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ORIGINAL PAPER

Removal of pharmaceuticals and personal care products from domestic wastewater using rotating biological contactors

N. Delgado[1,2] · A. Navarro[3] · D. Marino[1,2] · G. A. Peñuela[4] · A. Ronco[1,2]

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Abstract

New emerging contaminants are being released into the environment. Some of these are not removed in conventional wastewater treatment plants. Therefore, the aim of this study was to evaluate a pilot-scale rotating biological contactor (RBC) as a technology applicable to the removal of emerging pollutants. The work focused on two pharmaceutical compounds in particular (carbamazepine and sildenafil citrate) and a personal care product (methylparaben). The behavior of the system was evaluated for the individual addition and mixture of the three compounds, using organic loads similar to those of secondary and tertiary treatment effluent. The working flow rate was 70 mL/min, and the concentration of the compounds was 200 µg/L. The organic load was evaluated at different times. Dissolved oxygen, temperature, conductivity and pH were measured in situ on each sampling. Removal efficiencies greater than 98% were achieved for methylparaben, but less than 20% for pharmaceuticals. The average removal efficiency was 95% for the organic matter in the different tests. In this investigation, it was observed that the RBC allowed the effective removal of emerging contaminants which have flat molecular geometry and essential chemical elements, such as methylparaben. However, its effectiveness was poor in removing contaminants formed from more complex structures such as carbamazepine and sildenafil citrate. The operational stability of the system was not affected when these compounds were incorporated in amounts similar to those found in wastewater.

Keywords Biodegradation · Biofilm · Carbamazepine · Emerging contaminants · Methylparaben · Sildenafil citrate

Introduction

The natural water cycle has an important purification capacity. However, it does not eliminate many pollutants of anthropic origin, due to their high concentration, persistence and solubility in water, several of them remaining for a long time in the environment. Those that are toxic also endanger the life of aquatic organisms. In domestic wastewater, a large variety of chemical substances that were previously not detected due to their low concentration have been quantified thanks to the development of analytical techniques making this possible (Gracia-Lor et al. 2012; Elorriaga et al. 2013; Sui et al. 2015). There are currently approximately 100,000 registered substances that are potentially dangerous, in addition to those new synthetic compounds that periodically enter the market, among which are therapeutic drugs, veterinary drugs, fragrances and cosmetics. (Petrie et al. 2015). The substances of greatest concern due to their potential toxicity include: antibiotics, estrogen steroids, antidepressants, calcium channel blockers, genotoxic and antiepileptic drugs. The last of these include phenytoin, valproate and carbamazepine, which are capable of initiating apoptosis in brains in development. Also in this category are parabens, which are antimicrobial preservatives used in cosmetics, toiletries, pharmaceuticals and even foodstuffs. Although the acute toxicity of these compounds is very low, there is data from literature showing a slight estrogenic activity (Stuart et al. 2012; Gavrilescu et al. 2015; Li et al. 2015b). It is

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✉ D. Marino
damianm@quimica.unlp.edu.ar

1 Centro de Investigaciones del Medio Ambiente, Facultad de Ciencias Exactas, Universidad Nacional de la Plata (UNLP), 47 y 115, 1900 La Plata, Argentina

2 Consejo Nacional de Investigaciones Científicas y Técnicas, La Plata, Argentina

3 Departamento de Ingeniería Química, Facultad de Ingeniería, UNLP, 47 y 115, 1900 La Plata, Argentina

4 Grupo GDCON, Universidad de Antioquia, Calle 70 No. 52-21, Medellín, Colombia

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for this reason that pollution of the environment by urban discharges is of growing concern due to the many risks that may arise, especially since many studies have shown that conventional treatment plants do little to remove some of these pollutants (Boyd et al. 2003; Stackelberg et al. 2004).

Currently, alternative treatment procedures are being studied. These include adsorption, advanced oxidation processes, alternative biological treatments and ultraviolet radiation. Biosorption involves the accumulation of contaminants by biological material either by metabolic or purely physical–chemical methods (Costley and Wallis 2001). Unlike physical and chemical treatments, biosorption generally does not imply high operating costs. In particular, solid wastes generated during treatment are concentrated in smaller volumes, which are easily treated (Bhide et al. 1996).

It has been demonstrated that rotating biological contactors (RBC) are effective processes to treat complex wastewater compared to other bioreactors (Pakshirajan and Kheria 2012). This is especially so because they offer a high surface area generated in the rotating disk, establishing good contact between the microbial species and the contaminants in the system. It is important to note that in biofilm processes such as RBC, the mean retention time of the microbial cells is decoupled from the hydraulic retention time, allowing higher organic loads and providing greater resistance to toxic shocks than suspension culture (Hassard et al. 2015). This is also an attractive form of technology due to its operational characteristics such as short hydraulic retention times, simple operation and design, high treatment efficiencies and low-energy requirements (Duque et al. 2011). In addition, it is important to emphasize that currently, in terms of treatment performance, operation and management costs, the combination of RBC with physical filtration and disinfection is considered to be the most economical and feasible solution to treatment and recycling gray water, when spaces are reduced (Nolde 2000; De Gisi et al. 2016).

These systems have been successfully used to treat different contaminants such as phenol, dyes and heavy metals (Yamaguchi et al. 1999; Alemzadeh and Vossoughi 2001, Malachova et al. 2013). In view of the fact that the treatment of emerging contaminants using rotating biological contactors has not been studied much (Vasiliadou et al. 2014), the present work analyzes the efficiency of these systems to remove two pharmaceuticals (carbamazepine and sildenafil) and a personal care product (methylparaben) in domestic wastewater. These compounds were chosen for their representativeness in surface water and wastewater (Gracia-Lor et al. 2012; Elorriaga et al. 2013).

This study was developed between November 2014 and November 2015, at the Center for Environmental Research (Centro de Investigaciones del Medio Ambiente-CIMA) and the Department of Chemical Engineering of the National University de La Plata.

Materials and methods

Chemical compounds

Two pharmaceuticals, carbamazepine (Cbz) and sildenafil citrate (Sil), and a personal care product—methylparaben (Mp)—were used. These were of pharmacopeia quality (Parafarm Drugstore, Saporiti, Argentina). Table 1 lists the physicochemical characteristics of the three compounds. Stock 1000 mg/L solutions were prepared by dissolving the compounds in 99.9% methanol.

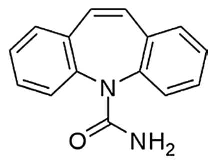
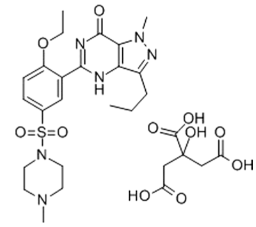
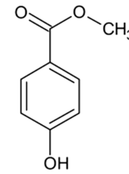
RBC experimental pilot plant system

The water used in this study was synthetic wastewater, which was prepared to simulate domestic wastewater. The characteristics of the preparation were modified from ISO 11733, as proposed by Ho and Sung (2010). Non-fat milk powder (135 mg/L), soluble starch (65 mg/L), sodium acetate (40 mg/L), yeast extract (10 mg/L), ammonium chloride (38 mg/L), Urea (21.5 mg/L) and monobasic potassium phosphate (22 mg/L) were dissolved in tap water to obtain a COD of 200 mg/L.

The inoculum was developed in batch in the laboratory under conditions of room temperature and constant stirring. 25 mL of bacterial consortium characteristic of domestic wastewater was added to 250 mL of the synthetic wastewater with a COD of 200 mg/L. This was done without exceeding 30% of the total volume of the vessel, in order to promote aeration and bacterial growth. After this culture was added to the RBC, which was maintained in batch with synthetic wastewater with the same organic load, allowing the growth of suspended biomass and its subsequent adherence on the disks. Daily purges of 1 L were carried out, and simultaneously, fresh synthetic wastewater was added to the reactor. With this stage concluded, a continuous flow rate of 70 ml/min and a COD of 100 mg/L were applied until a constant biofilm was obtained on the disks. Bacterial suspension development was determined by a spectrophotometer (UV-1203 UV/Vis) at a wavelength of 600 nm (Hazan et al. 2012).

The pilot-scale RBC (Fig. 1) consisted of twenty acrylic disks of rough surface (2 mm thick; 300 mm diameter; spaced 30 mm apart; 40% disk submergence, 1.5 rpm rotation). These were connected by a central axis of stainless steel that crossed them longitudinally, and contained in an acrylic tank of 20 L, which maintained their profile. The system was completed with a peristaltic pump used to load

Table 1 Physicochemical properties of the emergent contaminants selected

Properties	Carbamazepine	Sildenafil citrate	Methylparaben
Molecular structure			
Cs	$C_{15}H_{12}N_2O$	$C_{28}H_{38}N_6O_{11}S$	$C_8H_8O_3$
CAS	298-46-4	171599-83-0	99-76-3
M_w (g/mol)	236.09	666.70	152.05
W_s (mg/L) a 25 °C	18	3500	2500
pK_a	< 2.3; > 13.9	4.0; 8.8	8.4
Log K_{ow}	2.45	2.75	1.96
Log K_{oc}	2.7	3.5	2.4
H (atm m^3/mol)	1.08×10^{-10}	7.2×10^{-21}	2.23×10^{-9}

Cs, chemical structure; CAS, chemical abstracts service registry number; M_w, molar weight; W_s, water solubility; pK_a, negative logarithm of the ionization constant of an acid; Log K_{ow}, octanol–water partition coefficient; Log K_{oc}: soil organic carbon–water partitioning coefficient; H, Henry constant

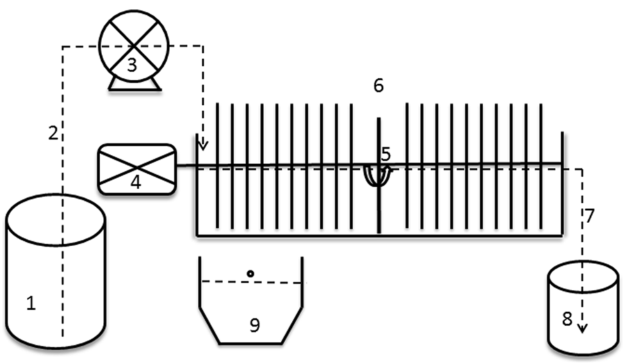


Fig. 1 Pilot-scale RBC: (1) 200 L tank containing the synthetic wastewater and place of incorporation of PPCPs; (2) affluent line; (3) peristaltic pump; (4) gear motor; (5) central axis; (6) RBC (tank and disks); (7) effluent line; (8) treated-water collector tank; (9) Transverse Section of the reactor

the effluent, which was contained in a feed tank of 200 L capacity (see Table 2).

The RBC system was evaluated without bacteria with the intention of determining the interaction of the pharmaceuticals and the personal care products (PPCPs) with the materials of the reactor. For this purpose, concentrations of 5 mg/L were diluted in distilled water with 0.01% sodium azide (used as an antimicrobial agent) (Rossner et al. 2009). Samplings for every solution were analyzed before being placed in contact with the reactor and 24 h after contact. These were determined by spectrophotometry with wavelengths of 285, 292 and 256 nm for Cbz, Sil y Mp, respectively. These samples were also evaluated at 24, 48 and 72 h without contact with the system to determine degradation by exposure to light.

After establishing the inoculum and maintaining the system under continuous and stable conditions, tests were carried out incorporating PPCPs both individually and in a mixture of the three compounds at concentrations of 200 µg/L. Organic loads of 300 and 30 mg/L were treated in the reactor, in order to evaluate the performance of PPCPs during the treatment of affluent with organic loads similar to those of a secondary and a tertiary treatment plant. The monitoring of the system was carried out both on the affluent and effluent, with sampling every 5 h during the first day in accordance with the hydraulic retention time, and 24 h subsequently until 168 h elapsed. The parameters measured to determine the water quality were pH, temperature, dissolved oxygen, electrical conductivity and COD (see Table 2). At the same time, samples to evaluate the concentrations of the incorporated PPCPs were taken and maintained at − 20 °C until the time of analysis. 0.5 cm^2 of biomass was extracted from disk 1 and disk 20, in order to determine the adsorption of PPCPs on the biofilm. These were kept at − 20 °C in 1 mL of distilled water.

The concentration of PPCPs was determined in the laboratory by HPLC–MS chromatographic analysis with a C18 CSH column with electro-spray ionization source at positive SIM mode for each compound. 0.1% formic acid and methanol were used as solvents and each characteristic ion was followed in accordance with Elorriaga et al. (2013). The same procedure was performed for the analysis of the adsorption of PPCPs on biofilm.

Table 2 Reactor design parameters, physicochemical parameters measured and experimental conditions evaluated

Reactor parameter	Dimension	Physicochemical parameters	Sampling time (h)
Number of disks	20		5[a]
Disk thickness	2 mm		10
Disk diameter	300 mm	pH	24
Spaced between disks	30 mm	Temperature	48
Disk submergence	40%	Dissolved oxygen	72
Disk rotation rate	1.5 rpm	Electrical conductivity	96
Tank volume	20 L	COD	120
Flow rate	70 mL/min		144
Feeding tank volume	200 L		168
Experimental conditions			
Test	COD (mg/L)	Addition of PPCPs	Concentration of PPCPs (μg/L)
Test 1	300	Separated	200
Test 2	300	Mixture	200
Test 3	30	Mixture	200

[a]Hydraulic retention time

Dissolved oxygen, pH, conductivity and temperature were determined in situ using a multiparameter sonde (WA-2017SD). The COD was performed according to the 5220 method proposed in the Standard Methods (APHA 1998).

Results and discussion

RBC initial development

The initial concentration of the Cbz, Sil y Mp solutions was 5.0, 4.7 and 5.1 mg/L, respectively. After 24 h in contact with the system, the respective concentrations were 5.6, 4.6 and 5.3 mg/L. Accordingly, adsorption on the reactor was not determined. Meanwhile, PPCPs were not affected by photodegradation, since the average values in the solutions containing the PPCPs exposed to light after 24, 48 and 72 h were the same as those obtained in the solutions prepared initially.

The inoculum development at laboratory-scale took 24 h. This volume was later added to the RBC. The highest density of microorganism suspension occurred on the third day by batch-operating, with the subsequent adhesion of this on the disks. Bacterial growth in the suspension was detected until the 5th day, indicating the adhesion on disks and the stability of the batch system. The continuous affluent operation was performed with an organic load of 100 mg/L, until a biofilm with a constant thickness of about 3 mm and stable conditions were obtained (Cortez et al. 2008). Dissolved oxygen was maintained between 6.2 and 5.1 mg/L, pH between 7.47 and 7.11, temperature between 14.0 and 15.7 °C, and conductivity between 1.230 and 0.911 ms/cm. Removal efficiencies above 90% were reached for COD, and a good effluent quality was achieved after 7 days of operation.

PPCPs removal

After stable conditions were obtained in the reactor, its behavior was evaluated with organic loads of 300 and 30 mg/L in the affluent. The compounds were incorporated separately and as a mixture. The experiments carried out were named as follows: *Test-1*, in which the system behavior with a COD of 300 mg/L and the addition of separate PPCPs was studied; *Test-2*, with the same organic load but with a mixture of the three PPCPs; and *Test-3* with a COD of 30 mg/L and a mixture of the compounds (see Table 2).

Cbz particular analysis

The results obtained for *tests 1, 2* and *3* are presented in graphs A1, B1 and C1 of Fig. 2. There are no significant Cbz removals for any of the tests performed. In addition, if removal efficiency is calculated for each pair of samples (affluent and effluent), negative removals of up to 60% are obtained in some cases. Similar results have been reported in several works. In 2014 for example, negative removals of up to 316% were reported for Cbz in biological treatments (Verlicchi and Zambello 2014). Other studies have shown similar removal percentages (Onesios et al. 2009; Shaver 2011). Different reasons have been proposed to explain these

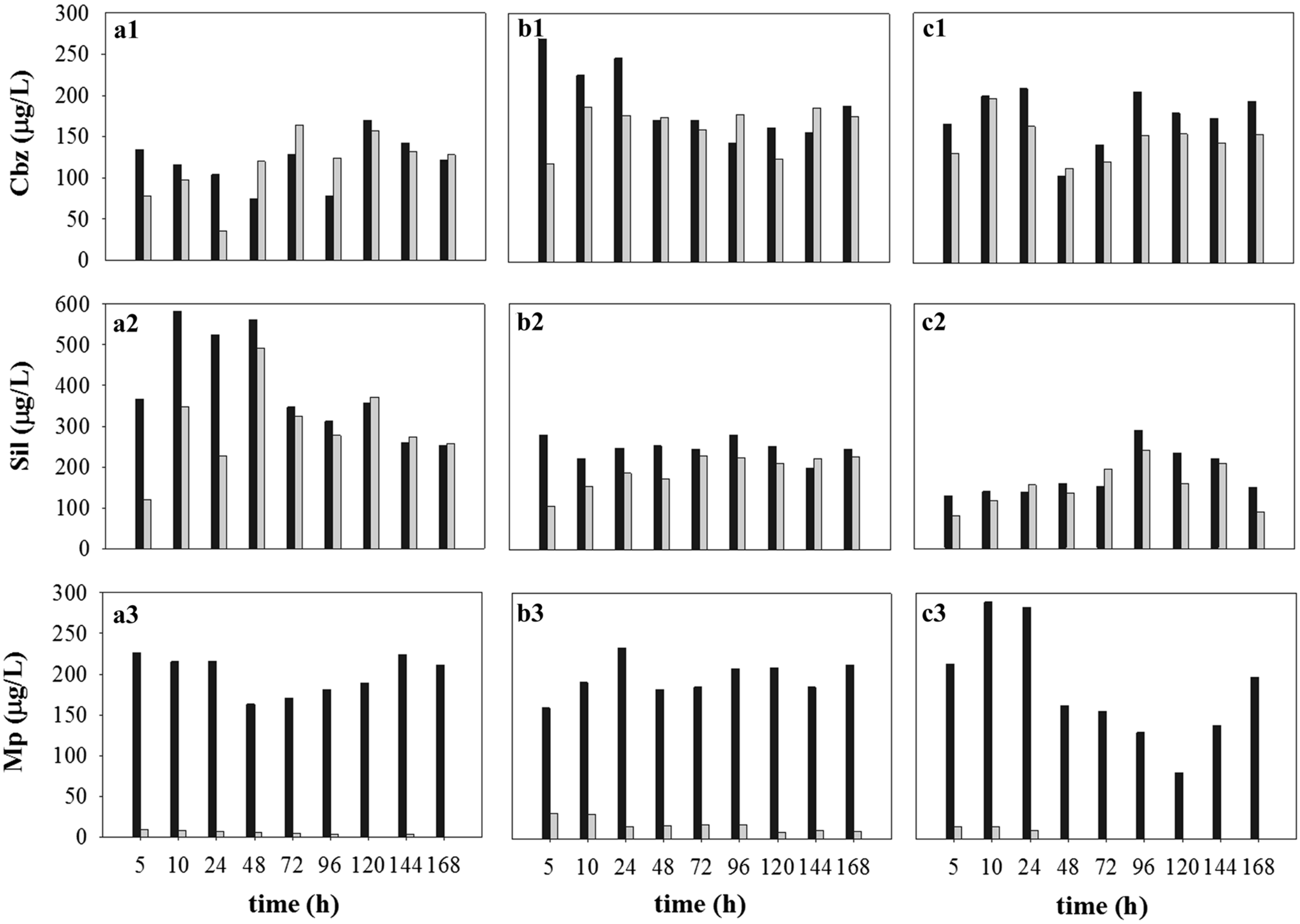


Fig. 2 Affluent (black bars) and effluent (gray bars) concentration of Cbz (1), Sil (2) and Mp (2), when an organic load of 300 mg/L is used and the compounds are incorporated individually (**a**), as a mixture (**b**); and for the mixture of the three compounds (**c**) with an affluent organic load of 30 mg/L

results. For example, the fact that PPCPs are entering wastewater treatment plants as conjugates which are then cleaved during treatment, leading to an apparent increase in concentration of the PPCPs of interest when influent and effluent concentrations are compared. In this study, however, the compounds were added directly to the synthetic wastewater, evidencing that there are other factors that generate negative values. Another cause reported is the partial evaporation of wastewater contained in the system, favoring the concentration of contaminants. However, in this work, there was no significant variation of the water level.

Majewsky et al. (2011) suggest taking into account the residence time distribution (RTD) instead of the hydraulic retention time (HRT), in which the flow rate, tank volume, mixing efficiency, concentration variability and distribution within the system are considered. For sampling times related to HRT or 24 h, which are the most common for the evaluation of treatment systems, the total xenobiotic load is not covered. Thus, samples taken every 24 h or less only work to determine removal of COD and easily biodegradable substances. The remaining fraction of COD is used as a model substance by determining RTD. Taking this consideration into account, a RTD of 72 h was determined for Cbz (see Fig. 3a). However, for this time removal, percentages of up to − 48% were obtained. Although these values were lower than those assessed with HRT, the disadvantage of negative values persisted.

Desorption of the PPCPs adsorbed in the suspended particles after the biological treatment may be another factor that affects the operation of the system. It could also affect analytical uncertainty, considering that at low concentrations instrumental errors can be great, resulting in an apparent rise in the substances studied, rather than an insignificant elimination of them during the treatment process (Shaver 2011; Verlicchi and Zambello 2014).

Therefore, the samples were analyzed using a paired *t* test based on the RTD, in order to verify whether there were statistically significant differences between concentrations in the affluent and effluent. The results were analyzed for a significance test of 0.05. Significant differences were

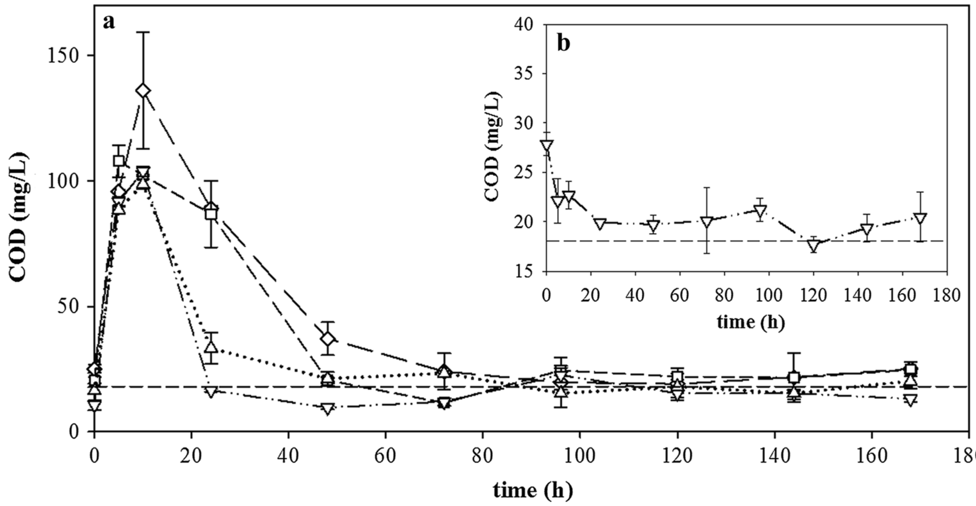


Fig. 3 The graph shows the behavior of COD effluent after incorporating Cbz (diamonds), Sil (squares) and Mp (upward triangles), both individually and when the three compounds are mixed (downward triangles) with 300 mg/L (**a**) and 30 mg/L (**b**) affluent organic loads. The constant dotted line represents the average of the remaining COD

not observed for *tests 1* and *2*, with *p* value scores of 0.795 and 0.176, respectively. Meanwhile, a *p* value of 0.004 was obtained for *test-3*, so average removal efficiency less than 20% was obtained. Similar results have been obtained for many biological treatments, in which elimination levels lower than 20 or 30% were achieved (Onesios et al. 2009; Matamoros et al. 2016). The low removal obtained for Cbz is due to its being a recalcitrant compound to bacterial degradation (Li et al. 2015a). Thus, although the organic load was reduced to 30 mg/L, significant removal efficiencies were not achieved.

Sil particular analysis

The results obtained for sildenafil citrate are listed in graphs A2, B2 and C2 in Fig. 2. When the removal efficiency for each pair of samples was analyzed taking into account the HRT, as in the case of Cbz, negative values up to − 28% were obtained.

A sampling time of 48 h was determined when the RTD was analyzed for Sil (Fig. 3a). The removals obtained of up to − 10% were inferior to those found with HRT. Significant differences were not found between affluent and effluent concentrations for *tests 1* and *3* when the paired *t* test was analyzed, with 0.058 and 0.077 *p* values, respectively. Meanwhile, for *test-2*, the *p* value was 0.018, indicating that there are statistically significant differences between the affluent and effluent concentration, with removal efficiencies less than 13%. Pilot or real-scale studies using microbial communities to remove Sil have not been reported.

Mp particular analysis

Unlike Cbz and Sil, high removal efficiencies were achieved for Mp. These results are presented in graphs A3, B3 and C3 in Fig. 2. Removal efficiencies greater than 98 and 95% were achieved for Mp and organic matter, respectively, when HRT was evaluated. As expected, in the paired *t* test, *p* values less than 0.001 were obtained. According to Majewsky et al. (2011), Mp should be an easily biodegradable compound since its removal efficiency can be determined considering only TRH. Matamoros et al. (2016) found removals of up to 69 and 74% using RBC and stabilization ponds, respectively. Thus, the reactor under study presented a 27% increase in efficiency, so is a viable option to removal Mp of the domestic wastewater containing organic loads of up to 300 mg/L.

The high removal efficiency achieved for Mp could be due to its molecular composition. It is formed only of essential chemical elements (C, H, O), which are easily degraded by microorganisms. In addition, this compound has a favorable stereochemistry (flat molecule) for its adsorption on the biofilm, easy dispersion in the wastewater (Log k_{ow} = 1.96) and adhesion on the bacterial layer considering that Log k_{oc} = 2.4. It is important to note that the pharmaceuticals present octanol–water and organic carbon–water partition coefficients similar to Mp, as shown in Table 1. However, these compounds were not efficiently removed from the system, indicating that one of its principal factors is due to its more complex molecular structure. These results are consistent with the studies reported by Kümmerer and Al-Ahmad (1997). In addition, Sil was recently reported as a recalcitrant compound (Grossberger et al. 2014), as well as carbamazepine (Li et al. 2015a), since they present chemical structures very stable and resistant to the biological attack, being therefore toxic for microorganisms. Also these compounds are formed by structural elements that rarely are in the nature. It is also known that the resistance of an aromatic compound to attack by microorganisms increases with the number of substituents it has, whatever its chemical nature. Therefore, although some studies were reported in which specific bacteria were acclimatized with these drugs as a carbon source, for periods longer than 30 days, they only found removal efficiencies below 20%, (Gauthier 2008; Grossberger et al. 2014; Kruglova et al. 2014), as well as the removals reported in the present study for a period of 168 h. However, De Felice et al. (2008) reported 99.9% removal efficiencies using specific strains on batch-laboratory test. Thus, inoculation of RBC with specific strains could be of interest in pilot-scale systems; however, it should be borne in mind that in real-scale systems, bacterial consortia coming from wastewater can easily inhibit their growth, taking its place on the surface of the disks (Pearce et al. 2003).

The impact of the PPCPs on treated water quality

High removal of organic matter was achieved during the process, with efficiencies higher than 95%. After the incorporation of the PPCPs, the system adapted progressively until reaching stable conditions, as shown in Fig. 3a. This performance could be due to the adaptation or selection of the microorganisms, which become resistant to the toxic effects of recalcitrant compounds. The time required to reach stable conditions was 48 h when sildenafil citrate and methylparaben were added. With the addition of Mp, the RBC was less disrupted. The greatest alteration occurred by incorporating Cbz, given that stable conditions were reached at 72 h. This longer time could be due to its toxic effect on the biofilm (Ferrari et al. 2003).

Similar results to those of the Mp treatment were observed when the mixture of compounds was incorporated with an affluent COD of 300 mg/L. However, stable conditions were obtained at 24 h. In this test, the system was able to stabilize rapidly, which may be due to horizontal gene transfer or a genetic drift that occurred when the biofilm was in contact with wastewater containing individual PPCPs (Singh et al. 2006). Similar behavior was presented for *test-3*, as can be seen in graph 3B, in which the system stabilizes at 24 h, confirming the adaptability of microorganisms to the compounds studied.

A correlation matrix of the coefficients and their probabilities was performed in order to analyze the effluent stability when PPCPs were added. Thus, for 9 measurements with n-2 freedom degrees, the correlation coefficient should be $r > 0.754$ using a 95% confidence interval. A good fit is indicated for p value < 0.05.

A significant positive correlation between Mp and COD removal efficiency was determined in all tests, as shown in Table 3. This correlation is because the Mp proved to be an easily biodegradable compound, being part of the organic material available. The same conclusion is suggested by Amin et al. (2010). Similar behavior was observed with the temperature and dissolved oxygen when Mp was incorporated in *test-1*. The positive correlation with temperature is due to higher temperatures increasing microbial growth and thus the degradation of Mp. In the same way, an increase in temperature entails a decrease in dissolved oxygen, displaying a negative correlation with this parameter. Significant correlations were not observed with any variables for Cbz and Sil. When an organic load of 300 mg/L is incorporated into the system, the pH has no influence on the degradation of methylparaben, while at low concentrations (30 mg/L) this parameter presents a positive correlation with Mp removal.

The dissolved oxygen was always above 2 mg/L, exceeding the recommended limit (Cortez et al. 2008). Values were between 3 and 8.7 mg/L, as can be seen in Fig. 4. A decrease in mean concentration was observed in the effluent, due to the biological stabilization of organic matter (Droste 1997). The pH remained within the recommended range for optimum development of bacteria (Pope 1978), with values between 6.5 and 8. The temperature also remained within the set range (10–32 °C) for correct

Table 3 Matrix to determine the PPCPs influence on effluent physicochemical parameters, linking the removal efficiency (%) with the probability and the (correlation coefficient)

Compounds	COD	pH	Temp. (°C)	D.O (mg/L)	Cond. (ms/cm)
Test-1 COD = 300 mg/L, analysis of each PPCP individually					
Mp	0.006 *(0.89)**	0.198	0.008 *(0.96)**	0.013 (*– 0.86)**	0.292
Cbz	NA	NA	NA	NA	NA
Sil	NA	NA	NA	NA	NA
Test-2: COD = 300 mg/L, analysis of the mixture of the 3 PPCPs					
Mp	0.003 *(0.86)**	0.382	0.884	0.983	0.941
Cbz	NA	NA	NA	NA	NA
Sil	0.805	0.102	0.852	0.295	0.263
Test-3: COD = 30 mg/L, analysis of the mixture of the 3 PPCPs					
Mp	0.002 *(0.88)**	0.008 (*0.81)**	0.434	0.327	0.633
Cbz	0.558	0.990	0.846	0.203	0.875
Sil	NA	NA	NA	NA	NA

NA Does not apply because there was no removal

*Significant cases

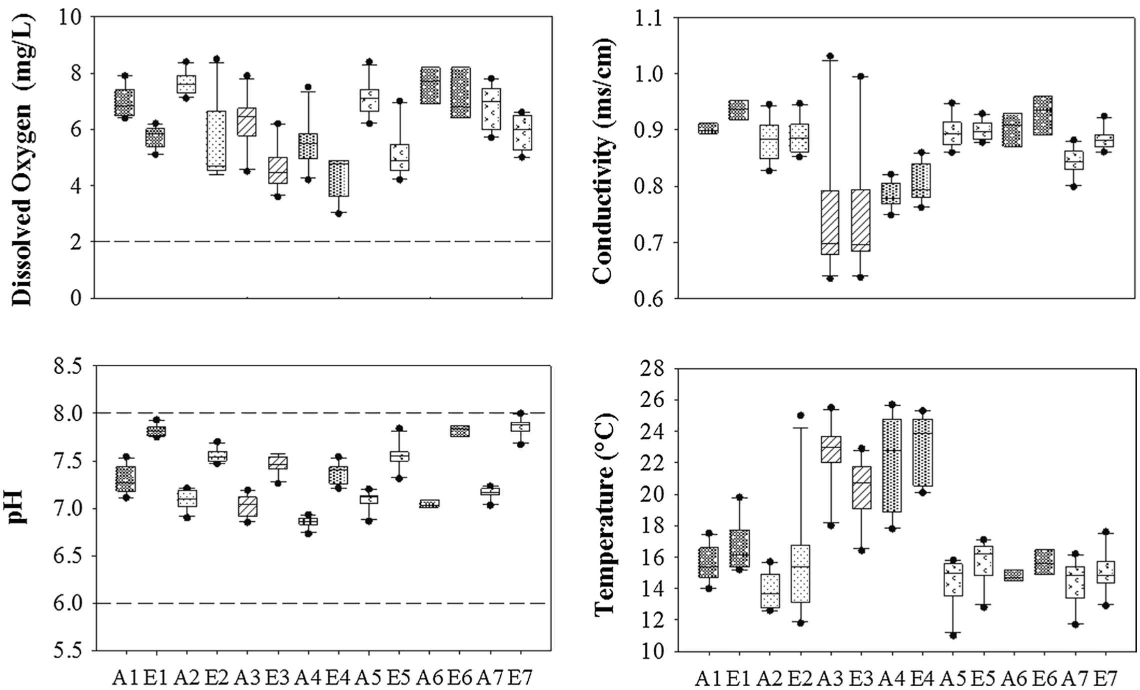


Fig. 4 Affluent (A) and effluent (E) physicochemical parameters for each test performed, with the reference limits indicated in dotted lines. Control system (1); methylparaben addition (2), carbamazepine addition (3), sildenafil addition (4) and the three compounds addition (5) with 300 m/L organic load, control system (6) and the three compounds mixture (7) with 30 mg/L COD

operation of the system (Cortez et al. 2008). This is an important factor for the RBC, because it affects the rate of the biological process. An increase in this entails an increase in microbial activity, and hence higher removal of the substrate. Large decreases can affect the biofilm stability, mainly in its initial stages (Costley and Wallis 2000). The conductivity values remained between 0.6 ms/m and 1.0 ms/cm. These results indicate that this parameter was not modified during treatment (see Fig. 4). High conductivities indicate the presence of impurities such as salts. These values are increased by domestic use of water, being situated normally in the range 1.0–2.0 ms/cm. Waters with levels below 1.2 ms/cm do not usually present any problems in the water surface (Shrestha and Kazama 2007). Therefore, it is concluded that effluent quality was not affected by the incorporation of pharmaceuticals and personal care products.

Adsorption percentage of PPCPs on biofilm

According to the results for adsorption on biofilm of *test-3*, it was determined that approximately 10% of the pharmaceuticals adhered to the biofilm on disks 1 and 20, as seen in Table 4. Mp adsorption was not determined at the different times sampled, confirming the biodegradation of this compound. Subsequently, a *t* test to compare the two groups alternately (disk 1 and disk 20 for each pharmaceutical, disk 1 and disk 20 for both pharmaceuticals) was performed. The difference in mean values was not sufficiently significant to reject the possibility that the discrepancy observed in Table 4 (Cbz and Sil) may be due to random sampling variability. The test probabilities were greater than 0.05 in all groups compared, with 95% confidence. Therefore, a statistically significant difference was not determined either between the amounts of Cbz or Sil adsorbed on disks 1 and 20 or for the amounts adsorbed of both pharmaceuticals on the same disk. Consequently, the low removals found for

Table 4 PPCPs adsorption percentage on biofilm

Time (h)	Cbz		Sil		Mp	
	Disk 1 (%)	Disk 20 (%)	Disk 1 (%)	Disk 20 (%)	Disk 1	Disk 20
24	8	5	10	7	–	–
72	10	8	17	10	–	–
168	4	6	24	11	–	–

Cbz and Sil can be attributed to the adsorption on biofilm and not to the degradation itself.

Conclusion

High removal efficiencies were obtained for methylparaben, although removals less than 20% were achieved for carbamazepine and sildenafil citrate. Therefore, in this study, it was determined that compounds with a favorable stereochemistry (flat molecule) to the adsorption on biofilm, and composed of essential chemical elements (C, H, O), such as Mp, are efficiently removed in the rotating biological contactors. However, it is necessary to investigate other processes for the removal of compounds of complex molecular structure such as the pharmaceuticals studied, because the decrease in the COD during the treatment process did not show an improvement in the removal of these compounds.

The quality of the effluent, measured through its general physicochemical parameters, was not affected by the incorporation of PPCPs in the system after conditions of stability were reached. Thus, removal percentages of up to 95% were achieved for organic matter during the different tests performed. Moreover, dissolved oxygen, temperature and pH remained within the reference limits for good system operation.

To determine the removal efficiency of complex compounds, when the rotating biological contactors are in stable conditions, the residual COD or the residence time distribution should be considered. In addition, a careful statistical analysis of the data must be taken into account to avoid erroneous results.

Rotating biological contactors have been extensively used for removal of organic matter and nutrients (nitrogen and phosphorus). However, their design has not been fully completed, and other studies such as the inoculation of specific strains should be carried out to evaluate the removal efficiency of emerging contaminants of complex molecular structure.

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