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Article

Comparison of Three Systems for Biological Greywater Treatment

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Abstract: Greywater consists of household wastewater excluding toilet discharges. Three systems were compared for the biological treatment of greywater at a similar hydraulic retention time of approximately 12–13 hours. These systems were aerobic treatment in a sequencing batch reactor, anaerobic treatment in an up-flow anaerobic blanket reactor and combined anaerobic-aerobic treatment (up-flow anaerobic blanket reactor + sequencing batch reactor). Aerobic conditions resulted in a COD removal of 90%, which was significantly higher than 51% removal by anaerobic treatment. The low removal in the anaerobic reactor may have been caused by high concentration of anionic surfactants in the influent (43.5 mg/L) and a poor removal of the colloidal fraction of the COD in up-flow anaerobic sludge blanket reactors. Combined aerobic-anaerobic treatment accomplished a COD removal of 89%, similar to the aerobic treatment alone. Greywater methanization was 32% for the anaerobic system and 25% for the anaerobic-aerobic system, yielding a small amount of energy. Therefore, anaerobic pre-treatment is not feasible and an aerobic system is preferred for the treatment of greywater.

Keywords: aerobic treatment; anaerobic treatment; greywater; surfactants; sludge yield

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1. Introduction

Greywater (*i.e.*, discharges from shower, hand basin, bath, laundry and kitchen) accounts for up to 75% of the wastewater produced in households [1]. It contains low concentrations of organic compounds (in terms of chemical oxygen demand (COD)), nutrients and pathogens compared to the more highly concentrated black (toilet)water [2]. Therefore, it makes sense to collect greywater, treat it separately and re-use it for irrigation, infiltration, washing or other non potable applications.

Currently, most greywater treatment systems installed are based on septic tanks in combination with constructed wetlands, sand filtration or compact aerobic systems such as membrane bioreactors, bio-rotors and biofilters [3].

Upflow anaerobic sludge blanket (UASB) reactors have been successfully applied in domestic wastewater treatment at tropical and semi-tropical conditions [4]. An important feature of this technology is the advantage of operating at a short hydraulic retention time (HRT) combined with longer sludge retention time (SRT) [5], and the possibility to combine the removal of COD with energy production as methane. Only few studies on anaerobic treatment of greywater are available [6,7]. Elmitwalli *et al.* [6] recommended anaerobic treatment as the first treatment step for greywater. They noted a COD removal of around 50% at 30 °C and an HRT of 6 hours was achieved applying a UASB reactor; in batch tests the maximum anaerobic degradability of greywater was 76% [6].

The process configuration where aerobic and anaerobic conditions are combined for greywater treatment has not been previously investigated. This configuration, however, has the possibility of combining the advantages of anaerobic and aerobic processes, and leading to optimal biological treatment of greywater.

Therefore, this study aims at comparing the biological treatment of greywater at lab-scale under anaerobic and aerobic conditions, and under the combination of these at similar overall HRT.

2. Experimental Section

2.1. Greywater source

Greywater was collected from the 32 houses of the Decentralized Sanitation and Reuse (DeSaR) demonstration project in Sneek, the Netherlands [8]. These 32 houses are equipped with vacuum toilets for the collection of concentrated black water, which is treated on-site. Greywater (the rest of the household wastewater) was collected separately. At the time of this study there was no treatment system on-site yet. Therefore, an autosampler (ASP-Station 2000, Endress + Hauser) was installed to take time proportional samples and store them at 4 °C. The capacity of the autosampler was 80 L. The total amount of water treated was about 5 m^3, therefore we treated 0.5% of the greywater from this site. These samples were transfered every 1–4 days from Sneek to our lab facilities in Leeuwarden and stored in a cold room (2–10 °C) before the water was treated in lab-scale reactors.

2.2. Experimental setup

Three systems were operated at a total HRT of 12-13 hours, based on the design HRT for a UASB reactor [5].

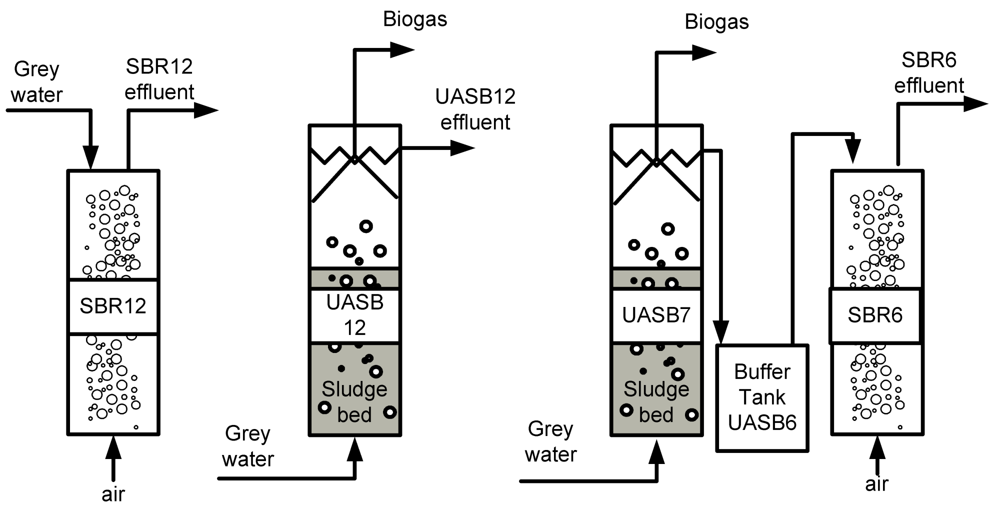
The aerobic system consisted of an SBR of 3.6 L. The anaerobic system consisted of a UASB reactor of 5 L. The combined anerobic-aerobic system consisted of a sequence of an SBR of 3.6 L operated at an HRT of 6 hours and a UASB of 5 L operated at an HRT of 7 hours. Figure 1 shows the scheme of the setup used in this study. Reactors are labeled after the reactor configuration (SBR or UASB), followed by a number representing the HRT in hours. Reactors SBR12 and UASB12 were operated for 168 days and reactors UASB7 and SBR6 were operated 84 days (from day 84–168).Table 1 shows the description of the operating cycles of the SBRs.

The temperature of the reactors was 32 ± 3 °C and it was maintained by means of a water bath connected to the double wall of the reactors.

Table 1. Operating cycles for aerobic SBRs.

	SBR12	SBR6
Cycle duration (h)	6	4
Filling (min)	15	10
Reaction time (h)	4.8	3.2
Settling time (min)	45	30
Emptying time (min)	15	10

Figure 1. Lab-scale reactors set up.



2.3. *Chemical analysis*

Influent and effluent were monitored every 2–3 days over a period of 168 days for total nitrogen, nitrate, total ammonium nitrogen (NH_3-N and NH_4^+), total phosphorus, total and dissolved COD and total and volatile suspended solids (VSS). These analyses were all conducted according to Standard Methods [9], suspended COD was defined by a particle size of >12–25 μm (Whatman Grade 589, black ribbon filter) and dissolved COD was defined below a particle size of 0.45 μm. Colloidal COD was calculated from the difference of the COD from the black ribbon filtered samples and the dissolved COD. Effluent from the reactors was collected over periods of 2–3 days, stored at 4–10 °C and then weighed and measured for the above mentioned parameters. This enabled an accurate measurement of flow and the collection of representative samples.

Influent and effluent concentrations of anionic surfactants were monitored every 2–3 days for a period of 2 months. Surfactants were measured with cuvette tests based on the MBAS Standard Method [9].

Biogas produced in the UASB reactors passed a washing bottle containing sodium hydroxide 15%w to remove CO_2 and was subsequently measured with gas counters (MGC, Ritter). Dissolved methane in the effluent was calculated according to Henry's law and taken into account in the methane production.

2.4. Determination of sludge yield

TSS and VSS concentration in the reactors were measured three times a week for the SBRs and twice a month for UASBs. Sludge yield was determined from a plot of cumulative production of volatile suspended solids versus the cumulative amount of COD removed. The yield was calculated with Equation 1 and 2. The amount of VSS in the influent was negligible, and therefore it was not used for the calculations.

$$yield = \frac{\Delta VSS_{reactor} + \Sigma VSS_{wasted} + \Sigma VSS_{effluent} - \Sigma VSS_{influent}}{\Sigma COD_{removed}} \tag{1}$$

$$COD_{removed} = V_{effluent} \cdot (COD_{in_{total}} - COD_{out_{filtered}}) \tag{2}$$

COD$_{in_{total}}$ is the total COD concentration in the influent. $COD_{out_{filtered}}$ is the COD concentration of the effluent filtered through $>$12–25 μm pore size paper filter (Black ribbon, 589/1 grade, Whatman). Removed COD was calculated as the product of the volume of effluent collected ($V_{effluent}$) and the difference between COD_{in} and $COD_{out_{filtered}}$ measured in the period of collection of that effluent volume. Total effluent COD was not used for the calculation of the yield, since suspended solids in the effluent were considered to be sludge.

SRT was calculated as the ratio between the amount of VSS in the reactor and the sludge production rate (g VSS/d) over the operational period t, see Equations 3 and 4.

$$SRT = \frac{VSS_{reactor}}{\textit{Sludge production rate}} \tag{3}$$

$$\textit{Sludge production rate} = \frac{\Sigma VSS_{wasted} + \Sigma VSS_{effluent}}{t} \tag{4}$$

The data presented in Table 3 have been statistically treated to minimize the influence of outliers in the interpretation. Outliers were defined as values outside the range of the mean $\pm$ 2*(standard deviation). For the calculation of yield, SRT, and mass balances all data were taken into account.

3. Results and Discussion

3.1. Characteristics of greywater

Average concentrations of COD in the influent were similar in the two test periods, *i.e.*, 830 mg/L (Table 2). Of the COD, the suspended fraction accounted for approximately 50% and the remaining part was equally distributed between the colloidal and dissolved fractions. Greywater in this study had a relatively high concentration of COD compared to other studies previously performed in regions of similar climate and customs. For example, in Germany COD concentrations of 640 mg/L [6] and 420 mg/L [10] have been measured. In Sweden concentrations have been reported of 588 mg/L [11] and at another location in the Netherlands 450 mg/L [8]. The higher COD concentrations in this study were probably due to the lower volume of greywater produced at this particular site in Sneek (70 L/pd *vs.* the Dutch average of 90 L/pd) [8].

Table 2. Characteristics (averages with standard deviations) of greywater used in this study (all values are in mg/L).

	days 1–168 average	days 84–168 average
COD total	833 ± 188	827 ± 204
COD suspended	411 ± 151	385 ± 167
COD colloidal	204 ± 58	227 ± 65
COD dissolved	224 ± 59	196 ± 52
Anionic surfactants		43.5 ± 6.5
Total N	41.2 ± 27.2	29.9 ± 11.0
NH_4-N	1.0 ± 0.7	0.6 ± 0.4
NO_3-N	0.12 ± 0.08	0.12 ± 0.07
Total P	6.6 ± 2.7	5.8 ± 1.4

Greywater was found to have a higher temperature than combined wastewater because of the use of warm water in showering, laundry and kitchen. Eriksson *et al.* [1] reports temperatures ranging between 18–38 °C. In this study, the temperature was set at 32 ± 3 °C.

3.2. General systems performance

Table 3 summarizes the operation and average treatment performance of the reactors. For the combined system the table shows the performance of the individual reactors (UASB7 and SBR6) and for the combination (UASB7 + SBR6). Mean VLR was similar for the 3 systems, with a value around 1.6 kg COD/m^{3}d. Reactor performances were stable with respect to COD removal and average COD concentrations in the effluent.

Under anaerobic conditions, the removal of COD averaged 51% in UASB12 and 39% in UASB7, whereas under aerobic conditions 90% of the COD was removed in SBR12 and 82% in SBR6. The combined anaerobic-aerobic system achieved a COD removal of 89% which is similar to the performance of the aerobic reactor operated at an HRT of 12 h. Average concentrations of COD in the effluents of each system were 82, 392 and 100 mg/L for the aerobic, anaerobic and anaerobic-aerobic system, respectively.

3.3. Removal of different COD fractions

Variation in the VLR was due to the variation in the COD concentration of influent, the COD fractions of the influent and HRT in the studied reactors (Figures 2, 4, 5, 3) over the operational period. In the figures, diagonal lines correspond with 100% removal. The data of the aerobic reactors approached this 100% removal efficiency for all COD fractions. Furthermore, the data suggest that in theory a further increase of the VLR (*i.e.*, a shorter HRT) might be possible while maintaining a high level of COD removal. It was noted, that at a VLR above 2 kg COD/m^3d a COD removal of 90% can be achieved (Figure 2). A higher VLR, however, would also imply higher sludge concentrations and settleability may therefore become a limiting factor.

In reactors UASB12 and UASB7, COD was poorly removed and was mainly accomplished by the removal of suspended COD, which was 79% for UASB12 and 72% for UASB7 (Figures 4 and 5). In another study on greywater, a total COD removal of 52% was achieved in an UASB operated at HRTs of 10 and 6 h [6]. The difference between the results of our study and those published by Elmitwalli and Otterpohl [6] is the higher concentration of COD in our influent and the greywater sampling procedure. The effluent COD in Elmitwalli and Otterpohl [6] was 310 mg/L at an HRT of 10 h and 327 mg/L at an HRT of 6 h. Despite of having higher influent COD, in our study the COD removal was lower, leading to effluent COD concentrations of 392 and 528 mg/L for UASB12 and UASB7, respectively.

The difference in HRT in the anaerobic reactors strongly affected the removal of the dissolved COD. A removal of 30% was achieved at an HRT of 12 h but only 12% at an HRT of 7 h. Similarly, 16% of the colloidal COD was removed at an HRT of 12 h and only 10% at an HRT of 7 h. The removal of suspended COD remained high at both HRTs, 79% at 12 h and 72% at 7 h.

Poor COD removal of the anaerobic reactors can be attributed to the limited removal of colloidal COD. It is known that the colloidal fraction of the COD is poorly removed in UASB reactors because it cannot be entrapped or flocculated in the sludge bed, and the HRT is not long enough for its degradation [12].

Table 3. Operation and performance of biological reactors for greywater treatment.

	SBR 12 (aerobic system)	SBR6	UASB12 (anaerobic system)	UASB7	UASB7 + SBR6 (anaerobic-aerobic system)
HRT (h)	11.7 ± 1.1	6.1 ± 0.8	12.3 ± 1.8	7.0 ± 2.0	13.17 ± 2.03
VLR (kg COD/m^3d)	1.6 ± 0.5	1.9 ± 0.4	1.7 ± 0.4	2.7 ± 0.8	1.5 ± 0.6
COD removal rate (kg COD/m^3d)	1.5 ± 0.4	1.5 ± 0.4	0.8 ± 0.3	1.1 ± 0.6	1.4 ± 0.5
SLR (kg COD/kg VSS d)	0.29 ± 0.07	0.6 ± 0.3	0.12 ± 0.04	0.23 ± 0.08	**
Sludge concentration (g VSS/L)	5.5 ± 1.1	3.3 ± 1.1	12.5 ± 2.4	12.7 ± 4.3	**
SRT (d)	15	379	392	97	**
Yield (kg VSS/kg COD)	0.12	0.06	0.08	0.18	0.18
COD removal (%)	90 ± 7	82 ± 06	51 ± 13	39 ± 15	89 ± 3
COD effluent (mg/L)	82 ± 47	100 ± 33	392 ± 85	528 ± 180	100 ± 33
Anionic surfactants (mg/L)	1.4 ± 1.2	1.3 ± 1.5	33.4 ± 4.1	35.9 ± 5.3	1.3 ± 1.5
Effluent total N (mg/L)	31 ± 20	26 ± 13	34 ± 17	32 ± 10	26 ± 13
Effluent NH4-N (mg/L)	0.35 ± 0.20	0.4 ± 0.1	4.7 ± 2.1	5.4 ± 2.4	0.4 ± 0.1
Effluent NO3-N (mg/L)	1.5 ± 1.4	22.6 ± 13.5	0.2 ± 0.1	0.2 ± 0.1	22.6 ± 13.5
Effluent total P (mg/L)	4.4 ± 2.4	5.8 ± 1.7	5.3 ± 1.5	6.1 ± 1.7	5.8 ± 1.7
Effluent VSS (mg/L)	45 ± 61	30 ± 26	7 ± 9	21 ± 23	30 ± 26
Removal total N (%)	35 ± 37	26 ± 27	15 ± 33	–1 ± 63	2 ± 56
NH4-N removal (%)	51 ± 47	92 ± 4	*	*	7 ± 86
Total P removal (%)	28 ± 50	31 ± 11	11 ± 28	1 ± 36	3 ± 44
Methane flow (NL/d)	**	**	0.76	0.8	0.8
Methane production (NL/m^3)	**	**	123	71.5	71.5

* No ammonium removal

** Not applicable

Figure 2. COD removal rate *vs.* volumetric loading rate in SBR12.

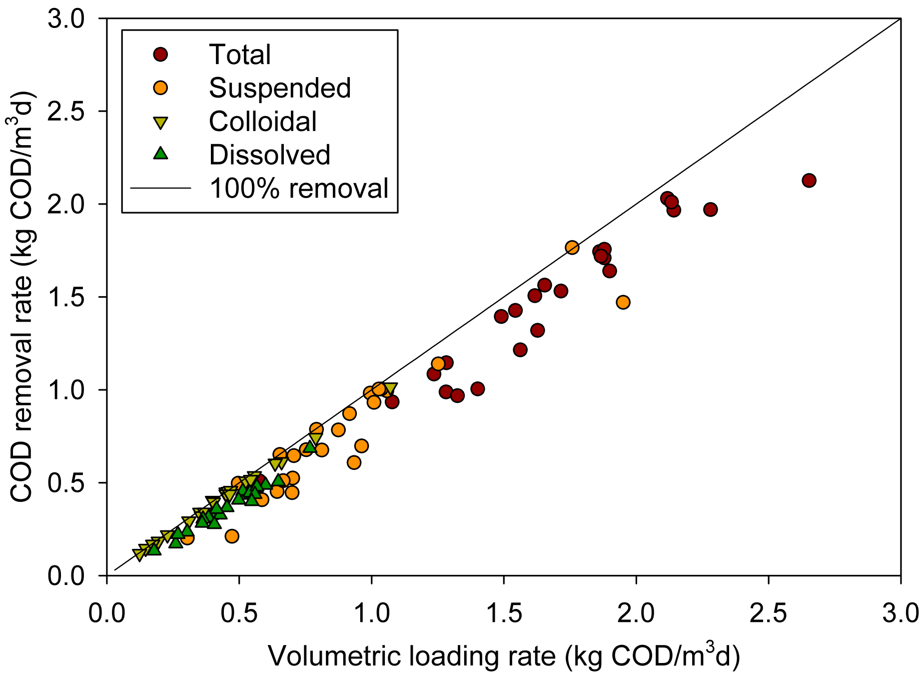


Figure 3. COD removal rate *vs.* volumetric loading rate in SBR6.

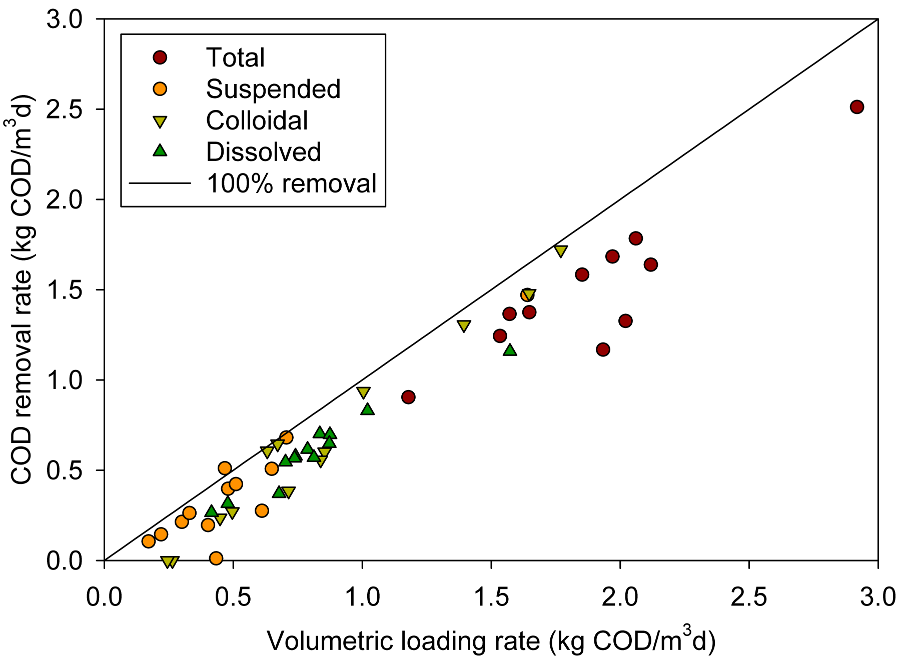


Figure 4. COD removal rate *vs.* volumetric loading rate in UASB12.

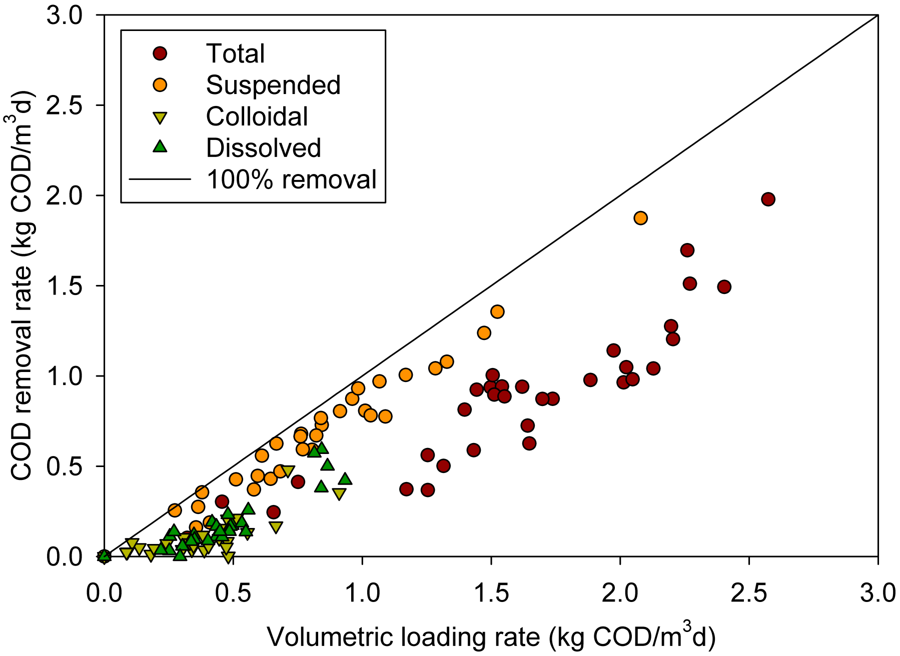
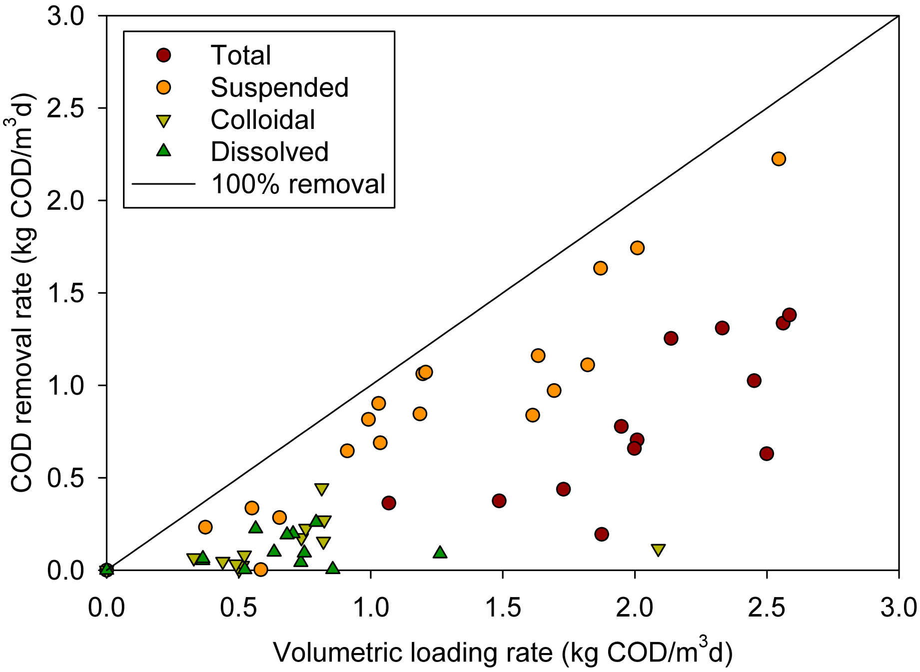


Figure 5. COD removal rate *vs.* volumetric loading rate in UASB7.

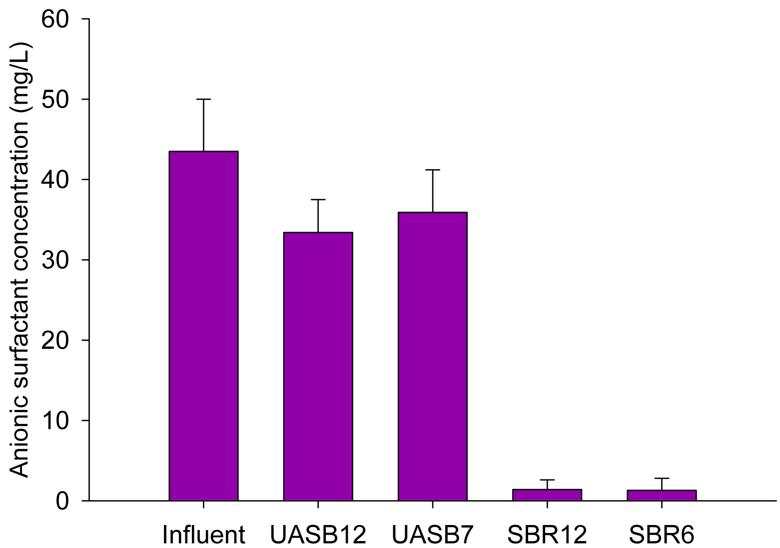


100% removal line is used as a reference, points laying at the proximity of this line indicate high COD removal efficiency.

3.4. Surfactant effect

The performance of the UASB reactors was most likely affected by the high concentration of anionic surfactants, which constituted about 15% of the influent COD. The average influent concentration of surfactants was 43.5 mg/L, of which UASB12 and UASB7 only removed 24 and 17%, respectively (Figure 6). In contrast, the aerobic SBR's removed 97% of the anionic surfactants, leading to average effluent concentrations of 1.4 and 1.3 mg/L in SBR12 and SBR6, respectively. Anionic surfactants are readily biodegradable under aerobic conditions as opposed to anaerobic conditions, at which they are unlikely to be degraded [13]. Removal that occurred in the UASB reactors was probably due to adsorption to the sludge particles. Furthermore, Garcia Morales *et al.* [14] found that acidogenic bacteria in anaerobic sludge were inhibited by linear alkylbenzene sulfonates (LAS, widely used anionic surfactants), with an EC50 of 19 mg/L. With the concentration of anionic surfactants in the range of 30–50 mg/L, the inhibition of hydrolysis likely occurred in these UASB reactors, leading to sludge accumulation.

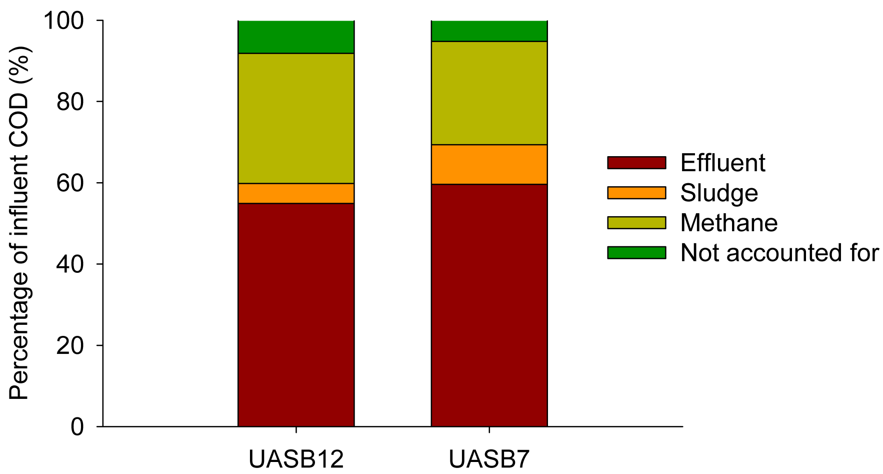
Figure 6. Influent and effluent concentrations of anionic surfactants in 4 bioreactors treating greywater.



3.5. COD mass balances

Mass balances of COD were done for the anaerobic reactors (Figure 7). The majority of the influent COD was not converted in the reactors and 55 and 60% was found in the effluent, respectively for UASB12 and UASB7. Methanization of the influent COD was 32% in UASB12 and 25% in UASB6. COD transformed into sludge was determined to be 5% in UASB12 and 10% in UASB7. A small percentage (8 and 5%) could not be accounted for.

Figure 7. Cumulative COD mass balance on anaerobic reactors UASB12 (period of 168 days) and UASB7 (period of 84 days).



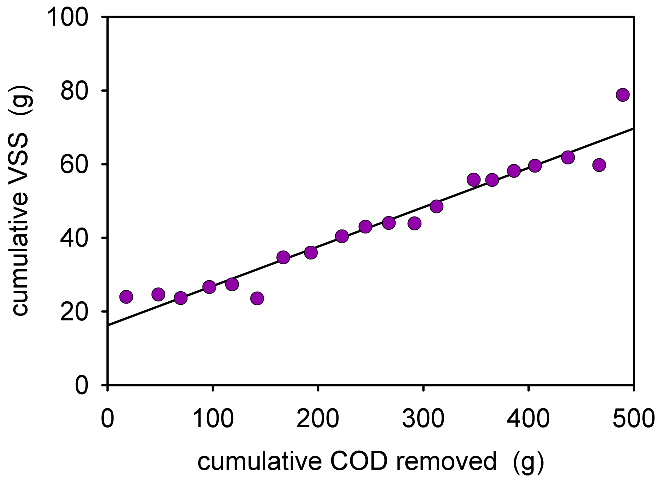
3.6. Sludge yield

The researched aerobic reactors exhibited a sludge yield of only 0.06 and 0.12 g VSS/g COD for SBR6 and SBR12, respectively. Figure 8 shows the yield curve for reactor SBR12. SRT at SBR6 was 378 days which explains the low sludge yield measured in this reactor. In contrast to SBR6, the yield of SBR12 was accompanied by a relatively short SRT of only 15 days. Although this is a good feature of the process, it is not clear why the sludge yield is so low. Based on the total sludge production, the amount of VSS in the effluent was 43% for SBR12 and 95% for SBR6. These figures indicate a need for another reactor configuration to prevent sludge from washing out, e.g., a membrane bioreactor. Nevertheless, both aerobic reactors showed a very low sludge yield, compared to reported average values of 0.30–0.50 [15].

Anaerobic reactors had a sludge yield of 0.08 (UASB12) and 0.18 kg VSS/g COD (UASB7). From the total sludge production, 84 and 62% of the VSS was in the effluent, respectively. Part of the sludge yield cannot be attributed to the formation of biomass, and was caused by the accumulation of slowly biodegradable particulate organic matter in the sludge bed.

The difference between the yields in reactors UASB12 and UASB7 was probably due to lower hydrolysis of slowly biodegradable organic matter in the sludge bed at a higher VLR, and therefore a lower SRT. Nevertheless at an SRT of 97 days and a temperature of $32 \pm 3\,^{\circ}\mathrm{C}$ a complete degradation of biodegradable COD was expected [5]. The combined sludge yield for the anaerobic + aerobic system was 0.18 kg VSS/kg COD, which is higher than the aerobic system 0.12 kg VSS/kg COD and the anaerobic one (0.08 kg VSS/kg COD). Even though it has a low sludge yield, the anaerobic system cannot be selected for greywater treatment due to the poor COD removal, therefore the aerobic system is preferred.

Figure 8. Cumulative VSS produced as a function of cumulative COD removed in SBR12 for a period of 168 days.



3.7. Nutrient removal

Nutrient removal in all reactors was low and exhibited large variations. This resulted in effluent concentrations of nitrogen in a range of 26–34 mg/L and effluent phosphorus concentrations of 4.4–6.1 mg/L (Table 3). In spite of the rather long SRT of 15 days in SBR12, nitrification was hardly observed, as can be seen from the low nitrate concentration in the effluent (1.5 mg/L). Furthermore, differences in influent and effluent total nitrogen and ammonium concentrations strongly indicate that a large fraction of the nitrogen in the influent was organically bound. In sewage, most of the nitrogen is present as ammonium [16], this is probably because 90% of the nitrogen in sewage comes from urine [2], which is quickly converted into ammonium. In greywater, most of the nitrogen was organically bound, apparently limited ammonification took place and therefore it was not available for nitrification/denitrification in the SBR12. On the other hand, nitrification in SBR6 was significantly higher, with a mean nitrate concentration of 22.6 mg/L. The higher nitrification in SBR6 was possibly due to a higher ammonium concentration into SBR6 (5.4 $\pm$ 2.4 mg/L, as opposed to 1.0 $\pm$ 0.7 mg/L into SBR12) and the much longer SRT of 379 days (compared to the SRT of 15 days in SBR12).

Table 4 shows the mass of N and P that was removed in the aerobic reactors, along with the estimated mass of N and P required for biomass production (assuming 12% of VSS is constituted by N and 2% by P [15]). In SBR12, nutrient removal could be explained from this biomass production. In SBR6 however, the removal of nutrients could not be attributed to biomass production alone, indicating that a significant fraction of total N was removed with the sludge as non-biomass associated particles.

Table 4. Nutrient removal and calculated consumption by sludge production.

	mass of N removed (g)	mass of N in sludge	mass of P removed (g)	mass of P in sludge (g)
SBR12	9.88	8.48	1.15	1.41
SBR6	10.35	3.08	1.02	0.51

3.8. *Energy production*

Whether a combined system is worthwhile from the energetic point of view is an important aspect to consider. With a greywater production of 70 L/p·d and a COD of 830 mg/L as determined in this study, one person produces 58.1 g COD/d (48% of the COD load from household wastewater). The energy required for aerobic treatment of greywater treatment was estimated to be 8 kWh/p y, based on the energy requirements for aeration of municipal wastewater of 0.31 kWh/m^3 [17]. The methane amount that can be gained from an anaerobic-aerobic system as shown in this study is 5 L/p d, which represents 18 kWh/p y. For the aerobic post-treatment 4 kWh/p y are required, assuming the energy requirements for the aerobic post-treatment are 50% of those of aerobic greywater treatment, since 50% of COD was removed in the UASB reactor. The combined anaerobic-aerobic system, therefore, has a net energy production of 14 kWh/p y. In the context of the whole sanitation concept, black water mixed with kitchen refuse can produce 28 L methane/p·d, *i.e.*, 101 kWh/p y [18]. If anaerobic treatment of black water is installed, treatment of greywater with an aerobic system would consume 8% of the energy production of black water leaving an amount of 93 kWh/p y. On the other hand, treatment of greywater with an anaerobic-aerobic system could produce 14% more methane than by black water treatment alone, to an energy production of 115 kWh/p y. However, two UASB reactors are needed instead of one, which makes this treatment option unfeasible, given the limited energy gain. Therefore, the aerobic system is preferred for the treatment of greywater. A combined anaerobic-aerobic system may also have a positive effect on the removal of xenobiotic organic compounds, an aspect that is yet to be investigated.

4. Conclusions

- Aerobic treatment of greywater achieved 90% of COD removal and 97% removal of anionic surfactants at a HRT of 12 hours and a temperature of 32 ± 3 °C.

- The low sludge yield of the aerobic SBR (0.12 g VSS/g COD) operated at 32 ± 3 °C and an HRT or 12 hours makes it an attractive process for the treatment of greywater.

- Anaerobic treatment of greywater at an HRT of 12 hours and a temperature of 32 ± 3 °C reaches 51% removal of COD, with a greywater methanization rate of 32% and with a poor removal (24%) of anionic surfactants.

- A combined anaerobic-aerobic system operated at 32 ± 3 °C and at an HRT of 12 hours did not give an advantage, compared to aerobic treatment, in regard to the removal of COD from greywater and sludge yield. However, the benefits of this configuration depend on factors such as gas use and energy input.

- Based on COD removal, sludge yield and energy considerations, treatment of greywater in an aerobic system is preferred over an anaerobic system and a combined anaerobic-aerobic system.

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