

Characterization of secondary treated effluents for tertiary membrane filtration and water recycling

C. Ayache, M. Pidou, W. Gernjak, Y. Poussade, J.-P. Croué, A. Tazi-Pain and J. Keller

ABSTRACT

This study evaluates the impacts of water quality from three different secondary effluents on low pressure membrane fouling. Effluent organic matter (EfOM) has been reported by previous studies as responsible for membrane fouling. However, the contribution of the different components of EfOM to membrane fouling is still not well understood. In order to improve and optimize treatment processes, characterization and quantification of the organic matter are important. The characterization methods used in this study are liquid chromatography coupled with an organic detector (LC-OCD) and excitation emission matrix fluorescence spectroscopy (EEM). A bench-scale hollow fibre membrane system was used to identify the type of fouling depending on the feed water quality. Results showed no measurable dissolved organic carbon removal by the membranes for the three secondary effluents. Biopolymers and humic-like substances found in different proportions in the three effluents were partially retained by the membranes and were identified to contribute significantly to the flux decline of the low pressure membranes. The observed fouling was determined to be reversible by hydraulic backwashing for two effluents and only by chemical cleaning for the third effluent.

Key words | excitation emission matrix fluorescence spectroscopy, liquid chromatography organic carbon detector, membrane fouling, ultra-filtration, water recycling

C. Ayache (corresponding author)

M. Pidou

W. Gernjak

J. Keller

The University of Queensland,
Advanced Water Management Centre (AWMC),
St Lucia, Brisbane, QLD 4072,
Australia
E-mail: c.ayache@awmc.uq.edu.au

C. Ayache

A. Tazi-Pain

Veolia Environnement Recherche et Innovation,
Chemin de la Digue,
78603 Maisons Laffitte,
France

C. Ayache

J.-P. Croué

LCEE ENSIP, Université de Poitiers,
40 avenue du Recteur Pineau,
86022 Poitiers,
France

Y. Poussade

Veolia Water Australia,
PO Box 10819, Adelaide St Post Office,
Brisbane, QLD 4000,
Australia

J.-P. Croué

WDRC,
King Abdullah University of Science and
Technology,
Thuwal,
Kingdom of Saudi Arabia

INTRODUCTION

Municipal wastewater is a resource from which high-quality water can be produced and recycled after tertiary treatment by micro-filtration (MF) or ultra-filtration (UF) followed by reverse osmosis (RO) filtration. Therefore, water reuse becomes an additional source for producing water suitable for applications in industry and agriculture as well as for aquifer recharge or other indirect drinking water supply (Lee *et al.* 2006). Despite these water recycling systems being successful in producing excellent water quality, there is still much that remains to be learned in order to find the optimum integration of biological wastewater treatment with MF/UF and RO membranes design and operation. Membrane fouling is

an operational challenge in plants utilizing membrane technologies. Its occurrence can result in water production loss, integrity loss and poorer water quality, higher energy and chemical costs and shortened membrane life (Wilf 2010). Better knowledge of the effects of effluent quality on membrane fouling mechanisms is required in order to improve fouling prevention and management techniques. As effluent organic matter (EfOM) plays a major role in organic and colloidal fouling of low pressure membranes (Shon *et al.* 2006), its characterization and quantification is important. Several studies have already attempted to characterize the composition of EfOM (Jarusutthirak *et al.* 2002; Zheng *et al.* 2009).

EfOM is described as a combination of natural organic matter and soluble microbial products. Organic matter consists of a range of different compounds present in all secondary effluents, from large aliphatic to highly coloured aromatics. Some of this organic matter, consisting of a wide variety of chemical compositions and molecular sizes, is negatively charged (Świetlik *et al.* 2004). Also, organic matter present in waters consists of both hydrophilic and hydrophobic components. Soluble microbial products consist mainly of colloids such as proteins, polysaccharides and humic substances of microbial origin derived from biological wastewater treatment (Drewes & Croué 2002). This EfOM is still poorly understood in terms of its implications for membrane fouling. In addition, the amount and characteristics of the organic matter depends on climate (temperature, precipitation), environment and urbanization, as well as on the treatment process applied (Delpla *et al.* 2009). A change of water quality presents major challenges to the treatment process. Therefore, it is essential to study the role of EfOM in membrane fouling to achieve understanding of EfOM properties in water reuse. The present study focused on the impact of water quality on membrane fouling in reuse of treated municipal wastewater. The aim of this work was to perform advanced characterization of three effluents generated by wastewater treatment plants with very different designs and operating conditions in order to determine which organic fractions could specifically affect membrane fouling. Methods used in the characterization included standard methods such as the analysis of dissolved organic carbon (DOC), nitrogen and phosphate concentrations as well as advanced tools such as liquid chromatography coupled with an organic detector (LC-OCD) and excitation emission matrix fluorescence spectroscopy (EEM). A bench-scale hollow fibre membrane system was used to assess the fouling rate depending on the feed water characteristics.

METHODS

Feed water

The experiments were conducted with wastewater effluents collected at three different wastewater treatment plants primarily receiving municipal wastewater. The first was an

activated sludge plant (sludge age of 18–20 days) with enhanced biological phosphorus removal (EBPR). The second was a sequential batch reactor with a sludge age of 18 days (SBR). The third was an activated sludge plant performing short sludge age process (10 days) with rudimentary coagulation by dosing with ferric chloride (SSA). The effluents were stored at 4 °C for a maximum of 48 h before analysis. All fouling experiments were conducted at room temperature (23 °C).

Bench-scale filtration unit

UF experiments were performed at bench-scale using tailor-made polyvinylidene membrane mini modules (eight hollow fibres; nominal pore size = 0.04 µm) and a flexible vacuum pressure filtration system (Figure 1) developed at the Johns Hopkins University (USA) (Huang *et al.* 2007). This bench-scale filtration unit was operated under constant flux around 50–55 L/m²/h¹ (corrected at 20 °C) with the constant flow provided by a dual channel peristaltic pump allowing two membrane modules to be tested simultaneously. Another dual channel peristaltic pump was used to feed the column with the effluent.

Filtration tests were performed on 1 L secondary effluent samples in batch mode. Transmembrane pressure was recorded by a pressure sensor to measure the rate of membrane fouling during the filtration of various feed solutions. After membrane conditioning (20 mg/L HOCl –

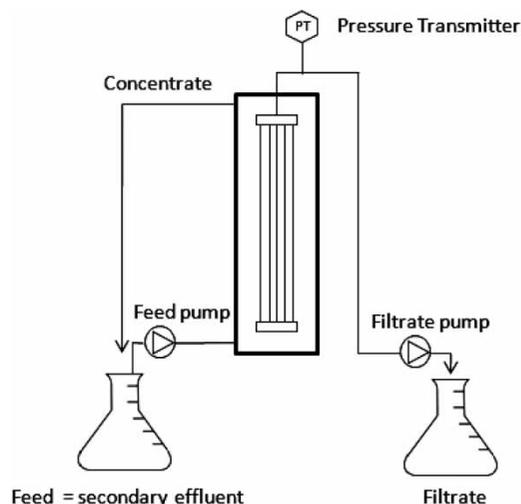


Figure 1 | Bench-scale filtration unit (as proposed by Huang *et al.* (2007)).

10 min of filtration followed by 30 min soaking and then rinsed by filtering MilliQ (MQ) water until transmembrane pressure stabilized) and prior to the filtration test, membrane specific permeability (L_{p0}) of each module was determined by filtering MQ water for a period of 30 min at the same permeate flux as used during the fouling experiment.

Several tests were used to evaluate the performance (permeability, selectivity, fouling potential) and efficiency (simple and complex organic compounds rejection, permeate quality) of the filtration process for the three selected secondary effluents. Continuous fouling tests and multi-cycle fouling tests were carried out. Continuous fouling tests consisted of a single filtration period of at least 8 h. Multi-cycle fouling tests consisted of 15 min filtration interspersed by 1 min of backwash. Backwashes were performed with MQ water in order to facilitate the analysis of the collected backwash samples.

At the end of each filtration test, chemical cleaning (200 mg/L HOCl – 30 min contact time) was performed to restore initial clean membrane conditions. Flux recovery was measured by conducting another MQ water filtration test to assess the efficiency of chemical cleaning.

Characterization methods

Feed, filtrate and backwash samples from the bench-scale filtration trials were characterized. Samples for standard methods and LC-OCD filtration were pre-filtered through a 0.45 μm filter (PM Separations).

Ammonia ($\text{NH}_4^+ + \text{NH}_3$), nitrate (NO_3^-) and phosphate (PO_4^{3-}) were analysed using a Lachat QuickChem8000 Flow Injection Analyser (Lachat Instrument, Milwaukee). DOC, total nitrogen and total phosphorus were analysed according to standard methods (APHA 1995).

The organic matter was characterized by an LC-OCD system (DOC-LABOR Dr Huber) equipped with a size exclusion chromatography column HW-50S filled with Toyopearl resin (pore size of 125 Å). Three detectors were installed in series in the sequence: UV detector (UVD), organic carbon detector (OCD) and organic nitrogen detector (OND). First, the UVD measured the spectral absorption coefficient at 254 nm. Second, the OCD oxidized all organic matter in a thin film UV reactor and the organic carbon

present in the sample could therefore be quantified from the CO_2 produced. Finally, the OND completely oxidized organic nitrogen (N_{org}) into nitrate and measured the amount of nitrate using a UVD at 220 nm.

Three-dimensional EEM data were collected using a luminescence spectrometer (PerkinElmer LS55, USA). In these studies, the EEM spectra were collected with corresponding scanning emission spectra from 280 nm to 500 nm at 5 nm increments by varying the excitation wavelength from 200 nm to 400 nm at 5 nm sampling intervals. The excitation and emission slits were maintained at 7 nm and the scanning speed was set at 1200 nm/min. A 290 nm emission cut-off filter was used in scanning to eliminate second order Rayleigh light scattering. The spectrum of MQ water was recorded as the blank enabling calculation of the area of the Raman peak and thus normalization to Raman units (RU) (Lawaetz & Stedmon 2009; Murphy *et al.* 2010). All samples were diluted with a predetermined dilution factor to avoid inner filter effects and assure linearity of the obtained results. EEM fluorescence was qualitatively attributed to compound classes as described by Chen *et al.* (2003).

RESULTS AND DISCUSSION

Effluent characterization

Physical-chemical characteristics of the feed water are presented in Table 1.

The median DOC content across the source types ranged from 5 to 10 mg/L for the EBPR and SBR effluent, respectively. As shown in Figure 2, DOC levels observed over the characterization period appeared less constant for the SBR with 50% of values in the second and third quartile ranging from 9 to 11 mg/L, compared to 4.5 to 5.5 mg/L and 7.5 to 8.2 mg/L for EBPR and SSA effluents. However, the variation of the data was closer to the minimum values, with a median at 10 mg/L, suggesting that the 18 mg/L maximum value was an anomaly in one sample, either due to a temporary problem in the operation of the wastewater treatment plant or an increase in the pollutant load.

The median nitrate values across the different secondary effluents evaluated ranged from 0.5 up to 5.5 mg/L in the

Table 1 | Physical-chemical characteristics of the feed water

	EBPR (<i>n</i> = 13)	SBR (<i>n</i> = 14)	SSA (<i>n</i> = 6)
DOC (mg/L)	4.9 ± 0.5	9.9 ± 2.4	7.7 ± 0.9
N_NH ₄ ⁺ (mg/L)	0.7 ± 0.8	0.1 ± 1.1	2.7 ± 2.6
N_NO ₃ ⁻ (mg/L)	0.6 ± 1.6	5.6 ± 1.3	3.8 ± 1.7
P_PO ₄ ³⁻ (mg/L)	2.4 ± 1.3	1.0 ± 0.8	2.1 ± 0.4
TKN (mg/L)	1.4 ± 1.0	1.6 ± 1.0	3.4 ± 1.8
TP (mg/L)	2.2 ± 1.2	0.8 ± 0.8	2.0 ± 0.6
Cl (mg/L)	281 ± 45	125 ± 12	281 ± 9
Ca (mg/L)	26.4 ± 2.6	22.2 ± 2.4	32.9 ± 7.5
K (mg/L)	15.8 ± 1.9	20.5 ± 2.0	21.4 ± 2.3
Mg (mg/L)	16.1 ± 1.7	9.4 ± 3.0	42.4 ± 11.9
Na (mg/L)	177 ± 27	124 ± 14	188 ± 4
Si (mg/L)	5.2 ± 0.4	7.2 ± 0.6	6.2 ± 0.3
Conductivity (µS/cm)	1500 ± 176	1458 ± 480	-
pH	7.0 ± 0.8	6.9 ± 0.4	-

TKN: total Kjeldahl nitrogen; TP: total phosphorus.

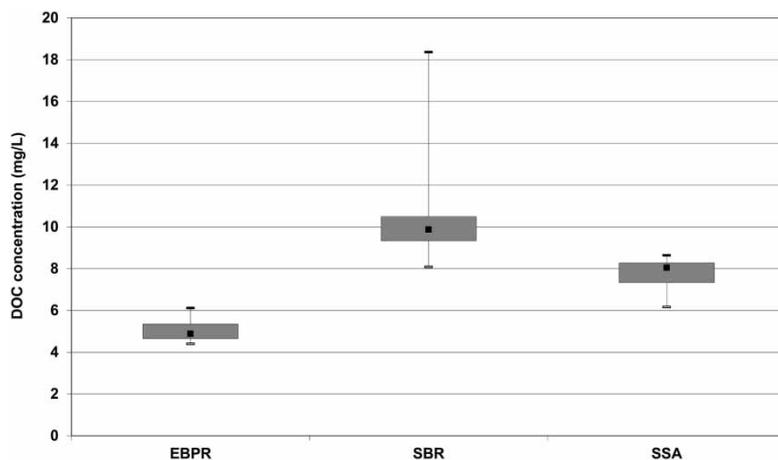


Figure 2 | DOC concentrations of the three secondary effluents. Data were analysed using box whisker plots. Vertical lines represent the limits of maximum and minimum values, the box the 25th to 75th percentile values, and the data point the median; *n* = 13 for EBPR; *n* = 14 for SBR; *n* = 6 for SSA.

EBPR and SBR effluents, respectively. The ammonia concentration for the SSA was around 2.5 mg/L, much higher than for the two other effluents, with a large box range between 0.5 and 4.5 mg/L (Figure 3).

With regard to phosphates and phosphorus content, the results shown in Figure 4 suggest that phosphate was the predominant form of phosphorus found in these effluents and that there was no detectable organic phosphorus. A box range from 2 to 3 mg/L of phosphate was measured for the EBPR with a median of about 2.5 mg/L. The SSA

process included coagulation by dosing ferric chloride for phosphate precipitation; however, the mean was around 2 mg/L. In comparison, the SBR had a lower concentration with a residual of about 1 mg/L.

Complex interaction phenomena between water constituents and membrane material influenced by system configuration, which vary from site to site, are responsible for fouling. It is well known that nutrient content is directly correlated to the level of biofouling (Vrouwenvelder *et al.* 2010; Xu *et al.* 2010). The characteristics of various types of

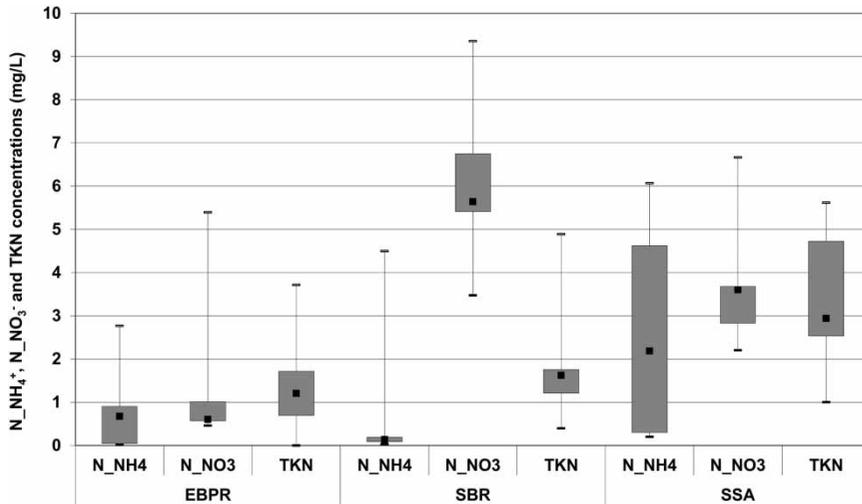


Figure 3 | Ammonia, nitrate and total Kjeldahl nitrogen (TKN) concentrations of the three secondary effluents. Data were analysed using box whisker plots. Vertical lines represent the limits of maximum and minimum values, the box the 25th to 75th percentile values, and the data point the median; $n = 14$ for EBPR; $n = 16$ for SBR; $n = 6$ for SSA.

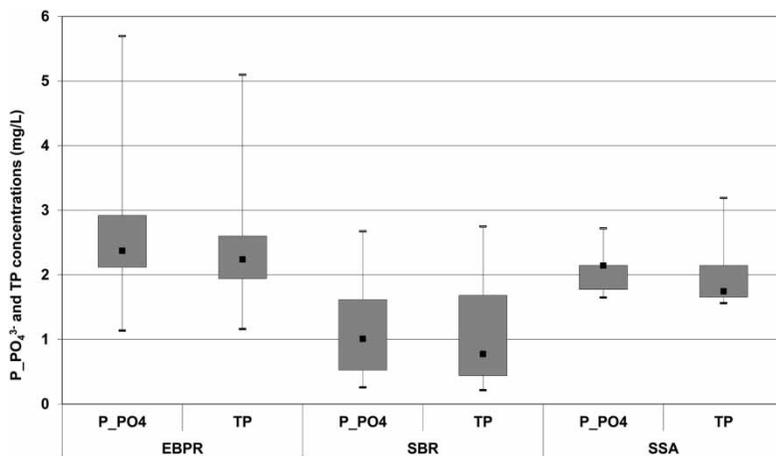


Figure 4 | Phosphorus and phosphate concentrations of the three secondary effluents. Data were analysed using box whisker plots. Vertical lines represent the limits of maximum and minimum values, the box the 25th to 75th percentile values, and the data point the median; $n = 14$ for EBPR; $n = 16$ for SBR; $n = 6$ for SSA.

effluents suggested that EBPR effluent will contribute the least to fouling in tertiary treatment with membrane applications. The SBR had a higher organic carbon level and the SSA had a higher phosphate level. Higher organic levels have been reported to cause more severe fouling of low-pressure membranes (Lozier *et al.* 2008). High phosphate concentration has been reported to increase the growth of microorganisms on the surface of high pressure membranes (Vrouwenvelder *et al.* 2010). Therefore, both SBR and SSA effluents were expected to contribute to organic fouling and to have a strong impact on the development of biofouling on tertiary membranes. As part of another study, a membrane

pilot plant is currently being investigated at the EBPR and SBR plants to confirm these results in real conditions.

Fluorescence spectrometry (Figure 5) clearly shows larger amounts of protein-like compounds in the secondary effluent from the SBR in comparison to the SSA or the EBPR (Ex: 220–230 nm/Em: 330–360 nm). Zheng *et al.* (2009) showed that the biopolymer concentration influences the filterability of corresponding water samples proportionally. Zheng's experiments demonstrated that pore blocking or cake/gel fouling were the main fouling mechanisms (Zheng *et al.* 2009). As a result, the SBR secondary effluent was likely to have a stronger fouling potential.

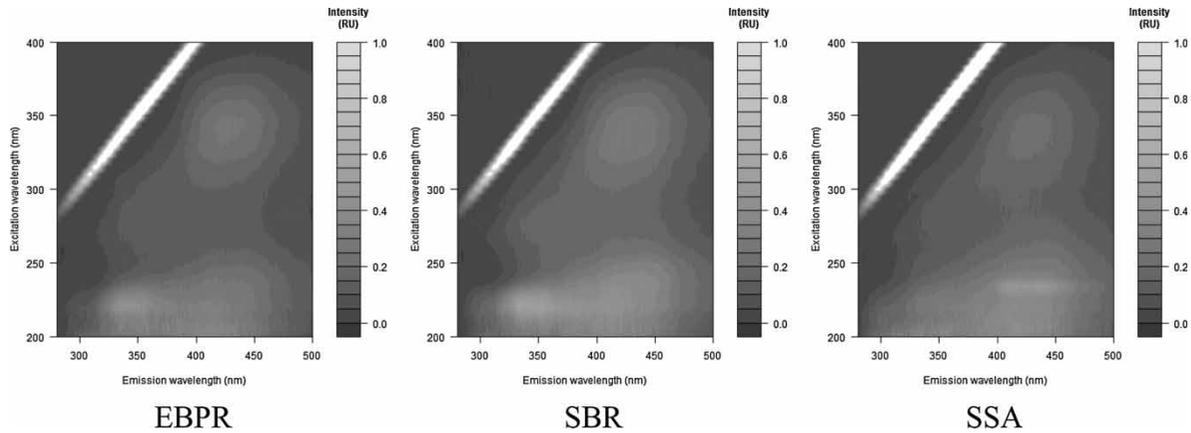


Figure 5 | Excitation-emission matrix plots for the three secondary effluents (Y-scale: 200–400 nm/X-scale: 280–500 nm). RU: Raman units.

This trend was confirmed by the LC-OCD results shown in Figure 6, with concentration of biopolymers (BP) up to double that in the SBR than SSA samples, and five times higher than the EBPR samples. Also, the concentration of humic-like substances (HS) in the SBR samples was three times higher than in the EBPR samples and double the concentration in the SSA samples. Peiris *et al.* showed that colloidal/particulate matter is considered to be mostly responsible for reversible fouling, and humic-like and protein-like substances are more responsible for irreversible fouling behaviour on UF membranes (polysulfone and polyethersulfone, GE Osmonics) (Peiris *et al.* 2010). Consequently, more significant irreversible fouling was expected with the effluent from the SBR.

The objective of the first part of the study was achieved with the identification and quantification of the different organic fraction levels in each secondary effluent. The EBPR effluent appeared to have the lowest concentration of BP and HS, which are known to cause fouling, in contrast to the SBR effluent which appeared to have the most favourable composition to lead to both reversible and irreversible fouling.

Filtration tests

The continuous filtration experiments confirmed the very good quality of the EBPR effluent (Figure 7) with regard to fouling, with a low permeability decrease of 7.1% after

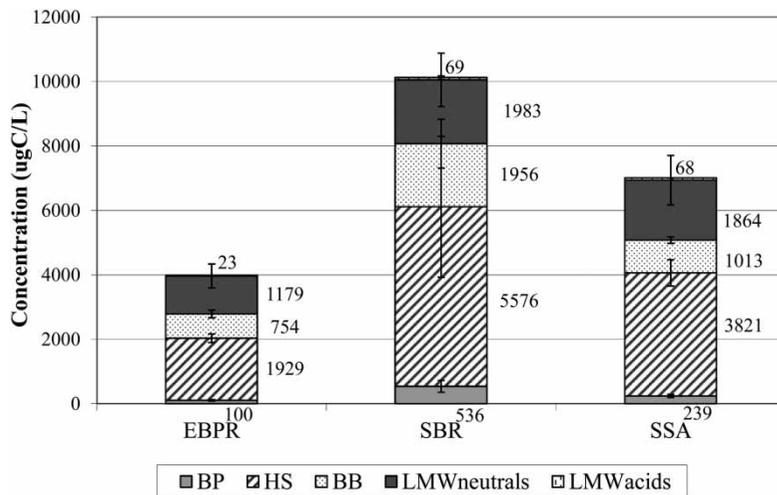


Figure 6 | Comparison of the different fractions in the three effluents. BP = biopolymers; HS = humic-like substances; BB = building block substances; LMW = low molecular weight. $n = 11$ for EBPR; $n = 10$ for SBR; $n = 6$ for SSA.

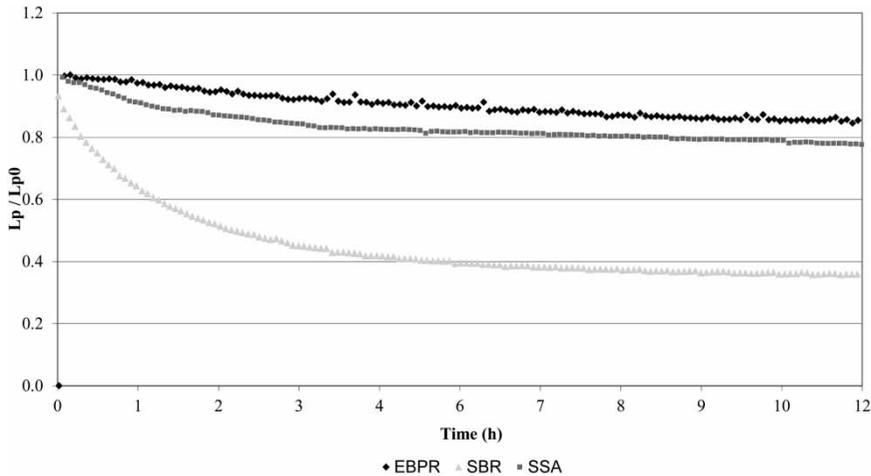


Figure 7 | Fouling potential during a long-term fouling test for the three secondary effluents (EBPR, SBR and SSA). L_p = permeability ($L/m^2 \cdot h \cdot bar$); L_{p0} = initial permeability ($L/m^2 \cdot h \cdot bar$).

Table 2 | Results of filtration test after a single long period of filtration of 12 h

	DOC feed (mg/L)	DOC filtrate (mg/L)	L_p decrease (%)	Flux recovery after backwash (%)
EBPR	5.0 ± 0.1	5.1 ± 0.1	7.1 ± 0.5	97.8 ± 0.2
SBR	8.8 ± 0.1	8.9 ± 0.1	60.5 ± 2.1	112.0 ± 2.0
SSA	8.2 ± 0.2	8.6 ± 0.2	29.5 ± 1.2	109.5 ± 0.5

12 h of continuous filtration (Table 2). As expected from the physical-chemical characteristics of the effluent, the SBR effluent resulted in a greater permeability decrease with a significant drop of 60.5% after $680 L/m^2$ of effluent had filtered through the membranes (Table 2). For the three effluents, no measurable DOC removal was observed between the feed and the filtrate (Table 2). Prior to DOC analysis, feed samples were passed through a $0.45 \mu m$ filter thereby quantifying only dissolved organic carbon.

With the three effluents, the observed fouling was mainly reversible and can be removed after backwash, with a recovery near to 100% in each case (Table 2). Flux recovery was slightly higher than 100% for the SBR and

the SSA. This unexpected result was likely due to insufficient conditioning of the membrane prior to the filtration test because of the different effectiveness of membrane conditioning at full- and laboratory-scale. Membrane permeability was also completely recovered by chemical cleaning. Therefore no chemically irreversible fouling was observed on this lab-scale unit under the experimental conditions.

Experiments after successive multi-cycle filtrations showed a more significant flux decline for the SBR as compared to the two other effluents tested. As for the continuous filtration, the decrease of the permeability of the SBR was significant with an average of 40% after 15 min of filtration.

Table 3 | Results of filtration tests after multi-cycle filtrations. Average and standard deviation for 10 cycles in duplicate

		DOC feed (mg/L)	DOC filtrate (mg/L)	L_p decrease (%)	Flux recovery after backwash (%)
EBPR	1	4.4 ± 0.2	5.1 ± 0.3	7.1 ± 1.4	99.8 ± 1.6
	2		5.0 ± 0.1	7.9 ± 2.3	99.9 ± 2.3
SBR	1	12.6 ± 0.1	12.1 ± 0.3	42.2 ± 2.0	83.8 ± 4.1
	2		12.3 ± 0.1	39.0 ± 3.2	82.2 ± 7.2
SSA	1	7.3 ± 0.2	7.1 ± 0.1	5.6 ± 2.5	100.2 ± 6.9
	2		7.7 ± 0.1	4.6 ± 2.1	100.3 ± 7.3

This rapid fouling can be explained by pore blocking phenomena as described by Kim *et al.* (2008). SBR feed water contained significantly more BP (including protein-

like substances and polysaccharides) and HS which accumulated on the membrane surface forming a cake layer, and the adsorption of small particles in the membrane

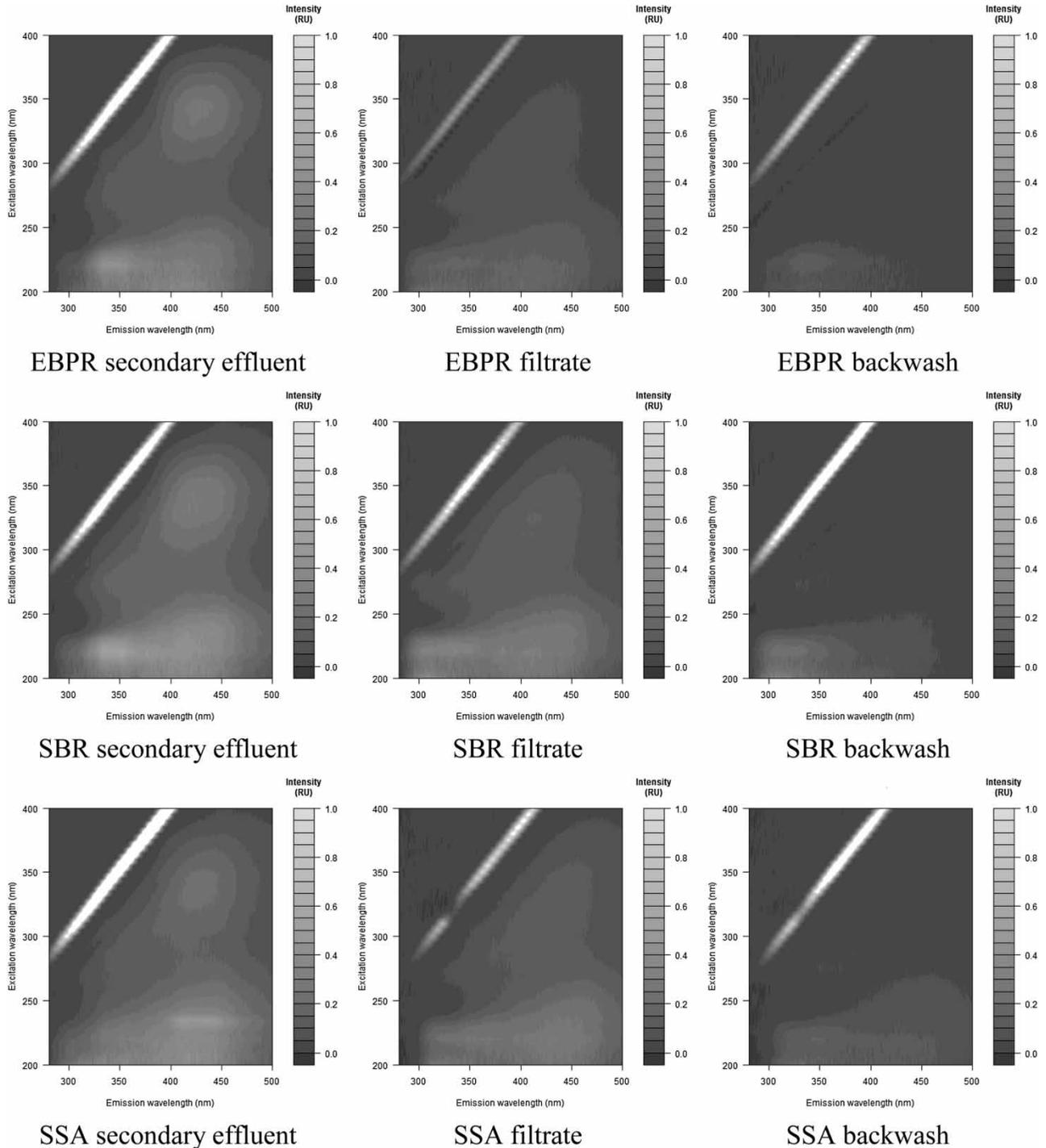


Figure 8 | Excitation-emission matrix plots for the feed/filtrate and backwash samples of the three secondary effluents (Y-scale: 200–400 nm/X-scale: 280–500 nm). RU: Raman units.

pores reduced filtration efficiency. Again the results showed that no significant DOC removal occurred, whatever the type of secondary effluent used (Table 3).

Recoveries after backwash are compared in Table 3. Close to 100% permeability recovery was observed for the EBPR and SSA effluents. Expected irreversible fouling for the SBR effluent based on characterization was observed: the results showed only 83% flux recovery. These results showed the significant contribution of the BP and HS in the hydraulically irreversible fouling. Again, membrane permeability was completely recovered with chemical cleaning for all three effluents, indicating that the SBR fouling was hydraulically irreversible but chemically reversible.

Foulant characterization

As shown in Figure 8, the use of fluorescence underlined the partial retention of high molecular weight compounds such as protein-like compounds by the UF membranes (Ex: 220–230 nm/Em: 330–360 nm). EEM profiles indicated that a fraction of humic-like compounds (Ex: 340 nm/Em: 430–440 nm) and fulvic-like compounds (Ex: 240 nm/Em: 430–440 nm) was also retained by the membranes. Backwash samples clearly showed that protein-like substances were the major part of the organics recovered from the membranes. This work supports literature data on low-membrane fouling showing that the high molecular weight organic materials comprised of hydrophilic components such as soluble microbial products and protein-like compounds were the major cause of membrane fouling (Jarusutthirak *et al.* 2002; Shon *et al.* 2006; Fan *et al.* 2008).

The LC-OCD confirmed these results (data not shown). As expected, membranes removed a fraction of the aromatic compounds, such as proteins and humic-like substances having a molecular weight above 1000 D. The filtrate had slightly lower intensity than the feed, meaning a minor part was retained by the membranes. Previous studies showed that these substances (organic colloids, polysaccharides and proteins) are partly retained by UF and MF membranes and are responsible for the observed fouling (Lee *et al.* 2004, 2008; Laabs *et al.* 2006). The backwash samples (operated with MQ water) showed a large removal of organic matter from the surface of the membranes, confirming backwash efficiency for removing accumulated material.

Characterization results showed that part of high molecular weight substances are partially retained by the membranes. In particular for the SBR, BP fraction decreased after UF filtration but was not removed in the backwash samples, indicating that BP were retained on the membrane surface and thus confirming their major role in membrane fouling. The experimental conditions run for 10 cycles only indicated a decrease in permeability in continuous filtration and in multi-cycle filtrations for the SBR effluent, confirming the characterization of a hydraulically irreversible fouling.

CONCLUSIONS

The characterization of three secondary effluents has demonstrated that they have different organic matter distribution. EBPR effluent appeared to be the one with the best water quality, with low organic carbon content and low concentration of BP. No major fouling was observed at bench-scale confirming its potential to be further treated with low-pressure membranes for water recycling. SSA and SBR effluents were more concentrated, especially SBR which had a high concentration of BP and HS and was expected to result in more irreversible fouling on low-pressure membranes. Bench-scale filtration experiments indicated a significant permeability decrease and an incomplete flux recovery after a backwash for the SBR effluent, confirming the hydraulically irreversible fouling properties. However, the fouling was reversible after a chemical cleaning. A comparison of lab-scale/pilot-scale fouling is currently under process and will be able to determine if there is a correlation between lab-scale characterization and full-scale membrane fouling.

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