Desalination and Water Treatment
www.deswater.com

21 (2010) 122–130
September

1944-3994/1944-3986
doi: 10.5004/dwt.2010.1294

Removal of drugs of abuse from municipal wastewater using reverse osmosis membranes

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Received 6 May 2009; accepted 26 March 2010

ABSTRACT

Drugs of abuse are important emerging contaminants due to their presence in water bodies following incomplete removal in wastewater treatment. This poses a threat to drinking water sources and has led to concerns about possible health effects, both to wildlife, ecosystems and humans. In recent years, there has been growing interest in the incorporation of reverse osmosis (RO) and nanofiltration (NF) membrane technologies into existing municipal and industrial wastewater treatment facilities as a quaternary treatment option. By improving effectiveness of wastewater treatment, the exposure of firstly, nature and secondly, humans to these compounds can be minimized. It has been suggested that these membrane techniques are suitable for cost-effective desalination and the removal of a wide range of low-molecular-weight (LMW) trace organic constituents, including drugs of abuse. This paper presents the detailed results of a feasibility assessment study on the removal of selected drugs and metabolites from secondary treated wastewater. The study was carried out at a small scale RO pilot plant. Three different FILMTEC^{TM} brackish water membranes were used; low energy (LE) membrane, high rejection membrane (BW30) and extra fouling resistant membrane (XFR). The results show that RO membranes can reduce the amount of target compounds in the effluent water. The obtained mean rejection values for the three membranes and six monitoring campaigns were as follows: 74–83% (caffeine), 49–63% (nicotine), 94–96% (cotinine), 98–99% (codeine), 98% (norcodeine), 81% (METH), 57–64% (MDA), 93–96% (MDMA) and 47–57% (MDEA). No major differences were observed between the three different membranes. This indicates that the low energy RO membrane provides the same removal efficiency as the other two membranes, thus demonstrating the potential for a less energy intensive RO plant operation.

Keywords: Reverse osmosis (RO); Drugs of abuse rejection; Removal efficiency

1. Introduction

Drugs of abuse constitute a new class of emerging environmental contaminants which has been identified

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in the aquatic environment, and which has been the subject of increasing interest from water specialists in recent years. The presence of these compounds either in unaltered state or as their main human metabolites in wastewater has been reported in many countries. Moreover, they are often only partially removed by wastewater treatment plants (WWTPs) using conventional treatments [1–6]. This incomplete elimination leads to the release of these compounds in surface receiving waters [3,5,6,9–12], which thus become contaminated. Since these waters can be used for drinking water production [12], it is important that drugs are eliminated through drinking water treatments. The use of reverse osmosis (RO) has proved to be effective at removing emerging contaminants in wastewaters. Several studies and reviews of behavior and factors involving rejection of emerging contaminants such as endocrine disrupting compounds (EDCs) and pharmaceuticals during nanofiltration (NF), ultrafiltration (UF) and RO treatment processes have been published [13–18]. The majority of these studies report research performed on a laboratory scale using artificially prepared water, whilst studies devoted to evaluating emerging contaminant rejection using real waste water matrix either in pilot or full-scale RO treatments are less frequent [19,20].

The aim of this present study was to examine the ability of different types of reverse osmosis membranes to eliminate drugs of abuse from waste water. Experiments were performed in a small RO pilot plant, situated in a WWTP in NE Spain, which treats only urban waste water. The secondary effluent following biological treatment was used as the RO feed. Feed and permeate samples were studied in order to obtain rejection values. Three types of RO membranes operating in parallel were tested for caffeine, nicotine, its metabolite cotinine, codeine, its metabolite norcodeine and amphetamine type substances (ATS), such as amphetamine (AMP), methamphetamine (METH), 3,4-methylenedioxyamphetamine (MDA), 3,4-methylenedioxymethamphetamine (MDMA) and 3,4-methylenedioxyethamphetamine (MDEA). The presence and rejection of target compounds in the effluent waste water was studied seasonally.

2. Materials and methods

2.1 Studied compounds

The compounds included in this study, their metabolites and some of their chemical characteristics can be found in Table 1. With the exception of caffeine, all of the studied compounds are listed by the U.S. National Institute on Drug Abuse (NIDA) as commonly abused drugs that can have potential health consequences [21]. Although caffeine could be classified as a stimulant drug, the reason it was included in the study was that it is often considered an anthropogenic tracer for wastewater contamination of surface waters [22]. Nicotine is extensively metabolized in the human body and cotinine is its major urinary metabolite. The half life of nicotine is two hours, whereas for cotinine, it is 20 h. and it is typically detected for several days in blood and urine. For this reason, cotinine, rather than nicotine, was used as indicator to assess exposure to tobacco [23]. Moreover, cotinine is also considered a chemical marker for domestic wastewaters [24].

AMP, METH, MDA, MDMA (ecstasy) and MDEA are stimulant drugs, primarily consumed for their ability to cause euphoria and feelings of exhilaration, although AMP and METH have also been approved for restricted medical use [25]. Codeine is an opiate and narcotic pain medicine, commonly used to treat mild to moderate pain. Following ingestion, codeine metabolizes to norcodeine.

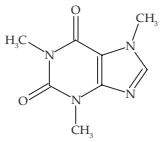
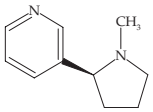
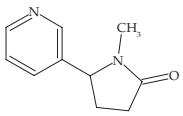
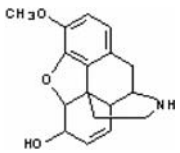
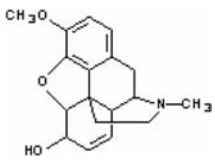
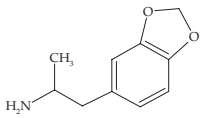
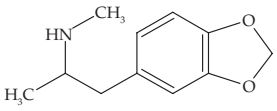
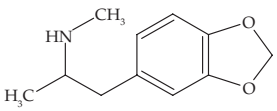
2.2. Membranes

Three different polyamide membranes were used in this study. The LE membrane is a low energy membrane widely employed for industrial and municipal[2] applications that operate at low pressure. The BW30 membrane is an industrial standard high rejection and high productivity brackish water membrane and XFR is a membrane providing advanced fouling resistance and targeted for use with waste water applications. The size of the elements was 2.5 inches in diameter and 14 inches in length, with an active membrane area of 0.54 m^2. Some characteristics of the membranes are summarized in Table 2. Molecular weight cut-off (MWCO) for each tested membrane was estimated to be around 100 Da. Three sets of new membranes were used, two sets for summer sampling and one set for the winter sampling.

2.3. Reverse osmosis pilot plant and experimental conditions

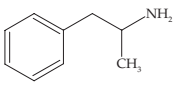
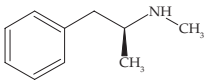
The pilot plant used in this experiment is located in a wastewater treatment plant (NE, Spain) treating municipal wastewater (16,500 m^3/d) originating from the nearby villages. The existing treatment scheme consists of a primary treatment, a secondary biological treatment and a tertiary treatment including chlorination, coagulation/flocculation, lamellar clarification and sand filtration. Following tertiary treatment, the purified water is used for rural irrigation purposes (i.e. landscaping, golf courses). In this experiment, the

Table 1
Properties of the studied compounds

Name	Structure	CAS no.	Molecular weight	pKa	Log K_{ow}
Caffeine		58-08-2	194.2	10.4	0.16
Nicotine		54-11-5	162.23	3.1	1.17
Cotinine		486-56-6	176.22	4.89	0.34
Norcodeine		467-15-02	285.34	9.23	0.69
Codeine		76-57-3	299.37	8.21	1.19
MDA		4764-17-4	179.21	8	1.82
MDMA		42542-10-9	193.24	8-10.5	2.28
MDEA		82801-81-8	207.26	8.5	2.77

(continued)

Table 1 (*continued*)

Name	Structure	CAS no.	Molecular weight	pKa	Log K_{ow}
AMP		300-62-9	135.1	10	1.76
METH		537-46-2	149.1	9.5	2.22

feed water was collected after secondary treatment and subjected to RO treatment.

The presence and rejection of the studied compounds was assessed seasonally because large variations in the population served occur during summertime. Six sampling campaigns were carried out in June and August 2008, and February, March, June and July 2009. For sample collection, 1,000 mL grab samples from the outlet of the secondary treatment (feed of RO pilot plant) and permeates from each membrane tested (LE; BW30 and XFR) were collected in amber glass bottles. Samples were stored at 4°C, filtered through glass microfiber GF/A filters (Whatman, UK) and analyzed within a week.

2.4. Analytical standards and reagents

Standard solutions (1 mg/mL in methanol) of AMP, METH, MDA, MDMA, MDEA, caffeine, codeine, norcodeine, nicotine and cotinine were used. Standard solutions of each compound and deuterated analogues, AMP-d_8, METH-d_9, MDA-d_5, MDMA-d_5, MDEA-d_5, $^{13}C_3$-caffeine, codeine-d_6, nicotine-d_4 and cotinine-d_3 were purchased from Cerilliant (Austin, TX, USA).

2.5. Analytical methodology

The target compounds were analyzed following a previously published method [26] based on solid-phase extraction ultra-performance liquid chromatography-tandem mass spectrometry (SPE-UPLC/MS/MS). Briefly, 100 mL of water spiked with deuterated standards (100 ng/L) was enriched by SPE on Oasis-HLB cartridges (Waters, Milford, USA). Cartridges were washed with 5% methanol aqueous solution, dried with nitrogen and eluted using methanol.

Compounds were analyzed by UPLC-MS/MS. An Acquity BEH C_{18} column (100 mm × 2.1 mm, 1.7 μm) and solvent A: acetonitrile with 0.1% formic acid and solvent B: 30 mM formic acid/ammonium formate (pH 3.5), were used for optimum separation of target compounds. The UPLC was coupled to a Quattro Micro triple quadruple mass spectrometer (Waters) operating in positive electrospray ionization mode. Acquisition was performed in selected reaction monitoring (SRM) mode, where the protonated molecular ion of each compound was the precursor ion. Two transitions per compound were employed as required by the EC [27].

The quantification and confirmation transitions have been published elsewhere [26,28]. Run-to-run ($n = 6$ in one day) and day-to-day precisions ($n = 6$ in five days) were evaluated using a drug-free wastewater matrix spiked at 1.5 μg/L for nicotine and cotinine and 80 ng/L for the rest of compounds. The results obtained, calculated as %RSD (relative standard deviation), were lower than 6% and 10%, respectively.

Table 2
Characteristics of the membranes used in this study

Name	Manufacturer	Material	Stabilized NaCl rejection (%)*	Permeate Flow rate (m^3/h)*	MWCO (Dalton)
LE	Dow	Polyamide	99.3	0.64	~100
BW30	Dow	Polyamide	99.5	0.58	~100
XFR	Dow	Polyamide	99.65	0.63	~100

*Stabilized salt rejection (NaCl) and permeate flow are specified under the following conditions: 2,000 ppm NaCl, 25°C, 5% recovery and pressures of 10.3 bar (LE) and 15.5 bar (BW30 and XFR).

Table 3
Feed water characteristics and experimental conditions during the sampling campaigns

Parameter	June 08	August 08	February 09	March 09	June 09	July 09
Temperature (°C)	24	28	17	18	26	29
pH	7.3	7.4	7.4	7.1	7.2	7.4
Conductivity (μS/cm)	2160	2042	2515	2041	2105	2260
Permeate flow[a] (L/h)	6.2/6.8/6.5	7.6/8.1/8.1	8.9/9.1/9.1	6.5/6.9/6.7	6.9/9.9/9.5	8.9/7.5/9.3
Recovery[a] (%)	4/4/4	4/5/3	4/5/5	4/4/4	4/5/6	5/5/5
Feed pressure (bar)	8.6	7.4	9.0	9.0	8.0	7.4
Salt rejection[a] (%)	97.5/97.5/98.5	97.5/97.5/98.1	99.2/99.4/99.4	98.4/98.7/98.7	97.9/98.5/98.8	97.7/97.6/98.0

[a] LE/BW30/XFR membranes.

Recoveries calculated by spiking a blank wastewater ($n = 6$) with the analytes and the deuterated analogs ranged from 70% (AMP) to 101% (MDEA). Limits of quantification (LOQs) ranged from 0.2 ng/L (MDEA) to 5 ng/L (codeine).

3. Results and discussion

3.1. Occurrence of studied compounds in the WWTP

Table 4 summarizes the concentration ranges, mean concentrations and LOQs for each compound in both feed and permeates obtained with the three membranes tested during the six monitoring campaigns. All studied compounds except MDEA, AMP and METH were continuously detected in the secondary treated waste water, indicating that the conventional biological process alone is not sufficient to remove them from waste streams. Some of the studied compounds showed a clear increase during the summer season, which can be related to the fact that the region is a tourist destination where the population (and thus the waste water load) increases significantly during the holiday period.

Caffeine was found in high concentrations in feed samples (following the secondary treatment), with levels ranging from 75 to 4106 ng/L. As with some other compounds, higher concentrations were observed during the summertime. These values are lower than those found in a survey of 42 WWTPs in NE Spain, where median concentrations were 54 and 1.3 μg/L in wastewater influents and effluents respectively, and concentrations ranged from 700 ng/L to 209 μg/L (influents), and 30 ng/l–44 μg/L (effluents) [4]. Caffeine is efficiently removed in conventional WWTPs with values usually higher than 95% [4,22,29].

Nicotine and its metabolite cotinine were found in all campaigns. Concentrations in the feed water ranging from 42 to 203 ng/L for nicotine, and 69 to 239 ng/L for cotinine were measured. Cotinine was found at higher concentrations than nicotine in five of the six campaigns, and their presence in the treated waste water was fairly stable throughout the

Table 4
Concentrations (ng/L) of drugs of abuse (maximum, minimum, mean and median) in the RO feed and permeates of LE, BW30 and XFR membranes (n=6)

	LOQs	Feed	Permeate LE	Permeate BW30	Permeate XFR
Compound	ng/L	Min-Max (Mean/Median)	Min-Max (Mean/Median)	Min-Max (Mean/Median)	Min-Max (Mean/Median)
Caffeine	2.0	74.7-4106 (1056/569)	13.2-130 (49.1/39.5)	16.7-201 (67.0/47.7)	13.6-132 (56.3/40.5)
Nicotine	1.0	42.5-203 (79.0/57.3)	16.1-35.0 (27.6/31.2)	17.1-30.7 (23.3/22.5)	17.0-47.3 (31.1/33.7)
Cotinine	1.5	69.2-239 (120//105)	1.66-13.5 (6.62/6.12)	1.64-8.74 (4.93/3.54)	2.38-10.3 (6.48/6.51)
Norcodeine	2.1	8.26-79.9 (53.5/65.5)	<LOQ	<LOQ	<LOQ
Codeine	5.0	275-1201 (750/727)	<LOQ-9.30 (<5.0)	<LOQ	<LOQ-5.99 (<5.0)
MDA	0.8	3.33-28.4 (11.3/7.21)	1.23-10.5 (4.71/2.86)	0.88-10.3 (4.41/2.38)	0.76-10.0 (4.52/2.76)
MDMA	0.3	5.51-97.0 (29.8/21.8)	<LOQ-4.32 (1.19/0.74)	<LOQ-2.38 (0.87/0.49)	<LOQ-1.81 (0.72/0.30)
MDEA	0.2	<LOQ-1.10 (0.63/0.70)	<LOQ-1.00 (0.38/0.20)	<LOQ-0.80 (0.28/0.20)	<LOQ-0.90 (0.38/0.20)
AMP	1.0	<LOQ-5.08 (1.31/<1.0)	<LOQ	<LOQ	<LOQ
METH	0.7	<LOQ-4.15 (1.63/<0.7)	<LOQ	<LOQ	<LOQ

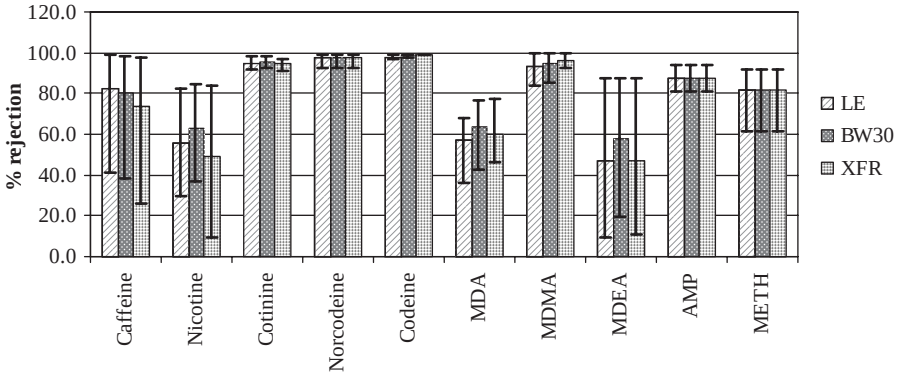


Fig. 1. Rejection efficiencies (%) of drugs of abuse in wastewater by three RO membranes (n=6).

campaigns, with a marked increase in cotinine in August 2008. Significant elimination of cotinine occurs during conventional wastewater treatment. For instance, in the NE of Spain elimination values of between 70% and 99% has been reported [4], and similar efficiencies (90–99%) were achieved in Swiss WWTPs with cotinine concentration ranges in treated wastewater varying from ~10 to 600 ng/L [24].

Codeine and norcodeine were also found in all the six sampling campaigns. Concentrations in feed water ranged from 275 to 1200 ng/L for codeine and 8.3 to 71.2 ng/L for norcodeine. These values are higher than levels measured in a survey of 15 WWTPs in the same region with median values of 69 and 39 ng/L for codeine and 6 and 2.5 ng/L for norcodeine in influents and effluents, respectively [28]. The poor average elimination of codeine (~ 33%) in conventional WWTPs is explained by the cleavage of codeine conjugates to free codeine during wastewater treatment. Median codeine levels of 40, 54, 193 and 220 ng/L were also measured in WWTPs influents in eastern Spain, in Italy and in Germany [9,30,31], with elimination efficiencies ranging from 12% to 100%.

Amphetamine and methamphetamine were found in only two and three feed samples respectively at very low concentrations of up to 5.1 ng/L for AMP and 4.2 ng/L for METH. In contrast, the other three amphetamine type substances were regularly found in all feed samples with concentration values ranging from 3.3 to 28.4 ng/L for MDA and from 5.5 to 97 ng/L for MDMA. Ecstasy (MDEA) was present in almost all samples at very low concentrations with a maximum value of 1.1 ng/L. In general, the highest concentrations were found during the summer. Concentration levels of ATS in different wastewater influents and effluents have been reviewed recently [32], and amphetamine was the most prevalent, with concentrations of up to 5236 ng/L [33]. The average removal of ATS has been estimated to be in the range of 40–99% in WWTPs from NE Spain [4] and 57% in wastewaters located on the Ebro River (Spain) [34].

3.2. Reverse osmosis rejection efficiencies of drugs of abuse.

Concentrations of the studied compounds in the RO permeate samples during six sampling campaigns are summarized in Table 4. The rejection efficiency was calculated as shown below:

$$\text{Rejection} = \left(1 - \frac{C_p}{C_f}\right) * 100, \quad (1)$$

where C_p is the concentration in the permeate and C_f is the concentration in the feed.

When the concentrations of the studied compounds in the permeate samples were below the quantification limits (Table 4), rejection was calculated by considering permeate concentration as half the LOQ. Fig. 1 summarizes the rejection efficiencies of the target compounds by each membrane type (LE, BW30 and XFR). The rejection of the different compounds is discussed separately below, but some general trends were observed for all the compounds: (a) In general, the obtained rejection values are high, indicating that reverse osmosis is a suitable technology for removing drug contaminants from waste streams; (b) No major differences between the three membranes were observed, which is important since this implies that low energy membranes can be used for these compounds.

Mean rejection values in the range of 74–83% were found for caffeine in the three tested RO membranes with no apparent significant differences between them. Caffeine was eliminated by RO in four campaigns very well (mean 94%), but in March 09 and June 09

significantly lower rejections were obtained (55–70%). Rejection values for caffeine are in line with previously published values for BW30 membrane [35] and are better than those measured for cellulose acetate membrane SC-3100 (44%) and XLE (70%) [18], when measured in artificial solutions.

Nicotine was the only compound in which a significant rejection difference among the three membranes was observed. Mean nicotine rejection was 49%, 56% and 63% for XFR, LE and BW30, respectively. Moreover, a high variability between different samplings was observed, with rejections varying from 9% to 85%. The lowest rejections were obtained during the winter samplings (mean rejection 32% in February and March), whereas rejection improved significantly (55–85%) in summer campaigns. For cotinine, a very high rejection ($\sim$95%) was obtained in all campaigns and for the different membranes tested. These eliminations are similar to, or even better than those measured in conventional wastewater treatments [4,24].

Codeine and norcodeine were not usually detected in the studied permeate samples, even when they were present at high concentrations in the RO inlet water (Table 4), indicating a very high elimination percentage by RO membranes (>98%) for both compounds and all tested membranes during the six campaigns. These rejection values are significantly better than the elimination percentages published in the literature, which ranged from 12% to 100% [9,28,30,31].

The elimination percentages of amphetamine type compounds are shown in Fig. 1. The best results were found for MDMA, for which the rejection efficiency was from 93% to 96% without showing any difference between the observed rejection and the membrane type or season. These values compare favorably with elimination ranges of MDMA in WWTPs in the same region, which ranged from 50% to 99% [4]. In contrast, MDA and MDEA showed lower rejection values. For MDA, rejection was around 57–63% for the six campaigns with no apparent difference between membranes. As the concentration levels in the feed were generally very low ($\leq$ 1.1 ng/l), the rejection values for MDEA can only be roughly estimated (47–57%). Moreover, large variations of between 9% and 88% in rejection values measured in the six campaigns were observed, since low concentrations close to the LOQ affect the accuracy of the results. Methamphetamine (METH) was present in the three feed waters at low concentrations and was not detected in any of the tested permeate samples. AMP was only present in two feeds (February 09 and July 09) but, as with METH, was not detected in any of the tested permeate samples. For both compounds, relatively high rejection efficiencies (80–85%) were calculated.

3.3. Relationship between rejection and physical/chemical properties of target compounds.

Several factors can affect the solute rejection of a RO membrane. The three most important ones are the membrane properties (separation layer, pore size, topology, membrane surface charge, hydrophilicity), the characteristics of the feed water (chemical composition, concentration, pH, charge, molecular size, polarity and salt diffusivity) and operation conditions (temperature, pressure, recovery and flux). In practice, rejection always depends on all these factors. Whereas the rejection of inorganic ions can be well modeled and simulated with commercial simulation programs, the rejection of organic compounds is more difficult to model. Membrane manufacturers often use stabilized salt rejection to predict solute passage, but this cannot be directly related to rejection of organic molecules. Kimura et al. concluded in their research that salt passage could only be used as a rough estimation [18].

For organic molecules, especially uncharged compounds, the characteristic term most commonly used is their molecular weight, which can be related to transport through the membrane if it is assumed that this is mainly due to size exclusion. In our case, the molecular weight cut off (MWCO) of the tested membranes was approximately 100. Although all studied compounds in this study are larger than 100, rejection efficiencies higher than 90% were only observed for caffeine, cotinine, norcodeine, codeine and MDMA. Moreover, no relationship between mean rejection values and the three membranes or the molecular weight of the compounds, as previously observed by other authors, was obtained [18,36], indicating that other compound characteristics are more significant. Some researchers have used the polarity of the organic compounds to predict rejection [16,18], assuming that when hydrophobicity (log K_{ow}) increases, rejection also increases [37]. In our case, no significant correlation between them was observed.

It should be borne in mind that some of these compounds are ionized at the working pH (7.1–7.4). For charged compounds, the electrostatic and repulsive forces between the compound and the membrane surface must be considered, since these are the decisive factor affecting rejection. Therefore, all the properties mentioned above must be taken into account in order to explain the rejection of the compounds by RO membranes. In this context, Verliefde et al. [16] proposed eight categories to qualitatively predict the rejection of organic pollutants based on a scheme previously proposed by Bellona et al. [14] but using hydrophobicity as the primary solute parameter and also including molecular weight (MW) and pK_a values. According to

this scheme, caffeine, norcodeine, codeine, amphetamine and MDA belong to category 2 (log K_{ow} < 2; MW > MWCO; pK_a > pH), which comprises compounds with a steric hindrance effect as the rejection mechanism and a qualitative rejection prediction ranging from moderate to high. The obtained rejection values from (~60%) for MDA to 98% (norcodeine and codeine) agreed with the qualitative prediction. The group of compounds, MDMA, MDEA and METH belongs to category 4 (log K_{ow} > 2; MW > MWCO; pK_a > pH) for which hydrophobic interactions are the main rejection mechanisms, providing moderate qualitative rejection prediction values. The obtained values ranged from 47% (MDEA) to 93% (MDMA). The last group of compounds, formed by nicotine and cotinine (log K_{ow} < 2; MW > MWCO; pK_a < pH), belongs to category 6. For this group of compounds, charge repulsion is the main rejection mechanism and a high qualitative rejection is predicted. The value obtained for cotinine (~95% rejection) is in agreement with this prediction, although for nicotine a low rejection value was obtained (~55%). Nevertheless the qualitative predictions obtained using this approach are roughly in agreement with the observed values for the compounds studied.

4. Conclusions

This paper presents the results obtained from the study of RO membrane efficacy in removal of selected drugs and their metabolites, including caffeine, nicotine, cotinine, codeine, norcodeine and amphetamine type compounds such as amphetamine, methamphetamine, 3,4-methylenedioxyamphetamine (MDA), 3,4-methylenedioxymethamphetamine (MDMA) and 3,4-methylenedioxyethamphetamine (MDEA) from secondary treated wastewater.

The results show that RO membranes can significantly reduce the amount of the target compounds in the effluent water. The rejection values were as follows: 74–83% (caffeine), 49–63% (nicotine), 94–96% (cotinine), 98–99% (codeine), 98% (norcodeine), 81% (METH), 57–64% (MDA), 93–95% (MDMA) and 47–58% (MDEA). It has been shown that by taking certain physical and chemical properties into account, such as hydrophobicity, size and pKa values, the removal of organic compounds by RO can be roughly estimated using the models generated with laboratory studies.

The three evaluated membranes (LE, BW30 and XFR) provided similar removal efficiencies, supporting the use of the low energy RO membrane for the elimination of drugs of abuse by reverse osmosis with the advantage of making the operation of a RO plant less energy intensive. By incorporating a reverse osmosis unit into existing municipal waste water treatment facilities, the exposure of firstly, nature and secondly, humans to drugs of abuse can be minimized.

Notes

1. FILMTEC™ is a trademark of Filmtec.
2. The use of FILMTEC™ membranes for municipal applications is dependent upon local regulations.

Acknowledgements

This study was carried out within the framework of the SOSTAQUA project, led by AGBAR, aimed at developing technologies promoting sustainability of urban water cycle. The authors would like to acknowledge the support and funding received from the Spanish Government through the CDTI (Centro para el Desarrollo Tecnológico Industrial) under the INGENIO 2010 program, and the Ministerio de Ciencia y Tecnología (CTM2006-00753/TECNO). This study was also partially financed by R+i Alliance, a company established in France to select, fund and coordinate the execution of joint research, development and innovation projects carried out by the companies Lyonnaise des Eaux, United Water, AGBAR Group, Northumbrian Water and Suez Environment. The authors would also like to acknowledge the staff at Sorea (member of AGBAR group) WWTP for their contribution to the practical execution of the study as well as ACA (Agencia Catalana de l'Aigua) for authorizing the pilot plant installation at their premises.

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