Chapter 3

Efficient Removal Approach of Micropollutants in Wastewater Using Membrane Bioreactor

Berna Kırıl Mert, Nihan Ozengin,
Esra Can Dogan and Coskun Aydıner

Additional information is available at the end of the chapter

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Abstract

In the recent past years, micropollutants that are pharmaceutically active compounds (PhACs) have been used extensively and have been discovered in raw sewage, wastewater treatment plants, effluents, surface, and groundwater with concentrations from ng/L to several μg/L. Even though many of these compounds are still not determined online, monitoring technology improvements progressed. Today's wastewater treatment plants are not constructed to remove these micropollutants yet. Conventional activated sludge processes are used in the treatment of municipal wastewater but are not specifically designed for the removal of micropollutants. The remaining pharmaceuticals mix into surface waters. At that stage, they can adversely affect the aquatic environment and may cause issues for drinking water production. As the conventional methods are insufficient for removing the micropollutants, other alternative treatment methods can be applied such as coagulation-flocculation, activated carbon adsorption (powdered activated carbon and granular activated carbon), advanced oxidation processes, membrane processes, and membrane bioreactor. It has been observed that membrane bioreactor (MBR) can achieve higher and more consistent micropollutants removal. The removal of micropollutants is based on physicochemical properties of micropollutants and the conditions of treatment. Due to recent technical innovations and cost reductions of the actual membranes, the membrane bioreactor takes attention. In this study, membrane bioreactor experiments for micropollutants in drinking use, wastewater, and surface waters were investigated in detail based on literature investigations, and the feasibility of this method was evaluated.

Keywords: wastewater, removal, micropollutant, membrane bioreactor

1. Introduction

Pharmaceutical wastewater is one of the most important gateways of emerging pollutants (such as synthetic hormones including corticosteroids) to enter water bodies. During the last years, numerous studies have documented the presence of many of these substances at the level of microgram or nanogram per liter in raw water (i.e., stream/source water), in wastewater effluents, and even in finished drinking waters [1, 2]. As a consequence, pharmaceuticals are entering in the trophic chain and causing adverse ecological and human health effects [3].

Pharmaceuticals are not regulated at the moment in the EU, but the 2013 amendment of the Environmental Quality Standards Directive (2008/105/EC) contains a mechanism to collect high-quality data on concentration of compounds of environmental concern, the so-called watchlist. This list includes diclofenac, 17-beta-estradiol (E2), and 17-alpha-ethinylestradiol (EE2). For compounds on this list, it is likely that regulations will be developed in the future. This would mean that additional treatment of wastewater will be necessary to comply with these regulations [4].

Membrane bioreactor (MBR) technique is a promising alternative to conventional treatment, [5, 6], and its usage is increasingly for municipal wastewater treatment and reuse, and great concerns have been raised to some emerging trace pollutants found in aquatic environment in the last decade, notably the pharmaceuticals [7]. In that sense, recently a pilot MBR was innovatively applied leading to removal efficiencies over 95% of the chemical oxygen demand (COD). Furthermore, other lab-scale MBR studies have been focused not only in the removal of the bulk organic matter but also in the elimination of the specific organic micropollutants present in the raw wastewater [1].

In this study, we present a comprehensive review of the studies carried out in the literature with MBR of micropollutant residues in different wastewaters, and it is expected that these pollutants, which are highly biologically active and difficult to biodegrade, shed light on treatment strategies to improve biodegradation.

2. Sources of pharmaceutical micropollutants in the aquatic environment

Pharmaceuticals are important and indispensable elements of modern life. They are used in humans and animals, in agriculture and in water culture. The presence of pharmaceuticals in the environment first attracted the attention of the scientific community and the public in the 1970s. However, until the 1990s, little has been done about the presence, behavior, and effects of pharmaceuticals in the environment. During this time, environmental pollutants such as heavy metals, polycyclic hydrocarbons, dioxins, furans, pesticides, and detergents have been extensively studied. Endocrine system drugs and lipid-lowering drugs have been on the rise since the 1990s. After this date, many studies have been done in the USA and Europe for hormones and other pharmaceuticals [8–10].

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An important reason why so much care is taken with pharmaceutical products is that they have to produce a biological effect. They are made as stable as possible so that they can be stored for a long time and easily swallowed. The membranes are lipophilic enough to cross the membranes, and in order to reach the sites of action—especially those taken orally—drugs must be resistant to enzymes and must not hydrolyze at acidic pH values. They must be stable and have high mobility in liquid phase [11–13].

Because of these properties, active pharmaceutical ingredients/conversion products can be bioaccumulated and can cause effects in aquatic or terrestrial ecosystems.

The intake of drug active substances occurs in various ways. Starting from humans and animals, the active pharmaceutical ingredients reach the wastewater, soil, and groundwater and, if adequate treatment is not done, reach our drinking water. Pharmaceutical products can be roughly divided into two: medicinal products and veterinary drugs used by humans. Veterinary medicines are used in farm animal breeding and poultry production. Medicinal products used by humans reach sewage through urea and feces and from there to wastewater treatment plant. If xenobiotics are taken as an example, there are three possible behaviors of the substance: (i) the substance is completely mineralized to water and CO_2 (e.g., aspirin). (ii) The substance is lipophilic and does not easily fragment. So, some of the material is kept in clay. (iii) The substance is metabolized to a more hydrophobic than lipophilic form but becomes resistant. It cannot be removed in the treatment plant, and it is thrown away with wastewater and mixed with the receiving waters. If the metabolites are still biologically active, they also affect the aquatic organisms in the environment. Possible materials in clay, if the mud is laid on the field, may affect microorganisms and the useful ones. Medicinal substances used to support growth of animals in the stables are mostly fertile. These substances can affect soil organisms. The hydrophilic materials in the sewage sludge, which are scattered in the mouth, reach the aquatic environment by infiltrating with rain [11–13].

Pharmaceutical substances used for animals in the field are thrown directly to the ground via urea and feces. High local concentration affects soil organisms. It is also possible that medicinal substances spread over the surface are mineralized to the ground or reach the groundwaters.

They are used in fish farms and are directly confused with the receiving waters because the best way to treat fish with antibiotics and other medicines is to use feed additives. Because most of the feed additives are not eaten by the fish, they fall from the cages and accumulate in the seabed. These substances can affect aquatic organisms. An unknown part of the medical goods sold for human use is thrown into the toilet as waste by people and reaches the treatment plant by interfering with the sewage system [14–17].

Micropollutants consist of a vast and expanding array of anthropogenic as well as natural substances. These include pharmaceuticals, personal care products, steroid hormones, industrial chemicals, pesticides, and many other emerging compounds. Micropollutants are commonly present in waters at trace concentrations, ranging from a few ng/L to several μg/L. The "low concentration" and diversity of micropollutants not only complicate the associated detection and analysis procedures but also create challenges for water and wastewater treatment processes [2].

Sources of micropollutants in the environment are diverse, and many of these originate from mass-produced materials and commodities. **Table 1** summarizes the sources of the major categories of micropollutants in the aquatic environment.

Pharmaceuticals are thoroughly used to cure the diseases in humans and as veterinary drugs. These biologically active chemicals are treated as emerging contaminant due to their persistence and potential harmful impact on the aquatic ecosystem.

These refractory emerging contaminants (RECs) (analgesics, anti-inflammatories, antiepileptics, and antibiotics) fall into the class of endocrine-disrupting compounds, which continually enters into the aquatic environment in small concentration.

They remain active even in low concentrations and deteriorate water quality and have an adverse impact on the ecosystem and human health. The most common and persistent pharmaceutical products in the aquatic environment are summarized below.

2.1. Antibiotics

In recent years, global consumption and the use of antibiotics increase to >30% [18]. Antibiotics are generally treated as pseudo-persistent compound because of its continuous introduction in environment. The existence and release of antibiotics are inclined to be of specific concern since

Category	Important subclasses	Major sources	Nonexclusive
Pharmaceuticals	[a]NSAIDs, lipid regulator, anticonvulsants, antibiotics, β-blockers, and stimulants	Domestic wastewater (from excretion) Hospital effluents Runoff from [b]CAFOs and aquaculture	Sources that are not exclusive to individual categories include industrial wastewater (from product manufacturing discharges) Landfill leachate (from improper disposal of used, defective, or expired items)
Personal care products	Fragrances, disinfectants, UV filters, and insect repellents	Domestic wastewater (from bathing, shaving, spraying, swimming, etc.)	
Steroid hormones	Estrogens	Domestic wastewater (from excretion) Runoff from CAFOs and aquaculture	
Surfactants	Nonionic surfactants	Domestic wastewater (from bathing, laundry, dishwashing, etc.)	
Industrial chemicals Pesticides	Plasticizers, fire retardants Insecticides, herbicides, and fungicides	Industrial wastewater (from industrial cleaning discharges) Domestic wastewater (by leaching out of the material) Domestic wastewater (from improper cleaning, runoff from gardens, lawns, roadways, etc.) Agricultural runoff	

[a]NSAIDs, Nonsteroidal anti-inflammatory drugs.
[b]CAFOs, concentrated animal feeding operations.

Table 1. Sources of micropollutants in the aquatic environment.

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they are designed to kill and inhibit the growth of microorganism; thus, they will hinder the activity of beneficial microbes in wastewater treatment plant (WWTP) operation and involved in their removal. Moreover, for constant exposure to antibiotics, microbial community stay in wastewater improves resistant mechanism more readily than the rest of another microbial world. The presence of numerous antibiotic compounds was identified in untreated wastewater in both aqueous and solid phases. Overall, occurrence and persistence of antibiotics in water bodies increase concern; almost 90% of antibiotics consumed by human body were discharged via urine and feces [19].

2.2. Therapeutic hormones

Therapeutic hormones are the synthetic analog of animal or plant natural hormones, which affect the endocrine system and have impacts on human and animal health. The most frequently found hormones in the environment are estrogens. A synthetic estrogenic steroid is used as a birth control agent and in estrogen substitution therapies. Thus, estrogen and its metabolite become the abundant class of emerging pharmaceutical contaminants. The metabolite of 17b-ethinyl estradiol and estrone (E1) is one of the most powerful EDCs creating impacts in aquatic organisms. Their presence in the river environment causes adverse reproductive and developmental effect in nontargeted organisms [20]. Several studies confirmed that the presence of estrogen in both influent and effluent of municipal wastewater treatment plants at a concentration ranges from 5 to 188 ng/L and between 0.3 and 12.6 ng/L, respectively [19, 21].

2.3. Analgesic pharmaceuticals

Analgesic is the widely used drug for pain relaxation and to treat fever. Drugs belonging to the class of analgesics such as naproxen acetaminophen, ibuprofen, diclofenac, and meprobamate were treated as significant environment pollutants due to their persistence in the aquatic environment [22]. Almost, 15% of ibuprofen was excreted after administration and 26% as its metabolite. The metabolite of ibuprofen is more toxic to aquatic organisms than parental compound [23]. The presence of ibuprofen, diclofenac, naproxen, gemfibrozil, and hydrochlorothiazide in the river shows a concentration range from 2 to 18 ng/L. The occurrence of these xenobiotic compounds in natural water bodies represents a significant concern for human health as little information is available on the effect of long-term ingestion of these compounds through drinking water [19].

2.4. By-product and metabolites

Pharmaceutical compounds pass on a set of biochemical transformation in human and animal body and form polar, hydrophilic, and biologically active metabolites, which are discharged through urine and feces and enter WWTP. These active metabolites are accumulated in tissues of aquatic organisms. They have the potential to bind covalently to their cellular protein and may evoke an immune response or exert toxic effects [25]. These metabolites are reported to be 50% more toxic than their parental compounds. The poorly metabolized parental pharmaceutical substances undergo a transformation and affect the action of microbial community present in

the WWTP. These metabolites are persistent due to their weaker sorption potential and high mobility and, thus, detected in environmental samples [26].

Literature reported that the concentration of the metabolite in influent and effluent of WWTP is often higher than their parental compounds, and their fate depends on the environmental conditions such as salinity, temperature, pH, and microbial diversity [19, 27].

Many studies on removal of pharmaceutical compounds from wastewater have been conducted, and many treatment technologies of hospital wastewater treatment have been developed.

Treatment of pharmaceutical residues using MBR processes was discussed in the following sections.

3. General features of MBR systems

Membranes have been used for many years as biological treatment (aerobic and anaerobic) and solid–liquid separation methods in physical applications. Nowadays, these methods are increasingly attracted to the name of membrane bioreactors combined with biological wastewater treatment [28]. Membrane bioreactor technology is emerging as a mature technology around the world with many full-scale installations for municipal and different wastewater treatments [29–31]. The reactor is operated in a similar manner to a conventional activated sludge process, and there is no need for tertiary stages such as secondary purification and sand filtration. Low-pressure membrane filters such as microfiltration (MF) or ultrafiltration (UF) are used to separate wastewater from the activated sludge [32].

Several factors have been reported that may affect contamination in MBR membrane properties such as floc size, mixed liquid viscosity, mixed liquid viscosity, pH, solubility, associated polymeric compounds (EPS), pore size, porosity, surface charge, roughness, and hydrophilicity/hydrophobicity. Operating parameters such as hydraulic retention time (HRT), solid retention time (SRT), and food/mass (F/M) ratio do not have a direct effect on membrane contamination [33, 34]. They affect more sludge properties and therefore sludge filtration properties. Organic contamination is caused by contamination of the membrane during active sludge filtration compared to inorganic pollution [35].

3.1. MBR configuration

There are two membrane-type alternatives: the first option is submerged MBR configuration such as operating under a vacuum, instead of direct pressure. This configuration may be named immersed as the membrane is placed directly into the liquid. The second option is sidestream MBR configuration such as operating under pressure. In this approach, the membrane is separated from the bioreactor, and a pump is required for pushing the bioreactor effluent into the membrane system and permeates through the membrane. This configuration may be named external cross flow membrane. Flat sheet (FS) and hollow fiber (HF) membranes are generally used for submerged MBR configuration [36]. The two main MBR configurations involve either submerged membranes or external circulation (sidestream configuration) (**Figure 1**) [32].

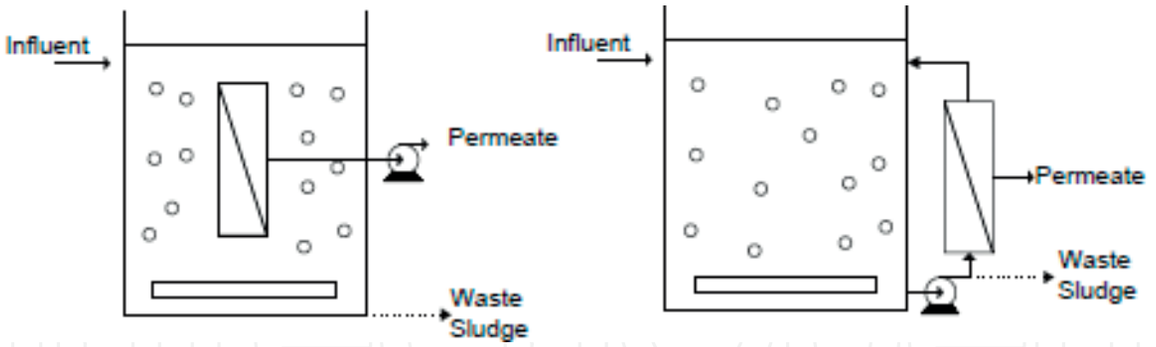

Figure 1. Configuration of MBR systems: (a) submerged (immersed) MBR and (b) sidestream (external) MBR configuration (adapted from [32, 37]).

Since submerged MBRs operate at lower operating fluxes, they have greater hydraulic efficiency due to greater permeability. Working with low flux is important in submerged MBR because this application minimizes membrane contamination or plugging. Membrane blockage is one of the major disadvantages of MBRs and requires cleaning mechanisms that increase cost and make operation difficult. While submerged MBRs require lower pumping costs than external MBRs, they require more aeration. The reason is that the aeration is the main method to prevent membrane clogging. In addition, low flux studies in submerged MBRs require more membrane surface area (and hence greater initial investment cost) when based on constant permeate flux production. Despite these disadvantages, however, the selected and implemented configuration for medium- and large-scale municipal wastewater treatment is the internal submerged MBR [38].

By the year 1990s, this existing accumulation has been rapidly increased by the MBR applications which are made as academic and field studies. MBR producers are Kubota from Japan, Zenon from Canada, Mitsubishi Rayon, and US Filtration [36, 39, 40] (**Table 2**).

Items	Zenon	Mitsubishi Rayon	Tianjin Motimo	Kubota[b]	Shanghai Zizheng
(1) Membrane module properties					
Polymer	PVDF	PE	PVDF	PE	PVDF
Filtration type	UF	MF	MF	MF	MF
Module	Hollow fiber	Hollow fiber	Hollow fiber	Flat sheet	Flat sheet
Hydrophilic	Yes	Yes	Yes	Yes	Yes
Outside diameter (mm)	1.95	—	1.00	490 (width)	460 (width)
Inside diameter (mm)	0.92	—	0.65	1000 (height)	1010 (height)
Fiber length (mm)	1650	663.5	1010	6 (thickness)	7 (thickness)
Pore size (μm)	0.04	0.4	0.2	0.4	0.2
Surface area (m^2)	23/module	105/module	20/module	0.8/panel	0.7/panel
Normal flux ($L/(m^2h)$)	25.5	10.3–16.7	15	25.5	20–30

Items	Zenon	Mitsubishi Rayon	Tianjin Motimo	Kubota[b]	Shanghai Zizheng
(2) MBR performance					
[a]MLSS (g/L)	12–30		<15	15–30	10–30
Aeration per module (m^3/h)	14	57–73	—		0.6/panel
SRT (d)	10–100		<60	>40	40
Sludge yield (kg MLSS/kg BOD)	0.1–0.3		—		0.26
BOD effluent (mg/L)	<2	2–6	—	3–5	
NH_3 effluent (mg/L)	<0.3		—	<2	<2
Cleaning method	Back pulse and relax	Relax	—	Relax	
Cleaning frequency (min/min)	0.5/15	2/12		1/60	
Recovery method	Chemical soak	Chlorine backwash		Chlorine backwash	
Recovery frequency	≥3 months	≥3 months		≥6 months	
Recovery location	Drained cell or in situ	In situ		In situ	

[a]MLSS, Mixed liquor suspended solids.
[b]Although Kubota was not found very active in China, it was still referenced here in order to compare flat-sheet membranes made in China and those made in other countries.

Table 2. Summary comparison of membranes used in full-scale MBRs and MBR performance (adapted from [39, 41, 42]).

3.2. Design and operating parameters

A number of parameters must be considered in order to activate an economically appropriate MBR system. These include membrane selection, membrane performance (permeate flow, transmembrane pressure, viscosity), biological performance of microorganisms (biomass concentration, ESS, HBS, F/M ratio), and economic factors (energy consumption, sludge treatment, and disposal cost). These parameters can influence each other, and a positive change can be observed in the other parameter by changing one parameter. For example, a high biomass concentration requires a long CIS, which in turn reduces the cost of sludge disposal and sludge disposal. On the other hand, at high sludge age, the cost of energy also increases as the sludge reaches a viscous structure, which leads to the decomposition of the organic fraction and the amount of oxygen needed to grow the microorganism [43–45].

These designed and operational parameters are used to design the reactor and to be able to differentiate in different configurations applied to the process, to give formulas which are used in the general working principles of MBRs, also in the definition and calculation.

The amount of liquid drained from the surface area of the membrane is called flux. MBRs are mostly 10–100 LMH flux values.

3.3. Advantage and disadvantage of MBR

The best feature of MBRs is that they can easily convert existing activated sludge systems into MBR systems. This can be accomplished by placing submerged membranes in the aeration tank [46]. Membrane bioreactor is separating biological treatment of microorganisms and secondary cleaners from one site to another. The feed water is mixed with the biomass, the mixture is filtered from the membrane, and the biomass is separated from the treated water. Conventional activated sludge (CAS) units compared to the same operational conditions to provide better recovery efficiency in the MBR. Using MBR has many advantages [22, 47] (**Table 3**).

At higher MLSS concentrations, the ability to work at higher SRT than conventional treatments, reduced biomass yield, higher quality waste, less hydraulic residence time and lower area footprint generation are advantages of MBRs compared to CAS units [48]. This means a small reactor volume and a reduction in the initial investment cost. They are also more resistant to sudden different hydraulic and organic loads and better respond to existing sustainability criteria for municipal wastewater systems [49]. Biomass separation is independent of the ability of the activated sludge to precipitate as it is achieved by microfiltration or ultrafiltration; in other words, there is no need for final sedimentation, no sludge swelling, and sedimentation problems caused by filament growth. Due to high MLSS concentrations, excess organic loading can be done in the system. MBRs are less likely to be negatively affected by nitrification or by business problems related to the toxic effects of toxic organisms [50]. Since the sludge from the membrane system is less than the conventional system, the storage requirement is also reduced [51].

MBRs are becoming increasingly common throughout the world, despite the fact that they can reduce their investment and operating costs and produce effluent that cannot be used despite their different reuse areas. One of the biggest causes of this is the clogging of the wastes, and the transmembrane pressure (TMP) increases to provide a constant flux. Occlusions may occur at the membrane surface or within the membrane pores. Membrane clogs

MBR	CAS
Meets sensitive discharge standards	Cannot meet sensitive discharge standards
Decreased reactor volume and foot print	Large area is required for the secondary clarifier
Used as a pretreatment for reverse osmosis (RO) and nanofiltration (NF) with good effluent quality	Less quality effluent is obtained
Complete retention of bacterial flocs by the membrane	Needs disinfection step
Biomass retention is achieved by the membrane	Biomass retention is accomplished by gravity
Operated at elevated solid retention time (SRT)	Usually operates with low SRT
Better removal efficiency for slowly biodegradable micropollutants	The low SRT in ASP cannot allow this
High MLSS (10–15 g L^{-1}) and low feed to microorganism ratio (F/M)	MLSS is about four times less than that of MBR
Long SRT and high MLSS imply low sludge yield	Low SRT and low MLSS imply high sludge yield

Table 3. Comparison of MBR and CAS (adapted from [52]).

are roughly divided mechanically into two: recycled (removal of the surface gel and cake layer by aeration or physical backwash) and irreversible (removal of dissolved or colloidal substances in the adsorptive pore accumulation and clogging by chemical cleaning) [53]. MLSS, particle size distribution, soluble microbial by-products, extracellular polymeric materials, viscosity, pore size, porosity, surface energy, electrical charge, hydrophilic/hydrophobic properties parameters are affecting clogging [54]. The formation of cake, which is unavoidable on the membrane surface, is one of the factors that cause the membrane to become contaminated. In a general system, the sidestream of the MBR shows a higher tendency to pollute than the submerged MBR. The reason is that the sidestream MBR needs high pump energy to generate high flux which will cause repetition of pollution when compared to the submerged MBR [37]. Tank reduces production, increases operating and maintenance costs, and requires a special extra cleaning and backwashing. Membrane replacement is challenging. There are more than 10 years of MBR systems. On the other hand, there are many systems that change after 4 years. The main causes are often pollution problems. When contamination is combined with high transmembrane pressure, this contamination is most irreversible, and therefore the chemical cleaning frequency should be increased. This leads to an increase in operating cost by reducing membrane life [51].

The main contributors to energy costs in MBR are sludge transfer, permeate production, and aeration which is often exceeding 50% of total energy consumption. Energy consumption of membrane-related modules was in the range of 0.5–0.7 kWh/m^3, and specific energy consumption for membrane aeration in flat sheet was 33–37% which was higher than in a hollow fiber system. Submerged membranes in MBR reduces the pumping energy requirement to 0.007 kWh/m^3 of permeate compared with sidestream membrane (3.0 kWh/m^3). Future trend of MBR might be focused on two aspects which are reduction of energy demand and membrane fouling [55].

4. Micropollutant treatment studies with MBR applications

Many analgesics such as ibuprofen, diclofenac, naproxen, and ketoprofen; lipid regulators such as bezafibrate and gemfibrozil; and carbamazepine for antiepileptic drugs were frequently found to be removed at concentrations above 1.0 mg/L in domestic wastewater and in MBR procedures [22].

While the removal rates of microcontaminants in MBR vary from one compound to another, these removal rates, sludge retention time (SRT), biomass concentration, temperature, pH value, class of microcontaminants and hydrophobicity, chemical structure, pKa etc. as well as their physico-chemical properties. The hydrophobic components are removed from the liquid phase by adsorption and, possibly, when the SRT is sufficiently high, to be removed between the biodegradation processes [56–58]. The compactness of the MBR system, the high organic load that can be applied, and the high SRT give good results in removing micropollutant [48]. When the pH value of the wastewater changes, it may affect the removal of micropollutants in the negative direction. On the other hand, the role of pH on sorption has been related with the dissociation of certain micropollutants (through the acid dissociation constant pKa), which can result in the generation of positively charged compounds (prone to interact with the negatively

charged surface of sludges) or anions (low interaction). Thus, the cationic species would be adsorbed by van der Waals-type interactions [59].

Wastewater temperature also plays an important role. WWTP with an average temperature of 15–20°C can be better suited for micropollutants such as in cold countries, which are often below 10°C in the USA. Summer and winter affect seasonal temperature changes, micro-degradation, and biodegradation [60]. Sorption has been correlated inversely with temperature in the case of the hormone 17α-ethinyl estradiol (EE2), with a reduction of Kd values of 20–25% when the temperature was increased from 10 to 30°C [59].

Studies have shown that compounds such as ibuprofen and antiseptic powder, methyl paraben, and galaxolide, an analgesic drug in hospital wastewater, do not have significant differences in effluent efficiency with activated sludge processes and MBR. MBR system was found to be efficient for hormones (e.g., estriol, testosterone, androstenedione) and certain pharmaceuticals (e.g., acetaminophen, ibuprofen, and caffeine) with approximately 99% removal [61, 62]. Experimental investigations show that the removal of such compounds from wastewater is 30–50% superior to that of conventional activated sludge process. In addition, the removal efficiencies of some compounds such as mefenamic acid, indomethacin, diclofenac, and gemfibrozil in MBR were 40%, 40%, 65%, and 32–42% [63, 64]. However, biodegradable erythromycin, TCEP, trimethoprim, naproxen, diclofenac, carbamazepine, and nonylphenoxyacetic acid have not been removed [47]. This is comparable to the results of previous studies which indicated very low elimination rates of diclofenac and carbamazepine in WWTP due to their recalcitrant nature processes in Germany. Hydrophilic compounds such as MBRs, acetaminophen, atenolol, iopromide, and sulfamethoxazole (calculated logP <2) (with the exception of sulfamethoxazole (> 62%) are more efficient than hydrophobic compounds. Hydrophobic compounds (calculated log P > 2) can largely be removed by active sludge biosorption in the MBR and in the middle, and longer holding scoops are formed in the bioreactor, resulting in a higher removal yield from the CAS process. However, some hydrophilic microspheres such as carbamazepine and diclofenac tend to be highly resistant to biological degradation in the treatment of CAS and MBR. The retention time of the hydrophilic and persistent micropollutants in the bioreactor is the same as the retention time of hydraulic retention (HRT), as the micropollutants can freely permeate MF and UF membranes. The duration of hydraulic retention in the MBR and the prolongation of the retention time of the sludge are dependent on the compound biosorption of some hydrophobics for the activated sludge, and it can be seen that the pollutants can improve the biodegradation [2, 65].

In the comparison between the two MBR modules used in this study (plate and frame versus hollow fiber), no difference in target compound removal was found [60, 65]. Some results can be negative efficiency. For example, González-Pérez et al. (2017) have worked on the system that has been operated with complex nitrification and ensured that the biodegradable organic material due to circulation is effectively retained [66]. By reducing the concentration increase in the diclofenac (DCF) in the aerobic bioreactor, negative removal efficiencies for DCF have been obtained. This was not observed in the anoxic reactor.

Membrane bioreactor applications for these pollutants in different wastewaters are presented in detail given in **Table 4**.

Operation mode/membrane configuration	Volume (L)/ temperature (°C)	Substrate	MLSS (g SS/L) Organic loading rate (kg COD/m^3.d)	Removal (%)			Reference
Full scale (VRM) Pilot Scale (Clear-box) PES membrane Plate and frame	350 L 4 °C	Syntetic wastewater	5 g/L		Full Scale	Pilot Scale	[67]
				Diltiazem	100	0	
				Acetaminophen	100	>95%	
				Estrone	10	100%	
				Carbamazepine	0	0	
Anoxic + aerobic hollow fiber (MF) PVDF Membrane submerged	5 L 25°C (±5°C)	Real wastewater	7 – 11 g/L 0.1 grBOD/grMLVSS	Bezafibrate Ketaprofen Furosemide Atenolol Propranolol Diltiazen Roxithromyan Clarithromyun Naproxen Ciprofloxacin Levofloxacin Tetracycline Triclosan Triclocarban	93 87 68 58 50 57 51 46 97 36 47 52 84 42		[63]
Anaerobic reactor+ external / hollow fiber Hybrid aerobic MBR	176 L 20 – 22 °C	Synthetic wastewater	0,6 g/L 1.7 grCOD/L.d	Sulfamed hoxazole Trimethoprim	> 84%		[68]
Anoxic+Aerobic+MBR 3,6m^3 + 8,8 m^3 + 3,5 m^3 Flat sheet (MF)			6,3 – 7.1 gr/L TSS 75 – 77 % VSS 0.83 – 0.98 kgCOD/m^2d	Ibuprofen Naproxen Ketoprofen $Diclofenac_A$ $Diclofenac_B$	98.12 98.2 92.26 20.70 -18.75		[66]
Flat Sheet MF Hallow Fiber UF + PAC	MF_{MBR} ➔ 30 L UF_{MBR} ➔ 185 L 20 – 22 °C	Synthetic wastewater	$3gr_{VSS}$/L 400 mgCOD/L 0.4 gr COD/L	Trimethoprim	MF% 50	UF % 40	[69]

Operation mode/membrane configuration	Volume (L)/ temperature (°C)	Substrate	MLSS (g SS/L) Organic loading rate (kg COD/m^3.d)	Removal (%)			Reference
				Sulfamed hoxazole	80	70	
				Erythromycin	80	90	
				Carbamazepine	0	0	
				Roxithromyan	70	> 95	
				Aceclofenac	30	60	
				Naproxen	90	90	
				Ibuprofen	90	> 95	
				Ethynilestradiol	90	> 90	
				Estradiol	95	> 95	
				Naproxen	98.2		
				Ketoprofen	92.26		
				$Diclofenac_A$	20.70		
				$Diclofenac_B$	-18.75		
Lab scale hollow fiber membrane model	3.2 L	PVDF submerged	1.66 gVSS/L 2.16 g+COD/Lday	Levo	98.7		[1]
				Betha-V	97.8		
				Betha-D	99.6		
				Medro	93.4		
Pilot-scale PES UF submerged flat sheet		Hospital effluent	2 g/L	Carbamazepine	−6		[70]
				Trimethoprim	96		
				Sulfamethoxazole	7		
				Atenolol	99		
(4.7 m^3) Microfiltration (MF) Flat Sheet (FS) membrane Module (3.6 m^3) Hollow fiber (HF) ultrafiltration membrane External configuration	20±2 °C	Real wastewater			FS MBR	HF MBR	[22]
				Analgesics and anti-inflammatory drugs			
				Ibuprofen	99.2	99.5	

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Operation mode/membrane configuration	Volume (L)/ temperature (°C)	Substrate	MLSS (g SS/L) Organic loading rate (kg COD/m^3.d)	Removal (%)			Reference
				Naproxen	90.7	91.6	
				Ketoprofen	43.9	44.0	
				Diclofenac	65.8	62.6	
				Mefenamic	40.5	35.5	
				Propyphenazone	64.5	60.7	
				Acetaminophen	99.8	99.9	
				Indomethacin	41.4	39.7	
				Anti-histamines			
				Ranitidine	44.2	29.5	
				Loratidine	<10	33.5	
				Famotidine	64.6	47.4	
				Anti-epileptic drug			
				Carbamazepine	<10	<10	
				Psychiatric drugs			
				Fluoxetine	98.0	98.0	
				Antibiotics			
				Erythromycin	43.0	25.2	
				Sulfamethoxazole	80.8	78.3	
				Ofloxacin	95.2	91.3	
				Trimethoprim	66.7	47.5	
				ß-blockers			
				Atenolol	76.7	69.5	
				Sotalol	53.1	30.4	
				Metoprolol	44.2	29.5	

Operation mode/membrane configuration	Volume (L)/ temperature (°C)	Substrate	MLSS (g SS/L) Organic loading rate (kg COD/m^3.d)	Removal (%)			Reference
				Propranolol	77.6	65.5	
				Hypoglycaemic agents			
				Glibenclamide	95.6	82.2	
				Lipid regulator and cholesterol lowering statin drugs			
				Gemfibrozil	42.2	32.5	
				Bezafibrate	90.3	88.2	
				Pravastatin	86.1	83.1	
				Hydrochlorothiazide	<10	<10	
Full-scale hollow fiber		Raw wastewater	7.5–8.5 g/L	Ibuprofen	~100		[71]
				Diclofenac	43		
				Carbamazepine	24		
				Sulfamethoxazole	60		
				Trimethoprim	30		
				Estrone,	~100		
				Estriol	~100		
				BisphenolA	~100		
Lab-scale hollow fiber submerged UF modüle		Synthetic wastewater	8.6–10 g/L	Ibuprofen	96.7		[72]
				Diclofenac	17.3		
				Paracetamol	95.1		
				Carbamazepine	13.4		
				Linuron	21.1		

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Operation mode/membrane configuration	Volume (L)/ temperature (°C)	Substrate	MLSS (g SS/L) Organic loading rate (kg COD/m^3.d)	Removal (%)		Reference
				Sulfamethoxazole	91.9	
				Ketoprofen	70.5	
				17β-estradiol	99.4	
				17α- ethynilestradiol	93.5	
				Triclocarban	>98.4	
				Naproxen	40.1	
				Bisphenol A	90.4	
				Sulfamethoxazole	91.9	
				Nonylphenol	99.3	
				Atrazine	4.4	
Flate and frame- type hollow fiber	1 m^3/d	Domestic wastewater		Hormones	Good	[60]
				Acetaminophen	99	
				Ibuprofen	99	
				Caffeine	99	
				Others	low	
A pilot-scale MBR flat-sheet membranes submerged	21 L	Municipal, hospital, and industrial wastewater		Aceclofenac Carbamazepine Diclofenac Enalapril Trimethoprim	~ 50 ~ 0 ~80 > 95 > 95	[49]
Flat- sheet (MF) membrane submerged MBR	21L (20 ± 2 °C)	Real wastewater (municipal, hospital and industrial)		***Analgesics and anti-inflammatory drugs***		[22]
				Naproxen	99.3	
				Ketoprofen	91.9	
				Ibuprofen	99.8	

Operation mode/membrane configuration	Volume (L)/ temperature (°C)	Substrate	MLSS (g SS/L) Organic loading rate (kg COD/m^3.d)	Removal (%)		Reference
				Diclofenac	87.4	
				Indomethacin	46.6	
				Acetaminophen	99.6	
				Mefenamic acid	74.8	
				Propyphenazone	64.6	
				Anti-ulcer agents		
				Ranitidine	95.0	
				Psychiatric drugs		
				Paroxetine	89.7	
				Antiepileptic drugs		
				Carbamazepine	-	
				Antibiotics		
				Ofloxacin	94.0	
				Sulfamethoxazole	60.5	
				Erythromycin	67.3	
				B-blockers		
				Atenolol	65.5	
				Metoprolol	58.7	
				Diuretics		
				Hydrochlorothiazide	66.3	
				Hypoglycaemic agents		
				Glibenclamide	47.3	
				Lipid regulator and cholesterol lowering statin drugs		

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Operation mode/membrane configuration	Volume (L)/ temperature (°C)	Substrate	MLSS (g SS/L) Organic loading rate (kg COD/m^3.d)	Removal (%)			Reference
				Gemfibrozil	89.6		
				Bezafibrate	95.8		
				Clofibric acid	71.8		
				Pravastatin	90.8		
Full-scale flat sheet		Hospital effluent		Ibuprofen	>80		[73]
				Carbamazepine	<20		
				Diclofenac	<20		
MBR Concept A	10 °C		10 g/L		Concept A	Concept B	[74]
MBR Concept B	10 °C		10 g/L				
				Paracetamol	>99	>99	
				Ibuprofen	>99	98.5	
				Ketoprofen	90.1	81.3	
				Naproxen	>97	90.2	
				Caffeine	99.7	99.5	
				Tetracycline	>95	>95	
				Atenolol	70.8	69.1	
				Bisoprolol	41.9	21.7	
				Metoprolol	18.8	20.1	
				Sotalol	-27.7	-33.0	
				Furosemide	16.3	5.6	
				Hydrochlorothiazide	27.5	2.4	
Pilot-scale MBR hollow fiber PVDF	1.3 m^3	Hospital wastewater	less than 13 g/L	Diclofenac Sulfamethoxazole Trimethoprim		0 0 0	[75]

Operation mode/membrane configuration	Volume (L)/ temperature (°C)	Substrate	MLSS (g SS/L) Organic loading rate (kg COD/m^3.d)	Removal (%)			Reference
				Carbamazepine		0	
				Tramadol		0	
				Naproxen		23.6	
				Propanolol		34.2	
				Ibuprofen		100	
				17b-Estradiol		100	
				Triclosan		100	
				Gemfibrozil		0	
Laboratory-scale MBR	Feed tank (50 L) MBR (15 L)		2.15 gCOD/L/d	Amelotin (AMTN)		20-40	[76]
Pilot-scale Anoxic+Aerobic MBR	Anoxic (13.8 L) Aerobic (11.7 L) hollow fiber Ultrafiltration membrane (18 ± 3 °C)		Anoxic (4.1 ± 0.5 and 2.7 ± 0.3 g/L) Aerobic (2.4 ± 0.8 g/L (MLSS))	Atenolol	Pilot	Full Scale	[77]
				Sulfamethoxazole	> 80	> 80	
				Caffeine	∼ 60	> 80	
				Naproxen	∼ 90	> 90	
				Ibuprofen	> 80	> 90	
				Paracetamol	> 90	> 90	
				Trimethoprim	> 90	> 90	
				Primidone	∼ 60	∼ 70	
				Diclofenac	∼ 20	0	
				Gemfibrozil	∼ 20	∼ 60	
				Carbamapazine	∼ 90	> 90	
				DEET	0	0	
				Diuron	∼ 80	> 90	
				Polyparaben	∼ 30	> 90	
				Amtriptyline	> 90	> 90	
				Estrone	0	∼ 30	
				Androsterone	> 90	> 90	
				Etiocholanolone	> 90	> 90	
				Triclosan	> 90	> 90	
				Triclocarban	∼ 60	∼ 60	
					> 90	∼ 50	

Table 4. Membrane bioreactor applications for micropollutants in various wastewaters.

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5. Integration of MBRs with other technologies

Membrane bioreactors (MBRs) have recently emerged with integrated MBR systems, along with other treatment technologies. The purposes of the integrated MBR are to improve qualities of permeates, mitigate membrane fouling, and enhance the stability of the treatment process. Recent studies have provided improvements in the degradation of micropollutants using integrated

Integrated technology of MBR	Advantages	Disadvantages and limitations
Advanced oxidation processes/electrocoagulation-MBR	Effective in removal of recalcitrant contaminants (pharmaceutical wastewater) Effective in removing colors Reduces the production of excess sludge Easy to operate Reduce membrane fouling	High capital and operational cost Not effective in treatment of wastewater with high TSS
FO-MBR	Produce good effluent quality Phosphorus recovery Low energy consumption as compared to conventional MBR Low fouling tendency compared to RO Effective in removal of trace organic contaminants Fouling is largely reversible Effective in treatment of wastewater with high TSS as compared to RO	Uncertainly of stability of membrane Increasing salinity/salt accumulation might decrease the microbial kinetics and water flux
RO-MBR	Low fouling tendency Cost of RO membrane is cheaper than FO membrane Low energy consumption as compared to conventional MBR	Not effective in treatment of high-salinity wastewater compared to FO
Membrane distillation	Enhances biodegradation of recalcitrant compounds Low sludge yield Higher effluent quality Excellent process stability Cost-effective compared to RO process Smaller footprint	Low removal of COD
Biofilm/bio-entrapped MBR	Reduces the concentration of suspended solids Reduce membrane fouling Improve nitrification and denitrification processes	Membrane fouling might be severe at the later stage of treatment
Granular MBR	Improve nitrification and denitrification processes High shock resistance capacity Reduce membrane fouling Smaller footprint	Membrane fouling might be severe at the later stage of treatment Long start-up period of granule formation

Table 5. Advantages and disadvantages of various integrated MBRs in wastewater treatment technology [55].

processes. There are several methods to reduce the membrane fouling of MBR such as optimization of HRT and SRT which were discussed in some review papers. These processes containing biofilm carriers, suspended/attached growth system, or cross-linked enzyme aggregates showed better removal of micropollutants, even on recalcitrant compounds such as CBZ [78].

The advantages and disadvantages of various integrated systems, such as advanced oxidation processes (AOPs) [79], reverse osmosis (RO-MBRs) [64], forward osmosis (FO-MBRs) [80], membrane distillation (MDBRs) [81], microbial fuel cells (MBR-MFCs) [7], anaerobic (AnMBRs) [82], biofilm (BF-MBR) [83], and granular (GMBR) membrane bioreactors [84] to demonstrate their ability to reduce membrane contamination, are given in the **Table 5**. Combined MBR process configurations and conventional biological therapies, as an alternative, resulted in more consistent results. As shown in the studies, the removal efficiency of each of the micropollutants is different for the different membrane technologies. The value ranges from close to zero to almost complete removal. For example, the removal efficiency of carbamazepine is less than 20% with ASP and MBR and up to 93% with MBR-NF and higher than 99% with MBR-RO, MBR-PAC, and MBR-GAC [52]. The use of combinations of different complementary technologies has produced promising results. Nonetheless, there is a lack of a holistic understanding of the nature of pollutants, their interactions, and some predictable relationships between the best available specific technologies. More practice is needed to evaluate the hybrid MBR systems proposed in the treatment of micropollutants [48].

6. Conclusions

In recent years, pharmaceutical products have been a cause for concern due to the persistence of their presence in aquatic environments. Drugs are known to be involved in a variety of aquatic environments, including domestic wastewater, hospital discharges, sewage treatment plants, and water treatment plants.

Pharmaceutical products can preserve their original concentrations and structures, or they can be mobilized for life in water matrices and converted to other active (or inactive) compounds. The presence of micropollutants in aqueous environments is an increasing concern due to their potentially harmful effects on aquatic life. Since this situation poses a serious danger to the environment, the treatment of these pollutants is very important.

As it is clear from this work, today's CAS is not sufficient for the destruction of many pharmaceutical substances in the wastewater of the AAT. For these pollutants, the use of MBR systems developed by adding membranes to CAS systems has begun to be used, and these are often more effective at removing pollutant concentrations than traditional biological treatment systems. MBR technology has become a reliable and valuable option with many advantages. However, in addition to its advantages, membrane fouling is a major obstacle to the development of these systems. To this end, it will be useful to focus on the reduction of energy demand and membrane contamination during operation, along with the development of integrated MBR systems, with future research. Further work is needed to assess which system actually makes more cost–benefit and to investigate the toxicity of micropollutants and the effect of working conditions after processing.

Author details

Berna Kırıl Mert[1]*, Nihan Ozengin[2], Esra Can Dogan[3] and Coskun Aydıner[4]

*Address all correspondence to: bkiril@sakarya.edu.tr

1 Sakarya University, Faculty of Engineering, Department of Environmental Engineering, Sakarya, Turkey

2 Uludag University, Faculty of Engineering, Department of Environmental Engineering, Bursa, Turkey

3 Kocaeli University, Faculty of Engineering, Department of Environmental Engineering, Kocaeli, Turkey

4 Gebze Technical University, Environmental Engineering Department, Gebze, Kocaeli, Turkey

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