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Review

Removal of Organic Micro-Pollutants by Conventional Membrane Bioreactors and High-Retention Membrane Bioreactors

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Abstract: The ubiquitous presence of organic micropollutants (OMPs) in the environment as a result of continuous discharge from wastewater treatment plants (WWTPs) into water matrices—even at trace concentrations (ng/L)—is of great concern, both in the public and environmental health domains. This fact essentially warrants developing and implementing energy-efficient, economical, sustainable and easy to handle technologies to meet stringent legislative requirements. Membrane-based processes—both stand-alone or integration of membrane processes—are an attractive option for the removal of OMPs because of their high reliability compared with conventional process, least chemical consumption and smaller footprint. This review summarizes recent research (mainly 2015–present) on the application of conventional aerobic and anaerobic membrane bioreactors used for the removal of organic micropollutants (OMP) from wastewater. Integration and hybridization of membrane processes with other physicochemical processes are becoming promising options for OMP removal. Recent studies on high retention membrane bioreactors (HRMBRs) such as osmotic membrane bioreactor (OMBRs) and membrane distillation bioreactors (MDBRs) are discussed. Future prospects of membrane bioreactors (MBRs) and HRMBRs for improving OMP removal from wastewater are also proposed.

Keywords: organic micropollutants; membrane bioreactor; forward osmosis (FO); membrane distillation (MD); wastewater

1. Introduction

Rapid population growth, combined with increased agricultural and industrial undertakings is resulting in increased water demand and sewage production [1]. Thus, these persistent drivers of water stress are prompting interest in advanced wastewater treatment techniques that utilize alternative water sources, such as domestic wastewater for water reclamation [2,3]. However, with growing interest in reclaimed water use, safety warrants for health risks—especially when the diverse nature of organic micropollutants found in reclaimed water—must be taken into account [4]. However, the ubiquitous presence of OMPs in reclaimed water and sewage is a significant hurdle to water reuse [5,6].

Over the past few years, the omnipresent occurrence of trace organic contaminants (TrOCs) organic micropollutants (OMPs), emerging contaminants (ECs), emerging substances of concern (ESOC), has been identified, due to their stability in the environment. Products such as pharmaceuticals and personal

care products (PPCPs), veterinary medicines, endocrine-disrupting chemicals (EDCs), x-ray contrasting agents, surfactants, industrial additives and formulations, agricultural pesticides, food additives, disinfection by-products, hormones and steroids, flame retardants, metabolic regulators, preservatives, perfluorinated compounds and nanomaterials are safety concerns in reclaimed water [4,7–11]. The term "emerging" is used not only to describe new, recently discovered, developed and consumed compounds, but it is also applied to substances already present in the environment, though they may have been only recently recognized as contaminants. Furthermore, prescribed discharge guidelines and statutory requirements regarding these compounds have not yet been established [9,12]. Organic micropollutants pose huge environmental threats due to possible risks associated with mutagenicity, carcinogenicity, teratogenicity and high bioaccumulation [1].

1.1. Occurrence, Fate and Transport of OMPs in WWTPs and Impact on Human and Environment

Recent reports suggest the presence of numerous OMPs in increasing concentrations in polluted water environment (raw wastewater, surface and groundwater and drinking water), and that OMPs have become a global issue of great importance for environmental protection strategies [7,13]. More recently, OMPs have generated increased concern among health authorities, industries and agricultural product manufacturers due to the associated risks to health of people and damage to the environment [8–10]. OMPs have been widely assessed in an environment (sediment, soils, atmosphere, sewage, surface, ground and drinking waters) and received increasing attention in recent years [7,10,14]. OMPs originate from either human activity, such as process effluents, discharges of treated effluents from sewage and hospital wastewater, agricultural runoff, septic tank or natural activities. Other anthropogenic sources include landfills, inappropriately disposed wastes, surface runoff, sewer overflow and leaking sewers [8,9,15] (Figure 1). Although OMPs are present in the environment at very low concentrations—only ranging from a few nanograms per liter (ng/L) to micrograms per liter (µg/L)—they may pose risks to humans and other living organisms [6,9].

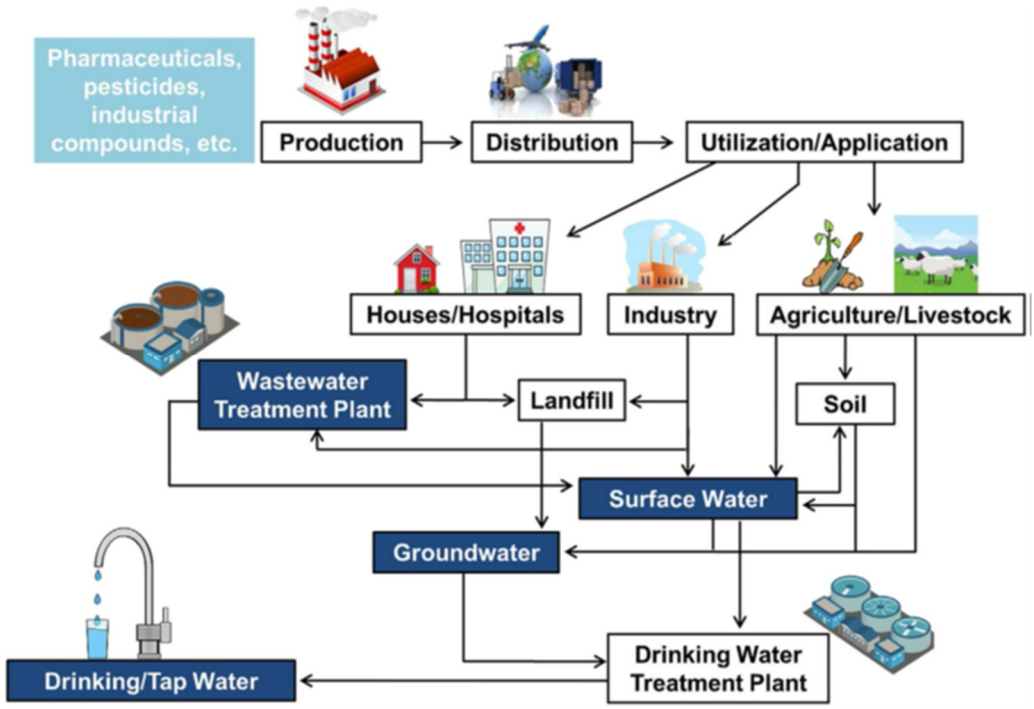

Figure 1. Representative sources and routes of micropollutants in the environment [16]. Reproduced with permission from [16], Copyright Water Research, 2016.

It has been reported that at 6.5 mg/L, the antibiotic ciprofloxacin had highest concentrations among more than 200 various pharmaceuticals detected in river waters all over the world. Single-compound

acute toxicity testing has found median sufficient levels (EC50s) for many OMPs to be < 1 mg/L [17]. For example, during spring and winter when infectious diseases spread rapidly, antibiotic consumption and discharge into aquatic environments increases. Those antibiotics accumulate in activated sludge; during summer, a peak concentration of around 9481.43 ng/g for the fluoroquinolone ofloxacin has been monitored in water bodies [18].

Nevertheless, accidental release of OMPs into the environment adversely affects several organisms such as flora and fauna, as well as human health [19,20]. Such endocrine-disrupting compounds (EDC) include endogenic hormones, mycoestrogenes—the organic compounds produced by fungi, micropollutants polycyclic aromatic hydrocarbons (PAHs), surfactants, pesticides, halo-organic compounds including dioxins, furans [7] and steroid estrogens (SEs), such as estrone (E1), estradiol (E2) and ethinylestradiol (EE2) [21]. A significant adverse effect on fish populations (*Pimephales promelas*) has been observed after exposure to 17-α-Ethinyl estradiol (EE2) at a concentration of 5 ng/L in 7 years, due to estrogenic activity that affects hormones in animals even at very low 0.1 ng/L concentration [9,22].

Findings suggest that some of industrial chemicals—nano/microplastics [23] and pesticides—pose some environmental and health concern. For instance, bisphenol A (BPA) is extensively used as a plastic additive and may pose health threats by entering the human body via different routes. There is increasing evidence that Bisphenol A (BPA) adversely affects reproduction and development systems, neural networks and cardiovascular, metabolic and immune systems [24]. Atrazine—one of the possible class-C carcinogens as detected by the United States-Environmental Protection Agency (US-EPA)—has also been found responsible for cancer in rats when exposed to high doses for extended periods [25].

The major point source for discharge of OMPs is from WWTPs. This can be attributed to the continuous presence of OMPs in the water bodies specifically close by urban dwellings [14,26,27]. More recently, OMPs have been identified in sewage and whole water bodies in North America, Europe, Asia and Africa [8]. Einsiedl et al. (2010) evaluated the fate and transport of OMPs in groundwater. They report that certain pharmaceuticals contaminated karst groundwater due to continual sewage discharge [28]. Also, in some advanced countries such as Germany, UK, Italy, Canada and the USA, OMPs (pharmaceuticals) were detected in their potable water samples [11]. To meet stringent discharge limits for treated effluent—and in order to produce reclaimed water—it is essential to design an efficient wastewater treatment technology [29]. Activated sludge processes are capable enough to remove certain OMPs, though many OMPs have shown non-biodegradable characters not removed by conventional processes [26].

1.2. Mitigation and Litigation of OMPs

For many OMPs, statutory limits are not clear. This does not mean that OMPs' presence in potable or ground water is safe. Unfortunately, the toxic effects of many OMPs have not been fully evaluated [11]. Both the US and European Union (EU) have to set statutory consented discharge limit for the release of OMPs into aquatic environments [8]. For example, twelve OMPs have been closely examined as emerging compounds by the European Union under the 2015's Water Framework Directive (WFD) [26]. Endocrine-disrupting compounds are already controlled and banned by European, North American and East Asian countries [30].

As per the World Health Organization (WHO), very stringent regulations are set up for drinking water standards for certain phenolic and PAH compounds, pesticide and herbicides. These force water suppliers to eliminate such OMPs from water to take care of health risk and secure environment [1]. In the United States, 11 disinfection by-products (DBPs) are regulated [10]. The maximum contaminant level of EPA for atrazine in water is 3 g/L [31,32]. Furthermore, pentachlorophenol (PCP) falls under the category of probable carcinogens, and hence its maximum contamination level of 1 mg/L has been kept by US EPA [33]. Triclosan is a raw material for toxic biocidal products, and is therefore banned in Europe; the US government has kept it under review [34]. However, World Health Organization (WHO) updated persistent OMPs into its guidelines for lack of representative statistical data for potable

water quality (WHO, 2011) [8]. Looking at the alarming threat posed by emerging micropollutant to human health, more stringent discharge standards are required for both sewage and industrial effluents in the future [35].

1.3. Membrane Bioreactors in Organic Micropollutants Removal

Conventional wastewater treatments targeting OMP removal face challenges with possible human health risk and risk to the environment [36,37]. The most widely utilized physical and chemical treatment processes are very financially demanding [38]. Current WWTPs are not designed to eliminate or degrade OMPs completely, many of these OMPs can pass through the treatment system and enter into the natural aquatic system because of their persistence [39,40]. The presence of refractory OMPs and their biodegradation by-products adversely affects biodegradation potential of bacteria present in activated sludge [34]. In addition, OMPs concentrations are uncertain in sewage and conventional wastewater treatment facilities are unable to efficiently remove OMPs to the extent for reuse applications [2]. For example, commonly detected OMPs such as ibuprofen showed biodegradable removal of 75% whereas Estrone (E1) and 17-α-Ethinyl estradiol (EE2) achieved 83% and 44% removal [41]. Furthermore, advanced OMP removal or destruction options include adsorption or ion exchange using activated carbon and ion exchange resins, ultraviolet (UV) disinfection and advanced oxidation processes (AOP) such as hydrogen peroxide oxidation, electrochemical advanced oxidation processes (EAOPs) such as Anodic oxidation (AO) and electro-Fenton (EF) [42] and photocatalytic degradation. However these processes involve high capital and energy costs and also require disposal of highly contaminated exhausted sorbent or problematic residues [33]. Further, membrane-based treatment processes such as membrane bioreactor, reverse osmosis, nanofiltration, forward osmosis and membrane distillation are promising alternative for OMP removal.

Microfiltration (MF)/ultrafiltration (UF) MBRs are techno-economically feasible and a most promising option in wastewater treatment. In MBRs, flatsheet or hollowfiber (HF) membranes are immersed into a bioreactor to achieve excellent and consistent micropollutant removal, compared to conventional activated sludge systems [3,14,18,43–45]. The worldwide suppliers and key players of MBRs include SUEZ Water Technologies & Solutions (formerly GE Water & Process Technologies) (France), Kubota (Japan), Beijing Origin Water Technology (China), Evoqua Water Technologies (US), Mitsubishi Chemical Aqua Solutions (Japan), Toray Industries (Japan), CITIC Envirotech Ltd. (Singapore), Koch Membrane Systems (US), Alfa Laval (Sweden), Triqua International (Netherlands), Veolia (France) and Newterra Canada) (https://www.Marketsandmarkets.com/Market-Reports/membrane-bioreactor-market-484.html). In 2017, SUEZ Water Technologies & Solutions acquired GE Water & Process Technologies, which strengthened the company's position in the water treatment field [46]. Kubota (Japan) was one of the early pioneers of the MBR concept and as of 2017 Kubota has supplied 5500 MBR systems of these 1500 MBR's are used in industrial wastewater treatment. Suez technology (formerly Zenon) produced four times better performance than Kubota in sewage treatment [47,48].

Several literature reviews report the removal efficacies of MBR systems in both sewage and industrial waste treatment [49–52]. However, this review aims at summarizing the recent advances, principally from 2015 to present, in standalone membrane biologic systems of MBRs and anaerobic MBRs (AnMBRs). This review further discusses factors affecting OMP removal by MBRs such as physicochemical properties of OMPs and operating parameters affecting MBR performance in OMP removal, referring to the most recent reports. Recently, high-retention membrane bioreactors (HRMBR) systems have been gaining momentum in wastewater treatment. This review also examines recent developments in forward-osmosis MBR (FO-MBR) and membrane distillation bioreactor (MDBR) for OMP removal. Finally, future perspectives for OMP treatment employing MBRs and HRMBRs were evaluated.

2. MBR Types and Configuration

Both academia and industries research and development (R & D) efforts are focused on aerobic and anaerobic membrane bioreactor (AnMBR), enzymatic membrane bioreactor and baffled membrane bioreactor [53]. In aerobic MBRs oxygen from supplied air acts as an essential medium for the bacterial growth while anaerobic is done without oxygen (no external air supplied). This leads to different bacteria strain in aerobic and anaerobic processes. Anaerobic process can easily be optimized for wastewater having high organic loading. Yet, maintaining low temperature for huge feed volume in mesophilic range poses a challenge. Moreover, anaerobic processes are not as efficient for high chemical oxygen demand (COD) removal as well as they exhibit more fouling potential than aerobic MBRs [54]. In wastewater treatment organic and nutrient removal are essential. For total nitrogen (TN) removal, denitrification was performed. To achieve TN removal denitrification requires anoxic conditions so anoxic tank is placed before or after aerobic tank. In order to achieve enhanced biologic phosphorus removal, anaerobic tank is incorporated in treatment train [55].

The two mains basic MBR configurations involve either submerged membranes or external circulation (side-stream configuration) (Figure 2). Submerged MBR configuration operates under subatmospheric pressure instead of hydraulic pressure. In this arrangement, membrane is placed inside the bioreactor and it is known as submerged MBR. In this design, pure water is obtained through MF or ultrafiltration UF membranes from mixed liquor and process is operated under low hydraulic pressure. The second configuration is also known as side stream or external cross flow MBR in which hydraulic pressure is applied. The membrane unit is isolated from bioreactor and an additional recirculation pump is employed to circulate bioreactor mixed liquor forced through membrane and pure permeate is obtained [52]. The side stream configuration deliberately separates bioreactor from external membrane thus reducing membrane maintenance. However, operating cost increases due to mixed liquor recirculation pump installation [54]. For this submerged configuration, flat sheet (FS) and hollow fiber (HF) membranes are ideal choices [52,56]. The submerged MBR process has less operating expenditure (OPEXm) due to elimination of mixed liquor recirculation pump than the side stream MBR system and this makes submerged membrane bioreactor (SMBR) as attractive option in wastewater treatment [54,55]. The membrane fouling involves deposition of impurities such as sludge flocs, colloidal particles and inorganic solutes, into membrane pores and onto the membrane surface and forms cake layer [57]. Membrane fouling simply incurs additional operational and repair costs due to rapid pressure drop, increased cleaning cycles and consumption of chemicals and it deteriorates permeate quality and reduces quantity (flux) [58]. Due to membrane fouling permeate flux decreases with time and fouling leads to frequent cleaning which incurs operating cost and process downtime [57].

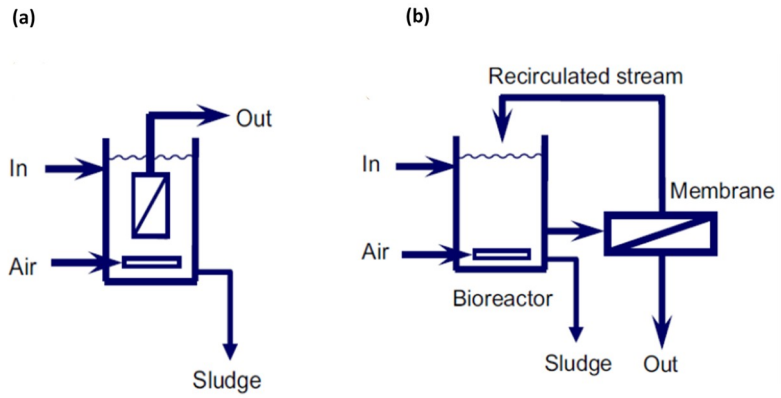

Figure 2. Configurations of a membrane bioreactor: (**a**) immersed and (**b**) sidestream [59]. Reproduced with permission from [59], Copyright Journal of Membrane Science, 2011.

2.1. Aerobic Membrane Bioreactors in OMPs Removal

Membrane bioreactors (MBRs) are nowadays very popular in sewage treatment and among industries for water reclamation due to their best permeate quality, less space requirement and reduced sludge management cost than activated sludge processes [60,61]. MBR is capable to reject bacteria and suspended solids, produce high purity permeate and flexible enough to operate with inflow variations. In comparison to activated sludge process MBR produces permeate with very low organics and accomplish reduction in OMPs at great extent [34,62,63]. The mechanisms for eliminating OMPs by MBR are complex and include volatilization, size exclusion, electrostatic repulsion or adsorption [39,64]. Further, OMP removal depends on physicochemical peculiarities of OMPs, membrane characteristics such as pore diameter, molecular weight cut-off (MWCO) and zeta potential, membrane-solute interactions and feed properties [64]. The microfiltration/ultrafiltration membrane of an MBR can retain all suspended solids leading to higher mixed liquor suspended solids (MLSS) concentration, long sludge retention time (SRT). This system also provides the opportunity to develop different bacterial consortia thereby producing pure permeates while efficiently accomplishing the removal of moderately biodegradable OMPs [14,27]. MF/UF-MBR are also more capable of rejecting OMPs and viruses than activated sludge processes [65,66].

Mutamim et al. (2013) have suggested that in MBRs to obtain pure water quality and to alleviate membrane fouling several operating parameters need to be optimized such as hydraulic retention time (HRT), solid retention time (SRT), mix liquor suspended solid (MLSS), food to microorganism (F/M) ratio, transmembrane pressure (TMP) and flux (J). Furthermore, fouling factors including membrane synthesis and morphology (types, orientation and physical properties), biomass characteristics and MBR operation (HRT, SRT, etc.) need to be taken into consideration because they are the major factors that affect MBR process [54].

In Table 1, a summary of reports for OMP removal employing MBRs is listed. It is clear that MBRs can more efficiently remove OMPs than conventional biologic treatment. MBRs achieved around 100% removal of PPCPs such as salicylic acid and propylparaben. Further, MBRs also can remove OMPs like beta blockers at 70–80% and atenolol can be removed by up to 97% [67]. The authors report that membrane bioreactor-reverse osmosis (MBR–RO) successfully removed >99% of azithromycin, clarithromycin, erythromycin, ofloxacin, sulfamethoxazole, diazepam, lorazepam, famotidine, ranitidine and clopidogrel [67]. In another study, Luo et al. (2015a) observed that moving bed bioreactor-membrane bioreactor (MBBR–MBR) combined process was effective in OMP removal. However, low removal efficiency is reported for ketoprofen (16.2%), carbamazepine (30.1%), primidone (31.9%), bisphenol A (34.5%) and estriol (39.9%). Nonetheless, the same study found that hybrid MBBR–MBR system could effectively remove most of the selected micropollutants [68].

Table 1. Organic micropollutants (OMPs) removal for aerobic membrane bioreactors (MBRs).

Micropollutants	% Removal											
	A-[5]	B-[68]	C-[68]	D-[69]	E-[70]		F-[71]	G-[71]	H-[72]		I-[73]	J-[74]
					MF	UF			SRT (∞)	SRT (27d)		
17α-ethynylestradiol	96	56	71	-	-	-	-	-	-	-	-	100
17β-estradiol	98	93	94	-	-	-	-	-	100	100	-	100
4-n-nonylphenol	-	90	90	-	-	-	-	-	-	-	-	
4-p-nonylphenol	-	-	-	-	-	-	-	-	-	-	-	100
4-t-nonylphenol	-	-	-	-	-	-	-	-	-	-	-	100
4-tert-Butylphenol	98	55	62	-	-	-	-	-	-	-	-	
4-tert-Octylphenol	97	82	84	-	-	-	-		-	-	-	100

Table 1. *Cont.*

Micropollutants	% Removal											
	A-[5]	B-[68]	C-[68]	D-[69]	E-[70]		F-[71]	G-[71]	H-[72]		I-[73]	J-[74]
					MF	UF			SRT (∞)	SRT (27d)		
Acetaminophen	-	90	90	95	-	-	-	-	-	-	-	-
Amitriptyline	95	-	-	-	-	-	34	-	-	-	-	-
Androsterone	-	-	-	-	-	-	98	98	-	-	-	-
Atenolol	-	-	-	59	-	-	92	85	-	-	-	-
Atrazine	30	-	-	-	-	-	-	-	-	-	-	-
Bezafibrate	-	-	-	93	-	-	-	-	-	-	-	-
Benzophenone	97	-	-	-	-	-	-	-	-	-	-	-
Bisphenol A	96	39.9	80	-	-	-	-	-	-	-	-	-
Caffeine	-	-	-	96	-	-	94	91	-	-	-	-
Carbamazepine	70	16.2	21	-	94	92	2	1	−94.5	−6.8	-	-
Ciprofloxacin	-	-	-	87	-	-	-	-	-	-	-	-
Codeine	-	-	-	-	-	-	-	-	-	-	71.9	-
Cyclophosphamide	-	-	-	-	-	-	-	-	-	-	59.5	-
DEET *	90	-	-	-	-	-	97	84	-	-	-	-
Diazepamdzp	-	-	2	-	-	-	-	-	-	-	-	-
Diclofenac	70	42	43	36	80	90	57	15	−50.1	−270.2	-	-
Diltiazem	-	-	-	58	-	-	-	-	-	-	-	-
Diuron	-	-	-	-	-	-	96	25	-	-	-	-
Enterolactone	91	-	-	-	-	-	-	-	-	-	-	-
Erythromycin	-	-	98	-	98	100	-	-	-	-	-	-
Estradiol	-	-	-	-	99	99	-	-	-	-	-	-
Estriol	91	34.5	90	-	-	-	-	-	-	-	-	-
Estrone	99	80	100	-	-	-	98	96	-	-	-	100
Ethinylestradiol	-	-	-	-	92	93	-	-	-	-	-	-
Etiocholanolone	-	-	-	-	-	-	98	98	-	-	-	-
Fenoprop	60	25	26	-	-	-	-	-	-	-	-	-
Fluoxetine	-	-	92	-	-	-	-	-	-	-	-	-
Gemfibrozil	97	80	72	-	-	-	89	83	45.8	−84.6	-	-
Ibuprofen	99	90	98		91	92	97	95	100	100	-	-
Ifosfamide	-	-	-	-	-	-	-	-	-	-	49.3	
Ketoprofen	96	30.1	72	87	-	-	-	-	-	-	-	-
Levofloxacin	-	-	-	82	-	-	-	-	-	-	-	-
Mefenamic acid	-	-	-	60	-	-	-	-	-	-	-	-
Metronidazole	97	18	35		-	-	-	-	-	-	-	-
Naproxen	97	70	80	97	89	98	95	85	82.3	23.6	-	-
Octocrylene	80	-	-	-	-	-	-	-	-	-	-	-
Paracetamol	-	-	-	-	-	-	98	97	-	-	-	-

Table 1. *Cont.*

Micropollutants	% Removal											
	A-[5]	B-[68]	C-[68]	D-[69]	E-[70]		F-[71]	G-[71]	H-[72]		I-[73]	J-[74]
					MF	UF			SRT (∞)	SRT (27d)		
Pentachlorophenol	91	80	80	-	-	-	-	-	-	-	-	-
Polyparaben	-	-	-	-	-	-	97	97	-	-	-	-
Primidone	58	31.9	69	-	-	-		13	-	-	-	-
Roxithromycin	-	-	96	51	98	100	-	-	-	-	-	-
Salicylic acid	97	88	92	-	-	-	-	-	-	-	-	-
Sulfamethoxazole	-	-	99	-	80	70	85	56	78.5	−43.9	75	
Triclocarban	-	-	-	95	-	-	51	94	-	-	-	
Triclosan	98	92	97	94	-	-	62	60	100	100	-	
Trimethoprim	-	-	99	-	97	91	64	55	80.1	24.6	-	
β-Estradiol-17-acetate	97	92	94	-	-	-	-	-	-	-	-	

	Type of Influent	Membrane with Operating Conditions
A-[5]	Synthetic wastewater (MBR)	Lab-scale MBR, Hollow fiber polyvinylidene difluoride (PVDF) MF membrane with a pore size of 0.4 µm and area 0.074 m^2, MLSS: 5 g/L, dissolved oxygen (DO): 5 mg/L, SRT: 20 d, HRT: 27 h.
B-[68]	Synthetic wastewater (MBR)	PVDF hollow fiber microfiltration (MF) membrane modules pore size of 0.2 µm and surface area of 0.2 m^2, HRT: 6 h, SRT: infinite, MLSS: 2.27–7.38 g/L
C-[68]	Synthetic wastewater (MBBR + MBR)	PVDF hollow fiber MF membrane modules pore size of 0.2 µm and surface area of 0.2 m^2 HRT: 24 h; SRT: infinite; MLSS: 2.27–7.38 g/L Polyurethane sponge cubes (S28/80R, Joyce Foam Products; dimension of 2 cm × 2 cm × 2 cm as biofilm carriers.
D-[69]	Real WWTP South Korea Anoxic + aerobic	Submerged hollow fiber MF PVDF membrane, pore size of 0.4 µm, total surface area 0.04 m^2, cycles of 7 min on and 1 min of relaxation. HRT: 11 h, Temperature: 25 °C, pH: 6.8, MLSS: 7–11 g/L.
E-[70]	Synthetic wastewater (MBR + PAC)	A flat sheet membrane (Kubota, pore size 0.45 mm) MF PVDF membrane, the pore size of 0.4 µm and ultrafiltration (UF) hollow fiber membrane (Zenon ZW-20, pore size 0.045 mm), cycles of 7.5 min on and relaxation time of 1.25 min for MF, while 7 min on and backwashing of 0.5 min for UF, HRT: 24 h, Temperature 20–22 °C. pH: 7.5, MLSS: 3 g/L.
F-[71]	Real wastewater (Full-scale plant MBR)	PVDF flat sheet MF membranes, total surface area 4800 m^2, HRT: 1.5–1.7 d, SRT: 25 d, T: 20–22 °C, pH: 7.5, MLSS: 9 g/L.
G-[71]	Real wastewater (Pilot-scale plant) (Anoxic-Aerobic MBR)	Hollow fiber UF membrane (Zeweed-10, pore size 0.04 µm, total surface area 0.93 m^2, HRT: 1.5 d, SRT: 25 d, T: 20–22 °C, pH: 7.1–7.4, MLSS: 2.4 g/L, aerobic DO: 2.5–5 mg/L, p H 7.14, anoxic DO: 0.25 mg/L pH: 7.43, T: 18 °C

Table 1. *Cont.*

Micropollutants	% Removal										
	A-[5]	B-[68]	C-[68]	D-[69]	E-[70] MF	E-[70] UF	F-[71]	G-[71]	H-[72] SRT (∞)	H-[72] SRT (27d)	I-[73]	J-[74]
H-[72]	Real WWTP Pilot-scale MBR set-up Bangkok, Thailand	PVDF Hollow fiber membrane, pore size 0.4 µm, total surface area 36 m2, 7 min on & 1 min off, HRT 3 h, SRT 27 d, MLSS 13 g/L, DO 1–4 mg/L, pH 6.8.										
I-[73]	Pilot scale MBR Hospital effluent (Marseille, France)	Hollow fiber Polysulfone membrane, 100 kDa MWCO, total surface area 0. 0.4 m2, HRT 16–40 h, SRT ∞ d, temperature 25 °C. MLSS 6.8 g/L, DO 2 mg/L, temperature 25 °C.										
J-[74]	MBR pilot plant Real wastewater	Hollow-fiber UF membrane module (Zenon, Zee-Weed® 500 modules), surface area 46.5 m^2, HRT 9 h, SRT 100 d, MLSS 15 g/L.										

N,N-diethyl-m-toluamide-(DEET) *.

OMP degradation and transfer into aquatic environment depends on the electron donating or withdrawing groups as well as hydrophobicity of OMPs [75]. The major physicochemical properties of OMPs include volatility, solubility, molecular weight, hydrophobicity (Kow), sludge adsorption and biodegradation, electron-withdrawing group (EWG) and electron-donating groups (EDG) [47]. Luo et al. (2015a) evaluated the removal of 30 OMPs and report that more than 85% removal was observed for hydrophobic OMPs. Hydrophobic OMPs can easily adsorb on the sludge particles that increases its retention time in the reactor leads to the better biodegradation [68]. Luo et al. (2015b) report that the large pore size of a microfiltration membrane was probably responsible for poor removal of hydrophilic in this study [76]. In another report, Prasertkulsak et al. (2016) operated pilot scale MBR at very short HRT of 3 h and with very short start-up time. The experimental results revealed the importance of immediate adsorption of the recalcitrant pharmaceutical compounds onto the colloidal particles in supernatant of MBR sludge and subsequently removed by membrane filtration [72]. The removal of diclofenac, sulfamethoxazole, carbamazepine, gemfibrozil was mostly found to be negative during MBR operation. For example, influent and effluent concentration for diclofenac have been reported as 3.81 mg/L and 5.72 mg/L, showing negative removal of −50.1%. Alvarino et al. (2014) report that sulfamethoxazole could be highly eliminated under oxygen free condition due to the presence of electron-withdrawing like sulfonyl group so their biodegradation under aerobic condition would be limited [77]. Carbamazepine, a moderate hydrophobic compound, was found to accumulate in supernatant (adsorption onto colloidal particles) and they were also partially detected in the membrane filtrate resulting in obtaining negative removals during MBR operation [72]. In another study, OMP removal was examined with and without powdered activated carbon (PAC) addition and employing two different membranes—flat sheet and hollowfiber, respectively. Trimethoprim, Carbamazepine and Diazepam achieved good removal by addition of PAC that could be related to the log D of the compound. However, PAC saturation and exhaust capacity is dependent on the ionic charge of the micropollutants [70].

Park et al. (2017) observed that triclocarban, ciprofloxacin, levofloxacin and tetracycline showed more affinity towards sludge particles in the bioreactor. However, authors also report that higher biodegradation was also governing mechanism. Moreover, higher removal efficiencies of beta-blockers

such as atenolol (58%), propranolol (50%) and diltiazem (57%) have been reported with lab-scale MBR than anaerobic/anoxic/oxic process which demonstrated lower removal for atenolol (43%), propranolol (17%) and diltiazem (35%), respectively [69]. It was deduced that biosorption on sludge surface was potential removal mechanism for beta-blockers [69]. In an MBR study, Prasertkulsak et al. (2016) report that gemfibrozil and carbamazepine were refractory molecules and highly persistent to biotransformation [72]. In another study, Hamon et al. (2018) examined the removal of three most consumed anticancer drugs namely ifosfamide, fluorouracile and cyclophosphamide and a pain killer codeine and antibiotic sulfamethoxazole were studied in submerged MBR. Biodegradation was successful removal mechanism for sulfamethoxazole and codeine achieving 79% and 95% removal efficiencies, respectively while ifosfamide and cyclophosphamide showed moderate elimination of less than 40%. However, for all selected pharmaceuticals of this study more than 89% removal were achieved due to intense membrane fouling. Needless to mention that membrane fouling led to high bio sorption of OMPs thereby prolonged their retention in bioreactor [73]. Arola et al. (2017) studied pilot-scale MBR and membrane bioreactor-nanofiltration (MBR-NF) system using real sewage. MBR-NF system with nanofiltration (NF270) membrane outperformed MBR achieving 84% of removal for OMPs except caffeine and hydrochlorothiazide. The MBR process found incapable with persistent OMPs such as carbamazepine and diclofenac compounds [78]. Alvarino et al. (2017) also report (Table 1) that biodegradation was the governing mechanism for naproxen, ibuprofen and hormones removal in MBRs with PAC addition [70]. Albeit, partial biodegradation and partial sorption onto powdered activated carbon surface were the removal mechanism observed for OMPs such as erythromycin and roxithromycin. Actually, membrane type influenced the removal of diclofenac and roxithromycin. It was noted that biosorption/biodegradation occurred in the cake layer of the membrane hence OMP removal. Park et al. (2017) demonstrated that biodegradation played a significant role for compounds adsorbed to the sludge as well as for recalcitrant OMPs. Author report that bezafibrate, ketoprofen and atenolol showed better removal [69]. In another study Sahar et al. (2011) report that in an MF-MBR process salicylic acid, metronidazole, ketoprofen, naproxen, primidone and ibuprofen six model OMPs showed more than 85% removal which was higher than conventional MBRs. The authors correlated this higher hydrophilic OMP removal with $–NH_2$- and –OH-like EDGs in their structure. Further, they report that strong EDGs were easy to attack by aerobic consortia [79]. In another hybrid MBR-MBBR study high removal efficiencies more than 80% for OMPs like nonylphenol, 17α-ethynylestradiol, 17β-estradiol, estrone, bisphenol A and triclosan were noted. It was further deduced that with those hydrophobic OMPs better removal were achieved than hydrophylic OMPs, such as diclofenac, ibuprofen and sulfamethoxazole. It is well established that hydrophobic OMPs possess natural tendency to adsorb to the sludge led to high removal. However, by pH adjustment (pH < pKa) and thus more acidic environment may have enhanced removal of hydrophilic compounds such as sulfamethoxazole in the same study [53,68].

High MLSS concentrations in bioreactor can provide more surface area and can favor biodegradation by enhancing retention time for OMPs as reported for pharmaceutical compound [72]. As reported by Sahar et al. (2011), removal efficiencies of 85% for roxithromycin and 92% for clarithromycin were obtained in MBR process due to higher biomass concentration of 10 g/L [79]. When operated in activated sludge process with typical 2.3–2.5 g/L biomass concentration, lower removals of 65% for roxithromycin and 78% for clarithromycin are reported [69]. Prasertkulsak et al. (2016) noticed that pharmaceutical degrading microorganisms were developed and led to remarkable biotransformation for OMPs in hospital wastewater even at very low HRT of 3 h in pilot-scale MBR [72]. Abargues et al. (2012) compared performance of full- and pilot-scale-membrane bioreactors (MBRs) in OMP removal. The authors report that better removal was accomplished for sulfamethoxazole, trimethoprim, diclofenac, diuron and amitriptyline in full-scale MBR. Both MBRs demonstrated excellent OMP removal in final permeate, meeting Australian Guidelines for Water Recycling, except caffeine, estrone and triclosan compounds [74]. Furthermore, alternative anaoxic and oxic redox conditions established link between removal of eight OMPs and total nitrogen removal [71]. Other

studies report improved removal of diclofenac, ethinylestradiol, triclosan and ibuprofen with varying redox conditions [80]. MBR–RO is a promising alternative for producing high quality permeate for water recycling. Report suggested that when treating 31 OMPs reuse quality water was obtained though organics from soluble microbial products (SMP) and extracellular polymeric substances (EPS) and other salts were responsible for membrane fouling adversely affected process performance [5].

2.2. Anaerobic MBR (AnMBR) for OMPs Removal

Another arrangement known as anaerobic MBR (AnMBR), has become an attractive option for energy-neutral wastewater treatment. AnMBR is a hybrid process that integrates anaerobic process and membrane-based separation. AnMBR process converts organics present in sewage into biogas (methane) by biologic transformation employing basic sequences, such as hydrolysis, acidogenesis, acetogenesis and methanogenesis [81]. AnMBR has gained attention as more energy-efficient and effective process in contrast to aerobic MBR. Aerobic MBR utilizes huge energy to maintain dissolved oxygen and for membrane scouring. AnMBR could become energy saver by methane generation or even be a positive energy system by producing biogas for beneficial usage [82,83]. The anaerobic process offers more challenges in terms of process disturbance because of retarding compounds, such as heavy metals, chlorinated hydrocarbons and cyanides frequently found in raw sewage [82]. The anaerobic process is less popular in sewage treatment due to fact that it rarely meets the required discharge standards specifically at low temperature. Also, it is too difficult to maintain sufficient slow growing MLSS with dilute wastewater and low HRT conditions [84]. Mitigation of membrane fouling and efficient dissolved methane recovery should be the key area for future developments in AnMBR technology [85].

Table 2 describes the fundamental difference between AnMBR and MBR. Anaerobic digestion was employed as an attractive option for excess sludge stabilization from conventional biologic process. The fate of OMPs present in settled sludge depends on characteristics and availability of EWGs and EDGs in the OMP when stabilized sludge is targeted for soil conditioning or in farm. The process governed by bacterial community consumes organics and OMPs in sludge as a food source and converts into carbon dioxide [67]. In order to remove OMPs, combined redox conditions can be applied as demonstrated by Alvarino et al. (2016), when operated anaerobic and aerobic reactors in tandem [86]. Authors report that aerobic conditions are favorable for most of the OMP removal. Certain OMPs such as trimethoprim a pharmaceutical compound essentially needed anaerobic treatment [80]. Some recent AnMBR studies for OMP removal are summarized in Table 3.

Table 2. Comparison between anaerobic membrane bioreactors (AnMBRs) and MBRs for wastewater treatment (Adapted from [81]). Copyright Bioresource Technology, 2018.

Feature	AnMBR	MBR
Energy consumption (kWh/m^3)	0.03–5.7 [a]	~2 [b]
Biomass concentration (g/L) [c]	10–40	5–20
Organic loading rate (kg COD/L/d)	0.17–35.5	0.25–0.8
Organic removal efficiency (%)	>90	>95
Hydraulic retention time (hours)	>8	4–8
Water flux, liters per square meter per hour (LMH)	5–12	20–30
Sludge retention time (d)	>100	5–20
Operational temperature (°C)	20–50	20–30

[a] Energy consumption was calculated for submerged AnMBR treating wastewater with strength between 0.27 and 10 g COD/L; [b] Energy consumption was calculated for submerged MBR-treating wastewater with strength between 0.3 and 1.0 g COD/L; [c] Biomass concentration was based on mixed liquor suspended solids content.

Table 3. OMP removal in AnMBR and major operating conditions.

Micropollutants	% Removal						
	A-[87]	**B-[88]**	**C-[89]**	**D-[90]**	**E-[74]**	**F-[86]**	**G-[80]**
17α-Estradiol	-	-	27	-	-	-	-
17α-Ethynylestradiol	-	-	15	-	100	-	-
17β-Estradiol	-	-	60	-	100	-	-
4-(tert-octyl)) phenol	-	-	-	-	0	-	-
4-p-nonylphenol	-	-	-	-	0	-	-
4-n-nonylphenol	94	96	-	-	-	-	-
Amitriptyline	99	90	47	77	-	-	-
Androstenedione	-	-	-	-	-	-	-
Androsterone	-	-	16	-	-	-	-
Aspartame	-	91	-	-	-	-	-
Atenolol	77	-	98	-	-	-	-
Atrazine	32	32	6.8	4	-	-	-
Bisoprolol	-	-	-	30	-	-	-
Benzophenone	-	62	-	-	-	-	-
Bisphenol A	99	4	32	15	-	-	81
Butylparaben	-	-	-	81	-	-	-
Caffeine	90	60	77	20	-	-	-
Carazolol	-	50	-	-	-	-	-
Carbamazepine	50	10	4.8	4	-	38	10
Celestolide	-	-	-	-	-	48	
Clozapine	99	81	28	75	-	-	-
DEET	99	10	1.4	5	-	-	-
Diazepam	54	20	-	6	-	38	2
Diazinon	93	91	-	79	-	-	-
Diclofenac	3	5	1	-	-	40	23
Dilantin	-	6	21	-	-	-	-
Diuron	62	16	-	7	-	-	-
EE2	-	-	-	-	-	12	-
Erythromycin	-	-	-	-	-	53	98
Enalapril	-	-	37	23	-	-	-
Estradiol E2	-	-	-	-	-	59	-
Estriol	-	-	1	-	-	-	-
Estrone	-	-	1		100	82	100
Ethinylestradiol	-	-	-	-	-	82	-
Etiocholanolone	-	-	52	-	-	-	-
Fluoxetine	-	-	-	-	-	22	92
Galaxolide	-	-	-	-	-	84	-
Gemfibrozil	18	11	13	-	-	-	-

Table 3. *Cont.*

Micropollutants	% Removal						
	A-[87]	**B-[88]**	**C-[89]**	**D-[90]**	**E-[74]**	**F-[86]**	**G-[80]**
Hydroxyzine	-	95	13	-	-	-	-
Ibuprofen	41	7	1	3	-	81	98
Ketoprofen	38	15	15	-	-	-	-
Linuron	88	90	11	62	-	-	-
Meprobamate	-	15	6.6	-	-	-	-
Metformin	-	-	99	-	-	-	-
Naproxen	75	51	70	40	-	92	77
Nonylphenol	-	96	99	80	-	-	-
Octylphenol	-	-	70	-	-	-	-
Omeprazole	99	97	20	-	-	-	-
Oxybenzone	-	98	-	-	-	-	-
Paracetamol	86	50	58	15	-	-	-
PFOS	-	64	-	-	-	-	-
Phenylphenol	-	57	-	42	-	-	-
Primidone	25	-	1.8	0	-	-	-
Propylparaben	-	70	-	-	-	-	-
Roxithromycin	-	-	-	-	-	42	96
Simazine	54	72	-	35	-	-	-
Sucralose	-	-	-	8	-	-	-
Sulfamethoxazole	99	95	95	-	-	99	99
TCEP	-	10	-	4	-	-	-
Testosterone	-	-	99	-	-	-	-
t-nonylphenol	-	-	-	-	0	-	-
t-Octylphenol	-	80	-	77	-	-	-
Tonalide	-	-	-	-	-	47	-
Triamterene	82	34	-	-	-	-	-
Triclocarban	95	89	37	71	-	-	-
Triclosan	70	62	90	59	-	-	97
Trimethoprim	98	82	35	94	-	-	99
Verapamil	-	-	99	-	-	-	-
α-ethinylestradiol	-	-	-	-	-	-	83
β-estradiol	-	-	-	-	-	-	100

	Wastewater	Membrane Type	HRT, SRT (d)	MLSS, MLVSS * (g/L)	T (°C)	Methane Yield
A-[87]	Synthetic wastewater	External ceramic membrane module (NGK, Japan), pore size1 µm, surface area 0.09 m^2, the cycle of 14 min on and 1 min off	4, 180	10	35	0.2 L CH4/g COD, 61% CH4, 5.4 L/d.

Table 3. *Cont.*

Micropollutants		% Removal						
		A-[87]	**B-[88]**	**C-[89]**	**D-[90]**	**E-[74]**	**F-[86]**	**G-[80]**
B-[88]	Synthetic wastewater	External ceramic membrane module (NGK, Japan), pore size 0.1 µm, surface area 0.09 m^2, cycle of 14 min on and 1 min off		5, 180	15, 10	35	0.2 L CH4/g COD (600 mg/L SO4 2-addition)	
C-[89]	Synthetic wastewater	Hollow-fiber membrane (Siemens Water Technologies, pore size 0.04 µm, total area of 0.0245 m^2		0.25, 30	Not measured	30	Not measured	
D-[90]	Synthetic wastewater	Side-stream MF membrane module (NGK, Japan), pore size 0.1 µm, effective area of 0.09 m^2, 14 min suction and 1 min relaxation		5, 140	16–22, 11.2	35	0.4–0.6 L/g COD, 58–65% CH_4	
E-[74]	Real wastewater pilot-plant (Valencia, Spain),	Hollow-fiber l ceramic UF membrane module (PURON® Koch Membrane Systems (PUR-PSH31), pore size 0.05 µm, effective area of 0.09 m^2, 14 min suction and 1 min relaxation		0.75–1, 80	na	Room temp		
F-[86]	Synthetic wastewater UASB ** + aerobic MBR	Submerged hollow fiber membrane (ZW-10 Zenon UF module), the pore size of 0.04 µm surface area 0.9 m^2, cycles of 7 min and 0.5 min of relaxation. Anaerobic temperature 20 to 22 °C. pH 7.5		MBR 0.5, 60 UASB 0.5, na				
G-[80]	Synthetic wastewater The pilot plant in Spain	Submerged HF membrane (ZW-10 Zenon UF module), the pore size of 0.04 µm, surface area 0.9 m^2, cycles of 7 min and 0.5 min of relaxation. UASB p H 7.1 ± 0.2		MBR 0.25 d, na UASB 0.75	6.9, 10.5	18.2–23.5		

Mixed liquor volatile suspended solids (MLVSS) *; upflow anaerobic sludge blanket (UASB) **.

As reviewed by Wijekoon et al. (2015) in an anaerobic process too low removal was achieved for an OMP such 17a–ethinylestradiol. Other OMPs—octyl phenol and nonylphenols—were also showed poor degradation [87]. In contrast, other researchers accomplished 20% removal for 17a–ethinylestradiol [89]. Those contradictory findings could be related to the wide variety of anaerobic microbes responsible for OMPs biodegradation. Though methanogenic archaea are major community for anaerobic degradation other electron acceptors such as sulfate-reducing, iron-reducing and nitrate-reducing microbial communities also plays significant role in OMP removal [87]. Alvarino et al. (2019) report that AnMBR improved up to 80% removal for 15 OMPs. In addition, they distinguished refractory compounds to biodegradation such as carbamazepine, diazepam and diclofenac. Further, naproxen, sulfamethoxazole, trimethoprim and fluoxetine were easily biodegradable compounds under anaerobic conditions. The authors further report that certain compounds were removed with combined UASB and the post-treatment reactor such as β-estradiol E2, α-ethinylestradiol EE2, erythromycin and erythromycin [80].

Salt accumulation, membrane module integrity, fouling of membrane and presence of refractory compounds are considered as major bottleneck for evolution of AnMBR in micropollutants removal during sewage treatment [81]. Wijekoon et al. (2015) have evaluated fate of 27 OMPs in AnMBR. They deduced that removal of OMPs can be correlated to their hydrophobicity and molecular structures. In particular, hydrophobic OMPs could be easily adsorbed on to the sludge and more than 70% removal for such OMPs had been reported. On the other hand electron donating hydrophilic OMPs such as sulfamethoxazole, carbamazepine, linuron, omeprazole and atrazine having hydroxyl and amine bearing nitrogen atoms in their structure shown high removal [87]. Another report by Hai et al. (2011) also confirmed improved process performance in anoxic process compared to aerobic process with regard to carbamazepine removal [91]. On the contrary, halogenated compounds such as chlorine molecule and amide like hydrophilics with EWGs had shown recalcitrant behavior in AnMBR process [80].

Monsalvo et al. (2014) evaluated OMP removal in AnMBR process and deduced that 50–90% removal were accomplished. However, nine OMPs showed over 90% removal. They also postulate OMP removal mechanism by AnMBR in detail, such as the impact of biodegradation, sorption and physicochemical properties (hydrophobicity, presence of EDG and EWG). As can be seen in Figure 3, easily biodegradable OMPs show better removal in AnMBR. Certain OMPs such as estriol, primidone, 17a-ethynylestradiol, 17a-estradiol, 17b-estradiol and rosterone and testosterone showed higher attachment onto the sludge having an overall 69 ng OMPs/g total solids (TS) sludge sorption capacity for OMPs. Nevertheless, analysis of variance (ANOVA) revealed a non-linear correlation for OMPs sludge sorption and Kow and the Log D values. Furthermore, authors noticed that AnMBR process showed better removal for EWDG containing compounds having higher Log D values as compared to too low removal (<21.4%) for OMPs bearing strong EWG [89]. Wijekoon et al. (2015) evaluated OMP removal in their work and deduced that more than 27% removal could be accomplished for the entire set of hydrophobic compounds. The authors co-related the removal of OMPs by AnMBR to their physicochemical properties, particularly hydrophobicity and molecular structure. Their results showed that all hydrophobic compounds out of 27 OMPs were removed by>70%. On the other hand, hydrophilic OMPs having strong electron withdrawing groups in their structure were found recalcitrant to the biodegradation. The authors further note that AnMBR processes achieved better performance than aerobic process for those OMPs such as linuron and caffeine with either nitrogen or sulfur in their structure. Enhanced removal for such compounds can be correlated to the nitrogen and sulfur reducing microbes available in AnMBR [87].

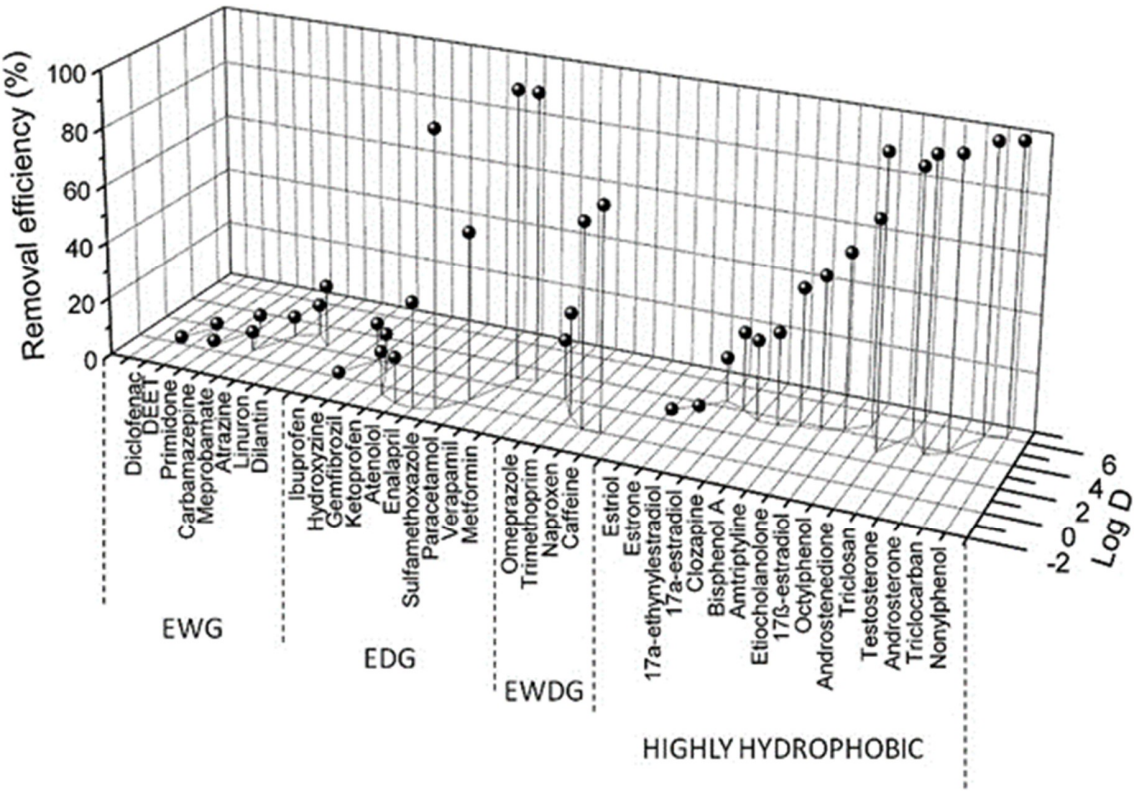

Figure 3. Effect of functional groups and hydrophobicity on the removal of OMPs in the AnMBR [89]. Reproduced with permission from [89], Copyright Water Research, 2014.

In another study, Abargues et al. (2012) compared the removal of the alkylphenols (4-(1,1,3,3-tetramethyl butyl)phenol, 4-p-nonylphenol and technical nonylphenol) and the hormones (estradiol (E2), estrone (E1), ethynylestradiol (EE2) in a pilot-scale submerged anaerobic membrane bioreactor (AnMBR) and report that hormones (E1, E2 and EE2) were below detection limit in the soluble fraction of AnMBR. The AnMBR favorably transformed alkylphenol polyethoxylates into alkylphenol polyethoxylates degradation into alkylphenols under anaerobic conditions. This improved biotransformation of alkylphenol polyethoxylates could be attributed to anaerobic conditions of 25h HRT that led to enhance time for biodegradation as well as high sludge sorption due to low Kow of the compound [74].

MBR for biofuel production is still a new concept whereby only a few industries employ anaerobic biologic process for wastewater treatment [82]. Song et al. (2016) investigated effects of increased salt concentration on OMP removal in anaerobic membrane bioreactor (AnMBR). It has been reported that salt accumulation up to 15 g/L (as sodium chloride (NaCl)) adversely affected AnMBR performance in terms of methane production and hydrophilic OMPs. The authors further report that salt accumulation had no pronounce effect on high removal of hydrophobic OMPs [92].

3. High Retention Membrane Bioreactors (HRMBR)

Organic micropollutants (OMPs) have been largely revealed in sewage, hospital and industrial wastewater at concentrations of up to several micrograms per liter [93]. Though many reports claimed more efficient and reliable OMP removal in MBRs than activated sludge process. However, persistent, high molecular weight hydrophilic OMPs still showed poor removal in MBR treatment [93,94]. While reverse osmosis can efficiently remove low molecular weight compounds, the OMPs rejected by this technique (RO concentrate) are not always recycled to bioreactor (MBR) to achieve more degradation. Thus, the organic retention time (ORT) of those micropollutants are not independent of the HRT of the MBR process [35].

Recent progress in advancing sewage purification and reclamation motivated research efforts towards novel technologies of high retention membrane bioreactors (HRMBRs) that showcased advanced wastewater treatment technology. Instead of deploying multiple treatment processes, water reclamation could be achieved in a HRMBR offers small footprint [95]. These mainly include the integration of NF, FO and MD membranes to the conventional MBRs. Some of them are osmotic membrane bioreactor (OMBR), membrane distillation bioreactors (MDBR), bio-electrochemical membrane reactor (BEMR) which can be an efficient and a safer 'multiple-barrier approach' in sewage treatment and specifically to achieve high removal of OMPs [81,82,95]. In addition to achieving the rejection and prolonging the retention time of refractory OMPs for further biodegradation, HRMBRs are less energy intensive due to natural osmosis phenomena thus alleviating the greenhouse gas (GHG) emission issue during sewage reclamation process [35,95]. Mert et al. (2018) appraised that OMP showed varying removal efficiency for different membrane technologies (0–100% removal). For instance, carbamazepine an anticonvulsant medication compound is quite persistent, which showed, less than 20% with removal in conventional sludge process and MBR [52]. However, integrated MBR-NF MBR–RO system accomplished 93% and 99% removal, respectively. In recent years both academia and industry shown increasing attention with regards to development of HRMBR [95].

The combined MBR can produce better quality permeate, lessen membrane fouling and thereby reduced cleaning cycles [82]. Nonetheless, HRMBR technology has certain drawbacks that need to be resolved prior to commercialization. Salt accumulation due to reverse salt diffusion from draw side to feed side, flux decline with time due to salt accumulation and concentration polarization, membrane fouling and membrane deterioration due to bacterial attack are major concerns. Many novel aspects are considered to overcome those shortcomings such as progress in developments in novel diversified bacterial consortia and advanced membranes [95]. In the following sections two major HRMBR systems, namely osmotic membrane bioreactors (OMBR) and membrane distillation bioreactors (MDBR) performance in wastewater treatment was discussed.

3.1. Osmotic Membrane Bioreactor (OMBR) for OMPs Removal

One of the promising options is to integrate submerged forward osmosis membrane with bioreactor known as an osmotic membrane bioreactor (OMBR) was materialized as an attractive alternative for sewage treatment and reuse [96,97]. OMBRs can produce clean permeate and efficiently degrade nutrients and OMPs as well as toxic pollutants like phenols. Another advantages are low fouling propensity of FO membranes [98,99]. OMBRs present certain benefits such as low fouling propensity, less numbers of cleaning cycles and therefore less operating cost and energy intensive process [58,97]. Further, it is possible to offer longer contact time for refractory compounds in OMBR due to the size exclusion mechanism it can retain any compound having molecular cutoff weight (MWCO) larger than forward osmosis membrane pore size [82].

For now, salt accumulation from draw solute to feed solution is a major issue in OMBR operation. FO membrane is semipermeable and hence salt leakage is an unavoidable phenomenon that relates molecular weight of draw solution. OMBR operation at high sludge retention, coupling with MF/UF membrane could be an option to mitigate salinity build up [100]. Recently combining OMBR with electrodialysis (ED) was suggested [53].

As previous research suggests, OMBR offers excellent OMP removal due to synergy between FO rejection followed by biotransformation in the bioreactor at long HRT and SRT. FO membrane rejection mechanism for OMP removal can be attributed to electrostatic repulsion, hydrated radius, molecular weight cut-off, retarded forward diffusion, Henry's law constants and hydrophilic (log D). Further, in the bioreactor OMP removal involves biodegradation in presence of diverse microbial communities and sludge sorption [101]. Table 4 shows some of the recent work of OMBR in OMP removal. Lay et al. (2012) successfully achieved >96% removal of 4 pharmaceutical compounds in OMBR that showed varying biologic process removal efficiency. For instance, ibuprofen showed highest removal (>90%) and below detection limits of the instrument followed by 40–90% moderate removal for naproxen.

Nonetheless, diclofenac had too low removal of < 40% and almost no biodegradation was observed for very persistent carbamazepine (0%) [102]. Holloway et al. (2014) used real wastewater as feed to the UF-OMBR (hybrid ultrafiltration-osmotic membrane bioreactor) system to examine OMP removal and deduced that among 20 OMPs, the UF-OMBR system was found efficient to remove 15 OMPs well below the analytical instrument detection limit. Forward osmosis membrane has shown higher removal of hydrophilic OMPs than hydrophobic OMPs. They noted that when UF system was offline for two weeks removal of bisphenol A, DEET, TCEP and sulfamethoxazole decreased, due to salinity build-up [36]. In another OMBR–RO operation performed by Luo et al. (2017), salt accumulation has negatively affected process performance by both reducing and changing microbial consortia thereby increased forward osmosis membrane fouling due to increased SMP and EPS [5].

Table 4. Removal of OMPs in OMBR and major operating conditions.

Wastewater	Membrane with Operating Conditions	Flux (LMH)	Salinity (g/L)	Micropollutants Investigated	% Removal	References
Synthetic Wastewater	Flat-sheet aquaporin FO membrane (Aquaporin Asia, Singapore), area 0.012 m^2, 0.5 M NaCl DS *, SRT 20 d, temperature 22 °C, HRT 24–36 h, MLSS 6.8 g/L, DO 2 mg/L	14–10	4	Clofibric acid	94	[104]
				Salicylic acid	100	
				Ketoprofen	100	
				Fenoprop	93	
				Naproxen	100	
				Metronidazole	98	
				Ibuprofen	100	
				Primidone	84	
				Diclofenac	99	
				Gemfibrozil	99	
				Propoxur	99	
				Enterolactone	80	
				Carbamazepine	92	
				pentachlorophenol	98	
				DEET	99	
				Estriol	94	
				Atrazine	90	
				Ametryn	93	
				Amitriptyline	98	
				Benzophenone	100	
				4-tert-Butylphenol	100	
				Oxybenzone	99	
				Estrone	100	
				bisphenol A	100	
				17α-ethynylestradiol	98	
				17β-estradiol	99	
				Triclosan	99	
				β-Estradiol-17-acetate	100	
				4-tert-Octylphenol	99	
				Octocrylene	98	

Table 4. *Cont.*

Wastewater	Membrane with Operating Conditions	Flux (LMH)	Salinity (g/L)	Micropollutants Investigated	% Removal	References
Synthetic Wastewater (OMBR)	TFC ** Flat-sheet FO membrane (Hydration Technology Inc.), area 0.03 m^2, 0.5 M NaCl DS, SRT 20 d, temperature 21 °C, MLSS 5.5 g/L, DO 5 mg/L	8–3	4	Clofibric acid Salicylic acid Ketoprofen Fenoprop Naproxen Metronidazole Ibuprofen Primidone Diclofenac Gemfibrozil Propoxur Enterolactone Carbamazepine pentachlorophenol DEET Estriol Atrazine Ametryn Amitriptyline Benzophenone 4-tert-Butylphenol Oxybenzone Estrone bisphenol A 17α-ethynylestradiol 17β-estradiol Triclosan β-Estradiol-17-acetate 4-tert-Octylphenol Octocrylene	99 100 97 97 98 99 99 100 100 98 98 96 97 98 98 95 90 94 99 99 99 100 100 98 100 100 100 100 100	[6]
Synthetic Wastewater (OMBR+MF)	CTA # Flat-sheet FO membrane (Hydration Technology Inc.-(HTI)), area 0.014 m^2, 1 NaCl DS, PVDF MF membrane Mitsubishi Rayon Engineering (Tokyo, Japan) area 0.074 m^2, pore size 0.4 µm, temperature 22 °C, HRT 24 h, MLSS 2–3.3 g/L, DO 5 mg/L	MF 1.6–2.6 FO 1.7 (steady)	0.4	Salicylic acid Clofibric acid Metronidazole Fenoprop Ketoprofen Naproxen Primidone Ibuprofen Propoxur Diclofenac Enterolactone Carbamazepine Gemfibrozil Amitriptyline DEET Estriol Atrazine pentachlorophenol Ametryn Benzophenone 4-tert-Butylphenol Estrone bisphenol A Oxybenzone 17α-ethynylestradiol 17β-estradiol β-Estradiol 17-acetate 4-tert-Octylphenol Triclosan Octocrylene	95 89 90 85 99 100 100 100 70 80 82 65 97 90 99 100 20 100 90 98 97 100 85 98 100 100 99 98 97 92	[76]

Table 4. *Cont.*

Wastewater	Membrane with Operating Conditions	Flux (LMH)	Salinity (g/L)	Micropollutants Investigated	% Removal	References
Synthetic Wastewater (OMBR+MF)	TFC Flat-sheet FO membrane (Hydration Technology Inc.), area 0.056 m^2, 0.5 M NaCl DS, PVDF MF membrane area 0.12 m^2, pore size 0.20 µm, SRT 30 d, temperature 25 °C, pH 7.5, HRT 4.2–6.6 h, MLSS 6 g/L, DO 6 mg/L	FO 6.06–8.14, MF 7.23–9.24		Bezafibrate Indomethacin Ketoprofen Gemfibrozil Ibuprofen Trimethoprim Atenolol Sulfamethoxazole	98 100 94 99 100 92 98 96	[101]
Synthetic wastewater	Flat-sheet, thin-film composite HTI-TFC-FO area 0.03 m^2, MLSS 5 g/L, DO 5 mg/L, SRT 20 d,HRT 27–60 h, 0.5 M NaCl DS	7–2	2–6	Clofibric acid Salicylic acid Ketoprofen Fenoprop Naproxen Metronidazole Ibuprofen Primidone Diclofenac Gemfibrozil Propoxur Enterolactone Carbamazepine pentachlorophenol DEET Estriol Atrazine Ametryn Amitriptyline Benzophenone 4-tert-Butylphenol Oxybenzone Estrone bisphenol A 17α-ethynylestradiol 17β-estradiol Triclosan β-Estradiol-17-acetate 4-tert-Octylphenol Octocrylene	98 98 97 96 98 98 99 98 98 98 99 98 98 98 98 93 90 97 96 98 98 99 99 97 98 99 98 98 98 90	[5]
Synthetic wastewater	Laboratory scale baffled OMBR, HTI-CTA FO membrane and PES hollow-fiber membrane module, pore size of 0.4 µm and area FO membrane 0.0264 m^2 and MF membrane 0.1 m^2, HRT 30 h, SRT 70 d MLSS 3.5 g/L,	7–5.5	2.5	Caffeine Atrazine Atenolol	94 51 100	[100]

Table 4. *Cont.*

Wastewater	Membrane with Operating Conditions	Flux (LMH)	Salinity (g/L)	Micropollutants Investigated	% Removal	References
Municipal wastewater	Pilot-scale hybrid UF-OMBR, UF hollow-fiberPVDF membrane module (Koch membrane) 0.03 µm pore size, area 0.44 m^2, FO plate-and frame Cassette (Hydration Technology), 1.2 m^2 area, MLSS 1.6–3.6 g/L, SRT-63-68 d, HRT 30 h, DS-0.7M NaCl,	4.7 steady	2 (UF subsystem on)	Acesulfame Acetaminophen Atenolol bisphenol A Caffeine DEET Diclofenac Diphenhydramine Fluoxetine Ibuprofen Naproxen Oxybenzone Propylparaben Sulfamethoxazole Surcalose Triclocarban Trimethoprim TCEP TCPP TDCP	100 100 100 87 100 96 100 100 100 100 100 100 100 99 100 100 100 98 99 100	[36]
Pharmaceutical wastewater	OMBR, FO flat sheet (Hydration Technology), 0.04 m^2 area, MLSS 7.2–8.1 g/L, SRT-63-68 d, HRT 30 h, DS-0.5 NaCl, SRT– 20 d, HRT-33 h (HTI)	2.7 steady	na	Diclofenac Naproxen Ibuprofen	98.4 98.3 97.1	[102]

Table 4. *Cont.*

Wastewater	Membrane with Operating Conditions	Flux (LMH)	Salinity (g/L)	Micropollutants Investigated	% Removal	References
Synthetic Wastewater	Lab-scale OMBR, CTA FO flat sheet (Hydration Technology), 0.02 m^2 area, PRO mode, DS- 1.5 M NaCl, MLSS 3.4–3.7 g/L	3 (Steady)	4.1	trimethoprim	32%	[103]
				diclofenac	30%	
				simazine,	02	
				atrazine,	20	
				diuron	22	
				Salicylic Acid	70	
				Paracetamol	100	
				Phenylphenol	90	
				Propylparaben	99	
				DEET	32	
				Caffeine	60	
				Ibuprofen	99	
				t-octylphenol	90	
				Primidone	58	
				Meprobamate	38	
				Nonylphenol	78	
				Naproxen	82	
				Carbamazepine	32	
				Linuron	70	
				Gemfibrozil	90	
				Dilantin	36	
				Triamteren	32	
				eSulfamethoxazole	80	
				Ketoprofen	82	
				pentachlorophenol	80	
				Atenolol	100	
				Estrone	98	
				17β-Estradiol	99	
				17α-Estradiol	99	
				Amitriptyline	99	
				Androstenedione	99	
				Estriol	98	
				Testosterone	100	
				Triclosan	90	
				Trimethoprim	33	
				Etiocholanolone	100	
				Androsterone	100	
				17α-Ethynylestradiol	99	
				Diazinon	99	
				Fluoxetine	99	
				Triclocarban	97	
				Clozapine	100	
				Omeprazole	83	
				Chlorpyrifos	99	
				HydroxyzineEnalapril	99	
				Risperidone	100	
				Simvastatin	82	
				Methotrexate	80	
				Verapamil	91	
				Simvastanin	100	

Draw Solution (DS) *; Thin Film Composite (TFC) **; Cellulose Triacetate (CTA) #.

Alturki et al. (2012) report >80% OMP removal for high molecular weight (>266 g/mol) molecules. They attributed this high removal to FO membrane rejection by size exclusion and also biodegradation. They also noted that biodegradation efficiency was negatively affected by salt accumulation in the bioreactor with time during continuous operation [103]. Luo et al. (2017) appraised

that >90% biodegradation was accomplished for all hydrophobic OMPs in OMBR–RO system [5]. Luo et al. (2015b) in their MF-OMBR work also noticed that out of 30 OMPs eleven hydrophobic OMPs were efficiently removed in OMBR and MF-MBR system, whereas adsorption onto the sludge was considered a dominant removal mechanism. Furthermore, authors report that OMPs such as salicylic acid (hydrophilic OMP) and bisphenol A and octocrylene (hydrophobic OMP) showed poorer removal by forward osmosis membrane in OMBR than MF-MBR which could be linked to the cake enhanced concentration polarization due to foulants cake layer on the membrane surface. Indeed, detailed investigations for such phenomena need to be performed to confirm such results [76].

To achieve efficient biodegradation of C and N atoms, conventional activated sludge (CAS) system was separated into anaerobic, anoxic and aerobic compartments operated in different sequences. Usually multiple reactor configuration is the most common however single-compartment with varying redox conditions may be used to save space. In single reactor configuration, partition was made by inserting baffles to divide reactor in anoxic and oxic zones, also by setting aeration cycle time anoxic and aerobic conditions can be achieved in a single bioreactor. This unique redox condition leads to entirely different microbial consortia capable to accomplish carbon and nitrogen biodegradation and stripping-off carbon (carbon dioxide (CO_2)) and nitrogen (e.g., nitrogen (N_2) and nitrous oxide (N_2O)) gases [3]. Pathak et al. (2018) recently examined a novel baffled osmotic membrane bioreactor-microfiltration integrated process to study the removal of three OMPs employing inorganic and organic draw solutes. Model OMPs showed better removal with organic draw solutes and in general baffled OMBR showed very high total nitrogen (>85%) and excellent OMP removal. Atrazine, a very refractory molecule and pesticide compound, had shown enhanced removal under unique redox environment under extended anoxic cycle time of 1.5 h that may have developed very different microbial community responsible for different enzyme secretion [100].

Zhang et al. (2017) evaluated removal of 30 different hydrophilic and hydrophobic OMPs in OMBR employing two different CTA and TFC membranes. The authors report that TFC membrane outperformed CTA membrane for OMP rejection and reduced load on combined RO process. Moreover, >95% removal was observed for hydrophobic OMPs with both CTA and TFC membranes. However, <80% removal was noted for hydrophilic OMPs such as clofibric acid, fenoprop, primidone, diclofenac, propoxur, carbamazepine, atrazine and ametrine. The authors further linked this to the recalcitrant EWGs (-Cl, -NO_2) in hydrophilic OMPs than EDGs (-NH_2 and –OH) in hydrophobic OMPs [6].

Similarly, Luo et al. (2018) observed that aquaporin FO membrane showed robustness and stability when integrated with activated sludge treatment. The authors report that OMBR achieved more than 80% removal for hydrophilic and refractory OMPs such as salicylic acid, ketoprofen, naproxen, metronidazole, ibuprofen, gemfibrozil, pentachlorophenol, DEET and ametrine. However, certain OMPs such as clofibric acid, fenoprop, rimidone, carbamazepine and atrazine showcased poor removals of less than 30% [104]. Luo et al. (2017) also observed that some hydrophilic OMPs such as clofibric acid, fenoprop, primidone, diclofenac, carbamazepine, atrazine and ametrine, had demonstrated too low removal of 20–70% in OMBR–RO process [5]. This removal difference could be further attributed to different EDG (e.g., amine and hydroxyl) and EWG (e.g., chloro, amide and nitro) functional groups in the molecular structure of these hydrophilic compounds [94,104]. In another lab-scale OMBR study [105] observed that a low MLSS concentration is not enough to effectively remove OMPs with both slow biodegradation and low molecular weight of the OMPs such as Trimethoprim. However, the removal rate of Trimethoprim was improved when biomass was increased [101].

3.2. Membrane Distillation Bioreactor (MDBR) for OMPs Removal

Membrane distillation incorporates hydrophobic microporous membranes that operate at low-temperature, involving solely transfer of water vapor from feed side to the distillate side through membrane pores. Due to gas-phase mass transfer, only volatiles could pass through the membrane and thus MD completely retains non-volatiles in feed solution [75,105]. A novel MDBR process combines thermophilic activated sludge membrane bioreactor (MBR) with the membrane distillation

(MD) process where usually direct contact membrane distillation module is immersed in a biologic reactor [49,106].

Hydrophobic membrane modules in MDBR are usually made of polypropylene (PP), polyvinylidene fluoride (PVDF) or polytetrafluoroethylene (PTFE) [106]. The advantages with MDBR consist of high organic removal in sewage reuse, less sludge production and least affected by salinity build up as in OMBR process, while complete rejection of salt can be achieved. MDBR offers advantages such as being less susceptible to membrane fouling, low installation cost and good performance under moderate thermophilic temperatures [82]. Compared to MBR and OMBR, in an industrial facility where both hot effluent and surplus heat is available MDBR could be an attractive emerging technology to be applied [107].

Wijekoon et al. (2014) noted that MDBR could achieve enhanced OMP removal compared to stand-alone MD process. Actually, in MD process OMP removal depends on the Henry's constant, H (vapor pressure) and the water partition coefficient (log D) of the OMPs to be removed. When MD process has a low (<2.5) 'pKH/log D' ratio poor OPMs removal is accomplished. But in MDBR even at low (<2.5) 'pKH/log D' ratio' higher OMP removal is possible due to the sorption of OMPs onto the sludge surface which prolongs OMPs retention followed by potential biodegradation [75,108].

In MDBR process OMP removal mechanism involves hydrophobic membrane rejection, sorption onto the sludge particles and biotransformation in bioreactor in the presence of thermophilic bacteria [75]. Table 5 presents some of the MDBR reports on OMP removal.

Table 5. Removal of OMPs in membrane distillation bioreactor (MDBRs) and major operating conditions.

Wastewater	Membrane with Operating Conditions	Temperature		MD Flux LMH	OMPs Investigated	% Removal	References
		Feed	Permeate				
Synthetic wastewater	MD-EMBR * reactor PTFE side stream MD Pore size 0.2 µm, 95–100 µM (DMP) **/min laccase, DO 3 mg/L	30	10	4 (Steady flux)		99	[108]
					Primidone	98	
					Ketoprofen	99	
					Naproxen	98	
					Gemfibrozil	98	
					Metronidazole	96	
					Diclofenac	99	
					Fenoprop	98	
					Ibuprofen	94	
					Ametrine	99	
					Clofibric acid	99	
					Carbamazepine	99	
					Octocrylene	99	
					Amitriptyline	92	
					Atrazine	95	
					Propoxur	99	
					Benzophenone	94	
					DEET	96	
					Enterolactone	97	
					Estriol	98	
					17α–Ethinylestradiol	94	
					Oxybenzone	99	
					Estrone	98	
					17β–Estradiol	98	
					17β-Estradiol-17-acetate		
					bisphenol A		
					Salicylic acid	96	
					pentachlorophenol	97	
					Triclosan	97	
					4-tert-Butylphenol	98	
						98	

Table 5. *Cont.*

Wastewater	Membrane with Operating Conditions	Temperature		MD Flux LMH	OMPs Investigated	% Removal	References
		Feed	Permeate				
Synthetic wastewater	Membrane distillation with an enzymatic bioreactor (MD-EMBR) PTFE side stream MD Pore size 0.22 µm, 95–100 µM (DMP)/min laccase, DO 3 mg/L	30	10	3.75	Sulfamethoxazole Carbamazepine Diclofenac Oxybenzone Atrazine	>99% for all	[20]
Synthetic wastewater	AnMBR-MD AnMBR MLSS 10 g/L, MLVSS 5 g/L, ceramic MF pore size 1 µm, area 0.09 m^2, HRT 4 d, 0.3 to 0.5 L/g COD, MD PTFE membrane, 0.2 µm pore size.	45	20		Caffeine Sulfamethoxazole Ketoprofen Trimethoprim Paracetamol Naproxen Primidone Ibuprofen Triamterene Carazolol TCEP Diclofenac Carbamazepine Gemfibrozil Simazine Amitriptyline Atrazine Diuron Propylparaben Linuron Clozapine Phenylphenol bisphenol A Diazinon Triclosan Triclocarban	99 89 99 99 99 97 99 99 98 97 92 75 90 99 79 99 74 99 91 93 99 80 85 99 85 95	[83]
Synthetic wastewater	PTFE side stream MD Bioreactor MLSS 5.3 g/L, pH 7.6, DO 2.8 mg/L, HRT 9.6 d, temperature 40 °C	40	14	1.2 (Steady flux) >95% removal for all OMPs	Clofibric acid Salicylic acid Ketoprofen Fenoprop Naproxen Ibuprofen Primidone Diclofenac Gemfibrozil Propoxur Carbamazepine pentachlorophenol Estriol Atrazine Ametryn Benzophenone Amitriptyline 4-Tert-butyphenol Oxybenzone Estrone 17α-Ethinylestradiol 17β-Estradiol Triclosan 17β-Estrodiol-17-acetate Octocrylene	100 96 99 97 100 100 100 95 98 100 96 97 98 96 99 97 99 98 99 100 99 100 98 100 97	[75]

Table 5. *Cont.*

Wastewater	Membrane with Operating Conditions	Temperature		MD Flux LMH	OMPs Investigated	% Removal	References
		Feed	Permeate				
Synthetic wastewater	OMBR-MD TFC-FO (HTI) membrane, 0.42 nm pore size, effective area of 300 cm^2, MLSS 6 g/L, HRT 30–40 h, SRT 20 d, DO 5 mg/L. PTFE MD membrane, 2 nm pore size.	40	20	6 (Steady flux)		99	[97]
					Clofibric acid	99	
					Salicylic acid	99	
					Ketoprofen	98	
					Fenoprop	99	
					Naproxen	95	
					Metronidazole	99	
					Ibuprofen	99	
					Primidone	98	
					Diclofenac	99	
					Gemfibrozil	99	
					Propoxur	90	
					Enterolactone	99	
					Carbamazepine	99	
					pentachlorophenol		
					DEET	99	
					Estriol	91	
					Atrazine	95	
					Ametrine	98	
					Amitriptyline	99	
					Benzophenone	98	
					4-Tert-butyl phenol	99	
					Oxybenzone	99	
					Estrone	99	
					bisphenol A	93	
					17α-Ethinylestradiol	98	
					17β-Estradiol		
					Triclosan	99	
					17β-Estradiol-17-acetate	98	
						98	
					Octocrylene	99	
						98	

Membrane Distillation-Enzymatic Membrane Boreactor (MD-EMBR) *; 2,6-dimethoxy phenol (DMP) **.

Wijekoon et al. (2014) concluded that both the salt accumulation and high temperature in bioreactor negatively influenced recalcitrant OMP removal in a membrane distillation–thermophilic bioreactor (MDBR). The hydrophilic compounds containing EWGs showed as low as 0 to 53% removal in thermophilic reactor due to their refractory nature during biologic process. Overall, MDBR process successfully removed more than 95% OMPs. However, all OMPs investigated were highly removed (>95%) by the MDBR system having more than 70% OMP removal by biodegradation. Membrane distillation contributed for certain OMPs rejection (42 to 94%) such as triclosan, fenoprop, atrazine, clofibric acid, diclofenac and carbamazepine. The authors further report that triclosan and octocrylene persistent hydrophobics were adsorbed on to the sludge led to better removal [75].

The synergy between the activated sludge and the MD membrane rejection contributed to 76% to complete removal of all 26 selected OMPs by the hybrid AnMBR-MD system. MD played a significant role in efficiently removing poorly degraded compounds from AnMBR such as primidone, ibuprofen, diclofenac and bisphenol A [83]. In another OMBR study, by integrating biologic process membrane distillation, the OMBR−MD combined system efficiently treated 30 OMPs successfully to extract reclaimed wastewater [97]. OMPs possessing EWG (e.g., -Cl_2, -NH_2 and –NO_2) in the molar configuration are non-biodegradable to biologic process. The removal of these persistent OMPs such as carbamazepine, clofibric acid, fenoprop, primidone, diclofenac, carbamazepine and atrazine were less than 40% by conventional MBR. Despite their persistence, more than 60% of removal was achieved due to the extended retention of such refractory OMPs in the OMBR-MD hybrid system [97]. Significant fouling of the MD unit and continuous flux decline was noticed due to the complete retention of SMP and EPS fractions and the inorganic salts accumulated in the MD feed solution [83].

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Another development is Laccase based enzymatic membrane bioreactor. Laccase can catalyze the degradation of a broad spectrum of pollutants including aromatic hydrocarbons, aliphatic amines and OMPs by using dissolved oxygen as a co-substrate. However, its larger-scale application is restricted by the lack of a reactor system, which can prevent washout of enzymes along with treated effluent [108]. In a recent study, Asif et al. (2017) developed a novel membrane distillation-laccase based enzymatic membrane bioreactor (MD-EMBR) system capable to retain laccase for OMP removal evaluation. They achieved 80 to 99% removal by biologic process in the MD-EMBR for oxybenzone and diclofenac, respectively [20]. In MD-EMBR operation at short HRT (12 h) significantly enhanced biodegradation was achieved due to lacassae presence. Compared to conventional biologic process 40% higher removal was noted specifically for refractory compounds such as carbamazepine. Moreover, MD-EMBR could achieve better OMP removal as compare to UF-OMBR. This is due to the retention of lacassae by MD-EMBR however UF-EMBRs could not retain laccase [20,108]. Similarly, in another such study, MD-EMBR achieved over 99% removal of some OMPs including 4-tert-octylphenol (pKH/logD = 0.98), octocrylene (pKH/log D = 1.21), 4-tert-butyl phenol (pKH/log D = 1.51), benzophenone (pKH/log D =1.83) and oxybenzone (pKH/log D = 2.1). This significant improvement can be attributed to the efficient degradation of these OMPs by laccase in MD-EMBR [108].

In MDBR, with course of time temperature polarization and settlement of non-volatiles negatively affected diversity and population of bacterial consortia that led to lowering Shannon index (1.75–2.53) observed which is lower than favorable Shannon index of more than 3 under operating conditions of 110 d SRT and without pH adjustment. Species from the kingdom fungi were observed to dominate in the MDBR. In spite of the lower biodiversity in the MDBR, there may be a chance that these particular niche species could be suitable for biodegradation of specific recalcitrant micropollutants [83]. Moreover, pH shift and lowered oxygen solubility with temperature increment are other potential factors which may affect OMP removal in MDBR [82].

4. Concluding Remarks and Future Perspectives

This paper reviewed recent research (2015-till date) concerning the practical application of membrane bioreactors (MBRs) and high-retention membrane bioreactors (HRMBRs) towards

the removal of OMPs from wastewater on both laboratory and pilot scale units. Challenges and apparent obstacles to the applications were discussed in the previous sections.

Environmental legislation all over the world should be tightened to include a wide range of OMPs in sewage and industrial treatments. Nevertheless, sound knowledge of their fate and transport during wastewater treatment and detailed environmental risk information is still missing [17]. Furthermore, standard operating procedures (SOPs) for OMPs measurement using analytical techniques and validation protocols should be established to prevent limitations and uncertainties in prevailing sampling methods [17,67]. Roos et al. (2012) suggested that exposure of OMPs such as pharmaceuticals to the environment should be assessed not only on sales statistics data, but also data on degradation, removal by CAS and MBR treatment and bioconcentration should be taken into account [109]. In short, ranking of OMPs to be included in legislative guidelines should be based on its risk potential [110]. To answer the growing concerns with emerging micropollutant, stricter discharge limits would likely be imposed on industrial effluents soon.

As noted by Huang and Lee (2015) recent R & D trends of MBR technology was shifted from process optimization and economic evaluation to the installation of new process architecture to enrich functional strains like nitrifiers and to applying MBR hybrid systems for achieving simultaneous removals of nutrients and OMPs [111]. Conventional MF/UF MBRs are already installed for wastewater treatment application. Therefore, scope exists to improve membrane morphologies and to integrate carefully with other established processes for better OMP removal [110]. Furthermore, low-pressure (MF and UF) membranes can be used in MBRs by addition of polymers or surfactants. While Polymer-enhanced and micellar-enhanced membrane processes can be efficiently used for the treatment of OMPs, little or no work has been reported, and scope exists to explore such techniques for OMP removal [110,112].

Some challenges with AnMBR for sewage treatment include the dilute nature and temperature difference of municipal wastewater, salinity build-up when diluted wastewater is preconcentrated, membrane fouling and stability and inhibitory substances (e.g., free ammonia and sulfide). However, commercialization of AnMBR at industrial scale is still pending due to membrane fouling and membranes sensitivity to toxicity [82]. Thus, future studies are required for the development of effective strategies to address these challenges for further development of AnMBR [81]. Furthermore, future trends should focus on novel ideas such as electrically enhanced AnMBR and electrically enhanced OMBR to further reduce energy consumption and ensure energy efficiency [53].

Due to the fewer reports of MBR and HRMBR treating real wastewater, a complete understanding of the OMP removal mechanisms is still missing. Thus, it is difficult to comment precisely on OMP removal solely by considering results obtained from lab-scale data with simulated water matrices, meaning that a true representation of real water matrices is required [110]. Nevertheless, in real effluents, it is difficult to predict OMP removal accurately from a blend of OMPs with varying concentration due to their simultaneous interactions [67,110].

A state-of-the-art HR-MBRs was successfully demonstrated from lab-scale experiments. Low flux, high salinity and membrane fouling and stability are critical issues for practical applicability of HRMBRs. Additionally, several technological challenges are associated with scaling-up of robustness and techno-economically feasible HRMBR at the pilot and commercial level to achieve high OMP removal and minimize toxic by-products [95]. Recently explored baffled osmotic membrane bioreactors can achieve simultaneous wastewater treatment, nutrient and OMP removal. Thus, this reactor design enables both aerobic and anoxic processes in an attempt to reduce the process footprint and energy costs associated with continuous aeration. Different redox conditions with extended anoxic cycle time can be linked with possible development of different microbial consortia responsible for diverse enzymes secretion responsible for efficient OMP removal [100,113]. Moreover, research efforts are in progress to make outer selective hollow fiber FO membrane modules. This FO modules are capable of producing around 14 LMH initial water flux during wastewater treatment. Compared to inner selective hollow fiber FO membranes outer selective hollow fiber membranes are more promising in

terms of higher sustained flux and less fouling propensity. However, both of those types of FO hollow fiber membrane reports are scarce and scope exists to scale –up and commercialize them [114].

Although anaerobic HRMBR was scarcely studied in the literature, given its potential for simultaneous wastewater treatment, biogas production and nutrient recovery and OMP removal potential this process is likely to be prioritized to be implemented at pilot and full-scale level [95]. Other novel concepts such as the coupling of electrically enhanced OMBRs with microbial fuel cell (MFC) for improvement in self-electricity generation should be tested in the future [52]. MFC alone leads to low-efficiency treatment and poor effluent quality due to limited biomass retention. Combination of MBR-MFC system known as an electrochemical membrane bioreactor (EMBR) offers a convincing option for wastewater treatment and energy recovery [82]. However, OMP removal employing EMBR studies are scarce [115] and scope exists to explore this process for micropollutant removal in cost-effective way.

Today, OMBR is still an emerging technology limited to lab-scale examinations. To accomplish better OMP removal with OMBR longer SRT is recommended. However, this leads to salinity build-up in the reactor. MF membranes must be incorporated in OMBRs for inorganic salt discharge. MF effluent may be potentially used for toilet flushing, gardening or green-wall irrigation agricultural irrigation where the presence of phosphorous and nitrogen are beneficial [101]. Actually, concentration factor (CF), which is defined as the concentration increase of inorganic salts in the bioreactor and it is a ratio of SRT to HRT during HRMBR operation is an important parameter for controlling and optimizing salinity buildup in HRMBRs [106,116]. Thus, an optimum CF value should be identified for balancing water recovery targets and salt accumulation in the bioreactor. Cost-effective treatment of membrane concentrates should be investigated. Further, microbial acclimation and the inoculation of halophilic microorganisms were put forward as feasible strategies to ensure a biologic treatment in the high saline environment. More important, easily biodegradable organically based draw solutes were tested and according to recent studies they do not contribute toward salinity build-up in the bioreactor [117]. Membrane fouling and energy consumption (aeration) are interconnected and considered as a major drawback in the application of MBR [82]. Further studies are necessary to ascertain the effects of the sludge cake layer on the rejection of OMPs—particularly the hydrophobic compounds, in the FO process [89,118].

The combinations of different complementary technologies have produced promising results. Nonetheless, there is a lack of a holistic understanding of the nature of pollutants, their interactions and predictable relationships between the best-available specific technologies [104]. More lab scale and pilot plant experiments should be performed to evaluate the performance of both hybrid MBRs and high retention MBRs for scaling-up and make them next-generation sustainable and techno-economically feasible technologies.

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