Recovery of particulate matter from a high-rate moving bed biofilm reactor by high-rate dissolved air flotation

Oscar Sanchez, Marc-André Labelle, Alain Gadbois, Edith Laflamme, Peter L. Dold, Antoine Laporte and Yves Comeau

ABSTRACT

High-rate biological wastewater treatment processes for carbon recovery are able to improve the energy balance and carbon footprint of water resource recovery facilities. Combination of a high-rate moving bed biofilm reactor (HR-MBBR) with a rapid flotation (HR-DAF), as a replacement for the 'A stage' of the A-B process, can achieve this objective. The main goal of this study was to maximize the capture of biodegradable particulate matter from an HR-MBBR effluent by an HR-DAF. A pilot-scale HR-DAF process was operated downstream of an HR-MBBR treating screened municipal wastewater. The particulate biodegradable matter recovery was evaluated by determining the total suspended solids (TSS) removal efficiency. TSS recovery in experiments without chemicals at low surface loading rates (<15 m/h) and high recycle ratio (>25%) was 94 ± 1%. By using a tannin-based polymer, the solids capture efficiency of the HR-DAF was slightly improved with TSS recovery reaching 96 ± 1% at a high SLR (at least 22 m/h) and low recycle ratio (14%). The anaerobic biodegradability of the tannin tested was determined to be 17%. The HR-DAF process downstream of an HR-MBBR gave a very good particulate matter recovery that offers a promising alternative to the A-B process for carbon recovery.

Key words | high-rate moving bed biofilm reactor, high-rate rapid dissolved air flotation, suspended solids, tannin

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INTRODUCTION

Wastewater engineering is gradually evolving from treatment of wastewater to the recovery of water resources such as water, energy (e.g., thermal and chemical), nutrients (e.g., phosphorus and nitrogen) and biosolids. Water resource recovery facilities (WRRFs) increasingly aim at becoming energy self-sufficient and having a low carbon footprint. Becoming energy-positive will require not only minimizing energy consumption but also maximizing energy recovery, mainly via organic matter methanogenesis (Jimenez *et al.* 2015; Ødegaard 2016).

High-rate biological wastewater treatment processes for carbon recovery such as done by the A-B process (Versprille *et al.* 1985; Wett *et al.* 2007) are of interest due to their ability to decrease energy demand for aeration and increase methane production during anaerobic digestion, thus improving the energy balance and the carbon footprint of WRRFs. The combination of a high-rate moving bed biofilm reactor (HR-MBBR) with a rapid solids separation process, as a replacement for the so-called 'A stage' of the A-B

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process, can achieve this goal, resulting in a compact and sustainable secondary treatment process.

In HR-MBBRs, clarification of the effluent is a challenge since the settleability of the biomass decreases with increasing organic loading (Ødegaard *et al.* 2000, 2010).

Conventional rate dissolved air flotation (DAF) systems (4–10 m/h) have been successfully used for separating particulate matter from MBBR effluents in many full-scale WRRFs. It is one of the most frequently used biomass separation processes for MBBR effluents, along with sedimentation. They are often combined with metal salts and/or cationic polymers (Ødegaard 2006; Ødegaard *et al.* 2010; Ivanovic & Leiknes 2012). The efficiency of a conventional rate DAF (4 m/h) downstream of an HR-MBBR was studied by Helness *et al.* (2005) at pilot scale and showed a suspended solids (TSS) recovery of 89%.

Innovative separation processes are needed to reduce energy costs, footprint and the addition of unbiodegradable chemicals. Thus, there is an increasing interest in natural-based chemicals, such as tannins. Previous experiments have revealed the high performance of tannins in wastewater treatment and they can be potential substitutes for metal coagulants and synthetic polymers (Ozacar & Sengil 2003; Beltran-Heredia & Sanchez-Martin 2009; Schubert *et al.* 2013; Brosseau *et al.* 2016).

In the last 15 years, high-rate DAF (HR-DAF) systems operating at surface loading rates of 15–30 m/h and greater have been developed. Conventional rate DAF is still used, but HR-DAF is an alternative which is now used in many drinking water treatment plants (Edzwald 2010). The efficiency of an HR-DAF system for the recovery of particulate matter in an HR-MBBR effluent, however, has not yet been reported.

In this work, an HR-DAF was operated at surface loading rates up to 22 m/h (including recycle flow) and a natural tannin-based polymer was used as a replacement for traditional metal coagulants. The main goal of this study was to maximize the capture of biodegradable particulate matter from an HR-MBBR effluent by HR-DAF for energy recovery via anaerobic digestion. The two specific objectives of this project were (1) to determine the particulate matter capture potential without chemical addition and by using a tannin-based low-molecular weight cationic polymer as sole coagulant and (2) to determine the biodegradability of the tannin-based polymer.

METHODS

A pilot-scale unit of the HR-DAF process called Spidflow® which uses an innovative micro-bubble air diffuser was provided by Veolia Water Technologies. This unit was operated during a 5-month period downstream of a 1.9 m^3 HR-MBBR treating screened (6 mm) municipal wastewater from the Repentigny WRRF (80,000 population equivalent) in Canada (Figure 1). The bioreactor was filled with Kaldnes K5 carriers (specific surface area of 800 m^2/m^3) occupying 50% of the bioreactor volume. The water temperature in

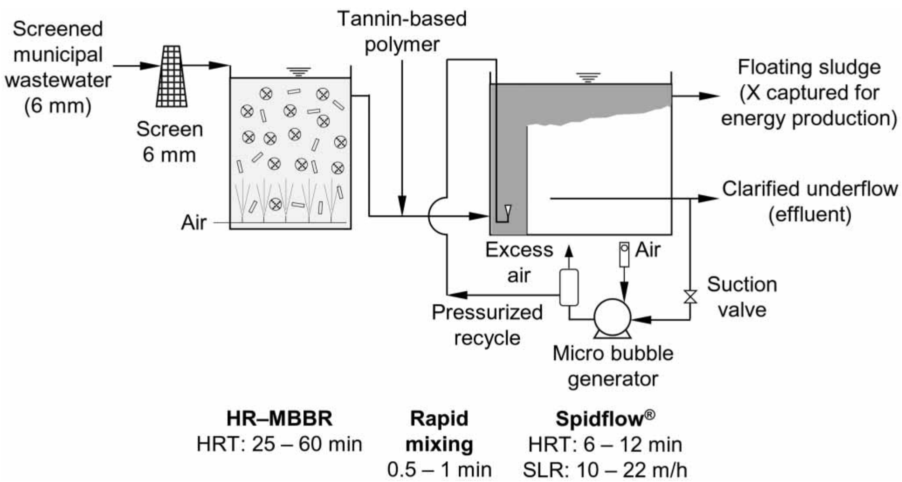

Figure 1 | Flow scheme of the high-rate process investigated.

the HR-DAF varied between 17 and 23 °C. The influent flowrate varied between 1.6 and 4.0 m^3/h, corresponding to hydraulic retention times (HRTs) of 25 to 60 min and organic loading rates (OLRs) of 20 to 65 g COD m^{-2} d^{-1} for the HR-MBBR, and a surface loading rate (SLR) of 10 to 22 m/h for the HR-DAF. The HR-DAF separation area was 0.22 m^2. The HRT and the OLR of the HR-MBBR, and the SLR and pressurized recycle ratio (R) of the HR-DAF system were controlled by the influent flowrate of the pilot unit.

The efficiency and performance of the HR-DAF process (Figure 2) were assessed without chemicals and with the use of a tannin-based polymer as sole coagulant. The particulate matter recovery potential of the HR-DAF process was evaluated in terms of TSS capture efficiency by collecting samples from the influent and effluent of the HR-DAF unit for each experimental run. The HR-DAF performance was evaluated in terms of the micro-bubble generator pressure (P), the SLR (including the recycle flow) and the pressurized recycle ratio for each experimental run.

First, the effect of the micro-bubble generator pressure (P) (3 to 6 bar) on the HR-DAF process efficiency was investigated. Second, an optimal dosage range of a tannin-based low-molecular weight cationic organic polymer (Hydrex 3818, Veolia Water Technologies) obtained from reforested *Acacia mearnsii* trees also known as Wattle extract was determined. Finally, the effect of the SLR and the pressurized water recycle ratio on the HR-DAF process solids capture efficiency and effluent quality performance was investigated. Carbonaceous BOD_5 ($CBOD_5$) and COD fractions were determined to characterize the colloidal matter capture efficiency and the secondary clarified effluent quality.

One of the most important flotation parameters is the air/solids (A/S) ratio (Ødegaard *et al.* 2010; Shammas *et al.* 2010; Metcalf & Eddy-AECOM 2014). It was evaluated using Equation (1) (Shammas *et al.* 2010; Metcalf & Eddy-AECOM 2014):

$$\frac{A}{S} = \frac{1.3C_s(fP_a - 1)R}{X_fQ} \quad (1)$$

where A/S is the air to solids ratio, mL/mg; C_s is the air solubility at 1.0 atm pressure and operating temperature, mL/L; X_f is the concentration of SS in the influent, mg/L; P_a is the absolute saturation pressure, atm absolute; f is the fraction of air dissolved at pressure P_a, usually 0.5; R is the pressurized recycle flowrate, m^3/d; and Q is the mixed-liquor flowrate, m^3/d. The factor 1.3 is the weight in milligrams of 1 mL of air.

A portion of the HR-DAF effluent was recycled, pressurized and partly saturated with air using a micro-bubble generator (Karyu Turbo Mixer Nikuni). This device has a pumping mechanism that generates pressurized air enriched water by negative pressure sucking air and mixing it with water at the same time. The pressurized recycle was mixed

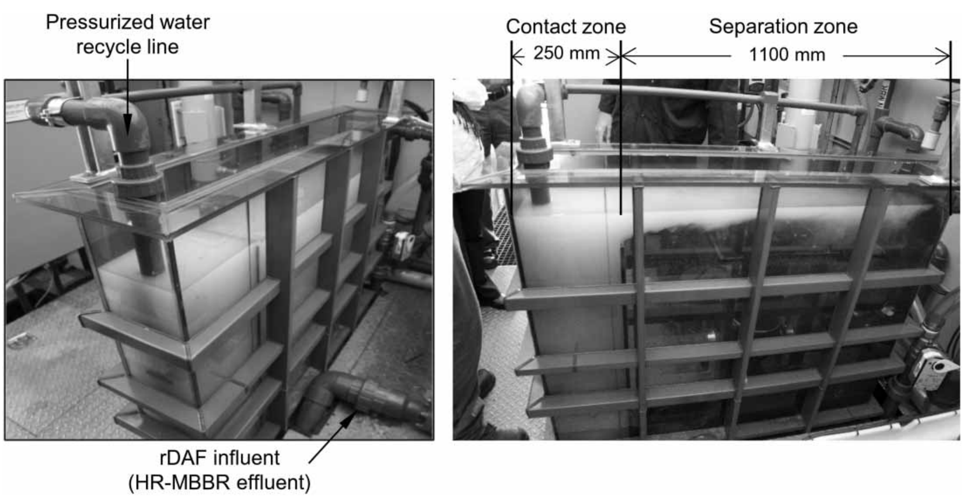

Figure 2 | Photograph of the pilot-scale HR-DAF process (the influent line to the tank is under the floor).

with the non-pressurized main stream just before the entrance to the flotation chamber. The recycle ratio was influenced by pilot influent flowrate and by pressurized recycle flow, the latter being proportional to micro-bubble generator pressure. The recycle ratio was calculated as follows:

$$R(\%) = \left(\frac{Q_r}{Q_{in}}\right) \times 100 \qquad (2)$$

where Q_r is the pressurized recycle flow and Q_{in} is the wastewater flow into the HR-DAF process. The coagulant was dosed with a variable speed peristaltic pump into a four-inch pipe ahead of three 90° elbows located upstream of the inlet of the HR-DAF unit. The rapid mixing time was controlled by the pilot influent flowrate, resulting in a contact time (coagulation-flocculation step) of 30 to 60 seconds.

Grab samples were collected into 1 L plastic bottles from the influent at the beginning and end of each experimental run to check variations in influent characteristics. When stable operating conditions were met, the HR-DAF process was run for a time equal to three times the nominal HRT in the HR-DAF unit. During the transition period, the floating sludge was scraped and regularly removed from the surface of the unit by using a manual sludge scraper. Then, the floating sludge was accumulated at the surface of the separation chamber over a period of 10 minutes. Samples were collected from the floating sludge to determine their solids content. At the same time, 10-minute composite samples were collected from the clarified effluent (using 2 L plastic bottles).

The biochemical methane potential (BMP) assay was performed in batch tests to determine the portion of tannin-based polymer that could be biologically degraded under anaerobic conditions. First, the COD of Hydrex 3818 was determined analytically (g COD/g Hydrex 3818). The BMP of the substrate (Hydrex 3818) was then evaluated (mL CH_4/g COD added) for which an inoculum was obtained from the Repentigny WRRF anaerobic digester. The BMP protocol followed in this study was based on the principles described by Angelidaki *et al.* (2009) and Hansen *et al.* (2004). The tannin-based polymer and the active anaerobic inoculum were added to 160 mL serum bottles. Initial inoculum to substrate ratio (I:S) was 1.8 g COD/g COD. The amount of substrate was 1.06 g COD per bottle resulting in a substrate concentration of 8.3 g COD substrate/L. pH was measured, and bottles were vented with N_2 and sealed immediately using rubber septa and aluminum crimp caps. Once sealed, the bottles were placed in an incubator and maintained at 35 °C. For each BMP trial, two additional bottles with distilled water but without substrate were included to determine the background methane production from the inoculum. The BMP assay was ended when the cumulative biogas curve reached the plateau phase, typically after 30 days. Cumulative pressure was measured inside the bottles using a digital manometer Ashcroft D625, every day at the beginning of the test and at longer time intervals as the BMP assay progressed. Finally, the volume of biogas was calculated at standard conditions from the pressure data, according to the ideal law of gases:

$$PV = nRT \qquad (3)$$

where P, V, n, and T are, respectively, absolute pressure (kPa), volume (m^3), moles, and temperature (K) of the gas; and R is the universal gas constant (8.3144 kPa L K^{-1} mol^{-1}). A sample of biogas was taken from the serum bottles to determine the methane content by gas chromatography with a SCION 456-GC by Bruker. The biodegradability of substrate was estimated by the ratio between the degradable COD and total COD added. Degraded COD was calculated from the observed specific methane yield and the theoretical 0.35 L CH_4/g COD at standard conditions (Metcalf & Eddy-AECOM 2014).

TSS, COD and $CBOD_5$ were determined according to APHA *et al.* (2012). Glass microfibre 1.2 μm filters (Whatman® 934-AH™, GE Healthcare Life Sciences, GBR) were used for TSS and filtered COD (colloidal and soluble COD, CS_{COD}). Soluble COD (S_{COD}) was determined by the flocculated-filtered COD test (Mamais *et al.* 1993) using 0.45 μm cellulose membrane filters (MF-Millipore™, EMD Millipore, USA). The following definitions were used for COD size fractionation into particulate X_{COD}: >1.2 μm, colloidal C_{COD}: 0.04 to 1.2 μm and soluble S_{COD} < 0.04 μm fractions.

RESULTS AND DISCUSSION

Micro-bubble generator pressure

For experiments without chemicals, recovery of TSS from the HR-MBBR effluent increased as the pressure increased from 3 to 6 bar. Maximum TSS recovery was 95% at an SLR of 11 m/h (Figure 3). In experiments with a tannin-based coagulant, recovery of TSS increased as the pressure increased from 3 to 4 bar and above this critical pressure, the HR-DAF efficiency did not increase. Maximum TSS recovery was 97% at an SLR of 21 m/h (Figure 3). DAF facilities have been typically designed and operated in the range of 4–6 bar since the 1960s (Edzwald 2010). Optimal saturation pressures of 4–5 bar are usually selected for typical wastewater systems (Shammas *et al.* 2010). Saturation pressures of about 2 bar with high efficiency were reported by Feris & Rubio (1999) in a pilot-scale DAF of $Fe(OH)_3$ precipitates at SLRs close to 4 m/h by lowering the air/water surface tension in the saturator by adding a surfactant in the saturator.

Optimal micro-bubble generator pressures observed in this work were within the typical operating range reported for conventional DAF facilities. Without chemicals, the optimal saturation pressure were close to 6.0 bar while with a tannin-based coagulant, it was about 4.0 bar. The different micro-bubble generator pressures correspond to different water recycle flowrates and consequently to different pressurized water recycle ratios. Similar efficiencies to those obtained without chemicals at a pressure of 6 bar, an SLR of 11 m/h and a R ratio of 40%, were obtained by adding a tannin-based coagulant at a pressure of 3 bar, an SLR of 19 m/h and a R ratio of 12%.

From an energy consumption point of view, the use of a tannin-based coagulant allowed reduction of the operating pressure of the HR-DAF unit, thus suggesting a potential reduction of energy requirements. Potential energy reductions could be quantified and supported by a life-cycle cost statement in future work.

Tannin-based polymer as sole coagulant

The TSS recovery efficiency of the HR-DAF increased up to a dose of 17–20 mg/L of tannin-based coagulant at an SLR of 20 m/h and with a mixing time (coagulation-flocculation step) of 0.5–1 minute. The appearance of a TSS recovery plateau of 96–97% over this critical dose indicates that it was the optimal dose (Figure 4). Without coagulant, the recovery of TSS at a high SLR was poor (74%). The pH of the HR-MBBR effluent was 7.3 ± 0.1 and it was not affected by tannin addition. The SLR range is due to the fact that not all the tests were performed at the same influent flowrate of the pilot unit.

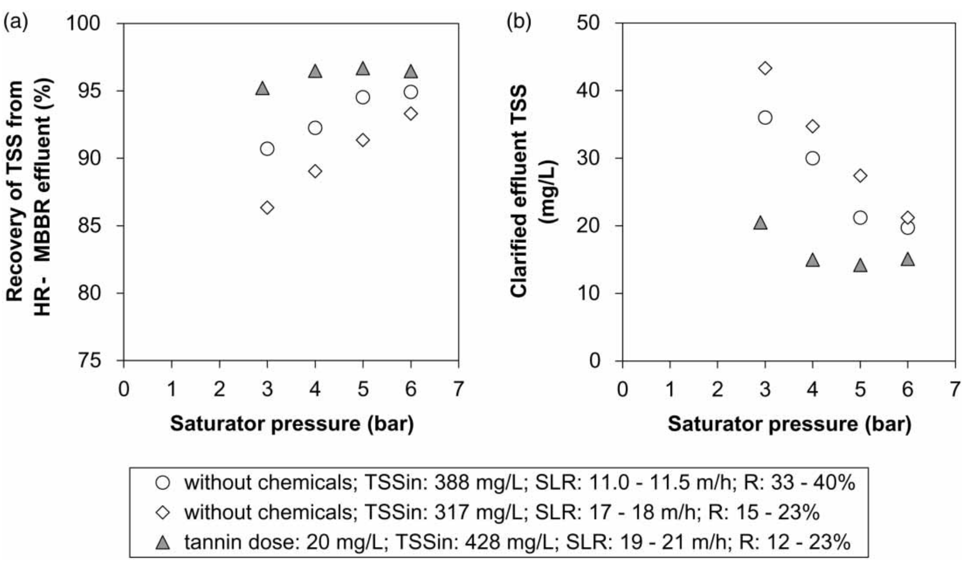

Figure 3 | Effect of micro-bubble generator pressure on TSS recovery (a) and effluent TSS (b). Operating conditions are summarized in the box.

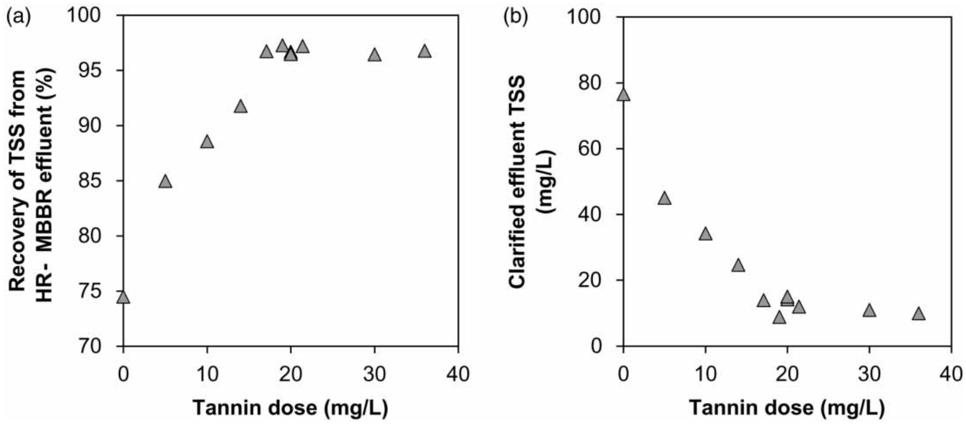

Figure 4 | Effect of tannin as sole coagulant on (a) TSS recovery and (b) effluent TSS. Influent TSS: 362 ± 63 mg/L; P: 5 bar; SLR: 19 ± 3 m/h.

The TSS recovery efficiency was similar to that observed by Brosseau *et al.* (2016) for an HR-MBBR/enhanced flotation process and higher than that reported by Melin *et al.* (2002), who reported a value 80% from a laboratory-scale HR–MBBR using a coagulation/flocculation/DAF system with low combined dosages of iron and cationic polymer, a rapid mixing time of 0.5–1 min and a flocculation time of 20 min. A TSS recovery of 89% was reported by Melin *et al.* (2004) and Helness *et al.* (2005) with a pilot-scale unit; they concluded that the flocculation time in a high-rate process can be short (7 min) with a low dosage of iron and cationic polymer.

Low-molecular weight polymers as sole coagulants for the HR-DAF were efficient for TSS recovery, in agreement with the results of Pilipenko (2007), but in contrast to those of Melin *et al.* (2002) and Ødegaard *et al.* (2004). Although the design of the rapid mixing step can be improved, the results show that the coagulation-flocculation residence time upstream of the HR-DAF unit can be very short, as suggested by Helness *et al.* (2005).

Effect of surface loading rate and recirculation ratio

Without chemicals, an HR-DAF TSS recovery of 94 ± 1% was obtained at low SLRs (<15 m/h) and high recycle ratios (>25%), corresponding to HR-MBBR OLRs of <50 g COD m^{-2} d^{-1} and HRTs of >35 minutes at an operating pressure of 6 bar. With a tannin-based low-molecular weight cationic polymer, an HR-DAF TSS recovery of 96 ± 1% was obtained at high SLRs (<22 m/h) and low recycle ratios (down to 14%), corresponding to HR-MBBR OLRs of <65 g COD m^{-2} d^{-1} and HRTs of >25 minutes at an operating pressure of 4 bar (Figures 5 and 6).

The effect of SLR and R on dry solids content of the floating sludge both without chemicals and with tannin-based low-molecular weight cationic polymer was not clear. The average dry solids content of the floating sludge was 44 ± 8 g/L without chemicals and 43 ± 8 g/L with tannin-based coagulant, indicating that the influence of tannin-based coagulant on floating sludge dry content was not significant.

The removal efficiency for TSS on a full-scale WRRF presented by Ødegaard *et al.* (2010) averaged 96% at nominal conventional SLRs ranging from 4 to 8 m/h which was similar to that obtained in this work when tannin-based polymer was used. The removal efficiency for TSS obtained in this work, without chemicals as with the tannin-based polymer, was higher when compared to pilot-scale results reported by Helness *et al.* (2005), which averaged 89% using low dosages of iron and cationic polymer at a constant SLR of 4 m/h.

A/S ratio defined as the mass of air precipitating per unit mass of wastewater solids is one of the most important flotation parameters (Ødegaard *et al.* 2010; Shammas *et al.* 2010; Metcalf & Eddy-AECOM 2014). If less than the optimum amount of air is employed in the DAF system, the efficiency of solids removal is reduced. If too much air is used, power is wasted. Therefore, it is important to optimize this variable by adjusting the pressure and the pressurized recycle flowrate (Shammas & Bennett 2010; Shammas *et al.* 2010).

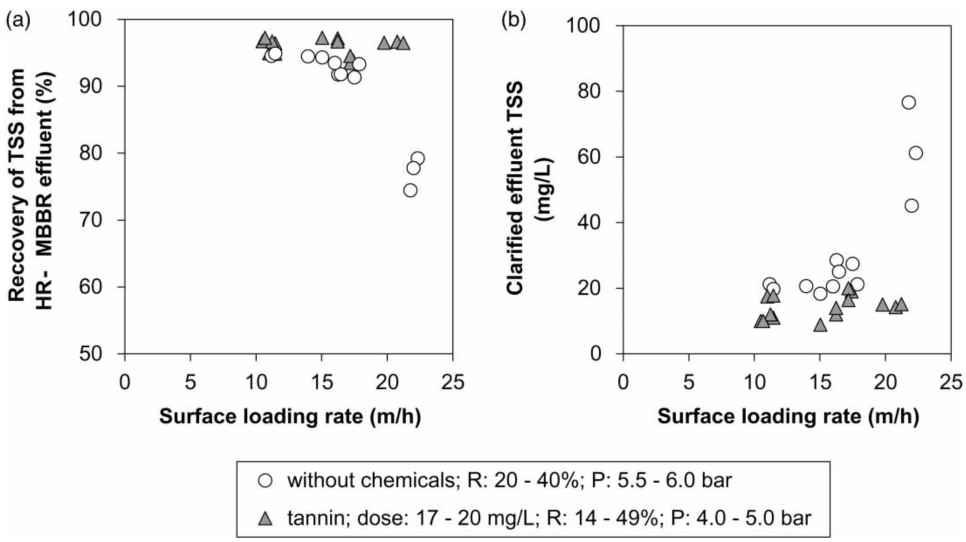

Figure 5 | Effect of surface loading rate on (a) TSS recovery and (b) effluent TSS. Influent TSS: 344 ± 54 mg/L.

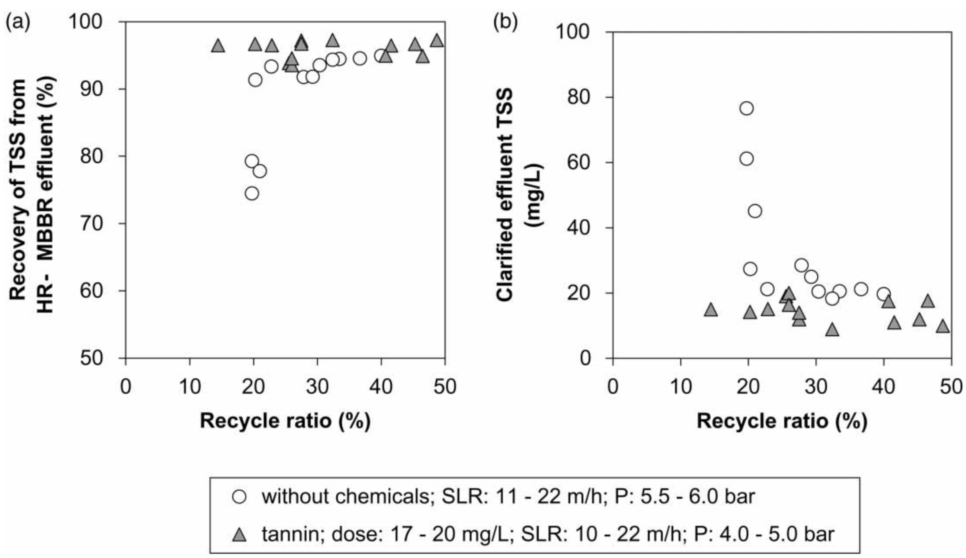

Figure 6 | Effect of recycle ratio rate on (a) TSS recovery and (b) effluent TSS. Influent TSS: 344 ± 54 mg/L.

Without chemicals, the efficiency of the HR-DAF decreased as the SLR approached 20 m/h (Figure 5). With the tannin-based low-molecular weight cationic polymer, the HR-DAF TSS recovery was stable throughout the range of SLRs tested. Therefore, to determine the optimal A/S ratio without chemicals, tests were conducted at SLRs up to 18 m/h. Without chemicals, a TSS recovery of 94 ± 1% was observed for A/S ratios of 0.03 to 0.05 g/g, and below this range of values the efficiency of HR-DAF decreased (Figure 7). With a tannin-based low-molecular weight cationic polymer, the optimal A/S ratio was around 0.01–0.02 g/g and a recovery of 96 ± 1% was observed for this range of values. Increasing the A/S ratio above this range of values did not have a significant effect on the efficiency of the HR-DAF.

The removal efficiency for TSS obtained in this work, both without reagents as with the tannin-based polymer, was somewhat higher than that observed at pilot scale by

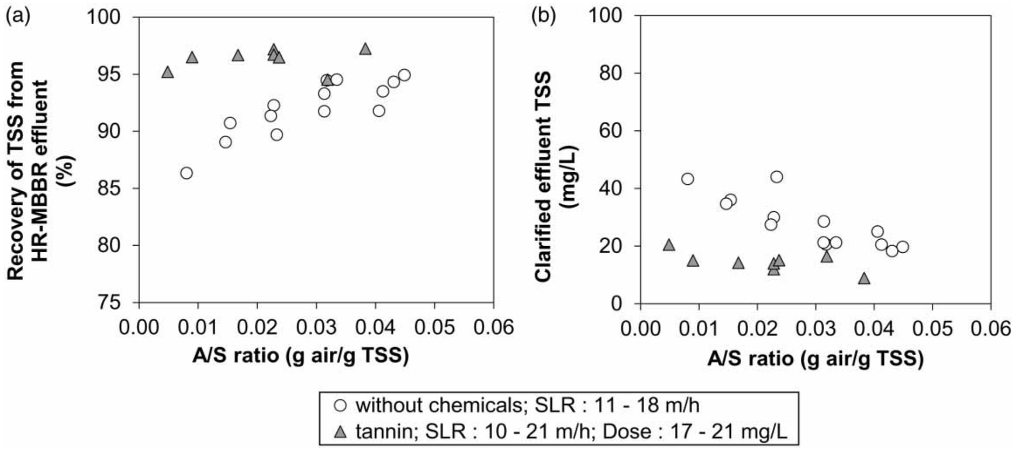

Figure 7 | Effect of air/solids ratio on (a) TSS recovery and (b) effluent TSS. Influent TSS: 344 ± 54 mg/L.

Helness *et al.* (2005), who obtained a mean SS recovery of 89% using low doses of iron and cationic polymer at a A/S ratio greater than 0.1. Lower A/S ratios were obtained with tannin addition, thus suggesting the potential for increased energy savings.

Secondary clarified effluent quality

Secondary effluent concentrations of less than 25 mg $CBOD_5$/L were achieved in two experimental runs at an HR-DAF SLR of 15 m/h (Table 1). $CBOD_5$ concentrations of 14 ± 3 mg/L and 15 ± 3 mg/L were observed in the clarified effluent with tannin-based polymer addition and without chemicals, respectively. Secondary effluent $CBOD_5$ reached in this study was similar to that reported in a pilot-scale study by Helness *et al.* (2005), who demonstrated that a secondary standard of 25 mg $CBOD_5$/L could be reached in a process with an HR-MBBR, flocculation HRT of 7 minutes and a conventional DAF (4 m/h). Colloidal matter, present at a very low concentation (7 mg/L) did not seem to be recovered efficiently by an HR-DAF with a tannin-based low-molecular cationic polymer (Table 1).

Biodegradability of tannin coagulant

The specific COD of the tannin used was 1.015 ± 0.006 g COD/g Hydrex 3818. The BMP test indicated that the

Table 1 | Organic matter fractionation of HR-MBBR and HR-DAF effluents with and without tannin-based polymer

			Effluent HR-MBBR (HR-DAF influent)	Clarified underflow without chemicals[a]		Clarified underflow tannin-based polymer[b]	
Parameter	Symbol	Unit	Value	Value	Treatment efficiency	Value	Treatment efficiency
Total suspended solids	TDS	mg/L	331 ± 6	18 ± 1	95%	9 ± 1	97%
Total 5-d carbonaceous BOD	BOD	mg/L	154 ± 19	15 ± 3	90%	14 ± 3	91%
Total COD	COD	mg/L	347 ± 7	59 ± 1	83%	50 ± 1	86%
Particulate COD	X_{COD}	mg/L	304 ± 3	16 ± 1	95%	11 ± 1	97%
Colloidal COD	C_{COD}	mg/L	7 ± 1	12 ± 2	–	7 ± 1	0%
Soluble COD	S_{COD}	mg/L	37 ± 1	32 ± 2	14%	33 ± 1	10%

[a]P: 5.5 bar; R: 32%; SLR 15 m/h.
[b]P: 5.0 bar; R: 32%; SLR 15 m/h. Dose: 19 mg tannin/L.

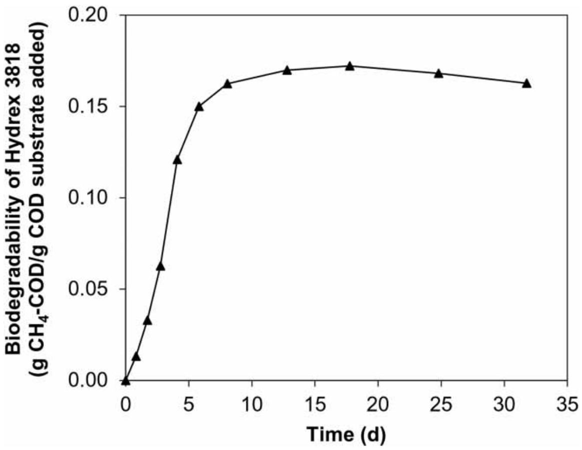

Figure 8 | Biodegradability of the tannin Hydrex 3818.

anaerobic biodegradability of Hydrex 3818 at a concentration of 8,300 mg/L, increased up to an incubation time of 8 days before reaching a plateau of 0.17 g CH_4-COD/g COD substrate added (Figure 8) with an initial I:S of 1.8 g COD/g COD. Thus, at 17% g COD/g COD, the biodegradability of Hydrex 3818 was quite poor.

The aerobic biodegradation of a high tannin-containing wastewater (4,900 mg/L) from a leather industry was inefficient in a laboratory-scale test by He *et al.* (2007), who concluded that its biodegradability varied greatly with its tannin content and that a dilution pre-treatment was necessary to improve the biotreatability of this kind of wastewater. The anaerobic biodegradation of two condensed (Wattle and Quebracho) tannin extracts (1,000 mg/L of tannins) by upflow anaerobic sludge blanket (UASB) reactors presented by Lopez-Fiuza *et al.* (2003) gave 20%, a similar result to that obtained for Hydrex 3818 in this work. They also observed a significant increase in tannin biodegradability when the condensed tannin extract concentrations were lower.

The poor biodegradability of Hydrex 3818 observed in this work was similar to reported values for tannin extracts present in high tannin-containing wastewaters. Further Hydrex 3818 BMP assays should be performed at lower substrate concentration. Because the tannin concentration in the floating sludge from the HR-DAF process would be lower than that used in the BMP test in this work, an increase in the biodegradability of Hydrex 3818 could be expected. A comparison of floating sludge biodegradability both without chemicals and with tannin should be the subject of future work.

CONCLUSIONS

The high-rate DAF process downstream of an HR-MBBR gave a very good particulate matter recovery. Without chemicals, an HR-DAF TSS recovery of 94 ± 1% was obtained at SLRs up to 15 m/h and at an HR-MBBR OLRs of less than 50 g COD m^{-2} d^{-1}. A/S ratios of 0.03 to 0.05 g air/g SS were required to achieve this efficiency, corresponding to recycle ratios higher than 25% and an operating pressure of at least 6 bar. The use of tannin-based low-molecular weight cationic polymer at an optimal dose of 17–20 mg/L for pre-treatment increased the HR-DAF TSS recovery to 96 ± 1%. Its application at very low contact times made the HR-DAF less dependent on the HR-MBBR OLR by achieving stable efficiency for SLRs of at least up to 22 m/h while reducing its needs in air at 0.01 to 0.02 g air/g SS, corresponding to recycle ratios as low as 14% and an operating pressure of 4 bar. The biodegradability of tannin-based polymer Hydrex 3818 under anaerobic conditions was only 17%, at an initial inoculum to substrate ratio of 1.8 g COD/g COD and at an initial concentration of 8,300 mg/L of tannin. Thus, the HR-DAF is a high-rate wastewater treatment process that is a promising alternative to the A-B process for carbon and energy recovery via methanogenesis. Other BMP tests and a comparison of energy requirements and carbon recovery without chemicals and with the addition of tannin could be considered.

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