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# Resource recovery from wastewater by anaerobic membrane bioreactors: Opportunities and challenges

Xiaoye Song University of Wollongong, xs245@uowmail.edu.au

Wenhai Luo University of Wollongong, China Agricultural University, wl344@uowmail.edu.au

Faisal I. Hai University of Wollongong, faisal@uow.edu.au

William E. Price University of Wollongong, wprice@uow.edu.au

Wenshan Guo University of Technology Sydney, wguo@uts.edu.au

See next page for additional authors

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### Resource recovery from wastewater by anaerobic membrane bioreactors: Opportunities and challenges

#### Abstract

This review examines the potential of anaerobic membrane bioreactor (AnMBR) to serve as the core technology for simultaneous recovery of clean water, energy, and nutrient from wastewater. The potential is significant as AnMBR treatment can remove a board range of trace organic contaminants relevant to water reuse, convert organics in wastewater to biogas for subsequent energy production, and liberate nutrients to soluble forms (e.g. ammonia and phosphorus) for subsequent recovery for fertilizer production. Yet, there remain several significant challenges to the further development of AnMBR. These challenges evolve around the dilute nature of municipal wastewater, which entails the need for preconcentrating wastewater prior to AnMBR, and hence, issues related to salinity build-up, accumulation of substances, membrane fouling, and membrane stability. Strategies to address these challenges are proposed and discussed. A road map for further research is also provided to guide future AnMBR development toward resource recovery.

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#### Authors

Xiaoye Song, Wenhai Luo, Faisal I. Hai, William E. Price, Wenshan Guo, Hao H. Ngo, and Long D. Nghiem

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7	Xiaoye Song <sup>a</sup> , Wenhai Luo <sup>b</sup> , Faisal I. Hai <sup>a</sup> , William E. Price <sup>c</sup> , Wenshan Guo <sup>d</sup> , Hao
8	H. Ngo <sup>d</sup> , Long D. Nghiem <sup>d*</sup>
9	<sup>a</sup> Strategic Water Infrastructure Laboratory, School of Civil Mining and
10	Environmental Engineering, University of Wollongong, Wollongong, NSW 2522,
11	Australia
12	<sup>b</sup> Beijing Key Laboratory of Farmland Soil Pollution Prevention and Remediation,
13	College of Resources and Environmental Sciences, China Agricultural University,
14	Beijing, 100193, China
15	<sup>c</sup> Strategic Water Infrastructure Laboratory, School of Chemistry, University of
16	Wollongong, Wollongong, NSW 2522, Australia
17	<sup>d</sup> Centre for Technology in Water and Wastewater, University of Technology Sydney,
18	Ultimo, NSW 2007, Australia

<sup>\*</sup> Corresponding author: <u>duclong.nghiem@uts.edu.au</u>; Ph: +61 2 9514 2625.

#### 19 Abstract

This review critically discusses the potential of anaerobic membrane bioreactor 20 (AnMBR) to serve as the core technology for simultaneous recovery of clean water, 21 energy, and nutrient from wastewater. The potential is significant as AnMBR 22 treatment can remove a board range of trace organic contaminants relevant to water 23 24 reuse, convert organics in wastewater to biogas for subsequent energy production, and liberate nutrients to soluble forms (e.g. ammonia and phosphorus) for subsequent 25 recovery for fertilizer production. Yet, there remain several significant challenges to 26 27 the further development of AnMBR. These challenges evolve around the dilute nature 28 of municipal wastewater, which entails the need for pre-concentrating wastewater prior to AnMBR, and hence, issues related to salinity build-up, accumulation of 29 substances, membrane fouling, and membrane stability. Strategies to address these 30 challenges are proposed and discussed. A road map for further research is also 31 32 provided to guide future AnMBR development toward resource recovery. Keyword: Anaerobic membrane bioreactor (AnMBR); Wastewater treatment; 33 34 Resource recovery; Biogas; Water reuse.

#### 35 **1. Introduction**

36 In a paradigm shift towards the circular economy, wastewater can no longer be viewed as the culprit of environmental pollution but rather a source of valuable 37 38 resources, including clean water, renewable energy and nutrients. The economic value 39 of key resources in wastewater can help to offset the cost of wastewater treatment 40 (Burn et al., 2014). Indeed, reclaimed water has been considered as an alternative 41 source to augment clean water supply and address issues caused by water shortage (Shannon et al., 2008). Energy can be extracted from the organic content in 42 43 wastewater by anaerobic treatment to produce biogas, which is a renewable fuel. 44 Nutrients in wastewater can also be recovered to produce fertilizers for sustainable agriculture production, particularly given the finite availability of phosphorus from 45 46 mining (Koppelaar & Weikard, 2013). Recent interest in these resources has spurred 47 new research aiming to convert wastewater treatment plants into resource recovery facilities. 48

Nutrient recovery from wastewater can also reduce the maintenance cost of wastewater treatment facilities and avoid environmental impacts. During wastewater treatment, phosphate and ammonium (which are abundant in wastewater) can react with magnesium to form crystalline precipitate, known as struvite (MgNH<sub>4</sub>PO<sub>4</sub>·6H<sub>2</sub>O), causing blockage and scaling of plant equipment (Doyle et al., 2002). Moreover, both nitrogen and phosphorus are important contaminants that can result in eutrophication of natural waterways if they are discharged to the environment.

56 Membrane bioreactor (MBR) has been deployed at an increasing speed to advance

57 wastewater treatment and reuse on a global scale (Hai et al., 2014). MBR is an

58 integration of membrane filtration with conventional activated sludge (CAS)

59 treatment. Compared to CAS treatment, MBR exhibits several advantages, including

60 higher effluent quality, smaller footprint, as well as easier operation and management

61 (Judd, 2016). Indeed, MBR is more effective for the removal of trace organic

62 contaminants (TrOCs) than CAS treatment for advanced water reuse (Luo et al.,

63 2014). TrOCs occur ubiquitously in municipal wastewater and are of particular

concern to water reuse. It is noteworthy MBR is energy-intensive since aeration is
necessary for the growth and activity of activated sludge. Furthermore, energy and
nutrients in wastewater are dissipated as released gases (e.g. carbon dioxide and
nitrogen gas) in MBR treatment.

An alternative MBR configuration, namely anaerobic MBR or AnMBR, has also been 68 69 explored for energy neutral wastewater treatment (Gao et al., 2008; Verstraete et al., 70 2009). AnMBR integrates anaerobic digestion treatment with membrane filtration. During AnMBR treatment, organic substances in wastewater are biologically 71 72 converted to methane-rich biogas. The produced biogas can offset the energy demand 73 for wastewater treatment (McCarty et al., 2011). Since anaerobic treatment converts nutrients to chemically available forms (e.g. ammonia and phosphate), AnMBR can 74 also facilitate nutrient recovery via subsequent precipitation. Nevertheless, there 75 76 remain several significant challenges in the development of AnMBR for resource 77 recovery from wastewater, particularly municipal wastewater. These include low organic and nutrient contents in municipal wastewater as well as issues associated 78 79 with salinity build-up, membrane stability, membrane fouling, and the occurrence of 80 inhibitory substances.

In this paper, the performance of AnMBR for wastewater treatment and resource recovery is critically reviewed. Several key challenges to the further development of AnMBR are delineated. Potential strategies to address these challenges are proposed. This review paper provides important insight to the development of AnMBR for the management of water, energy, and nutrients.

#### 86 2. Fundamentals and configurations of anaerobic membrane bioreactor

#### 87 **2.1 Fundamentals of anaerobic membrane bioreactor**

AnMBR differs intrinsically from aerobic MBR in terms of the biological component.
The anaerobic biological process involves four integrated stages, namely hydrolysis,
acidogenesis, acetogenesis, and methanogenesis. Degradation of organic matter and
their conversion to biogas depend on the symbiotic relationship among the different

92 groups of microorganisms (e.g. fermentative bacteria, syntrophic acetogens, 93 homoacetogens, hydrogenetrophic methanogens and aceticlastic methanogens) (Chen et al., 2016). Of these microorganism groups, methanogens play arguably the most 94 important role for biogas production by converting intermediate products from 95 previous stages to methane gas. However, methanogens are slow-growing 96 97 microorganisms and can be easily washed out from conventional anaerobic bioreactors. By integrating membrane separation processes, commonly including 98 99 microfiltration (MF) and ultrafiltration (UF), the hydraulic retention time (HRT) can 100 be decoupled from sludge retention time (SRT). Thus, AnMBR can produce more 101 biogas than conventional anaerobic treatment (Liao et al., 2006). In many aspects (e.g. energy consumption, contaminant removal efficiency, and 102 103 volume throughput), AnMBR differs considerably from aerobic MBR (Table 1). Since 104 aeration is not required, AnMBR has a significantly lower energy input to the 105 bioreactor compared to aerobic MBR. In addition, the energy footprint of AnMBR can be offset by produced biogas (Smith et al., 2012). Nevertheless, Martin et al. (2011) 106 reported that the energy demand in submerged AnMBR varies considerably from 0.03 107 to 5.7 kWh/m<sup>3</sup> due to different energy requirements for gas sparging to control 108 109 membrane fouling. Indeed, AnMBR is usually operated at high biomass concentration 110 as well as long SRT and HRT to treat complex wastewater (Skouteris et al., 2012), 111 resulting in more severe membrane fouling in comparison with aerobic MBR. As such, the reported flux of AnMBR is commonly in the range between 5 and 12  $L/m^2h$ , 112 which is considerably lower than the flux of 20 - 30 L/m<sup>2</sup>h typically for full-scale 113 aerobic MBR (Wang et al., 2018). Without oxygen as an electron acceptor, anaerobic 114 115 digesters release electrons onto methane (CH<sub>4</sub>) rather than using them for microbial 116 growth. Thus, AnMBR produces less sludge than aerobic MBR (Liao et al., 2006). 117 Since anaerobic degradation is a slow process, AnMBR has a lower contaminant 118 removal efficiency and volume throughput (i.e. treatment capability) than aerobic 119 MBR.

Feature	AnMBR	MBR	Reference
Energy consumption (kWh/m <sup>3</sup> )	$0.03 - 5.7^{a}$	~ 2 <sup>b</sup>	Martin et al. (2011)
Biomass concentration (g/L) <sup>c</sup>	10 - 40	5-20	Liao et al. (2006); Shin and Bae (2018)
Organic loading rate (kg COD/L)	0.17 - 35.5	0.25 - 0.8	Hai et al. (2014); Shin and Bae (2018); Maleki et al. (2018)
Organic removal efficiency (%)	> 90	> 95	Lin et al. (2013); Judd (2016); Svojitka et al. (2017)
Hydraulic retention time (hours)	> 8	4 - 8	Stuckey (2012); Berkessa et al. (2018)
Water flux (L/m <sup>2</sup> h)	5 – 12	20 - 30	Wang et al. (2018)
Sludge retention time (day)	> 100	5 - 20	Liao et al. (2006); Skouteris et al. (2012)
Operational temperature (°C)	20 - 50	20 - 30	Martinez-Sosa et al. (2011); Hai et al. (2014)

120 <b>Table 1:</b> Con	nparison between	AnMBR and MBR	for wastewater treatment.
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<sup>a</sup> Energy consumption was calculated for submerged AnMBR treating wastewater with strength between 0.27 and 10 g COD/L.

<sup>b</sup> Energy consumption was calculated for submerged MBR treating wastewater with strength between 0.3 and 1.0 g COD/L.

<sup>c</sup> Biomass concentration was on the basis of mixed liquor suspended solids content.

#### 124 **2.2** Configurations of anaerobic membrane bioreactor

There are several AnMBR configurations depending on the anaerobic treatment 125 process (Figure 1). Excellent reviews of anaerobic bioreactors for AnMBR are 126 127 available in the literature (Skouteris et al., 2012; Ozgun et al., 2013; Chen et al., 2016). 128 Common anaerobic bioreactors for AnMBR include up-flow anaerobic sludge blanket 129 (UASB), completely stirred tank reactor (CSTR), and anaerobic fluidized bed 130 bioreactor (AFBR) (Figure 1A - C). Of these reactors, CSTR is the most frequently used configuration for AnMBR due to its ease of construction and operation. UASB 131 132 can retain biomass mostly in the bottom zone of the bioreactor, thus, the effluent 133 passed through the membrane unit has low suspended solids concentration, which may help alleviating membrane fouling. In UASB, the produced biogas can be 134 captured through a gas/liquid/solid separator. AFBR contains granular media (e.g. 135 activated carbon or sponge) suspended in the reactor by the upward velocity of the 136 treated fluid (Kim et al., 2011). 137

AnMBR can be operated in either side-stream or submerged mode (Figure 1 D – F). In the side-stream AnMBR, membrane module is integrated outside of the bioreactor. Mixed liquor in the bioreactor is transferred to the membrane unit for clean water extraction. In the submerged AnMBR, membrane unit can be directly immersed into the bioreactor (Figure 1 E) to extract treated water through the membrane.

The submerged AnMBR can be deployed as a two-stage system by submersing the 143 144 membrane module in a chamber separated from the working bioreactor (Figure 1F). 145 The two-stage AnMBR configuration facilitates membrane maintenance and cleaning 146 by intensive shear force and chemicals. Retentate from the membrane tank can also be 147 recirculated to the anaerobic reactor for further contaminant biodegradation. As such, 148 the two-stage configuration can be potentially used for full-scale AnMBR applications. 149 Indeed, Shin and Bae (2018) reported that ten out of eleven recent pilot-scale AnMBR 150 studies have adopted the two-stage configuration. As a notable exception, Gouveia et 151 al. (2015b) developed a single-stage AnMBR system, in which a submerged 152 membraned housed at the supper part of the USAB reactor. In their study, two baffles

153 were placed between the three-phase (i.e. gas/liquid/solid) separator and the UF





Figure 1: Typical anaerobic bioreactors (A: up-flow anaerobic sludge reactor; B:
continuous stirred-tank reactor; C: anaerobic fluidized bed reactor) and their
integration with membrane separation process in the (D) side-stream, (E) submerged
and (F) external chamber modes.

160 Recent progress to advance wastewater treatment and reuse has resulted in the

161 emergence of high retention AnMBR systems. These mainly include anaerobic

162 membrane distillation bioreactors (AnMDBR) and anaerobic osmotic membrane

163 bioreactor (AnOMBR). By integrating with the MD or FO process, both AnMDBR

and AnOMBR can enhance the removal of contaminants for water reuse applications.

165 AnMDBR is an integration of membrane distillation (MD) and anaerobic treatment.

166 MD is a thermally driven separation processes, in which the thermal gradient between

167 a feed solution and distillate drives the transportation of water vapour through a

168 hydrophobic, microporous membrane. The competitive advantages of anaerobic

169 processes can be readily utilized when they are combined with the MD process,

170 because the thermophilic operation for anaerobic treatment can reduce extra heat

171 requirement for MD operation (Kim et al., 2015).

172 AnOMBR, which combines forward osmosis (FO) with anaerobic treatment, is also attractive for advanced wastewater treatment and reuse. In FO, water transports from 173 174 a feed solution, across the semi-permeable membrane, to a draw solution with the 175 osmotic pressure difference between these two solutions as the driving force. During AnOMBR operation, a desalination process, such as nanofiltration (NF) and reverse 176 177 osmosis (RO), can be used to regenerate the draw solution and produce clean water. 178 Compared to conventional MF and UF membranes, FO has higher selectivity, lower membrane fouling propensity and better membrane fouling reversibility (Xie et al., 179 2015). 180

#### 181 **3.** Anaerobic membrane bioreactors for water reuse and resource recovery

#### 182 **3.1 Organic removal**

The performance of AnMBR for water reuse has been extensively studied in recent
years. AnMBR is best suited for the treatment of wastewater with a high organic
content. Indeed, there have been a number of pilot demonstration and full-scale

186 AnMBR systems for treating effluents from field crop processing (e.g. sauerkraut,

187 wheat, maize, soybean, and palm oil), dairy processing, and the beverage industry (e.g.

188 winery, brewery, and distillery) (Table 2).

189 Amongst complex contaminants in wastewater, TrOCs present arguably the most

190 vexing challenge to water reuse (Schwarzenbach et al., 2006). Recent studies have

also demonstrated that the removal of TrOCs by AnMBR varied significantly from

- negligible to more than 90% (Figure 2). TrOC removal by AnMBR is governed
- 193 mostly by intrinsic physiochemical properties of the compound. Monsalvo et al. (2014)

194 investigated the removal of 38 TrOCs by AnMBR and reported over 90% removal of

- nine compounds; while the others were removed by less than 50%. Wijekoon et al.
- 196 (2015) have successfully developed a predictive framework to assess the removal of

197 TrOCs by AnMBR, which relates the removal of TrOCs to their hydrophobicity and 198 molecular structures. Specifically, hydrophobic TrOCs were effectively removed by 199 more than 70% as they are prone to adsorb onto sludge for subsequent biodegradation 200 (Figure 2). High removal was also observed for hydrophilic compounds with electron 201 donating groups (e.g. hydroxyl and amine) and nitrogen in the molecular structure. By 202 contrast, hydrophilic compounds with electron withdrawing groups (e.g. chloro and 203 amide) were resistant to AnMBR treatment (Figure 2).



Figure 2: Removal of trace organic contaminants (TrOCs) by AnMBR. Results were extracted from previous studies (Monsalvo et al., 2014; Wijekoon et al., 2015). TrOCs were ordered based on their hydrophobicity, which could be determined by their effective octanol – water partition coefficient (i.e. Log D) at the mixed liquor pH of 7.

Wastewater	Bioreactor configuration	Membrane integration	Organic removal (COD, %)	Methane yield (L/kg COD)	Reference
Food wastewater	N.A	Side-stream MF	81 - 94	136	He et al. (2005)
Kraft evaporator condensate	UASB	Submerged MF	97 – 99	290 - 310	Lin et al. (2009)
Landfill leachate	N.A	Submerged UF	90	460	Zayen et al. (2010)
Real municipal	CSTR	Side-stream MF	86 - 88	300	Yue et al. (2015)
Pre-concentrated synthetic wastewater	N.A	Submerged MF	96	223	Dai et al. (2015)
Meat packing wastewater	N.A	Submerged MF	88 - 98	130 – 180	Galib et al. (2016)
Raw tannery	N.A	Submerged MF	90	160	Umaiyakunjaram and Shanmugam (2016)
Domestic wastewater +					
food waste-recycling wastewater	N.A	Submerged MF	97.9 – 99.3	200 – 220	Jeong et al. (2017)
Synthetic wastewater	UASB	Submerged MF	> 98	290	Berkessa et al. (2018)
Malting wastewater	CSTR	Submerged MF	90.2 - 94.1	308 - 345	Maleki et al. (2018)

**Table 2:** Examples of AnMBR performance regarding organic removal and methane production

<sup>\*</sup>UASB: up-flow anaerobic sludge reactor; CSTR: continuous stirred-tank reactor; N.A: information is not available.

#### 211 **3.2 Biogas production**

212 Chemical energy in wastewater in the form of organic carbon can be recovered by

AnMBR to produce biogas (Table 2). It has been well established that biogas

214 produced by AnMBR consists of more than 80% of CH<sub>4</sub> (Skouteris et al., 2012).

215 During AnMBR treatment, the CH<sub>4</sub> yield increases linearly with the organic loading

rate (Yeo et al., 2015). Under an optimized condition, AnMBR can convert up to 98%

217 of the influent COD into biogas, which is equivalent to seven times of the energy

required for system operation (Van Zyl et al., 2008). In practice, actual biogas yield is

219 considerably lower than the theoretical value, due to the high solubility of CH<sub>4</sub> in the

220 effluent and process inhibition caused by inhibitory substances.

221 CH<sub>4</sub> loss due to its solubility (22.7 mg/L) in the effluent is significant during AnMBR

treatment, particularly for low strength municipal wastewater (Smith et al., 2012). Liu

et al. (2014) reported that dissolved  $CH_4$  in permeate was approximately 45% of total

224 produced CH<sub>4</sub> at 30 °C when AnMBR was used for treating municipal wastewater

with COD of 200 mg/L. Similar results were also reported by Yeo et al. (2015) who

 $226 \qquad observed that \ 24-58\% \ of \ total \ produced \ CH_4 \ was \ dissolved \ in \ the \ permeate \ during$ 

227 AnMBR treatment and Yue et al. (2015) who demonstrated that AnMBR could

remove 86 – 88% COD from municipal wastewater (influent COD of approximately

330 mg/L), but 67% of the produced CH<sub>4</sub> was dissolved in the mixed liquor and then

released via permeate. Galib et al. (2016) reported that the dissolved CH<sub>4</sub>

231 concentrations decreased from 54 to 25 mg/L when the organic loading rate of

wastewater increased from 0.4 to  $3.2 \text{ kg COD/m}^3$ d, due to the enhanced biogas yield

at the high organic loading rate.

234 Dissolved CH<sub>4</sub> in the permeate does not only reduce the energy efficiency of AnMBR

treatment, but also contribute to global warming as the greenhouse potency of CH<sub>4</sub> is

236 25 times higher than carbon dioxide. Vacuum packed towers, bubble columns and

237 forced drafted aerators can be used to remove CH<sub>4</sub> from anaerobically treated effluent

238 (Crone et al., 2016). These processes require a large physical footprint to ensure

239 sufficient contact time for gas stripping and prevent operational problems, such as 240 flooding and channelling (Sethunga et al., 2018). Membrane separation process has also been proposed recently to advance dissolved CH<sub>4</sub> recovery from anaerobic 241 effluents. Cookney et al. (2016) demonstrated a hollow fibre membrane contactor that 242 243 could recover more than 98.9% dissolved CH<sub>4</sub> from AnMBR effluent. However, membrane separation process for the recovery of dissolved CH<sub>4</sub> from anaerobic 244 effluents is still in the early stage and its economic viability and process safety have 245 246 not been fully evaluated. Overall, the dissolution of CH4 in effluent is still a major limiting factor to the deployment of AnMBR for low strength wastewater (Liu et al., 247 2014). 248

#### 249 **3.3 Nutrient removal and recovery**

During AnMBR treatment, nutrient removal depends largely on microbial assimilation and is limited due to low biomass yields of anaerobic microbes. Dai et al. (2015) reported that AnMBR could only remove 10% of the total nitrogen. On the other hand, anaerobic treatment liberates nitrogen and phosphorus in the form of ammonium ( $NH_4^+$ ) and phosphate ( $PO_4^{3-}$ ), respectively, thus facilitating their recovery through subsequent precipitation.

Integrating complementary processes with AnMBR may be necessary to enhance 256 nutrient recovery from AnMBR effluent. These processes include membrane 257 processes (Jacob et al., 2015), ion exchange (Liu et al., 2016), electrodialysis (Xie et 258 259 al., 2016), and photosynthetic bioreactor (Gonzalez et al., 2017). Deng et al. (2014) demonstrate that natural zeolite as an absorbent can be used to economically remove 260 NH4<sup>+</sup> from AnMBR effluent. Jacob et al. (2015) reported 90% removal of COD and 261 ammonium nitrogen from AnMBR effluent by a direct contact MD process. Similar 262 263 results were reported by Song et al. (2018b) who demonstrated the complementarity between AnMBR and MD for TrOC removal. It is noteworthy that a reduction of 264 NH<sub>4</sub><sup>+</sup> removal was observed in their study due to its transportation through the MD 265 membrane via ammonia evaporation. This issue can be potentially addressed using a 266 267 FO and MD hybrid system, where the FO membrane can effectively reject NH<sub>4</sub><sup>+</sup>

while MD can be used to regenerate the draw solution and produce clean water. Xie et al. (2014) has successfully demonstrated the feasibility of the FO and MD hybrid

270 system for nutrient recovery (as struvite) and clean water production from digested

sludge centrate.

272 Effective nutrient removal can be achieved by high retention AnMBR systems. Chen

et al. (2014b) demonstrated that AnOMBR could remove total phosphorus (TP) and

 $NH_4^+$  by 100% and 62%, respectively. The observed complete TP removal was

attributed to the high rejection of  $PO_4^{3-}$  ions by the FO membrane given their negative

charge and large hydrated radius (Holloway et al., 2007).

# 4. Factors underlying key challenges to further develop anaerobic membrane bioreactors

Despite the high potential of AnMBR for resource recovery from wastewater, there remain some challenges, particularly for treating municipal sewage. They include the dilute nature and temperature difference of municipal wastewater, salinity build-up when diluted wastewater is preconcentrated, membrane fouling and stability, and inhibitory substances (e.g. free ammonia and sulphide) (Figure 3). Thus, future studies are required for the development of effective strategies to address these challenges for further development of AnMBR.



Figure 3: Key challenges and their potential strategies to the development of AnMBR
for wastewater treatment and resource recovery.

289 4.1 Dilute nature of wastewater

290 Municipal wastewater has low concentrations of organic substances (for energy recovery) and even lower concentration of nitrogen and phosphorus (for nutrient 291 recovery). A moderate wastewater strength (> 1000 mg COD/L) is necessary to 292 293 maintain effective activity of anaerobic digester for adequate biogas yield and 294 removal of organic pollutants from wastewater (Verstraete et al., 2009). Similarly, ammonium and phosphate concentrations should be higher than 5 g NH<sub>4</sub>-N/L and 50 295 296 mg/L, respectively, for economically efficient recovery by conventional processes, 297 such as ion exchange and chemical precipitation. However, municipal wastewater 298 typically contains ammonium and phosphate less than 0.1 g NH<sub>4</sub>-N/L (Mulder et al., 299 2013) and 10 mg/L (Yuan et al., 2012). Thus, the pre-concentration of municipal wastewater is required prior to AnMBR treatment for the waste-to-resource strategy. 300 Membrane separation can be used to pre-concentrate wastewater to produce high 301 quality water and simultaneously enrich non-water components for subsequent 302

303 recovery. Currently used membrane processes include MF, UF and RO. As an

304 example, Dai et al. (2015) have successfully used an UF – RO hybrid system to

305 pre-concentrate municipal wastewater for elevating COD and nitrogen concentrations

to the levels suitable for AnMBR treatment.

307 FO is a promising membrane process for wastewater pre-concentration due to its high

selectivity, low fouling propensity, and high fouling reversibility (Xie et al., 2013;

Ansari et al., 2016). Ansari et al. (2016) demonstrated that FO could concentrate

municipal wastewater by more than eight times to a COD range (> 1000 mg/L)

311 suitable for biogas production in anaerobic treatment. Higher concentration factors

312 could be achieved when municipal wastewater was further diluted during rainy

seasons (Ansari et al., 2016). FO can be integrated with a desalination process (e.g.

RO and MD) for draw solution regeneration and clean water production (Xie et al.,

315 2013). When the recovery of the draw solution, such as seawater, is not needed, FO

316 can also be operated in the energy efficient osmotic dilution mode (Ansari et al.,

317 2016).

Pre-concentration of wastewater may entail several issues to AnMBR. In addition to 318 319 organic matter, pre-concentrating wastewater can enrich inhibitory substances, such as inorganic salts, ammonia, and sulphate. Salt accumulation in wastewater is significant 320 321 when using FO as the pre-concentration process due to its reverse salt flux. Inhibitions 322 of these substances to AnMBR are discussed in the following sections. Moreover, phosphorus may precipitate in the anaerobic reactor due to the enriched content of 323 phosphorus, calcium, and magnesium in pre-concentrated wastewater (Chen et al., 324 325 2014a), thereby resulting in significant membrane scaling in AnMBR and complications for subsequent phosphorus recovery as the availability of phosphorus in 326 liquid phase is reduced. 327

328 **4.2 Temperature** 

AnMBR can be operated under either thermophilic  $(50 - 60 \degree C)$  or mesophilic  $(30 - 40 \degree C)$  conditions (Martinez-Sosa et al., 2011; Gouveia et al., 2015a). Psychrophilic

331 condition (< 20 °C) is generally not suitable for municipal wastewater treatment. Thus, anaerobic treatment of municipal wastewater is still a challenge for cold regions, 332 where significant energy is required to heat wastewater to a mesophilic condition. 333 334 AnMBR operation at low temperature can result in several negative issues, including aggravated membrane fouling, slow contaminant biodegradation, and high CH<sub>4</sub> 335 solubility in the effluent. Hydrolysis of particulate matter into dissolved molecules is 336 337 limited at low temperature, leading to the accumulation of suspended solids in the reactor and a decrease in methanogenic activity. Martinez-Sosa et al. (2011) observed 338 339 an increase in the total suspended solids content and soluble COD in the bioreactor 340 when the temperature of AnMBR was reduced from 35 to 20 °C, resulting in severe membrane fouling and decreased CH<sub>4</sub> production. The decreased CH<sub>4</sub> production 341 could also be attributed to its increased solubility in the effluent when the temperature 342 decreased to 20 °C. In addition, the mixed liquor viscosity also increased as the 343 temperature decreased, thus requiring more energy for mixing and pumping. 344

#### 345 **4.3 Salinity build-up**

346 Saline wastewater is a challenge to biological treatment. Indeed, AnMBR 347 performance in terms of biogas production and organic removal decreases when treating highly saline feed, such as wastewater from seafood processing and cheese 348 production (Dereli et al., 2012). High salinity could result in enzyme inhibition, cell 349 activity decline, and plasmolysis to anaerobic microbes, thereby negatively affecting 350 351 the anaerobic digestion process (Chen et al., 2008). For instance, Ng et al. (2014) reported that the CH<sub>4</sub> yield of AnMBR was reduced to less than 160 L/kg COD<sub>removed</sub> 352 353 when treating pharmaceutical wastewater due to the disrupted ordinary metabolic 354 functions and degradation kinetics under saline concentrations. Song et al. (2016) also 355 reported the adverse effects of increase salinity (up to 15 g/L NaCl) on COD removal 356 and biogas production of AnMBR.

- 357 Microbial acclimatization could lead to the succession of halotolerant and even
- halophilic bacteria to recover AnMBR performance (Dereli et al., 2012). Jeison et al.

(2008) revealed that long-term adaption resulted in better salt tolerance, with the
observed 50% activity inhibitory concentration (IC50) value for acetotrophic
methanogenesis at approximately 25 g/L NaCl. Munoz Sierra et al. (2018) also
reported the robustness of AnMBR to short-term, step-wise increase of salinity up to
20 g/L NaCl with significant variation in the microbial community. It is noteworthy
that salinity increase exacerbated membrane fouling by reducing sludge particle size
in their study.

#### 366 4.4 Inhibitory substances

AnMBR is susceptible to the accumulation of inhibitory substances, such free 367 368 ammonia and sulphate, in wastewater. Ammonia is generated by the biodegradation of 369 nitrogenous compounds, mostly in the form of protein in wastewater, during anaerobic digestion (Chen et al., 2008). Ammonia toxicity (> 3500 mg/L) to anaerobic 370 371 digester can be attributed to direct inhibition to the activity of cytosolic enzymes as well as an increase in the intracellular pH and/or the concentration of other cations, 372 such as potassium (Kanai et al., 2010). The observed inhibition was due to free 373 ammonia in solution rather than the ammonium ions, whose equilibrium 374 concentrations are dependent on pH and temperature (Chen et al., 2008). Indeed, free 375 ammonia is more toxic than ionised ammonia, because it can penetrate through the 376 377 cell membrane and thus result in the disruption of cellular homeostasis, potassium 378 deficiency and/or proton imbalance. A higher temperature and pH value can exacerbate the inhibition by releasing more free ammonia (Meabe et al., 2013). 379 380 High sulphate concentration can also inhibit AnMBR performance. Such inhibition 381 can be attributed to the competition between sulphate reducing bacteria 382 (approximately 2 g COD/g SO<sub>4</sub>-S<sub>removed</sub>) and methanogenic microbes for available carbon (Chen et al., 2016). Moreover, sulphate can induce the precipitation of 383 non-alkaline metals in anaerobic reactors, reducing their availability as 384 micro-nutrients for methane producing microbes (Stefanie et al., 1994; Siles et al., 385 386 2010). In addition, sulphate reduction produces hydrogen sulphate (H<sub>2</sub>S), which is a 387 corrosive, malodourous, and toxic gas (Muyzer & Stams, 2008; Sarti & Zaiat, 2011;

Park et al., 2014).  $H_2S$  can readily penetrate through microbial cell membrane and denature native proteins inside the cytoplasm producing sulphide and disulphide cross-links between polypeptide chains (Siles et al., 2010).

Sufficient organic supply can mitigate the inhibition of free ammonia and sulphate to 391 AnMBR. Meabe et al. (2013) reported that longer SRT in AnMBR could allow for 392 393 sufficient acclimatization of biomass to resist ammonia inhibition. Thus, no critical 394 ammonia inhibition was observed for both mesophilic and thermophilic AnMBR in their study. Tian et al. (2018) recently developed a stepwise acclimation strategy to 395 allow anaerobic communities to adapt to 10 g  $NH_4^+$ -N/L in mesophilic CSTR. The 396 397 negative impact of sulphate is also insignificant provided that the ratio of COD and  $SO_4^{2-}$  is above 10 (Rinzema & Lettinga, 1988). In some cases, sulphate addition is 398 beneficial to methane production by boosting the degradation of propionic acid (Li et 399 al., 2015). Song et al. (2018a) investigated the effect of sulphate increase on the 400 401 performance of AnMBR and reported that basic biological performance of AnMBR was not affected by the increased sulphate concentration when the influent 402  $COD/SO_4^{2-}$  ratio was maintained higher than 10. Nevertheless, H<sub>2</sub>S content in the 403 404 produced biogas increased significantly and membrane fouling was exacerbated with 405 sulphate addition (Song et al., 2018a). Thus, some physicochemical techniques (e.g. striping, pH adjustment, coagulation, and precipitation) should be applied to reduce 406 sulphate load to AnMBR to secure biogas quality and sustain membrane performance 407 (Yuan & Zhu, 2016). 408

#### 409 **4.5 Membrane fouling**

Membrane fouling is a persistent challenge to advance AnMBR given membrane
material costs and energy demands for fouling control and cleaning. Fouling results
from the accumulation of inorganic and organic foulants internally in membrane pores
and externally on the membrane surface. Membrane fouling can reduce flux, increase
transmembrane pressure, and consequently necessitate chemical cleaning or
membrane replacement. The primary foulants of interest in AnMBR include
suspended biomass, colloidal solids, SMP, EPS, attached cells, and inorganic

precipitates, such as struvite (Smith et al., 2012). Jun et al. (2017) reported that
long-term operation (around 700 days) of AnMBR encountered frequent, sudden
irreversible fouling due to biologically induced mineral scaling, thus, intense chemical
cleaning was required to recover membrane permeability.

Membrane fouling during AnMBR treatment is governed mainly by membrane 421 422 properties and operational conditions (e.g. water flux, temperature, HRT, and SRT), 423 hydrodynamics, and sludge characteristics. For instances, Lin et al. (2009) shown that the filtration resistance in thermophilic AnMBR was about 5 - 10 times higher than 424 425 that of the mesophilic system when operated under similar hydrodynamic conditions. 426 This observation was due to more SMP, biopolymer clusters, and fine flocs (< 15 mm) under the thermophilic condition. Huang et al. (2011) reported that a decrease in HRT 427 enhanced biomass growth and SMP accumulation, while longer SRT reduced the 428 429 flocculation of particulates and particle size, thereby aggravating membrane fouling. 430 Thus, membrane fouling in AnMBR can be potentially mitigated to some extent by optimising the operational conditions. 431

Several techniques have been developed to control and clean membrane fouling 432 during AnMBR operation. In the side-stream AnMBR, high cross-flow velocity can 433 reduce foulant build-up on the membrane surface; while fouling control is typically 434 435 accomplished through biogas sparging for the submerged configuration. Stuckey 436 (2012) reported that the addition of powdered or granular activated carbon could effectively reduce membrane fouling in AnMBR, however, their long-term effects 437 membrane integrity have yet been investigated. In addition, wastewater pre-treatment, 438 439 membrane relaxation, and sub-critical flux operation can also control membrane fouling for AnMBR. 440

441 Despite effective strategies to control fouling, membrane cleaning is still necessary.

442 Membrane cleaning includes physical, chemical, and biological schemes. Physical

443 membrane cleaning can be achieved by backwashing, surface flushing, and

444 ultrasonication (Lin et al., 2013). Chemical cleaning is necessary to further remove

445 fouling layers using suitable agents, such as sodium hypochlorite, hydrochloric acid,

446 nitric acid, citric acid, sodium hydroxide, and EDTA for target foulants.

447 Chemically-assisted backwashing has also been developed to enhance membrane

448 cleaning for AnMBR. Nevertheless, chemicals that can diffuse back to the bioreactor

449 may inhibit the microbial activity and then biological performance of AnMBR. Mei et

450 al. (2017) reported that utilising 12 mmol/L NaOH to assist in-situ membrane

451 backflush did not adversely affect AnMBR treatment performance given the alkali

452 consumption by anaerobic biomass and buffering capacity of the mixed liquid.

#### 453 **4.6 Membrane stability**

454 Chemically and biologically stable polymeric materials are commonly used to

455 fabricate robust membranes for MBR applications. These polymeric materials mainly

456 include polytetrafluoroethylene, polyvinylidenefluoride, and polypropylene

457 (Alkhudhiri et al., 2012). Thus, membrane degradation is not a concern for

458 conventional MBR using the existing low retention UF or MF membranes. By

459 contrast, membrane integrity is a major issue to FO when integrating with biological460 processes.

461 Currently commercial FO membranes are made of either cellulose or polyamide.

462 Chen et al. (2014b) observed a sudden increase in the electrical conductivity of the

463 mixed liquor (over 20 times) after an AnOMBR using a CTA FO membrane was

464 operated for 76 days. They also attributed this observation to membrane

465 biodegradation or hydrolysis in the bioreactor.

466 Both cellulose and polyamide membranes are susceptible to biological and chemical

467 degradation. Cellulose membrane itself can become a substrate for microbial growth.

468 Polyamide TFC membranes appears to be more persistent to biodegradation and

469 hydrolysis than cellulose based membranes (Choi et al., 2005). Nevertheless, some

470 microbial species, such as strains of *Pseudomonas* sp., in activated sludge may

471 biodegrade polyamides by producing extracellular enzymes to hydrolyse amide bonds

472 (Yamano et al., 2008). On the other hand, polyamide membrane is more susceptible to

473 chemical attack by oxidising agents such as chlorine (Simon et al., 2009).

474 Membrane stability determines the product water quality and the sustainability of

475 AnMBR. Thus, it is essential to develop techniques to prevent biological and

476 chemical degradation of membranes in AnOMBR operation. New and robust

477 membrane materials are required to facilitate the integration of FO with AnMBR for

478 resource recovery. Module modification to allow for in-situ membrane cleaning can

also potentially control membrane biodegradation (Choi et al., 2002).

480 **5. Future perspectives** 

AnMBR has a proven capability and can offer a unique opportunity to achieve
simultaneous wastewater treatment and resource recovery. However, the adoption and
commercialisation of AnMBR at industrial scale is still pending due to the challenges
discussed above. Thus, future research should be dedicated to address these issues for
the further development of AnMBR (Figure 3).

486 FO is a promising approach to produce clean water and pre-concentrate wastewater to the level suitable for AnMBR treatment (Ansari et al., 2017). Yet, FO technology is 487 488 still in the early stage of development and requires research efforts for the realisation 489 of full-scale implementation. Moreover, wastewater pre-concentration results in the 490 enrichment of some inhibitory substances (salts, free ammonia, and sulphate) to 491 AnMBR. Thus, techniques for the removal of these inhibitory substances should be 492 developed to secure the performance of AnMBR for treating concentrated wastewater. 493 Membrane fouling in AnMBR is often more severe than aerobic MBR due to the 494 absence of aeration and lower sludge filterability (Skouteris et al., 2012). Thus, 495 advanced techniques to control membrane fouling during AnMBR operation should 496 be developed in addition to the optimisation of operational parameters. Using a low 497 fouling alternative, such as FO, is a potential strategy, which can also enhance contaminant removal in comparison to MF and UF membranes that are commonly 498 499 used for AnMBR.

Compared to membrane fouling, little is known about the stability of membranes
during AnMBR operation. In AnMBR, membranes are exposed to the biologically

active conditions with biomass concentration typically higher than 10 g/L. Moreover,
given the severity of membrane fouling in AnMBR operation, frequent membrane
cleaning with harsh chemicals may be necessary to maintain water production. Thus,
it is important to understand membrane degradation in AnMBR operation and develop
mitigation strategies to prolong membrane lifespan.

507 Several techniques have been proposed to further purify AnMBR effluent for clean 508 water production and/or nutrient recovery. They include membrane filtration, ion 509 exchange, electrodialysis, biological processes (e.g. photosynthetic bioreactor), 510 advanced oxidation processes, and electrocoagulation. Nevertheless, further work is 511 needed to evaluate the techno-economic feasibility of these processes in integration 512 with AnMBR to determine an appropriate framework that can facilitate practical 513 application of AnMBR for wastewater treatment and resource recovery. Moreover, the 514 agronomic availability of recovered nutrients should be assessed to emphasize AnMBR potential for resource recovery from wastewater. 515 516 Recovering dissolved CH<sub>4</sub> from effluent is also strategically important to broaden

517 AnMBR applications towards low organic content wastewater. Recent studies have demonstrated that the promise of membrane-based processes for the recovery of 518 519 dissolved CH<sub>4</sub> from AnMBR effluent (Cookney et al., 2016; Crone et al., 2016; 520 Sethunga et al., 2018), while their economic feasibility has not yet been fully 521 evaluated. Moreover, micro-porous membranes used for gas stripping are threatened by membrane fouling and wetting. As such, continued efforts should be devoted to the 522 development of gas-permeable membranes suitable for CH<sub>4</sub> fraction from AnMBR 523 effluent. 524

#### 525 6. Conclusion

AnMBR has the potential to revolutionise current wastewater treatment facilities for simultaneous recovery of clean water, energy, and nutrients. Such revolution can be accelerated by continued efforts to concentrate municipal wastewater to the level suitable for AnMBR treatment and subsequent resource recovery. Issues associated

530	with salinity build-up, membrane stability and fouling, and the occurrence of
531	inhibitory substances (e.g. free ammonia and sulphate) need to be addressed to
532	advance AnMBR for water reuse and resource recovery. Successful recovery of clean
533	water, energy and nutrient also requires the integration between AnMBR and other
534	complementary processes.
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