





Application of alternative carriers without protected surface in moving bed biofilm reactor for domestic wastewater treatment

Bruno de Oliveira Freitas ^{a,b,*}, Luan de Souza Leite ^b, Maria Teresa Hoffmann ^b, Antonio Wagner Lamon^b and Luiz Antonio Daniel ^b

^a Department of Environmental Engineering, Federal University of Technology-Paraná (UTFPR), Av. dos Pioneiros, 3131, 86036-370, Londrina, Paraná, Brazil

^b Department of Hydraulics and Sanitation, São Carlos School of Engineering, University of São Paulo, Av. Trabalhador São-Carlense, 400, 13566-590, São Carlos, São Paulo, Brazil

*Corresponding author. E-mail: brunofreitas@utfpr.edu.br

 B de OF, 0000-0003-2693-7806; L de SL, 0000-0002-2108-2960; MTH, 0000-0003-2695-3275; LAD, 0000-0002-1765-4209

ABSTRACT

Biological reactors with immobilized biomass on free carriers have provided new perspectives for wastewater treatment, once they reduce the system size and increase the treatment capacity. In this study, the performance of three Moving Bed Biofilm Reactors (MBBRs) using different carriers (with and without protected surface area) were evaluated for domestic wastewater treatment in continuous flow. Each MBBR (*i.e.*, R1, R2, and R3) was filled at a ratio of 50% with high-density polyethylene (HDPE) carriers with different characteristics: both R1-K1 and R2-Corrugated tube with protected surface and R3-HDPE flakes without protected surface. Chemical oxygen demand (COD) removal of 80 ± 5.0 , 80 ± 3.5 , and $78 \pm 2.4\%$ was achieved by R1, R2, and R3, respectively. The oxygen uptake by biofilm attached on the carriers was 0.0079 ± 0.0013 , 0.0033 ± 0.0015 , and $0.0031 \pm 0.0026 \mu\text{g DO}\cdot\text{mm}^{-2}$ for the K1, corrugated tube, and HDPE flakes, respectively. No significant differences were observed between the performance of the three MBBRs in terms of physico-chemical parameters (alkalinity, pH, and dissolved inorganic carbon) and COD removal. Results showed that the carrier type and its characteristics (total area and with/without protected area) did not affect the organic matter removal. Thus, the carrier without a protected surface in MBBR could be a promising low-cost option for domestic wastewater treatment.

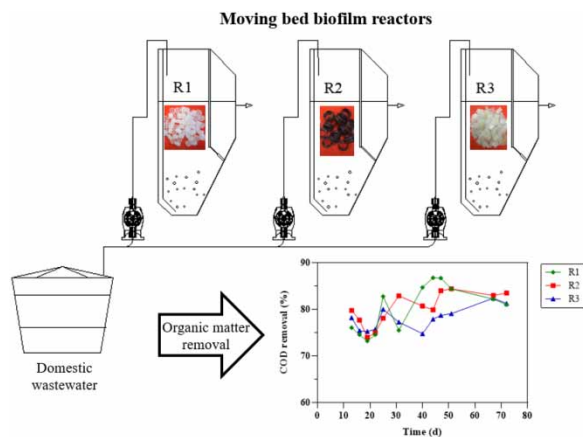
Key words: bench-scale reactor, nitrification, organic matter removal, plastic flakes carrier, protected surface

HIGHLIGHTS

- MBBRs using different carriers (with and without protected surface area) were evaluated for wastewater treatment.
- COD removal of 80 ± 5.0 , 80 ± 3.5 , and $78 \pm 2.4\%$ was achieved by R1, R2, and R3, respectively.
- No significant differences were observed between the three MBBRs' performance.
- Carrier without protected surface in MBBR could be a promising low-cost option.

This is an Open Access article distributed under the terms of the Creative Commons Attribution Licence (CC BY 4.0), which permits copying, adaptation and redistribution, provided the original work is properly cited (<http://creativecommons.org/licenses/by/4.0/>).

GRAPHICAL ABSTRACT



1. INTRODUCTION

Increasing urbanization, hydrological uncertainty, and limited water resources demand a more advanced technology to preserve water quality. In this scenario, the moving bed biofilm reactor (MBBR) stands out as an alternative technology to conventional activated sludge reactors for the treatment of domestic and industrial wastewater (di Biase *et al.* 2019).

The MBBR has been widely applied for wastewater treatment over the last two decades due to its advantages of operating in different treatments (aerobic, anaerobic, and anoxic), adapting in existing facilities under organic overloading, and developing smaller treatment systems (Mcquarrie *et al.* 2011; Piculell *et al.* 2014; Vieira *et al.* 2014; di Biase *et al.* 2019). The basic principle of this process is the growth of active biofilm on the surface of plastic carriers, which move freely inside the reactor (filling ratio up to 70%) and have a density very close to water density. The presence of carriers increases the treatment performance per working reactor volume (Ødegaard *et al.* 1994; di Biase *et al.* 2019).

In developing countries like Brazil, in which less than half of the population is served with wastewater treatment (Brasil 2019), the main drawback to MBBR application is the carrier cost, which limits the widespread use of this technology. For instance, conventional activated sludge was up to 100% more economical than IFAS/MBBR due to the cost of CAPEX carriers (Oliveira *et al.* 2013).

Different carriers are available in the market to be applied in the MBBR and they differ in shape, density, size, and surface available for biofilm formation (Ødegaard *et al.* 1994; Mcquarrie *et al.* 2011; di Biase *et al.* 2019). Carriers are usually designed to have a large protected surface on which biofilm can grow, such as K1–K5, sponges-HDPE, polyurethane foam (PUF), PUF-PP, and Mutag Biochip (Ødegaard *et al.* 1994; Araujo Junior *et al.* 2013; Bassin *et al.* 2016; Saidi *et al.* 2017; Sonwani *et al.* 2019). However, alternative carriers with low-cost and recycled material have been employed in MBBRs to overcome the economic limitations, such as a recycled corrugated tube, recycled plastic, and cigarette filter rods from tobacco factories (Wolff *et al.* 2005; Sabzali *et al.* 2012; Tombola *et al.* 2019).

The studies about MBBR for wastewater treatment applied commercial or alternative carriers with a protected surface (Levstek & Plazl 2009; Piculell *et al.* 2014; Zinatizadeh & Ghaytooli 2015; Bassin *et al.* 2016; Wang *et al.* 2018; Sonwani *et al.* 2019; Zhao *et al.* 2019; Massoompour *et al.* 2020). However, to the best of our knowledge, there is no study addressing the application of carriers without a protected surface for wastewater treatment, even though they are simpler and cheaper than the commercial carriers. Despite the well-known advantages of carriers with a protected area, carriers without a protected surface may also lead to a good MBBR performance for domestic wastewater treatment.

Herein, the treatment of domestic wastewater by MBBRs was investigated by using carriers with and without a protected surface. The MBBRs' performances were evaluated between one commercial (K1) and two alternative carriers (Corrugated tube and HDPE flakes). The main goals of this paper were: (1) to evaluate the nitrification process; (2) to assess the organic matter removal; and (3) to evaluate the consumption of dissolved oxygen by biofilm using a microsensor.

2. MATERIAL AND METHODS

2.1. Carrier

Three different carriers (K1, corrugated tube, and HDPE flakes) were used as support material for attached biofilm bioreactors. The characteristics of carriers used in this research are given in Table 1.

Table 1 | Characteristics of carriers employed in the MBBRs

Reactor	Carrier	Shape	Size (mm)	Material	Porosity (%)	Total specific surface area ($\text{m}^2\cdot\text{m}^{-3}$)	Protected surface	Total surface area (m^2) ^a
R1	K1	Cylinder	9.1 × 7.3 ($\varnothing \times L$)	Polyethylene	87.5	500	Yes	2.51
R2	Corrugated tube	Cylinder	19 × 9.6 ($\varnothing \times L$)	Polyethylene	82.8	390	Yes	1.46
R3	HDPE flakes	Rectangle	8.3 × 11.9 (W × L)	Polyethylene	60.9	1,000	No	3.75

Note: \varnothing , Diameter; W, width; L, length.

^aTotal surface area provided by carriers for biofilm growth in each MBBR.

The reactors R1, R2, and R3 were filled with 50% of carriers in the reaction zone according to Zinatizadeh & Ghaytooli (2015).

2.2. Experimental setup

The study was carried out in three lab-scale reactors operating in parallel (see Figure 1), which were fed with raw domestic wastewater from a sanitary sewer located at the University of São Paulo (São Carlos, São Paulo state, Brazil). The physico-chemical characterization of the raw wastewater is summarized in Table 2. Each acrylic reactor had a volume of 7.5 and 1.7 L for aeration tank and decanter, respectively. A sieve was placed in the decanter inlet to keep the carriers in the reaction zone of the MBBR. The mixed liquor recirculation was not performed because the reactors were operated as MBBRs (Wei *et al.* 2016).

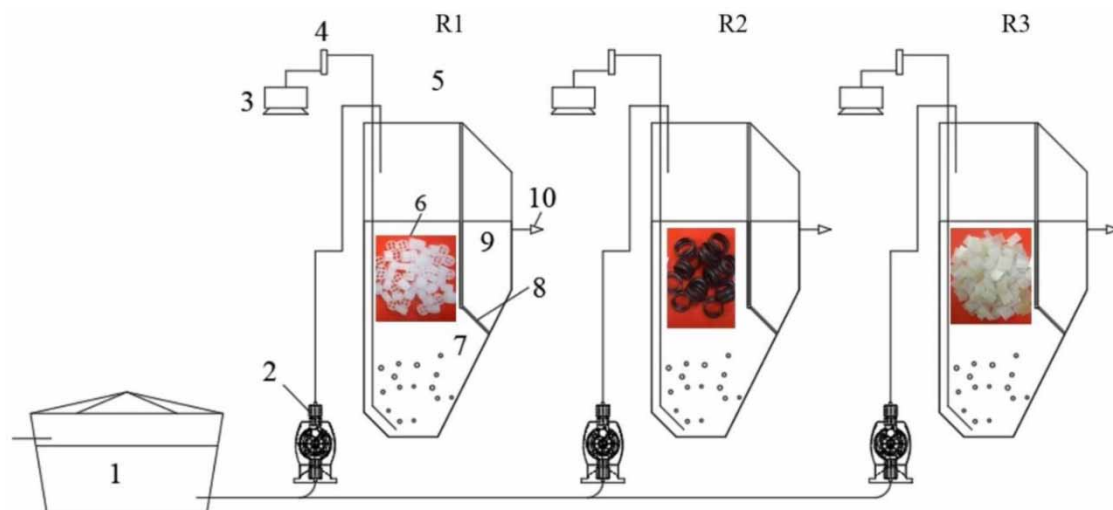


Figure 1 | Schematic of the experimental setup for continuous flow MBBRs. (1) storage tank of raw wastewater, (2) feed pump, (3) air blower, (4) rotameter, (5) MBBR, (6) carriers, (7) aerobic tank, (8) sieve (opening size of 5 mm), (9) decanter, and (10) MBBR effluent.

The MBBRs were continuously fed using metering pumps (ProMinent Concept Plus, Germany) at a flow rate of $3.75 \text{ L}\cdot\text{h}^{-1}$, which resulted in a hydraulic retention time (HRT) of 2 h. A low HRT is usually used in MBBR (0.5–3 h) and this value was adopted based on previous studies (Ødegaard 2006; Piculell *et al.* 2014; Metcalf & Eddy 2016; Bassin *et al.* 2016). The reactors were aerated by an air blower (Boyu, ACQ-003, China) at an air flow rate of $5 \text{ L}\cdot\text{min}^{-1}$. A coarse bubble diffuser was used to provide aeration and mixing inside the bioreactors, maintaining the dissolved oxygen (DO) concentration higher than $3 \text{ mg}\cdot\text{L}^{-1}$ (Collivignarelli *et al.* 2019).

Table 2 | Characterization of raw domestic wastewater

Parameter	n	Mean \pm SD	Maximum	Minimum
tCOD (mg·L ⁻¹)	12	306 \pm 44	378	253
Total solids (mg·L ⁻¹)	24	448 \pm 81	632	270
pH	32	7.36 \pm 0.19	7.98	6.98
Alkalinity (mg CaCO ₃ ·L ⁻¹)	32	177 \pm 20	212	136
NH ₃ (mg N·L ⁻¹)	32	42 \pm 7.8	56	26
PO ₄ ³⁻ (mg P·L ⁻¹)	9	4.45 \pm 1.7	7.10	0.58
tCOD/NH ₃ ratio	–	7.15	–	–

Note: N, number of analyses; SD, standard deviation.

The sludge excess from the decanter was discharged once a week. A sludge volume ranging from 1 to 2 L was removed each time, with a mean concentration of 1,039 \pm 343 mg TS·L⁻¹ and 620 \pm 266 mg VTS·L⁻¹. The MBBRs were operated at room temperature (24 \pm 5 °C). The reactors were operated for 80 days and samples (influent and effluent) were taken once a week to be analyzed in terms of physico-chemical parameters and organic matter.

The MBBRs were started up without a previous inoculum to evaluate the potential of each carrier to the biofilm formation using the microorganisms present in domestic wastewater.

2.3. Physico-chemical analyses

The MBBR influent and effluent were characterized by pH, alkalinity, dissolved inorganic carbon (DIC), total chemical oxygen demand (tCOD), and soluble chemical oxygen demand (sCOD). For DIC and sCOD analyses, the wastewater samples were previously filtered into 0.45 μ m fiberglass membrane. Total Solids (TS) and Volatile Total Solids (VTS) were monitored only in the mixed liquor and raw wastewater. The dissolved oxygen (DO) measurements were done using a portable oximeter (YSI, EcoSence DO 200A, USA). The ammonia nitrogen and orthophosphate were monitored only in the raw wastewater. All analyses were performed in duplicate and according to APHA (2012).

2.4. Microsensor application for oxygen depletion test

To measure the DO consumption due to the organic matter oxidation by the biofilm, experiments were carried out using a mini respirometric cell with a capacity of 15 mL. The microsensor Clark-type DO was developed and built according to Gonzalez *et al.* (2011), and it consisted of two glass plates, a rubber o-ring, an extravasation point of wastewater excess, and access to the DO microsensor.

The respirometric analyses were performed following the steps: (1) the raw wastewater samples were filtered into a 1.2 μ m membrane to remove the biological flocs, which can interfere in the tests, and the filtered sample was fully saturated with DO before the tests using an aquarium air blower and porous stone; (2) 15 mL of saturated wastewater sample was put into the respirometric cell to measure the DO concentration without the carriers (blank); and then (3) the DO concentration was measured in samples containing 15 mL saturated wastewater and the carriers. In all tests, the DO concentration was monitored for 10 min and the data were collected every one second.

2.5. Calculations

The MBBR performances were evaluated in terms of organic matter removal (or COD removal) and were expressed as CODr (%) according to Equation (1).

$$CODr (\%) = \frac{(tCOD_i - sCOD_e)}{tCOD_i} \times 100 \quad (1)$$

where tCOD_i is the tCOD of the wastewater influent (mg·L⁻¹) and sCOD_e is sCOD of the effluent (mg·L⁻¹).

The Volumetric Organic Loading rates (VOL) ($\text{gCOD}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$) of the influent wastewater were calculated according to Equation (2).

$$VOL = \frac{OL}{V_r} \quad (2)$$

where OL (organic loading) is the influent organic loading ($\text{kgCOD}\cdot\text{d}^{-1}$) and V_r is the working volume of MBBRs (m^3).

The Surface Organic Loading rates (SOL) ($\text{gCOD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) of the influent wastewater were calculated according to Equation (3).

$$SOL = \frac{OL}{CS} \quad (3)$$

where CS (carrier surface) is the total surface area (m^2) available for biofilm growth in each MBBR (see Table 1).

2.6. Statistical analyses

Statistical analyses were performed using GraphPad Prism software (version 6.01, USA). The ANOVA and Tukey's test were used to compare the influent and effluent wastewater quality with a significance level of 0.05.

3. RESULTS AND DISCUSSION

3.1. Nitrification

The concentration of alkalinity, pH, and DIC (bicarbonate) is usually affected by the biological oxidation of the ammonia nitrogen. The hydrogen ion production by nitrogen oxidation can decay the values of pH, alkalinity, and DIC during the reactor operation (Metcalf & Eddy 2016; Guerrero & Zaiat 2018). Souza *et al.* (2018) checked that without the alkalinity addition to the raw wastewater, the concentration of nitrogen ammonia increased in the effluent of a nitrification system. Based on this finding, it is expected that the nitrification process is not effective in matrices with low buffering capacity (e.g., domestic wastewater) and the occurrence will decay the alkalinity significantly. However, the nitrification process was not observed in the three MBBRs by the results found for alkalinity, pH, and DIC (Figure 2). The alkalinity values observed were $187 \pm 55 \text{ mg CaCO}_3\cdot\text{L}^{-1}$ for the influent and 191 ± 53 , 193 ± 53 , $186 \pm 44 \text{ mg CaCO}_3\cdot\text{L}^{-1}$ for R1, R2, and R3 effluent, respectively. The pH values found were 7.5 ± 0.2 for the influent and 7.4 ± 0.2 , 7.5 ± 0.2 , and 7.4 ± 0.2 for R1, R2, and R3 effluent, respectively. The DIC concentration observed was $50 \pm 4 \text{ mg}\cdot\text{L}^{-1}$ for the influent and 52 ± 6 , 56 ± 11 , and $53 \pm 4 \text{ mg}\cdot\text{L}^{-1}$ for R1, R2, and R3 effluent, respectively. No significant differences were found for alkalinity, pH, and DIC concentration observed in the influent and effluent from the three MBBRs (Tukey's test, $p > 0.05$). These

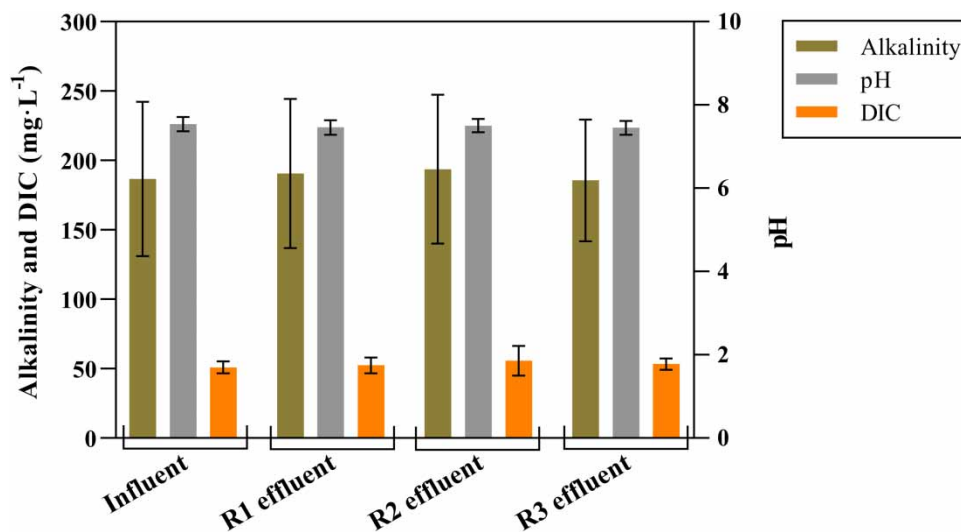


Figure 2 | Average concentration of alkalinity, pH, and DIC found in the wastewater influent and the effluent from the three MBBRs.

findings are not in accordance with those observed previously at nitrification bioreactors for domestic wastewater treatment that decreased pH value, alkalinity, and inorganic carbon (Yuan *et al.* 2020; Bressani-Ribeiro *et al.* 2021; Freitas *et al.* 2021).

For efficient nitrification in attached biofilm, the organic matter concentration must be removed before the establishment of nitrifying organisms, once heterotrophic organisms have a higher biomass yield than nitrifying bacteria and it dominates the carrier's surface (Metcalf & Eddy 2016). Due to that, the organic matter concentration must be maintained low for ammonia nitrogen oxidation in reactors treating both organic and nitrogen compounds (Campos *et al.* 1999). In this study, the reactors were operated with a high COD/N ratio (7.15) once no pre-treatment was applied, which explained the absence of the nitrification process during the operation. A high concentration of organic compounds in the oxidation-nitrification zone leads to a competition for oxygen uptake in the biofilm between heterotrophic and autotrophic organisms for organic matter and ammonia oxidation, respectively (Azimi *et al.* 2007), with an advantage to the first one over the nitrification process.

The absence of the nitrification process may also be caused due to the low HRT used in the tests. The MBBRs were operated at HRT of 2 h, which is lower than 7–8 h and 10–13 h required for the duplication of ammonia-oxidizing-bacteria (AOB) and nitrite-oxidizing-bacteria (NOB), respectively (Peng & Zhu 2006). Bassin *et al.* (2016) observed that the HRT reduction from 6 to 3 h had a negative impact on nitrogen oxidation, even though both organic and nitrogen loading rates were kept constant during the operation. Based on these results, a high concentration of COD and low HRT may impact negatively on the growth and attachment of nitrifying organisms on the carriers.

3.2. Organic matter removal

Organic matter is still the main pollutant of sewage in developing countries like Brazil (Brasil 2019). Hence, the main goal of domestic wastewater treatment is to reduce the organic loading before it is released into the environment. The influent and effluent COD concentration and the MBBRs performance over the operation time are shown in Figure 3.

The COD concentration was $306 \pm 44 \text{ mg}\cdot\text{L}^{-1}$ for the influent and 62 ± 24 , 61 ± 19 , and $67 \pm 14 \text{ mg}\cdot\text{L}^{-1}$ for R1, R2, and R3 effluent, respectively. The overall performance for organic matter removal was 80 ± 5.0 , 80 ± 3.5 , and $78 \pm 2.4\%$ for R1, R2, and R3 effluent, respectively. Significant COD removal was observed for the three MBBRs (Tukey's test, $p < 0.05$), and no significant difference was observed between the three MBBRs' performance during the experimental operation (Tukey's test, $p > 0.05$).

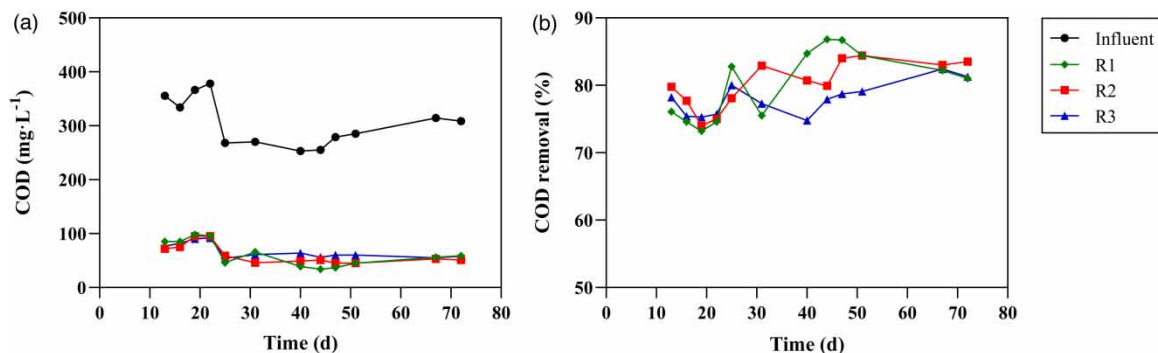


Figure 3 | (a) The COD concentration of the influent and MBBR effluents (R1, R2, and R3), and (b) the COD removal over the operation period.

A good performance was achieved since the beginning of MBBR operation due to the fast growth of the biofilm on the carriers' surface, which is typically observed in aerobic reactors (Yang *et al.* 2018). The results found are in agreement with other studies that achieved stable biofilm growth and high COD removal (75–82%) at the beginning of operation (6–17 days) (Gonzalez *et al.* 2011; Dias *et al.* 2018). However, the overall COD removals observed in the three MBBRs (78–80%) were lower than those reported for aerobic MBBR (85–88%) (Azizi *et al.* 2013; Zinatizadeh & Ghaytooli 2015). The slight difference in the performance may happen due to the limited hydrolysis of the particulate organic matter present in the influent, which depends on the amount of biomass present in the MBBR and the HRT applied in the bioreactor (Ødegaard *et al.* 2000). Then, with the use of low HRT (2 h) it is difficult to achieve a better organic matter removal due to the negative effect on the hydrolysis

process, as previously observed by Wang *et al.* (2018). The increase of HRT could be a strategy to improve the COD removal. For instance, Dias *et al.* (2018) found the average COD removal of 88% at HRT of 19 h.

Nevertheless, the results found in this study are in agreement with other studies which evaluated the MBBRs' performance by organic matter removal (Table 3). Wei *et al.* (2016) investigated three lab-scale MBBRs for real wastewater treatment at different carrier filling ratios of 40, 50, and 60%. COD removal of 70% was achieved by the three reactors and this performance was lower than obtained in this study (78–80%) with a carrier filling ratio of 50%.

Table 3 | Comparison of COD removal carried out in MBBR with different conditions

Wastewater	tCOD influent (mg·L ⁻¹)	Volume (L)	HRT (h)	Filling ratio (%)	Type of carrier	Carrier area (m ² ·m ⁻³)	RE (%)	Reference
Real wastewater	236–389	2,000	2.6–19	60	Recycled (PP)	112–610	80–88	Dias <i>et al.</i> (2018)
Residential	632	11	3	40	Corrugated tube (PP)	350	85	Azizi <i>et al.</i> (2013)
Synthetic wastewater	500–8,000	2.2	8	50	Kaldnes K1	500	45–95	Aygun <i>et al.</i> (2008)
Synthetic wastewater	300–350	8.66	7	40–60	Carrier cylindrical (HDPE)	Nr	70	Wei <i>et al.</i> (2016)
					K1 (HDPE)	500	80	This study
Domestic wastewater	306	7.5	2	50	Corrugated tube (HDPE)	390	80	
					Flakes (HDPE)	1,000	78	

Note: Nr, not reported; PP, Polypropylene; RE (%), organic matter removal.

It is important to note that all studies (Table 3) were carried out in MBBRs with different volumes, wastewater, HRT, carrier filling ratio, type, and surface area of carriers. Besides these operational differences, similar performances were obtained. These results indicate that MBBR filling with alternative carriers without protected surface (HDPE flakes) can remove organic matter as well as an alternative (corrugated tube) or commercial carrier (K1) widely employed for wastewater treatment.

3.3. Volumetric and surface organic loading rates

The reactors were fed with the same raw domestic wastewater and influent flow rate, which results in the same VOL influent of $3.60 \pm 0.50 \text{ kgCOD} \cdot \text{m}^{-3} \cdot \text{d}^{-1}$. The removal of 2.92 ± 0.30 , 2.93 ± 0.35 , and $2.86 \pm 0.40 \text{ kg COD} \cdot \text{m}^{-3} \cdot \text{d}^{-1}$ were achieved for R1, R2, and R3, respectively. The VOL influent and VOL removal are shown in Figure 4. The better VOL removal occurred in the R2 filled with the corrugated tube, with a close performance to those achieved by R1 or R3.

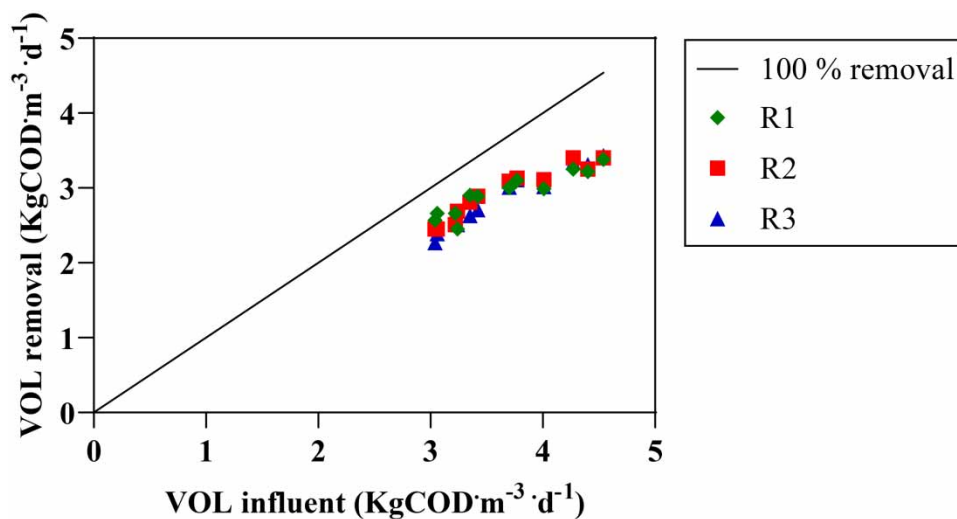


Figure 4 | Volumetric Organic Loading (VOL) influent and VOL removal for the three reactors (R1, R2, and R3).

The VOL influent applied in this study ($3.60 \pm 0.5 \text{ kgCOD}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$) agrees with previous studies using MBBRs for wastewater treatment. VOL values ranging from 0.8 to $5.2 \text{ kg COD}\cdot\text{m}^{-3}\cdot\text{d}^{-1}$ were applied to MBBRs with COD removals ranging from 90 to 95% (Marques *et al.* 2008; Bassin *et al.* 2016). Despite the lower performance reached by MBBRs (78–80%) here, the mentioned studies used synthetic wastewater which is more easily degradable than real domestic wastewater used in this study.

It is usual to correlate the organic matter loading applied to MBBR with the total surface area of the carriers, due to its surface availability for the growth of attached biofilm (Ødegaard *et al.* 2000). The SOL influent and SOL removal are shown in Figure 5. Surface organic loading of 11 ± 1.6 , 19 ± 2.7 , and $7 \pm 1.1 \text{ gCOD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ were applied to R1, R2, and R3, respectively. In this study, the SOL influent differences were due to the total surface area available by carriers in each MBBR (see Table 1). The surface organic loading removal of 8.7 ± 0.9 , 15 ± 1.8 , and $5.7 \pm 0.8 \text{ gCOD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ were achieved by R1, R2, and R3, respectively.

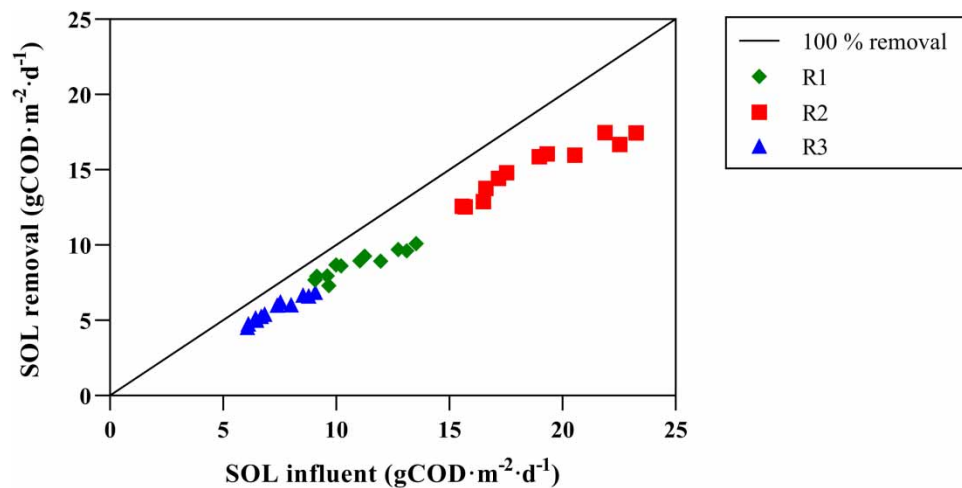


Figure 5 | Surface Organic Loading (SOL) influent and SOL removal for the three reactors (R1, R2, and R3).

Vieira *et al.* (2014) evaluated a pilot-scale MBBR to treat pulp and paper mill wastewater and the SOL influent of $43.8 \text{ g BOD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ was applied to the reactor. This value is much higher than the SOL influent of this study ($7\text{--}19 \text{ g COD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$), which can be explained by the filling ratio and wastewater quality. The authors used a filling ratio of 10% to treat a high COD influent of $1,384 \text{ mg}\cdot\text{L}^{-1}$, leading to a reduced total carrier surface available and a high SOL influent. The SOL influent in this study are according to the values applied to the MBBRs ($3.2\text{--}12.8 \text{ g COD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) (Bassin *et al.* 2016). The SOL influent extremely high can difficult the wastewater treatment by biological processes. Aygun *et al.* (2008) observed that the SOL increment from 6 to $96 \text{ g COD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ reduced the COD removal from 95.1 to 45.2%.

Ødegaard (2006) recommended that SOL values should not exceed $20\text{--}25 \text{ g sCOD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ in MBBRs, which corresponds to $65\text{--}85 \text{ g tCOD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ in typical wastewater in high-rate systems. The SOL values applied in this study are very much lower than the recommended values, which makes the operation of the MBBRs used in this study a low-middle rate (values higher than $6 \text{ gCOD}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) (Aygun *et al.* 2008). Carriers with different specific surface and low substrate loading rates also lead to similar performance (Wei *et al.* 2016), once a less specific surface is subjected to a higher SOL rate and also to a high COD removal (Ødegaard *et al.* 2000). These results confirm a high correlation between VOL applied, as well as SOL values, and the COD degradation rates achieved by the reactor.

3.4. Respirometric analysis of the biofilm

The respirometry assays aimed to evaluate the DO consumption by the attached biofilm to each type of carrier. The biofilm growth on the carrier surface leads to an increment in the fluid density and consequently, the residence time of the air bubble inside the bioreactor treating wastewater also increases (Collivignarelli *et al.* 2019). The DO consumptions by biofilm on the carriers are shown in Figure 6.

No significant DO variation was observed for wastewater filtered samples (blank) and the values ranged from 6.5 to $6.7 \text{ mg}\cdot\text{L}^{-1}$. The DO concentration decreased from 6.73 to $5.28 \text{ mg}\cdot\text{L}^{-1}$ for K1 carrier, from 6.73 to $6.10 \text{ mg}\cdot\text{L}^{-1}$ for corrugated tube carrier, and from 6.82 to $6.5 \text{ mg}\cdot\text{L}^{-1}$ for HDPE flakes carrier. Then, the DO

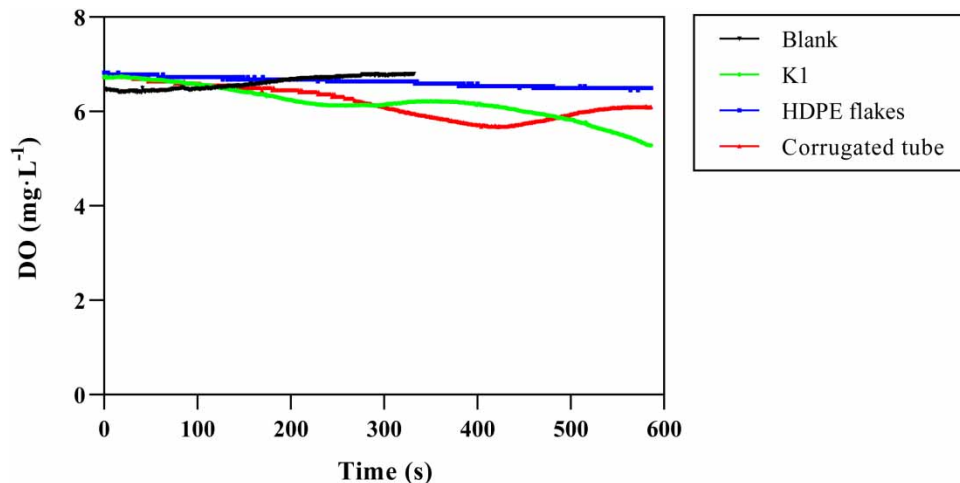


Figure 6 | Dissolved oxygen depletion by attached biofilm on the three carriers.

consumption observed were 0.022, 0.009, and 0.005 mg of DO for K1, corrugated tube, and HDPE flakes, respectively. These values evidence the activity of the biofilm attached to the carriers, and also that the K1 carriers showed the highest surface oxygen uptake.

The surface DO consumptions of 0.0079 ± 0.0013 , 0.0033 ± 0.0015 , and $0.0031 \pm 0.0026 \mu\text{gDO}\cdot\text{mm}^{-2}$ were observed for K1, corrugated tube, and HDPE flakes, respectively. HDPE flakes showed the lowest DO consumption, mainly due to the lack of a protected surface. The protected surface area present in the corrugated tube and K1 carrier has more ability to be colonized by biofilm (Aygün *et al.* 2008; Torresi *et al.* 2017; Tombola *et al.* 2019).

In this work, the oxygen consumption demonstrated in the respirometry essay is due to the biological organic matter oxidation by heterotrophic microorganisms attached to the carrier surface (Metcalf & Eddy 2016). Then, it was expected a higher COD removal for K1 carriers due to the highest DO consumption. However, as previously shown, no statistical difference was observed for COD removal obtained in three MBBRs. This difference may happen due to irregular and heterogeneous growth of active biofilm on the carrier's surface (Denkhaus *et al.* 2007). Furthermore, the protected surface of carriers can also show weak correlations with the COD removal by biofilm under some conditions (Dias *et al.* 2018). Hence, it is not reasonable to assume that all available surfaces of carriers in each reactor was colonized by active microorganisms.

The results showed that the COD removal was independent of the oxygen uptake obtained in the tests. Moreover, the results found for alternative carriers without protected areas are similar to those reported in the literature for a carrier with a protected area (Azizi *et al.* 2013; Wei *et al.* 2016; Santos *et al.* 2020).

CONCLUSIONS

The present study investigated the organic matter removal from raw domestic wastewater by MBBR using carriers with (K1 and corrugated tube) and without (HDPE flakes) protected surface. The nitrification process was not observed during the operation of the bioreactors. The COD removal of 80 ± 5.0 , 80 ± 3.5 , and $78 \pm 2.4\%$ was achieved by R1, R2, and R3, respectively. The oxygen uptake by biofilm attached on the carriers was 0.0079 ± 0.0013 , 0.0033 ± 0.0015 , and $0.0031 \pm 0.0026 \mu\text{g DO}\cdot\text{mm}^{-2}$ for the K1, corrugated tube, and HDPE flakes, respectively. No significant differences were observed between the three MBBRs performance in terms of physico-chemical parameters (alkalinity, pH, and dissolved inorganic carbon) and COD removal. Thus, the results showed that the carrier type and its characteristics (total area and with/without protected area) did not affect the organic matter removal.

ACKNOWLEDGEMENTS

This work was supported by the São Paulo Research Foundation (FAPESP) for the research assistance [Proc. 2017/00088-6], National Council for Scientific and Technological Development (CNPQ) for PhD scholarship [Proc. 141476/2016-8 and 302412/2017-4], and the Coordination for the Improvement of Higher Education Personnel- Brazil (CAPES) [Finance Code 001].

DECLARATION OF INTEREST STATEMENT

No potential conflict of interest.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES

- APHA 2012 Standard Methods for Examination of Water and Wastewater, 22nd edn. American Public Health Association, American Water Works Association, Water Environment Federation, Washington, DC.
- Araujo Junior, M. M., Lermontov, A., Araujo, P. L. d. S. & Zaiat, M. 2013 Reduction of sludge generation by the addition of support material in a cyclic activated sludge system for municipal wastewater treatment. *Bioresour. Technol.* **143**, 483–489. <https://doi.org/10.1016/j.biortech.2013.06.032>.
- Aygun, A., Nas, B. & Berktaş, A. 2008 Influence of high organic loading rates on COD removal and sludge production in moving bed biofilm reactor. *Environ. Eng. Sci.* **25**, 1311–1316. <https://doi.org/10.1089/ees.2007.0071>.
- Azimi, A. A., Hooshyari, B., Mehrdadi, N. & Nabi Bidhendi, G. 2007 Enhanced COD and nutrient removal efficiency in a hybrid integrated fixed film activated sludge process. *Iran. J. Sci. Technol. Trans. B Eng.* **31**, 523–533. <https://doi.org/10.22099/ijstc.2007.752>.
- Azizi, S., Valipour, A. & Sithebe, T. 2013 Evaluation of different wastewater treatment processes and development of a modified attached growth bioreactor as a decentralized approach for small communities. *Sci. World J.* **2013**, 1–8. <https://doi.org/10.1155/2013/156870>.
- Bassin, J. P., Dias, I. N., Cao, S. M. S., Senra, E., Laranjeira, Y. & Dezotti, M. 2016 Effect of increasing organic loading rates on the performance of moving-bed biofilm reactors filled with different support media: assessing the activity of suspended and attached biomass fractions. *Process Saf. Environ. Prot.* **100**, 131–141. <https://doi.org/10.1016/j.psep.2016.01.007>.
- Brasil 2019 Brasil. Ministério do Desenvolvimento Regional. Secretaria Nacional de Saneamento – SNS. Sistema Nacional de Informações sobre Saneamento: 24º Diagnóstico dos Serviços de Água e Esgotos – 2018. SNS/MDR, Brasília, p. 180. il.
- Bressani-Ribeiro, T., Almeida, P. G. S., Chernicharo, C. A. L. & Volcke, E. I. P. 2021 Inorganic carbon limitation during nitrogen conversions in sponge-bed trickling filters for mainstream treatment of anaerobic effluent. *Water Res.* **201**, 117337. <https://doi.org/10.1016/j.watres.2021.117337>.
- Campos, J. L., Garrido-Fernández, J. M., Méndez, R. & Lema, J. M. 1999 Nitrification at high ammonia loading rates in an activated sludge unit. *Bioresour. Technol.* **68**, 141–148. [https://doi.org/10.1016/S0960-8524\(98\)00141-2](https://doi.org/10.1016/S0960-8524(98)00141-2).
- Collivignarelli, M. C., Abbà, A. & Bertanza, G. 2019 Oxygen transfer improvement in MBBR process. *Environ. Sci. Pollut. Res.* **26**, 10727–10737. <https://doi.org/10.1007/s11356-019-04535-1>.
- Denkhaus, E., Meisen, S., Telgheder, U. & Wingender, J. 2007 Chemical and physical methods for characterisation of biofilms. *Microchim. Acta* **158**, 1–27. <https://doi.org/10.1007/s00604-006-0688-5>.
- Dias, J., Bellingham, M., Hassan, J., Barrett, M., Stephenson, T. & Soares, A. 2018 Influence of carrier media physical properties on start-up of moving attached growth systems. *Bioresour. Technol.* **266**, 463–471. <https://doi.org/10.1016/J.BIORTECH.2018.06.096>.
- di Biase, A., Kowalski, M. S., Devlin, T. R. & Oleszkiewicz, J. A. 2019 Moving bed biofilm reactor technology in municipal wastewater treatment: a review. *J. Environ. Manage.* **247**, 849–866. <https://doi.org/10.1016/j.jenvman.2019.06.053>.
- Freitas, B. O., Leite, L. S. & Daniel, L. A. 2021 Chlorine and peracetic acid in decentralized wastewater treatment: disinfection, oxidation and odor control. *Process Saf. Environ. Prot.* **146**, 620–628. <https://doi.org/10.1016/j.psep.2020.11.047>.
- Gonzalez, B. C., Spínola, A. L. G., Lamon, A. W., Araujo, J. C. & Campos, J. R. 2011 The use of microsensors to study the role of the loading rate and surface velocity on the growth and the composition of nitrifying biofilms. *Water Sci. Technol.* **64**, 1607–1613. <https://doi.org/10.2166/wst.2011.716>.
- Guerrero, R. B. S. & Zaiat, M. 2018 Wastewater post-treatment for simultaneous ammonium removal and elemental sulfur recovery using a novel horizontal mixed aerobic-anoxic fixed-bed reactor configuration. *J. Environ. Manage.* **215**, 358–365. <https://doi.org/10.1016/j.jenvman.2018.03.074>.
- Levstek, M. & Plazl, I. 2009 Influence of carrier type on nitrification in the moving-bed biofilm process. *Water Sci. Technol.* **59**, 875–882. <https://doi.org/10.2166/wst.2009.037>.
- Marques, J. J., Souza, R. R., Souza, C. S. & Rocha, I. C. C. 2008 Attached biomass growth and substrate utilization rate in a moving bed biofilm reactor. *Brazilian J. Chem. Eng.* **25**, 665–670. <https://doi.org/10.1590/S0104-66322008000400004>.
- Massoompour, A. R., Borghei, S. M. & Raie, M. 2020 Enhancement of biological nitrogen removal performance using novel carriers based on the recycling of waste materials. *Water Res.* **170**, 115340. <https://doi.org/10.1016/j.watres.2019.115340>.
- Mcquarrie, J., Boltz, J., Mcquarrie, J. P. & Boltz, J. P. 2011 Moving bed biofilm reactor technology: process applications, design, and performance moving bed biofilm reactor technology: process applications, design, and performance. *Water Environ. Res.* **83**, 560–575. <https://doi.org/10.2175/106143010X12851009156286>.
- Metcalf & Eddy 2016 *Tratamento de efluentes e recuperação de recursos*, 5a. edn. McGraw-Hill, New York, NY.
- Ødegaard, H. 2006 Innovations in wastewater treatment: the moving bed biofilm process. *Water Sci. Technol.* **53**, 17–33. <https://doi.org/10.2166/wst.2006.284>.

- Ødegaard, H., Rusten, B. & Westrum, T. 1994 A new moving bed biofilm reactor – applications and results. *Water Sci. Technol.* **29**, 157–165. <https://doi.org/https://doi.org/10.2166/wst.1994.0757>.
- Ødegaard, H., Gisvold, B. & Strickland, J. 2000 The influence of carrier size and shape in the moving bed biofilm process. *Water Sci. Technol.* **41**, 383–391. <https://doi.org/10.2166/wst.2000.0470>.
- Oliveira, D. V. M. d., Volschan, I. & Piveli, R. P. 2013 Avaliação comparativa entre custos dos processos MBBR/IFAS e lodo ativado para o tratamento de esgoto sanitário. *Rev. DAE* **61**, 46–55. <https://doi.org/10.4322/dae.2014.110>.
- Peng, Y. & Zhu, G. 2006 Biological nitrogen removal with nitrification and denitrification via nitrite pathway. *Applied Microbiology and Biotechnology* **73** (1), 15–26. <https://doi.org/10.1007/s00253-006-0534-z>.
- Piculell, M., Welander, T. & Joñsson, K. 2014 Organic removal activity in biofilm and suspended biomass fractions of MBBR systems. *Water Sci. Technol.* **69**, 55–61. <https://doi.org/10.2166/wst.2013.552>.
- Sabzali, A., Nikaeen, M. & Bina, B. 2012 Performance evaluation of cigarette filter rods as a biofilm carrier in an anaerobic moving bed biofilm reactor. *Environ. Technol. (United Kingdom)* **33**, 1803–1810. <https://doi.org/10.1080/09593330.2011.646317>.
- Saidi, A., Masmoudi, K., Nolde, E., El Amrani, B. & Amraoui, F. 2017 Organic matter degradation in a greywater recycling system using a multistage moving bed biofilm reactor (MBBR). *Water Sci. Technol.* **76**, 3328–3339. <https://doi.org/10.2166/wst.2017.499>.
- Santos, A. D., Martins, R. C., Quinta-Ferreira, R. M. & Castro, L. M. 2020 Moving bed biofilm reactor (MBBR) for dairy wastewater treatment. *Energy Reports* **6**, 340–344. <https://doi.org/10.1016/j.egy.2020.11.158>.
- Sonwani, R. K., Swain, G., Giri, B. S., Singh, R. S. & Rai, B. N. 2019 A novel comparative study of modified carriers in moving bed biofilm reactor for the treatment of wastewater: process optimization and kinetic study. *Bioresour. Technol.* **281**, 335–342. <https://doi.org/10.1016/j.biortech.2019.02.121>.
- Souza, T. S. O., Okada, D. Y. & Foresti, E. 2018 Proof of concept and improvement of a triple chamber biosystem coupling anaerobic digestion, nitrification and mixotrophic endogenous denitrification for organic matter, nitrogen and sulfide removal from domestic sewage. *Bioprocess Biosyst. Eng.* **41**, 1839–1850. <https://doi.org/10.1007/s00449-018-2006-0>.
- Tombola, R., Buttiglieri, G., Auset, M. & Gonzalez-Olmos, R. 2019 Recycled corrugated wire hose cover as biological carriers for greywater treatment in a sequential batch biofilm reactor. *J. Environ. Manage.* **240**, 475–484. <https://doi.org/10.1016/j.jenvman.2019.02.116>.
- Torresi, E., Escolà Casas, M., Polesel, F., Plósz, B. G., Christensson, M. & Bester, K. 2017 Impact of external carbon dose on the removal of micropollutants using methanol and ethanol in post-denitrifying Moving Bed Biofilm Reactors. *Water Res.* **108**, 95–105. <https://doi.org/10.1016/j.watres.2016.10.068>.
- Vieira, D., Oliveira, M. D., Rabelo, M. D. & Nariyoshi, Y. N. 2014 Evaluation of a MBBR (moving bed biofilm reactor) pilot plant for treatment of pulp and paper mill wastewater. *Int. J. Environ. Monit. Anal.* **2**, 220–225. <https://doi.org/10.11648/j.ijema.20140204.15>.
- Wang, Y., Liu, Y., Feng, M. & Wang, L. 2018 Study of the treatment of domestic sewage using PVA gel beads as a biomass carrier. *J. Water Reuse Desalin.* **8**, 340–349. <https://doi.org/10.2166/wrd.2017.181>.
- Wei, Y., Yin, X., Qi, L., Wang, H., Gong, Y. & Luo, Y. 2016 Effects of carrier-attached biofilm on oxygen transfer efficiency in a moving bed biofilm reactor. *Front. Environ. Sci. Eng.* **10**, 569–577. <https://doi.org/10.1007/s11783-015-0822-x>.
- Wolff, D. B., Ochoa, J. C., Paul, E. & Ribeiro Da Costa, R. H. 2005 Nitrification in hybrid reactor with a recycled plastic support material. *Brazilian Arch. Biol. Technol.* **48**, 243–248. <https://doi.org/Doi 10.1590/S1516-89132005000400030>.
- Yang, Y., Shao, Z., Du, J., He, Q. & Chai, H. 2018 Enhancement of organic matter removal in an integrated biofilm-membrane bioreactor treating high-salinity wastewater. *Archaea* **2018**, 1–8. <https://doi.org/10.1155/2018/2148286>.
- Yuan, C., Wang, B., Peng, Y., Li, X., Zhang, Q. & Hu, T. 2020 Enhanced nutrient removal of simultaneous partial nitrification, denitrification and phosphorus removal (SPNDPR) in a single-stage anaerobic/micro-aerobic sequencing batch reactor for treating real sewage with low carbon/nitrogen. *Chemosphere* **257**, 127097. <https://doi.org/10.1016/j.chemosphere.2020.127097>.
- Zhao, Y., Yuan, Q., He, Z., Wang, H., Yan, G., Chang, Y., Chu, Z., Ling, Y. & Wang, H. 2019 Influence of carrier filling ratio on the advanced nitrogen removal from wastewater treatment plant effluent by denitrifying MBBR. *Int. J. Environ. Res. Public Heal.* **16**, 1–12. <https://doi.org/10.3390/ijerph16183244>.
- Zinatizadeh, A. A. L. & Ghaytooli, E. 2015 Simultaneous nitrogen and carbon removal from wastewater at different operating conditions in a moving bed biofilm reactor (MBBR): process modeling and optimization. *J. Taiwan Inst. Chem. Eng.* **53**, 98–111. <https://doi.org/10.1016/j.jtice.2015.02.034>.

First received 17 August 2021; accepted in revised form 23 December 2021. Available online 11 January 2022