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# Removals of Organic Micropollutants in Anaerobic Membrane Bioreactor in Wastewater Treatment: Critical Review

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## Abstract

Anaerobic membrane bioreactor (AnMBR) is a promising technology for achieving an energy-saving or even energy-positive wastewater treatment process as it produces high effluent quality and renewable energy in the form of methane. Nevertheless, concerns on inflowing organic micropollutants (OMPs) caused by various human and industrial activities into AnMBR system are proliferating. The extent to which the removal of OMP in the AnMBR is understood should vary greatly depending upon the removal pathways mainly involved such as sorption into biomass, transformation, or membrane filtration. This review paper describes the fate and removal mechanisms of OMPs in AnMBR system. Although the overall performance of AnMBR treating various wastewaters has been observed under such reduced conditions, understanding and modeling the removal mechanisms of OMPs in this type of reactor still requires many works. Elucidating the removal mechanisms of OMPs will lead to the improvements in designs and operations of AnMBR system while optimizing performances and saving energy.

## 27 **Keywords**

28 Organic micropollutants, anaerobic membrane bioreactor, wastewater, biodegradation,  
29 adsorption, absorption, membrane separation

## 30 **Introduction**

31 Organic micropollutants (OMPs) in domestic wastewater are generated mainly by  
32 various human and industrial activities, covering a wide range of contaminants including  
33 pharmaceuticals, pesticides, personal care products (PCP), industrial chemicals, hormones,  
34 and other emerging compounds. While the OMPs are present at trace amounts in the  
35 wastewater ranging from ng/L to  $\mu\text{g/L}$ , tremendous efforts have been made to better  
36 understand the fate and transport of the OMPs in water bodies significantly with considering  
37 their significant risks to human health and eco-environmental security<sup>1</sup>. Those risks include  
38 short-term or long-term toxicity, environmental persistence, antibiotic resistance, endocrine  
39 disruption etc<sup>2</sup>. Nevertheless, the individual or collective behavior of the OMPs in domestic  
40 wastewater treatment plant (WWTP) require much attention since the information on their  
41 removal behaviors through the biological treatment process are still limited particularly under  
42 anaerobic conditions.

43 Interests in anaerobic membrane bioreactor (AnMBR) are growing in domestic  
44 wastewater treatment or high strength wastewater rapidly since it enables synergistic effects  
45 on wastewater managements and resource recovery<sup>3,4</sup>. The AnMBR is to combine anaerobic  
46 bioreactor with membrane filtration. The key advantages offered by the AnMBR process are  
47 to uncouple hydraulic retention time (HRT) and solid retention time (SRT). Compared to  
48 conventional aerobic biological treatment such as activated sludge process, the AnMBR

49 allows (1) low sludge production (up to 90 %)<sup>5</sup>, (2) low energy consumption and (3)  
50 production of bioenergy in the form of methane<sup>6</sup> as well as (4) production of high effluent  
51 quality (in terms of suspended solids, turbidity and microorganisms) thanks to membrane  
52 filtration. Nevertheless, understanding the removal of OMPs in AnMBR system still requires  
53 many works.

54 Both sorption and biotransformation play primary roles in the removals of OMPs  
55 during operation of AnMBR<sup>7</sup>. Sorption of OMPs from the aqueous phase to biomass should  
56 occur due to hydrophobic interactions and electrostatic interactions (cation bridging and  
57 exchange)<sup>1, 8</sup>. Biotransformation is the process whereby microbes decompose organic  
58 pollutants. In AnMBR, porous membrane such as microfiltration (MF) or ultrafiltration (UF)  
59 is often used to retain the biomass from wastewater. However, rejection efficiency of the  
60 OMP through MF or UF may not be very high because the size of OMP molecule is often  
61 smaller than the pore size of membrane which is in the range from 0.1 to 0.01  $\mu\text{m}$ .  
62 Nevertheless, it allows the retention of colloidal organic carbon which are largely bound to  
63 OMP. During membrane filtration, fouling caused by the deposit of organic matter on  
64 membrane and/or within membrane pores is an unavoidable phenomenon. It is also known  
65 that higher concentration of OMPs in wastewater can result in the formation of denser and  
66 more compact structure of fouling layer on membrane<sup>9</sup>. This is because that the fouling layer  
67 formed on membrane surface can often play a role as a secondary membrane to improve  
68 OMPs rejection<sup>10</sup>.

69 Nowadays, there has been an upsurge of interests in AnMBR in which biomass  
70 carriers are suspended for domestic wastewater treatment<sup>11</sup>. The carriers are added into the  
71 AnMBR not only for providing extra surface area for the growth of fixed biomass but also for

72 inducing mechanical membrane cleaning thanks to scouring actions. In anaerobic fluidized  
73 bed membrane bioreactor termed as AFMBR, granular activated carbon (GAC) or powdered  
74 activated carbon (PAC) or polyurethane sponge are often used as fluidized media<sup>4, 11,</sup>  
75 <sup>12</sup>. These media are fluidized by recirculating the bulk wastewater alone through the reactor  
76 without biogas sparging. As a result, operational energy can be reduced significantly<sup>4</sup>. The  
77 AFMBR is reported to have superior performance in the removal of pharmaceuticals  
78 compared to that from the conventional activated sludge process treating a real domestic  
79 sewage<sup>13</sup>. However, understanding the pathways on degrading the OMPs through AFMBR  
80 system needs more experimental works and practical applications with real wastewaters<sup>14</sup>.

81 In this paper, the removal mechanisms of OMPs in the biological wastewater  
82 treatment process are critically reviewed, mainly focusing on anaerobic systems. This review  
83 is expected to provide a vehicle by which an understanding of OMPs fate in biological  
84 wastewater treatment can be used to develop a new design of the AnMBR reactor to improve  
85 the OMPs removal performance.

## 86 **Organic micropollutants in AnMBR**

### 87 *Organic micropollutants (OMPs) issues in wastewater*

88 Many chemicals from human activities are present in trace amounts in natural  
89 environments (e.g., plasticizers, detergents, hydrocarbons, solvents, pesticides, cosmetics,  
90 and drugs). Their concentrations rarely exceed a few hundred nanograms per liter. While  
91 the risks associated with chronic exposure to these substances are still widely discussed by  
92 scientists, there is considerable research showing that at these concentrations, some  
93 substances will affect the behavior of aquatic organisms and human health. As a result, these  
94 substances are called "micropollutants" Wastewater treatment plants (WWTPs) are

95 considered to be one of the primary sources of micropollutants to aquatic environments.  
96 Since the year of 2000, the European regulation, for example, has encouraged the reduction  
97 of emissions in application of the objectives set by the Water Framework Directive (WFD).  
98 At the European level, a list of so-called "priority" substances (whose discharges are to be  
99 reduced) and "dangerous priority" substances (whose discharges are to be deleted) was  
100 published in 2001 and updated in 2008 and 2013. In parallel, many research teams are  
101 studying other micropollutants, so-called "emerging" substances (not yet regulated), due to a  
102 lack of knowledge about exposure levels and/or their toxicity to aquatic environments.

103         Considerable diversity of organic micropollutants is measured in urban wastewaters,  
104 being the mirror of the vital production and consumption of chemicals of human beings.  
105 These OMPs include a large variety of substances families (volatile organic compounds,  
106 detergents, plasticizers, flame retardants, pesticides, hormones, solvents, pharmaceuticals,  
107 PCPs, Polycyclic aromatic hydrocarbon (PAH), surfactants etc) with various physico-  
108 chemical properties (hydrophilic/ hydrophobic, from low to high molecular weight) and their  
109 own removal pathway (Table 1)<sup>1, 4, 11, 15-17</sup>.

110         The WWTPs are designed for the removal of organic matter and nutrients.  
111 Concomitantly they are able to remove some of these organic micropollutants thanks to (bio)  
112 transformation and sorption onto sludge<sup>18-21</sup>. However, some of them are recalcitrant, and  
113 effluent discharges from the WWTP constitute one of the main inputs of OMPs into the  
114 environment<sup>1, 17, 22</sup>. In order to reduce these discharges, tertiary treatments like advanced  
115 oxidation processes (AOPs), the addition of activated carbon, membrane filtration processes  
116 and biological treatments such as wetlands, biofilters, algae reactor, MBR, MBBR, etc., were  
117 designed and operated from lab-scale to full-scale<sup>15, 19, 23-29</sup>. It seems that the main removal

118 mechanisms along WWTP are transformation (biotic or abiotic) and sorption (onto sludge,  
119 carriers, membranes) and that operational parameters such as redox conditions, HRT, SRT,  
120 temperature, pH, type and quantity of (co)substrates play a significant role onto  
121 biotransformation by driving (a) the diversity of the microbial community and the metabolic  
122 pathways (long SRT, low to high redox may increase microbial and metabolic diversity), and  
123 (b) the microbial activity (higher temperature may increase the biological kinetics).  
124 Concentrations of OMPs in the influent and effluent of WWTP vary spatially and temporally  
125 (regional, daily, seasonal) according to industrial and domestic uses<sup>1</sup>. The spectrum of  
126 molecules found in sewage is very wide as exemplified in Table 1<sup>30</sup>. This includes  
127 pharmaceuticals, endocrine-disrupting chemicals (EDC), personal care products, surfactants,  
128 pesticides, etc, with a large spectrum of physico-chemical properties. Some of them can  
129 provide negative impacts on the wild-life in water bodies such as feminization of fish and  
130 mussels<sup>31</sup>. The concentrations found in influents range from the order of ng/L for  
131 pharmaceuticals<sup>17</sup> to mg/L particularly for detergents<sup>32</sup>. In conventional wastewater treatment  
132 plants, the concentrations are generally lower in secondary effluent than those in the primary  
133 effluent<sup>33</sup>. For some pharmaceuticals, however, the concentrations in secondary effluents can  
134 be higher than those detected in the primary effluent due to the deconjugation or the  
135 production of by-products through aerobic biological transformation<sup>34</sup>. which are generally  
136 found in domestic sewage.

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143 **Table 1.** Types of organic micropollutants <sup>1, 35</sup>

<b>OMPs</b>	<b>Classes</b>	<b>Major Sources</b>	<b>Examples</b>
Pharmaceuticals	Antibiotic, antidiabetic, analgesic, anticonvulsant, stimulant, veterinary drug	Excretion, hospital effluents, farmland waste	Acetaminophen, androstenedione, amoxicillin, cbz, dcf, ibp, keto, penicillin, smx, paracetamol, timolol, salicylic acid, etc.
Personal care products (PCPs)	Antiseptic, disinfectant, fragrance and synthetic musk, stimulant, UV filter, insects repellent	Shower, swimming, bathing, shaving, or industrial waste	Benzophenone, caf, diltiazem, chlorophene, triclosan, methylbenzylidene, chlorophene, tonalide, etc.
Steroids and hormones	Endocrine disruptive chemicals (EDCs)	Excretion, hospital effluents, farmlands, aquaculture	Estradiol, estrone, diethylstilbestrol, progesterone, testosterone, etc.
Pesticides	insecticide, herbicide, fungicide	Domestic and agriculture uses	Diuron, mecoprop, MCPA, terbutylazine, etc
Detergent, surfactant	Cationic, anionic, non ionic, perfluorinated compounds (PFCs)	Bathing, laundry, dishwashing, households, dilutants, dispersants	Alkylphenol ethoxylates, alkylphenols (nonylphenol and octylphenol), perfluorooctane sulfonate, perfluorooctanoic acid
Others	Plasticizer, flame retardant, hydrocarbon, solvent	Leaching out of the material, improper cleaning, run-off from the garden, roadway, etc	Phthalates, polybrominated compounds, dioxin and furans, polycyclic hydrocarbons, trichloroethylene, benzene, toluene, etc






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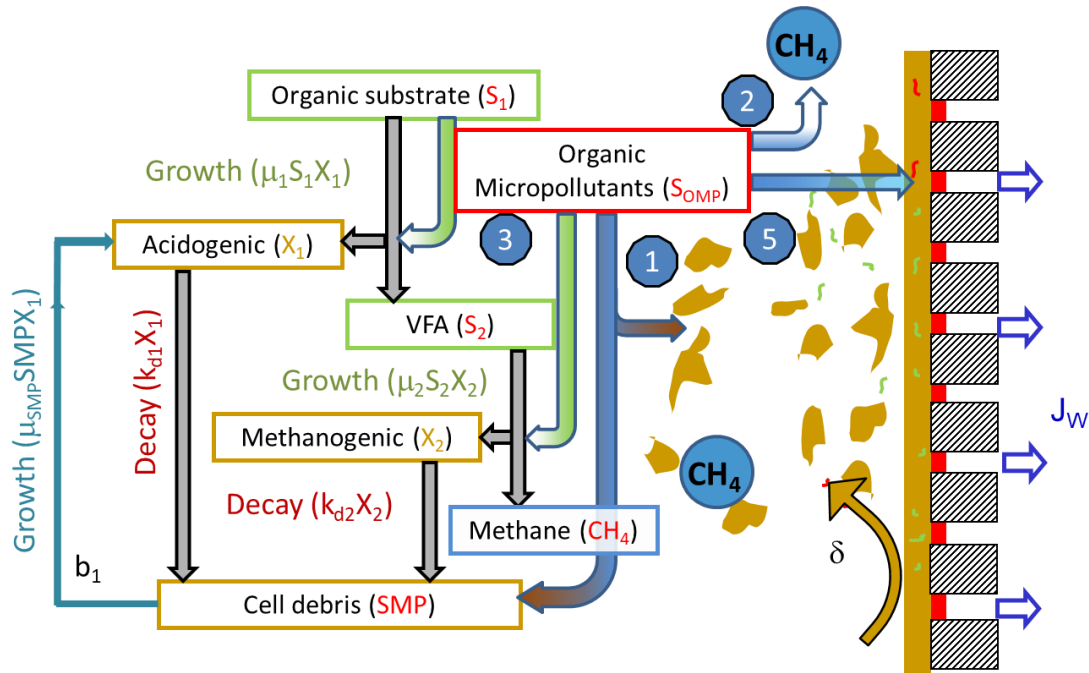
155 **Removal pathways of organic micropollutants**

156 According to micropollutant characteristics such as hydrophobicity, molecular weight,  
 157 volatile properties, biodegradation, and aromaticity, etc., several removal pathways allowing  
 158 their removal from the liquid phase can be considered as shown in Table 2. The figure 1  
 159 addresses the different items listed in Tab.2 in the global removal pathways in anaerobic  
 160 membrane bioreactor, which is discussed in more detail below.

161 **Table 2. Removal pathway of organic micropollutants (OMPs)** <sup>1, 35</sup>

	n°1	n°2	n°3	n°4	n°5
Pathway					
Mechanisms	Coagulation/Flocculation. Sorption Precipitation	Volatilization	Biological oxidation (Biodegradation)	Chemical oxidation	Liquid/Liquid separation (Membrane)
Variables	Partition coefficient Solubility constant : $K_{d,OMP} (L^3.M^{-1}), S$	Henry's law constant : $k_H$ $(ML^{-4}T^{-2}N^{-1})$	Biodegradation Kinetics constant : $k_{bio,OMP} (T^{-1})$	Kinetics constant : $k_{O3,OMP} (M^{-1}.s^{-1})$	Molecular weight Charge : $MW_{OMP}$
Influencing parameters	Particle size and charge, concentration and type of organic matter, hydrophobicity of compounds	Gas sparging	Composition of microbial community, redox conditions SRT, HRT	Oxidant level	Permeate flux, membrane fouling SRT, HRT
Strong influence of the pathway on	PAH, PCB, PBDE, fluoroquinolones	Naphthalene, trichloroethylene	Paracetamol, hormones, ibp	Cbz, dcf	smx, cbz, trim, amitriptyline

162



**Fig 1.** Removal pathway of organic micropollutant in anaerobic membrane bioreactor

*Sorption to biomass or adsorbents*

Sorption to the biomass has been suggested as one of the primary mechanisms affecting the removal of the OMPs in anaerobic treatment<sup>36</sup>. In fact, the sorption of OMP to biomass should be differentiated into two mechanisms : (1) Sorption in which the OMPs move from aqueous phase and (2) penetration into the lipophilic cell membrane, which is driven by their hydrophobicity. The OMPs are also sorbed onto surface of biomass due to electrostatic interactions between the biomass with negative surface charge and OMPs with positive surface charge<sup>37</sup> or negative one thanks to cation bridging. Suarez *et al.* (2008) reported that the sorption of OMPs by the biomass could be determined by the value of the Octanol-Water partitioning coefficient ( $K_{ow}$ ). In case of ionizable compounds, the  $K_{ow}$  can be modified into distribution partitioning coefficient ( $D_{ow}$ ) by taken in account the acid

176 disassociation constant (pKa) as shown in Eq.(1)<sup>38</sup>. On the other hand, field trials make it  
 177 possible to obtain the solid-water partitioning coefficient ( $K_d$ ), also known as the real ratio  
 178 between concentration in solid and liquid phases at equilibrium condition. This coefficient is  
 179 used to predict the fraction of the OMPs sorbed on the biomass<sup>38</sup>, as shown in Eq.(2).

$$180 \quad D_{ow} = \frac{K_{ow}}{1+10^{pH-pKa}} \quad \text{or} \quad D = \frac{K_d}{1+10^{pH-pKa}} \quad (1)$$

$$181$$

$$182$$

$$183 \quad K_d = \frac{C_s}{C_L \cdot X_{TSS}} \quad (2)$$

184  
 185 Where:

186  $K_{ow}$  : octanol-water partitioning coefficient

187  $K_d$  : solid-liquid partition coefficient (L/g)

188  $X_{TSS}$  : Concentration of volatile suspended solids (g/L)

189  $C_s$  : the concentration in the solid phase ( $\mu\text{g/L}$ )

190  $C_L$  : the concentration in the liquid phase ( $\mu\text{g/L}$ )

191

192 In 2010, Barret *et al.* described a three-compartment model for the sorption of OMPs into  
 193 sludge. They distinguished OMPs present in the sludge into; the freely dissolved ( $C_{\text{free}}$ , mg/L),  
 194 the sorbed to Dissolved and Colloidal Matter ( $C_{\text{DCM}}$ , mg/g<sub>DCM</sub>), and the sorbed to particles  
 195 ( $C_{\text{part}}$ , mg/g<sub>PART</sub>)<sup>39</sup>. This three compartment model is more adapted to a membrane separation  
 196 step (AnMBR). Indeed, the retention of DCM depends on molecular cut-off of membrane and  
 197 properties which provides precious information to predict OMP fate in AnMBR more  
 198 accurately. At equilibrium condition, the three-compartment system can be explained by  
 199 equations below:

200  $K_{part} = \frac{C_{part}}{C_{free}} \quad (4)$

201  $K_{DCM} = \frac{C_{DCM}}{C_{free}} \quad (5)$

202 Then, the concentration of apparently dissolved micropollutant ( $C_L$ ), which is the sum of  
 203 freely dissolved and sorbed to DCM ( $C_L = C_{free} + C_{DCM} [DCM]$ ).

204

205 
$$K_d = \frac{C_{part}}{C_L} = \frac{C_{free} \cdot K_{part}}{C_{free} + C_{DCM} [DCM]} = \frac{K_{part}}{1 + K_{DCM} [DCM]}$$

206 Where:

207 [DCM] : Dissolved and Colloidal Matter ( $g_{DCM}/L$ )

208  $K_{DCM}$  : Equilibrium constant of OMPs sorption to DCM ( $L/g_{DCM}$ )

209  $K_{part}$  : Equilibrium constant of OMPs sorption to particles ( $L/g_{Part}$ )

210 Thus, when the separation step is based on an ideal membrane (Total Retention of [DCM]),  
 211 the concentration of OMP in the effluent is equal to  $C_{free}$  ( $C_{free} = C_L - C_{DCM} [DCM]$ ) whereas  
 212 the value of  $C_L$  is reached when a settler is used.

213 It has been found that the OMP removal by sorption not only depends on their  
 214 physicochemical characteristics but also on sludge's characteristics, such as the biomass  
 215 conformation and particle size, as well as operational conditions<sup>39</sup>. Consequently, the  
 216 AnMBR enhanced sorption and biotransformation of some OMPs. In fact, the  
 217 physicochemical characteristics of the membrane will enhance the sorption capacity by  
 218 adding extra solid-liquid interface. The hydrophobic membranes are indeed able to retain  
 219 lipophilic OMPs, whereas charged membranes can retain OMPs due to electrostatic  
 220 interaction<sup>18</sup>.

221 Carrier materials are also often added into an AnMBR to facilitate biofilm growth  
222 and reduce membrane fouling. Both of them can be achieved by providing high specific  
223 surface area and mechanical scouring actions, which is driven by fluidizing the media along  
224 the membrane surface. Recently, activated carbon particles such as PAC or GAC have been  
225 mainly investigated as carrier materials or fluidized media in AnMBR. The addition of PAC  
226 or GAC into the AnMBR thus helps increase removal efficiency of soluble micropollutant. It  
227 was found that about 100 mg/L of PAC dosage played a crucial role in adsorbing organic  
228 contaminant to overcome competitive adsorption with other organics present in bulk  
229 wastewater<sup>40</sup>. Xiao et al. (2017) investigated the removal efficiency of the pharmaceutical  
230 compounds in AnMBR with and without the addition of PAC<sup>41</sup>. The PAC addition improved  
231 the removal of the pharmaceutical compounds such as *Sulfamethoxazole* (Smx) and *Triclosan*  
232 (Tcs) during operational period. This enhancement was explained by the fact that a local  
233 substrate concentration could be increased in the mesoporous structure not only provided by  
234 carbon particles but also to their high adsorption capability. Therefore, the biodegradation  
235 became more thermodynamically favorable<sup>41</sup>. However, it was also found that the removal  
236 efficiency of *Carbamazepine* (Cbz) and *diclofenac* (Dcf) was very low, which is less than  
237 5 % because those chemicals are refractory against biodegradation<sup>41</sup>. Similar results were  
238 found by Wei et al., (2016) demonstrating the limited adsorption capacity provided by the  
239 GAC for the removal of OMPs during extended operational time with AnMBR<sup>40</sup>. Terzyk et  
240 al., (2003)<sup>42</sup> claimed that the abiotic reaction between phenol and PAC should affect  
241 irreversible adsorption<sup>40, 42</sup>. The abiotic transformation itself can be occurred by several steps.  
242 Firstly is the creation of strong complexes between the surface lactone and carbonyl groups  
243 of the carbon and phenolic group. Secondly is polymerization by the *superoxo* ion formed  
244 during the oxygen adsorption into PAC. However, oxygen is very limited in the AnMBR, thus

245 strong complexes between Cbz and Dcf and the surface groups of the carbon particles may be  
246 dominant in transformations of those micropollutants. During AnMBR operation, the carbon  
247 particles should also be occupied by the OMPs, thus mitigating the biotransformation of Cbz  
248 and Dcf <sup>42</sup>. On the other hand, the removal of the OMPs in the AnMBR can occur initially  
249 after adding the PAC followed by a gradual increase of OMP concentration to a certain level.  
250 As operation time progressed, the surface groups of PAC were occupied by the OMPs, thus  
251 decreasing biotransformation of Cbz and Dcf <sup>42</sup>.

252 On the other hand, Wei *et al.* (2016) <sup>40</sup> reported that the reduction of OMPs only  
253 occurred initially after the PAC addition in AnMBR followed by a gradual increase of OMPs  
254 concentration to the level before PAC addition. It was also predicted that the low dosage of  
255 PAC (100 mg/L) played a significant role for this limited adsorption capability considering  
256 the sorption competition with other bulk organics<sup>40</sup>. Granular activated carbon (GAC)  
257 particles were added as carrier materials for the growth of biofilm and physical scouring to  
258 clean membrane surface. More than 90 % in removal efficiency was achieved at 5 hr of total  
259 HRT <sup>43</sup>. Sorption into GAC particles as much as biofilm formation on GAC and its associated  
260 biodegradation form the basis of these two crucial pathways responsible of AFMBR  
261 pharmaceuticals removal<sup>34</sup>.

## 262 *Volatilization*

263 Volatilization of OMPs from dissolved to gas compartment is mainly determined by  
264 physicochemical properties of the OMPs. The volatility of OMPs can be characterized by the  
265 value of Henry's law constant ( $k_H$ ) in which  $k_H$  value ranging from  $10^{-2}$  to  $10^{-3}$  mol/(m<sup>3</sup>.Pa)  
266 corresponds to the high tendency of volatilization<sup>1</sup>. Suarez *et al.* (2008)<sup>38</sup> reported that  
267 volatilization of OMPs from estrogens and pharmaceuticals group is nearly negligible<sup>38</sup>.

268 However, it can also act as the major removal pathway for semi-volatile or non-  
 269 biodegradable OMPs. In addition, volatilization behavior is intensified in the wastewater  
 270 treatment with the biogas production or membrane sparging and temperature. In fact,  
 271 Feigenbrugel *et al.* (2004)<sup>84</sup> reported that the behaviour of Henry's law constants as a  
 272 function of temperature can be expressed with a Van't Hoff equation. Thus, when the  
 273 temperature increase from 25°C to 45°C, the volatilization also will increase by a factor of  
 274 8.44 for metalachlor and 12 for diazinon. The equation of an equilibrium condition in the  
 275 interface between the atmospheric and the wastewater surface is described as below in Eq  
 276 (6)<sup>38, 44</sup>.

$$277 \quad R_{\text{vol, surf}} = -V K_L a_{\text{surf}} C_f \frac{H_c}{H_c + (k_l/k_g)_{\text{surf}}} \quad (6)$$

278 In which:

279  $R_{\text{vol, surf}}$  : Rate of Volatilization ( $\text{gd}^{-1}$ )

280  $V$  : Volume ( $\text{m}^3$ )

281  $H_c$  : dimensionless Henry's law constant of compound (expressed as  $\text{m}^3$  of  
 282 wastewater/ $\text{m}^3$  of air)

283  $C_f$  : concentration of compound ( $\text{mg L}^{-1}$ )

284  $a_{\text{surf}}$  : specific interface surface area ( $\text{m}^{-1}$ )

285  $k_L$  : liquid transfer mass coefficient ( $\text{m d}^{-1}$ )

286  $k_G$  : gas transfer mass coefficient ( $\text{m d}^{-1}$ )

287  $K_{L a_{\text{surf}}}$  : the overall surface-desorption gas-transfer coefficient ( $\text{d}^{-1}$ )

288  $(k_l/k_g)_{\text{surf}}$  : Ratio of the liquid-phase mass-transfer coefficient to the gas-phase mass-transfer  
 289 coefficient for the surface volatilization

290

### 291 *Biological transformation*

292 Biotransformation of OMPs has been explored because microbial cells should have  
 293 the ability to breakdown the molecular structure of OMPs either totally (which is called

294 mineralization) or partially with the production of transformation products (TPs). These  
295 biotransformations can be divided into two reactions: (1) the metabolic reactions where the  
296 OMPs are used as carbon (anabolism) and/or energy (catabolism) sources for their cell  
297 development (growth, maintenance)<sup>21</sup> and (2) the co-metabolic reactions where the OMPs  
298 could not sustain growth but are transformed by side reactions catalyzed by broad-spectrum  
299 non-specific enzymes, in this case a primary substrate is needed<sup>45</sup>. In the case of organic  
300 micropollutants present at very low concentration in WWTP, it is expected that the OMPs  
301 could not sustain growth and that the primary substrate is needed. Models were developed in  
302 order to describe this co-metabolic reaction based on the original model developed by Criddle  
303 (1993)<sup>46</sup> and underlying the beneficial effect of the primary substrate<sup>47-49</sup>. Despite the various  
304 redox conditions occurring in WWTP, the biotransformation under aerobic conditions has  
305 been more studied than the one under anoxic or strictly anaerobic conditions<sup>50,51</sup>. Indeed, the  
306 microbial and functional diversity of anaerobic ecosystems met in WW, sludge and livestock  
307 treatments, may lead to the biotransformation of OMPs<sup>36,39</sup>. The most observed reactions are  
308 the reductive dehalogenation and the cleavage of ether bonds mainly observed with simple  
309 molecules using pure or enrichment cultures<sup>39,50,52</sup>. However in complex anaerobic systems  
310 treating sludge or livestock effluent (lab/pilot/industrial-scale), many compounds have been  
311 shown to be well removed under these reduced conditions like naproxen, atenolol, loratidine,  
312 miconazole, tramadol, domperidone, azithromycine, trimethoprim, tylosine, some  
313 sulfonamides (table 3), however (i) large variability in the removal rates depending on  
314 sampling and experimental conditions, (ii) no apparent relationship with their sorption  
315 capacity (Eq. 1) ,  $\text{Log}K_d$  or  $\text{Log}D$  explaining the partition between aqueous and particulate  
316 phases but not transformation which would be barely linked to their molecular structure and  
317 the presence of electron donating functional groups like  $-\text{NH}_2$ ,  $-\text{OH}$ ,  $-\text{CH}_3$  and (iii) no



318 knowledge on their real elimination pathways (abiotic/biotic, major or minor structural  
319 modification) since transformation products are rarely identified. Very recently, Spielmeyer et  
320 al., (2017)<sup>53</sup> have quantified transformative products (TPs) and assessed their antimicrobial  
321 activity for three sulfonamides like sulfadimethoxine, sulfamethoxypyridazine,  
322 sulfamethoxazole (SMX) during anaerobic digestion of manure under batch reactor spiked  
323 with the molecules [43]. Two TPs were isolated by coming from demethylation of  
324 sulfadimethoxine and were also subsequently removed. Comparison with autoclaved systems  
325 supported the hypothesis of biotic reactions. For sulfamethoxypyridazine, one TP was  
326 identified also coming from demethylation that was not further degraded during the batch  
327 reactor operation. SMX seemed to be biotransformed through the hydrogenation of the  
328 double bond and reductive cleavage of the heterocycle; a hydrogenation step was also  
329 mentioned by Alvarino et al., (2016)<sup>18</sup> for SMX. Another TP was also detected but coming  
330 from abiotic transformation. They also observed transformation of CTC to iso-CTC,  
331 transformation that occurred abiotically. Mono and polycyclic aromatic compounds (benzene,  
332 phenol, low molecular weight PAH) anaerobic transformation were also quite well  
333 documented in literature with description of metabolic routes and isolation of degrading  
334 microorganisms<sup>54</sup>.

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340 **Table 3.** The range of OMPs removal under anaerobic conditions. Underlined OMPs are the  
 341 one found in only one class of biological removal ( 28, 31, 39, 53, 55-73)

Class of removal	<30	<30-70>	>70
Sludge <sup>1</sup>	para, cbz, dcf, ibp, flx, <u>acid salicylic</u> , <u>gem</u> , ofl, nor, cip, LAS, <u>NP</u> , NP2EO, <u>PAH</u> , PCB, E1, E3, T, $\alpha$ EE2, $\alpha$ E2, $\beta$ E2, DEHP, BBP, DEP, BPA, ahtn, hhcb, triclosan, triclocarban, <u>diuron</u> , <u>benzotriazole</u> , <u>clozapine</u> , <u>benzophenone</u> , <u>iopromide</u> , <u>bisoprolol</u>	<u>pfoa</u> , pfos, para, cbz, propra, smx, cefo, esci, lido, vera, citalopram, keto, ibp, dcf, <u>diazepam</u> , roxi, ctc, ofl, nor, cip, LAS, NP2EO, PCB, E1, E3, T, $\alpha$ EE2, $\alpha$ E2, $\beta$ E2, DEHP, BBP, DEP, DnBP, BPA, ahtn, hhcb, triclosan, triclocarban	pfos, para, cbz, propra, smx, <u>azi</u> , cefo, esci, lido, <u>lora</u> , <u>mico</u> , <u>trama</u> , vera, <u>domp</u> , dcf, ibp, <u>ate</u> , <u>caf</u> , <u>trim</u> , <u>nap</u> , <u>oxybenzone</u> , roxi, otc, flx, citalopram, <u>furosemide</u> , <u>clofibric acid</u> , keto, nor, cip, NP2EO, NP1EO, E2, E1, $\alpha$ EE2, DEP, DnBP, BPA, ahtn, hhcb, triclosan
Livestock effluent <sup>2</sup>	sulfathiazole, sulfamethazine, sulfadiazine, <u>sulfaguanidine</u> , sulfamerazine, <u>sulfapyridine</u> , <u>monensine</u> , <u>doxycycline</u> , <u>tetracycline</u>	<u>oxytetracycline</u> , <u>sulfachloropyridazine</u> , sulfathiazole	<u>smx</u> , sulfamerazine, sulfadiazine, <u>sulfadimethoxine</u> , <u>sulfamethoxypyridazine</u> , <u>trimethoprim</u> , <u>tylosine</u> , <u>florfenicol</u> , <u>ampicillin</u> , <u>chlortetracycline</u>

342  
 343 **Note:** Cip: Ciprofloxacin, nor: Norfloxacin, ofl: Ofloxacin, para: Acetaminophen/paracetamol, az  
 344 i: Azithromycin, cbz: Carbamazepine, cefo: Cefoperazone, domp: Domperidone, esci: Escitalopr  
 345 am, glyben: Glybencyclamide, iver: Ivermectine, lido: Lidocaine, lora: Loratadine, mico: Micon  
 346 azole, propra: Propranolol, smx: Sulfamethoxazole, trama: Tramadol, vera: Verapamil, pfoa: per  
 347 fluorooctanoate, pfos: Perfluorooctane sulfonate, E1: Estrone, E3: Estriol, T: Testosterone,  $\alpha$ E2:  
 348 17- $\alpha$ -estradiol,  $\beta$ E2: 17- $\beta$ -estradiol,  $\alpha$ EE2: 17- $\alpha$ -ethinylestradiol, NP: nonylphenol, NP1EO: Nonyl  
 349 lphenol monoethoxylate, NP2EO: Nonylphenol diethoxylate, OP: octylphenol, BBP: Benzyl but  
 350 yl phthalate, DEHP: Diethyl hexyl phthalate, DEP: diethyl phthalate, DnBP: Di(n)butyl phthala  
 351 te, LAS: linear alkylbenzene sulfonate, dcf: diclofenac, ibp: ibuprofen, keto: ketoprofen, Roxi: R  
 352 oxithromycin, Ate: Atenolol, Caf: Caffeine, Trim: Trimethoprim, Nap: Naproxen, otc: oxytetrac  
 353 ycline, ctc: chlortetracycline, flx: fluoxetine, gem: gemfibrozil, PAH: Polycyclic aromatic hydro  
 354 carbons, PCB: polychlorobiphenyls, ahtn, hhcb: Tonalide, galaxolide, BPA: Bisphenol A, 4-OP:  
 355 4-(1,1,3,3-tetramethylbutyl)phenol, t-NP: technical-nonylphenol, 4-NP: 4-n-nonylphenol

356 It is assumed that biotransformation occurs only in the sludge phase when the OMP  
 357 is transferred from the liquid phase through several mechanisms. Firstly, sorption-desorption  
 358 can limit the biotransformation of a highly hydrophobic compound, and secondly small size  
 359 of OMPs allows them to diffuse through outer membrane in the microbial cell.  
 360 Biotransformation rate is strongly dependent upon the OMPs physicochemical and structural  
 361 properties, the enzymatic activities, the microbial diversity, and the environmental and  
 362 operational parameters <sup>36</sup>. The anaerobic digestion of sludge can be described by using a two-  
 363 steps model; first hydrolysis to particulate matter ( $X_s$ ) followed by biodegradation of soluble  
 364 substrate ( $S_s$ ) to biogas <sup>47</sup>.



368 In which, biomass growth rate is linked to soluble substrate uptake and modeled with Monod-  
 369 type kinetics:

370  $\mu = \mu_{max} \frac{S_s}{K_s + S_s}$  (9)

371 Where  $\mu_{max}$  (1/d) is the maximum bacterial growth rate, and  $K_s$  (gCOD/L) is the half-saturation  
 372 constant associated with the soluble substrate  $S_s$  <sup>47</sup>.

373 In 1993, Criddle modeled the cometabolism equation as shown in Eq. (1) between a growing  
 374 and non-growing substrate by biomass <sup>46</sup>. This equation was then modified by Delgadillo-  
 375 Mirquez *et al.* (2011) in which OMPs are considered as the non-growing substrate <sup>47</sup>.

376  $r_{bio} = \left( T_c \frac{\mu}{Y} + k_c \right) \left( \frac{C_{biog}}{K_{sc} + C_{biog}} \right) X$  (10)

377 Where

378  $r_{\text{bio}}$  = biotransformation rate constant

379  $T_c$  = OMPs transformation capacity ( $\mu\text{g}_{\text{OMP}}/\text{g}_{\text{COD-Ss}}$ ) standing for cometabolic interaction  
380 between the soluble substrate and OMPs metabolism,

381  $k_c$  = the maximum specific rate of OMPs in the absence of primary substrate ( $\mu\text{g}_{\text{OMP}}/\text{g}_{\text{COD-}}$   
382  $x.d$ )

383  $K_{\text{sc}}$  = the half saturated constant of OMP in monod formalism ( $\mu\text{g}_{\text{OMP}}/\text{L}$ )

384  $\mu$  = the growth rate (1/d)

385  $Y$  = the growth yield ( $\text{g}_{\text{COD-X}}/\text{g}_{\text{COD-S}}$ )

386  $C_{\text{bio}}$ = the concentration of soluble substrate to be biodegraded

387 Co-metabolisms is a process by which the OMPs are fortuitously biodegraded by using the  
388 enzyme produced during the metabolisms of other compounds<sup>74</sup>. The co-metabolisms always  
389 involve external organic compound as carbon and energy sources to partially or entirely  
390 biodegrade the OMPs in wastewaters. The co-metabolisms enable the biodegradation of  
391 OMPs which are far below the concentration which can be used as energy and carbon sources  
392 by microbial cells<sup>74</sup>. Structure analog is a chemical compound which is similar chemical  
393 structure to the target biodegradable OMP compound. Addition of structural analogue  
394 compound can encourage the growth and activity of microbial cells to degrade organic target  
395 compound<sup>75</sup>. Structural analogue influences biodegradation by acting as an inducer of  
396 catabolic pathway and substrate for co-metabolic in biodegradation of OMPs<sup>76</sup>.

397

398 *Liquid-liquid separation*

399 Membranes in AnMBR can always provide a useful tool to reject OMPs through membrane  
400 pore matrix. In most of the AnMBR system, porous membranes such as microfiltration or

401 ultrafiltration membrane are used directly into anaerobic bioreactor contributing to the  
402 removal of OMP as a result of size exclusion and charge repulsion where the  
403 physicochemical characteristics of the membrane and OMPs determine rejection capacity.  
404 However, the rejection efficiency of OMPs may not be too high due to smaller size of OMPs  
405 than the pore size of membrane applied<sup>77</sup>. Alvarino et al.(2018) report that hydrophobic  
406 membrane is able to retain lipophilic OMPs, whereas charged membranes can retain OMPs  
407 by electrostatic interaction<sup>18</sup>. In fact, with pore sizes in the range 0.01-0.1 $\mu$ m, the membrane  
408 sieving effect concerns only pollutants with a molecular weight higher than the molecular  
409 weight cut-off (MWCO) of membrane. Nevertheless, it has been reported that the removal of  
410 micropollutants may also be achieved by MF/UF due to the rejection/sorption by the foulants  
411 that act as a secondary barrier on membranes<sup>78, 79</sup>. Moreover, membrane allows also the  
412 retention of a significant part of the DOM and its associated OMP sorbed. As a result, the  
413 overall removal efficiency via biodegradation and direct rejection by membrane were  
414 increased<sup>40</sup>. Monsalvo et al. (2014)<sup>80</sup> characterized the distribution of trace organics within  
415 the fouling layer which is formed on the membrane in AnMBR and found that 17  $\alpha$ -  
416 ethinylestradiol, estrone, octyphenol, and bisphenol A, were retained mostly due to the layer  
417 of membrane fouling<sup>80</sup>. Rejections of the OMPs by membrane also increase the concentration  
418 polarization on membrane surface. Therefore, threshold value of wall-concentration of OMPs  
419 can be achieved to activate their biological degradation and thus shortening their adaptation  
420 time. Thus, the presence of a fouling layer on the membrane surface in AnMBR affect the  
421 rejection of OMPs due to the pore blockage, cake enhanced concentration-polarization, the  
422 change in surface charge of membrane and hydrophilicity<sup>78</sup>. Membrane fouling in AnMBR  
423 can be classified into (1) biofouling, (2) organic fouling, (3) colloidal fouling and (4)  
424 inorganic fouling<sup>81</sup>. Each type of membrane fouling formed can result in the different

425 rejection efficiency of OMPs. The presence of organic fouling on membrane surface  
426 improved the rejection of OMPs due to the enhancement of steric hindrance by the organic  
427 cake layer on membrane<sup>82</sup>. However, sometimes, the rejection can be decreased after organic  
428 fouling or colloidal fouling is formed on membrane<sup>79, 83</sup>.

### 429 **Global removal efficiency of OMPs by AnMBR and enhanced AnMBR**

430

431 Studies on the OMP removal in AnMBR have particular relevance to biodegradation,  
432 adsorption, and membrane filtration. Dutta et al., (2014)<sup>43</sup> observed the removal of the  
433 pharmaceuticals in anaerobic fluidized bed membrane bioreactor (AFMBR) as a second stage  
434 for the treatment of the effluent produced by anaerobic fluidized bed bioreactor (AFBR)  
435 treating a municipal wastewater<sup>43</sup>. Monsalvo et al. (2014)<sup>80</sup> investigated removal efficiency of  
436 38 OMP compounds including pharmaceuticals, personal care products, endocrine disruptors,  
437 and pesticides in AnMBR system<sup>80</sup>. Only 9 out of 38 OMPs were removed at higher than  
438 90 %, and the rest of them was removed only less than 50 % efficiency. With synthetic  
439 municipal wastewater, six OMPs such as *amitriptyline*, *diphenhydramine*, *flx*, *smx*, *TDCPP*,  
440 and *trim* were biodegraded more easily higher than 80 % of removal efficiency. However,  
441 *atrazine*, *cbz*, *DEET*, *Dilantin*, *primidone* and *TCEP* showed more refractory characteristics  
442 against biodegradation yielding less than 40 % removal efficiency. *Acetaminophen*, *atenolol*  
443 and *caf* required a prolonged time of about 45 hr for their adaptation to AnMBR environment.

444 The removal efficiencies of OMPs under anaerobic conditions should be influenced  
445 by operational parameters. The longer solid retention time (SRT) can often result in higher  
446 biomass and microbial diversity, thus improving the biodegradation of OMPs<sup>37, 84</sup>. However,  
447 the impact of SRT on removal efficiency was dependent upon the types of OMPs. Carballa et  
448 al (2007) observed higher removal efficiency of pharmaceuticals by longer SRT, but no

449 influence of SRT on personal care products (PCPs) during anaerobic digestion<sup>85</sup>. The  
450 biodegradation of some pharmaceuticals was correlated with the methanogenic rate and the  
451 biomass structure which can improve intra-molecular diffusion. Reactor hydrodynamic and  
452 biomass characteristics determine the sorption efficiency. The increase in the HRT could  
453 increase the sorption of PPCPs into biomass by providing enough contact time to reach  
454 sorption equilibrium <sup>86</sup>. Also, the OMP removal by sorption is dependent upon the biomass  
455 conformation and particle size distribution. Decreasing the HRT in AnMBR at extending  
456 SRT results in high biomass concentration, causing severe membrane fouling and  
457 deterioration of the effluent quality<sup>14</sup>. Obviously, trade-off in selecting HRT and SRT in  
458 AnMBR should exist between OMP removal efficiency and fouling mitigation. However,  
459 membrane fouling which is an inevitable phenomena in AnMBR should also be influenced  
460 significantly by operating parameters and types of foulants present. Thus, further  
461 optimization to control strategies for AnMBR needs to be conducted from mechanistic  
462 knowledge of fouling and probably developing process models to adaptation in practice<sup>87</sup>.

463 Bacterial population should be one of the key factors in the removal of OMPs. It was  
464 observed that antibiotic-type OMPs were more readily biodegraded by the anaerobic MBR  
465 rather than aerobic MBR systems<sup>7, 88</sup>. This could be attributed to a lower abundance of  
466 antibiotic resistant genes in the anaerobic environment<sup>3, 88</sup>. In addition, adsorption process  
467 into biomass is enhanced under anaerobic conditions due to higher concentration of soluble  
468 compound along with accumulation potential of micropollutants<sup>13, 89</sup>. Understanding the role  
469 of anaerobic stages such as acidogenesis and methanogenesis on the removal of OMPs should  
470 be importantly considered. Carnerio et al., categorized the contribution into three groups such  
471 as OMPs with a biotransformation significantly higher in acidogenic condition, OMPs  
472 equally transformed in both stages and the OMPs with biotransformation higher in

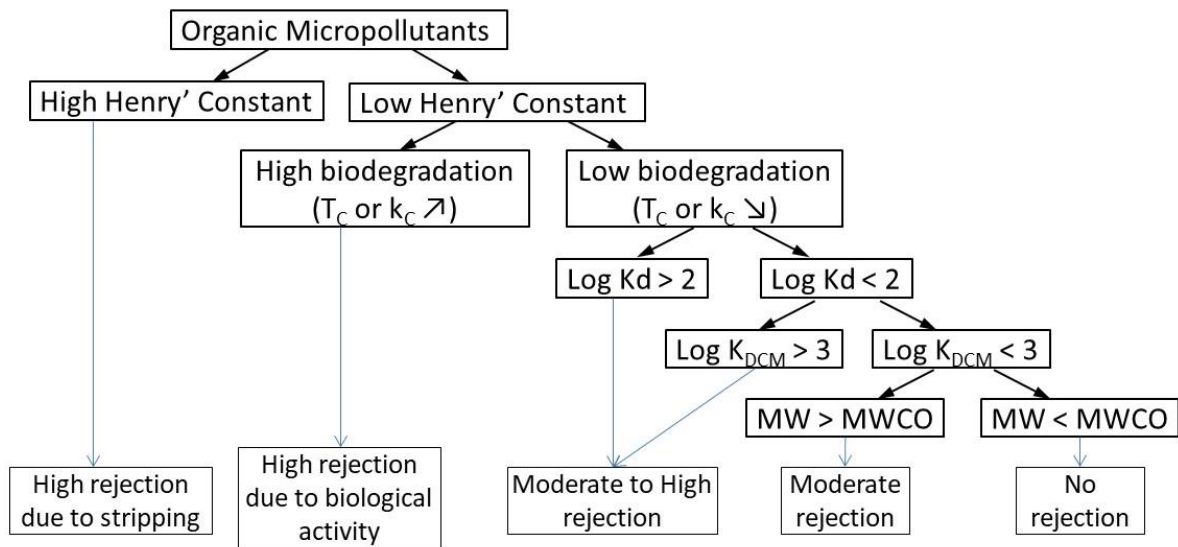
473 acetogenic/methanogenic conditions<sup>90</sup>. Nevertheless, more works are needed to elucidate  
474 dominant contribution of each microbial stage to the main metabolic pathways and products  
475 formed during the OMP biotransformation in AnMBR<sup>18</sup>.

476 The AnMBR is often combined with other unit technology for pre or post-treatment  
477 such as ozonation<sup>91</sup>, nanofiltration<sup>40</sup>, forward osmosis<sup>92</sup>, and membrane distillation<sup>93</sup>. In  
478 addition, the AnMBR is combined with carrier media such as GAC or PAC to improve the  
479 removal efficiency of the OMPs through biofilm growth<sup>41</sup>. Kaya et al., (2017)<sup>91</sup> combined  
480 ozonation as a pretreatment for the AnMBR system treating the wastewater containing  
481 *etodolac* from pharmaceutical industry<sup>91</sup>. Although activated carbon particles provide high  
482 surface area for biofilm formation and intrinsic adsorption capability, the breakage of them  
483 during AnMBR operation play roles as potential foulants against membrane  
484 performance<sup>94</sup>. Therefore, alternative media which are cost-effective to overcome the weakness  
485 of activated carbon will need to be developed for AnMBR. Here, the ozonation was used as a  
486 pretreatment to avoid sulfide inhibition due to oxidation of sulfate. The *Etodolac* was  
487 removed almost entirely with 99 % of removal efficiency by pre-ozonation<sup>91</sup>. Although  
488 overall reactor performance is not altered by the introduction of OMPs at low concentration,  
489 microbial community in AnMBR can be affected significantly<sup>88</sup>. The higher removal  
490 efficiency was observed by multiple antibiotic OMPs in the AnMBR as mentioned above, but  
491 the biodegradation associated with gene expression was impacted. Ozone has been known as  
492 very strong oxidant to removal micropollutants, but intermediates by-product can be present  
493 rather than their complete mineralization. Wang et al. observed that the AnMBR was adequate  
494 for the removal of the active ingredient of personal care products through both  
495 biotransformation and sorption phenomena onto the biosolids<sup>80</sup>. It was also found that the  
496 AnMBR resulted in effective removal of OMPs due to the fouling layer formed on membrane



497 which could play a role as a secondary membrane<sup>10</sup>. Removal diagram of OMP in AnMBR  
 498 is tabulated in Fig 2.

499



**Fig 2.** Removal diagram of OMP during anaerobic membrane bioreactor process<sup>95</sup>

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510 **Table 4.** Removal efficiency (%) of OMPs utilizing hybrid membrane bioreactor process<sup>18, 19,</sup>

511 31, 36, 40, 43, 53, 57-60, 64-68, 70, 73, 81, 88, 92, 93, 96, 97

Treatment	Removal efficiency (%)		
	Under 30 % removal (Moderate Rejection)	Between 30 and 70 % removal (Moderate to High Rejection)	Above 70 % removal (High Rejection due to Biological Activity and Stripping)
Anaerobic Membrane Bioreactor (AnMBR)	Ate, $\alpha$ E2, $\alpha$ EE2, atrazine, cbz, clozapine, DEET, dcf, Dilantin, E3, E1, gem, hydroxyzine, ibp keto, linuron, meprobamate, omeprazole, primidone	$\beta$ E2, Amytriptyline, BPA, enalapril, etiochlolanolone, paracetamol, triclocarban, trim	Androstenedione, androsterone, caf, metformin, naproxen, nonylphenol, OP, smx, T, triclosan, vera
Two staged AFBR-AFMBR + GAC as carrier material	-	-	Sulfadiazine, smx, sulfathiazole, sulfamethazine. Erythromycin-H <sub>2</sub> O, clarithromycin, josamycin, roxithromycin, tylosin, nalidixic acid, flumequine, pipemidic acid, norfloxacin, ciprofloxacin, ofloxacin, cephalixin, cephradine, Trim, cbz, psychastimulants, caf, ibp, naproxen, ketoprofen, dcf
AnMBR with GAC/PAC	-	-	Trim, smx, cbz, dcf, triclosan
AnMBR followed by	-	-	Etodolac

ozonation			
AnMBR followed by NF	-	-	Para, DEET, caf, atrazine, cbz, primidone, Dilantin, smx, diphenhydramine, ate, amitriptyline, TCEP, and flx
AnMBR followed by FO	-	-	Caf, ate, atrazine
AnMBR followed by MD	-	-	Caf, smx, keto, trim, para, nap, primidone, ibp, triamterene, carazolol, TCEP, dcf, cbz, gem, simazine, amitriptyline, atrazine, diuron, propylparaben, linuron, clozapine, phenylphenol A, diazinon, triclosan, triclocarban
SAnMBR followed by light, oxygen and microalgae treatment	-	-	4-OP, t-NP, 4-NP, BPA

512

513 It is always attractive to combine AnMBR with high-pressure driven membrane such  
514 as reverse osmosis for wastewater reuse purpose. Table 4 summarizes removal efficiency of  
515 OMPs in various hybrid anaerobic membrane bioreactor process. Cho et al. (1999) observed  
516 that either nanofiltration (NF) or reverse osmosis (RO) membrane indicated more than 85 %  
517 of the rejection efficiency of the biodegradable or non-biodegradable OMPs present in  
518 wastewater <sup>98</sup>. In addition, Wei et al., (2016) reported high rejection efficiency of 80-92%

519 with 15 OMPs from various OMPs sources such as pharmaceuticals, personal care products,  
520 and pesticides from integrated AnMBR-NF system<sup>40</sup>. The removal efficiency of the OMPs by  
521 integrated AnMBR-NF process (87 %) was higher than that by the NF membrane system  
522 alone. The fouling formed on the NF membrane by pretreated anionic ion exchange effluent  
523 can vary the effectiveness of the rejection of the OMPs due to their surface charge. The  
524 rejection of the OMPs, which is more positively charged, can be lower than that of the OMPs,  
525 which are more negatively charged due to electrostatic interactions between the foulants and  
526 membrane. Comparison of fouled membrane and non-fouled membrane on the rejection of  
527 OMPs has also been investigated. In spite of high removal efficiency of OMPs with RO or  
528 NF membrane followed by AnMBR, operational energy is still very high to provide very high  
529 hydraulic pressure to allow water passage through membrane. Recently, low-pressure driven  
530 membrane process with high rejection efficiency of OMPs from AnMBR effluent has been  
531 considered significantly. For instance, forward osmosis (FO) which is osmotically-driven  
532 membrane process are integrated to remove OMPs from wastewaters. Linares et al. compared  
533 the rejection of the 13 OMPs selected as target contaminants through the clean and fouled FO  
534 membrane treating secondary effluent<sup>99</sup>. The rejection of the OMPs was increased except for  
535 the hydrophilic neutral OMPs in the presence of membrane fouling. The higher rejection  
536 corresponded to higher hydrophilicity of the FO membrane, which is fouled rather than clean  
537 membrane. Negatively charged membrane surface caused by foulant composition increased  
538 adsorption capacity, thereby decreasing mass transport capacity accordingly<sup>99</sup>. Concern on  
539 FO process is that appropriate draw solution with high salt concentration is always needed to  
540 provide osmotic pressure to the membrane. Additional process to reconcentrate draw  
541 solution also needed in the sustainable operation of FO membrane. The effect of the foulant  
542 on the rejection of OMPs is also reported to be pore size-dependent. The effect of the humic

543 acid foulant on the rejection of OMPs was investigated by different NF membrane pore sizes  
544 <sup>9</sup>. The enhancement of OMPs rejection through NF270 membranes was associated with the  
545 pore blocking due to hydrophobic interactions between OMPs and membrane<sup>9</sup>. It was found  
546 that about 99 % of the removal efficiency of OMPs was achieved by combining forward  
547 osmosis membrane with AnMBR for the treatment of its effluent <sup>92</sup>. Transport of OMPs  
548 through RO membrane was influenced significantly by their surface charge. The rejection of  
549 the hydrophobic OMPs was improved owing to hydrophobic-hydrophobic repulsion as  
550 hydrophobic membrane was used to reject them<sup>92</sup>. Another study also showed that membrane  
551 distillation process complements AnMBR to improve the removal efficiency of 26 OMPs  
552 with 76 % of overall removal efficiency <sup>93</sup>.

553         The OMPs which are not degraded biologically can be removed by the absorption of  
554 photon from the wavelength found in sunlight, especially Visible Light, Infrared Radiation  
555 and UV light. Photo-oxidation involves the break-up of OMPs by photons into smaller pieces  
556 and the change of their shape into the form that can be altered by proteins denaturing and the  
557 supplement of other atoms or molecules<sup>100</sup>. Recently, there has been attempts to observe the  
558 influence of photo-oxidation to the removal of OMPs from the effluent treated by AnMBR<sup>89</sup>.  
559 Arbagues *et al.* (2013) observed the effect of light to the removal of four different  
560 micropollutants, such as 4-(1,1,3,3-tetramethylbutyl)phenol, technical-nonylphenol (t-NP), 4-  
561 n-nonylphenol (4-NP), and Bisphenol-A (BPA). Although the removal rate varied depending  
562 upon the types of OMPs used as compared to aerobic condition, almost complete removal of  
563 4-NP was observed by AnMBR-Photooxidation process <sup>89</sup>. Assessing degradation rate of  
564 EDC present in the effluents produced by AnMBR was also studied by using microalgae.

565 High dissolved oxygen concentrations produced by algae can enhance the degradation rates  
566 and as such light intensity should be one of dominant parameters<sup>101</sup>.

567 **Future perspectives and research efforts.**

568

569 This paper describes the state-of-the-art of removal behavior of OMPs observed in  
570 AnMBR for improving system designs and operations. Obviously, the AnMBR provides great  
571 way to produce high removal efficiency of the OMPs at low energy consumptions from  
572 various wastewaters. Although many efforts to better understand the removal mechanisms of  
573 the OMPs by AnMBR system have been conducted, more solid understanding of it needs to  
574 be achieved. Practical application of the AnMBR to remove OMPs from wastewaters put  
575 forth thus far by research community is still very limited. From this review, it is indeed that  
576 the removal of OMPs from the AnMBR has good correlation with operational conditions,  
577 thus further exploration to investigate these features systematically is desired. Although the  
578 extent of OMPs removal efficiency in AnMBR is understood, it varies greatly depending of  
579 the nature of the contaminants, far less works has been reported on real wastewaters. Because  
580 of fundamental differences among removal pathways, as well as modes of operation and  
581 reactor designs, modeling and experimental approaches to understanding dominant removal  
582 mechanisms need to be developed. Elucidating metabolic pathways during the  
583 biotransformation of OMPs should also need further studies particularly under various  
584 temperature regimes. It is always attractive to develop integrated AnMBR process, but more  
585 in-depth understanding on the removals of OMPs which may occur in pre- or post-treatment  
586 combined with the AnMBR reactor should help to optimize reactor design parameters and  
587 intensify the process.

588

589 **Conclusions**

590

591 Primary pathway affecting the removals of OMPs in AnMBR is the sorption which is  
592 driven by physicochemical characteristics of both contaminants and biomasses present in  
593 bulk and/or grown in suspended carriers. Longer SRT can increase the withdrawalss of OMPs  
594 due to higher biomass level and microbial diversity. In addition, the sorption into biomass can  
595 be improved by increasing HRT. Sorption-desorption between OMPs and biomass should be  
596 a rate-limiting step to determine the bio-transformative rate. Although intrinsic removal of the  
597 OMPs through the porous membrane often applied in AnMBR is very low, the biofilm  
598 formed on membrane surface can improve their removal significantly as much as the  
599 Dissolved and Colloidal Matter (DCM) retention. Similarly, biomass carriers such as carbon-  
600 based media used in AnMBR should contribute to the enhancement of biosorption and  
601 biotransformation of OMPs as operational time is increased. In addition, high temperature  
602 (around 40°C), biogas production and sparging significantly increased OMP volatilization  
603 according to Arrhenius analysis

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607

608 **References**

- 609 1. Y. Luo, W. Guo, H. H. Ngo, L. D. Nghiem, F. I. Hai, J. Zhang, S. Liang and X. C. Wang, A  
610 review on the occurrence of micropollutants in the aquatic environment and their fate and  
611 removal during wastewater treatment, *Sci. Total Environ.*, 2014, **473**, 619-641.
- 612 2. K. Fent, A. A. Weston and D. Caminada, Ecotoxicology of human pharmaceuticals, *Aquat.*  
613 *Toxicol.*, 2006, **76**, 122-159.
- 614 3. Y. M. Amha, M. Corbett and A. L. Smith, Two-Phase Improves Performance of Anaerobic  
615 Membrane Bioreactor Treatment of Food Waste at High Organic Loading Rates, *Environ.*

- 616 *Sci. Technol.*, 2019, **53**, 9572-9583.
- 617 4. J. Kim, K. Kim, H. Ye, E. Lee, C. Shin, P. L. McCarty and J. Bae, Anaerobic fluidized bed  
618 membrane bioreactor for wastewater treatment, *Environ. Sci. Technol.*, 2010, **45**, 576-581.
- 619 5. D. Jeison, W. Van Betuw and J. Van Lier, Feasibility of anaerobic membrane bioreactors for  
620 the treatment of wastewaters with particulate organic matter, *Separation Science and*  
621 *Technology*, 2008, **43**, 3417-3431.
- 622 6. P. L. McCarty, J. Bae and J. Kim, Domestic wastewater treatment as a net energy producer—  
623 can this be achieved? *Journal*, 2011.
- 624 7. M. Harb, E. Lou, A. L. Smith and L. B. Stadler, Perspectives on the fate of micropollutants in  
625 mainstream anaerobic wastewater treatment, *Current opinion in biotechnology*, 2019, **57**,  
626 94-100.
- 627 8. D. Cheng, H. Ngo, W. Guo, Y. Liu, J. Zhou, S. Chang, D. Nguyen, X. Bui and X. Zhang,  
628 Bioprocessing for elimination antibiotics and hormones from swine wastewater, *Sci. Total*  
629 *Environ.*, 2018, **621**, 1664-1682.
- 630 9. L. Zhu, Rejection of organic micropollutants by clean and fouled nanofiltration membranes,  
631 *J. Chem.*, 2015, **2015**.
- 632 10. D. Cheng, H. H. Ngo, W. Guo, Y. Liu, S. W. Chang, D. D. Nguyen, L. D. Nghiem, J. Zhou and  
633 B. Ni, Anaerobic membrane bioreactors for antibiotic wastewater treatment: performance  
634 and membrane fouling issues, *Bioresour. Technol.*, 2018, **267**, 714-724.
- 635 11. M. Aslam, A. Charfi, G. Lesage, M. Heran and J. Kim, Membrane bioreactors for wastewater  
636 treatment: a review of mechanical cleaning by scouring agents to control membrane  
637 fouling, *Chem. Eng. J.*, 2017, **307**, 897-913.
- 638 12. C. Chen, W. Guo, H. Ngo, Y. Liu, B. Du, Q. Wei, D. Wei, D. Nguyen and S. Chang, Evaluation  
639 of a sponge assisted-granular anaerobic membrane bioreactor (SG-AnMBR) for municipal  
640 wastewater treatment, *Renewable Energy*, 2017, **111**, 620-627.
- 641 13. M. Abargues, J. Ferrer, A. Bouzas and A. Seco, Fate of endocrine disruptor compounds in  
642 an anaerobic membrane bioreactor (AnMBR) coupled to an activated sludge reactor,  
643 *Environmental Science: Water Research & Technology*, 2018, **4**, 226-233.
- 644 14. J. Ma, R. Dai, M. Chen, S. J. Khan and Z. Wang, Applications of membrane bioreactors for  
645 water reclamation: micropollutant removal, mechanisms and perspectives, *Bioresour.*  
646 *Technol.*, 2018, **269**, 532-543.
- 647 15. S. Martin Ruel, J.-M. Choubert, H. Budzinski, C. Miège, M. Esperanza and M. Coquery,  
648 Occurrence and fate of relevant substances in wastewater treatment plants regarding  
649 Water Framework Directive and future legislations, *Water Sci. Technol.*, 2012, **65**, 1179-  
650 1189.
- 651 16. T. A. Ternes, M. Stumpf, J. Mueller, K. Haberer, R.-D. Wilken and M. Servos, Behavior and  
652 occurrence of estrogens in municipal sewage treatment plants—I. Investigations in



- 653 Germany, Canada and Brazil, *Sci. Total Environ.*, 1999, **225**, 81-90.
- 654 17. P. Verlicchi, M. Al Aukidy and E. Zambello, Occurrence of pharmaceutical compounds in  
655 urban wastewater: removal, mass load and environmental risk after a secondary  
656 treatment—a review, *Sci. Total Environ.*, 2012, **429**, 123-155.
- 657 18. T. Alvarino, S. Suarez, J. Lema and F. Omil, Understanding the sorption and  
658 biotransformation of organic micropollutants in innovative biological wastewater treatment  
659 technologies, *Sci. Total Environ.*, 2018, **615**, 297-306.
- 660 19. J. Margot, L. Rossi, D. A. Barry and C. Holliger, A review of the fate of micropollutants in  
661 wastewater treatment plants, *Wiley Interdisciplinary Reviews: Water*, 2015, **2**, 457-487.
- 662 20. M. Pomiès, J.-M. Choubert, C. Wisniewski and M. Coquery, Modelling of micropollutant  
663 removal in biological wastewater treatments: a review, *Sci. Total Environ.*, 2013, **443**, 733-  
664 748.
- 665 21. H. Siegrist and A. Joss, Review on the fate of organic micropollutants in wastewater  
666 treatment and water reuse with membranes, *Water Sci. Technol.*, 2012, **66**, 1369-1376.
- 667 22. S. C. Monteiro and A. B. Boxall, in *Rev. Environ. Contam. Toxicol.*, Springer, 2010, pp. 53-  
668 154.
- 669 23. A. Abtahi, S. M. Mazza, S. M. Ryno, E. K. Loya, R. Li, S. R. Parkin, C. Risko, J. E. Anthony and  
670 K. R. Graham, Effect of Halogenation on the Energetics of Pure and Mixed Phases in  
671 Model Organic Semiconductors Composed of Anthradithiophene Derivatives and C60, *J.*  
672 *Phys. Chem. C.*, 2018, **122**, 4757-4767.
- 673 24. M. Bourgin, B. Beck, M. Boehler, E. Borowska, J. Fleiner, E. Salhi, R. Teichler, U. Von Gunten,  
674 H. Siegrist and C. S. McArdell, Evaluation of a full-scale wastewater treatment plant  
675 upgraded with ozonation and biological post-treatments: Abatement of micropollutants,  
676 formation of transformation products and oxidation by-products, *Water Res.*, 2018, **129**,  
677 486-498.
- 678 25. M. E. Casas and K. Bester, Can those organic micro-pollutants that are recalcitrant in  
679 activated sludge treatment be removed from wastewater by biofilm reactors (slow sand  
680 filters)?, *Sci. Total Environ.*, 2015, **506**, 315-322.
- 681 26. J. Garcia-Ivars, L. Martella, M. Massella, C. Carbonell-Alcaina, M.-I. Alcaina-Miranda and M.-  
682 I. Iborra-Clar, Nanofiltration as tertiary treatment method for removing trace  
683 pharmaceutically active compounds in wastewater from wastewater treatment plants,  
684 *Water Res.*, 2017, **125**, 360-373.
- 685 27. R. Mailler, J. Gasperi, Y. Coquet, A. Buleté, E. Vulliet, S. Deshayes, S. Zedek, C. Mirande-Bret,  
686 V. Eudes and A. Bressy, Removal of a wide range of emerging pollutants from wastewater  
687 treatment plant discharges by micro-grain activated carbon in fluidized bed as tertiary  
688 treatment at large pilot scale, *Sci. Total Environ.*, 2016, **542**, 983-996.
- 689 28. R. Mailler, J. Gasperi, Y. Coquet, S. Deshayes, S. Zedek, C. Cren-Olivé, N. Cartiser, V. Eudes,

- 690 A. Bressy and E. Caupos, Study of a large scale powdered activated carbon pilot: Removals  
691 of a wide range of emerging and priority micropollutants from wastewater treatment plant  
692 effluents, *Water Res.*, 2015, **72**, 315-330.
- 693 29. V. Matamoros, J. García and J. M. Bayona, Organic micropollutant removal in a full-scale  
694 surface flow constructed wetland fed with secondary effluent, *Water Res.*, 2008, **42**, 653-  
695 660.
- 696 30. Y. Wang, W. Gao, Y. Wang and G. Jiang, Suspect screening analysis of the occurrence and  
697 removal of micropollutants by GC-QTOF MS during wastewater treatment processes, *J.*  
698 *Hazard. Mater.*, 2019, **376**, 153-159.
- 699 31. J. Malmberg and J. Magnér, Pharmaceutical residues in sewage sludge: effect of  
700 sanitization and anaerobic digestion, *Journal of environmental management*, 2015, **153**, 1-  
701 10.
- 702 32. A. Bergé, L. Wiest, R. Baudot, B. Giroud and E. Vulliet, Occurrence of multi-class surfactants  
703 in urban wastewater: contribution of a healthcare facility to the pollution transported into  
704 the sewerage system, *Environmental Science and Pollution Research*, 2018, **25**, 9219-9229.
- 705 33. J. Wang, Z. Tian, Y. Huo, M. Yang, X. Zheng and Y. Zhang, Monitoring of 943 organic  
706 micropollutants in wastewater from municipal wastewater treatment plants with secondary  
707 and advanced treatment processes, *Journal of Environmental Sciences*, 2018, **67**, 309-317.
- 708 34. D. L. McCurry, S. E. Bear, J. Bae, D. L. Sedlak, P. L. McCarty and W. A. Mitch, Superior  
709 removal of disinfection byproduct precursors and pharmaceuticals from wastewater in a  
710 staged anaerobic fluidized membrane bioreactor compared to activated sludge, *Environ.*  
711 *Sci. Technol. Lett.*, 2014, **1**, 459-464.
- 712 35. M. Arslan, I. Ullah, J. A. Müller, N. Shahid and M. Afzal, in *Enhancing cleanup of*  
713 *environmental pollutants*, Springer, 2017, pp. 65-99.
- 714 36. L. Gonzalez-Gil, M. Mauricio-Iglesias, D. Serrano, J. M. Lema and M. Carballa, Role of  
715 methanogenesis on the biotransformation of organic micropollutants during anaerobic  
716 digestion, *Sci. Total Environ.*, 2018, **622**, 459-466.
- 717 37. J. Sipma, B. Osuna, N. Collado, H. Monclús, G. Ferrero, J. Comas and I. Rodriguez-Roda,  
718 Comparison of removal of pharmaceuticals in MBR and activated sludge systems,  
719 *Desalination*, 2010, **250**, 653-659.
- 720 38. S. Suárez, M. Carballa, F. Omil and J. M. Lema, How are pharmaceutical and personal care  
721 products (PPCPs) removed from urban wastewaters?, *Rev. Environ. Sci. Bio/Technol.*, 2008,  
722 **7**, 125-138.
- 723 39. M. Barret, D. Patureau, E. Latrille and H. Carrère, A three-compartment model for  
724 micropollutants sorption in sludge: Methodological approach and insights, *Water Res.*,  
725 2010, **44**, 616-624.
- 726 40. C.-H. Wei, C. Hoppe-Jones, G. Amy and T. Leiknes, Organic micro-pollutants' removal via

- 727 anaerobic membrane bioreactor with ultrafiltration and nanofiltration, *J. Water Reuse*  
728 *Desalin.*, 2016, **6**, 362-370.
- 729 41. Y. Xiao, H. Yaohari, C. De Araujo, C. C. Sze and D. C. Stuckey, Removal of selected  
730 pharmaceuticals in an anaerobic membrane bioreactor (AnMBR) with/without powdered  
731 activated carbon (PAC), *Chem. Eng. J.*, 2017, **321**, 335-345.
- 732 42. A. P. Terzyk, Further insights into the role of carbon surface functionalities in the  
733 mechanism of phenol adsorption, *J. Colloid Interface Sci.*, 2003, **268**, 301-329.
- 734 43. K. Dutta, M.-Y. Lee, W. W.-P. Lai, C. H. Lee, A. Y.-C. Lin, C.-F. Lin and J.-G. Lin, Removal of  
735 pharmaceuticals and organic matter from municipal wastewater using two-stage anaerobic  
736 fluidized membrane bioreactor, *Bioresour. Technol.*, 2014, **165**, 42-49.
- 737 44. K.-C. Lee, B. E. Rittmann, J. Shi and D. McAvoy, Advanced steady-state model for the fate  
738 of hydrophobic and volatile compounds in activated sludge, *Water environment research*,  
739 1998, **70**, 1118-1131.
- 740 45. B. E. Rittmann and J. A. Manem, Development and experimental evaluation of a  
741 steady-state, multispecies biofilm model, *Biotechnol. Bioeng.*, 1992, **39**, 914-922.
- 742 46. C. S. Criddle, The kinetics of cometabolism, *Biotechnol. Bioeng.*, 1993, **41**, 1048-1056.
- 743 47. L. Delgadillo-Mirquez, L. Lardon, J.-P. Steyer and D. Patureau, A new dynamic model for  
744 bioavailability and cometabolism of micropollutants during anaerobic digestion, *Water Res.*,  
745 2011, **45**, 4511-4521.
- 746 48. E. Fernandez-Fontaina, M. Carballa, F. Omil and J. Lema, Modelling cometabolic  
747 biotransformation of organic micropollutants in nitrifying reactors, *Water Res.*, 2014, **65**,  
748 371-383.
- 749 49. B. G. Plósz, C. Vogelsang, K. Macrae, H. H. Heiaas, A. Lopez, H. Liltved and K. H. Langford,  
750 The BIOZO process—a biofilm system combined with ozonation: occurrence of xenobiotic  
751 organic micro-pollutants in and removal of polycyclic aromatic hydrocarbons and nitrogen  
752 from landfill leachate, *Water Sci. Technol.*, 2010, **61**, 3188-3197.
- 753 50. A.-K. Ghattas, F. Fischer, A. Wick and T. A. Ternes, Anaerobic biodegradation of (emerging)  
754 organic contaminants in the aquatic environment, *Water Res.*, 2017, **116**, 268-295.
- 755 51. C. Grandclement, I. Seyssiecq, A. Piram, P. Wong-Wah-Chung, G. Vanot, N. Tiliacos, N.  
756 Roche and P. Doumenq, From the conventional biological wastewater treatment to hybrid  
757 processes, the evaluation of organic micropollutant removal: a review, *Water Res.*, 2017,  
758 **111**, 297-317.
- 759 52. H. Smidt and W. M. de Vos, Anaerobic microbial dehalogenation, *Annu. Rev. Microbiol.*,  
760 2004, **58**, 43-73.
- 761 53. A. Spielmeyer, F. Stahl, M. S. Petri, W. Zerr, H. Brunn and G. Hamscher, Transformation of  
762 sulfonamides and tetracyclines during anaerobic fermentation of liquid manure, *J. Environ.*  
763 *Qual.*, 2017, **46**, 160-168.

- 764 54. R. Rabus, M. Boll, J. Heider, R. U. Meckenstock, W. Buckel, O. Einsle, U. Ermler, B. T. Golding,  
765 R. P. Gunsalus and P. M. Kroneck, Anaerobic microbial degradation of hydrocarbons: from  
766 enzymatic reactions to the environment, *J. Mol. Microbiol. Biotechnol.*, 2016, **26**, 5-28.
- 767 55. Ç. Akyol, S. Aydin, O. Ince and B. Ince, A comprehensive microbial insight into single-stage  
768 and two-stage anaerobic digestion of oxytetracycline-medicated cattle manure, *Chem. Eng.*  
769 *J.*, 2016, **303**, 675-684.
- 770 56. J. A. Álvarez, L. Otero, J. Lema and F. Omil, The effect and fate of antibiotics during the  
771 anaerobic digestion of pig manure, *Bioresour. Technol.*, 2010, **101**, 8581-8586.
- 772 57. L. T. Angenent, M. Mau, U. George, J. A. Zahn and L. Raskin, Effect of the presence of the  
773 antimicrobial tylosin in swine waste on anaerobic treatment, *Water Res.*, 2008, **42**, 2377-  
774 2384.
- 775 58. O. A. Arikan, L. J. Sikora, W. Mulbry, S. U. Khan, C. Rice and G. D. Foster, The fate and effect  
776 of oxytetracycline during the anaerobic digestion of manure from therapeutically treated  
777 calves, *Process Biochem.*, 2006, **41**, 1637-1643.
- 778 59. M. Carballa, F. Omil and J. M. Lema, Calculation methods to perform mass balances of  
779 micropollutants in sewage treatment plants. Application to pharmaceutical and personal  
780 care products (PPCPs), *Environ. Sci. Technol.*, 2007, **41**, 884-890.
- 781 60. A. Ezzariai, M. Hafidi, A. Khadra, Q. Aemig, L. El Fels, M. Barret, G. Merlina, D. Patureau and  
782 E. Pinelli, Human and veterinary antibiotics during composting of sludge or manure:  
783 Global perspectives on persistence, degradation, and resistance genes, *J. Hazard. Mater.*,  
784 2018, **359**, 465-481.
- 785 61. L. Gonzalez-Gil, M. Papa, D. Feretti, E. Ceretti, G. Mazzoleni, N. Steimberg, R. Pedrazzani, G.  
786 Bertanza, J. Lema and M. Carballa, Is anaerobic digestion effective for the removal of  
787 organic micropollutants and biological activities from sewage sludge?, *Water Res.*, 2016,  
788 **102**, 211-220.
- 789 62. S. M. Mitchell, J. L. Ullman, A. L. Teel, R. J. Watts and C. Frear, The effects of the antibiotics  
790 ampicillin, florfenicol, sulfamethazine, and tylosin on biogas production and their  
791 degradation efficiency during anaerobic digestion, *Bioresour. Technol.*, 2013, **149**, 244-252.
- 792 63. S. A. Mohring, I. Strzysch, M. R. Fernandes, T. K. Kiffmeyer, J. Tuerk and G. Hamscher,  
793 Degradation and elimination of various sulfonamides during anaerobic fermentation: a  
794 promising step on the way to sustainable pharmacy?, *Environ. Sci. Technol.*, 2009, **43**,  
795 2569-2574.
- 796 64. M. Muller, S. Combalbert, N. Delgenès, V. Bergheaud, V. Rocher, P. Benoît, J.-P. Delgenès, D.  
797 Patureau and G. Hernandez-Raquet, Occurrence of estrogens in sewage sludge and their  
798 fate during plant-scale anaerobic digestion, *Chemosphere*, 2010, **81**, 65-71.
- 799 65. M. Narumiya, N. Nakada, N. Yamashita and H. Tanaka, Phase distribution and removal of  
800 pharmaceuticals and personal care products during anaerobic sludge digestion, *J. Hazard.*

- 801 *Mater.*, 2013, **260**, 305-312.
- 802 66. N. Paterakis, T. Chiu, Y. Koh, J. Lester, E. McAdam, M. Scrimshaw, A. Soares and E. Cartmell,  
803 The effectiveness of anaerobic digestion in removing estrogens and nonylphenol  
804 ethoxylates, *J. Hazard. Mater.*, 2012, **199**, 88-95.
- 805 67. H. V. Phan, R. Wickham, S. Xie, J. A. McDonald, S. J. Khan, H. H. Ngo, W. Guo and L. D.  
806 Nghiem, The fate of trace organic contaminants during anaerobic digestion of primary  
807 sludge: A pilot scale study, *Bioresour. Technol.*, 2018, **256**, 384-390.
- 808 68. V. G. Samaras, A. S. Stasinakis, N. S. Thomaidis, D. Mamais and T. D. Lekkas, Fate of  
809 selected emerging micropollutants during mesophilic, thermophilic and temperature co-  
810 phased anaerobic digestion of sewage sludge, *Bioresour. Technol.*, 2014, **162**, 365-372.
- 811 69. A. Spielmeier, B. Breier, K. Großmeier and G. Hamscher, Elimination patterns of worldwide  
812 used sulfonamides and tetracyclines during anaerobic fermentation, *Bioresour. Technol.*,  
813 2015, **193**, 307-314.
- 814 70. E. Trably, N. Delgènes, D. Patureau and J. Philippe Delgènes, Statistical tools for the  
815 optimization of a highly reproducible method for the analysis of polycyclic aromatic  
816 hydrocarbons in sludge samples, *Int. J. Environ. Anal. Chem.*, 2004, **84**, 995-1008.
- 817 71. E. Trably, D. Patureau and J. Delgenes, Enhancement of polycyclic aromatic hydrocarbons  
818 removal during anaerobic treatment of urban sludge, *Water Sci. Technol.*, 2003, **48**, 53-60.
- 819 72. V. Varel, J. Wells, W. Shelver, C. Rice, D. Armstrong and D. Parker, Effect of anaerobic  
820 digestion temperature on odour, coliforms and chlortetracycline in swine manure or  
821 monensin in cattle manure, *Journal of applied microbiology*, 2012, **112**, 705-715.
- 822 73. G. Zeeman, K. Kujawa, T. De Mes, L. Hernandez, M. De Graaff, L. Abu-Ghunmi, A. Mels, B.  
823 Meulman, H. Temmink and C. Buisman, Anaerobic treatment as a core technology for  
824 energy, nutrients and water recovery from source-separated domestic waste (water), *Water*  
825 *Sci. Technol.*, 2008, **57**, 1207-1212.
- 826 74. T. C. Hazen, Cometabolic bioremediation, *Consequences of Microbial Interactions with*  
827 *Hydrocarbons, Oils, and Lipids: Biodegradation and Bioremediation*, 2018, 1-15.
- 828 75. W. Brunner, F. H. Sutherland and D. D. Focht, Enhanced Biodegradation of Polychlorinated  
829 Biphenyls in Soil by Analog Enrichment and Bacterial Inoculation 1, *J. Environ. Qual.*, 1985,  
830 **14**, 324-328.
- 831 76. L. J. Shaw and R. G. Burns, Biodegradation of organic pollutants in the rhizosphere, *Adv.*  
832 *Appl. Microbiol.*, 2003, **53**, 1-60.
- 833 77. E. Sahar, I. David, Y. Gelman, H. Chikurel, A. Aharoni, R. Messalem and A. Brenner, The use  
834 of RO to remove emerging micropollutants following CAS/UF or MBR treatment of  
835 municipal wastewater, *Desalination*, 2011, **273**, 142-147.
- 836 78. L. D. Nghiem, P. J. Coleman and C. Ependiller, Mechanisms underlying the effects of  
837 membrane fouling on the nanofiltration of trace organic contaminants, *Desalination*, 2010,

- 838 **250**, 682-687.
- 839 79. L. D. Nghiem and S. Hawkes, Effects of membrane fouling on the nanofiltration of  
840 pharmaceutically active compounds (PhACs): mechanisms and role of membrane pore size,  
841 *Sep. Purif. Technol.*, 2007, **57**, 176-184.
- 842 80. V. M. Monsalvo, J. A. McDonald, S. J. Khan and P. Le-Clech, Removal of trace organics by  
843 anaerobic membrane bioreactors, *Water Res.*, 2014, **49**, 103-112.
- 844 81. A. R. Verliefde, E. R. Cornelissen, S. Heijman, I. Petrinic, T. Luxbacher, G. Amy, B. Van der  
845 Bruggen and J. Van Dijk, Influence of membrane fouling by (pretreated) surface water on  
846 rejection of pharmaceutically active compounds (PhACs) by nanofiltration membranes, *J.*  
847 *Membr. Sci.*, 2009, **330**, 90-103.
- 848 82. K. V. Plakas, A. Karabelas, T. Wintgens and T. Melin, A study of selected herbicides  
849 retention by nanofiltration membranes—the role of organic fouling, *J. Membr. Sci.*, 2006,  
850 **284**, 291-300.
- 851 83. H. Y. Ng and M. Elimelech, Influence of colloidal fouling on rejection of trace organic  
852 contaminants by reverse osmosis, *J. Membr. Sci.*, 2004, **244**, 215-226.
- 853 84. E. Fernandez-Fontaina, I. Pinho, M. Carballa, F. Omil and J. M. Lema, Biodegradation kinetic  
854 constants and sorption coefficients of micropollutants in membrane bioreactors,  
855 *Biodegradation*, 2013, **24**, 165-177.
- 856 85. M. Carballa, F. Omil, T. Ternes and J. M. Lema, Fate of pharmaceutical and personal care  
857 products (PPCPs) during anaerobic digestion of sewage sludge, *Water Res.*, 2007, **41**,  
858 2139-2150.
- 859 86. T. Alvarino, S. Suarez, J. Lema and F. Omil, Understanding the removal mechanisms of  
860 PPCPs and the influence of main technological parameters in anaerobic UASB and aerobic  
861 CAS reactors, *J. Hazard. Mater.*, 2014, **278**, 506-513.
- 862 87. Á. Robles, M. V. Ruano, A. Charfi, G. Lesage, M. Heran, J. Harmand, A. Seco, J.-P. Steyer, D. J.  
863 Batstone and J. Kim, A review on anaerobic membrane bioreactors (AnMBRs) focused on  
864 modelling and control aspects, *Bioresour. Technol.*, 2018, **270**, 612-626.
- 865 88. M. Harb, C.-H. Wei, N. Wang, G. Amy and P.-Y. Hong, Organic micropollutants in aerobic  
866 and anaerobic membrane bioreactors: Changes in microbial communities and gene  
867 expression, *Bioresour. Technol.*, 2016, **218**, 882-891.
- 868 89. M. Abargues, J. Ferrer, A. Bouzas and A. Seco, Removal and fate of endocrine disruptors  
869 chemicals under lab-scale posttreatment stage. Removal assessment using light, oxygen  
870 and microalgae, *Bioresour. Technol.*, 2013, **149**, 142-148.
- 871 90. R. B. Carneiro, L. Gonzalez-Gil, Y. A. Londoño, M. Zaiat, M. Carballa and J. M. Lema,  
872 Acidogenesis is a key step in the anaerobic biotransformation of organic micropollutants, *J.*  
873 *Hazard. Mater.*, 2019, 121888.
- 874 91. Y. Kaya, A. M. Bacaksiz, H. Bayrak, Z. B. Gönder, I. Vergili, H. Hasar and G. Yilmaz, Treatment

- 875 of chemical synthesis-based pharmaceutical wastewater in an ozonation-anaerobic  
876 membrane bioreactor (AnMBR) system, *Chem. Eng. J.*, 2017, **322**, 293-301.
- 877 92. Y. Kim, S. Li, L. Chekli, Y. C. Woo, C.-H. Wei, S. Phuntsho, N. Ghaffour, T. Leiknes and H. K.  
878 Shon, Assessing the removal of organic micro-pollutants from anaerobic membrane  
879 bioreactor effluent by fertilizer-drawn forward osmosis, *J. Membr. Sci.*, 2017, **533**, 84-95.
- 880 93. X. Song, W. Luo, J. McDonald, S. J. Khan, F. I. Hai, W. E. Price and L. D. Nghiem, An  
881 anaerobic membrane bioreactor–membrane distillation hybrid system for energy recovery  
882 and water reuse: Removal performance of organic carbon, nutrients, and trace organic  
883 contaminants, *Sci. Total Environ.*, 2018, **628**, 358-365.
- 884 94. M. Aslam and J. Kim, Investigating membrane fouling associated with GAC fluidization on  
885 membrane with effluent from anaerobic fluidized bed bioreactor in domestic wastewater  
886 treatment, *Environmental Science and Pollution Research*, 2019, **26**, 1170-1180.
- 887 95. C. Bellona, J. E. Drewes, P. Xu and G. Amy, Factors affecting the rejection of organic solutes  
888 during NF/RO treatment—a literature review, *Water Res.*, 2004, **38**, 2795-2809.
- 889 96. O. A. Arikan, Degradation and metabolization of chlortetracycline during the anaerobic  
890 digestion of manure from medicated calves, *J. Hazard. Mater.*, 2008, **158**, 485-490.
- 891 97. M. Lim, R. Ahmad, J. Guo, F. Tibi, M. Kim and J. Kim, Removals of micropollutants in staged  
892 anaerobic fluidized bed membrane bioreactor for low-strength wastewater treatment,  
893 *rocess Saf. Environ. Prot.*, 2019, **127**, 162-170.
- 894 98. J. Cho, G. Amy and J. Pellegrino, Membrane filtration of natural organic matter: initial  
895 comparison of rejection and flux decline characteristics with ultrafiltration and  
896 nanofiltration membranes, *Water Res.*, 1999, **33**, 2517-2526.
- 897 99. R. V. Linares, V. Yangali-Quintanilla, Z. Li and G. Amy, Rejection of micropollutants by clean  
898 and fouled forward osmosis membrane, *Water Res.*, 2011, **45**, 6737-6744.
- 899 100. E. Yousif and R. Haddad, Photodegradation and photostabilization of polymers, especially  
900 polystyrene, *SpringerPlus*, 2013, **2**, 398.
- 901 101. M. Abargues, J. Giménez, J. Ferrer, A. Bouzas and A. Seco, Endocrine disrupter compounds  
902 removal in wastewater using microalgae: Degradation kinetics assessment G RA PHICAL  
903 AB STRACT.

904

905