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Membrane fouling of a hybrid moving bed membrane bioreactor plant to treat real urban wastewater

Jaime Martín-Pascual[a], Patricia Reboleiro-Rivas[b], María M. Muñio[c], Jesús González-López[d], José M. Poyatos[e,*]

[a] Department of Civil Engineering, University of Granada, 18071 Granada, Spain
[b] Institute of Water Research, University of Granada, 18071 Granada, Spain
[c] Department of Chemical Engineering, University of Granada, 18071 Granada, Spain
[d] Institute of Water Research, University of Granada, 18071 Granada, Spain
[e] Department of Civil Engineering, University of Granada, 18071 Granada, Spain

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ABSTRACT

The influence of operative variables in the performance of an ultrafiltration membrane in a hybrid moving bed membrane bioreactor treating real urban wastewater was studied in relation to the fouling rate and the recovery of permeability with a multivariable statistical analysis. Twenty-one cycles of operation were studied in relation to the filling ratio, flux, temperature, biomass concentration in a pilot-scale experimental plant. The pilot plant consisted of three units of ZW-10 submerged membrane and 20, 35 and 50% of K1 of Anoxkaldnes were used as carrier. The statistical analysis has shown that the most influential variables in the performance of the membrane were temperature and flux. Transmembrane pressure ranged from 22 kPa to 68 kPa, increasing with the MLSS, BFSS and flux and decreasing with temperature. The presence of biofilm negatively affected the performance of the membrane in relation to the fouling rate; this varied between 0.26 and 1.22 kPa/day, and was found to increase when viscosity and BFSS increased and temperature decreased.

1. Introduction

Advanced technologies regarding wastewater treatment are necessary to preserve water quality and satisfy the limits imposed on the effluent from municipal wastewater treatment plants (WWTPs) by the Water Framework Directive [4]. Biological processes are a cost-effective and environmentally friendly alternative [25] that allow the complete treatment of the wastewater [5]. However, the efficiency of biological processes can be improved with the use of membranes. Membrane separation processes are widely used in water desalination, biochemical processing, industrial wastewater treatment, food and beverage production, and pharmaceutical applications [10]. Membrane bioreactors (MBR), which replace the settling tank with membrane filtration [11,35], represent an attractive treatment technology in wastewater management, since they produce a high quality effluent with not much surface demand [15]. Due to their unique advantages such as product quality, high efficiency, ease of operation and small footprint, membrane bioreactors have become state-of-the-art in wastewater treatment and are becoming increasingly popular [6,41].

In practical application processes, the efficiency of membrane filtration and separation is limited by concentration polarisation and membrane fouling problems [14]. Membrane fouling has been the main drawback to the wide application of MBR because it causes decreasing permeate flux or increasing transmembrane pressure [37], resulting in decreasing membrane performance and increasing frequent membrane cleaning and energy consumption [8,41]. The major factors affecting fouling, described by [9], are biochemical kinetic parameters, temperature, membrane characteristics, the characteristics of mixed liquor, operational style and reactor hydraulic conditions. The mixed liquor suspended solids concentration (MLSS) and flux of the membrane affect membrane fouling in MBR processes [29,30]. Therefore, membrane fouling mechanisms are very complicated due to the complex rheological and physiological characteristics of mixed liquors [2].

An alternative to managing this problem is the hybrid moving bed biofilm reactor (MBBR)–MBR. Using this technology, the biofilm system may reduce the concentration of suspended solids

* Corresponding author.
E-mail addresses: jmpascual@ugr.es (J. Martín-Pascual), preboleiro@ugr.es (P. Reboleiro-Rivas), mmmunio@ugr.es (M.M. Muñio), jgl@ugr.es (J. González-López), jpoyatos@ugr.es (J.M. Poyatos).

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and improve the extent of membrane fouling [38], providing optional strategies for minimising the problem of fouling [12], in comparison with MBR. The hybrid MBBR technology has emerged as a compact treatment alternative to conventional activated sludge reactors for the treatment of municipal and industrial wastewater [27]. This system combines suspended biomass and biofilm processes inside the biological reactor for biofilm growth [26]. Therefore, they include positive aspects of the growth of suspended and attached biomass and aim to partially mitigate the fouling concerns in relation to MBR systems and the settleability issues regarding MBBR systems [18]. In contrast to most biofilm processes, the whole volume can be used for biomass growth [7]. Another important advantage of hybrid MBBR system is that the filling ratio of biofilm carriers in the reactor may be subject to preferences [33]. Martín-Pascual et al. [22] showed that the COD and BOD_5 removals in a hybrid MBBR-MBR working under a lower concentration of MLSS were similar to those obtained in an MBR working with high MLSS concentrations due to the presence of suspended and attached biomass.

The aim of the research was to study the influence of operative variables filling ratio, flux, dynamic viscosity, temperature and biofilm suspended solids concentration (BFSS) on the performance of an ultrafiltration membrane in a complex technology such as a hybrid moving bed membrane bioreactor to treat real urban wastewater throughout the fouling rate and the recovery of the permeability after the backwashing and chemical cleaning with a statistical multivariate analysis.

2. Material and methods

2.1. Experimental procedure

2.1.1. Description of the pilot-scale experimental plant

A pilot-scale experimental plant of hybrid MBBR-MBR was used in this research. The pilot plant was located in the WWTP Los Vados of Granada (Spain). The influent was taken from the outlet of the primary settler of the WWTP, so, the pilot plant was fed with real urban wastewater pretreated with a conventional physical pretreatment and primarily settled. As shown in Fig. 1, the experimental plant had two bioreactors: a bioreactor with an operating volume of 358 L and a tank with three submerged Zenon® hollow fibre ultrafiltration membrane units. In the first part, biodegradation took place and presented a 20, 35 and 50% filling ratio (rate between the apparent carrier volume and the operational volume of the bioreactor) of carriers. In the second part, solid separation was carried out and with an operating volume of 87 L.

The modules used were ZW-10, the configuration of which is outside/in hollow fibre with a nominal membrane surface area of 0.93 m^2, a nominal pore size of 0.04 μm and an absolute pore size of 0.1 μm. The carriers used were K1, developed by AnoxKaldnes® AS (Norway). This carrier is made of high-density polyethylene and shaped into small cylinders with a cross inside the cylinder and "fins" on the outside; this provides a greater surface-to-volume ratio. Their density values range from 0.92 to 0.96 g cm^{-3} and the specific biofilm surface area in bulk is 500 m^2 m^{-3}; it has been recognised that the biofilm is negligible on the outer surface of K1 carriers [33].

In order to maintain the concentration of biomass in both reactors, a recycling pump with a constant flow of 90 L/h took the sludge from the membrane tank to the hybrid MBBR. In this way, the characteristics of the suspended biomass were similar and the Solids Retention Time (SRT) could be defined for the global system. The excess sludge was extracted under constant flow in each phase according to the SRT.

2.1.2. Operating conditions

During the study, the submerged membrane units were operated at a constant flux using a suction pump in each cycle, and varying the transmembrane pressures (TMP). The cyclic mode

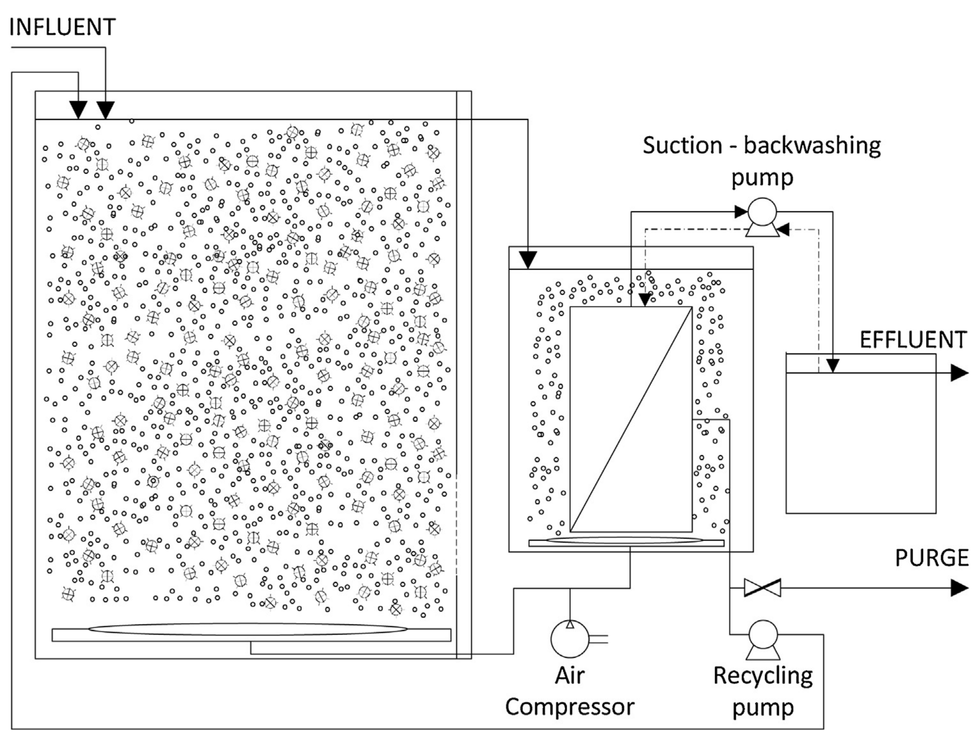

Fig. 1. Schematic diagram of the pilot plant of hybrid MBBR-MBR used with a cylindrical tank in which k1 of Anoxkaldness carriers were contained in a 20, 35 and 50% of filling ratio and a rectangular tank in which three units of ZW10 of Zenon were submerged. Dashed line represents the backwashing flux.

Table 1
Filling ratio, hydraulic retention time (HRT), solid retention time (SRT) and BOD5 of influent and effluent of each cycle tested in the research.

Cycle	Filling ratio (%)	HRT (h)	SRT (days)	BOD_5 Influent (mg/L)	BOD_5 Effluent (mg/L)
1	20	10	12.71	320 ± 80	31 ± 9
2	20	10	12.71	407 ± 95	37 ± 9
3	20	10	12.71	415 ± 62	26 ± 11
4	20	24	24.72	359 ± 76	8 ± 3
5	20	10	22.25	300 ± 65	6 ± 2
6	20	10	22.25	279 ± 71	7 ± 3
7	20	10	22.25	247 ± 42	5 ± 2
8	20	10	22.25	275 ± 53	6 ± 3
9	20	10	22.25	312 ± 74	6 ± 4
10	20	24	55.63	314 ± 82	4 ± 3
11	35	24	18.54	367 ± 73	10 ± 9
12	35	24	18.54	378 ± 58	5 ± 1
13	35	10	8.56	379 ± 67	14 ± 5
14	35	10	17.80	317 ± 87	9 ± 3
15	35	10	17.80	296 ± 58	6 ± 1
16	35	24	56.53	301 ± 69	5 ± 2
17	50	10	13.91	274 ± 124	5 ± 3
18	50	10	18.59	291 ± 101	8 ± 6
19	50	10	18.59	403 ±n52	17 ± 8
20	50	24	49.44	411 ± 39	23 ± 5
21	50	24	49.44	398 ± 37	21 ± 11

of operation of the membrane primarily consisted of a filtration period of 9.67 min with 0.33 min of backwash, and the air scouring of the membrane being continuously applied.

The average influent presented an organic matter concentration of 334 ± 88 mg/L of DBO_5 and 454 ± 104 mg/L of COD, the suspended solids of the influent wastewater were 119 ± 36 mg/L of totals and 106 ± 32 mg/L of volatiles with a pH of 7.64 ± 0.26 and a conductivity of 1237 ± 236 μS/cm. The dissolved oxygen during the research was controlled in the hybrid MBBR, maintaining an average value of 1.92 ± 0.82 mg/L.

Twenty-one cycles of operation were studied in the present research in relation to the filling ratio, flux, SRT, temperature, MLSS and BFSS (Table 1 and Table 2). The cycles were ordered chronologically, increasing the filling ratio and the MLSS; the durations depended on the operative conditions tested, as they were higher when the temperature was higher and the MLSS and flux were lower. These cycles operated with 20% (cycles from 1 to 10), 35% (cycles from 11 to 16) and 50% (cycles from 17 to 21) of the filling ratio and under two different fluxes of the membrane: 18.96 L/h (cycles 1, 2, 3, 5, 6, 7, 8, 9, 13, 14, 15, 17, 18 and 19) and 45.5 L/h (cycles 4, 10, 11, 12, 16, 20 and 21), corresponding to HRT of 10 and 24 h, respectively. SRT ranged between 8.56 days (cycle 13) and 56.53 days (cycle 16) according to the other operative condition which fixed the SRT to maintain the biomass concentration approximately constant. Data of BOD_5 of influent and effluent are shown in Table 1. As shown by Table 2, the rest of the operative variables showed different values, which presented statistically significant differences. The different homogenous subsets of the HSD of Tukey of ANOVA test, with $\alpha = 0.05$ in each variable, are shown as superscript; the cycles with different superscripts presented statistically significant differences, and those cycles were statistically homogeneous. The temperature changed between 5.5 ± 2.17 and 28.24 ± 1.48 °C. The MLSS varied from 1721 ± 795 to 4717 ± 87 mg/L, changing the BFSS between 1492 ± 462 and 6608 ± 989 mg/L of carrier. Prior to the beginning of each cycle, organic cleaning with hypochlorite (200 mg/L) was performed for 4 h.

2.2. Physical and chemical determination

Samples of 1 L from the feed tank, biological reactor and permeate were taken as three replicates every 24 h for analytical determination. BOD_5 were determined according to the American Public Health Association, the American Water Works Association and the Water Environment Federation (APHA-AWWA-WEF) method. The MLSS were determined by gravimetric methods [1]. Tests on carrier samples were carried out in order to establish the amount of biomass attached to the carriers [21]. This biomass was quantified as BFSS in relation to the bioreactor volume.

The viscosity of the mixed liquor was measured at different temperatures with a viscosimeter (Brookfield, Model LVDVE) using a spindle number 18 and a Small Sample Adapter (SSA) at 60 rpm. The relationship between temperature (T) and dynamic viscosity (μ) was determined using Andrade's equation (Eq. (1)), fitting Andrade equation coefficients (A and B):

$$\mu = A\ \exp(-T/B) \tag{1}$$

Since mixed liquor has non-Newtonian rheology, the viscosity obtained is the apparent viscosity under the temperature experimented by the sludge in the pilot plant.

2.3. Statistical analysis

The data obtained throughout this study were analysed using a computer-assisted statistics program, SPSS 20 for Windows. A least significant differences test (LSD test) was used to measure the differences between the different operational conditions studied (temperature, MLSS, and BFSS). Normality tests of the data were performed using the Shapiro-Wilk test, since the dataset was smaller than 2000 elements. An analysis of variance (ANOVA) was used to assess the homogeneity of variance, with a significance level of 5% ($P < 0.05$).

Moreover, a multivariable statistical analysis using the software Canoco for Windows version 4.5 was used to quantify the influence of the variables on the parameters of the behaviour of the membrane and to obtain variables with the greatest influence on the working of the membrane. A Detrended Correspondence Analysis (DCA), the most appropriate ordination statistical analysis, was carried out in order to obtain gradient lengths. DCA revealed that the longest ordination axis was lower than three, so the distribution of the model was linear. Redundancy Analysis (RDA) was used due to the fact that the distribution of the model was linear, as described by the statistical method recommended by [17]. Statistical significance was tested using a Monte Carlo test with 499 permutations and a selected significance level of 0.05.

2.4. Fouling rate modelling

A simple model of the fouling rate (FR) can be proposed considering the operative variable tested. Judd [42] proposed a linear model between fouling rate and flux (J) and [23] described a linear relationship of the permeability with the temperature (T) and dynamic viscosity. Dynamic viscosity of the sludge depends of both MLSS and BFSS, so, FR can be modelled linearly as Eq. (2) shows.

$$FR = A\ J + B\ T + C\ MLSS + D\ BFSS + E \tag{2}$$

Being A, B, C, D and E the fit constants which were obtained with solver function of Microsoft Excel considering the function to minimize the quadratic difference between the empirical and modelled data.

Table 2
Average of the filling ratio, flux (J), temperature, MLSS and BFSS of each cycle tested in the research. The different homogenous subset of the HSD of Tukey of ANOVA test done with α=0.05 in each variable are shown as superscript. So, the cycles with different subsets in a data presented statistical significance differences in this variable.

Cycle	Filling ratio (%)	J (L/h)	Temperature (°C)	MLSS (mg/L)	BFSS (mg/L of carrier)
1	20	18.96	$9,36 \pm 1.95^{b, c}$	1721 ± 795^{a}	$1492 \pm n462^{a}$
2	20	18.96	$11,24 \pm 2.01^{c},^{d}$	$2017 \pm 403^{a,b,c}$	$2117 \pm 218^{a,b}$
3	20	18.96	$16,58 \pm 2.81^{f,g}$	$2632 \pm 467^{a,b,c}$	$2524 \pm 299^{a,b}$
4	20	45.50	$20,81 \pm 3.98^{h}$	$2576 \pm 713^{a,b,c}$	$3814 \pm 618^{c,d,e}$
5	20	18.96	$27,07 \pm 0.94^{i,j,k}$	$4009 \pm 1039^{e,f,g}$	$4634 \pm 498^{e,f,g,h}$
6	20	18.96	$28,24 \pm 1.48^{k}$	$3969 \pm 1044^{e,f}$	$3216 \pm 1099^{b, c, d}$
7	20	18.96	$24,64 \pm 1.08^{i,j}$	$2226 \pm 652^{b,c}$	$4121 \pm 706^{d,e,f}$
8	20	18.96	$19,78 \pm 2.39^{g,h}$	$4503 \pm 392^{f,g,h}$	$4970 \pm 306^{f,g,h,i}$
9	20	18.96	$14,70 \pm 1.57^{e,f}$	$4099 \pm 384^{g,h}$	$4989 \pm 691^{f,g,h,i}$
10	20	45.50	$11,25 \pm 2.34^{c,d}$	$4484 \pm 380^{g,h}$	6608 ± 989^{j}
11	35	45.50	$9,19 \pm 2.26^{b,c}$	$2127 \pm 883^{a,b}$	$2843 \pm 1524^{b,c}$
12	35	45.50	$12,85 \pm 2.45^{d,e}$	$3187 \pm 191^{c,d}$	$4366 \pm 171^{e,f,g}$
13	35	18.96	$18,16 \pm 3.86^{g,h}$	3216 ± 214^{c}	$5475 \pm 313^{g,h,i,j}$
14	35	18.96	$26,59 \pm 2.24^{i,j,k}$	$4496 \pm 363^{e,f,g,h}$	$5404 \pm 257^{g,h,i}$
15	35	18.96	$27,43 \pm 1.81^{j,k}$	$4698 \pm 251^{g, h}$	$5249 \pm 396^{f,g,h,i}$
16	35	45.50	$24,15 \pm 2.34^{i}$	$4552 \pm 496^{g,h}$	$4729 \pm 300^{e,f,g,h}$
17	50	18.96	$18,39 \pm 1.93^{g,h}$	$2561 \pm 473^{b,c}$	$4189 \pm 2113^{d,e,f}$
18	50	18.96	$11,50 \pm 4.60^{c,de}$	$3635 \pm 897^{d,e}$	$5898 \pm 595^{i,j}$
19	50	18.96	$7,69 \pm 3.82^{a,b}$	4713 ± 168^{h}	$5518 \pm 266^{h,i,j}$
20	50	45.50	$9,71 \pm 4.57^{b,c,d}$	4717 ± 87^{h}	$5269 \pm 210^{f,g,h,i}$
21	50	45.50	$5,50 \pm 2.17^{a}$	4616 ± 40^{h}	$4622 \pm 189^{e, f, g, h}$

3. Results and discussion

3.1. Evolution of transmembrane pressure in the hybrid MBBR-MBR system

The evolution of TMP is shown in Fig. 2, which represents the average daily values of the TMP to obtain the flux during the research. The data of each cycle, before and after the periodic backwashing, are shown in Fig. 2a and b, respectively. These values presented a wide range of values, which varied from 22 kPa (cycle 16) to 67 kPa (cycle 19) after the backwashing and from 25 to 68 kPa in the same cycles before the backwashing. Comparing these subfigures, it can be observed that the recovery of the membrane permeability after the backwashing decreases when

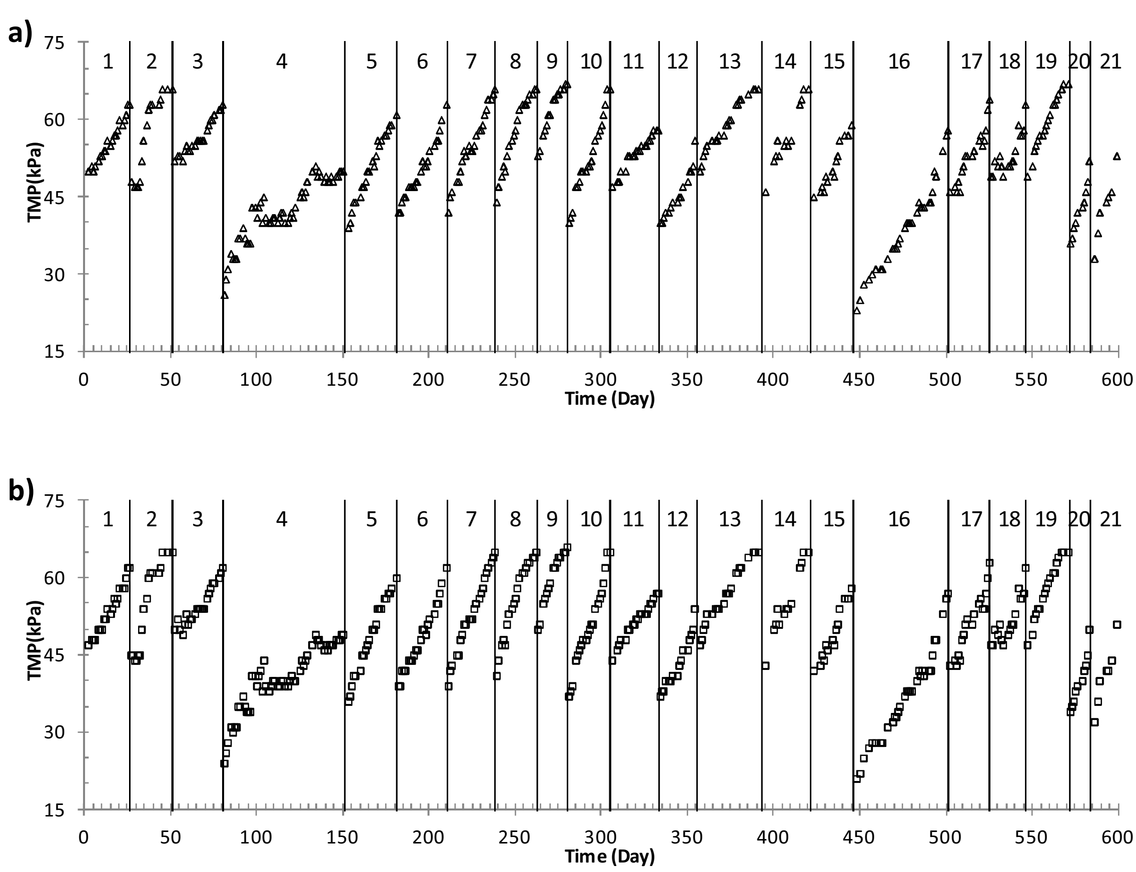

Fig. 2. Evolution of transmembrane pressure (TMP) through the 21 cycles between chemical cleaning of the research. (a) represents the TMP previous to the backwashing and (b) after backwashing.

the fouling is higher, decreasing the average recovery from 6.79 ± 1.46% at the beginning of the cycles to 2.06 ± 0.78% at the end.

The variations in TMP are caused by the change in operative variables (Table 2). The effect of each variable independent of the rest is difficult to separate due to the fact that the pilot plant worked with a real effluent and it was exposed to typical fluctuations of temperature in a WWTP [15]. However some common patterns in the performance of the membrane can be observed in relation to some of the operative variables. In the values obtained, the effect of MLSS and flux was clear: TMP increases with flux and MLSS. In cycles with similar temperatures, MLSS and BFSS, and different fluxes, such as cycles 3 and 4 or 19 and 20, the decrease in TMP with the increase in flux from 18.96 to 45.50 L/h is patented. The same effect can be observed in relation to the MLSS; comparing cycles 17 and 18 with similar BFSS and the same flux, but MLSS more than 1000 mg/L higher, it is observed that the TMP is lower in the case of the lower MLSS.

Temperature is one of the most influential variables in the performance of a membrane; fluctuation patterns of the filterability are consistent with seasonal temperature fluctuations [15], and deterioration of the activated sludge filterability in winter is a common observation in MBR installations treating domestic sewage [31,20,24,36]. In this research, as the pilot plant worked in a constant flux mode, these fluctuations were observed in the variation of the TMP showed in the different cycles of Fig. 1. Goosen et al. [10] showed that the flux can be improved with the change in the feeding temperature due primarily, although not completely, to viscosity effects on the water. Therefore, the operation of the membrane can be sensitive to changes in feed temperature [6,13,28]. This aspect can be shown by comparing cycles such as 8 and 9; these cycles operated with the same flux and similar MLSS and BFSS, however the TMP needed to obtain the flux was higher in cycle 9, in which the average temperature was 5.08 °C lower, even though the MLSS was slightly higher. Moreover, it can be observed that the cycles with an average temperature higher than 25 °C (cycles 5, 6, 14 and 15) presented TMP values that were relatively lower in spite of the higher concentration of MLSS. Indeed, the effect of temperature in the TMP needed to maintain the flux was observed during some cycles such as cycle 4, in which an increase of the TMP on approximately the 100th day was experimentally caused by a significant decrease of the temperature by about 6 °C. This fact is in accordance with previous research with the same membrane units, which showed that an increase of 5 °C could cause an increase of permeate flux of about 20% at temperatures lower than 20 °C [23].

3.2. Effect of the operative variables in the fouling of the membrane in the hybrid MBBR-MBR system

The fouling rate determines the frequency of chemical cleaning of the membrane. In order to study the effects of the operative variables on the fouling of the membrane, the permeability was obtained from the TMP needed to maintain the flux in each cycle. The values of permeability prior to chemical cleaning in each cycle are shown Table 3, in which the average fouling rate and the recovery of permeability are also indicated.

Two different ranges of final permeability are clearly differentiated in relation to the flux of the cycle; however, some differences could be observed among the cycles with the same flux. With a flux of 45.50 L/h m^2 (cycles 4, 10, 11, 12, 16, 20 and 21), the highest value was obtained under an MLSS of 2576 ± 713 mg/L and a temperature of 20.81 ± 3.98 °C (cycle 4), decreasing the permeability when the MLSS increases and the temperature decreases. In contrast, the minimum value with 45.50 L/h m^2 was obtained in cycle 10, which operated with average MLSS and temperature values of 4484 ± 380 mg/L and 11.25 ± 2.34 °C, respectively. With a flux of 18.96 L/h m^2, the final permeability was about 0.3 L/h m^2 kPa, varying between 0.28 L/h m^2 kPa in cycles 9 and 19 with higher MLSS (4099 ± 384 and 4713 ± 168 mg/L, respectively) and low temperatures (14.70 ± 1.57 and 7.69 ± 3.82 °C, respectively) and 0.32 L/h m^2 kPa in the cycle with an average MLSS value of 4698 ± 251 mg/L and temperature of 27.43 ± 1.81 °C (cycle 15).

To quantify the fouling rate, TMP values before and after chemical cleaning were measured. The fouling rate was obtained from the slope between the variation in TMP and the duration of the cycle. As shown in Table 3, this rate varied between 0.26 and 1.22 kPa/day, with the experimental changes caused by variations in the operative variables. The fouling rates obtained are supported by those obtained by some authors [31,34], although they are superior to those obtained by others. [29], with the same membrane and influent operating with a conventional MBR, for slightly higher permeate flux values, obtained a fouling rate that

Table 3
Values of initial pressure to get the flux of the membrane, fouling rate after backwashing and recovery of the membrane permeability after a chemical cleaning with sodium hypochlorite and backwashing.

Cycle	Final permeability (L/m^2 h kPa)	Fouling Rate (kPa/day)	Recovery after chemical cleaning with sodium hypochlorite(%)
1	0.30	0.51	23.81
2	0.29	0.94	21.21
3	0.30	0.36	26.95
4	0.91	0.26	67.50
5	0.31	0.78	31.15
6	0.30	0.67	33.33
7	0.29	0.80	33.33
8	0.29	0.94	19.70
9	0.28	0.83	40.30
10	0.69	1.02	28.79
11	0.78	0.41	31.03
12	0.81	0.64	62.79
13	0.29	0.45	30.30
14	0.29	0.75	31.82
15	0.32	0.70	33.45
16	0.78	0.56	66.95
17	0.30	0.69	23.44
18	0.30	0.51	22.22
19	0.28	0.78	46.27
20	0.88	1.22	36.54
21	0.86	1.18	37.74

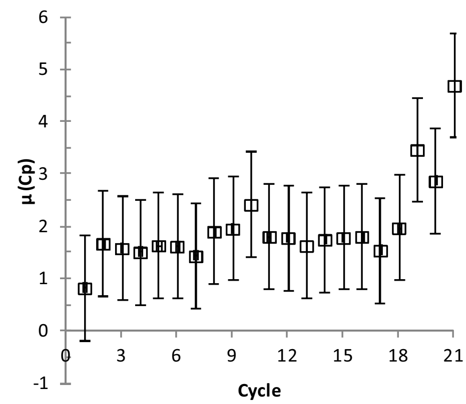

Fig. 3. Average and standard deviation values in each cycle of dynamic viscosity of the sludge.

was half of those obtained in this research using similar values for temperature and MLSS.

The temperature of the mixed liquor has an important effect on the fouling of a membrane [3]. The fouling rate decreases when the temperature increases, obtaining the highest values in cycles with lower temperatures, e.g. in cycles with a medium concentration of MLSS and low flux, under 20% of the filling ratio (cycles 2 and 3). Moreover, an important decrease in the fouling rate (from 0.94 to 0.36) was observed with a temperature increase of 5.36 °C.

The fouling rate also has a strong dependency with the MLSS and flux (fouling rate is higher when MLSS and flux increases). The effect of the flux can be observed by comparing cycles with similar temperatures and MLSS, e.g. cycles 15 and 16; here, the fouling rate was 21% higher in the cycle with the higher flux. With respect to the MLSS, comparing cycles 10 and 11, it is possible to observe that the fouling rate decreases (from 1.02 to 0.41 kPa/day) when the MLSS decreases (from 4.5 to 2.1 g/L).

Rosenberger et al. [32] observed that mixed liquor suspended solids (MLSS) constituted a major parameter influencing apparent viscosity in membrane bioreactor fouling. The viscosity was measured at different temperatures and obtained at a process temperature by Andrade's Equation (Eq. (1)) and was determined by the real temperature of the sludge in the pilot plant. Fig. 3 shows the viscosity in each cycle. The viscosity of the sludge obviously increased when the temperature decreased, e.g. between cycles 20 and 21, the reduction in average temperature of about 5 °C highly increased the viscosity due to the fact that the average temperature was lower than 10 °C while the MLSS was relatively constant. Therefore, as the mixed liquor and temperature are related with regard to viscosity, the dynamic viscosity was considered the representative variable of the feeding sludge in the performance of the membrane under different conditions tested.

In relation to the effect of the filling ratio and BFSS, researches comparing the fouling in membranes with flocs and biofilm obtained similar results. According to Luo et al. [19], the hybrid MBBR-MBR could reduce the membrane fouling propensity in relation to a conventional MBR as the transmembrane pressure (TMP) development was relatively slow. However, [38] stated that, under the same operational conditions, the fouling rate of hybrid MBBR-MBR was three times higher than of conventional MBR due to the presence of extracellular polymeric substances and proteins. Lee et al. [16] obtained a fouling rate that was seven times higher in systems with biofilm than with flocs; this could be due to a lower

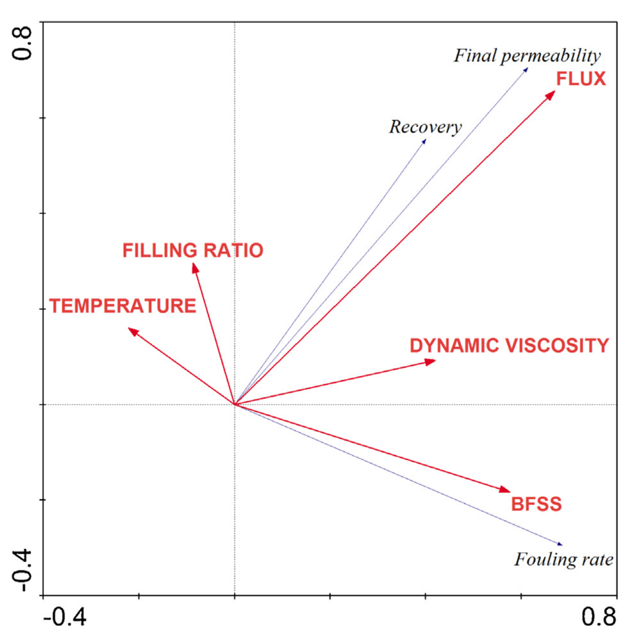

Fig. 4. Graphic representation of the multivariable analysis used to study the relationship between flux, temperature, dynamic viscosity, filling ratio and BFSS as variables, and final permeability (previous permeability before chemical cleaning), recovery of permeability after chemical cleaning with sodium hypochlorite and fouling rate of the membrane as species for the conditions tested in this study.

roughness in the layer formed on the membrane in the systems with only flocs. As Table 3 shows, in the present research, the highest values of fouling rate were obtained in the cycles with a higher filling ratio; however, these cycles were also the ones with the lower average temperature. Therefore, to establish the relationship separately is difficult, as it is necessary to use a multivariable analysis.

The recovery of permeability after chemical cleaning with sodium hypochlorite varied between 19.70 and 67.50%, with these differences being a consequence of the variability of the operative variables. The temperature, MLSS and flux influence the recovery of the membrane, as can be observed in the data shown in Table 3. Under the same filling ratio and similar temperature and MLSS, the recovery increases with the flux, e.g. in cycle 3, the recovery was 26.95%, whereas in cycle 4, in which the main variation was the increased flux, the recovery was 67.5%. Under the same and similar temperatures, recovery increases with the concentration of MLSS. However, the influence of the filling ratio is difficult to quantify since its effect seems to be cushioned by the temperature, as cycles with higher filling ratios presented lower average temperatures; therefore, both effects were studied in the multivariate analysis.

A redundancies analysis was performed to determine the relationships between the operational variables of the pilot plant (temperature, filling ratio, flux, dynamic viscosity and BFSS) and the behaviour of the membrane (Fig. 4). The analysis represented 62.4 and 97.8% of accumulated variance of the relationships between species and variables in the first and second axis, respectively. The Monte-Carlo test indicated that flux y temperature showed statistically significant differences, with P-values of 0.002 and 0.080, respectively. Therefore, these variables are the most influential with regard to the performance of the membrane.

On the one hand, the multivariable analysis showed that both final permeability and recovery after chemical cleaning were similarly influenced by the operative variables. The flux had a strong positive effect; both parameters increased when the flux increased. However, the temperature did not significantly affect the final permeability or recovery. Moreover, the rest of the variables (filling ratio, BFSS and viscosity) showed only a slightly positive influence. On the other hand, fouling rate presented a strongly positive correlation with viscosity and BFSS and a negative correlation with temperature; this means that the fouling rate increases with the viscosity and BFSS, and decreases when the temperature increases. Eq. (3) shows the fitting of the fouling rate with the variables. It is observed that the only variable that affects negatively is the temperature as previously was discussed. The correlation rate of the model was higher than 0.6 due to the difficult to fit the empirical data to a linear regression.

$$FR\,(\mathrm{kPa\ day^{-1}}) = 3.89\ 10^{-5}\ J - 8.80\ 10^{-3}\ T + 1.34\ 10^{-4} + 6.69\ 10^{-6}\ BFSS + 3.62\ 10^{-1} \quad (3)$$

As a result, the biofilm in the mixed liquor negatively affected the performance of the membrane in relation to the fouling rate. These facts are in accordance with the results obtained by Yang et al. [40] in a comparative study on membrane fouling between membrane-coupled MBBR and conventional MBR. It was observed that the fouling rates were much higher in the MBBR systems compared to the conventional MBR system, which indicated that membrane fouling potential was related not only to the concentration of soluble microbial products, but also to their sources and characteristics. Moreover, several similar studies have demonstrated less membrane fouling in conventional MBR in relation to the use of a moving bed. Lee et al. [16] observed that the rate of membrane fouling of the attached growth system was about 7-times higher than that of the suspended growth system, despite similar characteristics of the soluble fraction from the two reactors as a consequence of a better filtration performance with suspended growth; this was explained by the formation of dynamic membranes with suspended solids. Yang et al. [38] detected a rate of membrane fouling with moving bed that was three times higher than the one observed in conventional MBR. The thick and dense cake layer formed on the membrane surface was speculated to be caused by the filamentous bacteria in the moving bed membrane bioreactor [39].

4. Conclusion

Given the results obtained in a hybrid Moving Bed Membrane Bioreactor pilot plant treating urban wastewater in real conditions (MLSS between 1721 ± 795 and 4698 ± 251 mg/L and BFSS between 1492 ± 462 and 6608 ± 989 mg/L of carrier; temperatures between 5.50 ± 2.17 and 28.24 ± 1.48 °C; regimes of 18.96 and 45.50 L/h flux; and three filling ratios (20, 35 and 50%)), the following conclusions were made:

- The operative variables with more influence on the performance of the membrane were flux and temperature.
- Although the mechanisms of fouling are complex, it increases when MLSS and BFSS increase and temperature decreases. The fouling rate varied between 0.26 and 1.22 kPa/day.
- The recovery of permeability after chemical cleaning ranged between 19.70 and 67.50%, being higher when the viscosity, filling ratio and flux were also higher.
- The permeability before chemical cleaning showed two different ranges in relation to the flux modified by the effect of the other operative variables, increasing with filling ratio, viscosity and BFSS.

Considering the above, the MLSS reduction in a hybrid MB-MBR with respect to conventional MBR increased the fouling rate of the membrane, possibly as a consequence of the sources and characteristics of the global biomass in the system with a worse filtration performance; this could be explained by the formation of dynamic membranes with suspended solids or by the filamentous bacteria in the moving bed membrane bioreactor.

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