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Evaluation of the effectiveness of horizontal subsurface flow constructed wetlands for different media

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Abstract

Two media bed (gravel and Filtralite NR) were tested in a mesocosm to evaluate the removal of organic matter (as chemical oxygen demand (COD)), ammonia (NH_4-N), nitrite, nitrate and solid matter (as total suspended solids (TSS)) for a synthetic wastewater (acetate-based) and a domestic wastewater. The use of Filtralite allowed average removal rates (6–16.8 g COD/(m^2·day), 0.8–1.1 g NH_4-N/(m^2·day) and 3.1 g TSS/(m^2·day)) and removal efficiencies (65%–93%, 57%–85% and 78% for COD, NH_4-N and TSS, respectively), higher than that observed in the experiments with gravel. The applied loads of COD, ammonia, nitrate and TSS seem to influence the respective removal rates but only for the treatment of domestic wastewater with higher correlation coefficients for Filtralite. Regardless the type of media bed and the type of wastewater, nitrate was completely removed for nitrogen loading rates up to 1.3 g NO_3-N/(m^2·day). There was no evidence of the influence of nitrate loads on the removal of organic matter.

Key words: media bed; constructed wetlands; pollutants removal rates

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Introduction

The media bed of horizontal subsurface flow (HSSF) constructed wetlands is normally based on gravel and frequently present clogging problems which are related to both its properties (e.g., porosity and specific surface area) and the characteristics of the wastewater.

Light-expanded clay aggregates (LECA) has been presented as alternative media bed to reduce the clogging problem or to increase the treatment capacity since presents both higher porosity and specific surface area, which allow a good biofilm adhesion and a high hydraulic conductivity. Several studies have reported a good removal of ammonia and nitrate (Vilpas et al., 2005; van Deun and van Dyck, 2008; Albuquerque et al., 2009a) in LECA-based HSSF beds.

The HSSF beds may be seen as anoxic/anaerobic biofilm reactors as most of the biological removal pathways occur in the presence of low concentration of oxygen (< 2 mg/L). The atmospheric oxygen diffusion through the upper soil layer into the bed is low (Vymazal and Kropfelova, 2008) and the oxygen release through the plants into the rhizosphere is also low (Kadlec and Wallace, 2008) and is automatically consumed by the biofilm installed in the media bed, roots and rhizomes (Randerson, 2006) due to the presence of high oxygen demand wastewaters.

Organic matter is removed by aerobic, anoxic and anaerobic pathways, filtration and precipitation. Solid matter is removed through filtration and sedimentation into the media bed. Nitrogen (present as organic nitrogen, ammonia and nitrate) is removed through biological removal pathways (e.g., nitrification and denitrification), plant uptake, volatilization, filtration, sedimentation, adsorption and microbial assimilation (Kadlec et al., 2000; Kadlec and Wallace, 2008). Nitrification needs dissolved oxygen (DO) concentrations above 2 mg/L (Metcalf and Eddy, 2003), which cannot be guaranteed in HSSF beds. However, some authors have reported the occurrence of nitrification for DO concentrations below 2 mg/L (Paredes et al., 2007; Dong and Sun, 2007; Albuquerque et al., 2009a). When pH is below 6.5 the free ammonia concentration becomes too low for sufficient growth of the ammonia oxidizers whilst a high pH (> 7.5) benefits the ammonia oxidizers and ammonia volatilization (> 8.5) (Kadlec and Wallace, 2008; Vymazal and Kropfelova, 2008). Denitrification occurs in anaerobic/anoxic environments with availability of organic compounds.

The HSSF beds for domestic wastewater treatment normally use organic loading rates (OLR) from 5 to 25 g COD/(m^2·day), N loading rates (NLR) from 1 to 5 g

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N/(m^2·day), solids loading rates (SLR) from 1 to 20 g TSS/(m^2·day), hydraulic loading rates (HLR) from 2 to 20 cm/day and hydraulic retention times (HRT) from 3 to 10 days (Kadlec et al., 2000; Kadlec and Wallace, 2008; Vymazal and Kropfelova, 2008; Albuquerque et al. 2009b). Nitrogen removal in most HSSF beds is normally low (40%–50%) as compared to organics (> 90% as COD or BOD$_5$) and solids (> 60% as TSS).

The objective of this work was to evaluate the effectiveness of a HSSF mesocosm on the removal of organic matter, nitrogen and solid matter using two media beds (gravel and Filtralite) from different sources of nitrogen and a synthetic wastewater (acetate-based) and a domestic wastewater (after primary treatment).

1 Materials and methods

1.1 HSSF mesocosm

A HSSF mesocosm with 2 m × 0.8 m (length × width) and 0.2 m of media height was used (Fig. 1). The experiments were carried out with gravel (40–70 mm, specific surface area of 700 m^2/m^3 and bed porosity of 0.4) and a LECA with the commercial name of Filtralite NR (4–8 mm, specific surface area of 1250 m^2/m^3 and bed porosity of 0.45). The bed was colonised with common reed (*Phragmites australis*). Nine sampling points (PI1 to PI9) were provided along the bed to collect water samples for analytical measurements. Three piezometers (PIEZ1, PIEZ2 and PIEZ3) were included to evaluate the evolution of head losses in three sections of the bed.

1.2 Experimental procedure

Three sets of experiments were executed for different wastewaters and change of the media bed. Series with gravel (G) and Filtralite (F) took 22 and 20 weeks, respectively. Loads for Series G1, G2, F1 and F2 (only organic carbon source and ammonia or nitrate sources): 10–11.3 g COD/(m^2·day) (approximately 300 mg COD/L), 1–1.1 g N/(m^2·day) (approximately 30 mg N/L as NH_4-N or NO_3-N). Loads for Series G3 and F3 (domestic wastewater): 17–20 g COD/(m^2·day) (300–500 mg COD/L), 0.8–2 g NH_4-N/(m^2·day) (20–50 mg NH_4-N/L), 0.1–0.2 g NO_3-N/(m^2·day) (3–5 mg NO_3-N/L), 3.5–4.5 g TSS/(m^2·day) (100–120 mg TSS/L). The two first Series were performed with synthetic wastewater (based on sodium acetate, ammonia chloride and potassium nitrate solutions) whilst primary treated domestic wastewater was used in the last Series. The flow rate was kept 1 L/hr (i.e., HLR of 4 cm/day for gravel and 3.5 cm/day for Filtralite; HRT of 5.1 days for gravel and 5.7 days for Filtralite over the effective bed area).

In each series the bed was operated during 1 week to attain steady state conditions followed by weekly water sampling (6 and 5 experiments in Series G and Series F, respectively) at the influent and points PI2, PI5 and PI8 to

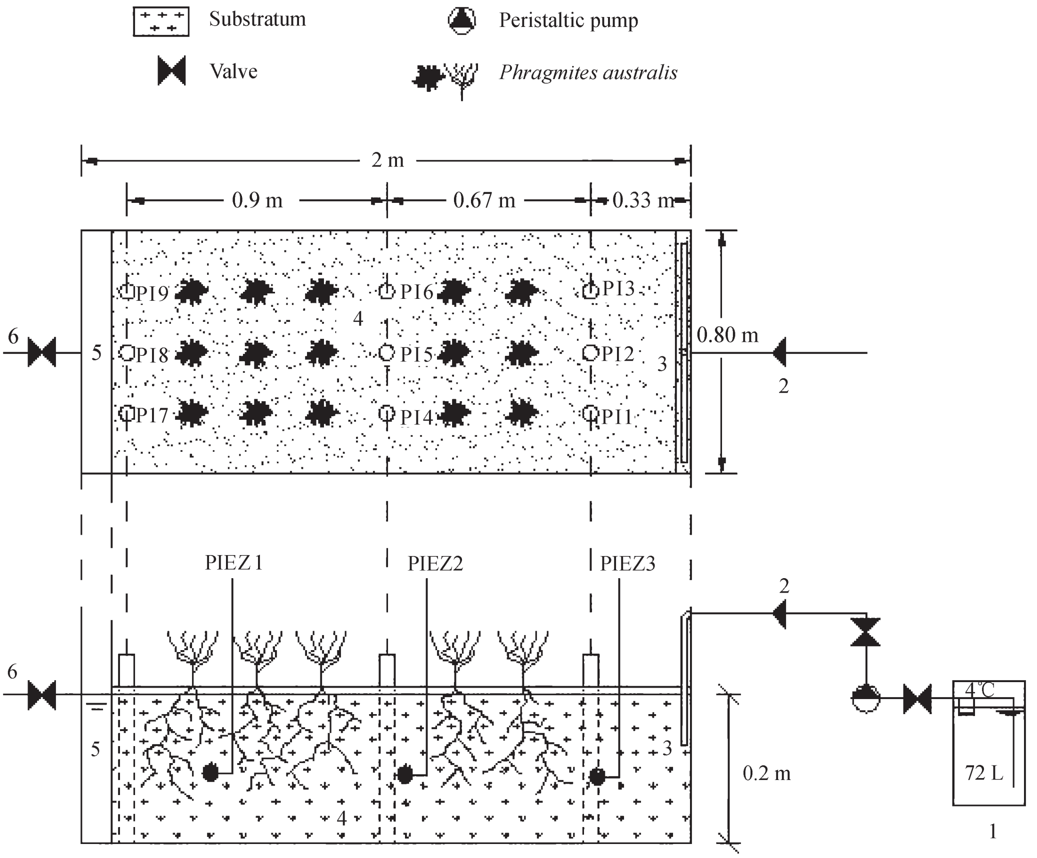

Fig. 1 Horizontal subsurface flow (HSSF) mesocosm. (1) reservoir; (2) inlet/influent; (3) feeding system; (4) HSSF bed; (5) outlet; (6) effluent; PIEZ1, PIEZ2, PIEZ3: piezometers; PI1 to PI9: sampling points.

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evaluate the pH, DO, temperature, COD, NH_4-N, NO_2-N and NO_3-N. The hydrostatic pressure in the piezometers PIEZ1, PIEZ2 and PIEZ3 was measured weekly. The temperature in the laboratory was kept constant at approximately 20°C.

1.3 Synthetic wastewater

Synthetic wastewater included an organic carbon source (sodium acetate solution), a nitrogen source (ammonia chloride or potassium nitrate solution) and a mineral source as proposed by Dang et al. (1989). All the solutions were prepared as concentrated ones according to the following composition: buffer solution (8.50 g KH_2PO_4 + 21.75 g K_2HPO_4 + 33.40 g $Na_2HPO_4 \cdot 7H_2O$ + 1.70 g NH_4Cl/L), magnesium sulphate solution (22.50 g $MgSO_4 \cdot 7H_2O$/L), calcium chloride solution (36.43 g $CaCl_2 \cdot 2H_2O$/L), iron chloride solution (0.25 g $FeCl_3 \cdot 6H_2O$/L), oligoelements solution (0.04 g $MnSO_4 \cdot 4H_2O$ + 0.06 g H_3BO_3 + 0.04 g $ZnSO_2 \cdot 7H_2O$ + 0.032 g $(NH_4)_6 \cdot Mo_7O_{24} \cdot 4H_2O$ + 0.0555 g EDTA ($C_{10}H_{14}N_2Na_2O_8 \cdot 3H_2O$) + 0.0445 g $FeCl_3 \cdot 6H$/L), sodium acetate solution (113.4 g $C_2H_3O_2Na \cdot 3H_2O$/L, which gives 50 g COD/L), ammonia chloride solution (76.41 g NH_4Cl/L, which gives 20 g N/L) and potassium nitrate solution (144.4 g KNO_3/L, which gives 20 g N/L).

The mineral medium was prepared by diluting the concentrated solutions in the following proportions: 2 mL/L (buffer solution), 0.2 mL/L (magnesium sulphate solution), 0.2 mL/L (calcium chloride solution), 0.2 mL/L (iron chloride solution), 0.2 mL/L (oligoelements solution), 6 mL/L (sodium acetate solution) and 1.5 mL/L (ammonia chloride and potassium nitrate solutions). The feeding solution was kept in a storage tank (ISCO FTD 220, Italy) at a constant temperature of 4°C and pumped to the bed through a peristaltic pump MCP CA4 (ISMATEC, Switzerland).

1.4 Analytical methods

The pH, DO and temperature were measured directly through the probes SenTix 41 and CellOx 325 connected to a Multi 340i meter (WTW, Germany). The COD was evaluated by closed reflux digestion and titrimetric method (APHA, 1998). Concentrations of NH_4-N, NO_2-N and NO_3-N were obtained using the cuvette-tests LCK 302 (47–130 mg NH_4-N/L), LCK 303 (2–47 mg NH_4-N/L), LCK 304 (0.015–2 mg NH_4-N/L), LCK 341 (0.015–0.6 mg NO_2-N/L), LCK 342 (0.6–6 mg NO_2-N/L), LCK 339 (0.23–13.5 mg NO_3-N/L) and LCK 340 (5–35 mg NO_3-N/L), following standards DIN 38406-E 5-1 (ammonia), DIN 38405 D10 (nitrite) and DIN 38405-9 (nitrate), and the CADAS 50 spectrophotometer UV-Vis (HACH-LANGE, Germany). TSS and VSS were determined according to the method by APHA (1998).

2 Results and discussion

2.1 Operating conditions and performance

Results for the 3 series are presented in Table 1. There was no detection of NO_2-N and NO_3-N in the 3 sampling points (all the values were below the detection limit as shown for nitrate).

The hydrostatic pressure measured in the 3 piezometers slight increased in the series with domestic wastewater (G3 and F3) due to high influent TSS. Although influent TSS concentrations were higher in the experiments with Filtralite (average of 133 mg/L against 112 mg/L observed in the experiments with gravel), the head losses were lower and the average removal efficiency (RE) of TSS was high

Table 1 Results for each series

Parameter	Point	G1	G2	G3	F1	F2	F3
pH	Influent	7.0–7.4	7.0–7.2	7.0–7.2	7.1–7.3	7.6–8.0	6.9–7.3
	PI2	7.5–8.0	8.6–9.0	7.1–7.3	7.3–7.9	8.3–8.9	7.2–7.8
	PI5	7.3–7.7	7.7–8.5	7.0–7.1	7.7–8.1	8.2–8.5	7.2–8.0
	PI8	7.3–7.9	7.5–8.2	6.9–7.0	7.5–8.0	8.2–8.2	7.1–7.7
DO (mg/L)	Influent	2.1 ± 1.0	2.0 ± 1.0	0.3 ± 0.1	2.4 ± 1.3	2.2 ± 1.1	1.8 ± 1.0
	PI2	0.1 ± 0	0.3 ± 0	0.1 ± 0	0.2 ± 0	0.1 ± 0	0.2 ± 0
	PI5	0.1 ± 0	0.2 ± 0	0.1 ± 0	0.1 ± 0	0.1 ± 0	0.2 ± 0
	PI8	0.1 ± 0	0.2 ± 0	0.1 ± 0	0.1 ± 0	0.1 ± 0	0.1 ± 0
COD (mg/L)	Influent	283 ± 14	285 ± 25	518 ± 59	316 ± 34	263 ± 76	507 ± 133
	PI2	151 ± 46	142 ± 24	227 ± 20	203 ± 22	118 ± 20	94 ± 14
	PI5	122 ± 46	158 ± 51	202 ± 11	197 ± 13	112 ± 17	57 ± 8
	PI8	91 ± 43	126 ± 33	154 ± 20	94 ± 32	97 ± 5	32 ± 16
NH_4-N (mg/L)	Influent	27.7 ± 1.1	–	53.6 ± 4.5	38.7 ± 3.2	–	35.5 ± 3.7
	PI2	19.6 ± 1.1	–	39 ± 3.5	23.7 ± 3.9	–	23.3 ± 5
	PI5	18.0 ± 1.1	–	39 ± 2.8	21.6 ± 3.4	–	14.5 ± 4.7
	PI8	14.5 ± 2.2	–	31.1 ± 1.8	15.3 ± 3.7	–	5 ± 2.1
NO_3-N (mg/L)	Influent	–	32.7 ± 1.4	0.5 ± 0.1	–	21.3 ± 5.3	0.3 ± 0.1
	PI2	–	< 0.231*	0.3 ± 0.1	–	< 0.231*	< 0.23*
	PI5	–	< 0.231*	< 0.23*	–	< 0.231*	< 0.23*
	PI8	–	< 0.231*	< 0.23*	–	< 0.231*	< 0.23*
TSS (mg/L)	Influent	–	–	113 ± 26	–	–	115 ± 58
	PI2	42 ± 25	45 ± 38	42 ± 18	22 ± 7	16 ± 7	34 ± 15
	PI5	15 ± 14	52 ± 33	40 ± 7	40 ± 7	25 ± 6	25 ± 10
	PI8	16 ± 13	22 ± 7	49 ± 18	23 ± 7	19 ± 6	29 ± 14

* Lower than the limit of the cuvette-test LCK 339.

Average values and confidence interval (calculated for a confidence level of 95% and 6 values for Series G and 5 values for Series F).

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for the type of media bed (70% against 51% observed in the experiments with gravel). However, the observed head losses for both media bed were low and only noted in the PIZ1 piezometer as shown in Fig. 2 and it can be assumed that the bed clogging was insignificant and had no influence on the bed effectiveness.

The bed was DO limited in all the experiments (< 0.3 mg/L), which was expected since both wastewaters had high levels of oxygen demand and the DO was rapidly consumed through aerobic respiration and chemical oxidation. The pH in the series with ammonia was below 8, despite the RE of NH_4-N has been significantly different: 45% (G1), 43% (G3), 57% (F1) and 85% (F3). This circumstance resulted from the buffer effect created by the simultaneous removal of acetate which, by generating alkalinity, compensated the consumption of alkalinity through nitrification. In series with nitrate (G2 and F3) pH increased significantly (up to 9) due to the production of alkalinity associated to acetate oxidation and denitrification reactions.

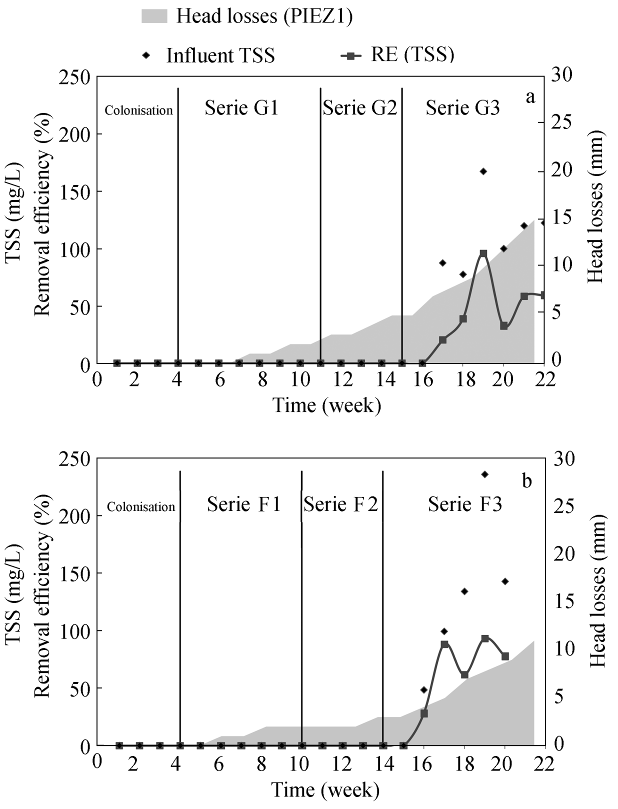

Fig. 2 Evolution of TSS and head losses gravel (a) and filtralite (b). RE: removal efficiency.

2.2 Mass removal rates

The mass removal rates (g/(m^2·day)) were calculated for COD (r_{CQO}), NH_4-N ($r_{NH_4\text{-}N}$), NO_3-N ($r_{NO_3\text{-}N}$) and TSS (r_{TSS}) based on influent and effluent (sampling point PI8) concentrations, flow rate and effective areas of each bed as shown in Table 2.

Regardless the type of bed material, COD removal was similar in the experiments with synthetic wastewater. However, the corresponding RE was lower in the experiments with gravel. When the bed was fed with domestic wastewater r_{COD} increased 24% from the experiments with gravel to the ones with Filtralite. The introduction of nitrate in Series G2 and F2 had no effect on COD removal. Although nitrate was completely removed in the first section of the bed (Influent-PI2) COD removal rates and the respective RE were lower than the ones observed in the experiments with ammonia.

Higher COD removal was expected in Series G2 and F2 because it is presumed that, since the bed was in anoxic conditions, the removal of nitrate should have occurred mainly by denitrification. These results suggest that denitrifiers may have used not only organic carbon from wastewater. Kuai and Verstraete (1999) and Ahn (2006) pointed out that alternative nitrate removal pathways such as autotrophic denitrification uses hydrogen, sulfur compounds, ammonia, nitrite and nitrate as energy source and inorganic carbon sources. Some chemolithoautotrophi bacteria as *Nitrosomonas* are able to nitrify and denitrified in low concentrations of oxygen.

Regardless of the type of wastewater the removal of ammonia was higher in the presence of Filtralite, especially when the bed was fed with domestic wastewater. The RE increased approximately 50% when the media bed was changed from gravel to Filtralite. In the experiments with gravel, the removal of ammonia was lower and quite similar for both types of wastewaters. However, as DO in the bed was limited in all assays and knowing that the autotrophic nitrifiers and aerobic heterotrophic microorganisms compete for the same electron acceptor (oxygen), the latter ones with both higher specific growth rates and cell yield coefficients (Metcalf and Eddy, 2003), it seems therefore unlikely that there was sufficient oxygen flux

Table 2 Applied loads and mass removal rates

Media bed	Series	OLR	r_{CQO}	NLR	$r_{NH_4\text{-}N}$	NLR	$r_{NO_3\text{-}N}$	SLR	r_{TSS}
		(g/(m^2·day))		(g/(m^2·day))		(g/(m^2·day))		(g/(m^2·day))	
Gravel	G1	11.3 ± 0.6	7.7 ± 1.7	1.1 ± 0.1	0.5 ± 0.1	–	–	–	–
	G2	11.4 ± 1	6.4 ± 1	–	–	1.3 ± 0.2	1.3 ± 0.1	–	–
	G3	20.7 ± 2.4	14.5 ± 3	2.1 ± 0.2	0.9 ± 0.2	0.1 ± 0	0.1 ± 0	4.5 ± 0.2	2.6 ± 0.2
Filtralite	F1	11.2 ± 1.2	8.3 ± 2	1.4 ± 0.2	0.8 ± 0.2	–	–	–	–
	F2	9.3 ± 2	6 ± 1	–	–	0.8 ± 0.1	0.8 ± 0	–	–
	F3	18 ± 4.7	17 ± 4	1.3 ± 0.3	1.1 ± 0.3	0.1 0	0.1 ± 0	4.1 ± 0.3	3.1 ± 0.2

OLR: organic loading rate; NLR: nitrogen loading rate; SLR: solid loading rate.
Average values and confidence interval (calculated for a confidence level of 95% and 6 values for Series G and 5 values for Series F).

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to drive the apparent ammonia removal rates through the conventional nitrification pathway. Paredes et al. (2007) pointed out that HSSF beds present oxidation-reduction conditions to allow alternative microbiological ammonia pathways (e.g., autotrophic anaerobic oxidation or heterotrophic nitrification), which was already observed in some studies (Kuai and Verstraete, 1999; Dong and Sun, 2007). Regardless of both the type of bed material and the type of wastewater, the nitrate removal was complete for NLR below 1.3 g NO_3-N/(m^2·day) and occurred in the section Influent-PI2.

In most of the experiments the pH ranged from 6.5 to 8.5, which are values considered appropriate for organic carbon oxidation, nitrification, denitrification and other non-conventional removal pathways, such as anaerobic autotrophic ammonia oxidation and autotrophic denitrification (Ahn, 2006; Dong and Sun, 2007; Kadlec and Wallace, 2008).

The filtration capability of both media bed only was evaluated in the experiments with domestic wastewater (Series G3 and F3). The results show that Filtralite seems to have a higher capacity for TSS removal. When Filtralite was introduced in the bed the RE increased more than 26% comparing with the RE observed form gravel.

The removal of COD, ammonia, nitrate and TSS occurred mainly in the initial section of the bed (Influent-PI2) in the types of bed material and wastewater. This circumstance suggests that HSSF beds allow a better oxygenation near the feeding point, which helps the removal of organic matter and ammonia as well as provide a higher filtration capability for solid matter removal, especially in the presence of Filtralite. Section Influent-PI2 was not colonized with plants and the additional oxygenation of the medium through roots or the removal of nitrogen by plant uptake was neglected.

A regression analysis between applied and removed loads shows that there was no significant linear correlation in the experiments with synthetic wastewater. The only linear relationship was observed for OLR and r_{COD} in the presence of Filtralite (R^2 = 0.6 for Series F1 and R^2 = 0.64 for Series 2, in both cases with $p < 0.05$). A significant linear correlation was observed between applied and removed loads for COD, NH_4-N, NO_3-N and TSS for both bed materials in the treatment of domestic wastewater with statistical significance ($p < 0.05$). Figure 3 shows the results for COD and TSS. The higher correlation coefficients were observed for Filtralite (R^2 = 0.99 for COD, R^2 = 0.88 for NH_4-N and R^2 = 0.93 for TSS, in any case with $p < 0.05$).

Therefore, regardless of the type of bed material, in the presence of domestic wastewater COD, NH_4-N, NO_3-N and TSS loads influence the respective removal rates, for the range of applied loads in this study.

There was no linear correlation between r_{COD} and the $r_{NO_3\text{-}N}$. Therefore, there is no statistical evidence to say that the COD removal was influenced by nitrate removal or that the organic load had influence on nitrate removal, as it would be expected if nitrate was removed only via denitrification. Therefore, these findings reinforce the conviction that nitrate removal may also have occurred through non-conventional removal pathways.

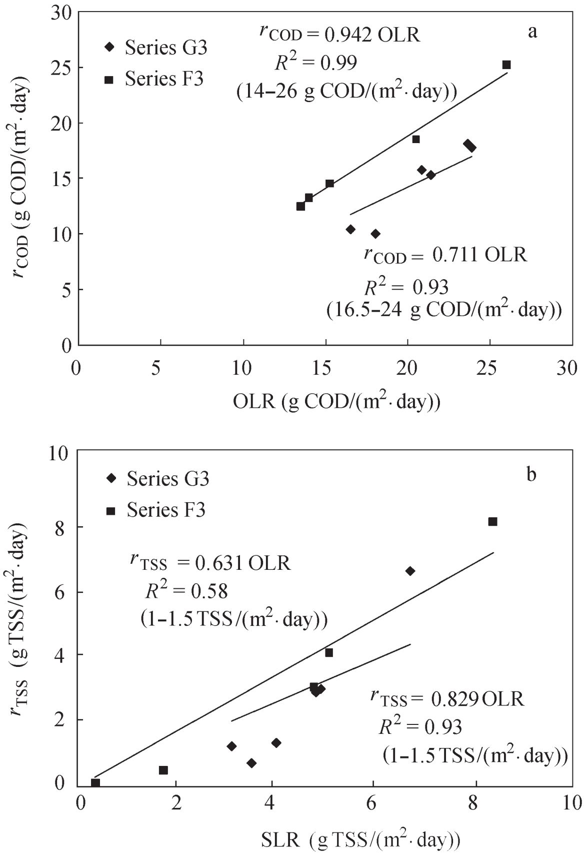

Fig. 3 Relationships between applied and removed loads of COD (a) and TSS (b).

3 Conclusions

HSSF beds Filtralite-based presented both higher removal rates and removal efficiencies of COD, ammonia and TSS than gravel did. The characteristics of Filtralite seem to enable a quick development of biofilm with the ability to promote the removal of organic matter, ammonia, nitrate and solid matter at high rates. The oxygen limitation observed in both beds does not favor the aerobic removal of ammonia and organic matter, and the removal rates observed for ammonia and nitrate may not be only associated to conventional removal mechanisms. In both the type of bed material and the type of wastewater, nitrate removal was complete for NLR below 1.3 g NO_3-N/(m^2·day) and occurred entirely in the first section of the bed. For the range of applied OLR, NLR and SLR the incoming loads of COD, NH_4-N, NO_3-N and TSS seem to have significantly influenced their removal rates but only for the treatment of domestic wastewater with higher correlation coefficients for Filtralite.

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