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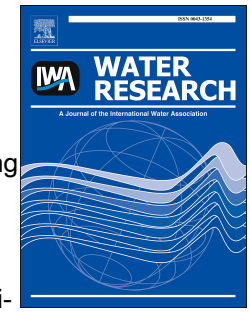

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PII: S0043-1354(16)30379-7

DOI: 10.1016/j.watres.2016.05.053

Reference: WR 12088

To appear in: *Water Research*

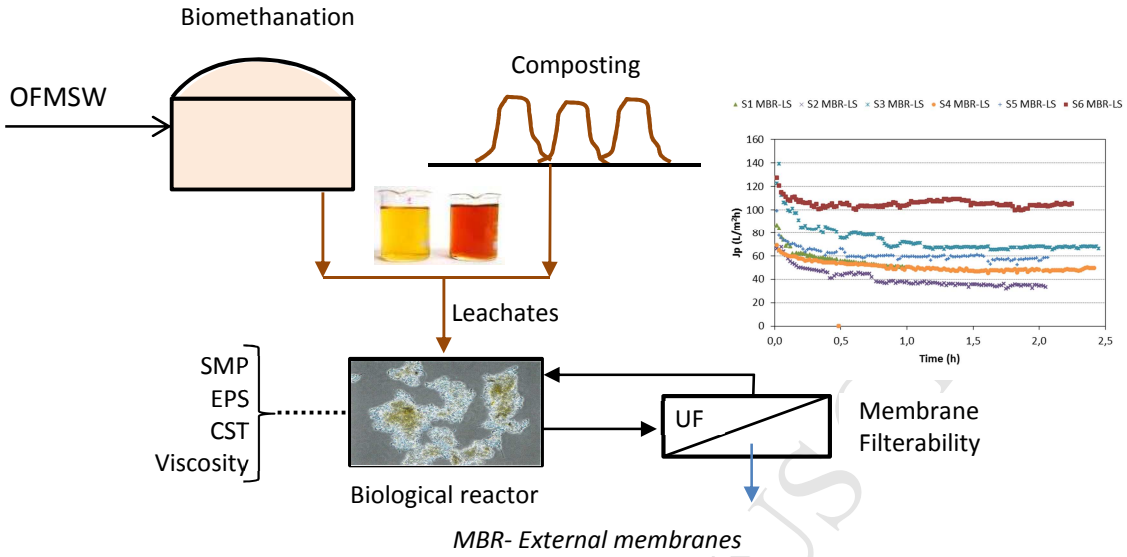
Received Date: 23 February 2016

Revised Date: 30 April 2016

Accepted Date: 16 May 2016

Please cite this article as: Zuriaga-Agustí, E., Mendoza-Roca, J.A., Bes-Piá, A., Alonso-Molina, J.L., Muñagorri-Mañueco, F., Ortiz-Villalobos, G., Fernández-Giménez, E., Comparison between mixed liquors of two side-stream membrane bioreactors treating wastewaters from waste management plants with high and low solids anaerobic digestion, *Water Research* (2016), doi: 10.1016/j.watres.2016.05.053.

Graphical abstract



COMPARISON BETWEEN MIXED LIQUORS OF TWO SIDE-STREAM MEMBRANE BIOREACTORS TREATING WASTEWATERS FROM WASTE MANAGEMENT PLANTS WITH HIGH AND LOW SOLIDS ANAEROBIC DIGESTION

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Abstract

In the last years, biological treatment plants for the previously separated organic fraction from municipal solid wastes (OFMSW) have gained importance. In these processes a liquid effluent (liquid fraction from the digestate and leachate from composting piles), which has to be treated previously to its discharge, is produced. In this paper, the characteristics of the mixed liquor from two full-scale membrane bioreactors treating the effluents of two OFMSW treatment plants have been evaluated in view to study their influence on membrane fouling in terms of filterability. For that, the mixed liquor samples have been ultrafiltrated in an UF laboratory plant. Besides, the effect of the influent characteristics to MBRs and the values of the chemical and physical parameters of the mixed liquors on the filterability have been studied. Results showed that the filterability of the mixed liquor was strongly influenced by the soluble microbial products in the mixed liquors and the influent characteristics to MBR. Permeate flux of MBR mixed liquor treating the most polluted wastewater was

considerable the lowest (around 20 $L/m^2 \cdot h$ for some samples), what was explained by viscosity and soluble microbial products concentration higher than those measured in other MBR mixed liquor.

***Keywords*:** MBR; leachate; digestate; municipal wastes; anaerobic digestion.

1. Introduction

Municipal waste treatment generates wastewaters that have to be treated before their disposal. In the case of sanitary landfills, the generated liquid effluent is named leachate. Leachates are very pollutants and they are characterized by very high organic matter and ammonium concentrations, among others. Other types of municipal waste treatment plants, which have gained importance in the last years, are the named biomethanation plants, which usually treat the previously separated organic matter fraction from the municipal solid wastes (OFMSW) (Cesaro et al., 2015). These plants consist of a pretreatment stage, an anaerobic digestion (AD) for methane production and a further composting of the solid fraction of the digestate (Tampio et al., 2015). As a result, two types of waste liquid effluents are produced, digestate liquor (liquid fraction from the anaerobically treated waste after being dehydrated) and leachate from the composting piles. Both effluents require treatment before being discharged to a municipal wastewater treatment plant.

Processes for leachate treatment have been summarized by some authors in review papers (Omar and Rohani, 2015; Pokhrel and Viraraghavan, 2004; Renou et al., 2008; Wiszniowski et al., 2006). It can be commented that all the types of wastewater processes (physical, chemical and biological processes) have been used for the leachates

management. Because of its high pollution load, high efficient processes or combination of different ones have to be applied.

Membrane bioreactor (MBR) is one of the processes that have gained importance in the leachate treatment. This process has been successfully applied to treat wastewaters with toxic compounds in low concentrations (Boonyaroj et al., 2012; Nghiem et al., 2009; Svojitka et al., 2009). Besides the treated water quality achieved, other advantage of MBRs in comparison with other technologies is the smaller footprint, since high biomass concentration can be maintained in the reactor (Judd, 2011).

Papers about MBR treating leachates have been mainly focused on the quality of the treated water (Ahmed and Lan, 2012; Lin et al., 2012). Ahmed and Lan (2012) also stated that the majority of the published papers related to landfill leachate treatment by MBRs are bench or pilot scale studies. Alvarez-Vazquez et al., 2004 carried out a comparison between the quality of the treated leachate with MBR and with other biological techniques. They concluded that MBRs usually offer high COD removal efficiencies for less biodegradable feeds at a much smaller footprint. Campagna et al. (2013) evaluated the size of the organic matter of a landfill leachate and the evolution of these fractions after a MBR and after a further nanofiltration stage. These fractions were related with their removal in the process (Campagna et al., 2013).

Table 1 summarizes the papers of several authors who reported results about the application of MBRs to landfill leachates. MBR size in terms of membrane surface, membrane configuration, leachate characteristics and mixed liquor characteristics have been included. As it can be observed, only little information is available about physic-chemical characteristics of the mixed liquor.

Table 1. Reported data in the literature about MBRs treating landfill leachate

The main operating problem for the application of MBRs to leachate treatment is membrane fouling. In general terms, this phenomenon is the main drawback of MBRs and fouling may be more severe in MBRs treating leachate due to its composition. This phenomenon could make the process unfeasible by increasing the transmembrane pressure (TMP) to achieve a sustainable flux. Membrane fouling mainly depends on membrane module design, wastewater composition, membrane characteristics, operation of the filtration process and operation of the biological process (Khongnakorn et al., 2007; Lyko et al., 2008; Meng et al., 2009). Once the MBR is working, only operating conditions can be modified; thereby the control of the mixed liquor characteristics will be of paramount importance to prevent or reduce the membrane fouling (Lin et al., 2014). According to literature, extracellular polymeric substances (EPS), both extracted from the cell wall of bacteria (eEPS) and soluble microbial products (SMP) are the main responsible for membrane fouling (Ding et al., 2015; Liu et al., 2012; Wang et al., 2009).

This is the reason why they should be analysed and controlled. However, unlike applications of MBR to municipal wastewater, only a few papers about mixed liquor characterization from industrial WWTPs are found. Only Sanguanpak et al., 2015 reported results about the influence of the leachate pH on EPS generation and consequently on membrane fouling.

Literature also lacks of papers dealing with the treatment of leachate from composting and digestate liquor by MBR technology. Although composition can be in rough outlines very similar to landfill leachates, specific works for effluents from anaerobic digestion (digestate liquor) plus aerobic digestion (leachate) OFMSW are required. Brown et al., 2013, detailed the elimination efficiencies of a great number of compounds in a MBR treating leachate from composting of the OFMSW.

In this paper, the operation of two full-scale MBRs with external membrane configuration (side-stream MBRs) treating effluents from two OFMSW plants is studied from the point of view of the mixed liquor characterization. Comparison is carried out in order to find out the differences in the MBR mixed liquors caused by the different type of process carried out in the OFMSW. Both plants consist of anaerobic digestion plus composting processes.

2. Materials and Methods

Full scale MBRs

Samples were obtained from two full-scale MBRs. MBRs treat the waste effluents from OFMSW plants, specifically from the AD and composting processes.

The difference between both plants is that the AD process in one plant is carried out by means of a high solids system (Dry-process, i.e. solids concentration higher than 15% (Li et al., 2011)) and the other one by means of a low solids system (Wet-process, i.e. solids concentration lower than 10%), Hereafter, MBRs will be referred to as MBR-HS and MBR-LS in order to distinguish them.

For both plants the MBR configuration is the same, i.e. membranes are external and mixed liquor is pumped from the biological reactor to the UF module (MBR recirculated). Membranes are multichannel tubular and the installed active surface is 127 m^2 and 72 m^2 in MBR-LS and MBR-HS, respectively. Biological reactor consists of one anoxic tank, two aerobic tanks and a final tank that can be operated aerobically or anoxically depending of the nitrogen removal efficiencies. Therefore, both plants were designed to eliminate both organic matter and nitrogen.

115 Sampling

116 Six samples, one per month, were taken from both MBRs. *Samples were maintained*
117 *refrigerated at 4ºC until they were processed (the day after the collection).* Sample
118 points were the influent to MBRs streams and the mixed liquor that was pumped to the
119 membranes from the last tank of the reactor.

120 Analyses of the influent

121 The following characterization parameters were measured: pH, conductivity, total
122 nitrogen (TN), soluble total nitrogen (sTN), ammonium nitrogen (NH_4^+-N), total COD
123 (tCOD), soluble COD (sCOD) and suspended solids (SS).

124 pH and conductivity were respectively measured with pH-Meter (GLP-21+) and EC-
125 Meter (GLP-31+) from CRISON (Spain). TN, sTN, NH_4^+-N, tCOD and sCOD were
126 determined spectrophotometrically by means of standard cell tests from Merck. Samples
127 had to be diluted so that no interferences were produced by salinity and colour.
128 Suspended solids were measured in duplicate according to APHA, 2005.

129 Ultrafiltration of the mixed liquor sample in laboratory

130 In order to compare the sludge filterability, 5 L of mixed liquor of each MBR were
131 ultrafiltrated in a laboratory plant equipped with a Rayflow 100 membrane module,
132 (Orelis, France) containing a flat-sheet membrane with an active surface of 100 cm^2.
133 Figure 1 represents the flowchart of the laboratory plant. The membrane used in every
134 test was a 150 kDa hydrophilic polyethersulfone membrane from Microdyn Nadir,
135 membrane characteristics are shown in Table 2. A new membrane was used for each
136 experiment, so that permeate flow were not influenced by the residual membrane
137 fouling from the earlier test.

Table 2: Membrane characteristics and filtration test conditions

Figure 1: Ultrafiltration laboratory plant scheme (150 kDa membrane)

Tests were carried out at the following operating conditions: 1 bar of TMP, 25ºC of temperature (T) and 2 m/s of crossflow velocity by means of a variable speed pump. The duration of each test was the necessary to reach the steady state (constant flux). The minimum duration considered was 2 hours. Experiments were carried out in total recycle mode of filtration, where both retentate and permeate streams were continuously recirculated into the feed tank. The permeate flux (J_p) was gravimetrically measured with an electronic weighing scale (KERN KB 2400-2N, 0.01 g accuracy, Germany) connected to a computer with a data acquisition software (Balance Connection SCD-4.0, Kern®). Data were recorded in the computer each minute. The permeate flux was monitored throughout the UF experiments according to Eq. (1), in order to determine the flux decline.

$$J_p = \frac{V_p}{A \cdot t} \qquad \text{(Eq. 1)}$$

Where Jp is the permeate flux ($L/m^2 \cdot h$), Vp is the permeate volume (L), A is the effective membrane area (m^2) and t is the sampling time (h).

Calculation of membrane filtration resistances

Total filtration resistance (R_T) has been calculated according to Eq. 2 (Bae and Tak, 2005). R_T (m^{-1}) can be expressed as the sum of the resistances caused by the membrane (R_m), the resistance that can be eliminated after rinsing (R_{rev}) and the remaining resistance (R_{irrev}).

$$R_t = \frac{TMP}{\mu \cdot J_p} \quad \text{(Eq. 2)}$$

Where TMP is the transmembrane pressure (Pa), μ is the viscosity (Pa·s) and J_p is the permeate flux by filtrating activated sludge at the steady state (L/·m^2·h).

Irreversible resistance (R_{irrev}) was calculated according to Eq. 3.

$$R_{irrev} = \frac{TMP}{\mu \cdot J_w} - R_m \quad \text{(Eq. 3)}$$

Where J_w is the membrane flux after water rinsing and R_m is the membrane resistance. Reversible resistance (R_{rev}) was calculated applying Eq. 4.

$$R_{rev} = R_t - R_{irrev} - R_m \quad \text{(Eq. 4)}$$

Mixed liquor characterization

The characterization of the mixed liquor was physical and chemical.

The physical parameters measured were mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS), capillary suction time (CST) and viscosity.

MLSS and MLVSS were analysed according to APHA (APHA, 2005). Capillary suction time (CST) was measured with the equipment Triton (304M model, United Kingdom). Due to the high MLSS concentration, samples were diluted with deionized water. Viscosity was measured with a rheometer from Haake RheoStress 1 (Thermo, Germany), equipped with concentric cylinder (Z34 DIN sensor) and operated at constant temperature (20ºC). Shear rate ($\dot{\gamma}$) was increased and decreased since 0 to 800 s^{-1}, in order to study eventual thixotropy.

The chemical characterization of the mixed liquor samples was performed by measuring both SMP and eEPS. SMP were determined after centrifuging at 12000 xg and filtering (0.45 microns). For eEPS extraction the cation exchange resin (CER) method (Frølund et al., 1996) was applied. In order to avoid interferences between the activated sludge and the analytical methods, sample dilutions were carried out until achieving 2gVSS/L (Ras et al., 2008). In both, SMP and eEPS, proteins (by BCA method) and carbohydrates (by Anthrone method) were analysed. Bovine serum albumin (BSA) (Sigma-Aldrich) and glucose (Panreac) were used as the protein and carbohydrate standards.

Respirometry test

Respirometry tests were carried out in a BM-Advance analyser from SURCIS (Spain). Respirometry is based on the oxygen consumption by the microorganisms from the activated sludge. The reactor vessel was filled with 1L of MBR activated sludge from MBR-LS or MBR-HS depending on the experiment to be held. The activated sludge was previously aerated during 24h to obtain endogenous conditions in the biomass. A dynamic experiment was performed by continuous stirring, aeration and recirculation between both sides of the vessel by means of a peristaltic pump, where the dissolved oxygen was continuously measured. Temperature was kept constant at 22ºC during the experiment through a Peltier cooler module. The heterotrophic biomass yield coefficient (Y_H) was calculated by Eq. 6.

$$Y_H = 1 - \frac{OC}{COD_{sodium\ acetate}} \quad \text{(Eq. 6)}$$

Where OC is the oxygen consumed by the microorganisms to biodegrade a substrate. To determine the OC a dynamic respirometry test was performed by adding a sodium acetate solution of 400 mg/L (COD = 300 mg/L). In order to determine the COD fractionation, different dynamic tests were carried out by adding in the vessel 15 mL of influent wastewater (for the total COD) and influent wastewater filtered by 0.45 µm (for the soluble fraction, rapidly biodegradable COD) to study their biodegradation.

Fluorescence in situ hybridization (FISH) and microscopic observation

Identification and estimation of filament abundance within the phylum Bacteroidetes were performed by applying SAP-309 probe (25% formamide) targeting the family Saprospiraceae (Schauer and Hahn, 2005). Samples were fixed in 4% paraformaldehyde at 4 ºC. The fixed biomass was washed three times with phosphate-buffered saline (PBS), and re-suspended in a 1:1 (v/v) volume of PBS and absolute ethanol and then stored at -20 ºC. The fixed samples were immobilized on gelatin-coated glass slides, air-dried, and consecutively dehydrated in 50%, 80% and absolute ethanol. Subsequently, 9 µl of hybridization solution was mixed with 1 µl (50 ng) of Tamra-labeled SAP-309 probe. Hybridization buffer and probe were applied to the slide and incubated at 46 ºC for 2 hours. After hybridization, the slides were incubated in the washing buffer for 15 min in a 48 ºC water bath (Rossetti et al., 2006). The slides were incubated with a 4´,6´-diamidino-2-phenylindole (DAPI) solution (final concentration 1 µg/ml) at 4 ºC for 15 min. Microscopic observation was performed using an epifluorescence microscope (Olympus BX50, Japan, equipped with a CCD camera (Olympus DP12, Japan).

3. Results

Influent wastewater and permeate characterization

Table 3 shows the characterization of the influent wastewaters to MBRs. Average values of the 6 samples and standard deviations have been included.

Table 3. Characterization of the influent wastewaters to MBRs (n = 6)

Differences between both wastewaters are considerable except for pH value. Thus, the influent to MBR-HS has a concentration of SS approximately three times higher than the MBR-LS, what is clearly due to the way of carrying out the anaerobic digestion. This also affects to the tCOD and TN values, in such a way that analysis showed almost the same relation between both wastewaters (tCOD and TN are 3.5 and 3.3 times higher in the influent to MBR-HS than in the influent to MBR-LS).

Concerning conductivity, it has to be highlighted that values were much higher in the influent to MBR-HS than in the influent to MBR-LS, though samples in MBR-HS showed less variable values.

In addition, sCOD, sTN and NH_4^+-N of influent to MBR-HS were also much higher than in the influent to MBR-LS. As expected, these values were in concordance with the tCOD and TN obtained for both plants.

In comparison with landfill leachates, the characteristics of MBR-LS are similar to those reported by authors whose works have been summarized in Table 1. However, SS concentrations in the MBR-LS influent were higher than in the landfill leachates. Concerning MBR-HS, concentrations of all measured parameters were higher than those reported for landfill leachates.

239 In general terms, it can be assumed that the high SS and COD concentrations in the
240 effluents from the OFMSW treatment plants require a biological process with high
241 biomass concentrations in the reactors and high hydraulic retention times, especially in
242 the case of the plant with high solids anaerobic digestion.

243 Ultrafiltration experiments

244 Membrane permeability was measured using deionized water as feed before each UF
245 test. The mean value of the membrane permeability measured was 500 L/($m^2 \cdot h$).
246 Membrane was discarded if the permeability value measured was 15% above or below
247 the mean value with the aim that all the tests were comparable.

248 In Table 4, filtration resistances are shown for each activated sludge sample from both
249 MBRs. As it can be observed, in samples from MBR-HS, the R_t was higher than in the
250 MBR-LS ones. This fact is in agreement with the results shown in figure 2.

251 **Table 4:** Membrane filtration resistances in the activated sludge from MBR-LS and
252 MBR-HS

253 It can also be observed that the highest contribution to the total resistance is caused by
254 the reversible membrane resistance in all the tests. That means that cake formation is the
255 main mechanism involved in membrane fouling. Comparing both MBRs, the difference
256 between the total resistances was due to the measured R_{rev} values.

257 Figures 2 shows the permeate fluxes of the ultrafiltration experiments for mixed liquor
258 samples from MBR-LS and MBR-HS, respectively.

259

Figure 2. UF laboratory tests: Evolution of permeate flux using mixed liquor samples from MBR-LS (Fig. 2a) and MBR-HS (Fig. 2b) as feed (TMP = 1 bar, T = 25ºC, v = 2 m/s)

It can be observed that samples from MBR-HS showed much lower filterability than samples from MBR-LS. In fact, in the case of MBR-HS only for S6 it was achieved a flux higher than 25 L/($m^2 \cdot h$), which is lower than the minimum flux obtained in the UF of samples from MBR-LS. The reason for this behaviour will be discussed in the next paragraphs once the physical and chemical characterizations of the mixed liquors are detailed.

Respirometric tests

Respirometric tests help determining the non-biodegradable organic fractions, which will remain in the bacterial flocs until they are taken out of the system in the sludge withdrawals. In the meanwhile, these fractions will collaborate to diminish the filterability of the mixed liquor contributing to the membrane fouling at the same time. In fact, though direct UF/MF of digestate liquor has been hardly studied, Camilleri-Rumbau et al., 2014 reported very low fluxes in the MF (0.2 microns of pore size) of digestate liquor from an anaerobic digester treating 50% of pig slurry, 15% of cattle manure, 10% chicken manure and 25% food waste. That is, fluxes were very similar to those obtained for the mixed liquor of MBR-HS. In general, it was observed that the more the SS in the influent is, the lower flux will be obtained in the UF of the mixed liquor. Table 5 shows the differences in the particulate non-biodegradable COD of both influents to MBRs.

The heterotrophic biomass yield coefficient (Y_H) calculated according to Eq. 6, was 0.674 and 0.71 for MBR-LS and MBR-HS, respectively. These values showed that the

284 microorganisms exhibit optimal growth when a rapidly biodegradable substrate is
285 added, in both activated sludges.

286 In the MBR-HS observed in Table 5, total and soluble COD values measured
287 spectrophotometrically were 70000 and 10400 mg/L, respectively. As the soluble
288 spectrophotometrically COD was very similar to the respirometric COD, it can be stated
289 that the great majority of soluble COD was rapidly biodegradable. However, the inert
290 particulate COD (difference between total and soluble inert COD) was very high. When
291 results are compared with those obtained for the MBR-LS, it can be observed that inert
292 particulate COD in MBR-HS was considerably higher than in MBR-LS. This can
293 explain the differences in the mixed liquors in terms of physical properties and
294 structure, and consequently the differences in filterability.

295 **Table 5:** COD fractionation obtained with the respirometric tests (S3)

296 Physical characterization and influence on mixed liquor filterability

297 *MLSS and MLVSS concentrations*

298 Table 6 shows the MLSS and MLVSS concentrations determined in the mixed liquor
299 samples for each MBR.

300 **Table 6.** Total and volatile suspended solids concentration in MBR-LS and MBR-HS
301 mixed liquors

302 As observed in Table 6, MLSS concentrations were variable in the period studied,
303 mainly in MBR-LS. Values ranged between 13.06 and 27.83 g/L for MBR-LS and
304 between 20.06 and 26.59 for MBR-HS. These values can be considered typical for a
305 recirculated MBR configuration, meanwhile a range between 8 and 18 g/L is typical for

306 submerged MBRs (Drews, 2010). The high concentrations are required to eliminate the
307 degradable COD.

308 Concerning the percentage of volatile solids, Table 6 shows that the initial values (S1,
309 S2 and S3) were the lowest ones. However, the MLVSS percentage increased when
310 MLSS diminished due to the sludge withdrawals. Thus, values between 74 and 80%
311 were reached.

312 On the other side, no relation between MLSS and sludge filterability was found. Thus,
313 the maximum fluxes were obtained with S6 both in MBR-LS (104 $L/m^2 \cdot h$) and also in
314 MBR-HS (24.6 $L/m^2 \cdot h$), corresponding to concentrations of MLSS of 23.62 and 20.83,
315 respectively. These lack of relation between filterability and MLSS have been reported
316 for submerged MBRs treating municipal wastewater (Lousada-Ferreira et al., 2015).

317 *Capillary suction time*

318 Capillary suction time (CST) was measured to evaluate the mixed liquor dewatering
319 capacity by filtration. Results showed that activated sludge from MBR-HS was less
320 dehydratable than the MBR-LS one, since CST values were very high (1629 s as
321 average value) in comparison with MBR-LS sludge (83.5 s). This difference can be
322 probably attributed to the considerable higher concentration of SMP_C in MBR-HS than
323 in MBR-LS. These positive correlations between CST and SMP_c have been reported by
324 (Reid et al., 2008; Sabia et al., 2013).

325 In addition,the mixed liquor with higher CST (MBR-HS) coincides with the mixed
326 liquor that more resistance to filtration (R_t) has (data collected in Table 4).

327

328 *Viscosity*

329 The activated sludge is a non-Newtonian fluid with a pseudo-plastic behaviour (Moreau
330 et al., 2009). Figure 3 shows the variation of the apparent viscosity (η) and the shear
331 stress (τ) with the shear rate ($\dot{\gamma}$) for the sample 2 from both MBRs.

332 The evolution of these rheological parameters was modelled using the Ostwald de
333 Waele model (solid line), where τ (shear stress) can be expressed as a function of $\dot{\gamma}$,
334 (shear rate) $\tau = K \cdot \dot{\gamma}^{n}$ and the apparent viscosity as $\eta = K \cdot \dot{\gamma}^{n-1}$, where the
335 parameters K and n are the consistency index and flow behaviour index, respectively.
336 For MBR-LS sample, the adjustment has been performed from 0 to 540 s^{-1}, since from
337 this shear rate on, the excessive turbulence was generatedd and Taylor vortices appear
338 (Ratkovich et al., 2013).

339 **Figure 3:** Comparison between apparent viscosities of the mixed liquors (S2) from both
340 MBRs

341 It can be observed that the apparent viscosity in the mixed liquor from MBR-HS was
342 considerable higher than the MBR-LS one, which could explain its lower filterability.
343 This behaviour was very similar in all the analysed samples (S1-S6). However, slightly
344 differences were observed with the MLSS.

345 *Chemical characterization and influence on mixed liquor filterability*

346 Figure4 show the protein and carbohydrate concentrations in SMP from both MBRs
347 mixed liquors.

348 **Figure 4.** Protein (Fig. 4a) and carbohydrate (Fig. 4b) concentration in SMP from both
349 MBRs mixed liquors

350 It can be observed clearly that SMP (both proteins and carbohydrates) concentration
351 was higher in MBR-HS than in MBR-LS. This fact can be mainly due to the higher
352 stress of the biomass in MBR-HS, caused by accumulation of non-biodegradable solids
353 (Hao et al., 2010) and high salinity (40.13 ± 4.65 mS/cm) (Jang et al., 2013) that can
354 lead to bacteria stress.

355 These different values of SMP were considerably amplified from S2, what coincides
356 with a pronounced increment of the VSS percentage in MBR-HS. Thus, S1 showed the
357 lowest differences in SMPs between mixed liquors. In that sample, biomass in MBR-HS
358 was considerably mineralized (only 57.92% of MLVSS). It can probably explain the
359 lower SMP concentration by cryptic growth phenomena. It means that bacteria, in
360 absence of degradable food or stress conditions, are able to use the residual cellular
361 material as food, i.e. SMPp and SMPc. Once organic load is increased by sludge
362 withdrawal, bacteria have more food available, the percentage of volatile solids
363 increases and bacteria do not degrade the SMP.

364 The higher concentration of SMP in MBR-HS in comparison with MBR-LS and the
365 influent wastewater composition are the causes that would explain the poor filterability
366 of the mixed liquor samples from MBR-HS. In fact, the high SMP concentrations in
367 MBR-HS samples do not allow appreciating differences among the fluxes obtained in
368 the filterability tests. Nevertheless, it seems that there is a relationship between SMP_C
369 concentrations and UF fluxes represented in Figure 2 in MBR-LS samples. In fact,
370 samples with the lowest SMPc concentrations (3 and 6) corresponded with the samples
371 with the highest flux values in the filterability tests.

372 If these SMP concentrations are compared with those determined by other authors for
373 MBRs treating landfill leachates, it can be mentioned that Sanguanpak et al., (2015)

374 reported concentrations lower than the SMP obtained in MBR-LS. In fact, the SMPp
375 concentrations measured in MBR-LS are around twice higher than those reported by
376 these authors. For SMPc the relationship was very similar.

377 From Figure 4, it has to be highlighted that the concentration difference between
378 proteins and carbohydrates is very high. Sabia et al., 2013 reported that ratio
379 SMPp/SMPc sharply increased with the sludge retention time (SRT). In fact, SMPc
380 were higher than SMPp for low SRT, whereas at high SRT the ratio SMPp/SMPc
381 reached values between 5 and 10. These results are in agreement with those obtained in
382 MBR-LS and MBR-HS, since both hydraulic and sludge retention times are high in
383 order to achieve the required COD removal efficiencies.

384 The mechanism that may cause this behaviour could be associated to the appearance of
385 microorganisms that degrade in a higher extent carbohydrates coming from cellular
386 debris.

387 As shown in Figure 5, the abundance of filamentous bacteria belonging to the
388 Bacteroidetes phylum was very high. Among them, Haliscomenobacter filaments were
389 observed as predominant ones.

390 **Figure 5.** (Fig. 5a) Filaments with a needle-like appearance similar to
391 *Haliscomenobacter* DAPI staining. (Fig. 5b) *Haliscomenobacter* filaments identified
392 with the probe SAP-309 using FISH technique (S3, MBR-HS)

393 *Haliscomenobacter* filaments are specialized bacteria involved in degradation of sugars,
394 e.g. glucose and *N*-acetylglucosamine, and may participate in the conversion of
395 lipopolysaccharides and peptidoglycan liberated by decaying cells (Kragelund et al.,

2008). Therefore, these bacteria are able to degrade carbohydrates, increasing the relationship between proteins and carbohydrates concentration.

Figure 6 shows the protein (P) and carbohydrate (C) concentrations in eEPS from both MBRs mixed liquors.

Figure 6. Protein and carbohydrate concentration in eEPS from both MBRs mixed liquors

Unlike SMPs, differences between eEPS from both MBRs mixed liquors were not found; thereby we can state that this parameter was not responsible for the different behaviour of the mixed liquors in the UF tests. Compared to the concentrations reported by Sanguanpak et al. (2015), the measured eEPS concentrations were lower. It is probably due to the low organic matter concentration available for the microorganisms in MBR-LS and MBR-HS. They assimilate rapidly the degradable organic matter adsorbed on the bacterial flocs.

Summarizing, the mixed liquors of MBR-LS and MBR-HS are characterized by high amounts of cellular debris that are responsible for the high SMP concentrations (especially in MBR-HS). This has no influence on the eEPS concentration, which is low and very similar in both MBRs due to the low organic loads and the lack of organic matter available for the microorganisms.

4. Conclusions

In this study, the mixed liquor characteristics of two full-scale MBRs treating effluents from OFMSW management plants have been compared. It can be concluded that the plant that uses high solids anaerobic digestion generates effluents with higher SS and

conductivity than the plant with low solids anaerobic digestion. Thus, influent characteristics were the most important factor influencing the mixed liquor filterability. The low filterability of the MBR-HS mixed liquor is explained mainly by the high viscosity, considerable higher than that measured for the mixed liquor of MBR-LS. Besides, biomass of MBR-HS is subjected to more stress than biomass of MBR-LS due to high non-biodegradable suspended solids concentration and salinity; whereby SMPs concentrations were higher than in MBR-LS. No differences between extracted EPS were detected.

As a general conclusion, and on the basis of the results obtained, a lower design flux should be considered for this type of plants, since fouling problems occur mainly due to the influent MBR characteristics and SMPs generated by bacteria of the mixed liquor.

Acknowledgements

This work was supported by the URBASER Company. Authors thank personnel of the full-scale MBR plants for providing samples.

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Table 1. Reported data in the literature about MBRs treating landfill leachate

Author	Scale (Membrane surface)	MBR configuration	Leachates characteristics	Physico-chemical characteristics of mixed liquor
Boonyaroj et al. (2012)	Pilot	Not detailed	pH = 7.4, COD = 9306 mg/L, NH_3-N = 138 mg/L, SS = 1240 mg/L, conductivity = 23.5 mS/cm (average values)	MLSS = 10000 – 12000 mg/L, sludge volume index = 30 – 60 mL/g
Campagna et al. (2013)	Full scale treating 2000 m^3/d	External (tubular)	COD = 16360 mg/L, NH_4^+-N = 2532 mg/L, conductivity = 33.9 mS/cm (average values)	Data not shown
Canziani et al. (2006)	Pilot (0.24 m^2)	External (ceramic membranes)	COD = 6316 mg/L, NH_4^+-N = 1497 mg/L (average values)	MLSS = 5000 – 8000 mg/L, Y = 0.67 gSS/gCOD
Hasar et al. (2009)	Lab (0.0390 m^2)	Submerged (hollow fiber)	pH = 6.45 – 6.50, sCOD = 8500 – 14200 mg/L, NH_4^+-N = 1100 – 2150 mg/L Mixed with domestic wastewater before feeding to MBR	MLSS = 4000-10000 mg/L
Hashisho et al. (2016)	Lab	Submerged (comparison between hollow fiber and flat sheet membranes)	pH = 8.43, COD = 5978 mg/L, NH_4^+-N = mg/L = 2464 , TN = 2543 mg/L (average values)	Data not shown
Litas et al. (2012)	Pilot	Submerged (flat sheet)	pH = 8.7, COD = 2544 mg/L, NH_4^+-N = mg/L = 269 , TN = 388 mg/L (average values). Diluted 1:1 with municipal wastewater	MLSS increasing between 2000 and 25000 mg/L, % of VSS = 84 – 70,

Rizkallah et al. (2013)	Pilot (0.929 m^2)	Submerged (hollow fiber)	pH = 7.26 – 7.91, COD = 9000 - 20000 mg/L, NH_3-N = 1800-4000 mg/L; TN = 2000 - 6000 mg/L, SS = 625 - 938 mg/L, VSS = 300 – 500 mg/L, conductivity = 38.2 – 50.4 mS/cm	Up to 7000 mg/L of MLVSS
Sanguanpak et al. (2015)	Lab (0.07 m^2)	Submerged (hollow fiber)	COD = 5445 mg/L (average value)	Zeta potential between -13 and -21 mV, SMP_p =98.8 – 132.2 mg/L, SMPc =24.3 – 44.6 mg/L, $eEPS_p$ =50.4 – 68.3 mg/gSS, $eEPS_c$ =18.4 – 29.4 mg/gSS, unsettled SS 18-80 mg/L, mean floc size = 54-58 microns [1]
Svojitka et al. (2009)	Lab (inflow rate (0.1 m^2)	External (tubular)	pH = 8.5, COD = 2200 mg/L, NH_4^+-N = 1200 mg/L; TN = 1258 mg/L	MLSS = 7100 – 11800 mg/L

[1] Ranges of average values obtained at tests varying pH

Table 2: Membrane characteristics and filtration test conditions

Active surface area (cm^2)	100 cm^2
Operating pressure (bar)	0-1 bar
Crossflow velocity (m/s)	2 m/s
Feed flow rate (L/h)	300 L/h
Membrane material	Polyethersulfone hydrophilic (PES)
Membrane pore size	0.04 µm
MWCO	150 kDa

Table 3. Characterization of the influent wastewaters to MBRs (n = 6)

	MBR-LS	**MBR-HS**
pH	7.99 ± 0.10	7.99 ± 0.10
Conductivity (mS/cm)	16.26 ± 6.97	40.13 ± 4.65
SS (mg/L)	4401 ± 1812	12940 ± 1301
tCOD (mg/L)	9430 ± 5944	32910 ± 7106
sCOD (mg/L)	3140 ± 1262	20927 ± 7635
TN (mg/L)	1879 ± 990	6267 ± 2666
sTN (mg/L)	790 ± 320	4890 ± 406
NH_4-N (mg/L)	767 ± 324	3990 ± 410

Table 4: Membrane filtration resistances in the activated sludge from MBR-LS and MBR-HS

MBR-LS	S1	S2	S3	S4	S5	S6
R_t (m^{-1})	8.01E+12	1.15E+13	6.50E+12	8.77E+12	6.93E+12	4.04E+12
R_{irrev} (m^{-1})	8.62E+11	5.23E+11	7.94E+11	2.19E+12	1.66E+12	4.12E+11
R_{rev} (m^{-1})	6.35E+12	9.96E+12	4.80E+12	5.79E+12	4.75E+12	2.90E+12
MBR-HS	S1	S2	S3	S4	S5	S6
R_t (m^{-1})	1.97E+13	1.99E+13	1.73E+13	2.16E+13	1.97E+13	1.64E+13
R_{irrev} (m^{-1})	9.86E+11	8.20E+11	1.36E+12	9.72E+11	6.31E+11	1.93E+11
R_{rev} (m^{-1})	1.80E+13	1.84E+13	1.51E+13	2.01E+13	1.82E+13	1.53E+13

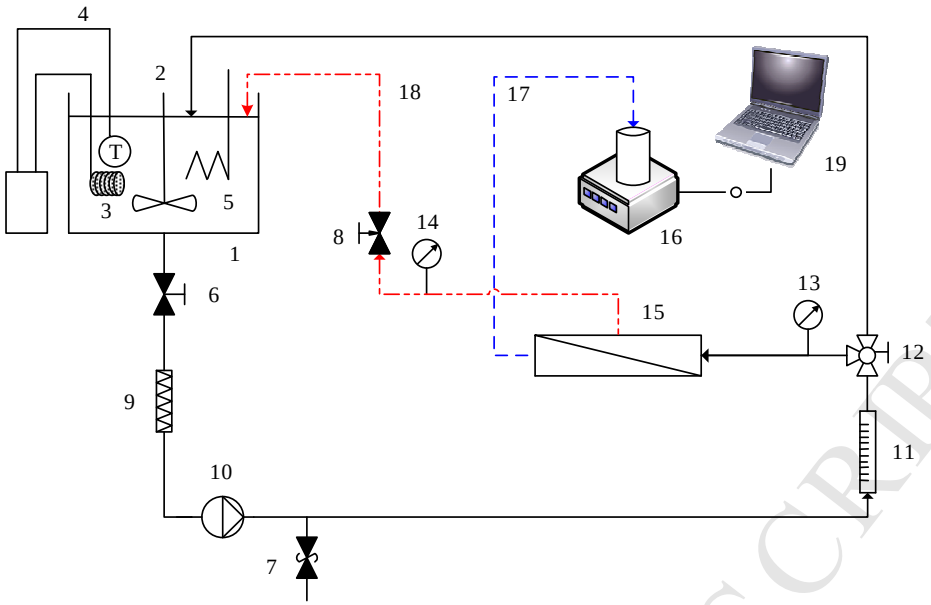
Table 5: COD fractionation obtained with the respirometric tests (S3)

	MBR-LS (mg/L)	**MBR-HS (mg/L)**
spCOD[1]		
Total	6580	70000
Soluble	2930	10400
rCOD[2]		
Total	4854	64088
Soluble	1931	10328
Inert calculated COD		
Total	1526	5912
Soluble	999	72

[1]spCOD: spectrofotometrically measured COD
[2]rCOD: respirometric measured COD

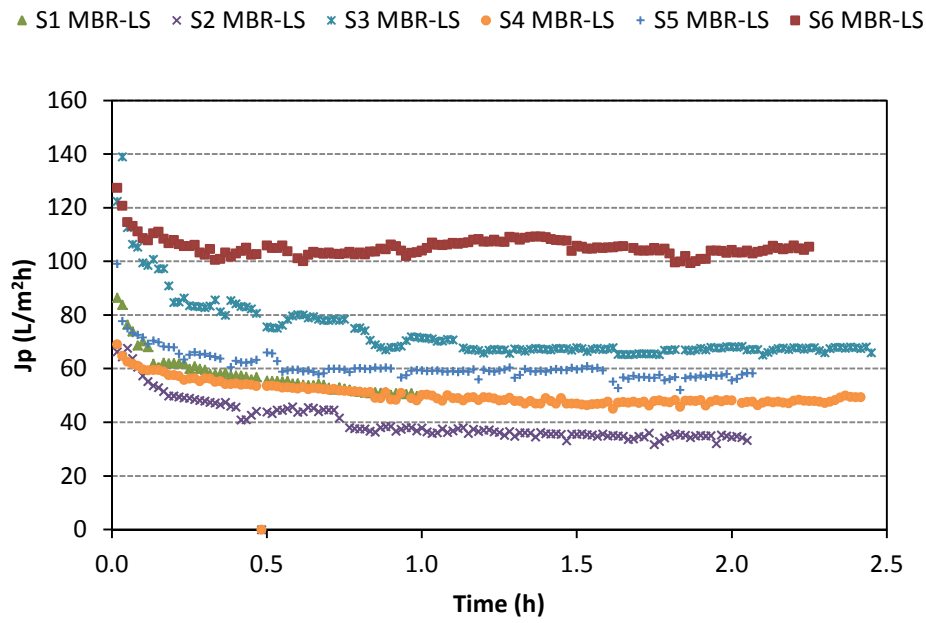
Table 6: Total and volatile suspended solids concentration in MBR-LS and MBR-HS mixed liquors

	MBR-LS			*MBR-HS*		
	MLSS (g/L)	*MLVSS (g/L)*	*%VSS*	*MLSS (g/L)*	*MLVSS (g/L)*	*%VSS*
S1	27.83	18.81	67.57	25.23	14.63	57.92
S2	13.06	10.07	77.15	26.59	16.79	63.63
S3	17.17	13.13	76.45	20.06	14.63	72.97
S4	19.38	15.10	77.90	21.67	16.69	77.02
S5	22.53	18.05	80.73	24.14	18.61	77.09
S6	23.62	18.68	79.14	20.83	15.50	74.42



1	Feed tank	9	Filter	16	Weighing scale
2	Stirrer	10	Pump	17	Permeate Stream
3	Immersion cooler	11	Flow meter	18	Retentate Stream
4	Thermometer	12	Three-way valve	19	Data acquisition
5	Heater	13,14	Manometer		
6-8	Valve	15	Membrane module		

Figure 1: Ultrafiltration laboratory plant scheme (150 kDa membrane)



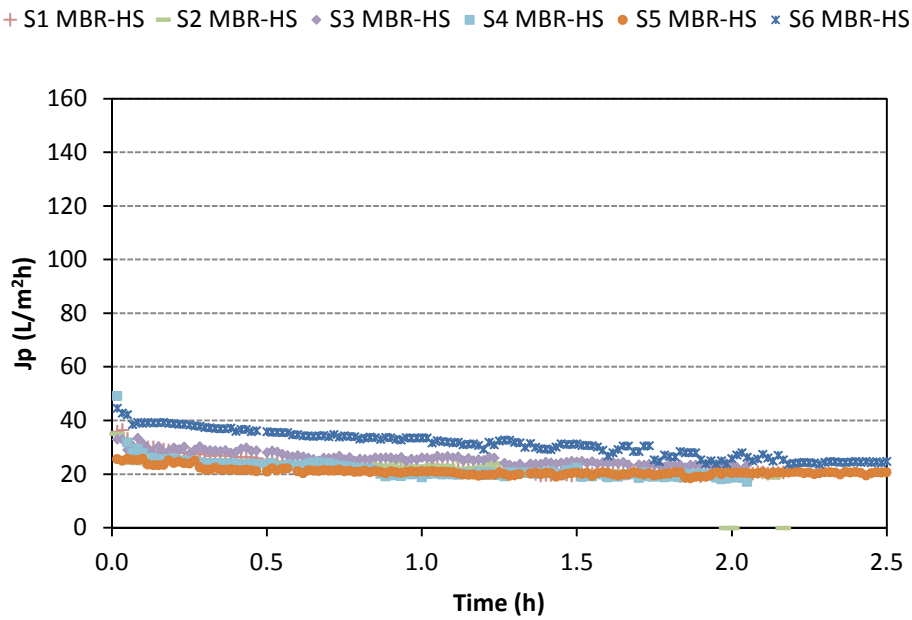


Figure 2. UF laboratory tests: Evolution of permeate flux using mixed liquor samples from MBR-LS (Fig. 2a) and MBR-HS (Fig. 2b) as feed (TMP = 1 bar, T = 25ºC, v = 2 m/s)

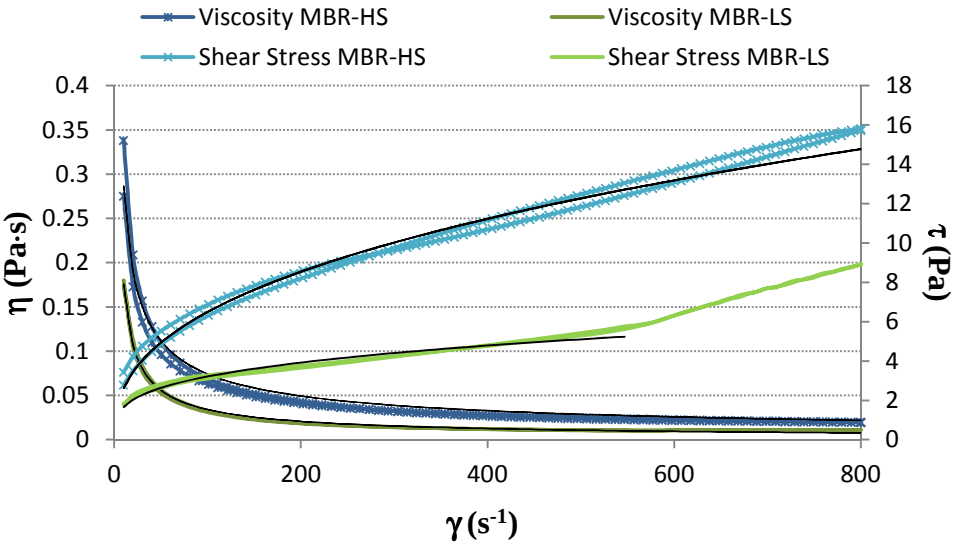


Figure 3. Comparison between apparent viscosities of the mixed liquors (S2) from both MBRs

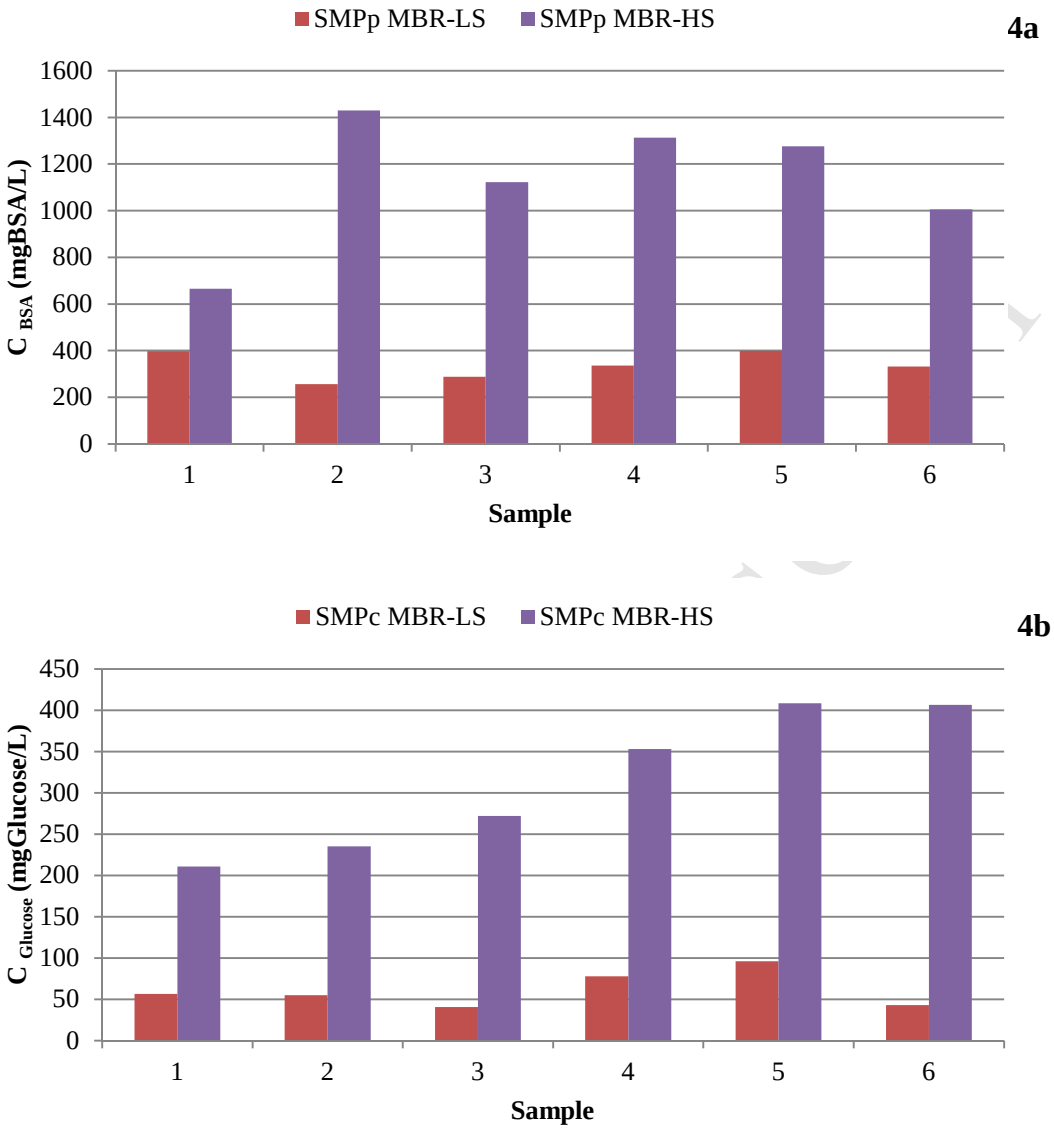


Figure 4. Protein (Fig. 4a) and carbohydrate (Fig. 4b) concentration in SMP from both MBRs mixed liquors

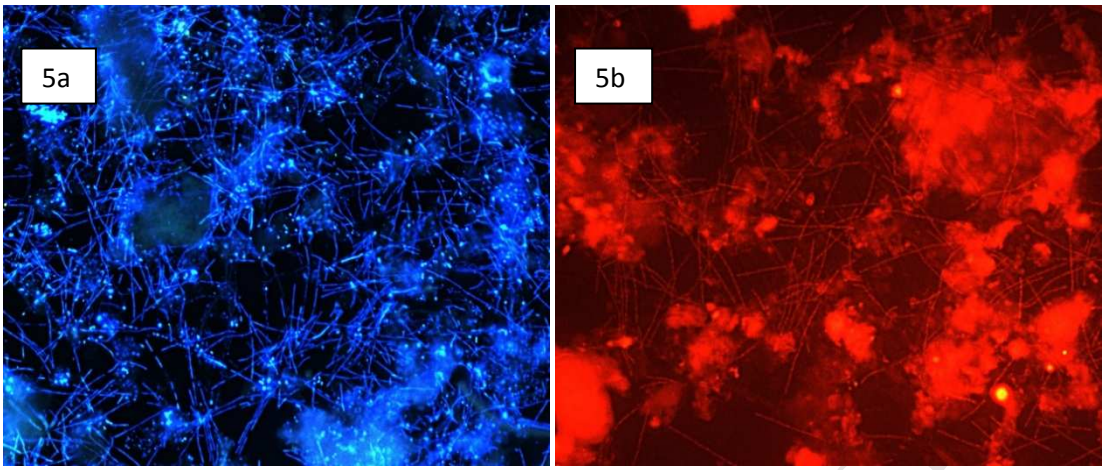


Figure 5. (Fig.5a) Filaments with a needle-like appearance similar to Haliscomenobacter DAPI staining. (Fig. 5b) Haliscomenobacter filaments identified with the probe SAP-309 using FISH technique (S3, MBR-HS)

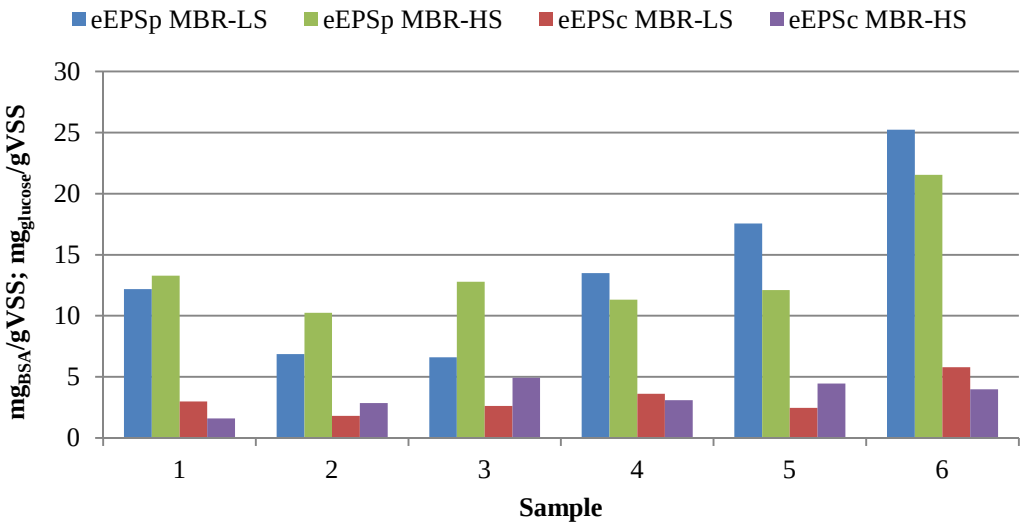


Figure 6. Protein and carbohydrate concentration in eEPS from both MBRs mixed liquors

HIGHLIGHTS

- Fouling in full-scale MBRs treating leachates of methane production plants is studied.
- Differences are found depending on methanation process (dry or wet).
- Membrane fouling is particularly severe for the dry process.
- High viscosity of the mixed liquor in dry process increases resistance to filtration.
- Differences in membrane fouling were not caused by extracted EPS.