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Removal and fate of micropollutants in a sponge-based moving bed bioreactor

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Keywords

bed, moving, bioreactor, sponge, removal, micropollutants, fate

Disciplines

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Abstract

This study investigated the removal of micropollutants using polyurethane sponge as attached-growth carrier. Batch experiments demonstrated that micropollutants could adsorb to non-acclimatized sponge cubes to varying extents. Acclimatized sponge showed significantly enhanced removal of some less hydrophobic compounds (log D < 2.5), such as ibuprofen, acetaminophen, naproxen, and estriol, as compared with non-acclimatized sponge. The results for bench-scale sponge-based moving bed bioreactor (MBBR) system elucidated compound-specific variation in removal, ranging from 25.9% (carbamazepine) to 96.8% (β-Estradiol 17-acetate) on average. In the MBBR system, biodegradation served as a major removal pathway for most compounds. However, sorption to sludge phase was also a notable removal mechanism of some persistent micropollutants. Particularly, carbamazepine, ketoprofen and pentachlorophenol were

found at high concentrations (7.87, 6.05 and 5.55 µg/g, respectively) on suspended biosolids. As a whole, the effectiveness of MBBR for micropollutant removal was comparable with those of activated sludge processes and MBRs.

Keywords: Micropollutants removal; Moving bed bioreactor (MBBR); Sponge; Attached-growth; Biodegradation.

1. Introduction

Micropollutants have been identified as an emerging group of contaminants which are frequently detected in the aquatic environment (Huerta-Fontela et al., 2011; Luo et al., 2014). They originate mainly from mass produced consumables used for medical care (pharmaceuticals), hygienic or cosmetic reasons (personal care products, PCPs), plant/crop protection (pesticides), and enhancement of the physical properties and performance of products (industrial chemicals). In addition, some micropollutants (e.g., steroid hormones) are of natural origin. The concentrations of micropollutants in raw sewage commonly range from a few ng/L to several µg/L, which essentially distinguish them from traditional contaminants (organic matter, nitrogen and phosphorus). Despite the low concentrations, micropollutants have been associated with a number of negative effects, such as toxicity and endocrine disrupting effects on aquatic organisms and antibiotic resistance of microorganisms (Baquero et al., 2008; Fent et al., 2006).

Treated effluent discharge from wastewater treatment plants (WWTPs) is an important pathway for micropollutants entering the aquatic environment. Hence, wastewater treatment is a vital barrier against the release of micropollutants. However,

due to the low concentrations and diverse physicochemical properties, micropollutants often experience inadequate removal during wastewater treatment. Consequently, many of these micropollutants, especially those that are polar and recalcitrant, can pass through the WWTPs and end up in the environment, thus posing harm to wildlife and humans. To yield a further reduction of micropollutant discharge, optimization and/or upgrade of conventional wastewater treatment processes (e.g. activated sludge) is of immense significance. Advanced treatments, such as activated carbon adsorption (Nguyen et al., 2013), membrane filtration (Yangali-Quintanilla et al., 2011), membrane bioreactor (MBR) treatment (Wijekoon et al., 2013), ozonation and advanced oxidation processes (De la Cruz et al., 2012), are able to significantly remove micropollutants from wastewater. Nevertheless, these technologies are usually associated with high operation cost and intractable problems, such as concentrated residues and membrane fouling in case of membrane-based techniques and formation of toxic by-products during advanced oxidation processes.

As promising alternatives to suspended-growth activated sludge processes, attached-growth processes are effective techniques for wastewater treatment, as they offer a number of advantages over activated sludge processes. One major advantage is that attached-growth processes facilitate the growth of slow-growing microorganisms, which are important for the removal of some micropollutants (Falås et al., 2012; Guo et al., 2012). Moreover, if controlled properly, attached-growth processes can have different redox conditions within the biofilm. The coexistence of oxic and anoxic conditions can not only facilitate nutrient removal, but also enhance the elimination of a wider spectrum of micropollutants. For instance, oxic condition improves the removal

of naproxen, ethinylestradiol, roxithromycin and erythromycin, while anoxic condition aids the degradation of carbamazepine, clofibric acid, diclofenac and iodinated X-ray contrast media (Drewes et al., 2001; Suárez et al., 2010; Zwiener and Frimmel, 2003).

The moving bed bioreactor (MBBR) is a simple yet effective and compact technology developed on the basis of attached-growth principle. In spite of their excellent performance in eliminating traditional contaminants (e.g., organic matter and nutrients), there is a limited amount of research on micropollutant removal in MBBR. Nonetheless, the results from some bench-scale or pilot-scale studies demonstrate MBBR as a potential technique for eliminating micropollutants from wastewater. Falås et al. (2012) compared the micropollutant removal in batch experiments with activated sludge and suspended biofilm carriers (K1, AnoxKaldnes). Although biofilm carriers and activated sludge exhibited similar removal rate constants for ibuprofen (around 2–5 L/g biomass·d) and naproxen (around 0.5–1 L/g biomass·d), significantly higher rate constants for diclofenac, ketoprofen, gemfibrozil, clofibric acid and mefenamic were found in the reactors with carriers (0.06–0.38, 0.9–3.6, 0.6–2.1, 0.05–0.17 and 0.08–0.48 L/g biomass·d, respectively) as compared with activated sludge (0–0.02, 0.01–0.32, 0.01–0.27, 0–0.04 and 0–0.06 L/g biomass·d, respectively). Their subsequent study (Falås et al., 2013) confirmed that a reactor with biofilm carriers (Bio-film Chip M, AnoxKaldnes) achieved rapid removals of diclofenac (1.3–1.7 L/g biomass·d) and trimethoprim (1.0–3.3 L/g biomass·d), while the elimination of both compounds in the suspended-growth sludge reactors was insignificant (≤0.1 L/g biomass·d).

Apart from the plastic biofilm carriers mentioned above, polyurethane sponge can also be considered as an ideal material for attached-growth microorganisms. Previous studies have proved the suitability of sponge-based MBBRs in removing organic matter, nitrogen and phosphorus (Chu et al., 2011; Ngo et al., 2008). However, to date, no effort has been directed towards investigating micropollutant removal by MBBR using sponge as attached-growth carrier. Therefore, this study aims to investigate the effectiveness of a sponge-based MBBR for removing five groups of micropollutants. Batch experiments using non-acclimatized and acclimatized sponge were conducted initially to evaluate short-term removal rates of the selected micropollutants. Subsequently, a continuous bench-scale MBBR was set up for a long-term assessment of micropollutant removal. The mass balance for each compound was also calculated to provide insight into its fate in the MBBR system.

2. Methods

2.1. Synthetic wastewater and sponge

A synthetic wastewater (simulating medium strength municipal wastewater primary effluent) spiked with micropollutants was used in this study. The synthetic wastewater contained chemical oxygen demand (COD) of 350–400 mg/L, total organic carbon (TOC) of 105–120 mg/L, ammonia (NH_4-N) of 17–20 mg/L, nitrite (NO_2-N) of 0–0.02 mg/L, nitrate (NO_3-N) of 0.4–1.1 mg/L and ortho-phosphate (PO_4-P) of 3.6–4.0 mg/L. $NaHCO_3$ or H_2SO_4 was used to adjust the pH in the MBBR to a constant value of 7. A set of 22 frequently detected and diversely structured compounds were selected (Nguyen et al., 2012) to represent five important groups of micropollutants, namely phamaceuticals and personal care products (PPCPs), pesticides, hormones and industrial

chemicals. A concentrated stock solution containing 100 mg/L of each micropollutant was prepared in pure methanol and kept in a freezer. The stock solution was added to attain an initial concentration of 5 µg/L for each compound during both batch and MBBR experiments. Reticulated porous polyurethane sponge cubes, with dimensions of 2 cm × 2 cm × 2 cm, were used as the attached-growth media in the study. The sponge has a density of 28–30 kg/m^3 with 80 cells per 25 mm. Before the experiments, the sponge was acclimatized (for attached microbial growth) using activated sludge fed with synthetic wastewater without the addition of micropollutants.

2.2 Experiment set-up and sample preparation

2.2.1. Batch experiments

Batch experiments were conducted using 15 L fully-aerated tanks (working volume = 10 L; aeration rate = 1.5 L/min; dissolved oxygen (DO) = 5–6 mg/L) filled with different volumetric ratios of acclimatized or non-acclimatized (virgin carrier without attached biosolids) sponge. Four filling ratios (0%, 10%, 20% and 30%) were initially examined. With the fixed aeration rate, unsatisfactory and non-uniform carrier circulation was observed at 30% filling ratio; therefore this ratio was excluded in subsequent investigations. Samples for micropollutant (300 mL), TOC (30 mL) and nutrient (30 mL) analyses were collected from the tanks at 0, 2, 6, 12 and 24 h during the 24-hour batch test. When the water sample was withdrawn from the tank, a corresponding quantity of sponge cubes were taken out simultaneously to maintain the initial filling ratios (10% or 20%).

2.2.2 MBBR system

A bench-scale MBBR system with a working volume of 40 L was used. The reactor was filled with 20 % (determined from the batch experiments) of acclimatized sponge cubes. Due to the refractory nature of some micropollutants, a prolonged hydraulic retention time (HRT), comparable to that in previous MBR studies (Nguyen et al., 2013; Wijekoon et al. 2013), was deemed necessary for efficient removal of the micropollutants. Thus, the HRT in this study was set at 24 h, which was longer than the typical HRTs (less than 15 h) applied in WWTPs. Accordingly, the reactor had a flow rate of 27.8 mL/min and a COD loading of 0.35–0.40 kg/ (m^3·d). To avoid excessive detachment of the biosolids within the sponge cubes, the aeration of the MBBR was adjusted to around 4 L/min to achieve gentle circulation of the sponge cubes. The DO concentration of the MBBR was in the range between 5.5 and 6.5 mg/L. Before the experiment with addition of micropollutants, the MBBR system was acclimatized to the synthetic wastewater (without addition of micropollutants) for 20 days until TOC, T-N, and PO_4-P removal became stable. After the acclimatization stage, micropollutant-bearing wastewater was continuously introduced to the MBBR and the investigation of micropollutant removal was carried out over a period of 100 days.

Every five days, 300 mL of influent and effluent aqueous samples were collected in duplicate for micropollutant analysis. Effluent samples were centrifuged at 3000 rpm for 30 min before filtration to improve filterability. The influent and centrifuged effluent samples were filtered with 1μm glass microfiber filter paper (47 mm DIA, Filtech) and acidified to pH 2 with 4 M HCl for subsequent solid phase extraction (SPE). To assess the extent of sorption of micropollutants on biosoilds (in suspension and on the sponge cubes), mixed liquor and sponge samples were withdrawn on Day 70 and Day 100. The

sludge within the sponge cubes was collected by squeezing the cubes. Subsequently, the micropollutants were extracted from sludge using the method previously described by Wijekoon et al. (2013).

2.3 Analytical methods

TOC of the influent and effluent was measured using a TOC analyzer (Analytikjena Multi N/C 2000). The analysis of COD and the measurement of biosolids (monitored as mixed liquor suspended solids, MLSS) and biomass (monitored as mixed liquor volatile suspended solids, MLVSS) concentrations were carried out according to Standard Methods (APHA, 1998). The attached-growth biosolids was obtained by hand squeezing the sponge cubes and rinsing the squeezed cubes with Mill-Q water. NH_4-N, NO_2-N, NO_3-N and PO_4-P were measured by spectrophotometric method using Spectroquant Cell Test (NOVA 60, Merck).

Oasis® HLB 6 cc cartridges were used for SPE of micropollutants from aqueous samples. After SPE, the micropollutants were eluted from the cartridges. The eluents were evaporated to dryness. The dry residues in the vials were derivatised, cooled to room temperature and subjected to GC–MS analysis using a Shimadzu GC–MS (QP5000) system. Details of sample preparation and GC–MS operation were described by Nguyen et al. (2012).

3. Results and Discussion

3.1 Batch experiments

Table 1 shows the variations of compound concentrations in the tanks with no sponge (blank experiment), non-acclimatized sponge (10% or 20%) and acclimatized sponge (10% or 20%) during the 24-h operation time. No or negligible micropollutant removals were observed during the blank experiment, indicating air stripping and photolysis did not contribute significantly to the elimination of micropollutants.

Table 1

With non-acclimatized sponge (control experiments), biodegradation of micropollutants was not expected to occur due to the absence of biomass on the sponge. Hence, sorption was the major removal pathway for these compounds. Unlike other plastic attached-growth carriers, which showed no sorption capacity for micropollutants (Falås et al., 2012), the results from experiments with non-acclimatized sponge revealed that sponge was able to adsorb, to varying extents, most of the studied micropollutants. This could be due to the presence of polar and non-polar functional groups in the structure of sponge material (Baldez et al., 2008). Moreover, the sorption of micropollutants onto sponge occurred promptly and reached the equilibrium within 2 hours. In the case of acclimatized sponge, the presence of attached biosolids (0.25 gMLSS/g sponge) resulted in biodegradation of micropollutants as well as sorption of these compounds onto biosolids. According to Falås et al. (2012), sorption onto biosolids is a rapid process and can reach equilibrium within 30 min for many micropollutants.

As shown in Table 1, bisphenol A, etrone, 17-β-estradiol, 17-α ethinylestradiol, 4-n-nonylphenol, 4-tert-octylphenol and triclosan were considerably eliminated (>80%)

during the first two hours in the experiments with either non-acclimatized sponge or acclimatized sponge, which indicates sorption played a significant role in the removal of these compounds. Acetaminophen, diclofenac, gemfibrozil, ibuprofen, ketoprofen, naproxen and salicylic acid were hardly removed (mostly < 20%) with non-acclimatized sponge but showed markedly improved reduction when acclimatized sponge was used. The large removal disparity between the use of non-acclimatized and acclimatized sponge might be attributed to two aspects, namely sorption onto biosolids on the sponge and biodegradation. The contribution of sorption onto biosolids could be estimated with the help of solid-water distribution coefficient (log D, as shown in supplementary data Table S1). Joss et al. (2005) reported that for compounds having a log D < 2.5, the sorption onto secondary sludge can be considered insignificant. As all the above mentioned compounds have log D values less than 2.5, their sorption onto biosolids could be deemed negligible. Hence, the removal of these compounds mainly resulted from biodegradation, while sorption (onto sponge and biosolids) was of fairly limited importance. Carbamazepine, fenprop, and metronidazole were poorly eliminated (mostly < 30%) in both cases. The results suggested that these compounds were resistant to biodegradation and sorption during the experiments.

In addition, filling ratio played an insignificant role in removing most micropollutants during the experiments with non-acclimatized sponge, indicating that the amount of sponge was not a limiting factor (i.e. sponge was in excessive amounts) for sorption of these micropollutants. However, the elimination of some compounds was enhanced by increasing the filling ratio of acclimatized sponge. For instance, acetaminophen, carbamazepine, diclofenac, fenoprop, gemifibrozil, ketoprofen, and

naproxen showed higher removals (76.3, 39.5, 76.1, 34.6, 83.5, 79.2 and 98.3% respectively) at the filling ratio of 20% compared with those (44.7, 17.1, 58.6, 14.5, 63.2, 47.6 and 60.8% respectively) at the filling ratio of 10%. The elevated removals at the higher filling ratio was probably because of the increased amount of attached microorganisms.

3.2 MBBR performance

3.2.1 Organic and nutrient removal

Fig. 1 shows the removal of TOC and nutrient in the MBBR system over a period of 100 days. Stable removal was achieved for TOC (92.6-95.2%) and the effluent TOC concentration was 5.9 ± 0.6 mg/L. NH_4-N removal was in the range between 73.6 and 87.0 % throughout the study. Despite the high DO concentration (5.5–6.5) in the reactor, a distinctive DO gradient occurred toward the core of the sponge, leading to an anoxic/anaerobic condition inside the sponge and thereby facilitating partial denitrification (Guo et al., 2010). As a result, the MBBR system was able to achieve a TN removal of 44.3 ± 10.3 %. Although the PO_4-P removal was relatively high (around 89%) during 20-day acclimatisation period, the MBBR only achieved around 35% removal for the study period. The high PO_4-P removal during the acclimatization period could be due to the use of phosphate for biomass growth as well as the phosphorus uptake by phosphate accumulating organisms (PAOs) under the aerobic condition (Guo et al., 2010). On the other hand, biomass growth on the sponge slowed down after acclimatization, which could contribute to the lower but stable PO_4-P removal. Overall, the effluent NH_4-N, NO_2-N, NO_3-N and PO_4-P concentrations were 3.0 ± 0.8, 0.03 ± 0.02, 5.9 ± 1.7 and 1.4 ± 0.7 mg/L, repectively.

The initial amounts of attached biosolids and biomass on the sponge was 0.25 gMLSS/g sponge and 0.23 gMLVSS/g sponge, respectively. The amount was decreased to 0.19 gMLSS/g sponge and 0.18 gMLVSS/g sponge within the first 20 days of operation owing to the loss of biosolids from the surface of the sponge cubes caused by aeration. Afterwards, with the stabilization of the MBBR system, a gradual increase in the attached biosolids and biomass concentrations was observed and on day 100, the biosolids and biomass concentrations on the sponge were around 0.43 gMLSS/g sponge and 0.40 gMLVSS/g sponge, respectively. During the experiments, some biosolids sloughed off due to the aeration as well as the friction and collision among sponge cubes, but such loss was fairly low. The MLSS and MLVSS in the mixed liquor were stable at 0.13 ± 0.05 and 0.12 ± 0.06 g/L, respectively. Hence, the attached biosolids on sponge can be considered as the principal contributor to the removal of pollutants.

Fig. 1.

3.2.2 Removal of the selected micropollutants

The removal efficiencies of the micropollutants are presented in Fig. 2. Among the pharmaceuticals, apparent variation in removals was observed. As the pharmaceuticals generally displayed low hydrophobicity (log D < 2.5), biodegradation (rather than sorption) was the major removal pathway of these compounds. Four of the studied PPCPs were efficiently eliminated (>80%): these were ibuprofen (93.7%), salicylic acid (91.1%), primidone (83.5%) and naproxen (81.1%). The high removal efficiency could be ascribed to the inclusion of strong electron donating (readily biodegradable) functional groups (e.g., –OH) in these compounds. Ketoprofen,

acetaminophen, metronidazole, and gemifibrozil were moderately removed (50-75%) in the MBBR. Diclofenac and carbamazepine, as opposed to other pharmacueticals, were resistant to MBBR treatment. The average removal of diclofenac in the MBBR was only 45.7% because of the refractory property induced by the inclusion of chlorine group in its molecule. Carbamazepine showed an even lower removal of 25.9%. Zhang et al. (2008) comprehensively surveyed the literature and confirmed that carbamazepine is persistent to biological transformation at low concentrations.

In the case of estrogenic hormones, the removal was consistently high (>85%), which could be attributed to the high hydrophobicity (log D > 3.2) of these compounds (except estriol) as previously suggested by Tadkaew et al. (2011). According to Andersen et al. (2003), estrogenic hormones could be efficiently biodegraded under nitrifying conditions. Hence, they could be successfully eliminated in the MBBR with high nitrification efficiency. Regarding industrial chemicals, the observed removals were generally high (>70%), especially for 4-tert-octylphenol (91.6%) and 4-n-nonylphenol (95.7%), as these compounds are commonly characterized by high hydrophobicity (log D>3.2). As for pesticides, fenoprop exhibited inefficient elimination (31.0%), whereas pentachlorophenol experienced much higher removal (78.9%). The poor removal of fenoprop could be attributed to its low hydrophobicity (log D = –0.13) and recalcitrance (Hai et al., 2011).

Fig. 2.

3.3 Fate of micropollutants during the MBBR treatment and application of mass balance

During the MBBR treatment, the micropollutants might be subjected to biodegradation, sorption and air stripping. Air stripping depends on the volatility and hydrophobicity of micropollutants (Cirja et al., 2008). Henry's constant (k_H) is used to determine volatility. The k_H ranging from 10^{-3} to 10^{-2} commonly indicates marked tendency of volatilization (Stenstrom et al., 1989). As can be seen from supplementary data Table S1, the k_H of most selected micropollutants are within the range of 10^{-4} to 10^{-11}, indicating a minor role of volatilization in the removal of these compounds. Two exceptions are 4-tert-butylphenol (3.61×10^{-2}) and 4-tert-octylphenol (1.98×10^{-3}). However, given the high hydrophobicity of these two compounds, removal by air stripping can also be considered to be insignificant for them. Therefore, it was assumed that the micropollutant removal only resulted from sorption and biological degradation.

Fig. 3 illustrates the concentrations of micropollutants sorbed to sludge. As shown in the figure, the compounds with lower removal efficiency generally accumulated to biosolids (in suspension and on the sponge cubes) to a greater extent, indicating that compound persistence is a factor that affects micropollutant sorption. For instance, carbamazepine, ketoprofen and pentachlorophenol were found at particularly high concentrations on suspended biosolids (7.87, 6.05 and 5.55 μg/g, respectively). There are two possible reasons for this. Firstly, the compounds with high persistence (low removal) were present at high concentrations in the aqueous phase, which may have resulted in high amounts of sorbed compounds at their sorption equilibriums. Secondly, after sorption to biosolids, the recalcitrant compounds were resistant to further biodegradation and therefore remained attached to the solids.

Fig. 3.

Hydrophobicity is another influential factor for micropollutant sorption. However, in this study, no evident correlation was found between compound hydrophobicity and its concentration on biosolids. Except for triclosan and 4-tert-octylphenol, most hydrophobic compounds were found at insignificant amounts on biosolids, possibly because of their higher biodegradability (Section 3.2). Additionally, Fig. 3 showed that suspended biosolids had higher accumulation of micropollutants than attached biosolids. Thus, the attached biosolids appears to possess better biodegradation capacity, possibly due to more diverse microbial community and the existence of different sludge characteristics on and inside the sponge carrier. Further studies are, however, required to substantiate this hypothesis.

To gain a better understanding of micropollutant removal in the MBBR system, a mass balance calculation was carried out over a period of 30 days (from day 70 to day 100), taking into consideration the sorption and biodegradation of micropollutants.

Micropollutant sorption was quantified based on the production of new biosolids, which was presumably responsible for the adsorptive removal of the compounds (Joss et al., 2005). In the MBBR, the production of new biosolids can be divided into two parts: one is the suspended biosolids which are continuously washed out and the other one is attached biosolids that are retained in the MBBR. Therefore, the load of compound removed via sorption can be calculated as follows:

$$L_{sol} = Q \cdot T \cdot MLSS \cdot C_{s,\,s} + \Delta SS \cdot C_{s,\,a} \qquad (1)$$

where L_{sol} is the load of micropollutant removed via sorption over the 30 days (ng), Q is the flow rate of the MBBR (L/day), T is the duration of the study period (day), MLSS is mixed liquor suspended biosolids concentration (g/L), $C_{s,\,s}$ is the concentration of micropollutant on the suspended biosolids (μg/g), ΔSS is the increased amount of attached biosolids over the study period (g), and $C_{s,\,a}$ is the concentration of micropollutant on the attached biosolids (μg/g).

The load of micropollutant removed via biological degradation was determined by calculating the difference of the reduction of micropollutant load (L_{inf} - L_{eff}, ng) and the load of sorbed micropollutant (Eq. 2).

$$L_{biol} = (L_{inf} - L_{eff}) - L_{sol} = Q \cdot T \cdot (C_{w,\,inf} - C_{w,\,eff}) - (Q \cdot T \cdot MLSS \cdot C_{s,\,s} + \Delta SS \cdot C_{s,\,a}) \quad (2)$$

where $C_{w,\,inf}$ and $C_{w,\,eff}$ are the average influent and effluent concentrations of micropollutants over the study period.

The average biosolids concentration in suspension during the 30-day period was 0.15 g/L. The initial and final attached biosolids concentrations were 0.37 and 0.43 gMLSS/g sponge, respectively. The total mass of sponge in the MBBR was 232 g, which was estimated by multiplying the total volume (8 L) of sponge by the density (29 g/L). Hence, the production of attached biosolids was 13.9 g. The calculated percentages of the micropollutant loads which were released from MBBR, sorbed to sludge and biodegraded were presented in Fig. 4. As shown in the figure, biodegradation served as the major removal pathway for most micropollutants. Even for some highly hydrophobic compounds, such as 4-tert-octylphenol, 4-n-nonylphenol, triclosan, β-Estradiol 17-acetate, sorption accounted for minor removals (<0.1 % to

5.5 %). The results are generally in good agreement with those obtained from an MBR, except that 50% the overall loading of triclosan accumulated onto the suspended biosolids of the MBR (Wijekoon et al., 2013). However, sorption was still significant for some persistent compounds (e.g., carbamazepine, diclofenac and fenoprop). In particular, despite the low hydrophobicity (log D=1.89), carbamazepine experienced similar degrees of biodegradations (16.5%) and sorption (15.1%) from day 70 to day 100. It is noteworthy that the sorption by acclimatized sponge played a significant role in the batch experiments (Table 1), while the continuous MBBR experiment showed a limited contribution from sorption (Fig. 4). This could be explained by the facts that 1) sorption is a more rapid process than biodegradation, thus sorption might act as an important removal mechanism within the short-term batch experiments; and 2) in the long-term experiment, the acclimatized sponge remained fully occupied by biomass over time, and the reduced sorption site resulted in the limited sorption efficiency.

Fig. 4.

3.4 Comparison between the MBBR and other treatment technologies for micropollutant removal

Table 2 compares the micropollutants removal in MBBR, suspended-growth activated sludge processes and MBRs. Generally, the effectiveness of MBBR for micropollutant removal was comparable with those of activated sludge processes and MBRs. Moreover, the MBBR seemed to be very effective in eliminating ibuprofen, metronidazole, naproxen, primidone, triclosan, estrone, 17-α ethinylestradiol, 4-n-nonylphenol, 4-tert-octylphenol and fenoprop. It is also noteworthy that the removal of diclofenac in the MBBR (46%) was significantly higher than that reported for a MBR

(20%) (Nguyen et al., 2012). Falås et al. (2012) demonstrated that the presence of biofilm carriers played an important role in the biodegradation of diclofenac. They found that biofilm carriers from full-scale nitrifying wastewater treatment plants had apparently higher removal rates per unit biomass for diclofenac in comparison with the activated sludge. Nevertheless, MBR systems had the ability for more efficient removal of ketoprofen, acetaminophen, gemifibrozil and 4-tert-butylphenol (Nguyen et al., 2013; Wijekoon et al., 2013). Although Servos et al. (2005) stated that activated sludge processes tended to have higher removal of estrogenic potentials (58–>99% with an average of 81%) than some attached-growth processes (0–75% with an average of 28%), such as trickling filters and rotating biological contactors, the MBBR in this study was able to achieve similar or higher elimination of the estrogenic hormones (85.2–96.8%) compared with the activated sludge processes.

Table 2

4. Conclusion

Batch experiments showed that non-acclimatized sponge cubes were able to adsorb varying amounts of micropollutants. Acclimatized sponge improved the removal of some less hydrophobic (log D < 2.5) compounds. The MBBR achieved varying removals for the selected micropollutants due to their diverse physicochemical properties. The removal efficiency is comparable with other processes (activated sludge and MBR). Biodegradation was shown to be the principal removal mechanism for most compounds. The micropollutants that were susceptible to biodegradation did not

significantly accumulate to the biosolids (regardless their hydrophobicity) and sorption was only considerable for compounds with high persistency (e.g., carbamazepine).

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References

1. Andersen, H., Siegrist, H., Halling-Sørensen, B., Ternes, T.A., 2003. Fate of estrogens in a municipal sewage treatment plant. Environmental Science and Technology 37, 4021–4026.
2. APHA, AWWA, WEF, 1998. Standard Methods for the Examination of Water and Wastewater, 20th ed. American Public Health Association, Washington, DC.
3. Baldez, E.E., Robaina, N.F., Cassella, R.J., 2008. Employment of polyurethane foam for the adsorption of Methylene Blue in aqueous medium. Journal of Hazardous Materials 159, 580–586.
4. Baquero, F., Martínez, J.L., Cantón, R., 2008. Antibiotics and antibiotic resistance in water environments. Current Opinion in Biotechnology 19, 260–265.
5. Chu, L., Wang, J., 2011. Comparison of polyurethane foam and biodegradable polymer as carriers in moving bed biofilm reactor for treating wastewater with a low C/N ratio. Chemosphere 83, 63–68.
6. Cirja, M., Ivashechkin, P., Schäffer, A., Corvini, P.F., 2008. Factors affecting the removal of organic micropollutants from wastewater in conventional treatment plants (CTP) and membrane bioreactors (MBR). Reviews in Environmental Science and Bio/Technology 7, 61–78.
7. De la Cruz, N., Gimenez, J., Esplugas, S., Grandjean, D., De Alencastro, L.F., Pulgarin, C, 2012. Degradation of 32 emergent contaminants by UV and neutral photo-fenton in domestic wastewater effluent previously treated by activated sludge. Water Research 46, 1947–1957.

8. Drewes, J.E., Fox, P., Jekel, M., 2001 Occurrence of iodinated X-ray contrast media in domestic effluents and their fate during indirect potable reuse. Journal of Environmental Science and Health, Part A 36, 1633–1645.
9. Falås, P., Longrée, P., la Cour Jansen, J., Siegrist, H., Hollender, J., Joss, A., 2013. Micropollutant removal by attached and suspended growth in a hybrid biofilm-activated sludge process. Water Research 47, 4498–4506.
10. Falås, P., Baillon-Dhumez, A., Andersen, H.R., Ledin, A., la Cour Jansen, J., 2012. Suspended biofilm carrier and activated sludge removal of acidic pharmaceuticals. Water Research 46, 1167–1175.
11. Fent, K., Weston, A.A., Carminada, D., 2006. Ecotoxicology of human pharmaceuticals. Aquatic Toxicology 76, 122–159.
12. Guo, W., Ngo, H., Vigneswaran, S., 2012. Enhancement of Membrane Processes with Attached Growth Media, in: Zhang, et al. (Eds.), Membrane Technology and Environmental Applications. American Society of Civil Engineers, New York, pp. 603–634.
13. Guo,W., Ngo, H.H., Dharmawan, F., Palmer, C.G., 2010. Roles of polyurethane foam in aerobic moving and fixed bed bioreactors. Bioresource Technology 101, 1435–1439.
14. Hai, F.I., Tessmer, K., Nguyen, L.N., Kang, J., Price, W.E., Nghiem, L.D., 2011. Removal of micropollutants by membrane bioreactor under temperature variation. Journal of Membrane Science 383, 144–151.
15. Huerta-Fontela, M., Galceran, M.T., Ventura, F., 2011. Occurrence and removal of pharmaceuticals and hormones through drinking water treatment. Water Research 45, 1432–1442.
16. Joss, A., Keller, E., Alder, A.C., Göbel, A., McArdell, C.S., Ternes, T.A., et al., 2005. Removal of pharmaceuticals and fragrances in biological wastewater treatment. Water Research 39, 3139–3152.
17. Kasprzyk-Hordern, B., Dinsdale, R.M., Guwy, A.J., 2009. The removal of pharmaceuticals, personal care products, endocrine disruptors and illicit drugs during wastewater treatment and its impact on the quality of receiving waters. Water Research 43, 363–380.

18. Lin, A.Y. –C., Yu, T.H., Lateef, S.K., 2009. Removal of pharmaceuticals in secondary wastewater treatment processes in Taiwan. Journal of Hazardous Materials 167, 1163–1169.
19. Luo, Y., Guo, W., Ngo, H.H., Nghiem, L.D., Hai, F.I., Zhang, J., et al., 2014. A review on the occurrence of micropollutants in the aquatic environment and their fate and removal during wastewater treatment. Science of The Total Environment 473–474, 619–641.
20. Ngo, H.H., Guo, W., Xing W. 2008. Evaluation of a novel sponge-submerged membrane bioreactor (SSMBR) for sustainable water reclamation. Bioresource Technology 99, 2429–2435.
21. Nguyen, L.N., Hai, F.I., Kang, J., Nghiem, L.D., Price, W.E., Guo, W., Ngo, H.H. and Tung, K.L. 2013. Comparison between sequential and simultaneous application of activated carbon with membrane bioreactor for trace organic contaminant removal. Bioresource Technology 130, 412–417.
22. Nguyen, L.N., Hai, F.I., Kang, J., Price, W.E., Nghiem, L.D., 2012. Removal of trace organic contaminants by a membrane bioreactor–granular activated carbon (MBR–GAC) system. Bioresource Technology 113, 169–173.
23. Servos, M.R., Bennie, D.T., Burnison, B.K., Jurkovic, A., McInnis, R., Neheli, T., et al., 2005. Distribution of estrogens, 17β-estradiol and estrone, in Canadian municipal wastewater treatment plants. Science of The Total Environment 336, 155–170.
24. Stenstrom, M.K., Cardinal, L., Libra, J, 1989. Treatment of hazardous substances in wastewater treatment plants. Environmental Progress 8, 107–112.
25. Suarez, S., Lema, J.M., and Omil, F., 2010. Removal of pharmaceutical and personal care products (PPCPs) under nitrifying and denitrifying conditions. Water Research 44, 3214–3224.
26. Tadkaew, N., Hai, F.I., McDonald, J.A., Khan, S.J., Nghiem, L.D., 2011. Removal of trace organics by MBR treatment: the role of molecular properties. Water Research 45, 2439–2451.
27. Wick, A., Fink, G., Joss, A., Siegrist, H., Ternes, T.A., 2009. Fate of beta blockers and psycho-active drugs in conventional wastewater treatment. Water Research 43, 1060–1074.

28. Wijekoon, K.C., Hai, F.I., Kang, J., Price, W.E., Guo, W., Ngo, H.H., Nghiem, L.D., 2013. The fate of pharmaceuticals, steroid hormones, phytoestrogens, UV-filters and pesticides during MBR treatment. Bioresource Technology 144, 247–254.
29. Yangali-Quintanilla, V., Maeng, S.K., Fujioka, T., Kennedy, M., Li, Z, Amy, G., 2011. Nanofiltration vs. reverse osmosis for the removal of emerging organic contaminants in water reuse. Desalination and Water Treatment 34, 50–56.
30. Zhang, Y., Geißen, S.-U., Gal, C., 2008. Carbamazepine and diclofenac: Removal in wastewater treatment plants and occurrence in water bodies. Chemosphere 73, 1151–1161.
31. Zwiener, C., Frimmel, F.H., 2003. Short-term tests with a pilot sewage plant and biofilm reactors for the biological degradation of the pharmaceutical compounds clofibric acid, ibuprofen, and diclofenac. Science of The Total Environment 309, 201–211.

Figure captions

Fig. 1. Organic and nutrient removal in the MBBR system

Fig. 2. Removal efficiency of micropollutants during the MBBR treatment. Each error bar represents the standard deviation of 20 measurements over 100 days.

Fig. 3. Average concentrations of micropollutants on the suspended and attached biosolids

Fig. 4. Fate of the studied micropollutants in the MBBR system.

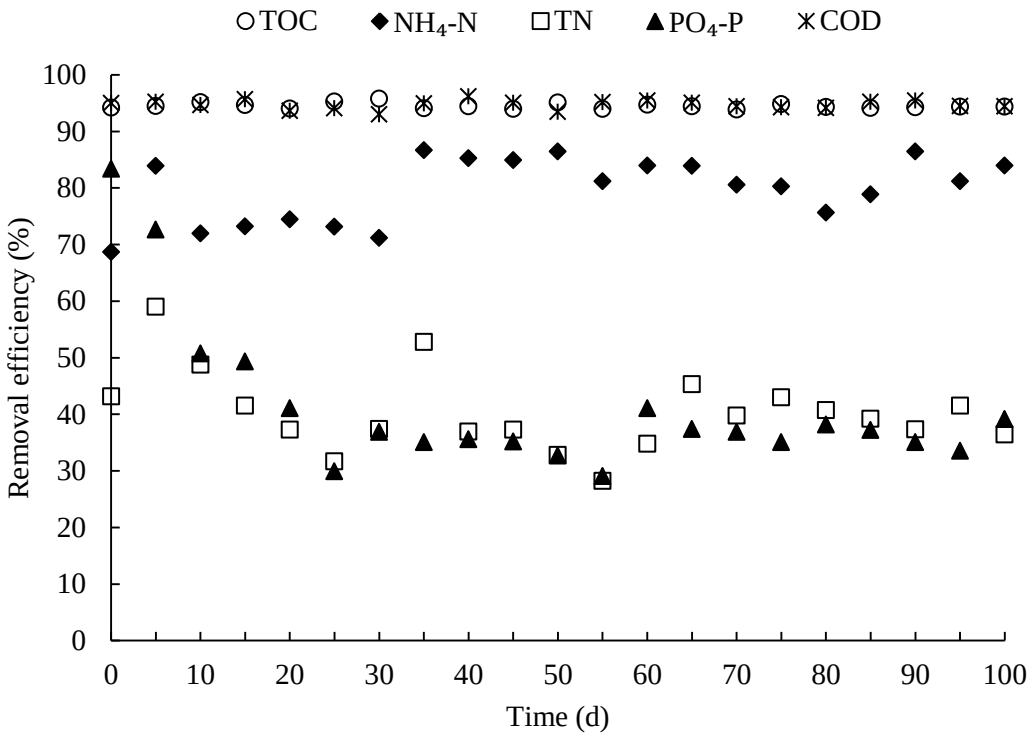
TOC
NH4-N
TN
PO4-P
COD
Removal efficiency (%)
Time (d)
0
10
20
30
40
50
60
70
80
90
100

Fig. 1.

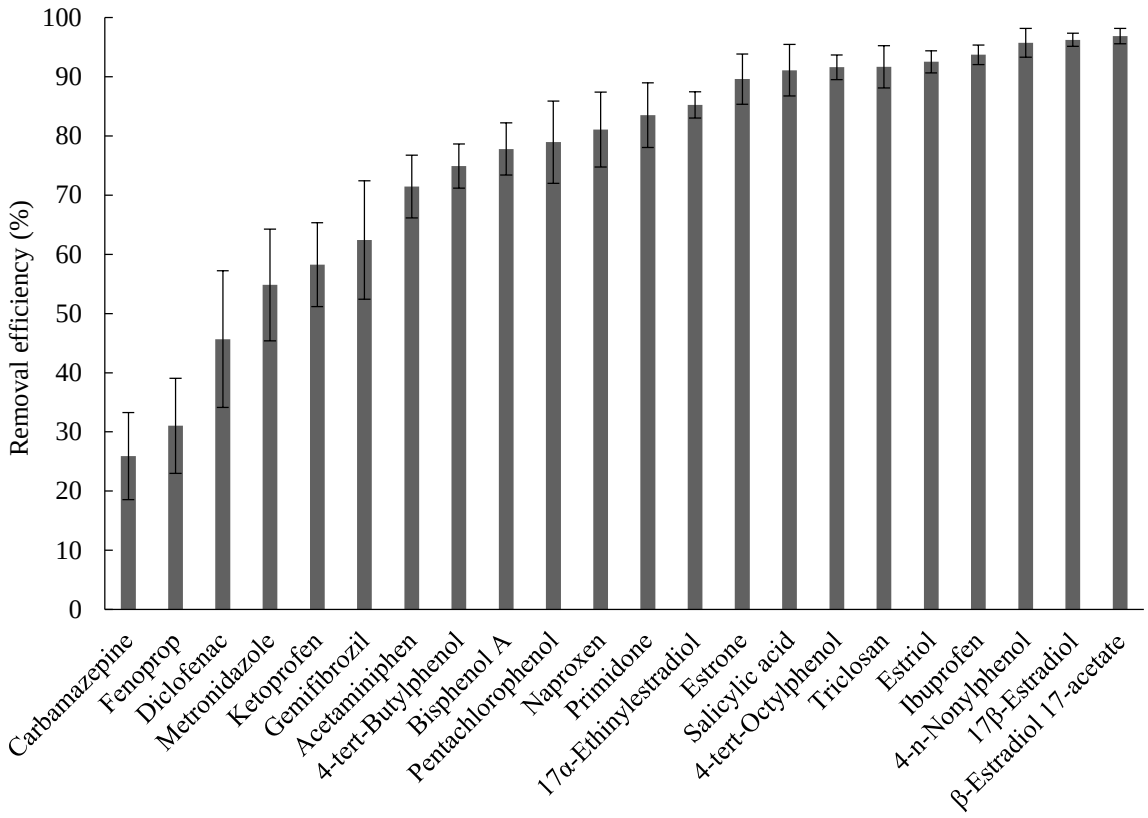
Removal efficiency (%)
100
90
80
70
60
50
40
30
20
10
0
Carbamazepine
Fenoprop
Diclofenac
Metronidazole
Ketoprofen
Gemifibrozil
Acetaminiphen
4-tert-Butylphenol
Bisphenol A
Pentachlorophenol
Naproxen
Primidone
17α-Ethinylestradiol
Estrone
Salicylic acid
4-tert-Octylphenol
Triclosan
Estriol
Ibuprofen
4-n-Nonylphenol
17β-Estradiol
β-Estradiol 17-acetate

Fig. 2.

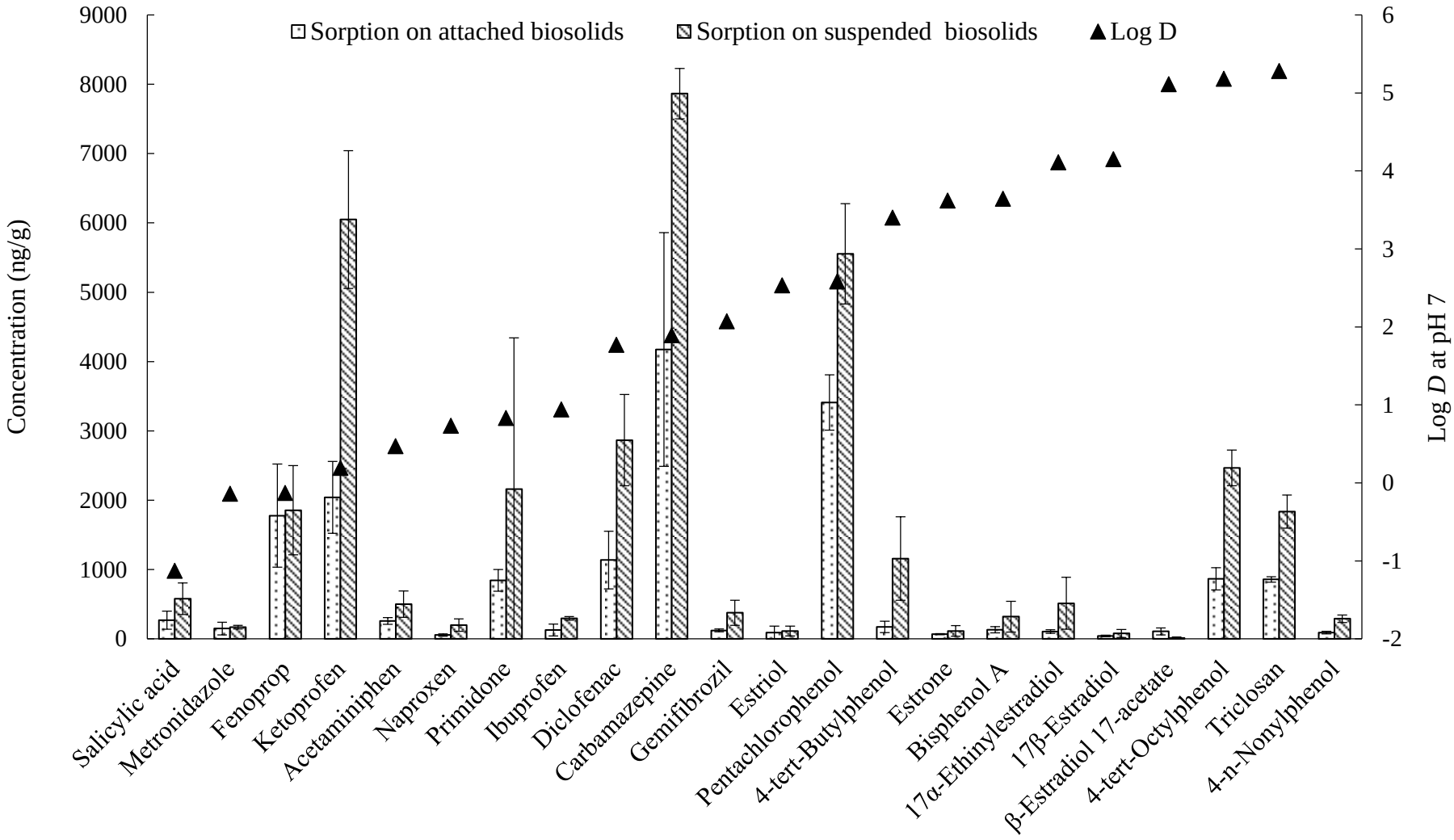
Sorption on attached biosolids
Sorption on suspended biosolids
Log D
Concentration (ng/g)
Log D at pH 7
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
6
5
4
3
2
1
0
-1
-2
Salicylic acid
Metronidazole
Fenoprop
Ketoprofen
Acetaminiphen
Naproxen
Primidone
Ibuprofen
Diclofenac
Carbamazepine
Gemifibrozil
Estriol
Pentachlorophenol
4-tert-Butylphenol
Estrone
Bisphenol A
17α-Ethinylestradiol
17β-Estradiol
β-Estradiol 17-acetate
4-tert-Octylphenol
Triclosan
4-n-Nonylphenol

Fig. 3.

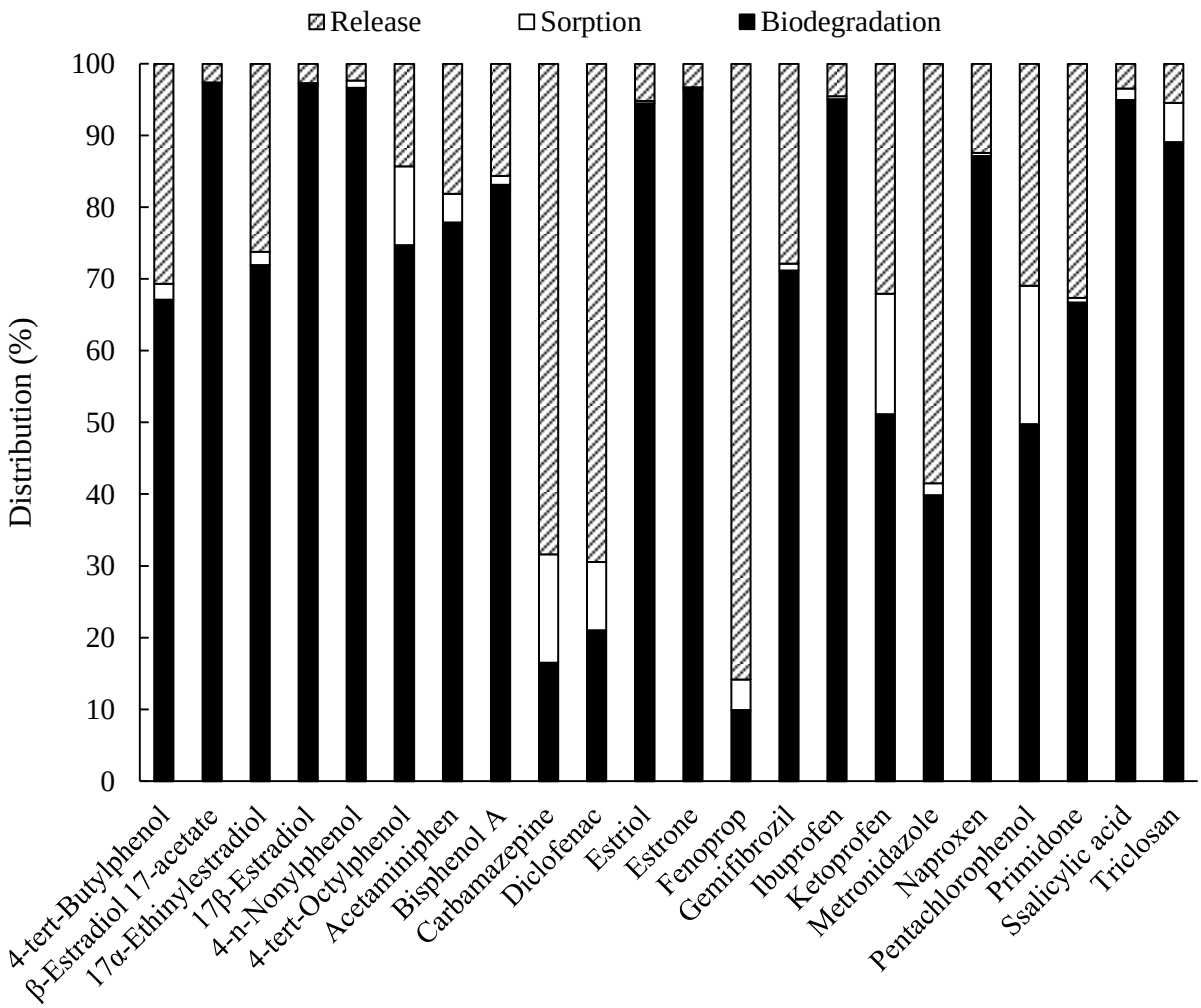
Release
Sorption
Biodegradation
Distribution (%)
100
90
80
70
60
50
40
30
20
10
0
4-tert-Butylphenol
β-Estradiol 17-acetate
17α-Ethinylestradiol
17β-Estradiol
4-n-Nonylphenol
4-tert-Octylphenol
Acetaminiphen
Bisphenol A
Carbamazepine
Diclofenac
Estriol
Estrone
Fenoprop
Gemifibrozil
Ibuprofen
Ketoprofen
Metronidazole
Naproxen
Pentachlorophenol
Primidone
Ssalicylic acid
Triclosan

Fig. 4.

Table captions

Table 1 The variation of micropollutants' concentrations (ng/L) during the batch experiments

Table 2 Comparison of micropollutant removal efficiency (%) in the MBBR and in other biological treatment technologies

Table 1
The variation of micropollutant concentrations (ng/L) during the batch experiments.

Compound[a]/ Condition[b]	0 h	2 h	6 h	12 h	24 h	% removal after 24h
ACM						
B	1666	1626	1598	1572	1629	2.2
N(10)	1394	1329	1354	1263	1214	12.9
N(20)	1373	1306	1334	1220	1237	9.9
A(10)	1420	1106	751	794	785	44.7
A(20)	1951	1577	602	491	463	76.3
BP						
B	3298	3172	2899	3071	3089	6.3
N(10)	2549	1394	983	1003	889	65.1
N(20)	2621	954	977	1029	943	64.0
A(10)	3571	2386	1966	1543	1397	60.9
A(20)	3591	1643	1583	1097	851	76.3
BPA						
B	3665	3591	3124	3472	3212	12.4
N(10)	3477	580	543	563	589	83.1
N(20)	3398	537	594	586	591	82.6
A(10)	4500	649	546	526	564	87.5
A(20)	4394	249	373	306	277	93.7
CBZ						
B	3181	3355	3090	2967	3037	4.5
N(10)	3340	2849	2711	2489	2383	28.7
N(20)	2988	2509	2371	2377	2202	26.3
A(10)	4463	3534	3574	3489	3699	17.1
A(20)	4951	3766	3711	3809	2997	39.5
DCF						
B	2745	2658	2798	2553	2599	5.3
N(10)	2474	2209	2174	2097	1966	20.5
N(20)	2984	1866	1729	2003	1916	35.8
A(10)	2220	1823	1817	1171	920	58.6
A(20)	2360	1291	1408	1011	563	76.1
E1						
B	2123	1842	1958	2154	2031	4.3
N(10)	1920	171	143	143	131	93.2

Compound/ condition	0 h	2 h	6 h	12 h	24 h	% removal after 24h
GFB						
B	3797	3612	3811	3537	3512	7.5
N(10)	2871	2751	2869	3117	2540	11.5
N(20)	3105	2583	2669	2760	2460	22.5
A(10)	3797	2903	2714	2517	1398	63.2
A(20)	3849	3443	1963	1389	634	83.5
IBP						
B	2940	2759	2885	2974	2943	1.0
N(10)	3051	2760	2897	2991	2814	7.8
N(20)	3057	2714	2660	2997	2920	4.5
A(10)	3669	2917	2263	1414	697	81.0
A(20)	3574	3354	719	623	480	86.6
KTP						
B	4759	4677	4816	4670	4623	2.9
N(10)	4251	4226	3971	4137	4060	4.5
N(20)	4112	3954	3949	3811	3889	5.4
A(10)	5146	4194	4569	4412	2694	47.6
A(20)	4869	5189	3825	2783	1014	79.2
MTZ						
B	1132	1207	1082	1053	1164	-2.8
N(10)	1100	1063	1054	1077	1037	5.7
N(20)	1121	960	1049	934	1006	10.3
A(10)	1551	1460	1426	1414	1238	20.2
A(20)	1591	1571	1435	1520	1334	16.2
NP						
B	1203	1106	934	1122	1006	16.4
N(10)	1200	149	117	74	20	98.3
N(20)	1001	117	74	83	48	95.2
A(10)	1297	71	126	102	76	94.1
A(20)	1089	94	192	111	123	88.7
NPX						
B	3609	3748	3776	3641	3745	-3.8
N(10)	3094	2717	2580	2789	2937	5.1

	N(20)	2212	83	77	74	102	95.4
	A(10)	2326	566	466	433	446	80.8
	A(20)	2246	69	286	183	160	92.9
E2							
	B	5340	5142	4833	4959	5461	-2.3
	N(10)	4446	460	377	366	346	92.2
	N(20)	4631	246	217	209	227	95.1
	A(10)	5177	154	80	120	133	97.4
	A(20)	4957	246	138	134	103	97.9
E2Ac							
	B	2114	2209	1934	2195	1953	7.6
	N(10)	2260	51	94	37	14	99.4
	N(20)	1998	51	23	0	9	99.5
	A(10)	1951	71	66	33	46	97.6
	A(20)	2194	69	111	0	34	98.5
E3							
	B	2671	2744	2644	2643	2609	2.3
	N(10)	2689	1994	1686	1777	1760	34.5
	N(20)	2534	1649	1380	1283	1219	51.9
	A(10)	2951	800	223	159	76	97.4
	A(20)	2794	1674	40	0	89	96.8
EE2							
	B	3256	3226	3386	3202	3180	2.3
	N(10)	2786	89	80	77	66	97.6
	N(20)	2822	46	43	34	31	98.9
	A(10)	3180	186	109	60	61	98.1
	A(20)	3163	14	97	97	123	96.1
FNP							
	B	2462	2227	2260	2376	2419	1.7
	N(10)	2217	2183	1900	2034	2097	5.4
	N(20)	2139	2057	2151	2160	1998	6.6
	A(10)	2571	2160	2151	2081	2198	14.5
	A(20)	2417	2394	1993	1683	1580	34.6
	N(20)	2867	2746	2631	2623	2594	9.5
	A(10)	3254	2714	2531	2102	1276	60.8
	A(20)	3303	3283	1802	740	57	98.3
OP							
	B	1857	1885	1954	2002	1843	0.8
	N(10)	1803	54	80	40	20	98.9
	N(20)	1808	80	31	34	37	98.0
	A(10)	1789	86	60	48	53	97.0
	A(20)	1943	66	61	34	43	97.8
PECP							
	B	2365	2413	2394	2086	2265	4.2
	N(10)	2417	2394	1993	1683	1580	34.6
	N(20)	2612	1097	940	943	671	74.3
	A(10)	2760	849	549	376	244	91.2
	A(20)	2697	1363	239	134	97	96.4
PRM							
	B	3194	3258	2965	3206	3010	5.8
	N(10)	3094	2069	1674	1834	1849	40.2
	N(20)	3111	1614	1366	1380	1262	59.4
	A(10)	3897	1600	1020	731	625	84.0
	A(20)	2549	1640	928	460	229	91.0
SA							
	B	4392	4496	4302	4272	3938	10.3
	N(10)	3706	3309	3397	3466	3340	9.9
	N(20)	3983	3129	3077	3020	3425	14.0
	A(10)	4034	1780	380	316	305	92.4
	A(20)	4046	1406	501	274	214	94.7
TCS							
	B	2491	2559	2481	2447	2237	10.2
	N(10)	2406	46	74	23	29	98.8
	N(20)	2105	60	29	11	10	99.5
	A(10)	2574	57	49	48	50	98.1
	A(20)	2546	69	84	40	63	97.5

[a] ACM: acetaminophen; BP: 4-tert-Butylphenol; BPA: bisphenol A; CBZ: carbamazepine; DCF: diclofenac; E1: estrone; E2: 17-β-estradiol; E2Ac: β-Estradiol 17-acetate; EE2: 17-α ethinylestradiol; E3: estriol; FNP: fenoprop; GFB: gemfibrozil; IBP: ibuprofen; KTP: ketoprofen; MET: metronidazole; NP: 4-n-nonylphenol; NPX: naproxen; OP: 4-tert-octylphenol; PECP: pentachlorophenol; PRM: primidone; SA: salicylic acid; TCS: triclosan;

[b] B: blank experiments (0 % fill ratio); N(10), N(20): non-acclimatized sponge with filling ratio (10% or 20%) in the parentheses; A(10), A(20): acclimatized sponge with filling ratio (10% or 20%) in the parenthes

Table 2
Comparison of micropollutant removal efficiency (%) in the MBBR and in other biological treatment technologies.

Compounds[a]		This study (MBBR, %)	Activated sludge[b] (%)	MBR[c] (%)
Pharmaceuticals	ACM	71.4 ±10.6	98.7-100	40–100
	CBZ	25.9 ± 14.7	<0-63.2	0–35
	DCF	45.7 ± 23.1	<0–81.4	0–87
	GFB	62.4 ± 20.0	<0–92.3	90–98
	IBU[d]	93.7 ± 3.3	72–100	50-99
	KTP	58.2 ± 14.2	10.8–100	52–92
	MET	54.8 ± 18.9	0–64.0	36–40
	NPX	81.1 ± 12.6	43.3–98.6	10–84
	PRM	83.5 ± 10.9	30-50	10–91
	SA	91.1 ± 8.7	89.6–100	93–98
	TCS	91.7 ± 7.1	71.3–99.2	70–99
Steroid hormones	**E1**	89.6 ± 8.5	74.8–90.6	96–99
	E2	96.2 ± 2.2	92.6–100	97–99
	E2Ac	96.8 ± 2.6	-	98–99
	EE2	85.2 ± 4.5	43.8–100	60–98
	E3	92.5 ± 3.7	100	83–97
Industrial chemicals	BP	74.9 ± 7.4	-	93–98
	BPA	77.8 ± 8.8	62.5–99.6	52–98
	NP	95.7 ± 4.9	21.7–99	87–97
	OP	91.6 ± 4.2	<0–96.7	97-98
Pesticides	**FNP**	31.0 ± 16.1	-	10–21
	PECP	78.9 ± 13.9	-	61–99

[a] ACM: acetaminophen; BP: 4-tert-Butylphenol; BPA: bisphenol A; CBZ: carbamazepine; DCF: diclofenac; E1: estrone; E2: 17-β-estradiol; E2Ac: β-Estradiol 17-acetate; EE2: 17-α ethinylestradiol; E3: estriol; FNP: fenoprop; GFB: gemfibrozil; IBP: ibuprofen; KTP: ketoprofen; MET: metronidazole; NP: 4-n-nonylphenol; NPX: naproxen; OP: 4-tert-octylphenol; PECP: pentachlorophenol; PRM: primidone; SA: salicylic acid; TCS: triclosan.
[b] Data from Kasprzyk-Hordern et al., 2009; Lin et al., 2009; Luo et al., 2014; Wick et al., 2009.
[c] Data from Nguyen et al., 2013.
[d] Listed in bold type are the compounds whose percentage removals are close to or above the maximum removals reported in activate sludge or/and MBR.