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

Nirenkumar Pathak 1, Van Huy Tran 1, Andrea Merenda 2, M. A. H. Johir 1, Sherub Phuntsho 1 and Hokyong Shon 1,∗

1 School of Civil and Environmental Engineering, University of Technology, Sydney, Post Box 129, Broadway, NSW 2007, Australia; Nirenkumar.Pathak@uts.edu.au (N.P.); VanHuy.Tran@student.uts.edu.au (V.H.T.); mohammed.johir@uts.edu.au (M.A.H.J.); Sherub.Phuntsho@uts.edu.au (S.P.)
2 Deakin University, Institute for Frontier Materials, Waurn Ponds, 3216 Victoria, Australia; a.merenda@deakin.edu.au

* Correspondence: Hokyong.Shon-1@uts.edu.au; Tel.: +61-447-332-707

Received: 5 March 2020; Accepted: 14 April 2020; Published: 24 April 2020

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](image_url)  
**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, mycoestrogens—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-Ethynyl 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.](image-url)
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 |
|-----------------------|-----------|
| 17α-ethynylestradiol  | 96        |
| 17β-estradiol         | 98        |
| 4-n-nonylphenol       | -         |
| 4-p-nonylphenol       | -         |
| 4-t-nonylphenol       | -         |
| 4-tert-Butylphenol    | 98        |
| 4-tert-Octylphenol    | 97        |

Note: The table data is not provided in the image.
Table 1. Cont.

| Micropollutants         | A- [5] | B- [68] | C- [68] | D- [69] | E- [70] | F- [71] | G- [71] | H- [72] | I- [73] | J- [74] |
|-------------------------|--------|---------|---------|---------|---------|---------|---------|---------|---------|---------|
|                         | MF     | UF      | SRT (so)| SRT (27d)|        |         |         |         |         |         |
| Acetaminophen           | -      | 90      | 90      | 95      | -       | -       | -       | -       | -       | -       |
| Amitriptyline           | 95     | -       | -       | -       | -       | 34      | -       | -       | -       | -       |
| Androsterone            | -      | -       | -       | -       | -       | 98      | 98      | -       | -       | -       |
| Atenolol                | -      | -       | 59      | -       | -       | 92      | 85      | -       | -       | -       |
| Atrazine                | 30     | -       | -       | -       | -       | -       | -       | -       | -       | -       |
| Bezfibrate              | -      | -       | 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      | -       | -       | -       |
| Diazepam/DPz            | -      | -       | 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      | -       | -       | -       | -       |
| Fenoproplone            | 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     | -       |
| Ilosfamide              | -      | -       | -       | -       | -       | -       | -       | -       | -       | 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                      | A- [5] | B- [68] | C- [68] | D- [69] | E- [70] | F- [71] | G- [71] | H- [72] | I- [73] | J- [74] |
|-------------------------------------|--------|---------|---------|---------|---------|---------|---------|---------|---------|---------|
|                                     | MF     | UF      | SRT (so)| 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 m², 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 m², 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 m², 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                        |
| Submerged hollow fiber MF PVDF membrane, pore size of 0.4 µm, total surface area 0.04 m², 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 m², 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 (Zeweeds-10, pore size 0.04 µm, total surface area 0.93 m², 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 | A- [5] | B- [68] | C- [68] | D- [68] | E- [70] | F- [71] | G- [71] | H- [72] | I- [73] | J- [74] |
|-----------------|--------|---------|---------|---------|---------|---------|---------|---------|---------|---------|
| % Removal       | MF     | UF      | SRT (so) | SRT (27d) |
| 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. |
| L- [73]         | Pilot scale MBR Hospital effluent (Marseille, France) | Hollow fiber Polysulfone membrane, 100 kDa MWCO, total surface area 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 m², HRT 9 h, SRT 100 d, MLSS 15 g/L. |

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, for 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 –NH2- 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 hydrophilic 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/m³)    | 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-Ethinylestradiol         | -      | -      | 15     | -      | 100    | -      | -      |
| 17β-Estradiol                | -      | -      | 60     | -      | 100    | -      | -      |
| 4-(tert-octyl)) phenol       | -      | -      | -      | -      | 0      | -      | -      |
| 4-p-nonylphenol              | -      | -      | -      | -      | 0      | -      | -      |
| 4-n-nonylphenol              | 94     | 96     | 47     | 77     | -      | -      | -      |
| Amitriptyline                | 99     | 90     | 47     | 77     | -      | -      | -      |
| Androstenedione              | -      | -      | 16     | -      | -      | -      | -      |
| Androsterone                 | -      | -      | 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     | -      | -      | -      | -      |
| 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 size 1 μm, surface area 0.09 m², 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 m², 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 m²) | 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 m², 14 min suction and 1 min relaxation | 5, 140 | 16–22, 11.2 | 35 | 0.4–0.6 L/g COD, 58–65% CH₄ |
| E-[74] Real wastewater pilot-plant (Valencia, Spain), | Hollow-fiber ceramic UF membrane module (PURON® Koch Membrane Systems (PUR-PSH31), pore size 0.05 µm, effective area of 0.09 m², 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 m², 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 m², cycles of 7 min and 0.5 min of relaxation. UASB p H 7.1 ± 0.2 | MBR 0.25 d, na | 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 as 17α-ethinylestradiol. Other OMPs—octyl phenol and nonylphenols—were also showed poor degradation [87]. In contrast, other researchers accomplished 20% removal for 17α-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 [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 EDC and EWG). As can be seen in Figure 3, easily biodegradable OMPs show better removal in AnMBR. Certain OMPs such as estriol, primidone, 17α-ethynylestradiol, 17α-estriadiol, 17β-estriadiol and roosterone 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 OMP’s 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].
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].

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.
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 naproen.
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 m², 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] |
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 m², 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 99 | | [6] |
|                     |                                    |            |                | Salicylic acid 100 | | |
|                     |                                    |            |                | Ketoprofen 97 | | |
|                     |                                    |            |                | Fenoprop 97 | | |
|                     |                                    |            |                | Naproxen 98 | | |
|                     |                                    |            |                | Metronidazole 99 | | |
|                     |                                    |            |                | Ibuprofen 99 | | |
|                     |                                    |            |                | Primidone 100 | | |
|                     |                                    |            |                | Diclofenac 100 | | |
|                     |                                    |            |                | Gemfibrozil 98 | | |
|                     |                                    |            |                | Propoxur 98 | | |
|                     |                                    |            |                | Enterolactone 96 | | |
|                     |                                    |            |                | Carbamazepine 97 | | |
|                     |                                    |            |                | pentachlorophenol 98 | | |
|                     |                                    |            |                | DEET 98 | | |
|                     |                                    |            |                | Estriol 95 | | |
|                     |                                    |            |                | Atrazine 90 | | |
|                     |                                    |            |                | Ametryn 94 | | |
|                     |                                    |            |                | Amitriptyline 99 | | |
|                     |                                    |            |                | Benzophenone 99 | | |
|                     |                                    |            |                | 4-tert-Butylphenol 99 | | |
|                     |                                    |            |                | Oxybenzone 100 | | |
|                     |                                    |            |                | Estrone 100 | | |
|                     |                                    |            |                | bisphenol A 98 | | |
|                     |                                    |            |                | 17α-ethynylestradiol 100 | | |
|                     |                                    |            |                | 17β-estradiol 100 | | |
|                     |                                    |            |                | Triclosan 100 | | |
|                     |                                    |            |                | β-Estradiol-17-acetate 100 | | |
|                     |                                    |            |                | 4-tert-Octylphenol 100 | | |
|                     |                                    |            |                | Octocrylene 100 | | |
| Synthetic Wastewater (OMBR+MF) | CTA # Flat-sheet FO membrane (Hydration Technology Inc.-HTI), area 0.014 m², 1 NaCl DS, PVDF MF membrane | MF 1.6–2.6 | 0.4 | Salicylic acid 95 | | [76] |
|                     |                                    | FO 1.7 (steady) |                | Clofibric acid 89 | | |
|                     |                                    |            |                | Metronidazole 90 | | |
|                     |                                    |            |                | Fenoprop 85 | | |
|                     |                                    |            |                | Ketoprofen 85 | | |
|                     |                                    |            |                | Naproxen 100 | | |
|                     |                                    |            |                | Primidone 100 | | |
|                     |                                    |            |                | Ibuprofen 100 | | |
|                     |                                    |            |                | Propoxur 70 | | |
|                     |                                    |            |                | Diclofenac 80 | | |
|                     |                                    |            |                | Enterolactone 82 | | |
|                     |                                    |            |                | Carbamazepine 82 | | |
|                     |                                    |            |                | Gemfibrozil 65 | | |
|                     |                                    |            |                | Amitriptyline 97 | | |
|                     |                                    |            |                | DEET 99 | | |
|                     |                                    |            |                | Estriol 99 | | |
|                     |                                    |            |                | Atrazine 20 | | |
|                     |                                    |            |                | pentachlorophenol 100 | | |
|                     |                                    |            |                | Ametryn 90 | | |
|                     |                                    |            |                | Benzophenone 98 | | |
|                     |                                    |            |                | 4-tert-Butylphenol 98 | | |
|                     |                                    |            |                | Estrone 100 | | |
|                     |                                    |            |                | bisphenol A 85 | | |
|                     |                                    |            |                | Oxybenzone 98 | | |
|                     |                                    |            |                | 17α-ethynylestradiol 100 | | |
|                     |                                    |            |                | 17β-estradiol 100 | | |
|                     |                                    |            |                | Triclosan 100 | | |
|                     |                                    |            |                | β-Estradiol-17-acetate 98 | | |
|                     |                                    |            |                | 4-tert-Octylphenol 98 | | |
|                     |                                    |            |                | Octocrylene 92 | | |
| 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 m², 0.5 M NaCl DS, PVDF MF membrane area 0.12 m², 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 98 | | [101] |
| Synthetic wastewater | Flat-sheet, thin-film composite HTI-TFC-PO area 0.03 m², 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 98 | Salicylic acid 98 | Ketoprofen 97 | Fenoprofen 96 | Naproxen 98 | Metromidazole 98 | Ibuprofen 99 | Primidone 98 | Diclofenac 98 | Gemfibrozil 98 | Propoxur 99 | Enterolactone 98 | Carbamazepine 98 | pentachlorophenol 98 | DEET 98 | Estradiol 93 | Atrazine 90 | Ametrisyline 90 | Amitriptyline 96 | Benzophenone 98 | 4-tert-Butyphenol 98 | Oxybenzone 99 | Estrone 99 | bisphenol A 97 | 17α-ethynylestradiol 98 | 17β-estradiol 99 | Triclosan 98 | β-Estradiol-17-acetate 98 | 4-tert-Octylphenol 98 | Octocrylene 90 |
| 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 m² and MF membrane 0.1 m², HRT 30 h, SRT 70 d, MLSS 3.5 g/L, | 7–5.5 2.5 | | Caffeine 94 | Atrazine 51 | Atenolol 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     | 4.7 steady | 2 (UF subsystem on) | Acesulfame                  | 100       | [36]       |
|                       | hollow-fiberPVDF membrane module   |            |                | Acetaminophen               | 100       |            |
|                       | (Koch membrane)                     |            |                | Atenolol                    | 100       |            |
|                       | 0.03 µm pore size, area 0.44 m², FO |            |                | bisphenol A                 | 87        |            |
|                       | plate-and frame Cassette (Hydration |            |                | Caffeine                    | 100       |            |
|                       | Technology),                         |            |                | DEET                        | 96        |            |
|                       | 1.6–3.6 g/L, SRT-63-68 d, HRT 30 h, |            |                | Diclofenac                  | 100       |            |
|                       | DS-0.7M NaCl,                       |            |                | Diphenhydramine             | 100       |            |
|                       |                                    |            |                | Flutoxetine                 | 100       |            |
|                       |                                    |            |                | Ibuprofen                   | 100       |            |
|                       |                                    |            |                | Naxprofen                   | 100       |            |
|                       |                                    |            |                | Oxycodone                   | 100       |            |
|                       |                                    |            |                | Propylparaben               | 100       |            |
|                       |                                    |            |                | Sulfamethoxazole            | 99        |            |
|                       |                                    |            |                | Surcalose                   | 100       |            |
|                       |                                    |            |                | Triclocarban                | 100       |            |
|                       |                                    |            |                | Trimethoprim                | 100       |            |
|                       |                                    |            |                | TCEP                        | 98        |            |
|                       |                                    |            |                | TCPP                        | 99        |            |
|                       |                                    |            |                | TDCP                        | 100       |            |
| Pharmaceutical        | OMBR, FO flat sheet (Hydration     | 2.7 steady | na             | Diclofenac                  | 98.4      | [102]      |
| wastewater            | Technology),                         |            |                | Naproxen                    | 98.3      |            |
|                       | 0.04 m² area, MLSS 7.2–8.1 g/L,     |            |                | Ibuprofen                   | 97.1      |            |
|                       | SRT-63-68 d, HRT 30 h, DS-0.5 NaCl  |            |                |                             |           |            |
|                       | SRT- 20 d, HRT-33 h (HTI)           |            |                |                             |           |            |
| 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 m² area, PRO mode, DS- 1.5 M NaCl, MLSS 3.4-3.7 g/L | 3 (Steady) 4.1 | | trimethoprim 32% | | |
| | | | | 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 | | |
| | | | | Diltiazem 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 | | |
| | | | | Trimeprin 33 | | |
| | | | | Ethinyl estradiol 99 | | |
| | | | | Diazinon 99 | | |
| | | | | Fluoxetine 99 | | |
| | | | | Triclocarban 97 | | |
| | | | | Clozapine 100 | | |
| | | | | Omeprazole 83 | | |
| | | | | Chlorpyrifos 99 | | |
| | | | | HydroxyazineEnalapril 99 | | |
| | | | | Risperidone 100 | | |
| | | | | Simvastatin 82 | | |
| | | | | Methotrexate 80 | | |
| | | | | Verapamil 91 | | |
| | | | | Simvastatin 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 (CO₂)) and nitrogen (e.g., nitrogen (N₂) and nitrous oxide (N₂O)) 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 clofibrate acid, fenoprofen, primidone, diclofenac, propoxur, carbamazepine, atrazine and ametrine. The authors further linked this to the recalcitrant EWGs (-Cl, -NO₂) in hydrophilic OMPs than EDGs (-NH₂ 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 clofibrate acid, fenoprofen, primidone, diclofenac, carbamazepine and atrazine showcased poor removals of less than 30% [104]. Luo et al. (2017) also observed that some hydrophilic OMPs such as clofibrate acid, fenoprofen, 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 Feed | MD Flux LMH | OMPs Investigated | % Removal | References |
|------------|----------------------------------|-----------------|-------------|-------------------|-----------|------------|
| Synthetic wastewater | MD-EMBR * reactor PTFE side stream MD | 30 | 10 | 4 (Steady flux) | Primidone 99, Ketoprofen 99, Naproxen 98, Gemfibrozil 98, Metronidazole 96, Diclofenac 99, Fenoprofen 98, Ibuprofen 94, Ametrine 99, Clotfibric 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-aceate bisphenol A 96, Salicylic acid 96, pentachlorophenol 97, Triclosan 97, 4-tert-Butylphenol 98 | [108] |

\[97\]
Table 5. Cont.

| Wastewater  | Membrane with Operating Conditions | Temperature | MD Flux LMH | OMPs Investigated | % Removal | References |
|-------------|------------------------------------|-------------|-------------|-------------------|-----------|------------|
| Synthetic wastewater | Membrane distillation with an enzymatic bioreactor (MD-EMBR) | 30 | 10 | 3.75 | >99% for all | [20] |
|            | PTFE side stream MD                 |             |             |                   |           |            |
|            | Pore size 0.22 µm, 95–100 µM (DMP)/min lacase, DO 3 mg/L |             |             |                   |           |            |
| Synthetic wastewater | AnMBR-MD                             | 45 | 20 |             |           |            |
|            | AnMBR MLSS 10 g/L, ceramic          |             |             |                   |           |            |
|            | MF pore size 1 µm, area 0.09 m², HRT |             |             |                   |           |            |
|            | 4 d, 0.3 to 0.5 L/g COD,            |             |             |                   |           |            |
|            | MD PTFE membrane, 0.2 µm pore size. |             |             |                   |           |            |
| Synthetic wastewater | PTFE side stream MD Bioreactor      | 40 | 14 |             |           |            |
|            | MLSS 5.3 g/L, pH 7.6, DO 2.8 mg/L, HRT 9.6 d, temperature 40 °C |             |             | 1.2 (Steady flux) | >95% removal |            |
|            | (Steady flux)                        |             |             |                   |           |            |

| OMPs | Removal |
|------|---------|
| Caffeine | 99 |
| Sulfamethoxazole | 89 |
| Ketoprofen | 99 |
| Trimethoprim | 99 |
| Paracetamol | 99 |
| Naproxen | 97 |
| Primidone | 99 |
| Ibuprofen | 99 |
| Triamterene | 98 |
| Carazolol | 97 |
| TCEP | 92 |
| Diclofenac | 75 |
| Carbamazepine | 90 |
| Gemfibrozil | 99 |
| Simazine | 79 |
| Amitriptyline | 99 |
| Atrazine | 74 |
| Diuron | 99 |
| Propylparaben | 91 |
| Linuron | 93 |
| Clozapine | 99 |
| Phenylphenol | 80 |
| bisphenol A | 85 |
| Diazinon | 99 |
| Triclosan | 85 |
| Triclocarban | 95 |

| OMPs | Removal |
|------|---------|
| Clofibric acid | 100 |
| Salicylic acid | 96 |
| Ketoprofen | 99 |
| Fenopro| 100 |
| Naproxen | 100 |
| Ibuprofen | 95 |
| Primidone | 98 |
| Diclofenac | 100 |
| Gemfibrozil | 96 |
| Propoxur | 97 |
| Carbamazepine | 98 |
| pentachlorophenol | 96 |
| Estriol | 96 |
| Atrazine | 99 |
| Ametryn | 99 |
| Benzophenone | 97 |
| Amitriptyline | 99 |
| 4-Tert-butyphenol | 99 |
| Oxybenzone | 100 |
| Estrone | 99 |
| 17α-Ethinylestradiol | 98 |
| 17β-Estradiol | 100 |
| Triclosan | 98 |
| 17β-Estradiol-17-acetate | 100 |
| Octocrylene | 97 |

References:
[20], [83], [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 cm², 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) | Clofibric acid 99 | [97] |
| | | | | 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 | |
| | | | | pantachlorophenol DEET 99 | |
| | | | | Estradiol 91 | |
| | | | | Atrazine 95 | |
| | | | | Ametrine 98 | |
| | | | | Amitriptyline 99 | |
| | | | | Benzophenone 98 | |
| | | | | 4-Tert-butyl phenol 99 | |
| | | | | Oxybenzone 99 | |
| | | | | Estrone 99 | |
| | | | | bisphenol A 93 | |
| | | | | 17α-Ethynylestradiol 98 | |
| | | | | 17β-Estradiol 98 | |
| | | | | Triclosan 99 | |
| | | | | 17β-Estradiol-17- acetate 98 | |
| | | | | Octocrylene 99 | |
| | | | | 98 | |

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 OMP's successfully to extract reclaimed wastewater [97]. OMPs possessing EWG (e.g., -Cl₂, -NH₂ and –NO₂) 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].
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, -NH2 and –NO2) 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].

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 laccase 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 laccasae 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, 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.

**Author Contributions:** Writing—original draft preparation, N.P.; Table and Figures preparation, V.H.T.; writing—review and editing, A.M.; M.A.H.J.; conceptualization and supervision, S.P.; H.S. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research was supported by a grant (code NRF-2017M1A2A2047369) from Climate Change Response Technology Development Program funded by National Research Foundation of Korea, Republic of Korea, the National Research Foundation of Korean Grant funded by the Korean Government (MSIP) (No. NRF-2015R1A5A7037825).

**Conflicts of Interest:** The authors declare no conflict of interest.

**References**

1. Tran, V.S.; Ngo, H.H.; Guo, W.; Zhang, J.; Liang, S.; Ton-That, C.; Zhang, X. Typical low cost biosorbents for adsorptive removal of specific organic pollutants from water. *Bioresour. Technol.* **2015**, *182*, 353–363. [CrossRef] [PubMed]

2. Luo, W.; Hai, F.I.; Price, W.E.; Elimelech, M.; Nghiem, I.D. Evaluating ionic organic draw solutes in osmotic membrane bioreactors for water reuse. *J. Membr. Sci.* **2016**, *514*, 636–645. [CrossRef]
3. Morrow, C.P.; Furtaw, N.M.; Murphy, J.R.; Achilli, A.; Marchand, E.A.; Hiibel, S.R.; Childress, A.E. Integrating an aerobic/anoxic osmotic membrane bioreactor with membrane distillation for potable reuse. *Desalination* 2018, 432, 46–54. [CrossRef]

4. Ma, X.Y.; Li, Q.; Wang, X.C.; Wang, Y.; Wang, D.; Ngo, H.H. Micropollutants removal and health risk reduction in a water reclamation and ecological reuse system. *Water Res.* 2018, 138, 272–281. [CrossRef]

5. Luo, W.; Pan, H.V.; Xie, M.; Hai, F.L.; Price, W.E.; Elimelech, M.; Nghiem, L.D. Osmotic versus conventional membrane bioreactors integrated with reverse osmosis for water reuse: Biological stability, membrane fouling, and contaminant removal. *Water Res.* 2017, 109, 122–134. [CrossRef]

6. Zhang, B.; Song, X.; Nghiem, L.D.; Li, G.; Luo, W. Osmotic membrane bioreactors for wastewater reuse: Performance comparison between cellulose triacetate and polyamide thin film composite membranes. *J. Membr. Sci.* 2017, 539, 383–391. [CrossRef]

7. Bodzek, M.; Konieczny, K. Membranes in organic micropollutants removal. *Current Org. Chem.* 2018, 22, 1070–1102. [CrossRef]

8. Hamza, R.A.; Iorhemen, O.T.; Tay, J.H. Occurrence, impacts and removal of emerging substances of concern from wastewater. *Environ. Technol. Innov.* 2016, 5, 161–175. [CrossRef]

9. Tran, N.H.; Gin, K.Y.-H. Occurrence and removal of pharmaceuticals, hormones, personal care products, and endocrine disrupters in a full-scale water reclamation plant. *Sci. Total Environ.* 2017, 599, 1503–1516. [CrossRef]

10. Priac, A.; Morin-Crini, N.; Druart, C.; Gavoille, S.; Bradu, C.; Lagarrigue, C.; Torri, G.; Winterton, P.; Crini, G. Alkylphenol and alkylphenol polyethoxylates in water and wastewater: A review of options for their elimination. *Arabian J. Chem.* 2017, 10, S3749–S3773. [CrossRef]

11. Lapworth, D.J.; Baran, N.; Stuart, M.E.; Ward, R.S. Emerging organic contaminants in groundwater: A review of sources, fate and occurrence. *Environ. Pollut.* 2012, 163, 287–303. [CrossRef] [PubMed]

12. Kosma, C.I.; Lambropoulou, D.A.; Albanis, T.A. Occurrence and removal of PPCPs in municipal and hospital wastewaters in Greece. *J. Hazard. Mater.* 2010, 179, 804–817. [CrossRef] [PubMed]

13. Curcio, E.; Profio, G.D.; Fontananova, E.; Drioli, E. Membrane technologies for seawater desalination and brackish water treatment. In *Advances in Membrane Technologies for Water Treatment: Materials, Processes and Applications*; Woodhead Publishing Series in Energy: Oxford, UK, 2015; pp. 411–441.

14. Wei, C.-H.; Wang, N.; HoppeJones, C.; Leiknes, T.; Amy, G.; Fang, Q.; Hu, X.; Rong, H. Organic micropollutants removal in sequential batch reactor followed by nanofiltration from municipal wastewater treatment. *Bioresour. Technol.* 2018, 268, 648–657. [CrossRef] [PubMed]

15. Pal, A.; He, Y.; Jekel, M.; Reinhard, M.; Gin, K.Y.-H. Emerging contaminants of public health significance as water quality indicator compounds in the urban water cycle. *Environ. Int.* 2014, 71, 46–62. [CrossRef]

16. Barbosa, M.O.; Moreira, N.F.F.; Ribeiro, A.R.; Pereira, M.F.R.; Silva, A.M.T. Occurrence and removal of organic micropollutants: An overview of the watch list of EU Decision 2015/495. *Water Res.* 2016, 94, 257–279. [CrossRef]

17. Petrie, B.; Barden, R.; Kasprzyk-Hordern, B. A review on emerging contaminants in wastewaters and the environment: Current knowledge, understudied areas and recommendations for future monitoring. *Water Res.* 2014, 72, 3–27. [CrossRef]

18. Zheng, W.; Wen, X.; Zhang, B.; Qiu, Y. Selective effect and elimination of antibiotics in membrane bioreactor of urban wastewater treatment plant. *Sci. Total Environ.* 2019, 646, 1293–1303. [CrossRef]

19. Song, H.L.; Yang, X.L.; Xia, M.Q.; Chen, M. Co-metabolic degradation of steroid estrogens by heterotrophic bacteria and nitrifying bacteria in MBRs. *J. Environ. Sci. Health Part A* 2017, 52, 778–784. [CrossRef]

20. Asif, M.B.; Nguyen, L.N.; Hai, F.L.; Price, W.E.; Nghiem, L.D. Integration of an enzymatic bioreactor with membrane distillation for enhanced biodegradation of trace organic contaminants. *Int. Biodeterior. Biodegrad.* 2017, 124, 73–81. [CrossRef]

21. Lloret, L.; Eibes, G.; Feijoo, G.; Moreira, M.T.; Lema, J.M. Degradation of estrogens by laccase from Myceliophthora thermophila in fed-batch and enzymatic membrane reactors. *J. Hazard. Mater.* 2012, 213–214, 175–183. [CrossRef]

22. Kidd, K.A.; Blanchfield, P.J.; Mills, K.H.; Palace, V.P.; Evans, R.E.; Lazorchak, J.M.; Flick, R.W. Collapse of a fish population after exposure to a synthetic estrogen. *Proc. Natl. Acad. Sci. USA* 2007, 104, 8897–8901. [CrossRef] [PubMed]
23. Enfrin, M.; Dumée, L.F.; Lee, J. Nano/microplastics in water and wastewater treatment processes – Origin, impact and potential solutions. *Water Res.* 2019, 161, 621–638. [CrossRef]
24. Cao, X.; Luo, J.; Woodley, J.M.; Wan, Y. Mussel-inspired co-deposition to enhance bisphenol A removal in a bifacial enzymatic membrane reactor. *Chem. Eng. J.* 2018, 336, 315–324. [CrossRef]
25. Ribaudo, M.; Bouzaher, A. Atrazine: Environmental Characteristics and Economics of Management. 1994; Available online: https://ideas.repec.org/p/ags/uerst/34011.html (accessed on 18 January 2020).
26. Alvarino, T.; Suarez, S.; Lema, J.; Omil, F. Understanding the sorption and biotransformation of organic micropollutants in innovative biological wastewater treatment technologies. *Sci. Total Environ.* 2018, 615, 297–306. [CrossRef] [PubMed]
27. Abegglen, C.; Joss, A.; McArdell, C.S.; Fink, G.; Schlüsener, M.P.; Ternes, T.A.; Siegrist, H. The fate of selected micropollutants in a single-house MBR. *Water Res.* 2009, 43, 2036–2046. [CrossRef]
28. Einsiedl, F.; Radke, M.; Maloszewski, P. Occurrence and transport of pharmaceuticals in a karst groundwater system affected by domestic wastewater treatment plants. *J. Contam. Hydrol.* 2010, 117, 26–36. [CrossRef]
29. Nguyen, L.N.; Hai, F.I.; Price, W.E.; Kang, J.; Leusch, F.D.; Roddick, F.; van de Merwe, J.P.; Magram, S.F.; Nghiem, L.D. Degradation of a broad spectrum of trace organic contaminants by an enzymatic membrane reactor: Complementary role of membrane retention and enzymatic degradation. *Int. Biodeterior. Biodegrad.* 2015, 99, 115–122. [CrossRef]
30. Sun, Y.; Huang, H.; Sun, Y.; Wang, C.; Shi, X.; Hu, H.; Kameya, T.; Fujie, K. Occurrence of estrogenic endocrine disrupting chemicals concern in sewage plant effluent. *Front. Environ. Sci. Eng.* 2014, 8, 18–26. [CrossRef]
31. Mukherjee, D.; Bhattacharya, P.; Jana, A.; Bhattacharya, S.; Sarkar, S.; Ghosh, S.; Majumdar, S.; Swarnakar, S. Synthesis of ceramic ultrafiltration membrane and application in membrane bioreactor process for pesticide remediation from wastewater. *Process Saf. Environ. Prot.* 2018, 116, 22–33. [CrossRef]
32. Bezbauhah, A.N.; Thompson, J.M.; Chisholm, B.J. Remediation of alachlor and atrazine contaminated water with zero-valent iron nanoparticles. *J. Environ. Sci. Health Part B Pestic. Food Contam. Agric. Wastes* 2019, 54, 518–524. [CrossRef]
33. Long, M.; EsraIlhan, Z.; Xia, S.; Zhou, C.; Rittmann, B.E. Complete dechlorination and mineralization of pentachlorophenol (PCP) in a hydrogen-based membrane biofilm reactor (MBfR). *Water Res.* 2018, 144, 134–144. [CrossRef] [PubMed]
34. Chtourou, M.; Mallek, M.; Dalmau, M.; Mamo, J.; Santos-Clotas, E.; Salah, A.B.; Walha, K.; Salvadó, V.; Montclus, H. Triclosan, carbamazepine and caffeine removal by activated sludge system focusing on membrane bioreactor. *Process Saf. Environ. Prot.* 2018, 118, 1–9. [CrossRef]
35. Goh, S.; Zhang, J.; Liu, Y.; Fane, A.G. Membrane distillation bioreactor (MDBR)—A lower greenhouse-gas (GHG) option for industrial wastewater reclamation. *Chemosphere* 2015, 140, 129–142. [CrossRef]
36. Holloway, R.W.; Regnery, J.; Nghiem, L.D.; Cath, T.Y. Removal of trace organic chemicals and performance of a novel hybrid ultrafiltration-osmotic membrane bioreactor. *Environ. Sci. Technol.* 2014, 48, 10859–10868. [CrossRef] [PubMed]
37. Nguyen, N.C.; Chen, S.S.; Nguyen, H.T.; Ray, S.S.; Ngo, H.H.; Guo, W.; Lin, P.H. Innovative sponge-based moving bed-osmotic membrane bioreactor hybrid system using a new class of draw solution for municipal wastewater treatment. *Water Res.* 2016, 91, 305–313. [CrossRef] [PubMed]
38. Singhal, N.; Perez-Garcia, O. Degrading organic micropollutants: The next challenge in the evolution of biological wastewater treatment processes. *Front. Environ. Sci.* 2016, 4, 36. [CrossRef]
39. Vásquez, E.; Trapote, A.; Prats, D. Elimination of pesticides with a membrane bioreactor and two different sludge retention times. *Tecnol. Y Cien. Del. Agua* 2018, 9, 198–212. [CrossRef]
40. Prasertkulak, S.; Chiemchaisri, C.; Chiemchaisri, W.; Yamamoto, K. Removals of pharmaceutical compounds at different sludge particle size fractions in membrane bioreactors operated under different solid retention times. *J. Hazard. Mater.* 2019, 368, 124–132. [CrossRef]
41. Kruglova, A.; Krákström, M.; Riska, M.; Mikola, A.; Rantanen, P.; Vahala, R.; Kronberg, L. Comparative study of emerging micropollutants removal by aerobic activated sludge of large laboratory-scale membrane bioreactors and sequencing batch reactors under low-temperature conditions. *Bioresour. Technol.* 2016, 214, 81–88. [CrossRef]
42. El Kateb, M.; Trellu, C.; Darwin, A.; Rivallin, M.; Bechelany, M.; Nagarajan, S.; Lacour, S.; Bellakhal, N.; Lesage, G.; Héran, M. Electrochemical advanced oxidation processes using novel electrode materials for mineralization and biodegradability enhancement of nanofiltration concentrate of landfill leachates. *Water Res.* **2019**, *162*, 446–455. [CrossRef]

43. Calero-Díaz, G.; Montecoliva-García, A.; Leyva-Díaz, J.C.; López-López, C.; Martín-Pascual, J.; Torres, J.C.; Poyatos, J.M. Impact of ciprofloxacin, carbamazepine and ibuprofen on a membrane bioreactor system: Kinetic study and biodegradation capacity. *J. Chem. Technol. Biotechnol.* **2017**, *92*, 2944–2951. [CrossRef]

44. Besha, A.T.; Gebreyohannes, A.Y.; Tufa, R.A.; Bekele, D.N.; Curcio, E.; Giorno, L. Removal of emerging micropollutants by activated sludge process and membrane bioreactors and the effects of micropollutants on membrane fouling: A review. *J. Environ. Chem. Eng.* **2017**, *5*, 2395–2414. [CrossRef]

45. Ibrahim, R.S.; Yuniarto, A.; Kamaruddin, S.N. The outlook on future MBR technologies. In *Sustainable Water Treatment: Innovative Technologies*; CRC Press LLC, Taylor & Francis Group: Boca Raton, FL, USA, 2017; pp. 81–92.

46. More, A. *Membrane Bioreactor (MBR) Market 2019 Global industry Size, Growth, Segments, Revenue, Manufacturers and 2025 Forecast Research Report*; The Express Wire: Riverside, CA, USA, 2019.

47. Hai, F.I.; Yamamoto, K.; Lee, C.-H. *Membrane Biological Reactors: Theory, Modeling, Design, Management and Applications to Wastewater Reuse*; Iwa Publishing: London, UK, 2018.

48. Mutamim, N.S.A.; Noor, Z.Z. Removal of micro-pollutants from wastewater through mbr technologies: A case study on spent caustic wastewater. In *Sustainable Water Treatment: Innovative Technologies*; CRC Press LLC, Taylor & Francis Group: Boca Raton, FL, USA, 2017; pp. 67–80. [CrossRef]

49. Yeo, B.J.L.; Goh, S.; Zhang, J.; Livingston, A.G.; Fane, A.G. Novel MBRs for the removal of organic priority pollutants from industrial wastewaters: A review. *J. Chem. Technol. Biotechnol.* **2015**, *90*, 1949–1967. [CrossRef]

50. Asif, M.B.; Ansari, A.J.; Chen, S.S.; Nghiem, L.D.; Price, W.E.; Hai, F.I. Understanding the mechanisms of trace organic contaminant removal by high retention membrane bioreactors: A critical review. *Environ. Sci. Pollut. Res.* **2019**, *26*, 34085–34100. [CrossRef]

51. Tran, N.H.; Urase, T.; Ngo, H.H.; Hu, J.; Ong, S.L. Insight into metabolic and cometabolic activities of autotrophic and heterotrophic microorganisms in the biodegradation of emerging trace organic contaminants. *Bioreor. Technol.* **2013**, *146*, 721–731. [CrossRef]

52. Mert, B.K.; Ozengin, N.; Dogan, E.C.; Aydiner, C. Efficient Removal Approach of Micropollutants in Wastewater Using Membrane Bioreactor. IntechOpen: London, UK, 2018; pp. 42–69.

53. Giwa, A.; Dindi, A.; Kujawa, J. Membrane bioreactors and electrochemical processes for treatment of wastewaters containing heavy metal ions, organics, micropollutants and dyes: Recent developments. *J. Hazard. Mater.* **2019**, *370*, 172–195. [CrossRef]

54. Mutamim, N.S.A.; Noor, Z.Z.; Hassan, M.A.A.; Yuniarto, A.; Olsson, G. Membrane bioreactor: Applications and limitations in treating high strength industrial wastewater. *Chem. Eng. J.* **2013**, *225*, 109–119. [CrossRef]

55. Radjenović, J.; Matošić, M.; Mijatović, I.; Petrović, M.; Barceló, D. Membrane bioreactor (MBR) as an advanced wastewater treatment technology. In *Handbook of Environmental Chemistry, Volume 5: Water Pollution*; Springer: Berlin/Heidelberg, Germany, 2018; Volume 5, S2, pp. 37–101.

56. Melin, T.; Jefferson, B.; Bixio, D.; Thoeeye, C.; De Wilde, W.; De Koning, J.; van der Graaf, J.; Wintgens, T. Membrane bioreactor technology for wastewater treatment and reuse. *Desalination* **2006**, *187*, 271–282. [CrossRef]

57. Cinar, O.; Kizile, A.; Isik, O.; Čemanovi, A.; Vera, M.A.; Duman, S. A review on dynamic membrane bioreactors: Comparison of membrane bioreactors and different support materials, transmembrane pressure. In *Proceedings of the International Conference on Engineering and Natural Sciences (ICENS)*, Sarajevo, Bosnia, 24–28 May 2016.

58. Wang, X.; Chang, V.W.C.; Tang, C.Y. Osmotic membrane bioreactor (OMBR) technology for wastewater treatment and reclamation: Advances, challenges, and prospects for the future. *J. Membr. Sci.* **2016**, *504*, 113–132. [CrossRef] [PubMed]

59. Judd, S. The status of membrane bioreactor technology. *Trends Biotechnol.* **2008**, *26*, 109–116. [CrossRef] [PubMed]

60. Wang, P.; Wang, Z.; Wu, Z.; Mai, S. Fouling behaviours of two membranes in a submerged membrane bioreactor for municipal wastewater treatment. *J. Membr. Sci.* **2011**, *382*, 60–69. [CrossRef]
61. Cornelissen, E.; Harmsen, D.; Dekorte, K.; Ruiken, C.; Qin, J.; Oo, H.; Wessels, L. Membrane fouling and process performance of forward osmosis membranes on activated sludge. J. Membr. Sci. 2008, 319, 158–168. [CrossRef]

62. Bui, X.T.; Vo, T.P.T.; Ngo, H.H.; Guo, W.S.; Nguyen, T.T. Multicriteria assessment of advanced treatment technologies for micropollutants removal at large-scale applications. Sci. Total Environ. 2016, 563–564, 1050–1067. [CrossRef] [PubMed]

63. Luo, Y.; Guo, W.; Ngo, H.H.; Nghiem, L.D.; Hai, F.I.; Zhang, J.; Liang, S.; Wang, X.C. A review on the occurrence of micropollutants in the aquatic environment and their fate and removal during wastewater treatment. Sci. Total Environ. 2014, 473–474, 619–641. [CrossRef]

64. Fonseca Couto, C.; Lange, L.C.; Santos Amaral, M.C. A critical review on membrane separation processes applied to remove pharmaceutically active compounds from water and wastewater. J. Water Process Eng. 2018, 26, 156–175. [CrossRef]

65. Rodriguez, F.A.; Poyatos, J.M.; Rebollo-Oriva, P.; Osorio, F.; González-López, J.; Hontoria, E. Kinetic study and oxygen transfer efficiency evaluation using respirometric methods in a submerged membrane bioreactor using pure oxygen to supply the aerobic conditions. Bioreour. Technol. 2011, 102, 6013–6018. [CrossRef]

66. Ahmed, M.B.; Zhou, J.L.; Ngo, H.H.; Guo, W.; Thomaidis, N.S.; Xu, J. Progress in the biological and chemical treatment technologies for emerging contaminant removal from wastewater: A critical review. J. Hazard. Mater. 2017, 323, 274–298. [CrossRef]

67. Luo, Y.; Jiang, Q.; Ngo, H.H.; Nghiem, L.D.; Hai, F.I.; Price, W.E.; Wang, J.; Guo, W. Evaluation of micropollutant removal and fouling reduction in a hybrid moving bed biofilm reactor-membrane bioreactor system. Bioreour. Technol. 2015, 191, 355–359. [CrossRef] [PubMed]

68. Park, J.; Yamashita, N.; Park, C.; Shimono, T.; Takeuchi, D.M.; Tanaka, H. Removal characteristics of pharmaceuticals and personal care products: Comparison between membrane bioreactor and various biological treatment processes. Chemosphere 2017, 179, 347–358. [CrossRef]

69. Alvarino, T.; Torregrosa, N.; Omil, F.; Lema, J.M.; Suarez, S. Assessing the feasibility of two hybrid MBR systems using PAC for removing macro and micropollutants. J. Environ. Manag. 2017, 203, 831–837. [CrossRef]

70. Phan, H.V.; Hai, F.I.; McDonald, J.A.; Khan, S.J.; Zhang, R.; Price, W.E.; Broeckmann, A.; Nghiem, L.D. Nutrient and trace organic contaminant removal from wastewater of a resort town: Comparison between a pilot and a full scale membrane bioreactor. Int. Biodeterior. Biodegrad. 2015, 102, 40–48. [CrossRef]

71. Prasertkulaks, S.; Chiemchaits, C.; Chiemchaits, W.; Tonaga, T.; Yamamoto, K. Removals of pharmaceutical compounds from hospital wastewater in membrane bioreactor operated under short hydraulic retention time. Chemosphere 2016, 150, 624–631. [CrossRef]

72. Hamon, P.; Moulin, P.; Ercole, L.; Marrot, B. Oncological ward wastewater treatment by membrane bioreactor: Acclimation feasibility and pharmaceuticals removal performances. J. Water Process Eng. 2018, 21, 9–26. [CrossRef]

73. Abargues, M.R.; Robles, A.; Bouzas, A.; Seco, A. Micropollutants removal in an anaerobic membrane bioreactor and in an aerobic conventional treatment plant. Water Sci. Technol. 2012, 65, 2242–2250. [CrossRef]

74. Wijekoon, K.C.; Hai, F.I.; Kang, J.; Price, W.E.; Guo, W.; Ngo, H.H.; Cath, T.Y.; Nghiem, L.D. A novel membrane distillation–thermophilic bioreactor system: Biological stability and trace organic compound removal. Bioreour. Technol. 2014, 159, 334–341. [CrossRef] [PubMed]

75. Luo, W.; Hai, F.I.; Kang, J.; Price, W.E.; Nghiem, L.D.; Elimelech, M. The role of forward osmosis and microfiltration in an integrated osmotic-microfiltration membrane bioreactor system. Chemosphere 2015, 136, 125–132. [CrossRef] [PubMed]

76. Alvarino, T.; Suarez, S.; Lema, J.; Omil, F. 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. [CrossRef] [PubMed]

77. Arola, K.; Hatakka, H.; Mänttäri, M.; Kallioinen, M. Novel process concept alternatives for improved removal of micropollutants in wastewater treatment. Sep. Purif. Technol. 2017, 186, 333–341. [CrossRef]
79. Sahar, E.; Messalem, R.; Cikurel, H.; Aharoni, A.; Brenner, A.; Godehardt, M.; Jekel, M.; Ernst, M. Fate of antibiotics in activated sludge followed by ultrafiltration (CAS-UF) and in a membrane bioreactor (MBR). Water Res. 2011, 45, 4827–4836. [CrossRef]
80. Alvarino, T.; Allegue, T.; Fernandez-Gonzalez, N.; Suarez, S.; Lema, J.M.; Garrido, J.M.; Omil, F. Minimization of dissolved methane, nitrogen and organic micropollutants emissions of effluents from a methanogenic reactor by using a preanoxic MBR post-treatment system. Sci. Total Environ. 2019, 671, 165–174. [CrossRef]
81. Song, X.; Luo, W.; Hai, F.I.; Price, W.E.; Guo, W.; Ngo, H.H.; Nghiem, L.D. Resource recovery from wastewater by anaerobic membrane bioreactors: Opportunities and challenges. Bioresour. Technol. 2018, 270, 669–677. [CrossRef] [PubMed]
82. Neoh, C.H.; Noor, Z.Z.; Mutamim, N.S.A.; Lim, C.K. Green technology in wastewater treatment technologies: Integration of membrane bioreactor with various wastewater treatment systems. Chem. Eng. J. 2016, 283, 582–594. [CrossRef]
83. Song, X.; Luo, W.; McDonald, J.; Khan, S.J.; Hai, F.I.; Price, W.E.; Nghiem, L.D. 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, 628–629, 358–365. [CrossRef]
84. Guo, W.; Ngo, H.H.; Chen, C.; Pandey, A.; Tung, K.-L.; Lee, D.-J. Anaerobic membrane bioreactors for future green bioprocesses. In Green Technologies for Sustainable Water Management; Cahter 25; American Society of Civil Engineers: Reston, VI, USA, 2016; pp. 867–901.
85. Lin, H.; Peng, W.; Zhang, M.; Chen, J.; Hong, H.; Zhang, Y. A review on anaerobic membrane bioreactors: Applications, membrane fouling and future perspectives. Desalination 2013, 314, 169–188. [CrossRef]
86. Alvarino, T.; Suárez, S.; Garrido, M.; Lema, J.; Omil, F. A UASB reactor coupled to a hybrid aerobic MBR as innovative plant configuration to enhance the removal of organic micropollutants. Chemosphere 2016, 144, 452–458. [CrossRef]
87. Wijekoon, K.C.; McDonald, J.A.; Khan, S.J.; Hai, F.I.; Price, W.E.; Nghiem, L.D. Development of a predictive framework to assess the removal of trace organic chemicals by anaerobic membrane bioreactor. Bioresour. Technol. 2015, 189, 391–398. [CrossRef]
88. Song, X.; Luo, W.; McDonald, J.; Khan, S.J.; Hai, F.I.; Guo, W.; Ngo, H.H.; Nghiem, L.D. Effects of sulphur on the performance of an anaerobic membrane bioreactor: Biological stability, trace organic contaminant removal, and membrane fouling. Bioresour. Technol. 2018, 250, 171–177. [CrossRef]
89. Monsalvo, V.M.; McDonald, J.A.; Khan, S.J.; Le-Clech, P. Removal of trace organics by anaerobic membrane bioreactors. Water Res. 2014, 49, 103–112. [CrossRef]
90. Song, X.; McDonald, J.; Price, W.E.; Khan, S.J.; Hai, F.I.; Ngo, H.H.; Guo, W.; Nghiem, L.D. Effects of salinity build-up on the performance of an anaerobic membrane bioreactor regarding basic water quality parameters and removal of trace organic contaminants. Bioresour. Technol. 2016, 216, 399–405. [CrossRef]
91. Hai, F.I.; Li, X.; Price, W.E.; Nghiem, L.D. Removal of carbamazepine and sulfamethoxazole by MBR under anoxic and aerobic conditions. Bioresour. Technol. 2011, 102, 10386–10390. [CrossRef]
92. Luo, W.; Phan, H.V.; Hai, F.I.; Price, W.E.; Guo, W.; Ngo, H.H.; Yamamoto, K.; Nghiem, L.D. Effects of salinity build-up on the performance and bacterial community structure of a membrane bioreactor. Bioresour. Technol. 2016, 200, 305–310. [CrossRef] [PubMed]
93. Phan, H.V.; Hai, F.I.; Kang, J.; Dam, H.K.; Zhang, R.; Price, W.E.; Broeckmann, A.; Nghiem, L.D. Simultaneous nitrification/denitrification and trace organic contaminant (TrOC) removal by an anoxic–aerobic membrane bioreactor (MBR). Bioresour. Technol. 2014, 165, 96–104. [CrossRef] [PubMed]
94. Tadkaew, N.; Hai, F.I.; McDonald, J.A.; Khan, S.J.; Nghiem, L.D. Removal of trace organics by MBR treatment: The role of molecular properties. Water Res. 2011, 45, 2439–2451. [CrossRef] [PubMed]
95. Luo, W.; Hai, F.I.; Price, W.E.; Guo, W.; Ngo, H.H.; Yamamoto, K.; Nghiem, L.D. High retention membrane bioreactors: Challenges and opportunities. Bioresour. Technol. 2014, 167, 539–546. [CrossRef]
96. Achilli, A.; Cath, T.Y.; Marchand, E.A.; Childress, A.E. The forward osmosis membrane bioreactor: A low fouling alternative to MBR processes. Desalination 2009, 239, 10–21. [CrossRef]
97. Luo, W.; Phan, H.V.; Li, G.; Hai, F.I.; Price, W.E.; Elimelech, M.; Nghiem, L.D. An osmotic membrane bioreactor-membrane distillation system for simultaneous wastewater reuse and seawater desalination: Performance and implications. Environ. Sci. Technol. 2017, 51, 14311–14320. [CrossRef]
98. Holloway, R.W.; Achilli, A.; Cath, T.Y. The osmotic membrane bioreactor: A critical review. Environ. Sci. Water Res. Technol. 2015, 1, 581–605. [CrossRef]

99. Lu, Y.; He, Z. Mitigation of salinity buildup and recovery of wasted salts in a hybrid osmotic membrane bioreactor-electrodialysis system. Environ. Sci. Technol. 2015, 49, 10529–10535. [CrossRef]

100. Pathak, N.; Li, S.; Kim, Y.; Chekli, L.; Phuntsho, S.; Jang, A.; Ghaffour, N.; Leiknes, T.; Shon, H.K. Assessing the removal of organic micropollutants by a novel baffled osmotic membrane bioreactor-microfiltration hybrid system. Bioresour. Technol. 2018, 262, 98–106. [CrossRef] [PubMed]

101. Zhu, W.; Wang, X.; She, Q.; Li, X.; Ren, Y. Osmotic membrane bioreactors assisted with microfiltration membrane for salinity control (MF-OMBR) operating at high sludge concentrations: Performance and implications. Chem. Eng. J. 2018, 337, 576–583. [CrossRef]

102. Lay, W.C.L.; Zhang, Q.; Zhang, J.; McDougald, D.; Tang, C.; Wang, R.; Liu, Y.; Fane, A.G. Effect of pharmaceuticals on the performance of a novel osmotic membrane bioreactor (OMBR). Sep. Sci. Technol. 2012, 47, 543–554. [CrossRef]

103. Alturki, A.; McDonald, J.; Khan, S.J.; Hai, F.I.; Price, W.E.; Nghiem, L.D. Performance of a novel osmotic membrane bioreactor (OMBR) system: Flux stability and removal of trace organics. Bioresour. Technol. 2012, 113, 201–206. [CrossRef] [PubMed]

104. Luo, W.; Xie, M.; Song, X.; Guo, W.; Ngo, H.H.; Zhou, J.L.; Nghiem, L.D. Biomimetic aquaporin membranes for osmotic membrane bioreactors: Membrane performance and contaminant removal. Bioresour. Technol. 2018, 249, 62–68. [CrossRef] [PubMed]

105. Curcio, E.; Drioli, E. Membrane distillation and related operations—A review. Sep. Purif. Rev. 2005, 34, 35–86. [CrossRef]

106. Phattaranawik, J.; Fane, A.G.; Pasquier, A.C.S.; Bing, W. A novel membrane bioreactor based on membrane distillation. Desalination 2008, 223, 386–395. [CrossRef]

107. Qin, L.; Zhang, Y.; Xu, Z.; Zhang, G. Advanced membrane bioreactors systems: New materials and hybrid process design. Bioresour. Technol. 2018, 269, 476–488. [CrossRef] [PubMed]

108. Asif, M.B.; Hai, F.I.; Kang, J.; Van De Merwe, J.P.; Leusch, F.D.; Price, W.E.; Nghiem, L.D. Biocatalytic degradation of pharmaceuticals, personal care products, industrial chemicals, steroid hormones and pesticides in a membrane distillation-enzymatic bioreactor. Bioresour. Technol. 2018, 247, 528–536. [CrossRef] [PubMed]

109. Roos, V.; Gunnarsson, L.; Fick, J.; Larsson, D.G.J.; Rudén, C. Prioritising pharmaceuticals for environmental risk assessment: Towards adequate and feasible first-tier selection. Sci. Total Environ. 2012, 421–422, 102–110. [CrossRef] [PubMed]

110. Ojajuni, O.; Saroj, D.; Cavalli, G. Removal of organic micropollutants using membrane-assisted processes: A review of recent progress. Environ. Technol. Rev. 2015, 4, 17–37. [CrossRef]

111. Huang, L.; Lee, D.J. Membrane bioreactor: A mini review on recent R&D works. Bioresour. Technol. 2015, 194, 383–388. [CrossRef] [PubMed]

112. Bodzek, M. Membrane technologies for the removal of micropollutants in water treatment. In Advances in Membrane Technologies for Water Treatment: Materials, Processes and Applications; Woodhead Publishing Series in Energy: Oxford, UK, 2015; pp. 465–517. [CrossRef]

113. Pathak, N.; Chekli, L.; Wang, J.; Kim, Y.; Phuntsho, S.; Li, S.; Ghaffour, N.; Leiknes, T.; Shon, H. Performance of a novel baffled osmotic membrane bioreactor-microfiltration hybrid system under continuous operation for simultaneous nutrient removal and mitigation of brine discharge. Bioresour. Technol. 2017, 240, 50–58. [CrossRef] [PubMed]

114. Tran, V.H.; Lim, S.; Han, D.S.; Pathak, N.; Akther, N.; Phuntsho, S.; Park, H.; Shon, H.K. Efficient fouling control using outer-selective hollow fiber thin-film composite membranes for osmotic membrane bioreactor applications. Bioresour. Technol. 2019, 282, 9–17. [CrossRef] [PubMed]

115. Ensano, B.M.B.; Borea, L.; Naddeo, V.; de Luna, M.D.G.; Belgiorno, V. Control of emerging contaminants by the combination of electrochemical processes and membrane bioreactors. Environ. Sci. Pollut. Res. 2019, 26, 1103–1112. [CrossRef]

116. Lay, W.C.; Liu, Y.; Fane, A.G. Impacts of salinity on the performance of high retention membrane bioreactors for water reclamation: A review. Water Res. 2010, 44, 21–40. [CrossRef] [PubMed]
117. Chekli, L.; Phuntsho, S.; Shon, H.K.; Vigneswaran, S.; Kandasamy, J.; Chanan, A. A review of draw solutes in forward osmosis process and their use in modern applications. Desalin. Water Treat. 2012, 43, 167–184. [CrossRef]

118. Nghiem, L.D.; Schäfer, A.I.; Elimelech, M. Removal of natural hormones by nanofiltration membranes: Measurement, modeling and mechanisms. Environ. Sci. Technol. 2004, 38, 1888–1896. [CrossRef]

© 2020 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).