Removal of organic micropollutants in anaerobic membrane bioreactors in wastewater treatment: critical review

Michael Lim, Dominique Steyer Patureau, Marc Héran, Geoffroy Lesage, Jeonghwan Kim

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

Michael Lim[1,2], Dominique Patureau[3], Marc Heran[4], Geoffroy Lesage[5] and Jeonghwan Kim[1]*

[1]Department of Environmental Engineering, Inha University, Incheon, Republic of Korea

[2]Department of Chemical and Materials Engineering, The University of Auckland, Auckland, New Zealand

[3]LBE, Univ Montpellier, INRA, Narbonne, France

[4]IEM, Univ Montpellier,, CNRS, ENSCM, Montpellier, France

*corresponding author: jeonghwankim@inha.ac.kr

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.

Keywords

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

Introduction

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

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

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

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

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

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

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

Organic micropollutants in AnMBR

Organic micropollutants (OMPs) issues in wastewater

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

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

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

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

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

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, salycilic 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, terbuthylazine, 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, solvant	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

Removal pathways of organic micropollutants

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

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

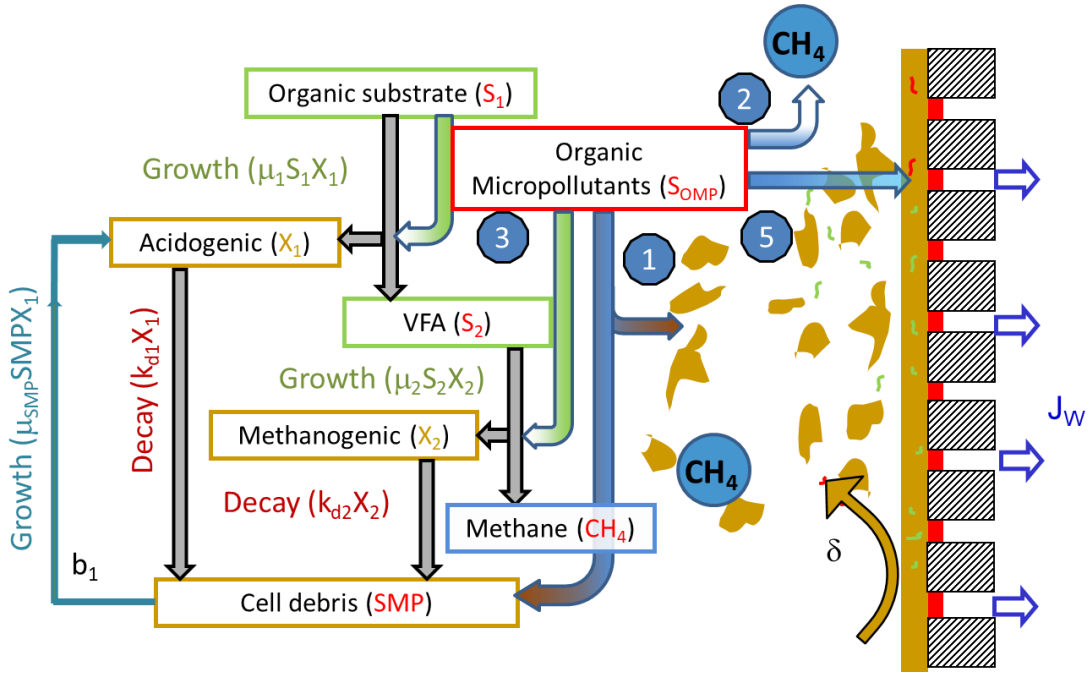

Fig 1. Removal pathway of organic micropollutant in anaerobic membrane bioreactor

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[36]. 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[37] 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

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

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

$$K_d = \frac{C_s}{C_L X_{TSS}} \qquad (2)$$

Where:

K_{ow} : octanol-water partitioning coefficient

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

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

C_S : the concentration in the solid phase (µg/L)

C_L : the concentration in the liquid phase (µg/L)

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

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

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

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

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

Where:

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

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

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

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

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

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

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

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

Volatilization

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

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

$$R_{vol.surf} = -V\ K_L\ a_{surf}\ C_f\ \frac{Hc}{Hc+(^{kl}/_{k_G})\ surf} \qquad (6)$$

In which:

$R_{vol.surf}$: Rate of Volatilization (gd^{-1})

V : Volume (m^3)

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

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

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

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

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

K_La_{surf} : the overall surface-desorption gas-transfer coefficient (d^{-1})

$(^{kl}/_{k_G})\ surf$: Ratio of the liquid-phase mass-transfer coefficient to the gas-phase mass-transfer coefficient for the surface volatilization

Biological transformation

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

mineralization) or partially with the production of transformation products (TPs). These biotransformations can be divided into two reactions: (1) the metabolic reactions where the OMPs are used as carbon (anabolism) and/or energy (catabolism) sources for their cell development (growth, maintenance)[21] and (2) the co-metabolic reactions where the OMPs could not sustain growth but are transformed by side reactions catalyzed by broad-spectrum non-specific enzymes, in this case a primary substrate is needed[45]. In the case of organic micropollutants present at very low concentration in WWTP, it is expected that the OMPs could not sustain growth and that the primary substrate is needed. Models were developed in order to describe this co-metabolic reaction based on the original model developed by Criddle (1993) [46] and underlying the beneficial effect of the primary substrate[47-49]. Despite the various redox conditions occurring in WWTP, the biotransformation under aerobic conditions has been more studied than the one under anoxic or strictly anaerobic conditions[50, 51]. Indeed, the microbial and functional diversity of anaerobic ecosystems met in WW, sludge and livestock treatments, may lead to the biotransformation of OMPs[36, 39]. The most observed reactions are the reductive dehalogenation and the cleavage of ether bonds mainly observed with simple molecules using pure or enrichment cultures[39, 50, 52]. However in complex anaerobic systems treating sludge or livestock effluent (lab/pilot/industrial-scale), many compounds have been shown to be well removed under these reduced conditions like naproxen, atenolol, loratidine, miconazole, tramadol, domperidone, azithromycine, trimethoprim, tylosine, some sulfonamides (table 3), however (i) large variability in the removal rates depending on sampling and experimental conditions, (ii) no apparent relationship with their sorption capacity (Eq. 1) , $LogK_d$ or LogD explaining the partition between aqueous and particulate phases but not transformation which would be barely linked to their molecular structure and the presence of electron donating functional groups like $–NH_2$, -OH, $-CH_3$ and (iii) no

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

Table 3. The range of OMPs removal under anaerobic conditions. Underlined OMPs are the 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, acid salicylic, gem, ofl, nor, cip, LAS, NP, NP2EO, PAH, PCB, E1, E3, T, αEE2, αE2, βE2, DEHP, BBP, DEP, BPA, ahtn, hhcb, triclosan, triclocarban, diuron, benzotriazole, clozapine, benzophenone, iopromide, bisoprolol	pfoa, pfos, para, cbz, propra, smx, cefo, esci, lido, vera, citalopram, keto, ibp, dcf, diazepam, roxi, ctc, ofl, nor, cip, LAS, NP2EO, PCB, E1, E3, T, αEE2, αE2, βE2, DEHP, BBP, DEP, DnBP, BPA, ahtn, hhcb, triclosan, triclocarban	pfos, para, cbz, propra, smx, azi, cefo, esci, lido, lora, mico, trama, vera, domp, dcf, ibp, ate, caf, trim, nap, oxybenzone, roxi, otc, flx, citalopram, furosemide, clofibric acid, keto, nor, cip, NP2EO, NP1EO, E2, E1, αEE2, DEP, DnBP, BPA, ahtn, hhcb, triclosan
Livestock effluent[2]	sulfathiazole, sulfamethazine, sulfadiazine, sulfaguanidine, sulfamerazine, sulfapyridine monensine, doxycycline, tetracycline	oxytetracycline, sulfachloropyridazine, sulfathiazole	smx, sulfamerazine, sulfadiazine, sulfadimethoxine, sulfamethoxypyridazine, trimethoprim, tylosine, florfenicol, ampicillin, chlortetracycline

Note: Cip: Ciprofloxacin, nor: Norfloxacin, ofl: Ofloxacin, para: Acetaminophen/paracetamol, azi: Azithromycin, cbz: Carbamazepine, cefo: Cefoperazone, domp: Domperidone, esci: Escitalopram, glyben: Glybencyclamide, iver: Ivermectine, lido: Lidocaine, lora: Loratadine, mico: Miconazole, propra: Propranolol, smx: Sulfamethoxazole, trama: Tramadol, vera: Verapamil, pfoa: perfluorooctanoate, pfos: Perfluorooctane sulfonate, E1: Estrone, E3: Estriol, T: Testosterone, αE2: 17-α-estradiol, βE2: 17-β-estradiol, αEE2: 17-α-ethinylestradiol, NP: nonylphenol, NP1EO: Nonylphenol monoethoxylate, NP2EO: Nonylphenol diethoxylate, OP: octylphenol, BBP: Benzyl butyl phthalate, DEHP: Diethyl hexyl phthalate, DEP: diethyl phthalate, DnBP: Di(n)butyl phthalate, LAS: linear alkylbenzene sulfonate, dcf: diclofenac, ibp: ibuprofen, keto: ketoprofen, Roxi: Roxithromycin, Ate: Atenolol, Caf: Caffeine, Trim: Trimethoprim, Nap: Naproxen, otc: oxytetracycline, ctc: chlortetracycline, flx: fluoxetine, gem: gemfibrozil, PAH: Polycyclic aromatic hydrocarbons, PCB: polychlorobiphenyls, ahtn, hhcb: Tonalide, galaxolide, BPA: Bisphenol A, 4-OP: 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].

365
366 $S_p \xrightarrow{k_{hyd}} S_s$ (7)

367 $S_s \xrightarrow{\mu} X + CH_4 + CO_2$ (8)

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

377 Where

r_{bio} = biotransformation rate constant

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

k_c = the maximum specific rate of OMPs in the absence of primary substrate ($\mu g_{OMP}/g_{COD\text{-}X}.d$)

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

μ = the growth rate (1/d)

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

C_{biov}= the concentration of soluble substrate to be biodegraded

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

Liquid-liquid separation

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

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

rejection efficiency of OMPs. The presence of organic fouling on membrane surface improved the rejection of OMPs due to the enhancement of steric hindrance by the organic cake layer on membrane[82]. However, sometimes, the rejection can be decreased after organic fouling or colloidal fouling is formed on membrane[79, 83].

Global removal efficiency of OMPs by AnMBR and enhanced AnMBR

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

The removal efficiencies of OMPs under anaerobic conditions should be influenced by operational parameters. The longer solid retention time (SRT) can often result in higher biomass and microbial diversity, thus improving the biodegradation of OMPs[37, 84]. However, the impact of SRT on removal efficiency was dependent upon the types of OMPs. Carballa et 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[85]. The
450 biodegration 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 quality[14]. 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 operationg 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[87].

463 Bacterial population should be one of the key fators 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 attriubuted 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

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

The AnMBR is often combined with other unit technology for pre or post-treatment such as ozonation[91], nanofiltration[40], forward osmosis[92], and membrane distillation [93]. In addition, the AnMBR is combined with carrier media such as GAC or PAC to improve the removal efficiency of the OMPs through biofilm growth[41]. Kaya et al., (2017)[91] combined ozonation as a pretreatment for the AnMBR system treating the wastewater containing *etodolac* from pharmaceutical industry[91]. Although activated carbon particles provide high surface area for biofilm formation and intrinsic adsorption capability, the breakage of them during AnMBR operation play roles as potential foulants agains membrane performance[94].Thereore, altnertive media which are cost-effective to overcome the weakeness of activate carbon will need to be developed for AnMBR. Here, the ozonation was used as a pretreatment to avoid sulfide inhibition due to oxidation of sulfate. The *Etodolac* was removed almost entirely with 99 % of removal efficiency by pre-ozonation[91]. Although overall reactor performance is not altered by the introduction of OMPs at low concentration, microbial community in AnMBR can be affected significantly[88]. The higher removal efficiency was observed by multiple antibiotic OMPs in the AnMBR as mentioned above, but the biodegradation associated with gene expression was impacted. Ozone has been known as very strong oxidant to removal micropollutants, but intermediates by-produce can be present rather than their complete minerization. Wang et al. observed that the AnMBR was adequate for the removal of the active ingredient of personal care products through both biotransformation and sorption phenomena onto the biosolids[80]. It was also found that the 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[10]. Removal diagram of OMP in AnMBR

498 is tabulated in Fig 2.

499

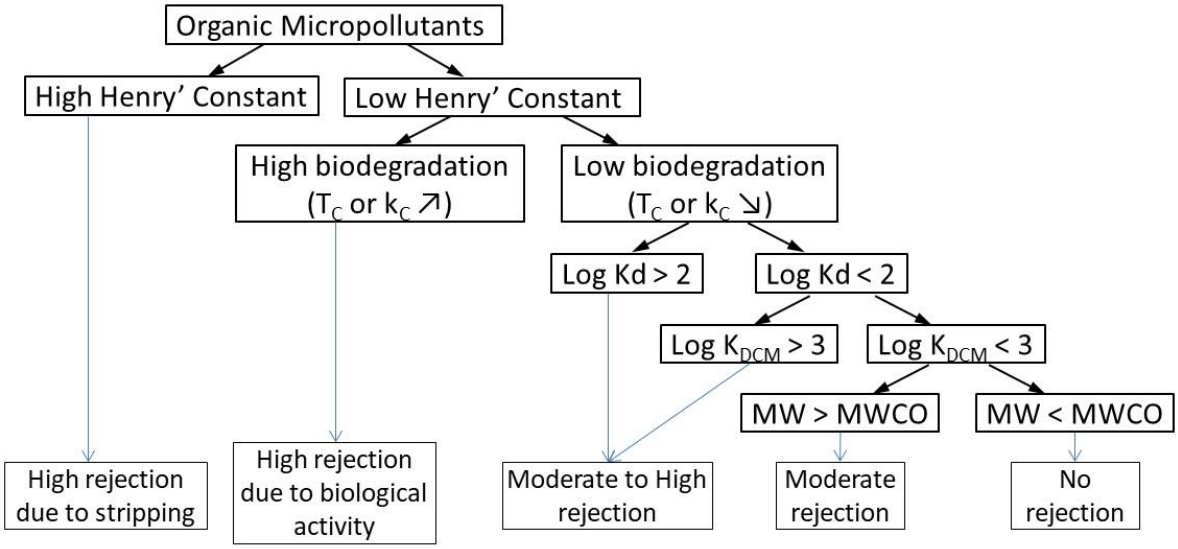

Fig 2. Removal diagram of OMP during anaerobic membrane bioreactor process[95]

500

501

502

503

504

505

506

507

508

509

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 Rejeciton due to Biological Acitivity and Stripping)
Anaerobic Membrane Bioreactor (AnMBR)	Ate, αE2, αEE2, atrazine, cbz, clozapine, DEET, dcf, Dilantin, E3, E1, gem, hydroxyzine, ibp keto, linuron, meprobamate, omeprazole, primidone	βE2, 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_2O, clarithromycin, josamycin, roxithromycin, tylosin, nalidixic acid, flumequine, pipemidic acid, norfloxacin, ciprofloxacin, ofloxacin, cephalexin, 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, amitriptytline, 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 system[40]. 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 mebrane 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[99]. 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[99]. 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 sustainabule 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 membrane[9]. 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 them[92]. 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 absoption 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[100]. Recently, there has been attempts to observe the
558 influence of photo-oxidation to the removal of OMPs from the effluent treated by AnMBR[89].
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 parameters[101].

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

Conclusions

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

Acknowledgment

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

References

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

Sci. Technol., 2019, **53**, 9572-9583.

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

anaerobic membrane bioreactor with ultrafiltration and nanofiltration, *J. Water Reuse Desalin.*, 2016, **6**, 362-370.

41. Y. Xiao, H. Yaohari, C. De Araujo, C. C. Sze and D. C. Stuckey, Removal of selected pharmaceuticals in an anaerobic membrane bioreactor (AnMBR) with/without powdered activated carbon (PAC), *Chem. Eng. J.*, 2017, **321**, 335-345.

42. A. P. Terzyk, Further insights into the role of carbon surface functionalities in the mechanism of phenol adsorption, *J. Colloid Interface Sci.*, 2003, **268**, 301-329.

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 pharmaceuticals and organic matter from municipal wastewater using two-stage anaerobic fluidized membrane bioreactor, *Bioresour. Technol.*, 2014, **165**, 42-49.

44. K.-C. Lee, B. E. Rittmann, J. Shi and D. McAvoy, Advanced steady-state model for the fate of hydrophobic and volatile compounds in activated sludge, *Water environment research*, 1998, **70**, 1118-1131.

45. B. E. Rittmann and J. A. Manem, Development and experimental evaluation of a steady-state, multispecies biofilm model, *Biotechnol. Bioeng.*, 1992, **39**, 914-922.

46. C. S. Criddle, The kinetics of cometabolism, *Biotechnol. Bioeng.*, 1993, **41**, 1048-1056.

47. L. Delgadillo-Mirquez, L. Lardon, J.-P. Steyer and D. Patureau, A new dynamic model for bioavailability and cometabolism of micropollutants during anaerobic digestion, *Water Res.*, 2011, **45**, 4511-4521.

48. E. Fernandez-Fontaina, M. Carballa, F. Omil and J. Lema, Modelling cometabolic biotransformation of organic micropollutants in nitrifying reactors, *Water Res.*, 2014, **65**, 371-383.

49. B. G. Plósz, C. Vogelsang, K. Macrae, H. H. Heiaas, A. Lopez, H. Liltved and K. H. Langford, The BIOZO process–a biofilm system combined with ozonation: occurrence of xenobiotic organic micro-pollutants in and removal of polycyclic aromatic hydrocarbons and nitrogen from landfill leachate, *Water Sci. Technol.*, 2010, **61**, 3188-3197.

50. A.-K. Ghattas, F. Fischer, A. Wick and T. A. Ternes, Anaerobic biodegradation of (emerging) organic contaminants in the aquatic environment, *Water Res.*, 2017, **116**, 268-295.

51. C. Grandclement, I. Seyssiecq, A. Piram, P. Wong-Wah-Chung, G. Vanot, N. Tiliacos, N. Roche and P. Doumenq, From the conventional biological wastewater treatment to hybrid processes, the evaluation of organic micropollutant removal: a review, *Water Res.*, 2017, **111**, 297-317.

52. H. Smidt and W. M. de Vos, Anaerobic microbial dehalogenation, *Annu. Rev. Microbiol.*, 2004, **58**, 43-73.

53. A. Spielmeyer, F. Stahl, M. S. Petri, W. Zerr, H. Brunn and G. Hamscher, Transformation of sulfonamides and tetracyclines during anaerobic fermentation of liquid manure, *J. Environ. Qual.*, 2017, **46**, 160-168.

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

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

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

of chemical synthesis-based pharmaceutical wastewater in an ozonation-anaerobic membrane bioreactor (AnMBR) system, *Chem. Eng. J.*, 2017, **322**, 293-301.

92. Y. Kim, S. Li, L. Chekli, Y. C. Woo, C.-H. Wei, S. Phuntsho, N. Ghaffour, T. Leiknes and H. K. Shon, Assessing the removal of organic micro-pollutants from anaerobic membrane bioreactor effluent by fertilizer-drawn forward osmosis, *J. Membr. Sci.*, 2017, **533**, 84-95.

93. X. Song, W. Luo, J. McDonald, S. J. Khan, F. I. Hai, W. E. Price and L. D. Nghiem, An anaerobic membrane bioreactor–membrane distillation hybrid system for energy recovery and water reuse: Removal performance of organic carbon, nutrients, and trace organic contaminants, *Sci. Total Environ.*, 2018, **628**, 358-365.

94. M. Aslam and J. Kim, Investigating membrane fouling associated with GAC fluidization on membrane with effluent from anaerobic fluidized bed bioreactor in domestic wastewater treatment, *Environmental Science and Pollution Research*, 2019, **26**, 1170-1180.

95. C. Bellona, J. E. Drewes, P. Xu and G. Amy, Factors affecting the rejection of organic solutes during NF/RO treatment—a literature review, *Water Res.*, 2004, **38**, 2795-2809.

96. O. A. Arikan, Degradation and metabolization of chlortetracycline during the anaerobic digestion of manure from medicated calves, *J. Hazard. Mater.*, 2008, **158**, 485-490.

97. M. Lim, R. Ahmad, J. Guo, F. Tibi, M. Kim and J. Kim, Removals of micropollutants in staged anaerobic fluidized bed membrane bioreactor for low-strength wastewater treatment, *rocess Saf. Environ. Prot.*, 2019, **127**, 162-170.

98. J. Cho, G. Amy and J. Pellegrino, Membrane filtration of natural organic matter: initial comparison of rejection and flux decline characteristics with ultrafiltration and nanofiltration membranes, *Water Res.*, 1999, **33**, 2517-2526.

99. R. V. Linares, V. Yangali-Quintanilla, Z. Li and G. Amy, Rejection of micropollutants by clean and fouled forward osmosis membrane, *Water Res.*, 2011, **45**, 6737-6744.

100. E. Yousif and R. Haddad, Photodegradation and photostabilization of polymers, especially polystyrene, *SpringerPlus*, 2013, **2**, 398.

101. M. Abargues, J. Giménez, J. Ferrer, A. Bouzas and A. Seco, Endocrine disrupter compounds removal in wastewater using microalgae: Degradation kinetics assessment G RA PHICAL AB STRACT.