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Removal of contaminants of emerging concern (CECs) using a membrane bioreactor (MBR): a short review

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Abstract

Contaminants of emerging concern (CECs), such as pharmaceuticals and personal care products (PPCPs) and as endocrine disrupting compounds, have recently recognized as the next set of pollutants due to their toxic effects on human health and aquatic organisms at very low concentrations. One of effective methods to remove these emerging contaminants present in the aquatic environment is a membrane bioreactor (MBR). In this review, 27 CECs belonged to diverse categories of PPCPs were surveyed from the point of view of the removal efficiency by several types of MBR modules with various operational conditions, such as a type of MBR, hydraulic retention time and sludge retention time. As a result, this review provided the overall ranges in the removal efficiency of 27 CECs by different MBR filtrations and modules. Certain categories of PPCPs such as analgesics/anti-inflammatory drugs (acetaminophen and ibuprofen), steroids/hormones (estriol and testosterone) and stimulant (caffeine) have relatively higher removal rates, while antimicrobial agent (TCEP) is rarely removed in the different MBRs. For further implementation of CEC removal by a MBR system, physical characteristics/biological fate of a wide variety of CECs, individual/synergistic effects which may occur during MBR operation, and application of advanced MBR technologies should be studied.

Keywords: Wastewater treatment, pharmaceuticals and personal care products (PPCPs), removal efficiency.

1. Introduction

Many studies have reported that novel organic compounds synthesized for various purposes, such as personal care (*i.e.* hand soap, sunscreen, shampoo and cosmetics etc.), agricultural activities, and human/animal health care, can threaten aquatic ecosystem (Lapworth *et al.*, 2012; Jiang *et al.*, 2013; Liu *et al.*, 2013). Due to the advancement of medical science, therapeutic compounds such as aspirin, arsphenamine and ephedrine, have been developed in early 20th and widely used as pain relievers and medicines against different types of illness (Tiwari *et al.*, 2016). A large consumption of these personal care products and pharmaceuticals (PPCPs) eventually contributes organisms in the aquatic environment to be exposed by contaminants of emerging concern (CECs). CECs are manmade pollutants and still remained less significant than their potential impacts due to the detection of these chemicals at infinitesimal concentrations; from µg/L to ng/L across an array of ecosystems. Recently, environmental scientists have dealt with the occurrence of CECs in surface water, water supply, wastewater, groundwater and sewage sludge in various regions including European, Asian, American and even African communities (Jiang *et al.*, 2013; Guo *et al.*, 2014; Subedi *et al.*, 2015; Hashim *et al.*, 2016; Fisch *et al.*, 2017; Fisch *et al.*, 2017; Krogh *et al.*, 2017; Kwon *et al.*, 2017).

One convincing technology to treat emerging contaminants including PPCPs in municipal wastewater treatment plants (WWTPs) is a membrane filtration with reverse osmosis (RO), nano-filtration (NF), ultra-filtration (UF) or micro-filtration (MF) (Kim *et al.*, 2007). Membranes are thin and permeable layers of material that can be used to remove contaminants in water by permitting the transmission of water at a different rate according to the porous size of the membrane (Visvanathan *et al.*, 2000; Benjamin *et al.*, 2013). As the technology of membrane filtration has been dramatically advanced, environmental scientists have made attempts to combine microporous or nanoporous membranes with the conventional activated sludge (CAS) system for solid and liquid separation, instead of using the secondary clarifiers (Shariati *et al.*, 2010; Chon *et al.*, 2011; Benjamin *et al.*, 2013). More recently, various integrated MBR modules, for example, advanced oxidation process combined with electrocoagulation MBR, reverse osmosis MBR (RO-MBR), forward osmosis MBR (FO-MBR), membrane distillation bioreactor (MDBR), biofilm/bio-entrapped MBR and granular MBR, have been developed to circumvent limitations of conventional MBRs (Neoh *et al.*, 2016). Eventually, this innovative technology have led to increase the removal efficiency of suspended solids, organic micropollutants and even CECs (Tan *et al.*, 2017).

For those reasons, extensive studies have reported the removal of CECs by different MBR system and modules (Kim *et al.*, 2007; Radjenovic *et al.*, 2007; Radjenović *et al.*, 2009; Chon *et al.*, 2011; Tiwari *et al.*, 2016). For instance, Kim *et al.* (2007) surveyed the occurrence of PPCPs in surface water, drinking water and wastewater in South Korea and measured the elimination efficiency using the RO and NF MBRs. Radjenovic *et al.* (2007) demonstrated that the MBR was more effective (removal rate > 80%) to treat the certain pharmaceuticals than the conventional activate sludge (CAS) system. Further, Radjenović *et al.* (2009) reported the better removal of pharmaceutically active compounds (PhACs), such as mefenamic acid, diclofenac and indomethacin, which were properly removed by CAS process.

The primary objective of this review is to compare the performances of an array of MBR modules with various operational conditions, and to evaluate the results from a wide range of the reported MBR studies in terms of removal efficiency of PPCPs belonged to CECs. The PPCPs evaluated in this review were included in analgesics/anti-inflammatory drugs, antibiotics, antiepileptic drug, β-blockers, blood lipid regulators, steroids/hormones (estriol and testosterone), stimulant, antimicrobial agent/disinfectant, flame retardant, and synthetic musks/fragrances. Therefore, providing information on overall removal rates with relatively well-/rarely removed PPCPs by MBR operational condition would be useful as a criterion for treatment strategy to effectively manage wastewater systems as well as to improve the sustainability of aquatic environment.

2. Membrane bioreactor

2.1. Classification of MBR

Membranes are generally divided into several groups according to the type of membrane and the application. Membranes are commonly classified as reverse osmosis (RO), nano-filtration (NF), ultra-filtration (UF), micro-filtration (MF) and particle-filtration regarding to the permeability of contaminants through the thin layers of the membrane (Benjamin *et al.*, 2013; Yoon, 2015). As the maximum pore dimensions of the membrane decrease, the permeability of contaminants through membrane usually decreases (Nath, 2017). Dead-end and the circular cross-flow filtration are two important types of filtration that should be considered. In dead-end filtration, the input flow runs perpendicular to the membrane whereas in cross-flow filtration, the input flow is parallel to the membrane (Shamsuddin *et al.*, 2015). The membrane configurations most widely used are hollow fiber and flat sheet module. The typical arrangement of a membrane bioreactor is represented as a submerged MBR, but a side-stream MBR is an alternative (Nath, 2017).

The material of the membrane usually can be polymeric, metallic or ceramic (Lin *et al.*, 2013). Polymeric membranes are made of a polymer monolith such as polyvinylidene difluoride (PVDF), polyethylene (PE), polypropylene and polyethersulfone (PES) which is the most broadly applied (Lin *et al.*, 2013). Moreover, polymeric membranes have the characteristics of being a single material, being self-supporting and being the only material used to construct hollow fiber membranes (Lee *et al.*, 2013). Metallic membranes have advanced hydraulic performance and fouling recovery. In addition, metallic membranes have more durable tolerance especially to high temperature and oxidation in comparison with polymeric membrane material (Kim *et al.*, 2007). Ceramic membranes is one of the most widely used material particularly for anaerobic MBR (AnMBR) system (Imasaka *et al.*, 1989; Chang *et al.*, 1994; Ghyoot *et al.*, 1997), due to their effective resistance to corrosion, abrasion, increased concentration polarization control, and fouling through backwashing (Ersu *et al.*, 2008). Ceramic membranes are needed to be supported with multiple materials and can be used for either hollow fiber or flat sheet configuration (Kumar *et al.*, 2015). However, as metallic membranes and ceramic membranes are much more expensive than polymeric membrane materials, there is limitation on the large scale implementation (Kumar *et al.*, 2015). Polymeric membranes are more economical for commercial applications (Lin *et al.*, 2013).

2.2. The characteristics of membrane bioreactor (MBR)

The membrane bioreactor (MBR) has three main advantages: 1) the water quality treated by a MBR is independent of the mixed liquor suspended solids (MLSS) (Yoon, 2015); 2) a secondary clarifier and a tertiary process are not necessary in the MBR system, because the MBR plays the important role of clarifier that is similar to a conventional activated sludge (CAS) system, thus the overall size of MBR plant can be significantly reduced (Howell, 2004; Shariati *et al.*, 2010); 3) a longer sludge retention time (SRT) allowed a MBR process to provide 2 to 5 times more active biomass than a CAS system, thus effluent water quality in a MBR system is considerably higher than that from a CAS process (Yamamoto *et al.*, 1989; Jefferson *et al.*, 2000). At the same time, the longer SRT improved active biological degradation due to the increased sludge concentration (Marrot *et al.*, 2004). However, the most significant concern is fouling during the operation of a MBR system. Fouling in a MBR system indicates that the accumulation of rejected materials on a membrane increases the resistance to transporting water through the membrane layers (Marrot *et al.*, 2004). Fouling can be controlled either physically or chemically, *i.e.* by backwashing with air and/or water, or by using chemicals like caustic soda, and oxidants including hydrogen peroxide (Shen *et al.*, 2015; Yoon, 2015). The performance of MBR process can be determined by several operating parameters. Hai *et al.* (2011) reported that the level of total organic carbon (TOC) and total nitrogen (TN) have significantly reduced at 45 °C rather than the temperature range of 10 °C to 35 °C in the bioreactor. Some PPCPs like acetaminophen, ketoprofen, naproxen, roxithromycin, sulfamethoxazole and trimethoprim have higher removal efficiency in the longer SRT (= 30-day) compare to 15-day of SRT (Tambosi *et al.*, 2010).

3. Contaminants of emerging concern (CECs) treatment in MBR

3.1. Contaminants of emerging concern (CECs)

Contaminants of emerging concern (CECs) can be any chemical compounds including industrial chemicals, persistent organic compounds (POPs), natural toxic compounds, and pharmaceuticals and personal care products (PPCPs). One major category, PPCPs are steadily being found in the aquatic environment at low concentrations, and have recently received significant attention by environmental scientists as well as policy makers (Kumar *et al.*, 2010; Archer *et al.*, 2017).

The general forms of PPCPs consumed by human are medicines, veterinary drugs, and cosmetic products (Sui *et al.*, 2017). The type of PPCPs is usually classified according to their applications. Pharmaceuticals can be analgesics and anti-inflammatory drugs, antibiotics, antiepileptic drugs, blood lipid regulators, *β*-blocks, stimulant, steroids and hormones (Ellis, 2006). As personal care products (PCPs), antimicrobial agents and disinfectants, artificial sweetener, cosmetics, flame retardants, insect repellants, synthetic musks, fragrances, and sunscreen UV filters have been widely used (Jiang *et al.*, 2013; Liu *et al.*, 2013). Further, PPCPs that can cause endocrine disruption (e.g., estrone (E1), estradiol (E2), testosterone and norgestrel) were described in Table 1.

PPCPs can have adverse effects in both humans and aquatic organisms. Even though the concentrations of PPCPs present in water bodies are as low as parts per trillion, the influence of PPCPs on neurobehavioral effects, inhibition of efflux pumps, and rapid inhibition of sperm activity have been observed (Wilkinson *et al.*, 2016; Yang *et al.*, 2017).

The endocrine system is an integrated system of glands and hormones that governs growth, development, reduction and metabolism (Ying *et al.*, 2004). The major endocrine glands are the pineal gland, the pituitary gland, the thyroid gland, the thymus, the adrenal gland, the pancreas, the ovary (female) and the testes (male) (Ying *et al.*, 2004; Holtz, 2006). EDCs can usually be absorbed into blood through food, skin or air, and disrupt the function of the endocrine glands by directly activating/blocking hormone receptors or by controlling hormone levels or hormone receptor concentrations (Tijani *et al.*, 2016; Archer *et al.*, 2017). The important chemicals of EDCs are pesticides, detergents, plasticizers, and a mixtures of unknown EDCs in wastewater (Dotan *et al.*, 2016). The most well-known examples of EDCs include dioxins, polychlorinated biphenyls (PCBs), dichlorodiphenyl trichloroethane (DDT), di-*n*-butylphthalate, bisphenol and diethylstilbestrol (DES), a synthetic estrogen (Giulivo *et al.*, 2016). The PCBs and dioxins cause immune alterations while dioxins and DDT result in diabetes and precocious puberty, respectively (Eskenazi *et al.*, 2017).

3.2. Pathway to aquatic environment and human effects of CECs

The occurrence of CECs including pharmaceutically active chemicals and endocrine disrupting compounds in the domestic and industrial wastewater have been recognized as a crucial environmental concern in ecological system (Kasprzyk-Hordern *et al.*, 2009). Gros *et al.* (2010) reported that over 3,000 different pharmaceuticals were used for human medications within daily human activity in the European Union (EU), resulting in a wide variety of CECs pathways into the aquatic environment (K'oreje *et al.*, 2016; Mandaric *et al.*, 2017).

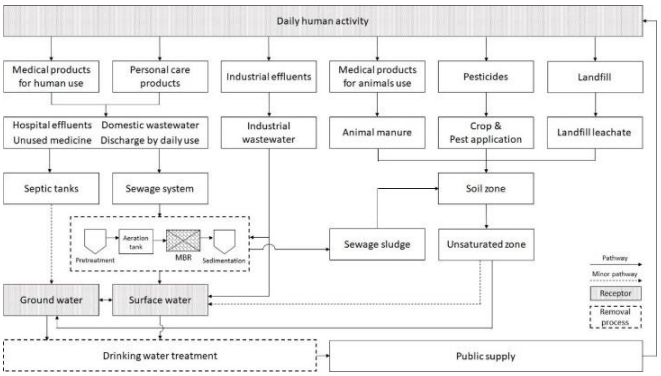

Figure 1. Possible pathways of CECs in the aquatic organisms (adapted from Ellis, 2006 and Stuart *et al.*, 2012)

According to previous studies on occurrence of CECs, discharge of treated water from WWTPs can be one of pathways into the water bodies due to the limited removal of pharmaceuticals by conventional secondary processes and/or sewage treatment plants (STPs) (Jiang *et al.*, 2013; Petrie *et al.*, 2015). Personal care products (PCPs) such as cosmetics, hair care products, tooth pastes and skin care products are also analogous to pharmaceuticals as shown in Figure 1. CECs can be also released from agricultural and rural point source due to the use of pesticides that can adversely affect crops, aquatic and soil ecosystems (Stuart *et al.*, 2012). Consequently, there is no doubt that humans can be potentially exposed to CECs through the unexpected pathways (Lu *et al.*, 2011).

Concerning the human exposure to CECs, the octanol-water partition coefficient (K_{ow}) and the solubility in water (S_w) should be considered. While the a target compound with a log K_{ow} lower than 4 is considered as hydrophilic, that with a log K_{ow} higher than 4 is hydrophobic (Meffe *et al.*, 2014). Pan *et al.* (2009) examined that hydrophobic contaminants such as diclofenac (log K_{ow} 4.51), estradiol (log K_{ow} 4.01), gemfibrozil (log K_{ow} 4.77) and mefenamic acid (log K_{ow} 5.12) generally showed the high sorption affinity particularly onto organic matters.

Table 1. List of general pharmaceutical and personal care products (Esplugas *et al.*, 2007; Kim *et al.*, 2009; Liu *et al.*, 2013)

PPCPs	Categories	Compounds	
Pharmaceuticals	Analgesics and anti-inflammatory drugs	Acetaminophen	Acetylsalicylic acid
		Antipyrine	Aspirin
		Diclofenac	Ethenzamide
		Fenoprofen	Hydrocortisone
		Ibuprofen	Indomethacin
		Ketoprofen	Mefenamic acid
		Naproxen	Propyphenazone
		Paracetamol	Triamcinolone
	Antibiotics	Ampicillin	Chloramphenicol
		Ciprofloxacin	Clarithromycin
		Erythromycin	Nalidixic acid
		Norfloxacin	Ofloxacin
		Roxithromycin	Sulfadiazine
		Sulfadimethoxine	Surfadimidin
		Sulfamethoxazole	Trimethoprim
	Antiepileptic drugs	Carbamazepine	Primidone
	Artificial sweeter	Aspartame	Cyclamate
		Saccharin	
	β-blockers	Atenolol	Metoprolol
		Propanolol	Sotalol
	Blood lipid regulators	Bezafibrate	Clofibrate
		Gemfibrozil	
	Cytostatic drugs	Cyclophosphamide	Ifosfamide
	Steroids & Hormones	Ethinyl estradiol (EE2)	Estradiol (E2)
		Estriol	Estrone (E1)
		Norethisterone	Norgestrel
		Testosterone	17-*β*-estradiol
	Stimulant	Caffeine	
	X-ray contrast media	Diatrizoate	Iomeprol
		Iohexol	Iopamidol
		Iopromide	
Personal care products	Antimicrobial agents/Disinfectants	Triclocarban	Triclosan
	Artificial sweetener	Acesulfame	Sucralose
	Cosmetic	Propylparaben	
	Flame retardants	Polybrominated diphenyl ethers (PBDEs)	
		Tri(2-chlorethyl) phosphate (TCEP)	
	Insect repellants	N,N-diethyl-m-toluamide (DEET)	
	Preservatives	Parabens	
	Sunscreen UV filters	Benzophenone	
		2-ethylhexyl-4-trimethoxycinnamate(EHMC)	
		4-methyl-benzilidine-camphor(4MBC)	
	Synthetic musks/Fragrances	Acetophenone	Galaxolide (HHCB)
		Indole	Isoborneol
		Isoquinoline	Nitromusks
		Methyl salicylate (tri) ethyl citrate	
		Toxalide (AHTN)	
		3-methyl-1(H)-indole (Skatole)	

4. Removal of CECs using MBRs

While a conventional activated sludge (CAS) treatment process was limited to remove emerging contaminants including PPCPs (Spring *et al.*, 2007), the application of MBR is expected to remove CECs at a higher efficiency than a CAS system (Clara *et al.*, 2005). There have been remarkable progress in the application of MBR technologies to wastewater treatment and reclamation, resulting in smaller footprint, higher separation efficiency and less sludge production (Neoh *et al.*, 2016). Further, advanced MBR technologies also become more attractive to overcome shortcomings such as lower removal rates of certain CECs, membrane fouling and energy consumption.

For example, Mascolo *et al.* (2010) reported that advanced oxidation processes (AOPs) and electrocoagulation processes with MBR provided the higher removal efficiency (20%-60% higher than conventional MBR) in pharmaceutical wastewater. Hybrid moving bed biofilm reactor-MBR (hybrid MBBR-MBR) contributes to reduce the concentration of suspended solids for membrane fouling mitigation without efficiency loss for treatment by allowing plastic carriers attached with microorganisms to freely move in the bioreactor (Leyva-Díaz *et al.*, 2013). Osmotic MBR has brought several advantages *e.g.* better water quality production and lower energy consumption. conventional MBR is not able to effectively remove some persistent hydrophilic contaminants, while osmotic membrane bioreactor can retain any micro-organic compounds through longer contact time for biodegradation (Tan *et al.*, 2015). Osmotic MBR can also minimize the use of energy through the osmotic driving force supplied by a draw solution (Wang *et al.*, 2014).

In this study, we reviewed a wide range of CECs removal performance, particularly elimination of PPCPs, using different MBR processes. As a result, the overall removal ranges of 27 CECs (analgesics and anti-inflammatory: acetaminophen, diclofenac, ibuprofen, indomethacin, ketoprofen, mefenamic acid, naproxen, propyphenazone; antibiotics: erythromycin, ofloxacin, roxithromycin, sulfamethoxazole, trimethoprim; antiepileptic drugs: carbamazepine; *β*-blocks: atenolol, metoprolol, propanolol, sotalol; blood lipid regulators: bezafibrate, gemfibrozil; steroids/hormones: estriol, testosterone; stimulant: caffeine; antimicrobial agent/disinfectant: triclosan; flame retardant: Tri(2-chloroethyl) phosphate (TCEP); synthetic musks/fragrances: galaxolide (HHCB), toxalide (AHTN)) by various MBR operating conditions and filtration have been summarized in Table 2. Acetaminophen, ibuprofen, estriol and caffeine are the well-treated contaminants (> 99%) regardless of the types of membrane films and modules (*e.g.* nanofiltration, ultrafiltration, hollow-fiber type and flat-sheet module), hydraulic retention time (HRT) and sludge retention time (SRT). However, indomethacin (13-65%), mefenamic acid (7.2-74.8%) and TCEP (0.3%) are not well-removed or have the comparably huge gap between different elimination rates. Despite of the same contaminant, there are significant differences in the removal efficiency as the treatment method and operation condition. For example, erythromycin, one of antibiotics, was removed about 4.5% when it was treated by hollow fiber module, whereas it was well-removed (about 90%) through hollow-fiber module with 12-hr of HRT and 72-day of SRT (Kim *et al.*, 2007; Reif *et al.*, 2008). Radjenović *et al.* (2009) and Chon *et al.* (2011) reported that naproxen, used as analgesic and anti-inflammatory drug, was removed over 78% with micro-filtration and nano-filtration. However, Radjenovic *et al.* (2007) demonstrated that more than 99% of naproxen was eliminated using two flat-sheet module with 14-hr of HRTs. Erythromycin and carbamazepine have the wide range in the removal efficiency, 4.5-91.0% and 4.4-93.0% respectively. Hence, it is clear that each chemical compound has different optimal condition for effective elimination, despite same MBR configuration and operation is applied. For example, analgesics/anti-inflammatory drugs (*i.e.* acetaminophen and ibuprofen), steroids/hormones (*i.e.* estriol and testosterone) and stimulant (*i.e.* caffeine) are relatively well-removed, while antimicrobial agent (TCEP) is difficult to be removed by the MBRs.

Regarding membrane types, micro-filtration, ultra-filtration and nano-filtration were mainly used in the studies we reviewed. In the case of diclofenac, sulfamethoxazole and carbamazepine, the removal efficiency by nano-filtration (97.0%, 90.0%, 93.0% respectively) are generally higher than ultra-filtration MBRs (32.9-50.6%, 61.4%, 4.4-12.5% respectively). However, the increase in removal efficiency of a PPCP are not necessarily correlated with the decrease in pore size (micro>ultra>nano) of membrane types. For instance, the removal efficiencies of naproxen were 87.5-93.9% (micro-filtration), 83.5% (ultra-filtration) and 78.0% (nano-filtration), while atenolol was removed by about 64.1-89.3%, 57.0-82.0% and 85.0% using micro-, ultra- and nano-filtrations, respectively.

SRT can be one of well-known parameters to evaluate the removal efficiency of aquatic contaminants during the treatment process. Tambosi *et al.* (2010) performed two membrane bioreactor pilot plants to remove highly consumed six pharmaceuticals (acetaminophen, ketoprofen, naproxen, roxithromycin, sulfamethoxazole and trimethoprim), particularly applying two different SRTs of 15-day and 30-day. As a result, higher removal ratios were observed for the longer SRTs due to the longer contact time for biodegradation. The other study showed that adsorption of contaminants into the sludge is directly associated with the sludge concentration in a MBR, since the higher concentration of sludge provides additional adsorption sites to contaminants (Schäfer *et al.*, 2002). For the further study, therefore, evaluation of the CECs removal efficiency according to the type/composition of sludge, and their physical characteristics/biological fate in MBR would be required for a better understanding on the adsorption and biodegradation mechanism of CECs in a MBR system.

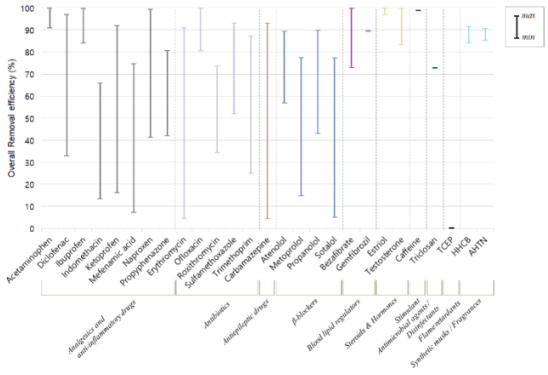

Figure 2. Overall range of removal efficiency applying different kinds of operational condition in MBR technology (summarized from Table 2)

Table 2. Removal efficiency of PPCPs in various operation condition with MBR systems

PPCPs	Removal %	Operation condition				Reference
		Type of module	MBR filter	HRTs (h)	SRTs (d)	
Pharmaceuticals						
Analgesics and anti-inflammatory drugs						
Acetaminophen	91.0	N.A	Nano-filtration	15.4	180±84.9	Chon *et al.* (2011)
	99.6-99.9	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	99.6	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	99.8-99.9	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
	> 99.9	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Diclofenac	32.9-50.6	N.A	Ultra-filtration	12-96	> 10	Clara *et al.* (2005)
	97.0	N.A	Nano-filtration	15.4	180±84.9	Chon *et al.* (2011)
	52.7-78.9	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	87.4	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	44.3-80.9	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Ibuprofen	96.9-99.2	N.A	Ultra-filtration	12-96	> 10	Clara *et al.* (2005)
	97.4-99.8	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	99.8	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	84.0-98.0	Hollow-fiber	N.A	12	72	Reif *et al.* (2008)
	97.9-99.8	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
	98.3	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Indomethacin	20.8-62.0	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	46.6	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	13.5-65.9	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Ketoprofen	16.2-71.6	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	91.9	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	23.4-64.3	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Mefenamic acid	16.8-64.2	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	74.8	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	7.2-63.8	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Naproxen	78.0	N.A	Nano-filtration	15.4	180±84.9	Chon *et al.* (2011)
	87.5-93.9	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	99.3	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	83.5	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
	41.2	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Propyphenazone	48.5-80.5	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	64.6	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	42.0-79.4	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Antibiotics						
Erythromycin	67.3	Two flat-sheet		14	Infinite*	Radjenovic *et al.* (2007)
	91.0	Hollow-fiber	N.A	12	72	Reif *et al.* (2008)
	4.5	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Ofloxacin	92.4-98.0	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	80.5-99.9	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Roxithromycin	34.4-73.5	N.A	Ultra-filtration	12-96	> 10	Clara *et al.* (2005)
Sulfamethoxazole	61.4	N.A	Ultra-filtration	12-96	> 10	Clara *et al.* (2005)
	90.0	N.A	Nano-filtration	15.4	180±84.9	Chon *et al.* (2011)
	68.6-93.0	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	60.5	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	52.0	Hollow-fiber	N.A	12	72	Reif *et al.* (2008)
	64.6-92.2	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
	70.1	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Trimethoprim	46.1-87.3	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	36.0	Hollow-fiber		12	72	Reif *et al.* (2008)
	25.0-70.0	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Antiepileptic drugs						
Carbamazepine	4.4-12.5	N.A	Ultra-filtration	12-96	> 10	Clara *et al.* (2005)
	93.0	N.A	Nano-filtration	15.4	180±84.9	Chon *et al.* (2011)
	9.0	Hollow-fiber	N.A	12	72	Reif *et al.* (2008)
β-blockers						
Atenolol	85.0	N.A	Nano-filtration	15.4	180±84.9	Chon *et al.* (2011)
	64.1-89.3	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	65.5	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	57.0-82.0	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Metoprolol	14.9-73.8	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	58.7	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	29.5-77.4	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Propanolol	65.4-89.8	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	43.1-87.9	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)

PPCPs	Removal %	Operation condition				Reference
		Type of module	MBR filter	HRTs (h)	SRTs (d)	
Sotalol	29.0-77.2	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	5.1-55.7	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Blood lipid regulators						
Bezafibrate	77.3-96.4	N.A	Ultra-filtration	12-96	> 10	Clara *et al.* (2005)
	80.2-99.8	Flat-sheet	Micro-filtration	15	N.A	Radjenović *et al.* (2009)
	95.8	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
	72.9-98.5	Hollow-fiber	Ultra-filtration	7.2	N.A	Radjenović *et al.* (2009)
Gemfibrozil	89.6	Two flat-sheet	N.A	14	Infinite*	Radjenovic *et al.* (2007)
Steroids & Hormones						
Estriol	> 96.9	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Testosterone	> 83.3	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Stimulant						
Caffeine	98.9	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Personal care products						
Antimicrobial agent/Disinfectant						
Triclosan	73.0	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Flame retardant						
TCEP	0.3	Hollow-fiber	N.A	N.A	N.A	Kim *et al.* (2007)
Synthetic musks/Fragrances						
Galaxolide (HHCB)	84.1-91.6	N.A	Ultra-filtration	12-96	> 10	Clara *et al.* (2005)
Toxalide (AHTN)	85.3-90.7	N.A	Ultra-filtration	12-96	> 10	Clara *et al.* (2005)

N.A: Not available

** No discharge of sludge*

5. Conclusion

A membrane bioreactor (MBR) is an advanced system that combines a membrane and a conventional activate sludge (CAS) system. Many studies have demonstrated that a MBR is more effective to remove CECs including PPCPs than a CAS system. More recently, MBR techniques and their integrated module have been adapted for beneficial elimination of emerging contaminants. In this study, we have aimed to review the removal efficiency of PPCPs according to MBR technologies applied with different modules. Hence, overall range of removal efficiency has been compiled for better understanding of wastewater compositions containing CECs. As a result, we came up with well-removed contaminants (*e.g.* acetaminophen, ibuprofen, estriol and caffeine); rarely-treated contaminants (*e.g.* TCEP) as well as huge range of removal efficiency (*e.g.* erythromycin and carbamazepine) using different types of MBRs. In general, analgesics/anti-inflammatory drugs (*i.e.* acetaminophen and ibuprofen), steroids/hormones (*i.e.* estriol and testosterone) and stimulant (*i.e.* caffeine) seem to have higher elimination rates compare to the other PPCPs groups, whereas antimicrobial agent (*i.e.* TCEP) is hardly treated in a MBR system.

Removal of CECs in a MBR system can be affected by various factors such as the type of modules, SRTs, HRTs, dilution factors, and the plant configuration. Based on the findings of this study, overall removal ranges that are useful as an indicator the performance of MBR systems were compiled according to MBR modules and filters adapted. The operational conditions for removal efficiency of PPCPs in a MBR system can vary because of biological fates, physical characteristics and biodegradation of the PPCPs, which are influenced by environmental factors in the system. By increasing SRT or the sludge concentration in a MBR, more PPCPs could be efficiently eliminated from the effluent, because long SRT gives enough time that more PPCPs could bind to the sludge in MBR for the biodegradation (Spring *et al.*, 2007). In addition, higher sludge concentration provides more adsorption sites to contaminants (Holbrook *et al.*, 2002; Tambosi *et al.*, 2010). Thus, relation between SRT, sludge concentration and MBR configuration could be important to develop effective a MBR system.

In this study, twenty-seven PPCPs were reviewed, even though there are numerous sort of CECs caused by both as pharmaceuticals and daily usage for personal cares. However, the result derived from this review regarding removal efficiency of PPCPs by MBR systems could provide fundamental information to assess a suitable MBR treatment systems for CECs and to investigate CECs in a MBR. Further, more detailed studies, for example, biodegradable fate of the frequently used PPCPs, combined or individual influence of operation conditions in a MBR system and catabolic enzymes through microbial communities should be needed. Studies on the removal of CECs using integrated MBRs should be carried out at laboratory scale to obtain information on their removal kinetics and mainly on the formation and degradation of by-products. Moreover, the fate and transport of CECs through the MBR treatment is required for effective elimination of CECs in MBR system.

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