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► To cite this version:

Michael Lim, Dominique Steyer Patureau, Marc Héran, Geoffroy Lesage, Jeonghwan Kim. Removal of organic micropollutants in anaerobic membrane bioreactors in wastewater treatment: critical review. *Environmental Science: Water Research and Technology*, 2020, 6 (5), pp.1230-1243. 10.1039/c9ew01058k . hal-02650858

HAL Id: hal-02650858

<https://hal.inrae.fr/hal-02650858>

Submitted on 21 Sep 2023

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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 security¹. Those risks include
38 short-term or long-term toxicity, environmental persistence, antibiotic resistance, endocrine
39 disruption etc². 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^{3,4}. 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 %)⁵, (2) low energy consumption and (3)
50 production of bioenergy in the form of methane⁶ 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 AnMBR⁷. Sorption of OMPs from the aqueous phase to biomass should
56 occur due to hydrophobic interactions and electrostatic interactions (cation bridging and
57 exchange)^{1, 8}. 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 μm .
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 membrane⁹. 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 rejection¹⁰.

69 Nowadays, there has been an upsurge of interests in AnMBR in which biomass
70 carriers are suspended for domestic wastewater treatment¹¹. 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^{4, 11,}
75 ¹². 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 significantly⁴. 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 sewage¹³. However, understanding the pathways on degrading the OMPs through AFMBR
80 system needs more experimental works and practical applications with real wastewaters¹⁴.

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)^{1, 4, 11, 15-17}.

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 sludge¹⁸⁻²¹. 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^{1, 17, 22}. 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^{15, 19, 23-29}. 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 uses¹. The spectrum of
126 molecules found in sewage is very wide as exemplified in Table 1³⁰. 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 mussels³¹. The concentrations found in influents range from the order of ng/L for
131 pharmaceuticals¹⁷ to mg/L particularly for detergents³². In conventional wastewater treatment
132 plants, the concentrations are generally lower in secondary effluent than those in the primary
133 effluent³³. 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 transformation³⁴. which are generally
136 found in domestic sewage.

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143 **Table 1.** Types of organic micropollutants ^{1, 35}

OMPs	Classes	Major Sources	Examples
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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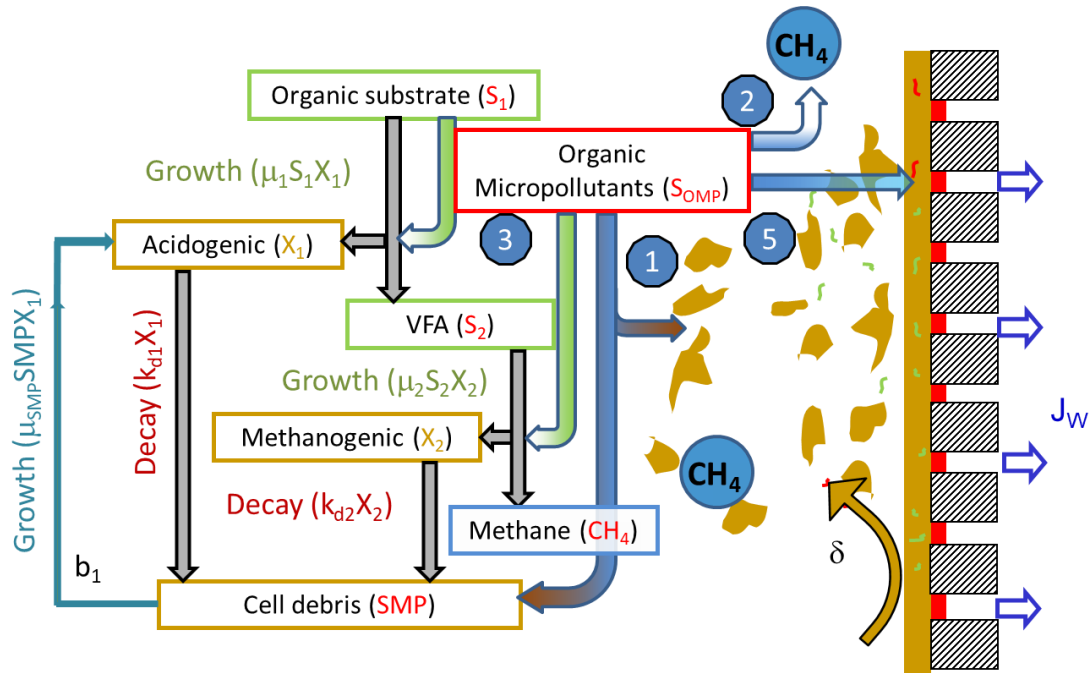
155 **Removal pathways of organic micropollutants**

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)** ^{1, 35}

	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

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163

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

164

165 *Sorption to biomass or adsorbents*

166 Sorption to the biomass has been suggested as one of the primary mechanisms
 167 affecting the removal of the OMPs in anaerobic treatment³⁶. In fact, the sorption of OMP to
 168 biomass should be differentiated into two mechanisms : (1) Sorption in which the OMPs
 169 move from aqueous phase and (2) penetration into the lipophilic cell membrane, which is
 170 driven by their hydrophobicity. The OMPs are also sorbed onto surface of biomass due to
 171 electrostatic interactions between the biomass with negative surface charge and OMPs with
 172 positive surface charge³⁷ or negative one thanks to cation bridging. Suarez *et al.* (2008)
 173 reported that the sorption of OMPs by the biomass could be determined by the value of the
 174 Octanol-Water partitioning coefficient (K_{ow}). In case of ionizable compounds, the K_{ow} can be
 175 modified into distribution partitioning coefficient (D_{ow}) by taken in account the acid

176 disassociation constant (pKa) as shown in Eq.(1)³⁸. 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 biomass³⁸, 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_{free} , mg/L),
 194 the sorbed to Dissolved and Colloidal Matter (C_{DCM} , mg/g_{DCM}), and the sorbed to particles
 195 (C_{part} , mg/g_{PART})³⁹. 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 conditions³⁹. 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 interaction¹⁸.

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 wastewater⁴⁰. Xiao et al. (2017) investigated the removal efficiency of the pharmaceutical
230 compounds in AnMBR with and without the addition of PAC⁴¹. 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 favorable⁴¹. 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 biodegradation⁴¹. 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 AnMBR⁴⁰. Terzyk et
240 al., (2003)⁴² claimed that the abiotic reaction between phenol and PAC should affect
241 irreversible adsorption^{40, 42}. 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 ⁴². 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 ⁴².

252 On the other hand, Wei *et al.* (2016) ⁴⁰ 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 organics⁴⁰. 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 ⁴³. 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 removal³⁴.

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/(m³.Pa)
266 corresponds to the high tendency of volatilization¹. Suarez *et al.* (2008)³⁸ reported that
267 volatilization of OMPs from estrogens and pharmaceuticals group is nearly negligible³⁸.

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)⁸⁴ 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)^{38, 44}.

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

278 In which:

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

280 V : Volume (m^3)

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

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

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

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

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

287 $K_{L a_{\text{surf}}}$: the overall surface-desorption gas-transfer coefficient (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)²¹ 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 needed⁴⁵. 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)⁴⁶ and underlying the beneficial effect of the primary substrate⁴⁷⁻⁴⁹. 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^{50,51}. 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^{36,39}. 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^{39,50,52}. 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)⁵³ 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)¹⁸ 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 microorganisms⁵⁴.

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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 ¹	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, α EE2, α E2, β 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, α EE2, α E2, β 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, α EE2, DEP, DnBP, BPA, ahtn, hhcb, triclosan
Livestock effluent ²	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, α E2:
 348 17- α -estradiol, β E2: 17- β -estradiol, α EE2: 17- α -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 ³⁶. 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 ⁴⁷.



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

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

371 Where μ_{max} (1/d) is the maximum bacterial growth rate, and K_s (g_{COD}/L) is the half-saturation
 372 constant associated with the soluble substrate S_s ⁴⁷.

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

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

377 Where

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

384 μ = the growth rate (1/d)

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

386 C_{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 compounds⁷⁴. 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 cells⁷⁴. 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 compound⁷⁵. Structural analogue influences biodegradation by acting as an inducer of
396 catabolic pathway and substrate for co-metabolic in biodegradation of OMPs⁷⁶.

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 applied⁷⁷. Alvarino et al.(2018) report that hydrophobic
406 membrane is able to retain lipophilic OMPs, whereas charged membranes can retain OMPs
407 by electrostatic interaction¹⁸. In fact, with pore sizes in the range 0.01-0.1 μ 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^{78, 79}. 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 increased⁴⁰. Monsalvo et al. (2014)⁸⁰ characterized the distribution of trace organics within
415 the fouling layer which is formed on the membrane in AnMBR and found that 17 α -
416 ethinylestradiol, estrone, octyphenol, and bisphenol A, were retained mostly due to the layer
417 of membrane fouling⁸⁰. 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 hydrophilicity⁷⁸. Membrane fouling in AnMBR
423 can be classified into (1) biofouling, (2) organic fouling, (3) colloidal fouling and (4)
424 inorganic fouling⁸¹. 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 membrane⁸². However, sometimes, the rejection can be decreased after organic
428 fouling or colloidal fouling is formed on membrane^{79, 83}.

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)⁴³ 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 wastewater⁴³. Monsalvo et al. (2014)⁸⁰ investigated removal efficiency of
436 38 OMP compounds including pharmaceuticals, personal care products, endocrine disruptors,
437 and pesticides in AnMBR system⁸⁰. 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^{37, 84}. 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 digestion⁸⁵. 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 ⁸⁶. 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 quality¹⁴. 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 practice⁸⁷.

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^{7, 88}. This could be attributed to a lower abundance of
466 antibiotic resistant genes in the anaerobic environment^{3, 88}. 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^{13, 89}. 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 conditions⁹⁰. 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 AnMBR¹⁸.

476 The AnMBR is often combined with other unit technology for pre or post-treatment
477 such as ozonation⁹¹, nanofiltration⁴⁰, forward osmosis⁹², and membrane distillation ⁹³. 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 growth⁴¹. Kaya et al., (2017)⁹¹ combined
480 ozonation as a pretreatment for the AnMBR system treating the wastewater containing
481 *etodolac* from pharmaceutical industry⁹¹. 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 performance⁹⁴. 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-ozonation⁹¹. Although
488 overall reactor performance is not altered by the introduction of OMPs at low concentration,
489 microbial community in AnMBR can be affected significantly⁸⁸. 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 biosolids⁸⁰. 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 membrane¹⁰. Removal diagram of OMP in AnMBR
 498 is tabulated in Fig 2.

499

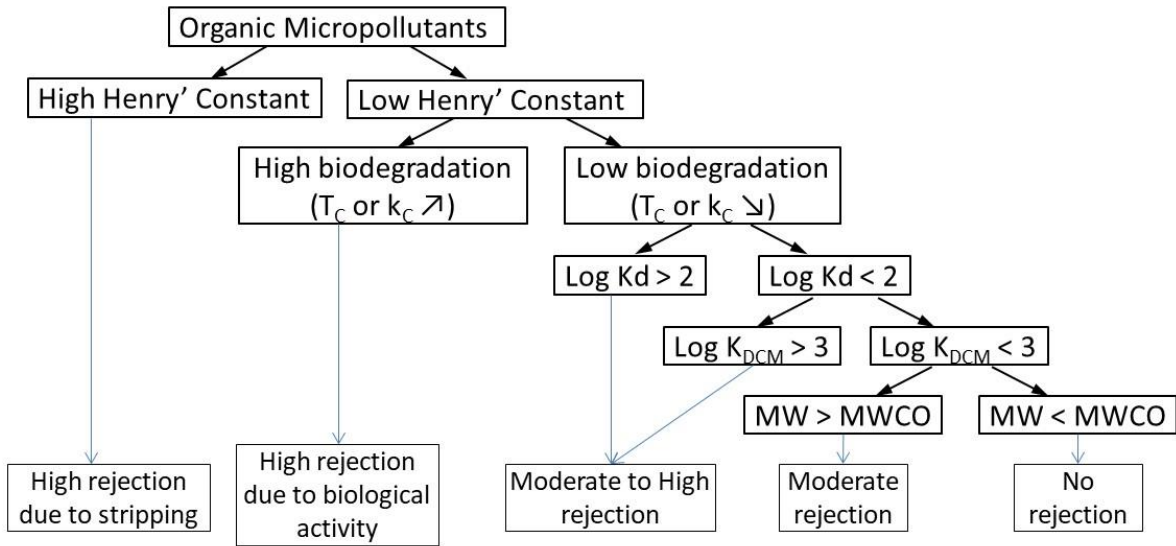


Fig 2. Removal diagram of OMP during anaerobic membrane bioreactor process⁹⁵

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510 **Table 4.** Removal efficiency (%) of OMPs utilizing hybrid membrane bioreactor process^{18, 19,}

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, α E2, α EE2, atrazine, cbz, clozapine, DEET, dcf, Dilantin, E3, E1, gem, hydroxyzine, ibp keto, linuron, meprobamate, omeprazole, primidone	β 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 ₂ 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 ⁹⁸. 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 system⁴⁰. 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 effluent⁹⁹. 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 accordingly⁹⁹. 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 ⁹. The enhancement of OMPs rejection through NF270 membranes was associated with the
545 pore blocking due to hydrophobic interactions between OMPs and membrane⁹. 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 ⁹². 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 them⁹². 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 ⁹³.

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 molecules¹⁰⁰. Recently, there has been attempts to observe the
558 influence of photo-oxidation to the removal of OMPs from the effluent treated by AnMBR⁸⁹.
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 ⁸⁹. 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 parameters¹⁰¹.

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

604 **Acknowledgment**

605 This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea
606 government(MSIT) (2019R1A2C1087530).

607

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