

1 **Performance of a novel baffled osmotic membrane bioreactor-microfiltration hybrid**  
2 **system under continuous operation for simultaneous nutrient removal and mitigation**  
3 **of brine discharge.**

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20 **Abstract**

21 The present study investigated the performance of an integrated osmotic and microfiltration  
22 membrane bioreactor system for wastewater treatment employing baffles in the reactor.  
23 Thus, this reactor design enables both aerobic and anoxic processes in an attempt to reduce  
24 the process footprint and energy costs associated with continuous aeration. The process  
25 performance was evaluated in terms of water flux, salinity build up in the bioreactor,  
26 organic and nutrient removal and microbial activity using synthetic reverse osmosis (RO)  
27 brine as draw solution (DS). The incorporation of MF membrane was effective in  
28 maintaining a reasonable salinity level (612-1434 mg/L) in the reactor which resulted in a  
29 much lower flux decline (i.e. 11.48 to 6.98 LMH) as compared to previous studies. The  
30 stable operation of the osmotic membrane bioreactor–forward osmosis (OMBR-FO)  
31 process resulted in an effective removal of both organic matter (97.84 %) and nutrient  
32 phosphate ( $\text{PO}_4\text{-P}$ ) 87.36 % and total nitrogen (TN) 94.28 % respectively.

33 **Keywords:** OMBR; microfiltration (MF); simultaneous nitrification-denitrification (SND);  
34 salinity build-up; biomass activity.

## 35 **1 Introduction**

36 With rapidly growing world population, the water scarcity issue is becoming critical and  
37 affects drinking water supplies, energy, food production, industrial output, and the quality  
38 of our environment at a global scale (Phuntsho et al., 2011). Hence, the emphasis on  
39 developing alternative approaches to supply ‘fit for purpose’ water is emerging. Therefore,  
40 an alternative water supply systems are becoming a visible practice in many water stressed  
41 regions (Memon & Ward, 2014). A paradigm shift is already taking place and low grade  
42 water such as grey water and sewage are now increasingly seen as a viable source of water,  
43 nutrient and energy rather than a waste (Memon & Ward, 2014; Wang et al., 2016a).  
44 However, there are still many challenges faced in wastewater treatment processes,  
45 especially in relation to nutrient and trace organic removal (Nguyen et al., 2016). In  
46 particular, nutrient removal is very important for water reuse, especially in water supply for  
47 outdoor use, to prevent water quality deterioration via eutrophication. This phenomenon is  
48 mainly caused by the presence of nitrogen and phosphorus in wastewater effluents, leading  
49 to algal blooms, oxygen deficiency, and ultimately an increased mortality of the aquatic  
50 species (Devia et al., 2015; Mun et al., 2011).

51 Conventional techniques for N and P removal from wastewater are based on physical and  
52 chemical methods. These techniques are not economical and do not facilitate nutrients  
53 recycle and reuse (Praveen & Loh, 2016). Fan et al. (1996) reported that perfect  
54 nitrification could be achieved in the membrane bioreactor (MBR) system. In order to  
55 achieve simultaneous nitrification-denitrification, a continuous aerated submerged MBR  
56 system for nitrification with a separated anoxic tank for denitrification was developed (Ahn  
57 et al., 2003). The energy required for sludge recirculation and mixing in an anoxic tank

58 accounts for 10–20% of the total energy consumption in a common MBR. This is in  
59 addition to the energy usage for aeration to mitigate membrane fouling which represents up  
60 to 60–70% of the total operating energy in MBRs (Kurita et al., 2015). To overcome these  
61 shortcomings, researchers introduced the alternating anoxic and oxic conditions in a  
62 submerged MBR by intermittent aeration for total nitrogen removal. However, in the  
63 intermittently aerated MBR, filtration operation is limited to the aeration periods, mainly to  
64 prevent membrane fouling (Song et al., 2010).

65 In recent years, more studies have shown that nitrification and denitrification could occur  
66 concurrently in one single reactor under aerobic conditions with low dissolved oxygen,  
67 through the so-called simultaneous nitrification and denitrification (SND) process. SND  
68 relies on concurrent aerobic  $\text{NH}_4\text{-N}$  oxidation and anoxic denitrification under identical  
69 operating conditions (Fu et al., 2009). Kimura and Watanabe (2005) have proposed a  
70 baffled membrane bioreactor, in which baffles are inserted in a submerged MBR, and the  
71 level of water in the reactor is controlled to facilitate simultaneous  
72 nitrification/denitrification without sludge recirculation. The inner zone of the baffles  
73 maintains an aerobic condition because of aeration, whereas the outer zone alternates  
74 between aerobic and anoxic conditions (Kimura & Watanabe, 2005). Thus, a baffled MBR  
75 offers advantages such as small footprint (no additional anoxic tank) and baffle design  
76 substitutes stirring of anoxic biomass and sludge recycle between oxic and anoxic tank. The  
77 baffles inserted in the MBR create circulation flows in the membrane tank and vigorously  
78 mix the biomass. This also improves the efficiency of mechanical cleaning (Kimura et al.,  
79 2007).

80 More recently, osmotic membrane bioreactors (OMBRs) have attracted growing interests in  
81 the field of low strength domestic/municipal wastewater treatment (Wang et al., 2016c).  
82 OMBR can potentially produce high quality reclaimed water for potable reuse, irrigation, or  
83 direct discharge in environmentally sensitive areas (Luo et al., 2015). OMBR have many  
84 advantages such as higher quality pure water, low and reversible fouling compared to  
85 pressure-driven membrane processes, minimum cleaning and energy efficient in the  
86 absence of any hydraulic pressure in comparison to the traditional membrane bioreactors  
87 (MBRs) (Luo et al., 2017; Wang et al., 2016b).

88 However, OMBR also has some limitations such as salinity build-up (i.e. accumulation of  
89 dissolved salts inside the bioreactor), internal concentration polarization and the energy  
90 associated with the DS recovery process (Nguyen et al., 2016). In order to mitigate the  
91 salinity build up, various approaches have been tested including operating at short sludge  
92 retention time (SRT) (Wang et al., 2014b). However, ammonia removal via biological  
93 treatment in the OMBR cannot be completed at low SRT since the nitrifying bacteria  
94 population would decrease due to their relative long generation time. In this case, the  
95 concentration of ammonia would accumulate in the mixed liquor and negatively impact on  
96 the microorganisms in the OMBR. Moreover, diffusion of high concentration ammonia  
97 across the FO membrane eventually leads to the deterioration of permeate quality (Wang et  
98 al., 2014b; Yap et al., 2012). Therefore, for long-term operation of the OMBR,  
99 incorporation of microfiltration (MF)/ultrafiltration (UF) membrane (Holloway et al.,  
100 2015a; Holloway et al., 2014) has been suggested as a promising option. MF/UF are also

101 helpful to mitigate salinity build up because these membranes could let the salts pass  
102 through but retain the activated sludge (Wang et al., 2014a).

103 The amount of concentrate produced from a desalination plant is a factor of the desalination  
104 recovery rate. Literature review shows that the concentrate produced from seawater reverse  
105 osmosis (SWRO) plants have up to two times more salt concentration than the receiving  
106 water since they generally operate at 50% recovery rate (Tularam & Ilahee, 2007). As  
107 reported by (Abualtayef et al., 2016), the potential harm of brine to the environment yields  
108 from either its higher than normal salinity compared to point of discharge, or due to  
109 pollutants that otherwise would not be present in the receiving waterbody. These pollutants  
110 include chlorine and other biocides, heavy metals, anti-scalants, coagulants, and cleaning  
111 chemicals (Abualtayef et al., 2016). In addition to the destructive saline properties of the  
112 concentrate, in the case of thermal desalination, the brine is usually hotter than the local  
113 recipient water body, a circumstance that has also been shown to cause further  
114 environmental damage, especially to fragile ecosystems such as corals. Due to these  
115 negative effects, direct disposal to seawater of RO concentrates is doomed to disappear  
116 (Perez-Gonzalez et al., 2012).

117 There are a few actual regulations, standards, or guidelines for brine discharges around the  
118 world particularly in the developed countries like the US, Australia and Israel. There is  
119 substantial variation in the specifics of the regulations, but almost all share two key  
120 elements: a salinity limit and a point of compliance expressed as a distance from the  
121 discharge. (Jenkins et al., 2012) reviewed RO concentrate discharge regulations and  
122 standards which have been applied around the world. These range from salinity increments

123 within 1 parts per thousand (ppt), 5%, or absolute levels such as 40 ppt. These limits  
124 typically apply at the boundary of a mixing zone whose dimensions are of order 50 to 300  
125 m around the discharge (Jenkins et al., 2012). In contrast, inland plants have to solve the  
126 problem of concentrate disposal without the possibility of their discharge to seawater, so  
127 the development of other management options is an urgent demand (Perez-Gonzalez et al.,  
128 2012). Although the TDS values of RO concentrates from brackish water desalting are  
129 significantly less than seawater TDS, they are typically greater than 10,000 mg/L, which  
130 makes them more compatible with ocean water than fresh waters. In this scenario OMBR  
131 could be a viable alternative. In this context, forward osmosis-membrane bioreactor (FO-  
132 MBR) is presented as an innovative technique to mitigate RO brine discharge. The FO-  
133 MBR process may also demonstrate a lower membrane fouling propensity than pressure-  
134 driven membrane processes such as conventional MBR. Membrane fouling in FO is  
135 relatively low, more reversible and can be minimized by optimizing hydrodynamics (Zhao  
136 et al., 2012).

137 The present study investigates for the first time the performance of an integrated osmotic  
138 and microfiltration membrane bioreactor system for municipal wastewater treatment  
139 employing baffles in the reactor. Thus, the single-stage reactor design employed here  
140 combines aerobic and anoxic processes to reduce the footprint and decrease energy costs of  
141 continuous aeration and sludge recycling in order to achieve simultaneous nitrification-  
142 denitrification. The process performance was investigated in terms of water flux and  
143 salinity build up, organic and nutrient removal and microbial activity using simulated RO  
144 brine as a DS. Further, as pointed out in several reviews (Wang et al., 2016a; Yap et al.,

145 2012) cellulose triacetate- forward osmosis (CTA-FO) membranes were predominantly  
146 utilized in early OMBRs studies. However, the loss of membrane rejection of CTA-FO  
147 membranes due to its biodegradation has been a major concern. In contrast, thin film  
148 composite (TFC) membranes possess better chemical/biological stability and separation  
149 properties. In spite of the increased interest in OMBR, there are limited reports of their  
150 operation with TFC membranes to investigate feasibility of this technology. Hence, a newly  
151 developed TFC FO membrane (Toray Korea) has been employed in the current study.

## 152 **2 Materials and methods**

### 153 **2.1 FO and MF membrane characteristics**

154 The FO membrane used in this study was a flat-sheet TFC polyamide (PA) membrane  
155 (Toray, Korea). The characteristics of this membrane are detailed in Table S1 (SI).  
156 Membranes were stored in distilled water at 4°C prior to use, and were oriented AL-FS  
157 (active layer facing feed solution) during the experiments with the feed solution being the  
158 OMBR mixed liquor. The membrane chemistry is proprietary, though it is believed that the  
159 TFC membrane has embedded polyester screen support and a negatively charge surface  
160 (Luo et al., 2016b). The submerged FO membrane module was custom designed and  
161 fabricated. Two stainless steel plates were attached to each side of the stainless steel block.  
162 The two FO membrane coupons were secured in place on each side of the stainless steel  
163 block with the stainless steel plates and then fixed using bolts and nuts. The channel in the  
164 membrane module ran through the stainless steel block having a width of 11 cm, a length of  
165 12 cm, and a depth of 0.5 cm. Mesh spacers were used on DS side to provide additional



166 support to the membrane and promote mixing of DS. Two ¼” nozzles were provided on  
167 each side of the stainless steel plates allowing DS to flow through the channel (Fig. S1  
168 supplementary information (SI)). The total effective FO membrane area was 264 cm<sup>2</sup>. The  
169 MF membrane was supplied by Uniqflux Membranes LLP, India and was made of  
170 polyethersulfone (PES) with a nominal pore size of 0.33 µm and an effective surface area  
171 of 1000 cm<sup>2</sup>. The characteristics of this membrane are shown in Table S1.

## 172 **2.2 Feed and draw solutions characteristics**

173 All the chemicals used in this research were of reagent grade (Sigma Aldrich, Australia).  
174 The influent water of the OMBR system was a synthetic municipal wastewater that  
175 consisted of 300 mg/L glucose, 50 mg/L yeast, 15 mg/L KH<sub>2</sub>PO<sub>4</sub>, 10 mg/L FeSO<sub>4</sub>, 60 mg/L  
176 (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, and 30 mg/L urea. The synthetic wastewater prepared daily and had  
177 concentrations of total organic carbon (TOC), ammonium nitrogen (NH<sub>4</sub>-N), total nitrogen  
178 and phosphate (PO<sub>4</sub>-P) of 100, 16, 28 and 3.5 mg/L, respectively. Sodium bicarbonate  
179 (NaHCO<sub>3</sub>) was used for alkalinity to maintain a neutral pH. DS were prepared by  
180 dissolving 64 g/L sodium chloride in deionized (DI) water. Osmotic pressure and  
181 diffusivity were obtained by OLI Stream Analyzer 3.2 (OLI System Inc., Morris Plains, NJ,  
182 USA). For 1.1 M NaCl, electrical conductivity and osmotic pressure are 91.26 mS/cm and  
183 51.78 atm, respectively.

184 **2.3 Baffled osmotic membrane bioreactor-microfiltration (OMBR-MF) system and**  
185 **operation**

186 A lab-scale baffled OMBR-MF system was used in this study and a schematic of the  
187 system is shown in Fig. S2 (SI). This hybrid system consisted of a feed solution reservoir, a  
188 plexiglass bioreactor with a submerged plate and-frame FO membrane cell and a hollow  
189 fiber MF membrane module, a concentrated DS reservoir and a diluted DS reservoir. The  
190 bioreactor tank (i.e., 24.5 cm length \* 15.5 cm width\* 40 cm height) had an effective  
191 volume of 11.5 L. On the three inside walls of the tank, plexiglass partition of 25 cm length  
192 was running from top to 5 cm above the bottom of the tank thus making hollow baffle box  
193 inside the tank with a the size of 18.5 cm length, 12.5 cm width and 25 cm height. The  
194 baffles were bent at approximately 30° angle in the end (3.5 cm length) to avoid dead zone  
195 formation and to attain thorough mixing of biomass. The volume ratio of the outer tank to  
196 the inner tank was approximately 1.9 when the water level was at the top of the inserted  
197 baffles (Fig. S3 SI). By changing the position of the level controller, the oxic and anoxic  
198 cycle times can be adjusted accordingly. The concept and operating details of the baffled  
199 reactor is discussed elsewhere by Kimura et al. (2007).

200 A plate-and-frame membrane module was prepared using commercial TFC FO membranes  
201 (Toray, Korea) and the module was immersed in the bioreactor tank for osmotic filtration.  
202 FO membrane samples were suspended vertically and parallel to the MF membrane module.  
203 The air diffuser was installed inside the oxic chamber of the bioreactor for oxygen supply at  
204 2 litres per minute (LPM) air-flow rate thus subjected both membranes to air scouring. The  
205 MF membrane, operated continuously, was driven by a peristaltic pump (Longer BT100 2J).

206 The MF permeate flux was changed manually in accordance with the change of FO water  
207 flux in order to maintain stable oxic and anoxic cycle times during the entire operation (i.e.  
208 38 days). A high resolution ( $\pm 0.1$  kPa) pressure sensor (Keller, Reinacherstrasse, Basel,  
209 Switzerland) was installed to record the trans-membrane pressure (TMP). Both of the  
210 concentrated and diluted DS reservoirs were placed on the weighing balance (Adam PGL  
211 15001) and connected to a computer. The weight difference between the diluted and  
212 concentrated DS was used to calculate the FO water flux. A water level controller was used  
213 to adjust the oxic-anoxic cycle time as well as to regulate the feed pump (peristaltic pump  
214 Longer WT600 2J) to feed synthetic wastewater to the bioreactor.

215 The seed sludge was collected from the recycled water facility at Central Park, Sydney,  
216 Australia. The sludge was acclimatized for a month prior to adding into the baffled OMBR-  
217 MF system. The OMBR-MF hybrid system was continuously operated for 38 days under  
218 similar conditions at a constant temperature of  $22 \pm 1^\circ\text{C}$ . The mixed liquid suspend sludge  
219 (MLSS) was adjusted to 4700 mg/L initially. Throughout baffled OMBR-MF operation, the  
220 sludge retention time (SRT) was controlled at 115 days by daily wasting 100 mL of mixed  
221 liquor from the bioreactor. The hydraulic retention time (HRT) for baffled OMBR+MF  
222 combined system was set at 30.25 h. A 1.1 M NaCl DS was used as simulated reverse  
223 osmosis (RO) brine. The concentrated DS was refilled twice a day and the diluted DS tank  
224 was emptied. The salt accumulation in the bioreactor was determined by monitoring the  
225 conductivity of the mixed liquor with a conductivity meter. The pH, total dissolved solids  
226 (TDS) and conductivity of the mixed liquor, permeate and DS were measured regularly  
227 (HACH, Germany). The operating conditions are listed in Table S2 (SI). No membrane

228 cleaning was conducted for both FO and MF membrane during the entire operation (i.e. 38  
229 days).

230 A detailed mass balance of the OMBR-MF hybrid system is also presented in Fig. S4 and  
231 Equations S1 to S11 (SI) to provide a better understanding of the salt accumulation  
232 phenomena occurring in the bioreactor.

## 233 **2.4 Analytical methods**

### 234 **2.4.1 Measurement of water flux**

235 The experimental water flux  $J_w$  (L/m<sup>2</sup> h) was calculated by measuring the net increase in  
236 diluted DS volume with time as follows:

$$J_w = \frac{\Delta V}{A\Delta t} \quad (1)$$

237 where  $\Delta V$  is the total increase in the volume of the permeate water (L) collected over a  
238 predetermined period,  $\Delta t$  (h) and  $A$  is the effective FO membrane area (m<sup>2</sup>).

### 239 **2.4.2 Biological parameters and basic water quality parameters**

240 The mixed liquid suspended solids (MLSS), mixed liquid volatile suspended solids  
241 (MLVSS) and the specific oxygen uptake rate (SOUR) of the mixed liquor in the OMBR  
242 were determined according to the APHA, AWWA, WEF (1998). The concentration of  
243 dissolved oxygen was measured by using a DO meter (Vernier, USA). TOC of the influent  
244 and effluent was measured using the Analytikjena Multi N/C 2000. Chemical oxygen  
245 demand (COD) was analysed according to standard methods (APHA, 1998). NO<sub>2</sub>-N, NO<sub>3</sub>-

246 N, NH<sub>4</sub>-N, TN and PO<sub>4</sub>-P were measured using Hach TNTplus™ reagent vials by  
247 photometric method (Spectroquant Cell Test, NOVA 60, Merck). Samples were diluted as  
248 necessary to minimize chloride interferences and ensure that analytes were within the  
249 desired range.

### 250 **2.4.3 SEM-EDX analysis**

251 The surface and cross-sectional morphologies of pristine and fouled membrane samples  
252 were observed by scanning electron microscopy and an energy diffusive X-ray (EDX)  
253 analyzer (SEM, Zeiss Supra 55VP, Carl Zeiss AG). Samples taken from each membrane  
254 were coated with gold. The SEM images were carried out at an accelerating voltage of 10  
255 kV, and different image magnifications at various areas were obtained for each sample.

## 256 **3 Results and discussion**

### 257 **3.1 Water flux and salinity build up in the baffled OMBR-MF hybrid system**

258 The OMBR-MF hybrid system was operated at constant DS concentration of approximately  
259 64 g/L (i.e., 1.1 M NaCl). Water flux of both FO and MF membranes as well as TDS  
260 concentration in terms of mg/L NaCl (equation S10 (SI)) as a function of time over the  
261 course of the OMBR-MF experiments are shown in Fig. 1. From the beginning of baffled  
262 OMBR-MF operation, MF was operated to mitigate the salinity build up in the reactor  
263 (equation S11 (SI)). Initial FO flux was 11.9 LMH and this flux varied in the range of  
264 11.54-6.98 LMH during the 38 days of continuous operation. During the first five days of  
265 operation, more than 9.5 LMH FO flux was observed and then decreased by 1 LMH during  
266 the first three weeks of operation. In between 18 to 30 days, the FO flux fluctuated around

267 8 LMH. In the final phase of the OMBR-MF operation, FO flux gradually decreased to  
268 around 7 LMH. Overall, an average of 8.56 LMH FO flux was achieved during the 38 days  
269 of continuous operation. The decrease in the FO water flux could be related to the internal  
270 concentration polarization (ICP) effect, salt accumulation and biofoulants accumulation on  
271 the membrane surface due to MLSS in the reactor. The FO flux decline during OMBR  
272 operation has also been reported by other researchers (Wang et al., 2014a).

273 Recently, Wang et al. (2016b) observed that the water flux in the OMBR with TFC FO  
274 membrane quickly reduced from about 15.3 LMH to approximately 8.0 LMH during the  
275 first 8 days of operation. The FO flux slightly decreased to the final value of about 3.0  
276 LMH while 22 mS/cm mixed liquor conductivity was reached at the end of the operation.  
277 In another continuous OMBR study an average 9 LMH flux was achieved over 30-day  
278 examination. However, in order to maintain 9 LMH flux, backwashing was performed at  
279 14, 21 and 28 days respectively (Achilli et al., 2009). In the present study, no membrane  
280 cleaning was performed during the 38 days of continuous operation and almost similar  
281 average flux (8.56 LMH) was obtained. Moreover, Holloway et al. (2015b) operated  
282 OMBR with UF membrane and reported that during the first three weeks, when OMBR  
283 was operated without UF membrane, flux declined considerably (down to 4.2 LMH) and  
284 thereafter, by incorporation of UF membrane, FO flux was found to increase and remain  
285 stable (4.8 LMH) for the rest of the study. In another MF-OMBR study by Wang et al.  
286 (2014b), the MF membrane was continuously operated under constant flux at 5 LMH but  
287 significant FO flux decline was reported during the first 30 days of operation after which  
288 almost stable FO flux (5.5 LMH) was obtained. In the present study, even though MF

289 membrane was operated in parallel to FO, the FO flux decline was gradual and relatively  
290 slow. Nevertheless, MF was operated at very low flux (1.1-2.4 LMH) while high FO flux  
291 was continuously achieved (average of 8.56 LMH). This also could be attributed to the high  
292 permeability of TFC PA membrane in comparison to the above cited studies that employed  
293 CTA membrane.

294 As reported by Luo et al. (2016a), salinity build-up in the bioreactor is an intrinsic  
295 phenomenon associated with OMBR operation and the rate of solute diffusion through the  
296 membrane depends on membrane selectivity, diffusion coefficient of the solute and on the  
297 concentration difference across the membrane (Holloway et al., 2015a; Nguyen et al.,  
298 2016). In practice, to prevent the inhibition of the microbial community activities due to  
299 reverse salt flux, the maximum bioreactor tank salinity must not exceed 2 g/L (Holloway et  
300 al., 2014; Nguyen et al., 2015). In this study, during the course of operation, the  
301 conductivity, TDS and pH (data not shown for pH) in the reactor, FO and MF permeates  
302 were measured regularly. Results showed that, due to simultaneous operation of MF  
303 membrane with FO, the TDS value did not increase significantly and remained almost  
304 stable in the mixed liquor (612-1434 mg/L). The salt permeating through the MF  
305 membrane was also helpful to maintain a high driving force between concentrated DS and  
306 mixed liquor. It could be seen that the trend of salinity (in terms of EC) variation of the  
307 reactor mixed liquor and MF permeate was similar (Fig. S5, SI) indicating that the  
308 incorporation of MF membrane to discharge the soluble salt could effectively decrease the  
309 salinity and further alleviate the salt accumulation in the OMBR.

310 The measured TDS concentration in the bioreactor changed from 612-1434 mg/L  
311 corresponding to 1.24-2.92 mS/cm during the first week of study and then after, an average  
312 TDS of less than 1200 mg/L was obtained during the rest of investigation time. This  
313 salinity range is well below the value reported in previous studies where a stable mixed  
314 liquor conductivity of approximately 5 mS/cm was observed during OMBR operation with  
315 continuous MF extraction (Luo et al., 2016a; Qiu et al., 2015; Wang et al., 2014b). Further,  
316 the result obtained here compares favourably with Luo et al. (2015) work as they reported  
317 that after a small increase in the first week, the mixed liquor conductivity stabilized at  
318 approximately 400 mg/L after incorporation of MF membrane. Similarly, (Holloway et al.,  
319 2015b) observed that TDS reached a peak concentration (approximately 8000 mg/L) during  
320 OMBR testing without UF membrane. This could be attributed to the small hydrated radius  
321 of monovalent ions (Na with a hydrated radius of 0.18 nm and Cl with a hydrated radius of  
322 0.19 nm) which could easily pass through the FO membrane (membrane pore size: 0.37  
323 nm) (Nguyen et al., 2016). In the previous study, as soon as the UF subsystem was operated  
324 in parallel to OMBR, the TDS concentration rapidly declined and remained constant at  
325 approximately 1000 mg/L until the end of the UFO-MBR investigation. After incorporation  
326 of UF membrane, a stable FO flux of 4.8 LMH was achieved over the duration of the  
327 investigation without a single membrane cleaning (Holloway et al., 2015b). Finally, in their  
328 work on OMBR, Alturki et al. (2012) observed a rapid increase in the mixed liquor  
329 conductivity from 0.27 to 8.27 mS/cm within seven days without housing the submerged  
330 MF/UF membrane in the bioreactor (Alturki et al., 2012). It should be emphasized that  
331 during the 38 days of baffled OMBR-MF test runs, both the FO and MF membranes were  
332 not offered any physical cleaning nor backwashing. The authors believe that during long-



333 term operation, FO biofouling could significantly affect the OMBR-MF hybrid system  
334 performance. In order to mitigate biofouling of FO membrane different cleaning techniques  
335 can be adopted. The first one could be the physical cleaning of the FO membrane. For this  
336 cleaning strategy, OMBR operation should be stopped and FO membrane module should be  
337 taken out from the system followed by cleaning with deionised water (DI) and then gentle  
338 cleaning with sponge ball. Since fouling layer in FO process is not compact (no applied  
339 pressure) (She et al., 2016), one step cleaning can recover the desired initial flux. When the  
340 flux recovery is not satisfactory then chemical cleaning can be performed. As reported by  
341 Holloway et al. (2015a), chemically enhanced osmotic backwashing is conducted by  
342 replacing the draw solution with a very low salinity base (NaOH) or acid (HCl) cleaning  
343 solutions, which are continuously recirculated on the draw solution side of the membrane.

#### 344 TOC and PO<sub>4</sub>-P removal

345 Biological process performance of the baffled OMBR-MF hybrid system was assessed with  
346 regards to the removal of basic contaminants (i.e. TOC, NH<sub>4</sub>-N, TN, and PO<sub>4</sub>-P), sludge  
347 production, and biological activity. TOC removal in reactor mixed liquor, MF permeate and  
348 diluted draw solution (FO) was 91.31 %, 93.51 % and 97.84 % respectively. The average  
349 TOC concentration in the feed, reactor, MF permeate and FO was 96.55 mg/L, 8.55 mg/L,  
350 6.25 mg/L and 2 mg/L respectively (Fig. 2 (a)). The removal of TOC from the OMBR FO  
351 channel was over 97% during the entire experimental period. As in the previous study Qiu  
352 and Ting (2013) noted that the overall removal rate of organic matter constantly reached up  
353 to 98% and the TOC in the DS was less than 5.0 mg/L during OMBR operation. Recently,

354 Wang et al. (2016a) also achieved 96% TOC removal with TFC membrane when treating  
355 140 mg/L fed TOC, with an average of 5.2 mg/L TOC reached in FO permeate.

356 In the current study, higher TOC removal has been achieved in both reactor and FO  
357 permeate as compared to (Wang et al., 2016b). Thus, biological degradation contributed  
358 greatly to reduce the concentration of TOC, overcoming the concentration process caused  
359 the FO membrane rejection. In the diluted DS and in the MF permeate, TOC concentration  
360 further decreased due to the FO and MF membrane rejection, respectively. Nevertheless,  
361 the reverse draw solute flux undesirably impacted the biological treatment of OMBR. In  
362 fact, TOC concentration in the bioreactor increased slightly at the beginning of OMBR  
363 operation (day 7 and 9 respectively). This observation is consistent with that reported by  
364 (Luo et al., 2016a; Luo et al., 2016b) and could be attributed to the high rejection of almost  
365 all the organic matter by the FO membrane, which caused a significant accumulation of  
366 non-degradable or/and refractory dissolved organic matter (DOM) within the bioreactor  
367 (Qiu & Ting, 2013).

368 It has been reported that, during OMBR operation, increase salinity in the bioreactor could  
369 inhibit the metabolic activity of biomass and plasmolysis causing the release of intracellular  
370 constituents and soluble microbial products (Wang et al., 2014a). This study also showed  
371 that the presence of high TDS can interfere with the oxygen transfer and affect the  
372 biological metabolism thereby reducing the capacity of the reactor to sustain shock loads.  
373 The incorporation of MF membrane in the present study successfully kept a salinity level  
374 well within the control (Fig. 1(a)) leading to high TOC removal efficiency (Fig. 2(a)).

375 Phosphate can be eliminated by two different mechanisms: assimilation and luxury uptake  
376 (Rosenberger et al., 2002). The baffled OMBR-MF system showed very stable and  
377 effective performance in achieving high removal of PO<sub>4</sub>-P with 81.22 %, 87.36 % and  
378 93.46 % removal efficiency achieved in reactor, MF and FO processes (Fig. 2 (b)). A  
379 relatively high and stable phosphorous removal was observed corresponding to the average  
380 0.23 mg/L obtained in FO permeate. In the baffled OMBR, mixed liquor suspension is  
381 cyclically exposed to aerobic and anoxic conditions, and enhanced biological phosphorus  
382 removal might have occurred to some extent. In the present study, the observed low DO  
383 concentration in the anoxic zone can have created a pseudo-anaerobic condition which  
384 would have favoured phosphorous release. During aerobic condition phosphorous uptake  
385 by bacteria could have happened. Thus, phosphorus removal can be achieved similarly to  
386 what was observed by (Kimura & Watanabe, 2005) Kimura and Watanabe (2005). Another  
387 possible explanation for the good removal of phosphorus is precipitation with inorganic  
388 substances. Aggregates of phosphorus and inorganic substances would settle in dead zones  
389 of the bottom of OMBRs (Rosenberger et al., 2002). In present study, salting out might  
390 have occurred and some potassium phosphate may have precipitated out with the salt  
391 transported in the reactor from draw solution. Thus, regular sludge withdrawal might have  
392 achieved phosphorus removal through biomass and settled phosphorus discharge from dead  
393 zones. Guo et al. (2008) achieved more than 98% of PO<sub>4</sub>-P removal in sponge-submerged  
394 membrane bioreactor. The explanation given was due to the sponge providing a good  
395 anoxic condition around the surface of the sponge and the anaerobic condition inside the  
396 sponge which makes the aerobic submerged membrane bioreactor able to achieve a higher  
397 removal efficiency of PO<sub>4</sub>-P (Guo et al., 2008). In our study, during the OMBR operation it

398 was observed that biomass clung to the outer baffle wall as well to the inside wall of the  
399 reactor in anoxic zone. Specifically, when switching over to anoxic cycle, the low DO  
400 concentration in the anoxic zone might have created anaerobic condition inside of the  
401 biomass that attached to the wall and phosphorous release would possibly have occurred.  
402 Qiu et al. (2015) reported 97.9% of phosphate phosphorus ( $\text{PO}_4\text{-P}$ ) rejection by the FO  
403 membrane in a hybrid microfiltration-forward osmosis membrane bioreactor (MF-FOMBR).  
404 In two studies by Nguyen et al. (2015 and 2016), their OMBR systems achieved more than  
405 99% and more than 98% phosphate ( $\text{PO}_4\text{-P}$ ) removal respectively. Moreover, Holloway et  
406 al. (2007)(Holloway et al., 2007) also showed that very high rejection of phosphate (99.6-  
407 99.9 %) by the FO membrane could be attained during concentration of anaerobically  
408 digested sludge centrate. Indeed, the FO membrane can almost completely reject  $\text{PO}_4\text{-P}$   
409 due to its negative charge and the relatively large radius diameter (0.49 nm) of the  
410 orthophosphate ion, which is the dominant phosphate species under the conditions tested  
411 (Aftab et al., 2015; Praveen and Loh, 2016).

412 The polyphosphate accumulating organisms are susceptible to saline conditions, and the  
413 increased osmotic pressure within their cells due to salt accumulation could diminish their  
414 phosphate accumulating capacity (Lay et al., 2010). Kinetics studies have suggested that  
415 nitrogen and phosphorus removal efficiency dropped to 20% and 62%, respectively, when  
416 salt concentration was 5% NaCl in the bioreactor (Nguyen et al., 2016). However, in this  
417 study,  $\text{PO}_4\text{-P}$  build-up in the bioreactor was not observed which can be attributed to  
418 reasonably low salinity as compared to other OMBR reports.

### 419 3.2 Nitrogen removal

420 In the proposed baffled OMBR-MF hybrid system, nitrification is carried out in the whole  
421 chamber when the liquid level was above the top of the inserted baffles (Fig. S2) while  
422 denitrification proceeds in the outer (anoxic) zone when the liquid level was low. Fig. 3 (a)  
423 shows the removal of  $\text{NH}_4\text{-N}$  during baffled OMBR-MF continuous operation. Initially,  
424 97% of  $\text{NH}_4\text{-N}$  was removed by the reactor, which then decreased down to 80% after 3  
425 days of operation as shown by the  $\text{NH}_4\text{-N}$  concentration increase observed in the reactor.  
426 The stability of nitrification was thus affected since the increase in supernatant  $\text{NH}_4\text{-N}$   
427 concentrations on day 7 (12.5 mg/L) and day 9 (8.9 mg/L) significantly affected effluent  
428 concentrations. The drop in the  $\text{NH}_4\text{-N}$  removal could be attributed to the effect of salinity  
429 on biomass. Additionally, the ammonia-oxidizing bacteria (AOB) are generally slow  
430 growing and more sensitive to changes in environmental conditions such as temperature  
431 and salinity (Qiu & Ting, 2013). Therefore, the activity of the AOB and nitrite-oxidizing  
432 bacteria (NOB) could easily be inhibited by elevation of salinity (non-halophilic bacteria)  
433 which hampered the biological conversion of  $\text{NH}_4\text{-N}$ . (Ye et al., 2009)(Ye et al., 2009)(Ye  
434 et al., 2009)(Ye et al., 2009)(Ye et al., 2009)(Ye et al., 2009)(Ye et al., 2009)(Ye et al.,  
435 2009)(Ye et al., 2009)(Ye et al., 2009)(Ye et al., 2009)(Ye et al., 2009)(Ye et al., 2009)(Ye  
436 et al., 2009)(Ye et al., 2009)(Ye et al., 2009)(Ye et al., 2009)Ye et al. (2009) also showed  
437 that a high salinity of 1.02 (W/V%) can be detrimental to the survival of many AOB and  
438 other bacteria. Besides, the survival bacteria were also shown to be strongly inhibited.  
439 Consequently, the  $\text{NH}_4\text{-N}$  removal efficiency decreased greatly. When the salinity level in  
440 the bioreactor was then stabilised after 11 days, the nitrifiers regained their potential to

441 remove  $\text{NH}_4\text{-N}$  and as a result the nitrifying activity was restored. In fact, after 19 days of  
442 operation, the supernatant  $\text{NH}_4\text{-N}$  concentration decreased significantly and the conversion  
443 of  $\text{NH}_4\text{-N}$  finally recovered to more than 97% till the end of the study.

444 Furthermore, although nitrifiers are slow growing bacteria, the proposed baffled OMBR-  
445 MF with a prolonged HRT of 30.25 hours and 115 days SRT was favourable for the  
446 relative long generation time of the nitrifying bacteria allowing better removal efficiencies.  
447 Aftab et al. (2015) also observed similar behaviour in their study when targeting  $\text{NH}_4\text{-N}$   
448 removal. In current study, the average  $\text{NH}_4\text{-N}$  concentration of 0.84 and 0.46 mg/L in the  
449 MF permeate and diluted DS was obtained respectively. Wang et al. (2016b) achieved  
450 about 97%  $\text{NH}_4\text{-N}$  removal in mixed liquor and 99%  $\text{NH}_4\text{-N}$  removal in FO permeate  
451 respectively. Most of the OMBR studies reported almost perfect nitrification. However, in  
452 this study complete nitrification has not been observed. It is worthwhile to note that in  
453 order to achieve simultaneous denitrification, air flow rate was kept quite low during  
454 baffled OMBR-MF operation. Therefore, in addition to salinity effect, low air flow rate  
455 might have created limited oxygen supply and non-homogeneous aeration; hence complete  
456 nitrification might have been compromised. In fact, Kurita et al. (2015) studied a baffled  
457 MBR and observed that, due to reduced aeration rate, the supply of oxygen to the biomass  
458 was apparently not sufficient; resulting in limited nitrification and an increased  
459 concentration of  $\text{NH}_4\text{-N}$  in the treated water. This result can also be correlated to the  
460 aerobic-anoxic cycle time (30 min-90 min, respectively). In fact, due to prolonged anoxic  
461 cycle time, denitrification performance possibly would have been improved though offset  
462 by the increase of  $\text{NH}_4\text{-N}$ . However, during the second and third week of baffled OMBR-

463 MF operation, reasonably high but unstable removal was observed and then from 20 day  
464 onwards, more than 95%  $\text{NH}_4\text{-N}$  removal in mixed liquor was achieved.

465 Total nitrogen concentration in the treated water was apparently lower than that in the feed  
466 water (Fig. 3 (b)). This reduction in TN was accomplished by denitrification due to the  
467 creation of anoxic conditions associated with the insertion of the baffles. Hence, the TN  
468 removal was well achieved under elevated salinity conditions and overall TN removal rate  
469 reached 94.28% in the diluted DS (FO permeate). Considering the all system, 70.38% of  
470 average TN removal could be achieved by the reactor (biological process) without  
471 recirculation of mixed liquor for 38 days continuous operation. Indeed, removal of nitrogen  
472 was significant without addition of external carbon, indicating the effectiveness of the  
473 proposed baffled OMBR-MF hybrid system. Fig. 3 (c) shows the changes in concentration  
474 of  $\text{NH}_4\text{-N}$ ,  $\text{NO}_2\text{-N}$ , and  $\text{NO}_3\text{-N}$  in the mixed liquor during the operation. Denitrification  
475 allows maintaining a relatively low  $\text{NO}_3\text{-N}$  concentration with an average concentration of  
476 5.12 mg/L in mixed liquor supernatant within the 38 days of operation. TN removal  
477 efficiency is also plotted in Fig. 3 (c). In the aerobic bioreactor, TN consumption occurs  
478 mainly through microbial assimilation. At the same time, nitrification converts  $\text{NH}_4\text{-N}$  to  
479 nitrite ( $\text{NO}_2\text{-N}$ ) and then nitrate ( $\text{NO}_3\text{-N}$ ) under aerobic conditions (Luo et al., 2016b).  
480 Incomplete nitrification is usually manifested by the detection of both  $\text{NH}_4\text{-N}$  and  $\text{NO}_x\text{-N}$   
481 in the bioreactor. From Fig. 3 (c), it is clearly seen that the removal of nitrogen in the  
482 baffled OMBR-MF was limited by nitrification especially during the first week of operation.  
483 However, considerably good denitrification has been achieved throughout the operation.  
484 Overall, TN removal was considerably high in this study.

485 Fig 3 (c) shows that, when  $\text{NO}_3\text{-N}$  concentration in the mixed liquor increases, the T-N in  
486 the treated water also increases and TN removal efficiency decreases. Also, in the baffled  
487 OMBR-MF system, the high rejection of the FO membrane prolonged the retention time of  
488  $\text{NO}_2\text{-N}$  and  $\text{NO}_3\text{-N}$  within the bioreactor, which also facilitated the removal of  $\text{NO}_2\text{-N}$  and  
489  $\text{NO}_3\text{-N}$  to nitrogen during the anoxic cycle. One explanation for the better TN removal  
490 performance was the improvement in the creation of an anoxic environment. This was  
491 confirmed by dissolved oxygen (DO) measurements in the anoxic zone. Fig. 4 shows the  
492 DO profile in outer zone of the baffles at two different depths in the aerobic and anoxic  
493 zone, respectively. The measured values shown in Fig. 4 were obtained on day 31, when  
494 good removal of nitrogen was observed. As can be seen in Fig. 4, at time ( $t= 0$  minute) a  
495 wastewater pump has started and thus wastewater was fed to the reactor. When water level  
496 in the reactor reached on top of the inserted baffles at desired height, the level controller  
497 sensor activated and then feed pump stopped automatically. After 30 min, the water level  
498 had dropped back to the top of the inserted baffles (end of aerobic cycle time). In order to  
499 achieve efficient denitrification in the system, creation of a good anoxic condition in the  
500 exterior zone of the reactor is essential. Further, the denitrification is the limiting step in the  
501 removal of nitrogen in the operation. In present work, a good anoxic condition was  
502 achieved in the outer zone ( $\text{DO} < 0.5$  mg/L as shown in Fig. (4)). The experimental data  
503 also confirmed that the outer zone worked as an anoxic reactor for a long period in the total  
504 operation time which probably explained the superior denitrification performance of the  
505 proposed baffled OMBR-MF system. Furthermore, the experimental results also proved  
506 that the creation of an anoxic environment could be achieved even in the bottom part of the  
507 reactor at 18 cm depth (Fig. (4)). This was probably due to the high concentration of MLSS



508 (4.7-6.1 g/L) which eventually promoted better denitrification. Several studies have  
509 indicated the negative effect of salinity on MLSS (as low as 1 g/L MLSS) which adversely  
510 led to poor MLVSS/MLSS ratio as low as 0.45 (Wang et al., 2014a). Finally, in the current  
511 study, a reasonably high C/N ratio of about 14 has been maintained in baffled OMBR-MF  
512 system which is favourable enough to prevail better denitrification and reduction of total  
513 nitrogen for baffled OMBR-MF hybrid system.

### 514 **3.3 Biomass activity**

515 Water extraction by the MF membrane from OMBR mixed liquor did not significantly  
516 impact biomass characteristics (Fig. 5). Since very little excess sludge was discharged  
517 everyday (i.e. 115 days SRT) from the bioreactor, the MLSS concentration improved with  
518 time during the operation. In the later stage (21-38 days), the growth rate of MLSS was  
519 steady about 6 g/L due to low sludge organic loading. The MLSS concentration in the  
520 reactor varied from 4.7 to 6.1 g/L over the 38 days of continuous operation. Some studies  
521 (Li et al., 2016) demonstrated that high salinity in the mixed liquor adversely impacts on  
522 the MLSS. For example, (Wang et al., 2016b) operated OMBR, employing both CTA and  
523 TFC membranes, at 0.76 g/L stable MLSS and the MLVSS/MLSS ratio dropped down to  
524 41% within 33 days operation. This finding was correlated with the increase salinity level  
525 in the reactor; (reaching up to 20 mS/cm), clearly showing the adverse effect of salinity on  
526 microbial activity. In another study, (Luo et al., 2016b) noted that a small but noticeable  
527 decrease in MLSS concentration was observed during OMBR operation with 0.5 M NaCl  
528 as a DS and yet at infinite SRT conditions.

529 Alturki et al. (2012) reported that build-up of salinity level up to 4.13 g/L of NaCl led to a  
530 gradual decrease in the ratio of MLVSS over MLSS from 0.87 to 0.66 after seven days of  
531 operation. The decrease in the MLVSS/MLSS ratio indicates that biological activity of the  
532 reactor may have deteriorated over time (Alturki et al., 2012). Further, an increase in the  
533 osmotic stress could result in the dehydration and plasmolysis of bacterial cells and thus  
534 reduce their viability (Luo et al., 2016a). However, in the present work, the relatively low  
535 salt concentration ( $< 1.5$  g/L over the entire operating time) in the bioreactor enabled the  
536 normal growth of the microbial community due to continuous salt bleeding by MF  
537 membrane and daily withdrawn mixed liquor (100 mL) from the bioreactor. Thus, high  
538 concentration of active biomass as well as a stable and high (i.e. above 0.84)  
539 MLVSS/MLSS ratio was maintained during the operation (Fig. 5).

540 The specific oxygen uptake rate (SOUR) has been widely used to understand the effect of a  
541 higher concentration of materials including salt on the activity of biomass in various  
542 aerobic processes (Choi et al., 2007). In this study, respiration test of the activated sludge  
543 showed a significant decrease in the SOUR from an initial 4.51 to 3.03 mg O<sub>2</sub>/g MLVSS/h  
544 during the first week of baffled OMBR-MF hybrid system, suggesting a deterioration of  
545 biological activity (Fig. 5). This decrease in SOUR could be well correlated to the increase  
546 in salinity during the first week of operation. These observations are consistent with  
547 previous studies especially within the first two weeks of operation and could be attributed  
548 to the inhibition of elevated bioreactor salinity on biomass growth and activity (Luo et al.,  
549 2016a; Reid et al., 2006). Indeed, as can be seen in Fig. 5, in the following weeks,

550 measured SOUR was well within the optimum range prescribed for MBR operation (i.e. 3-  
551 5 mg O<sub>2</sub>/g MLVSS/h) while the salinity level in the bioreactor was stabilised (Fig. 1(a)).

### 552 **3.4 Fouling behaviour**

553 Fig. S6 (a) (SI) shows the image of FO membrane before and after (fouled) 38 days of  
554 continuous operation. No significant foulant deposition has been observed on the  
555 membrane surface. Since the baffled OMBR-MF system was operated with the membrane  
556 active layer facing the feed side (AL-FS mode), foulant build up occurs on the active layer,  
557 where it could easily be removed by hydraulic shear force due to continuous aeration (Mi  
558 & Elimelech, 2008). This is different from what occurs with microporous membranes in  
559 traditional MBR, where the initial foulant deposition takes place within the porous structure  
560 of the membranes, and hydraulic shear forces cannot effectively remove the foulant (Qiu &  
561 Ting, 2013). Fig. S5 (b) (SI) shows SEM images of both pristine and fouled TFC  
562 membranes, respectively. It could be observed from SEM images that the surface of  
563 pristine TFC FO membrane is rougher and more rugged (valley like structure-  
564 Magnification x 5000). As shown in SEM observation for fouled membrane, compared  
565 with the original membrane, the active layer of fouled membrane surface was almost fully  
566 covered with a rather thin and compact gel-like fouling layer. The foulant layer was 2.38  
567 µm thick as deduced from the SEM image. This is smaller than the typical thickness of 20–  
568 50 µm fouling layer found in MBRs (Lay et al., 2011). It is worthwhile to note that the  
569 fouling layer on the FO membrane surface was very thin, and it had only a small effect on  
570 the water flux during the entire operation time. The EDX analysis for both pristine and  
571 fouled membrane (Fig. S6) was also performed. From EDX analysis of pristine membrane,

572 it can be seen that the TFC FO membrane surface mainly includes C, O and S. On the other  
573 hand, many inorganic elements such as Na, Ca, Fe, Al, Cl, P and Si were observed on the  
574 fouled membrane surface. The source for many of those inorganic elements was most  
575 probably the synthetic wastewater (e.g. Fe, P). The presence of Na and Cl could be  
576 correlated to the reverse salt diffusion from the DS.

#### 577 **4 Conclusions**

578 Primary findings drawn from this study can be summarized as follow:

- 579 • The performance of a novel baffled OMBR-MF hybrid system was examined for  
580 water flux, salinity build up and nutrient removal specifically SND was successfully  
581 achieved in a single reactor.
- 582 • An average 8.56 LMH FO flux was achieved during 38 days of continuous  
583 operation.
- 584 • MF membrane incorporation alleviated salinity build up and hence better  
585 performance was achieved.
- 586 • The dissolved oxygen profile during the aerobic-anoxic cycle confirmed < 0.5 mg/L  
587 oxygen favourable for denitrification.
- 588 • More than 97 % TOC, 87 % PO<sub>4</sub>-P and 94% TN removal was achieved from  
589 OMBR-FO channel.

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749 **Figure Captions**

750 Fig. 1. Variation of water flux and reactor salinity

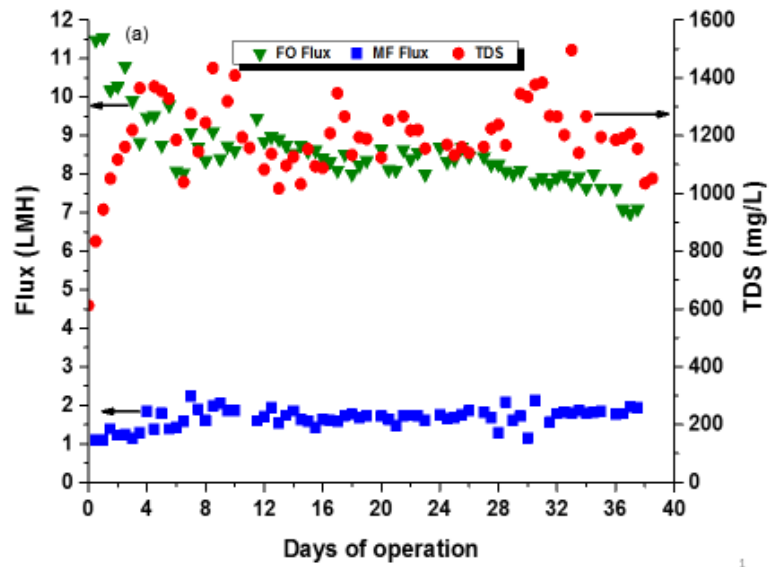
751 Fig. 2. Performance of baffled OMBR-MF for (a) TOC removal, (b) phosphate ( $\text{PO}_4\text{-P}$ )  
752 removal

753 Fig. 3. Performance of baffled OMBR-MF system for (a)  $\text{NH}_4\text{-N}$  removal, (b) total  
754 nitrogen (Devia et al.) removal, (c) variation of  $\text{NH}_4\text{-N}$ ,  $\text{NO}_2\text{-N}$ ,  $\text{NO}_3\text{-N}$  and TN  
755 removal in the bioreactor

756 Fig. 4 Dissolved oxygen (DO) profile with aerobic-anoxic cycle time in baffled OMBR-MF  
757 hybrid system

758 Fig. 5 Variation of MLSS, MLVSS and MLVSS/MLSS ratio and SOUR in baffled  
759 OMBR-MF hybrid system with time

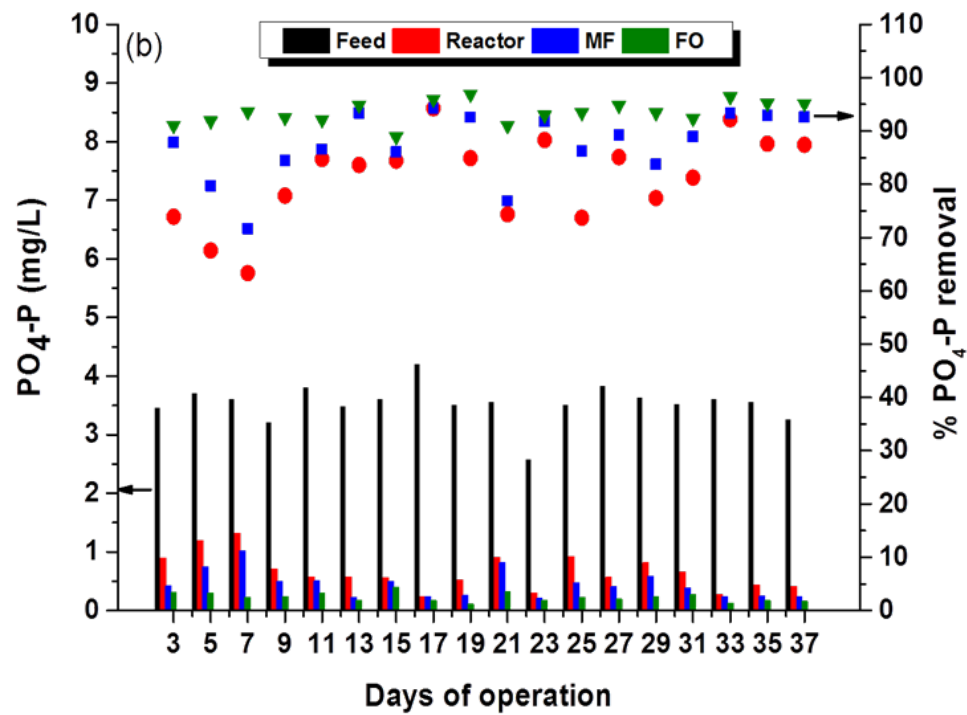
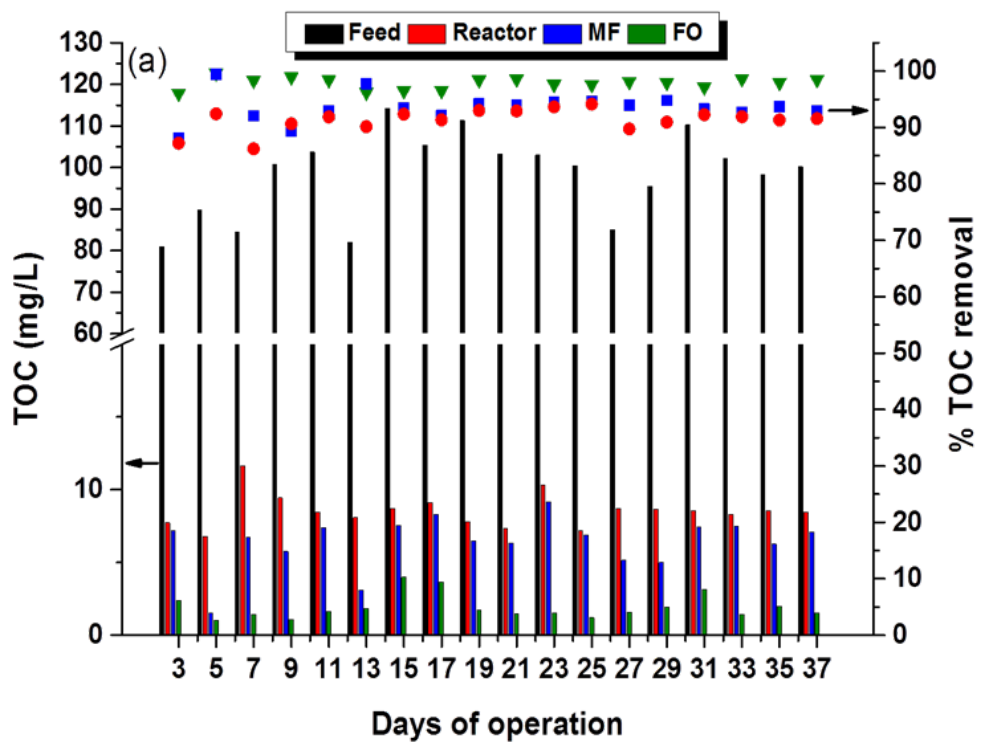
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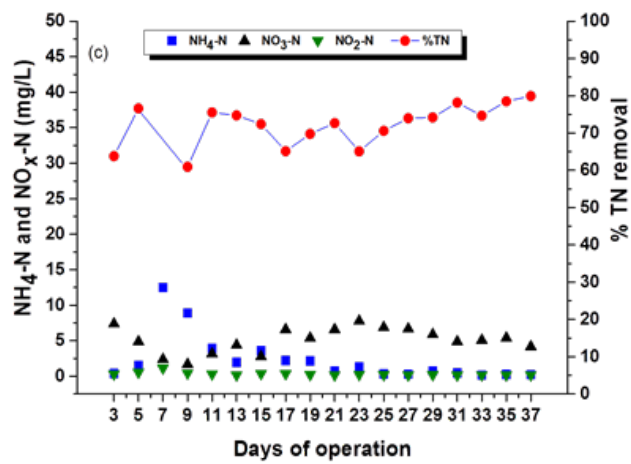
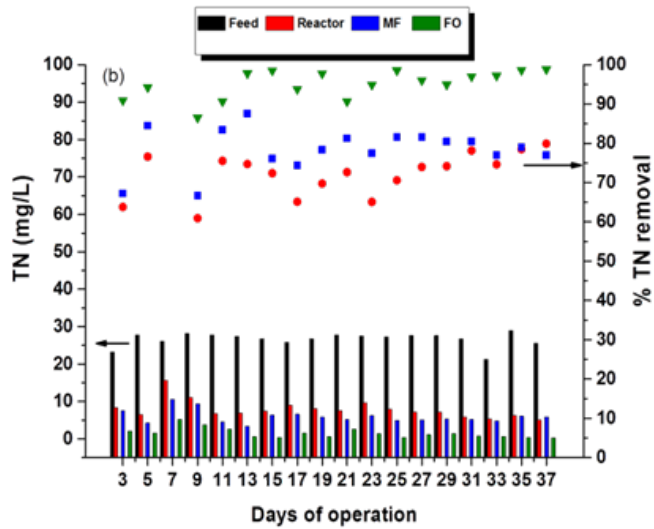
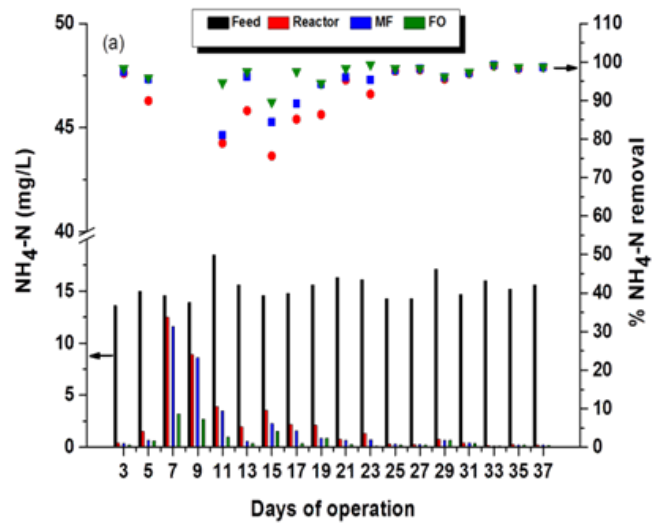
762 Fig. 1

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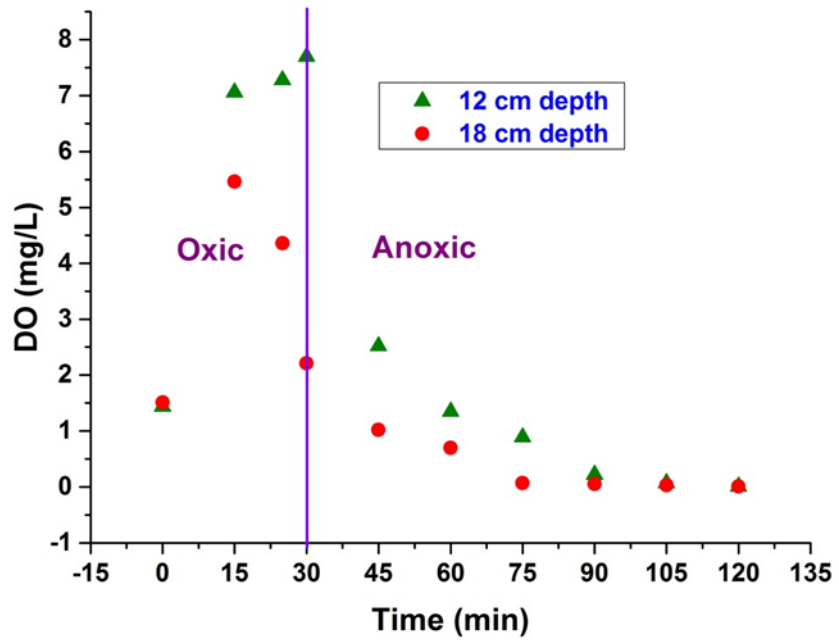
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765 Fig. 2 (a) and (b)



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767 Fig. 3 (a), (b) and (c)



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769 Fig. 4

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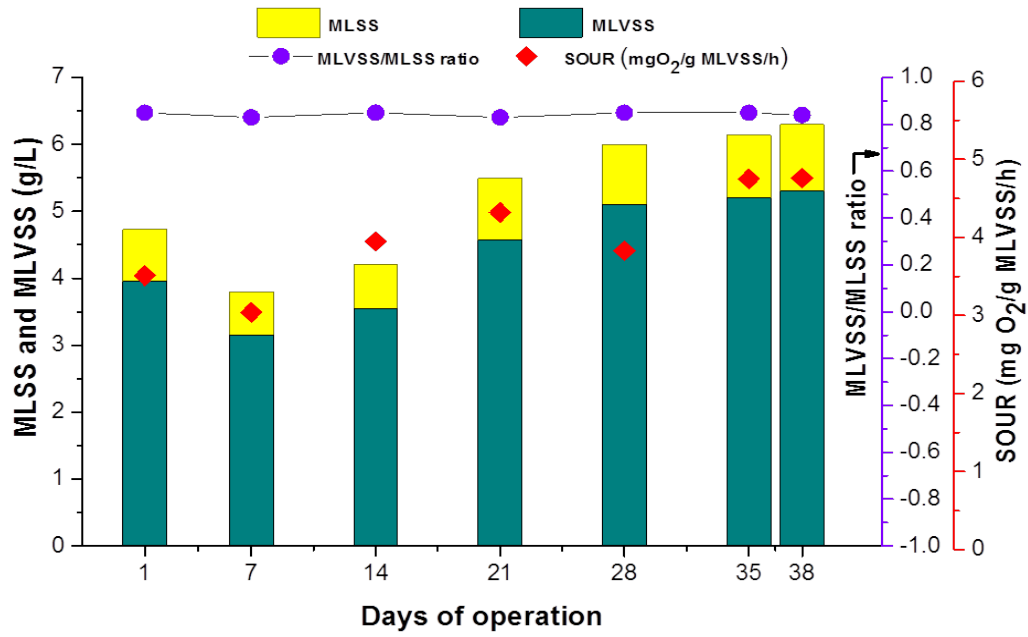
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779 Fig. 5