots

The inlet and outlet water concentrations were assessed for the first and the last day of the feeding periods using refrigerated samplers (Ponsel, ISCO 4700 and Hach, Bühler 2000). 24-hour flow-weighted composite samples were taken from the outlet while, for the inlet, 24-hour composite samples were obtained from one grab sample per batch. Intermediate 24-hour composite samples were taken from different depths (see Table 1) and analysed. Pore water was collected during infiltration by PVC gutters (9 cm and 30 cm of width and length, respectively), located at different depths within the filtration layer, and then stored into pre-acidified 25L polyethylene containers. Each pilot was evaluated for total and dissolved COD (COD_{tot} and COD_d, respectively), TSS, NH_4-N, NO_3-N and $CaCO_3$ using quick method tests (Hach).

Online measurements were also carried out for continuous monitoring of hydraulic and treatment performance dynamics. Inlet flows were determined using an electromagnetic flowmeter (Siemens, SITRANS MAG 5100W) whereas outlet flows were measured with ultrasonic probes (Pil, P43-F4V-2D1-D0-330E) by the rise of the level of effluent drained into a collecting tank. Nitrogen concentrations (NH_4-N and NO_3-N) were continuously monitored at the inlet and the outlet of each pilot at time intervals of 15 and 2 minutes, respectively, using ion selective electrodes (AN-ISE, Hach).

The monitored data were used to compare the performance of pilots. For this purpose, removal rates were calculated on mass basis considering the measured concentrations and the inlet volumetric flows.

Statistical analysis

Experimental results were statistically analysed using R software. Kruskal Wallis tests were carried out on the full set of data, in order to validate that all pilots received the same influent, while Wilcoxon and Student tests were used for pair-wise comparison of each pilot with the reference unit. Significant difference was established at p-value ≤ 0.05.

III) Results and Discussions

1) Validation of control pilot VFSt

The treatment performance of VFSt was compared to full-scale classical French first stage filters (Morvannou *et al.*, 2015) with respect to TSS, COD_{tot} and ammonium as shown in Figure 2. The results of Wilcoxon statistical comparison between VFSt and full-scale VFCWs confirmed that the reference pilot VFSt of the study could be considered as a standard filter. Moreover the pollutant concentrations in the inlet (TKN/COD_{tot}, TSS/COD_{tot} and $TKN/NH_4\text{-}N$) were in the range of what was reported from a survey of almost 3000 treatment plants of small French communities (Mercoiret *et al.*, 2010).

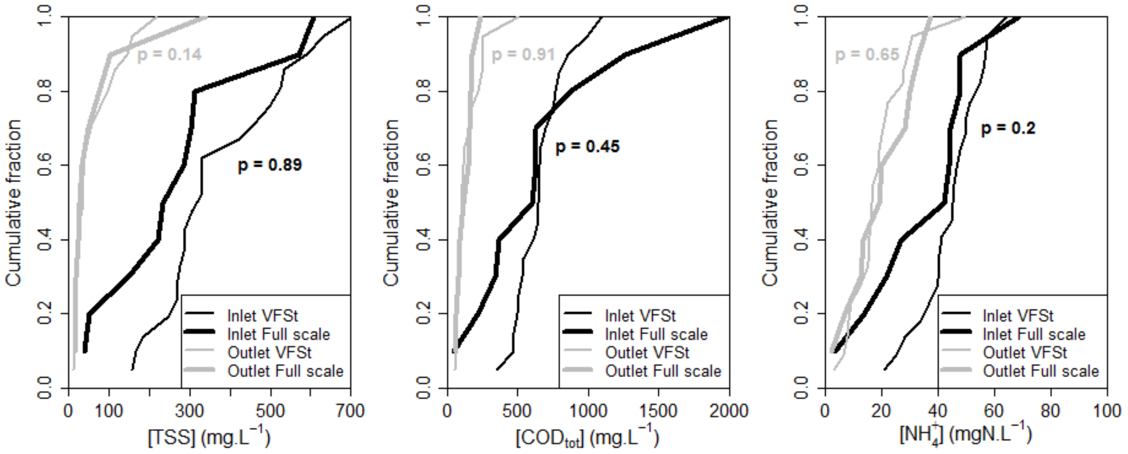

Figure 2 Influent and effluent composition of VFSt and Full scale treatment plants (depth of 40cm) (p-values are the outcome of a Wilcoxon test comparing data from pilot and full-scale systems)

Figure 3 shows the distribution of influent composition over the whole study without distinction between pilots. It can be observed that inlet composition during the study was similar for all pilots, as confirmed by the Kruskal Wallis test comparing average inlet concentrations of each pilot. It was therefore relevant to compare them with the VFSt filter. Nitrate concentrations however were significantly different. This can be explained by a few high values measured in the influent which modified the average value.

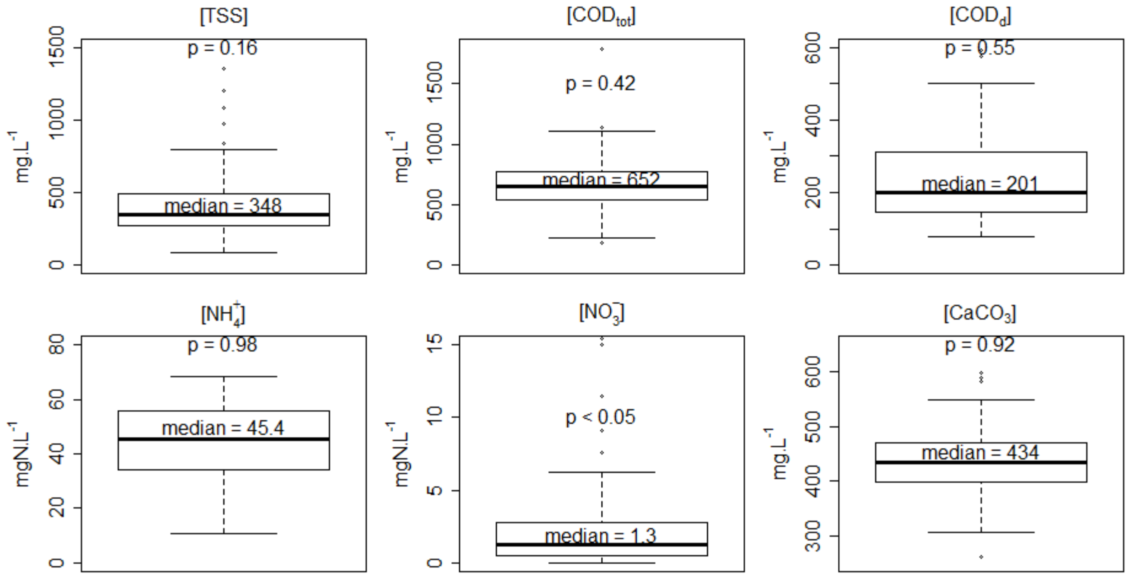

Figure 3 Influent wastewater composition during the study (note that one TSS outlier over 3000mg/L, one COD_{tot} outlier over 1500mgO_2/L, one COD_d outlier over 1000mgO_2/L and four NO_3^- outliers between 10 and 20mgN/L are not shown for visibility reasons). P-values are the outcome of a Kruskal Wallis test comparing data from pilot full dataset.

2) Influence of filtration depth

Increasing the filtration layer depth from 40 to 100 cm did not significantly improve TSS removal ($p = 0.09$). Median TSS removal efficiency was 92% and 91% for VFSt and VFG^+, respectively, falling within the range of removal rates usually observed for first stage filters in French VFCW system (Morvannou *et al.*, 2015, Paing and Voisin, 2005). Molle *et al.* (2005) and Paing & Voisin (2005) reported that TSS removal mostly occurred at the surface of the first stage filter. Figure 4a shows the effect of applied load on treatment efficiency. It can be seen that TSS removal was linear even for high loads. The lowest removal rates (especially for the VFSt at loads of 100 $g.m^{-2}.d^{-1}$) were obtained within the first five months after the commissioning period when the sludge deposit layer was still very thin. TSS removal efficiencies were thereafter higher than 90%. This observation confirmed the positive effect of the sludge deposit layer thickness on filtration performance (Molle *et al.*, 2005).

The effect of filtration depth on COD_{tot} removal was quite similar as for TSS (Figure 4b). This was mainly explained by the fact that most of COD_{tot} was under particulate form ($COD_d/COD_{tot} = 0.3$ in this study).

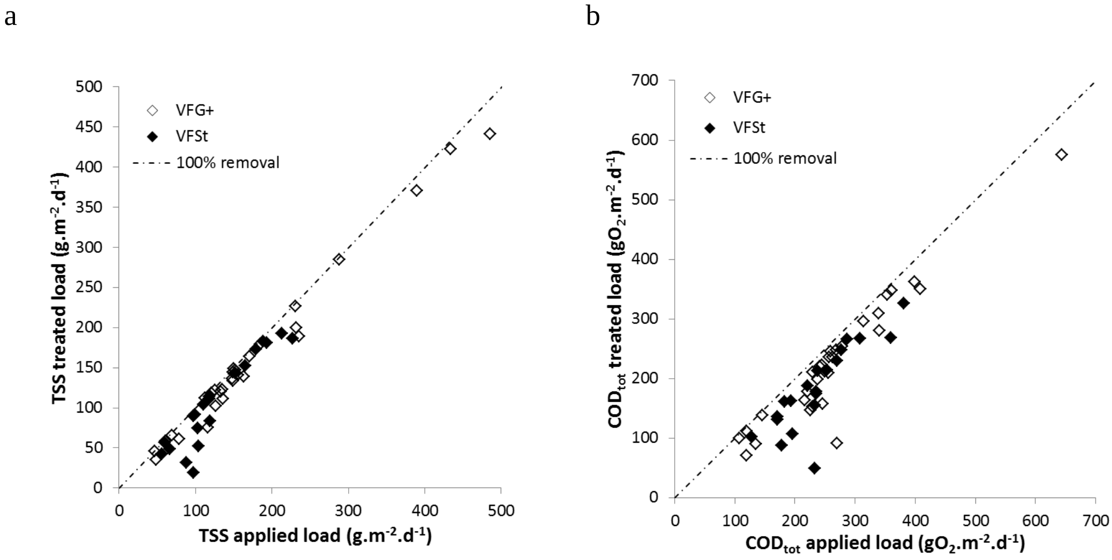

Figure 4 Treated TSS (a) and COD_{tot} (b) loads according to the applied TSS and COD_{tot} loads, respectively

Figure 5a showed that, within the range of COD_d loads applied in this study, the reference pilot VFSt performed similarly to the deep filter pilot VFG^+. COD_d removal was not statistically improved by increasing the filtration depth ($p = 0.06$) although a slightly better removal was observed for VFG^+ (59% and 66% for VFSt and VFG^+, respectively). Even though the implementation of a deeper filtration layer did not result in a statistically significant improvement of COD_d removal, it allowed a slight improvement of the outlet concentration (92.5 and 73.1 $mg.COD_d^{-1}$ on average for VFSt and VFG^+, respectively).

Around 60% of COD_d was degraded within the upper 20 cm of the filter as shown by the depth profile presented in Figure 5b. The removal rate then strongly decreased up to 40 cm-depth to become almost negligible with further depth. Morvannou *et al.* (2014)

reported that the heterotrophic community was mainly located in the sludge deposit and the upper part of the filtration layer in French first stage VFCW. Their similar performance in COD_d removal was consistent with the distribution of heterotrophic bacteria of Morvannou et al. (2014). Olsson (2011) carried out a similar experiment with VFCWs filled with different media (gravel or sand) and fed with pre-treated wastewater. The depth profile of total organic carbon (TOC) in sand revealed 68% removal at 20 cm-depth, which was very close to the COD_d profile observed in the present study. For gravel however, the profile was quite linear until 80 cm deep, suggesting that heterotrophic community can colonize deeper zones of filtration. This different depth profile with gravel may be explained by the fact that the gravels used in Olsson's work were coarser than in this study (4/8 mm and 2/6 mm, respectively) and the influent was pre-treated in a settling tank. In our study, the infiltration rate was thus probably lower. The similar depth profile between our study and the sand VFCW (1/3 mm) of Olsson shows the positive impact of sludge deposit on the hydraulics of the French systems (Molle *et al.*, 2006).

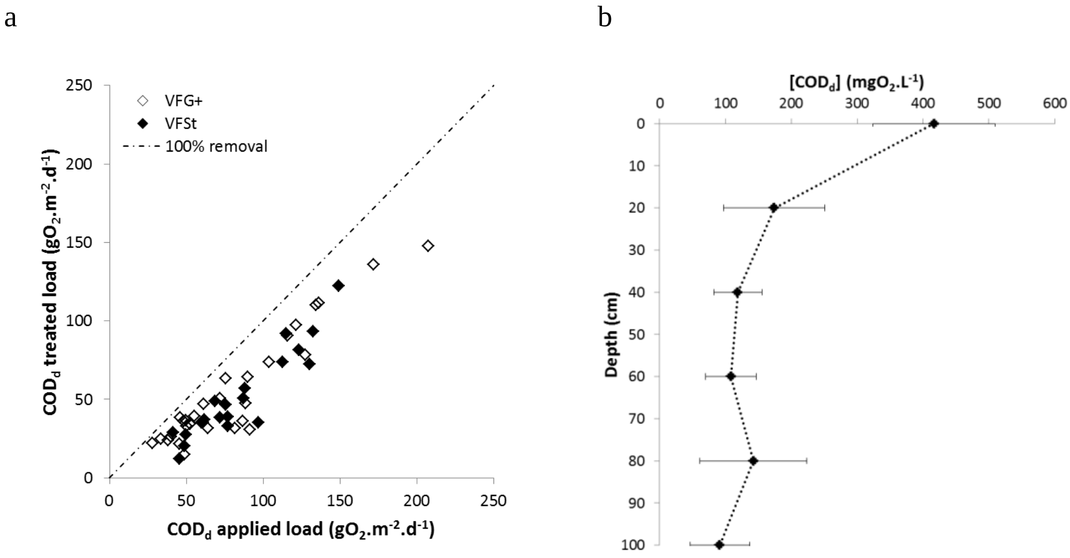

Figure 5 Treated COD$_d$ loads according to COD$_d$ the applied loads (a) and COD$_d$ depth profile during feeding cycle for the VFG$^+$ pilot (Six 24-hour composite samples) (b)

As illustrated in Figure 6a, ammonium removal efficiency was significantly improved by increasing the filtration depth ($p = 0.01$). It increased from 62% in 40cm-deep reference pilot VFSt to 81% in 100cm-deep VFG$^+$. Ammonium removal was linear within the applied load between 5 and 25 gNH$_4$-N.m^{-2}.d^{-1}.

We also observed a significantly different consumption of alkalinity ($p = 0.04$) and production of nitrate ($p = 0.001$). While VFSt had a mean nitrate production of 10.1 gN.m^{-2}.d^{-1} and removed 57.1 g.m^{-2}.d^{-1} of calcium carbonate on average, increasing the filtration depth from 40 cm to 100 cm improved the phenomena by almost 50% (14.1 gN.m^{-2}.d^{-1} and 75.9 g.m^{-2}.d^{-1} of nitrate production and CaCO$_3$ removal, respectively). These observations, along with the results on ammonium removal discussed above, revealed that a deeper filtration layer enhanced the nitrification rate.

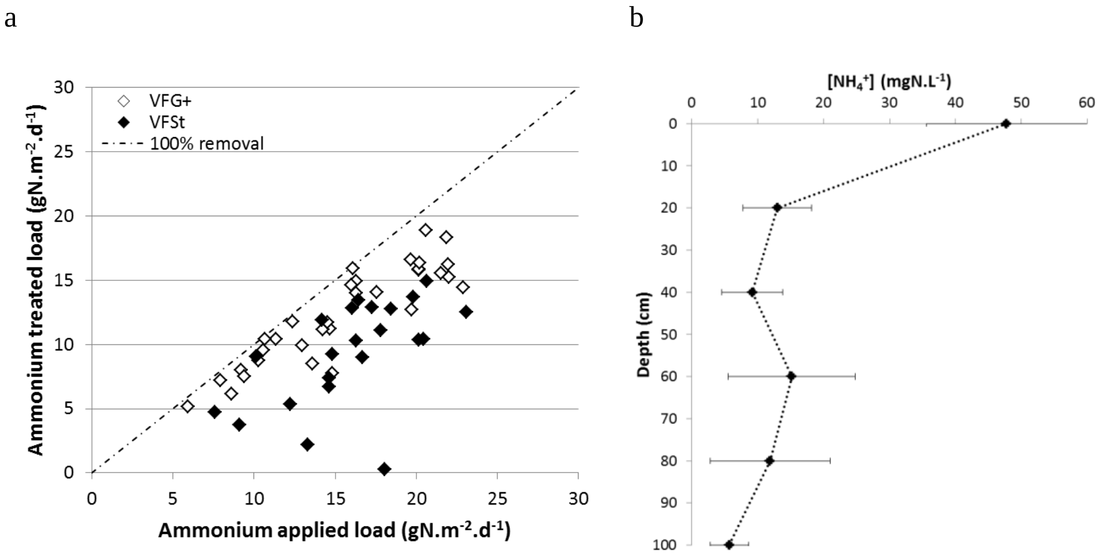

Figure 6 Ammonium treated loads according to the ammonium applied loads (a) and ammonium depth profile during feeding cycle for the VFG+ pilot (Six 24-hour composite samples) (b)

The depth profile of ammonium concentration (Figure 6b), carried out on 6 24-hour composite samples in the last stages of operation, showed that the upper 40 cm achieved about 75% of removed ammonium, while the overall performance was 87% at 100 cm deep. These results are in accordance with previously published works. Thus, Torrens *et al.* (2009) observed a higher TKN removal when increasing the filtration depth of a sand VFCW (from 69% to 78% at 25 cm and 65 cm, respectively) and Molle *et al.* (2008) reported a negligible improvement of TKN removal when increasing the filtration depth of the first stage of a French system from 60 cm to 80 cm. These observations may be attributed to the fact that autotrophic bacteria are mainly located in the sludge deposit and the upper 30 cm of the filtration layer as reported by Morvannou *et al.* (2014).

3) Effects of hydraulic and organic loads

Figure 7a presents the removal efficiency observed for TSS, COD_{tot}, COD_d and NH_4 in the reference (VFSt) and high load (VFHL) pilots. VFSt and VFHL showed similar TSS removal (92% and 84% respectively) indicating that hydraulic and organic loads had no significant influence on this parameter within the studied range ($p = 0.12$). Analytical data were exploited in terms of concentrations since the pilots did not receive identical loads.

COD_d removal was significantly impacted ($p=0.04$). It was reduced from 59% in VFSt to 44% in VFHL as shown in Figure 7a. This observation may be explained by the hydraulic changes induced by the increase of the loads. More frequent feedings resulted in an increase of ponding time, a decrease of water retention time (Molle *et al.*, 2006) and thus hindered oxygen renewal within the filter. This in turn was detrimental for aerobic microbial activity. The impact of hydraulic conditions is well described in Figure 7b which shows the COD_d removal in relation with loads. The removed load was lower with VFHL than with VFSt for similar applied organic loads.

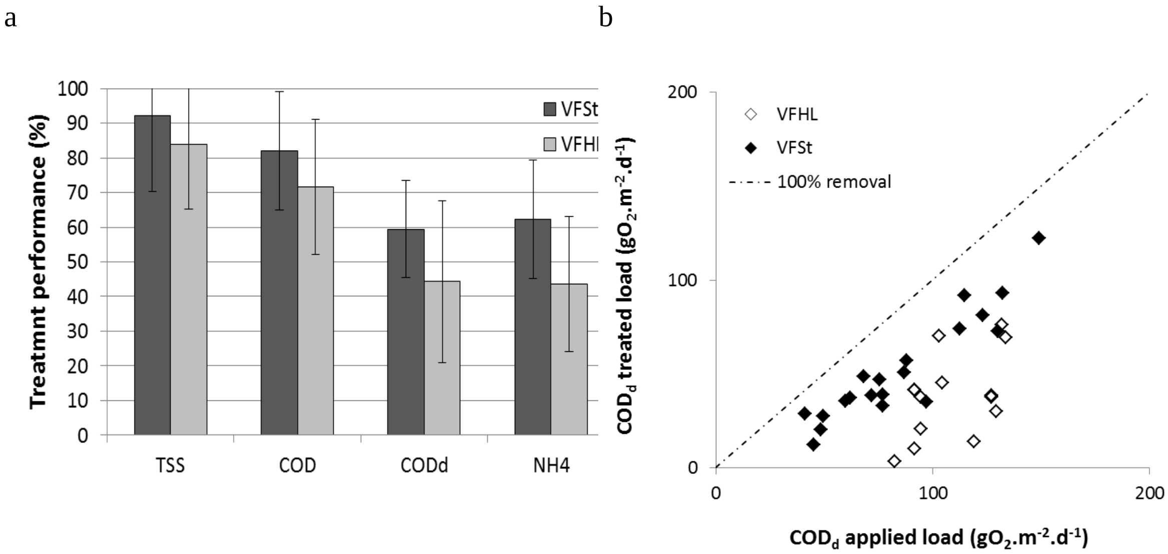

Figure 7 Treatment performance for global pollutants (a) and COD$_d$ treated loads according to the COD$_d$ applied loads (b) (Note that a selection in VFHL data was carried out in order to study treatment efficiency for similar organic loads but different hydraulic loads)

A significant reduction of ammonium removal ($p = 0.007$) occurred when increasing the hydraulic load from 0.36 cm.d^{-1} to 0.64 cm.d^{-1}. Performance dropped from 62% to 44% for VFSt and VFHL, respectively (Figure 7a). Ammonium removal related to applied loads in VFSt and VFHL is shown in Figure 8. For similar applied ammonium loads (between 5 and 25 gN.m^{-2}d^{-1}), VFHL exhibited lower removal capacity than VFSt. This might be explained by the lower oxygen transfer capacity of the system, lower water retention time as well as a higher saturation of ammonia adsorption sites due to less time for nitrification between batches.

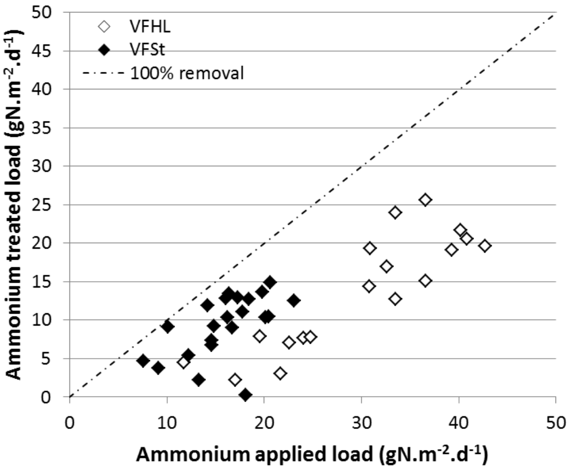

Figure 8 Ammonium treated loads according to the ammonium applied loads

4) Effect of the implementation of a sorbent material

The implementation of zeolite at the bottom of the filtration layer did not result in a significant improvement of ammonium removal ($p = 0.29$) regardless of the applied load as shown in Figure 9a. VFSt and VFZ achieved 62% and 68% of ammonium removal, respectively. This observation was consistent with Stefanakis and Tsihrintzis (2009) who reported that no significant improvement occurred in TKN removal by using zeolite in VFCW.

Knowing the cationic exchange capacities of zeolite (Erdoğan and Ülkü, 2011; Malekian *et al.*, 2011; Huang *et al.*, 2010; Ivanova *et al.*, 2010), we can observe that adsorption process was not efficient with the design used for VFZ. Since regeneration of sorption sites was expected to occur through nitrification of ammonium during the resting period, the progressive fouling of the media may not fully explain this lack of efficiency. The alkalinity concentrations and the pH values measured in VFZ effluent were favourable for nitrification (284 mg/L and 7.5, respectively). However, Figure 9b shows that almost no ammonium removal occurred in the zeolite layer. Different possible explanations can be drawn as preferential flows, short water retention times as well as the low ammonia concentration at this stage (< 10 mgN.L^{-1}). Nevertheless, Lahav and Green (1998) reported outlet ammonium concentrations lower than 1 mgN.L^{-1} with upflow mode columns fed at 40 mgN.L^{-1}. Such ammonium removal was possible by the implementation of large amount of chabazite (almost five times the amount in this study) with short contact time (2 minutes). It should be thus possible to improve the ammonium removal by increasing the zeolite fraction of the filtration layer.

It is also known from kinetic studies, carried out under static conditions for different contact times, that sorption increases with contact time until an equilibrium is reached (Huang *et al.*, 2010; Wen *et al.*, 2006). Therefore, limiting the outflow rate may be one possible option to improve the effect of zeolite by increasing contact time without adding more zeolite.

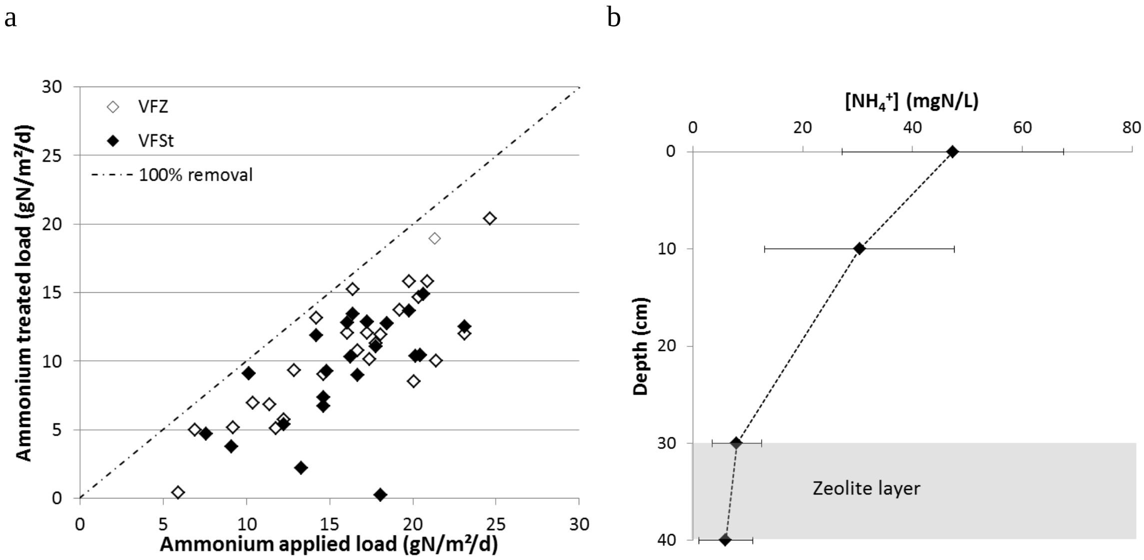

Figure 9 Treated ammonium loads versus applied ammonium loads (a) and depth profile of ammonium concentration during a feeding cycle (3 24-hour composite samples) (b) Note that the systems were drained vertical flow filters which were therefore operated under unsaturated conditions.

IV) Conclusion

This study aimed at identifying the leverage actions in order to reduce the treatment footprint of VFCW. The respective impact of design criteria and operation conditions on the ability of a 1st stage of VFCW to perform treatment of different pollutants (TSS, COD_d, ammonium), from raw domestic wastewater, were assessed for this purpose.

TSS removal was not affected by the studied modifications of design or operational parameters since it was mainly a surface mechanism. Therefore, reduction of the 1st stage surface would not result in a drop in particles treatment efficiency. Nevertheless, the decrease in surface of treatment (from 0.4 m²/p.e./bed to 0.25 m²/p.e./bed, respectively) would cause an increase in daily hydraulic load (from 0.36 $m.d^{-1}$ to 0.64 $m.d^{-1}$) which showed significant adverse effects on COD_d and ammonium removal (from 59% to 44% and from 62% to 44%, respectively) because of the shorter contact time as well as lower oxygen renewal within the filter.

The lower removal of COD_d and ammonium observed when decreasing the surface of the 1st stage may be partly counterbalanced by the implementation of deeper filtration layer. Ammonium removal was actually raised from 62% to 81% and COD_d removal was improved from 59% to 66% when filtration depth increased from 40 cm to 100 cm. Nevertheless, the relation between gain of performance and depth of filtration was low, especially when the filtration layer was deeper than 60 cm since the microbial community was mainly located in the upper part of the filtration layer. In addition, a deeper filtration layer enabled to maintain a more constant efficiency which might be valuable when fluctuation of performance is observed (i.e. when temperature variations, over year, are wide).

Furthermore, despite its theoretical ion exchange capacity, zeolite implementation in the filtration layer did not allow to reach the expected improvement of ammonium removal for the assessed characteristics of design and operation. Higher zeolite content might provide different conclusions but would result in prohibitive extra-costs (zeolite was almost 5 times more expensive than gravel). The implementation of such reactive material, as suitable alternative to intensification, should not be further considered unless the operation conditions allowed the optimal use of exchange capacity. Further studies are thus necessary to determine the best design and operational conditions for its efficient use.

In conclusion, it seems difficult to reach low discharge levels with a single stage of VFCW treating domestic wastewater. However, surface requirements may be reduced to 0.25 m²/p.e./bed if a second stage ensures the final treatment of remaining pollution and if filtration depth is also used as an adjustment parameter.

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