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Progress in Emerging Contaminants Removal by Adsorption/Membrane Filtration-Based Technologies: A Review

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ABSTRACT

This paper discussed the removal and the remediation of emerging contaminants (ECs) in water using adsorption and membrane filtration as a single and hybrid system. The classifications, sources, effects, detection techniques, and available technologies for ECs removal were discussed. Next, an overview of both adsorption and membrane filtration processes in terms of materials, separation mechanisms, factors affecting their performances, and their applications for ECs removals was provided. It was followed by a comprehensive review of the combination of the membrane and the adsorption processes with other physical, chemical, or biological treatments. Finally, progress in research on a hybrid system between membrane filtration and adsorption was discussed. The combination included adsorption as the pre-treatment, integrated adsorption/membrane filtration system, or adsorption as the post-treatment. Generally, the hybrid systems showed improved performance than a single system. Nonetheless, further studies are recommended for applications of those systems on a wide range of ECs removals and the scale-up issue.

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1. INTRODUCTION

Water bodies have been polluted not only by conventional pollutants but also by emerging contaminants (ECs). With advances in detection techniques, ECs have been found in the water even at low concentrations (ng/L to mg/L, or lower). The presence of ECs is not regulated yet; hence they are not appropriately monitored. Compounds belonging to ECs are extensive in numbers. The numbers are also developing with the discovery of new compounds. ECs are classified into pharmaceuticals and personal care products (PCPs). Their presence in water bodies is getting more attention from scientists, government agencies, and society. They negatively impact ecology and have drawbacks on human health and aquatic organisms (Rasheed *et al*., 2019).

Based on the SCOPUS database using keywords of “emerging contaminant,” 6326 publications on ECs from 2011 until 2020. The number of publications increased annually. Most of them were published by institutions based in the United States and China, as shown in **Figure 1**.

Production, usage, and application of chemicals and pharmaceuticals are usually associated with long-term environmental pollution and health problems (Kümmerer, 2011). When consumed, they can accumulate in humans, invertebrates, and other living organisms and persist in the food chain (Rodriguez-Narvaez *et al*., 2017). Some examples of major human/environmental health concerns of ECs include hormone activities, skin, brain, nervous system disruption, cancer, ecological toxicity, persistency, and accumulation (Smital, 2008).

The emergence of new contaminants has been reported globally. In European surface waters, pharmaceuticals were frequently detected in ppb level, in which there were 12 high-risk and 17 medium-risk compounds (Zhou *et al*., 2019b). In East and Southeast Asian countries, antibiotics were found in surface water (at concentrations of <1 ng/L – µg/L with median values of 10 - 100 ng/L). Yet, the lack of antibiotics monitoring in surface waters makes it difficult to know their distribution (Anh *et al*., 2020). In the Central Mexican Pacific, pharmaceuticals classified as non-steroidal anti-inflammatory drugs have also been detected. They included diclofenac, naproxen, ketorolac, and ibuprofen. Moreover, these compounds were also detected in muscle tissues of 14 fish species, which could end up in bioaccumulation in the environment and organisms (Arguello-Pérez *et al*., 2019). In another study, the presence of ECs in water bodies was found in Coruña, Galicia (NW Spain). From 53 target compounds, 19 compounds at concentrations of > 0.1 µg/L were detected in wastewater. The traditional wastewater treatment plant (WWTP) could not remove these chemicals efficiently (Rodil *et al*., 2012). The compounds with the highest concentration were ibuprofen, salicylic acid, and UV filter benzophenone. Some compounds (i.e., ibuprofen, tri (2-chloroethyl) phosphate, diphenyl phosphate, benzophenone-4, 2-phenyl benzimidazole-5-sulphonic acid, tri (chloropropyl) phosphate, atenolol, diethylhexyl phosphate, tri-n-butyl phosphate, and diclofenac) were also present at concentrations of 20-200 ng/L.

In many countries in Asia, North America, and Europe, several categories of ECs (i.e., anticonvulsants/antidepressants, antibiotics, antifungal/antimicrobial agents, steroid hormones, nonsteroidal anti-inflammatory drugs, beta-adrenoaceptor, blocking agents, X-ray contrast media, UV filters, stimulants, anti-itching drugs, insect lipid regulating drugs, repellents, plasticizers, artificial sweeteners) have also been detected in WWTPs (Tran *et al*., 2018).

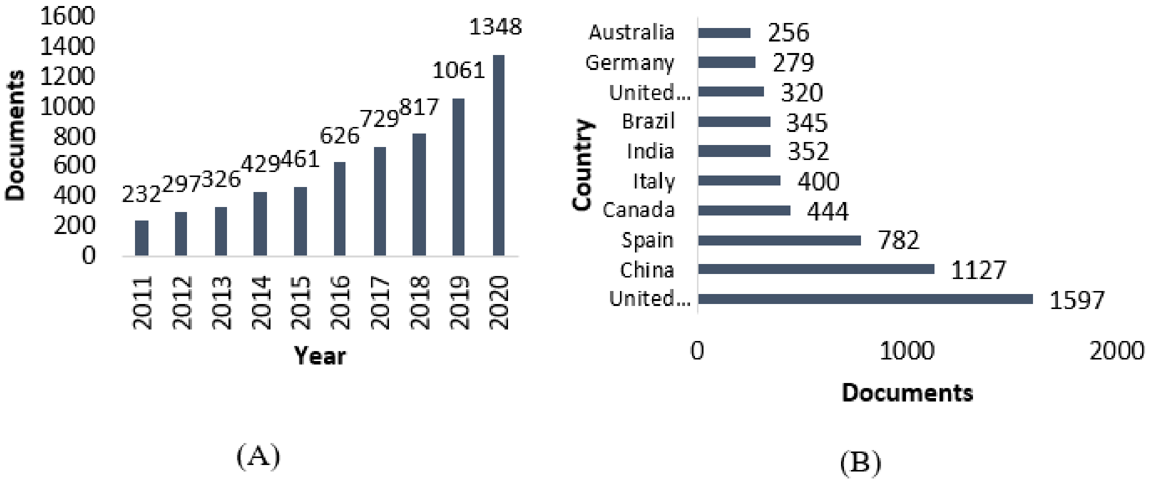

Figure 1. (A) The number of publications in SCOPUS by year (A) and by country (B) obtained using “emerging contaminant” as a keyword (Source: SCOPUS, search on 19th August 2021).

Conventional treatment technologies such as activated sludge, coagulation, sedimentation, flocculation could not thoroughly remove ECs (e.g., antibiotics, x-ray contrast media, beta-blockers, anticonvulsants) (Rasheed *et al.,* 2019; Tran *et al.,* 2018). Therefore, advanced technologies (e.g., membrane filtration, oxidation, adsorption) either as standalone or in combination have been assessed (Patel *et al.,* 2019; Rodriguez-Narvaez *et al.,* 2017). Adsorption has been widely used to remove hazardous inorganic and organic materials (Ahmed *et al.,* 2015). It is simple, cost-effective, and offers high removal efficiencies (Gopal *et al.,* 2014). In adsorption, adsorbent and adsorbate (contaminant) interact either physically or chemically. Other essential aspects of adsorption include adsorbent type, modeling, mode of operation, and regeneration (Dotto & McKay, 2020).

Membrane filtration is attractive for ECs removal. It offers high removal capability, low energy requirement, ease of scale-up, rapid kinetics, and a small carbon footprint. However, it still imposes several challenges such as membrane fouling, limited lifespan, insufficient rejection, chemical resistance, additional treatment of concentrates, and lack of tools for modeling and simulations (Van der Bruggen *et al.,* 2008).

The hybrid system comprising membrane filtration and adsorption is thus of interest which combines the advantage of both processes. Moreover, a hybrid system can be applied to overcome the shortcomings of each technology (Dhangar & Kumar, 2020). Both adsorption and membrane filtration can be combined with other treatments (i.e., physical, biological, or chemical) to improve their performances.

This review reported a systematic analysis of membrane filtration and adsorption-based technologies for ECs removals. In addition, this review discusses ECs classification, sources, effects, analysis, and general information about available technologies for ECs removals.

2. EMERGING CONTAMINANTS (ECs)

The advances in analytical techniques using gas chromatography or liquid chromatography-mass spectrometry (GC-MS/LC-MS) allow the detection of polar compounds such as pharmaceuticals, metabolites, and transformation products at low concentrations that could not be analyzed beforehand. These groups of compounds are called ECs. ECs are found in the environment at very low concentrations (in µg/L or lower). Hence, they are often also called organic micro-pollutants (Kümmerer, 2011). Henceforth, the term EC is used in this review.

2.1. Classification of Emerging Contaminants

There is no comprehensive definition and a complete list of compounds classified as ECs (Kümmerer, 2011). Each study categorized ECs into different classes. For instance, ECs were ranked into algal toxins, biocide and their transformation products, bioterrorism and sabotage agents, industrial chemicals, PCPs, and pharmaceuticals. Another report categorized ECs into pesticides, pharmaceuticals, PCPs, food additives, x-ray contrasting agents, steroids and surfactants, flame retardants, industrial compounds, and veterinary medicines (Qureshi *et al.*, 2020).

In general, the term ECs refers to three main categories. The first category consisted of substances that have recently entered the environment, such as industrial additives. The second category included substances that may have been in the environment for a long time but have recently detected and attracted much attention, such as pharmaceuticals. The third category was a group of substances that have been known, but their potential adverse effects on the environment and organisms have only recently been realized. The ECs categories that have attracted more attention and always existed in the ECs categorization in various studies are pharmaceuticals and PCPs.

The compounds classified into pharmaceuticals and PCPs have different physical and chemical properties. Pharmaceuticals and PCPs are manufactured to improve animals/humans' health and enhance human life quality. Pharmaceuticals include antibiotics, steroids, diuretics, non-steroid anti-inflammatory drugs, stimulant drugs, analgesics, antimicrobials, beta-blockers, antiseptics, hormones, lipid regulators, illicit drugs (e.g., amphetamines, cocaine). In contrast, PCPs include cosmetics, sunscreen agents, fragrances, domestic insect repellents, personal hygiene products, food supplements, and their metabolites, as well as transformation products. Some lotions and shampoos can contain up to 10-20 compounds (i.e., dyes, surfactants, preservatives, and others). Most of the PCPs are rinsed and drained into the sewage (Kümmerer, 2011).

2.2. Sources of Emerging Contaminants

The primary sources of ECs are wastewater, sewage sludge, municipal solid waste, households, livestock and poultry, hospital and industrial effluents, and urban runoff (Sophia & Lima, 2018). The paths of ECs reaching the water bodies are illustrated in **Figure 2.**

Most ECs were originated from industries, hospitals, households, animal husbandries, and urban runoff and flowed into WWTPs. Physical and chemical processes in WWTPs produce a sludge containing heavy metal concentrates and organic compounds. For example, Southeast Spain (Almeri'a) municipal WWTP had sludge that contained ECs. The analysis showed that there were 62 types of ECs detected (Klamerth *et al.*, 2013).

Pharmaceuticals and PCPs also entered the environment through drug residues excretion from the human body that later flowed into sewerage; externally used drugs or PCPs; or expired/unused pharmaceuticals/PCPs disposed of in the trash. Moreover, ineffective treatment in WWTPs also led to ECs discharge into rivers and ecosystems.

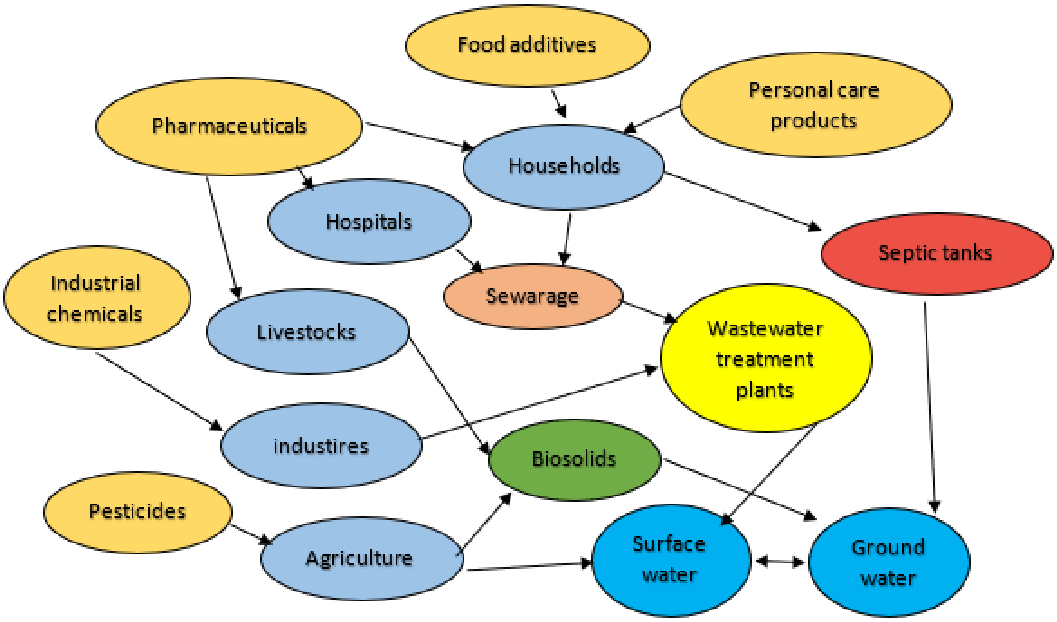

Figure 2. Sources and pathways of emerging contaminants.

2.3. Adverse effect of emerging contaminants

A summary of potential human health risks, bioaccumulation, toxic effects, and regulatory status of some ECs frequently found in the environment is shown in **Table 1.** ECs have shown either substantial, probable, or limited evidence on human/environmental health problems. Pharmaceuticals have shown strong evidence of congenital disabilities, developmental delays, hormone activity, and endocrine system (including liver). While, they have probable and limited evidence on the accumulation in wildlife and/or people, cancer, respiratory system, skin, reproduction and fertility, wildlife and environmental toxicity, brain, and nervous system. The toxicities of pharmaceutical products toward plants and alga are shown in **Table 2.**

Sulfachlorpyridazine, oxytetracycline, and diclofenac are toxic to duckweed/plant with EC50 of 2.33 mg/L, 4.92 mg/L, and 7.5 mg/L, respectively. While tiludronate, propranolol and metoprolol are toxic to algae with EC50 of 13.3, 5.8, and 7.3 mg/L, respectively.

ECs have shown potential adverse effects and need to be analyzed further. Some significant concerns of ECs on human's health include glucose metabolism, infertility, abortion, cholesterol, weight, fetal growth, allergy, cancer, uric acid, semen quality, acute effects (Lei *et al.*, 2015), nervous system syndrome, memory disruption, anemia, hypertension, carcinogenic, non-degradable, toxic in nature, oxidative stress, cardiovascular disorder, reproductive disorder, reduce IQs, and apoptosis (Rasheed *et al.*, 2019). The toxicity effect of ECs (i.e., furosemide and tramadol) was detected on Prague's surface water and WWTP to *Artemia salina*. They had a lethal concentration (LC50) of 225.01 mg/L and 14,000 mg/L for furosemide and tramadol, respectively (Diaz-Sosa *et al.*, 2020). Further research and regulation of ECs and their detrimental effects are needed to protect human health and ecology.

Table 1. The priority of major human/environmental health issues of the most prominent categories of ECs (Smital, 2008).

Health concern	**Chemical family**												
	Nonculturable biological	**Bisphenol & BADGE**	**Perfluoro-chemicals (PFCs)**	**Perchlorate**	**Pharmaceuticals**	**Nanomaterials**	**Fragrances (nitro- and polycyclic**	**Polychlorinated naphthalenes**	**Phthalates**	**Triclosan**	**Polybrominated diphenyl esthers**	**A Brominate dioxins and furans**	**Alkyl phenols**
Persistent accumulates in wildlife and/or people		++	++		+		++	++	++	++	++	+++	+ +
Birth defects and developmental delays		+	+	+	+++				+++		++		+
Cancer		+	+		+		+		+		+	+	
Respiratory system	+++				+	++			+++				+
Hematologic (blood) system				+									
Immune system (including sensitization and allergies)	+	++	+++			+	+		+++	+		+	
Skin	++	+			+		+	+++	+	+			
Reproduction and fertility		++	+++		++		+++	+	+++	+	+++	++	+ + +
Hormone activity		+++	+++		+++				+++		+++		+
Endocrine system (including liver)			+	+++	+++		+			+			+
Wildlife and environmental toxicity		++			++	+	+	++		+	+		+ + +
Kidney and renal system		+	+++										
Brain and nervous system			++		+	+			+++		+++		
USA, Canada, EU list of priority compound								√	√	√			√
OSPAR list		√					√	√	√		√		√

Note: Weight of evidence:+++ strong, ++ probable, + limited. Oslo and Paris Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR)

Table 2. Pharmaceutical product toxicity toward plant and alga (Basheer, 2018).

Pharmaceutical group	Substance	Organism affected		Long-Term Exposure (mg/L)	
		Duckweed/ Plant	Alga	EC 10	EC50
Anti-bacterial (sulfonamide)	Sulfamethoxazole	√		0.011	
	Sulfamethazine	√		>1.00	
	Sulfachlorpyridazine	√			2.33
	Sulfadimethoxine	√		>0.044	
Nicotine metabolite	Cotinine	√		> 1.00	
Anti-protozoal	Metronidazole		√	2.03	
Oestrogen	Ethinylestradiol		√	0.054	
Anti-bacterial (tetracycline)	Tetracycline	√		0.23	
	Oxytetracycline	√		0.788	
		√			4.92
	Chlortetracycline	√		0.036	
	Doxycycline	√		0.055	
Anti-bacterial (macrolide antibiotic)	Erythromycin	√		>1.00	
	Lincomycin	√		>1.00	
	Roxithromycin	√		>1.00	
	Tylosin	√		>1.00	
Fluvoxetine	Sertraline	√		>1.00	
Anti-diabetic (biguanide)	Metformin		√		>320
		√			110
Antihyperlipoproteinemic	Clofibric acid		√	5.40	
			√		115
		√			12.5
Anti-hyperlipidemic	Atorvastatin	√		0.085	
Anti-hypertensive	Captopril		√		168
		√			25
Bone resorption Inhibitor	Tiludronate		√		13.3

Table 2 (Continue). Pharmaceutical product toxicity toward plant and alga (Basheer, 2018).

Pharmaceutical group	**Substance**	**Organism affected**		**Long-Term Exposure (mg/L)**	
		Duckweed/ Plant	**Alga**	**EC 10**	**EC50**
			√		36.6
Non-steroid antiInflammatory drug	Naproxen		√		>320.0
		√			24.2
	Ibuprofen	√		>1.0	
		√			22.0
	Acetaminophen (paracetamol)	√		>1.0	
			√		72.0
	Diclofenac	√			7.5
			√		315.0
β-adrenergic receptor blocker	Propranolol		√		5.8
		√			114.0
	Metoprolol		√		7.3
		√			>320.0
Anti-bacterial	Trimethoprim	√		>1.0	
	Cephalexin	√		>1.0	
	Ciprofloxacin	√		0.106	
	Norfloxacin	√		0.206	
Anti-bacterial (aminogycoside)	Neomycin	√		>1.0	
	Streptomycin	√		>1.0	
Anti-epileptic	Carbamazepine		√		74.0
		√		>1.0	
		√			25.5

The presence of ketorolac, ibuprofen, estradiol, diclofenac, and pentachlorophenol ranged under toxic concentrations in the coastal zone of Mexico. It was found from an ecotoxicological analysis. These ECs can contaminate surface water and soil and later cause endocrine changes in organisms living in this ecosystem (Arguello-Pérez *et al.*, 2019). Endocrine disruption chemicals interfere endocrine system by mimicking, disrupting, and blocking the hormone's function (Bolong *et al.*, 2009).

2.4. Emerging Contaminants Analysis

The data on the ECs presence in water bodies are still scarce and limited. It is partly due to the unavailability of instruments for analysis. Hence, improvement of ECs analysis methods is significant in ECs' detection. Generally, ECs analysis can be classified based on the targeted compound, non-targeted compound, and unknown compound. The LC-MS/MS is commonly used to analyze ECs (Agüera *et al.*, 2013). Nonetheless, research is still needed, especially for detecting non-target/unknown ECs. New methods are required to allow a more efficient, low-cost, and time-saving analysis (Rodriguez-Narvaez *et al.*, 2017).

Figure 3 shows targeted compounds and non-targeted compounds analysis using LC-MS. The LC-QTOF-MS/MS method showed high-confidence results. In the targeted compound analysis, the high sensitivity of TOF-MS offered intelligent screening and quantitative analysis. In the study of non-targeted compounds, the data were obtained through processing data based on some parameters (i.e., full scan mode mass accuracy; MS/MS library data of spectral purity score grade; structure description and mass error of fragment ion). Although the commercial MS/MS library can accurately and simultaneously confirm the non-target compounds, it is recommended to use the library that contains 20,000-30,000 compounds to achieve higher accuracy (Bueno *et al.*, 2012). A pharmaceutical product is the standard category of ECs. The general steps for analyzing the ECs include sample collection, filtration, extraction, derivatization (if necessary), and LC-MS or GC-MS analysis (Fatta *et al.*, 2007).

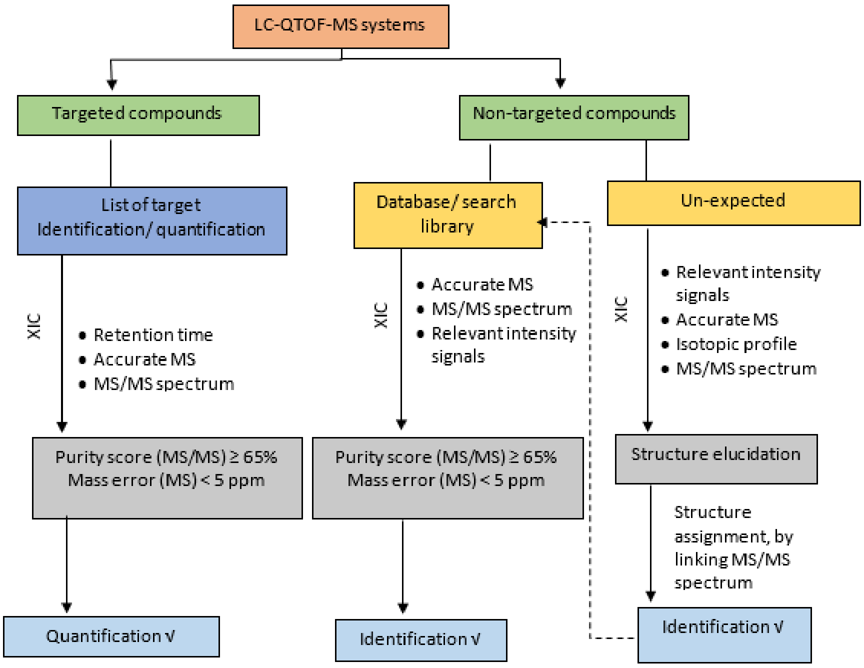

Figure 3. Targeted and non-targeted compound analysis (Bueno *et al.*, 2012).

2.5. Technologies for Emerging Contaminants Removal

ECs treatment technologies typically consist of physical, biological, and chemical treatments (**Figure 4).** Physical treatment does not employ a biological or chemical agent and does not change the biochemical properties of the ECs. Biological treatment is a process that involves living organisms or enzymatic degradation. Lastly, chemical treatment is a process that involves chemical reactions (Ahmed *et al.*, 2021).

Some effective treatments for ECs removal are biological treatment, membrane filtration, adsorption, and advanced oxidation processes. The biological treatment is the most widely used because of its availability, low cost, and environmentally friendly. However, it was reported to be less effective due to its poor biodegradability. In chemical treatment, ECs are converted into more stable or into compounds that are biodegradable through mineralization or conversion into inorganics (e.g., H_2O, CO_2, and N_2). It is the most effective in eliminating various ECs, although posing some drawbacks. In several studies, physical treatment has also been shown effective in pharmaceutical removals (Dhangar & Kumar, 2020).

Recently, technologies for ECs removal have been developed (Rodriguez-Narvaez *et al.*, 2017). Each technology offered different performances for a specific type of ECs. For instance, phosphorized carbonaceous adsorbent showed good performance for some ECs, with a removal efficiency of about 99%. Continuous ultrafiltration (UF) showed removals of less than 30%. Activated sludge and constructed wetland showed removal efficiencies of 10-95% for 14 types of ECs. Sono-fenton+photocatalysts showed different diethyl phthalate and dimethyl phthalate removal efficiencies of 35 and 47%, respectively.

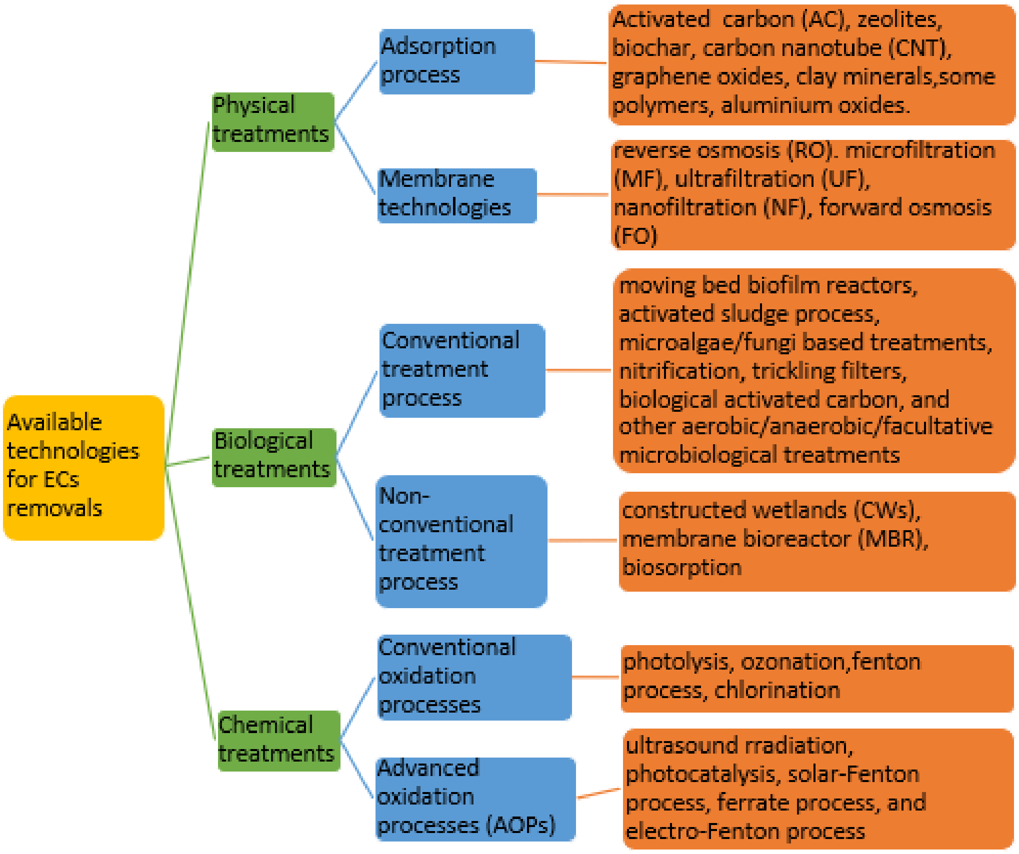

Figure 4. Available technologies for ECs removals (Dhangar & Kumar, 2020).

3. ADSORPTION

Adsorption is a surface phenomenon in which the solute (adsorbate) and the porous surface (adsorbent) interact through a particular mechanism (Rashed, 2013). Adsorption is one of the promising methods for ECs removal due to its simple operation, low cost, and high efficiency (Sophia & Lima, 2018). Several advantages of adsorption include (Rasheed *et al.*, 2019): high selectivity, high efficiency, facile processing, no harsh chemicals used, high productivity, cost-effective, easy post-treatment, and less disruptive.

Like other methods, adsorption also faces some challenges. They include the cost aspect of the entire adsorption process; only a few studies available on the fixed-bed system typically used in the industry; the ability to regenerate and reuse adsorbents in an environmentally friendly manner; the need to develop isotherm models for multi-component systems; and the application of adsorption methods in actual cases (Dotto & McKay, 2020).

The main stages in the adsorption studies for ECs removal are shown in **Figure 5**. The first stage involves choosing raw material for the adsorbent. It can come from various sources such as natural, synthetic, waste, etc. The second stage is the development of adsorbent material through physical or chemical processes, such as changing the particle size, acidification, structure modification, etc. The third stage is the adsorbent characterization that reveals the physical and chemical characteristics of the adsorbents. The fourth stage is the selection of ECs types and concentrations due to the wide range of ECs. The fifth stage is sorption studies, which include optimizing parameters that affect adsorbent performance and investigating the adsorption equilibrium and the adsorption kinetic. The final stage is desorption and regeneration studies, where the effectiveness of adsorbent is investigated when used many times.

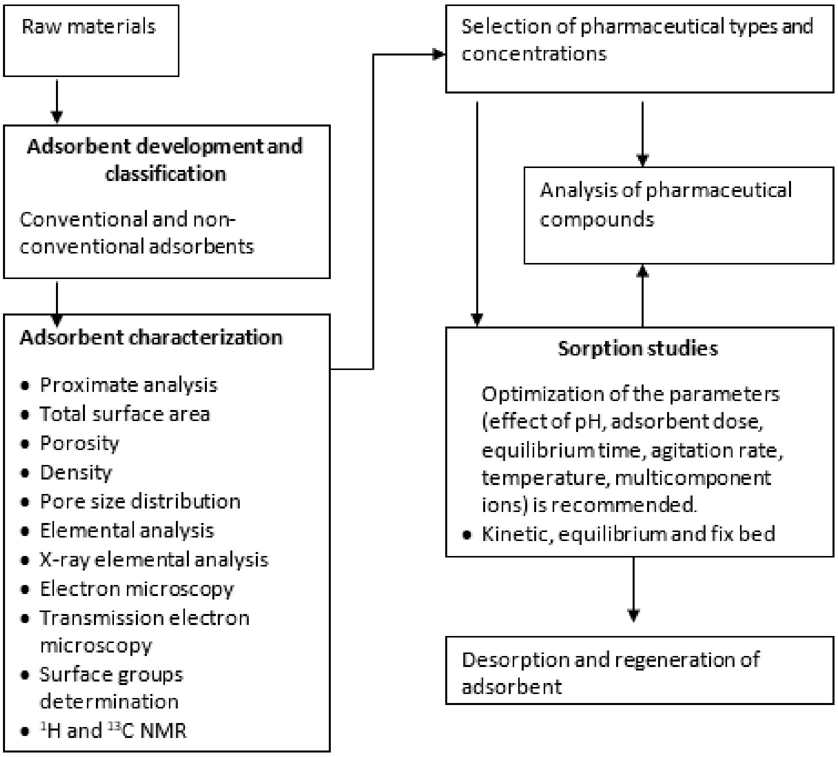

Figure 5. Adsorbent selection protocol for pharmaceutical product removals (Patel *et al.*, 2019).

3.1. Adsorbent Materials

The adsorbent's selection, characterization, and development are essential aspects in ECs removal via the adsorption process. Some critical criteria are cost and adsorbent availability, chemical stability, good physicochemical and textural characteristics, fast kinetic rate, mechanical stability, ability to regenerate or reuse, and high adsorption capacity (Dotto & McKay, 2020).

Adsorbents are generally classified into five categories (Singh *et al.*, 2018): natural, biomass, industrial waste, agriculture waste, and synthetic adsorbents. Low-cost adsorbents include the ones prepared from readily available materials (e.g., peat, natural zeolite, chitin/ chitosan, clay, coal, wood, etc.), industrial wastes (e.g., saw dust, sunflower stalk, nuts and fruits peels, straw, corncob waste, etc.), and agriculture wastes (e.g., palm oil ash, fly ash, blast furnace, red mud, shale oil ash, bagasse, and bagasse pith, bagasse fly ash, etc.).

An adsorbent is characterized in terms of absorption capacity, porosity, and pore structure. Natural adsorbents (e.g., clay, zeolite, charcoal) are cheap and abundant and can be modified to increase their adsorption capacity (Rashed, 2013). Agriculture wastes-based adsorbents have a porous structure and desirable functional groups (e.g., hydroxyl, carboxyl). They are also low-cost and renewable resources. Modification of agriculture waste-based adsorbents resulted in enhanced performance (Dai *et al.*, 2018). Industrial wastes such as fly ash, red mud, waste slurry are also potential adsorbents. The adsorption capacity of industrial waste adsorbents depends on adsorbent characteristics, adsorbate concentration, and adsorbent modification (Ahmaruzzaman, 2011).

Biosorbents can be classified as living (e.g., bacteria, algae, fungi, yeast) or non-living organic materials (e.g., wastes of agricultural and food industries) (Shamim, 2018). Biosorption is a physical or a chemical process (i.e., adsorption, electrostatic interactions, micro-deposition, chelation, ion exchange) that occurs in the cell wall before the adsorbate is assimilated. It has high selectivity and efficiency, as well as low cost.

Another category is synthetic adsorbents, such as nano adsorbents. It has a high surface area and consists of several categories based on their roles in the adsorption process related to surface properties and their functionalities. They are nanoparticles, silicon nanomaterials, nanofibers, xerogels and aerogels, polymer-based nanomaterials, nano clays, and carbonaceous nanomaterials (Khajeh *et al.*, 2013).

3.2. Adsorption Mechanism

Adsorbent and adsorbate interact in two ways: chemi- and physisorption. In the former, the adsorbate forms a monolayer. The adsorbate interacts with the external surface of the adsorbent, enters the internal pores through pore diffusion, and interacts with the active sites (Khulbe & Matsuura, 2018). The adsorbent and the adsorbate form a new electronic configuration through electron sharing or electron transfer, and chemical interaction occurs. In the latter, adsorbent and adsorbate interact through the Van der Waals force in solid-liquid or solid-gas systems (Khulbe & Matsuura, 2018). Electrostatic forces or Van der Waals forces occur without the transfer or sharing of electrons. The adsorbate retains its identity, although a surface force field may deform it.

The illustration of the adsorption process is shown in **Figure 6.** The process is related to pore surface areas, active sites**,** chemisorption, and/or physisorption. Cheng *et al.* (2021) explained that modified biochar could adsorb some ECs with different mechanisms: hydrophobic interaction (for tylosin), ion exchange (for tetracycline and endocrine-disrupting chemicals), electrostatic interaction, hydrogen bonding, and functional groups (for tetracycline and endocrine-disrupting chemicals), and π-π bond interaction (for ciprofloxacin, tetracycline, sulfate, and endocrine-disrupting chemicals).

The adsorption equilibrium state is reached when there is minimum solute adsorption from the bulk to the adsorbent. The adsorption amount (*qe*, mmol/g) under equilibrium is given in Equation [1].

$$qe = \frac{V\,(Co - Ce)}{M} \qquad (1)$$

where *V* is the volume of the solution (*L*); M is the adsorbent mass (*g*); *Co*, *Ce* are adsorbate concentrations in the initial and equilibrium condition, respectively (Rashed, 2013).

The adsorption isotherm is a function of the equilibrium concentration of the solution at a constant temperature. It is defined as the percentage of adsorbate per unit weight of adsorbent. Usually, the adsorption isotherm is given in the Langmuir or Freundlich model (Román *et al*., 2020), as shown in Equations [2] and [3], respectively.

$$qe = \frac{Q0 \text{ x } Ce \text{ x } Kl}{1 + (Ce \text{ x } Kl)} \qquad (2)$$

$$qe = K_fCe\ 1/n \qquad (3)$$

where *Kl* (L/mg) is the Langmuir constant; Q_0 (mg/g) is the monolayer maximum adsorption capacity; *qe* (mg/g) is the measured adsorption at *Ce*; K_f (mg/g)(L/mg)$^{1/n}$ is the Freundlich constant representing the adsorption capacity, and n is the constant depicting the adsorption intensity.

Adsorption kinetic shows the retention rate or solute release from solution to adsorbent's surface at certain conditions (i.e., the dose of adsorbent, pH, temperature, and flow rate). Examples of adsorption kinetics are pseudo-first-order, intra-particle diffusion, pseudo-second-order, and Elovich models.

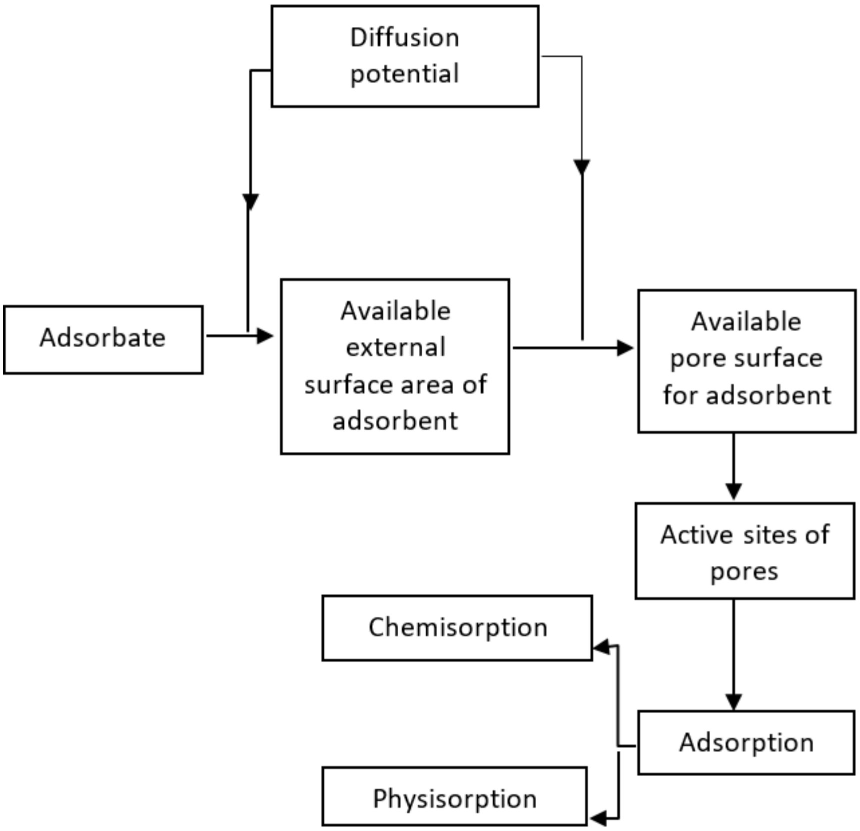

Figure 6. Adsorption pathway (Singh *et al*., 2018).

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3.3. Factors Affecting Adsorption

Internal and external factors influence the adsorption process. The internal factors include structure features (e.g., molecular weight, functional groups, morphology, pore size, surface area, etc.). The external factors include adsorbent dosage, adsorbate concentration, contact time, pH, temperature, and competitiveness.

Those factors need to be optimized to achieve desirable performances. A high concentration of adsorbate leads to lower adsorption efficiency. High temperature is preferred for endothermic reactions. Contact time relates to the saturation of all adsorbent active sites. The presence of competitive ions can also compete on the active site of the adsorbent. An adsorbent's chemical properties (e.g., functional group, ionization, steric effect) also influence the adsorption capacity (Román *et al.,* 2020). Pore size and surface area affect the number of available active sites on the adsorbent surface. There is also a relationship between pore size and surface area, in which smaller pore size results in higher surface area.

3.4. Adsorption for Emerging Contaminants Removal

Adsorption is a promising method for ECs removal. Research has shown the application of various types of adsorbents (e.g., nano-adsorbent, biochar, activated carbon, composite adsorbent) for ECs removals. The adsorbent surface can be modified chemically or thermally to convert it from a functional material to a multifunctional nano-adsorbent and increase its capacity to absorb ECs (Sophia & Lima, 2018).

Agricultural wastes have a prospect to be applied as an adsorbent. As such, it simultaneously reduces environmental waste and converts waste into functional and valuable materials. Physical and chemical modifications have been proved to improve their adsorption performances (Dai *et al.,* 2018). After being modified to increase their porosity and surface area, most of the agricultural and industrial wastes were used as adsorbents. These modifications included nano-structuring, carbonization, activation, milling, sieving, derivatization, and grafting techniques (Mo *et al.,* 2018).

The summary of reports on ECs removals using adsorbent is shown in **Table 3**. The performance of agricultural and industrial waste-based adsorbents for ECs removals depends on the type of ECs, modification of adsorbent material, and several external factors.

Peñafiel *et al.,* (2019) investigated ciprofloxacin removal by corn cob and rice husk-based adsorbent. The removals obtained by the corn cob was 56.3%, and rice husk was 59.7%. The optimum doses were 2 g/L for the corn cob and 6 g/L for the rice husk-based adsorbent. The performance was strongly influenced by pH with an optimum value of 6, and the mechanism was well described by the Freundlich isotherm and pseudo-second-order kinetic models.

Oyehan *et al.,* (2020) investigated phenol removal by a mesoporous fly ash-based adsorbent. It was coated by an ultrathin film of polydiallydimethyl ammonium chloride. The highest removal was about 95%. The mechanism was fitted by Freundlich, Langmuir and Temkin isotherm models with physisorption mechanism.

Biomass-based adsorbents (i.e., living or dead microorganisms and their components) can be utilized for the biosorption of ECs. However, the commercialization of this group of adsorbents is still limited. Various physical and chemical modifications led to higher costs and created new environmental problems (Fomina & Gadd, 2014).

Bankole *et al.,* (2020) investigated Ibuprofen, diclofenac, celecoxib removals using two wood-rot fungi: *Laetiporus sulphurous* and *Ganoderma applanatum*. The combination of *Laetiporus sulphurous* and *Ganoderma applanatum* biomassed showed better performance than the standalone.

Table 3. Emerging contaminants removal using adsorbents.

Category of adsorbent	Type of adsorbent	EC adsorbate	Adsorption Capacity	Adsorption isotherm model	Reference
Natural adsorbent	Granulated cork	Diclofenac, phenol, 2,4-dichlorophenol, methyl paraben, pentachlorophenol carbamazepine, ketoprofen, triclosan,2-nitrophenol and 2-chlorophenol, naproxen	2-chlorophenol (45%), 2,4-dichlorophenol (75%), methyl paraben (50%), 2-nitrophenol (55%), phenol (20%), pentachlorophenol (100%), naproxen (2%),triclosan (100%).sodium diclofenac (100%), ketoprofen (57%), carbamazepine(50%).	Freundlich and Langmuir	(Mallek *et al.*, 2018)
	Activated carbons obtained from peat	poly(acrylic acid)	265mg/g	-	
	magnetic poly(N-isopropylacrylamide)/chitosan hydrogel	hydrophilic sulfamethoxazole (SMZ) and hydrophobic bisphenol A (BPA).	SMZ = 33.95 mg/g BPA = 747.53 mg/g	Freundlich (for SMZ) Freundlich and Slips (for BPA)	(Wiśniewska & Nowicki, 2020)
	Thermally modified bentonite clay	Ciprofloxacin	114.4 mg/g	Langmuir	
	Zeolite	Phenol	23.3, 24.9, 23.8, 34.5, mg/g at 55, 35, 45, and 25^{o}C, respectively	Freundlich	(Zhou *et al.*, 2019a)
Agriculture waste	Grapefruit peel based biochar	Tetracyclin	37.92 mg/g	Langmuir	(Antonelli *et al.*, 2020)
	Hydrochar-derived magnetic carbon composite from sawdust	Roxarsone	588.2 mg/g	Langmuir	
	Corn cob	Ciprofloxacin	56.3%	Freundlich	(Yousef *et al.*, 2011)
	Rice husk	Ciprofloxacin	59.7%	Freundlich	

Table 3 (Continue). Emerging contaminants removal using adsorbents.

Category of adsorbent	Type of adsorbent	EC adsorbate	Adsorption Capacity	Adsorption isotherm model	Reference
	Activated carbons from peach stones	Carbamazepine, diclofenac, caffeine	caffeine (260 mg/g), carbamazepine (335 mg/g). Diclofenac adsorption capacity is lower than caffein	Sips	(Yu et al., 2020a)
	Modification and magnetization of rice straw derived biochar	tetracycline	98.33 mg/g	Langmuir, Freundlich, Temkin	
	Waste tea residue	Hydralazine hydrochloride pharmaceutical pollutant	131.63 mg/g	Langmuir and Freundlich	(Qureshi *et al.*, 2020)
	Pistachio shell coated with ZnO nanoparticles	tetracycline	95.06 mg/g	Freundlich	
Industrial waste	Mesoporous fly ash	phenol	~95%	Freundlich, Langmuir and Temkin	
	Biochar derived from hydrothermal carbonization of sugarcane bagasse	sulfamethoxazole	400 mg/g	Freundlich	(Peñafiel *et al.*, 2019)
	Bagasse fly ash	2-picoline	98%	Langmuir and Redlich–Peterson	
	Activated carbon from effluent beverage industry treatment plant sludge	ibuprofen, ketoprofen, and paracetamol	ibuprofen (105 mg/g), paracetamol (57 mg/g), and ketoprofen,(145 mg/g)	Sips	(Torrellas *et al.*, 2015)

Table 3 (Continue). Emerging contaminants removal using adsorbents.

Category of adsorbent	Type of adsorbent	EC adsorbate	Adsorption Capacity	Adsorption isotherm model	Reference
Biomass adsorbent	Chars from reprocessed wet olive mill waste pitted, olive tree pruning, olive stone.	Diclofenac, ibuprofen, triclosan.	Diclofenac (64%), ibuprofen (43%), triclosan (98%).	Freundlich	
	Novel modified red mud with polypyrrole	Phosphorus (P) and diclofenac (DCF)	DCF/P (115.7 mg/g), DCF (195 mg/g).	Freundlich	(Dai *et al*., 2020)
	Unburned materials obtained by combustion in a conical spouted bed of four types of vegetable biomasses of forestry residues, grass, and food industry.	amoxicillin	1:33,107 mg/g	Langmuir	
	Wood-rot fungi; *Laetiporus sulphureus* (LS, *Ganoderma applanatum* (GA).	Ibuprofen, diclofenac, celecoxib	ibuprofen (95%), diclofenac (96%), celecoxib (98,96%)	Langmuir and Temkin	(Patil *et al*., 2019)
	Yeast, *Saccharomyces cerevisiae,*	Phenol	27 mg/g	Langmuir	
	Pseudomonas aeruginosa immobilizedFe_3O_4-multiwalled carbon nanotubes bioadsorbent	2,4,6-trinitrophenol	100 mg/g	Langmuir	(Mohammed & Kareem, 2019)
	Fungal Strains from Municipal Wastewater	diclofenac	>98%	-	

Table 3 (Continue). Emerging contaminants removal using adsorbents.

Category of adsorbent	Type of adsorbent	EC adsorbate	Adsorption Capacity	Adsorption isotherm model	Reference
Nano-adsorbent	composite iron nano adsorbent.	Ibuprofen	= 92%	Freundlich, Langmuir,Dubinin-Radushkevich. Temkin and	(Oyehan *et al.*, 2020)
	Carbon nanotubes impregnated with metallic nanoparticles.	Glyphosate-based herbicides	43.66 mg/g	Sips	
	Surface modification of aluminum hydroxide Nanoparticles	emerging pesticide lindane	93.68%	A two-step adsorption model	(Prasannamedha *et al.*, 2021)
	Magnetic graphene/chitosan nanocomposite	2-naphthol	169.49 mg g−1	Freundlich	
	Polypyrrole-functionalized magnetic Bi_2MoO_6 nanocomposites	ketoprofen and indomethacin	ketoprofen (87.03%) and indomethacin (86.24%)	Langmuir	(Lataye *et al.*, 2008)
	magnetic silica-based nanoadsorbents	Carbamazepine, ibuprofen, diclofenac,	Diclofenac (83.0%) ibuprofen (63.5%), carbamazepine (< 3%).	Freundlich model	

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Dalecka *et al.,* (2020) investigated diclofenac removal from municipal wastewater by fungal strains: *Aspergillus luchuensis*, *A. luchuensis indicate*, and *Trametes versicolor*. The diclofenac removals by *Aspergillus luchuensis*, *A. luchuensis* and *Trametes versicolor* were >99.9, >98, and 100%, respectively. Each fungal strain required different pHs and incubation periods for achieving a good biosorption process.

Nano-adsorbent could remove ECs even at low concentrations (µg/L) under optimum temperature and pH. Moreover, the nano-adsorbent dose was low, and the time required for ECs removal was short (1-15 minutes). However, the application of nano-adsorbent is still limited. Therefore, further studies are still required to enhance the adsorption capacity, security level of materials, and application in various conditions (Basheer, 2018).

Nguyen *et al.* (2020) investigated lindane (pesticide) removal by using modified nanomaterial of aluminum hydroxide as adsorbent. The removal efficiency was 93.68% by applying an adsorbent dosage of 25 mg/L, adsorption time of 60 min, ionic strength of 10 Mm NaCl, and pH six.

4. MEMBRANE FILTRATION

Membrane-based processes have long been used in water and wastewater to remove microorganisms, organic materials, including emerging contaminants and other particles (Gómez-Espinosa & Arizmendi-Cotero, 2019). The membrane material can separate those constituents under specific driving forces. Various pressure-driven membrane processes (i.e., nanofiltration (NF), UF, reverse osmosis (RO)), and osmotically driven forward osmosis (FO) have been studied for ECs removal. The removal efficiency followed the order of the typical pore sizes being the highest for the one with the smallest one: RO≥FO> NF> UF. Retention of ECs by the membrane is affected by the size/ steric exclusion, hydrophobic/hydrophilic interactions (adsorption), electrostatic forces, or a combination thereof. Polar ECs have less retention than less polar ones. UF is less effective for ECs removals but can act as a pre-treatment before FO, NF, or RO. Further studies are needed to study the transfer mechanism and evaluate the effects of draw solution type, concentration, permeation rate, and foulant accumulation (Kim *et al.,* 2018).

4.1. Materials

Most of the commercial membranes were prepared from synthetic polymers. The materials often used for UF and MF are polysulfone, polyvinylidene fluoride, polyacrylonitrile, and polypropylene, including recently inorganic/ ceramic, which has been of great interest. The most common material for RO membrane is polyamide, while the materials for NF membrane are polysulfone, polyimide, and ceramic. The application of membrane technology for ECs removals is challenged by various limitations on membrane physicochemical characteristics and other factors that influence the separation process. Some membrane materials such as polymers, inorganic membranes, and others need further tests (Kárászová *et al.,* 2020).

Membranes can have a porous or dense structure. The separation of ECs using membranes is related to the solubility and diffusivity of ECs and the applied pressure (Gómez-Espinosa & Arizmendi-Cotero, 2019). Detailed classification of membrane materials can be found elsewhere (Pendergast & Hoek, 2011).

Generally, different membrane materials (e.g., polyamide, ceramic, cellulose triacetate) are pretty similar in the rejection of ECs, only differ in some ways. In RO membrane prepared from cellulose triacetate had a less significant effect on the electrostatic interaction than polyamide. The rejection of positively charged ECs is lower than negatively charged ECs when separated using ceramic membranes. In polyamide NF/RO membrane, the effect of positive/negative charge on ECs was not observed.

4.2. Separation Process

In membrane-based separation, membrane material acts as a physical barrier for pollutants/contaminants in the feed. The retention of contaminants is highly affected by the relative size of the pores to the contaminants. A membrane process requires a driving force (e.g., electrical force, concentration difference, pressure) to allow separation of a particular substance, as schematically illustrated in **Figure 7**. Membrane with tiny pores requires high transmembrane pressure to separate specific components from the feed and vice versa (Madhura *et al.*, 2018).

The transport through the membrane pores is influenced by the material, physicochemical characteristics, and membrane morphology. In general, the ECs-membrane interaction mechanism consists of size exclusion, electrostatic and/or hydrophobic interactions. The size exclusion mechanism occurs through sieving, where large ECs molecules are retained while smaller ECs molecules pass through the membrane pores. The relative sizes of the ECs and the free volume of the active membrane layer is a factor that determines the efficiency of the separation.

Electrostatic interaction is related to electrostatic attraction or repulsion between the ECs and the membrane material. If the membrane surface is negatively charged, then ECs with negative charged ECs will be rejected, and vice versa. Moreover, neutral ECs are not affected by electrostatic interactions.

Hydrophobic interactions between hydrophobic ECs and hydrophobic membranes can also affect the separation process. Hydrophobic ECs do not fully dissolve but suspend in with water. When presented in the feed, they could be absorbed into the hydrophobic membrane material and concentrated on its surface. The use of membranes (especially MF and UF) for ECs removals can be done in a combined process (Gómez-Espinosa & Arizmendi-Cotero, 2019). UF is effective for pathogen removal, and MF is effective for particulates removal. RO and NF operate at higher transmembrane pressure and can remove contaminants up to 0.0001 µm and 0.001 µm, respectively. The RO and NF processes can remove a wide range of contaminants and requiring more pre-treatments. NF and RO effectively remove ECs such as pesticides, pharmaceutical products, endocrines, algal toxins, and other similar substances.

Tables 4 and **5** compare NF and RO membranes and their performance for ECs removal. In ECs removals, RO has higher removal efficiency than NF. Generally, NF and RO membranes are different in some parameters, including adequate pore size, energy consumption, micropollutant removal, membrane availability, etc.

4.3. Factor Affecting Performance

The main parameter to judge membrane performance is permeability that is affected by membrane pore size, surface chemistry, morphology, porosity, thickness, etc. The increase in membrane thickness causes a decrease in permeation flux because of longer flow path across the membrane matric (Kárászová *et al.,* 2020). Membrane permeability is also influenced by hydrophilic/ hydrophobic characteristics and morphology of membrane surface. Higher porosity leads to lower intrinsic membrane resistance (Shamsuddin *et al.,* 2016).

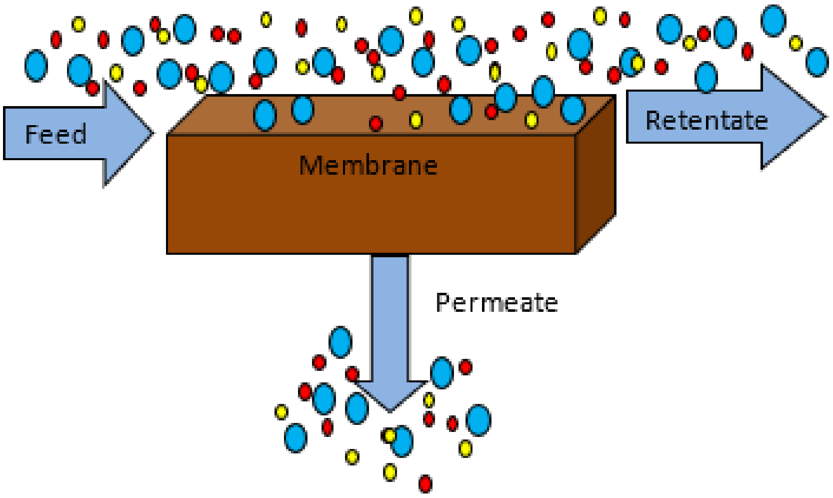

Figure 7. Schematic diagram of membrane separation process (Madhura *et al*., 2018).

Table 4. A general comparison between NF vs. RO (Yangali-Quintanilla *et al*., 2011).

Condition	NF	RO
Effective pore size (range)	1–2 nm	< 1 nm
Energy consumption	Low to moderate	High
Removal of salts	Moderate	High
Post-treatment for the addition of salts (ions)	Not necessary	Necessary
Removal of contaminants (micropollutants)	Low to high, depending on “tight” or “loose” membrane and type of contaminant	Low to high, depending on the type of contaminant
Membrane availability	Low to moderate	Plentiful
Types of membranes by manufacturer	Few	Many

Transport through a membrane is affected by both the feed solution characteristics and the membrane properties (Hammami *et al.,* 2017). In NF, ECs rejection is influenced by several factors. They are EC properties (i.e., molecular size, charge, hydrophobicity, polarity, diffusivity, solubility), membrane properties (i.e., surface charge, hydrophobicity, permeability, pore size), and membrane operating conditions (i.e., rejections/recovery, transmembrane pressure, flux, water feed quality) (Bolong et al., 2009). The efficiency of ECs removal is influenced by both the physicochemical characteristics of ECs and the membrane properties.

Membrane properties affect the interaction of the feed constituents and the membrane surface. Membranes can attract or repel water (i.e., hydrophilic and hydrophobic surfaces). The nature of the feed solution can also affect the membrane fouling propensity. The increased foulant material concentration in the feed lowers the permeability. The permeation rate is proportional to the trans-membrane pressure, in which higher pressure leads to higher fluid force.

Table 5. Removal efficiencies of specific ECs by NF and RO membranes.

Type	Chemical	NF Removal (%)	RO Removal (%)
Pharmaceuticals			
Analgesic	Ibuprofen	98	>98
	Naproxen	23	>95
Antibiotic	Trimethoprim	22	90
Muscle relaxant	Diazepam	55	>95
Steroid	17β-estradiol (estrogen)	20	90
	Testosterone (androgen)	60	95
Personal Care Products			
Antimicrobial	Triclosan	45	>96
Insecticide	DEET	75	>90
Surfactant	Nonylphenol	>99	>99

4.4. Membrane Fouling

One of the main challenges of membrane-based filtration is membrane fouling (Van der Bruggen *et al.,* 2008). It is caused by substance deposition on the membrane surface and/or in the membrane pores (Madhura *et al.,* 2018). It lowers the membrane flux and could alter the membrane surface hydrophobicity through the formation of the cake layer, the surface charge, traces contaminant adsorption, and the overall surface roughness (Kárászová *et al.,* 2020). Membrane fouling is affected by the foulant material characteristics, such as the functional groups, overall structure, size, etc. It dictates its physical/chemical interaction with the membrane material. (Shamsuddin *et al.,* 2016).

Based on the nature of the foulant materials, foulant materials can be classified into living cells (biological), organics, particulates, and inorganics (Bokhary *et al.,* 2018). Several parameters also influence membrane fouling, namely membrane properties, feed characteristics, and operational parameters. These parameters must be considered when designing a membrane process (Bokhary *et al.,* 2018).

There are several mechanisms of membrane fouling: adsorption, pore blocking, cake layer, and gel-layer formation. Some of them mostly co-occur. In the adsorption mechanism, the membrane and the foulant material interact, forming particles monolayer on the surface of the membrane. The layer causes additional hydraulic resistance and eventually lowers the hydraulic throughput. In the pore-blocking mechanism, the foulant blocks the membrane pores either wholly or partially, depending on the particle's relative size to the membrane pore size. In the cake layer fouling, particle deposition causes additional hydraulic resistance on the surface of the membrane. The firmly attached cake forms gel-layer on the membrane surface (Madhura *et al.,* 2018).

Several methods can be implemented to reduce the severity of membrane fouling as summarized in **Figure 8,** namely pre-treatment of the feed, membrane modification, operation conditions, and cleanings (physical or chemical methods).

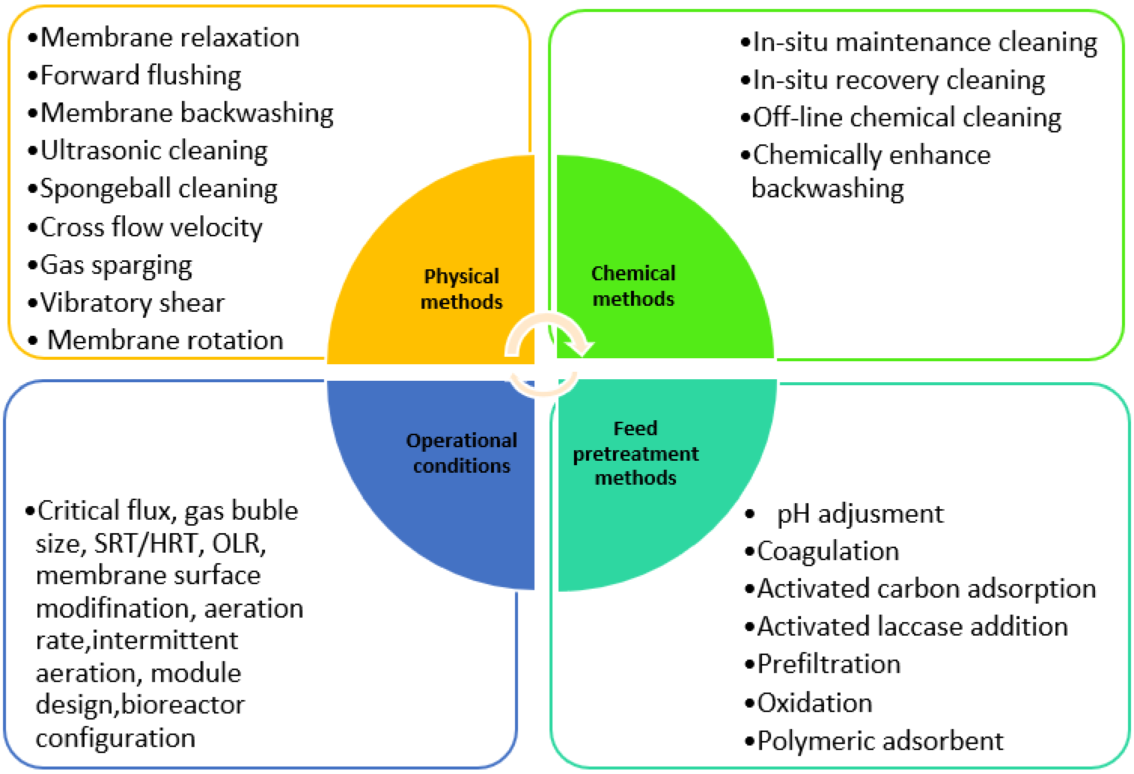

Figure 8. Strategies for membrane fouling control (Bokhary *et al*., 2018).

4.5. Application of Membrane Filtration for Emerging Contaminants Removal

Membrane-based processes (such as MF, UF, membrane bioreactor) have been proposed as the technologies ECs removals (Couto *et al.,* 2018). They have proven to offer good permeation and do not impose toxicity (i.e., excess chemicals) to the environment. Reports on membrane application for ECs removal showed that good selectivity could be achieved by implementing dense membrane (NF, RO, FO). Denser membrane (i.e., RO) had better selectivity in retaining ECs than more open pore membrane (i.e., NF). The mass transport through a dense membrane generally obeys the solution diffusion model. Porous membranes offer higher permeation but lower EC rejection (Kárászová *et al.,* 2020).

Table 6 summarizes the applications of various types of membrane processes for ECs removals. The application of membrane filtration for the separation or concentration of pharmaceuticals has been extensively investigated. Most of the studies employed commercial membranes (especially polyamide) with an NF process. RO membranes have an efficiency of ≥ 80% removals for most pharmaceuticals. For the UF membrane, the results varied widely (Shojaee Nasirabadi *et al.,* 2016).

Table 6. Application of membrane technology for emerging contaminant removals.

Process	Membrane material	ECs	Removal efficiency	References
UF	Polyether sulfone (PES)	39 high-occurrence CEC	< 30% except for amitriptyline (63%).	(Ferreiro *et al.*, 2020)
Polyelectrolyte multilayer (PEM) nanofiltration (NF)	Poly(sodium styrene sulfonate) + poly(diallyl dimethylammonium chloride)	Tetracycline, amoxicillin trihydrate, perfluorooctanesulfonic acid, perfluorooctanoic acid, and hydrochloride.	90%	(Wang *et al.*, 2021)
MBR–RO	RO: aromatic polyamide. MBR: flat sheet membranes (Kubota, porous size of 0.4 µm).	20 types of pharmaceuticals	> 99%	(Dolar *et al.*, 2012)
UF	Poly (vinylidene fluoride) (PVDF)-ZnO/Ag_2CO_3/Ag_2O nanocomposite membrane	Ibuprofen	5.27%	(Rosman *et al.*, 2020)
MF-FOMBR	FO: CTA-ES MF: Polyvinylidene fluoride	20 antibiotics	58.9-100%	(Qiu *et al.*, 2021)
UF	UF: CuO/TiO_2 ceramic	Ciprofloxacin	99.5%	(Bhattacharya *et al.*, 2019)
NF	NF: 270 and NF 90 (Filmtec–Minneapolis, MN). Thin-film (skin) of polyamide over a layer of polysulfone on a polyester support layer.	Norfloxacin	87-99.5%	(De Souza *et al.*, 2018)
NF	Chitosan-modified acrylic nanofiltration membrane	Diphenhydramine and mebeverine	diphenhydramine (97%) and mebeverine (~98%)	(Kamrani *et al.*, 2018)
NF	Polyamide Thin-Film Composite	Caffeine, theobromine, theophylline, amoxicillin, and penicillin G	amoxicillin (89%), caffeine (20%), theobromine (18%), penicillin G (70 %), theophylline (7%)	(Egea-Corbacho *et al.*, 2019)

Table 6 (Continue). Application of membrane technology for emerging contaminant removals.

Process	Membrane material	ECs	Removal efficiency	References
NF	Two thin film composite NF membranes: NF-200 and NF-90 (Dow-Filmtec), made of polyamide	18 ECs	ionic contaminants (97%), neutral contaminants (82%)	(Yangali-Quintanilla *et al.*, 2011)
RO	BW30-2540 made from polyamide thin-film composite	Caffeine, theobromine, theophylline, amoxicillin, and penicillin G	100%	(Lopera *et al.*, 2019)
RO	RE2521-SHF made from polyamide thin-film composite	Ciprofloxacin	> 90%	(Alonso *et al.*, 2018)
RO	ESPA2, ESPAB, andLFC3 made from polyamide thin-film composite	N-nitrosodimethylamine	80%	(Fujioka *et al.*, 2020)
FO	Polyamide thin film composite and polysulfone (PS)	24 ECs	> 93 %	(Salamanca *et al.*, 2021)
FO	thin film composite membrane with aquaporin proteins embedded in a polyamide active layer supported by a porous polysulfone support layer	21 ECs	> 80%	(Li *et al.*, 2021)
FO	thin-film composite membrane with aquaporin protein embedded in the poly-amide layer	N-nitrosodimethylamine (NDMA) and haloacetonitriles (HANs)	NDMA (31%), HANs (48–76%)	(Xu *et al.*, 2018)
FOMBR	cellulose triacetate	Ibuprofen	96.32%	(Yao *et al.*, 2021)
FO	commercial cellulose triacetate (CTA) based membranes and thin-film composite (TFC) polyamide-based membranes	Carbamazepine, diclofenac, ibuprofen and naproxen	naproxen (93%), Diclofenac (99%), ibuprofen (93%), carbamazepine (95%).	(Jin *et al.*, 2012)

Ferreiro *et al.* (2020) studied ECs removals in WWTP using the biological treatment, followed by UF as the tertiary treatment. The removals of 39 types of ECs using UF were ≤ 30%, except for amitriptyline (63.0%). UF showed good performance in other studies on phenol, atenolol, ciprofloxacin, amoxiline, and sulfamethoxazole removals (Bhattacharya *et al.*, 2019, 2020; Ali *et al.*, 2021; Shakak *et al.*, 2020).

Yangali-Quintanilla *et al.* (2011) compared NF and RO to remove 18 types of ECs. They found that the average removal efficiencies of neutral and ionic contaminants by NF were

82 and 97%, respectively. Meanwhile, average removal efficiencies by RO were 85 and 99%, respectively.

5. HYBRID SYSTEMS

Various treatment technologies (i.e., biological, physical, chemical treatments) have been investigated for ECs removal and showed good performance. However, hybrid systems have been developed due to challenges and limitations of each technology (e.g., contaminant sludge disposal, high retention time, high cost, limited removals of a wide range of ECs) (Dhangar & Kumar, 2020). Recently, studies of various hybrid systems for ECs removal have increased significantly (Ahmed *et al*., 2017). One of the hybrid systems is adsorption/membrane filtration-based technologies. This hybrid system has some advantages: rapid kinetic, low pressure-drop, better separation efficiency, easier control and handling, lower discharge volume, higher reusability, lower fouling rate (in some cases), and low-energy footprint, lower process cost, and potential use as bio-sorbents. In this section, an overview of the hybrid system involving adsorption or/and membrane is provided.

5.1. Adsorption-Based Hybrid Systems

Adsorption can be combined with biological treatments (e.g., activated sludge, MBR), chemical treatments (e.g., photo Fenton oxidation, ozonation), and other physical treatments (e.g., membrane filtration). **Table 7** shows a summary of various adsorption-based hybrid systems for ECs removal.

Tagliavini & Schäfer, (2018) evaluated polymer-based activated carbon adsorption+UF/NF for steroid removals. They compared adsorption+UF and adsorption+NF. The result showed that the role of adsorption in the adsoprtion+NF hybrid system was not significant. Nonetheless, the adsorption+UF hybrid system showed more potential due to its higher permeability and good performance, with a removal efficiency of > 90%.

Granzoto *et al*., (2021) investigated ECs removals (i.e., phosphate, tri-n-butyl phosphate, tris (chloropropyl), triphenyl phosphate) by using a hybrid system. It combined adsorption (granulated activated carbon (GAC)) and ozonation (O_3)/ UV-H_2O_2. They evaluated different configurations, namely UV/H_2O_2+GAC+UV/H_2O_2; O_3+GAC+O_3; UV/H_2O_2+GAC+O_3; O_3+GAC+UV/H_2O_2. The performance of the O_3+GAC+O_3 configuration showed a good result and was found to be cost-effective. This configuration could remove the ECs almost entirely with the removals from various units as follow: the first O_3 treatment (15%), GAC treatment (80%), and last O_3 treatment (100%).

Dwivedi *et al*., (2018) assessed a hybrid system combining Fenton and GAC to treat carbamazepine in raw wastewater. The process was optimized by Response Surface Methodology tools and found the optimum condition of Fenton pretreatment with the concentration of H_2O_2 (8.5 g/L) and pH (3.5). The maximum removal was 99.51 ±0.02%. This result was higher than the standalone Fenton treatment (with the removal of merely 49.39%).

Ferrer-Polonio *et al*., (2020) assessed a hybrid system consisting of adsorption (activated carbon) and biological (activated sludge) treatments. The targeted ECs were caffeine, ibuprofen, and acetaminophen. The adsorption process was effective for acetaminophen removal but not for caffeine and ibuprofen. The hybrid system of activated sludge + activated carbon could altogether remove the targeted ECs in 35 days.

Table 7. Application of adsorption-based hybrid systems for ECs removal.

Hybrid system	ECs	Removal efficiency	Reference
polymer-based activated carbon adsorptionwith UF/NF	Estradiol (E2)	> 90%.	(Tagliavini & Schäfer, 2018)
Ozone+ Granulated activated carbon (GAC)	Deuterated surrogate standard tris (phenyl) phosphate-D15 (TPHP-D15), Tri-n-butyl phosphate (TNBP), tris (chloropropyl) phosphate (TCIPP). triphenyl phosphate (TPHP)	100%	(Granzoto *et al.*, 2021)
Fenton pretreatment + granulated activated carbon (GAC)	Carbamazepine (CBZ)	99.51 ±0.02%.	(Dwivedi *et al.*, 2018)
Electrochemical+ Adsorption (granular activated carbon (GAC))	Iopromide (IPM), carbamazepine (CBZ), diatrizoate (DTR), DEET	DEET (40-57%), iopromide (22-46%), carbamazepine (15-34%) and diatrizoate (4-30%)	(Norra & Radjenovic, 2021)
sequencing batch reactor (SBR) + powdered composite adsorbent (CA)	Atenolol (ATN), ciprofloxacin (CIP) and diazepam (DIA)	DIA (95.5%), CIP (94.0%), ATN (90.2%)	(Mojiri *et al.*, 2020)
adsorption/photo Fenton oxidation+ Microbial Fuel Cell	Fumaric acid, succinic acid	40.8 %	(Civan *et al.*, 2021)
active carbon felt (ACF)+ electro-Fenton (EF)	Tetracycline	> 90%	(Zhang *et al.*, 2018)
Ozonation + activated carbon adsorption	28 ECs	80%	(Guillossou *et al.*, 2020)

Based on results summarized in **Table 7**, an adsorption-based hybrid system offered better performances than single systems. The combination between adsorption and biological/physical/chemical treatments shows higher removal efficiencies than a single system. The range of removal efficiency is quite diverse depending on the type of ECs and the treatment processes.

5.2. Membrane Filtration-Based Hybrid System

The applications of membrane filtration-based hybrid systems for ECs removal are shown in **Table 8**. Pathak *et al.*, (2018) assessed a combination of osmotic membrane bioreactor (OMBR) with MF to remove atenolol, caffeine, and atrazine. MF was required to solve the salt accumulation problems due to rejection by the FO, acting as the purging system. The process worked under oxic-anoxic conditions, and the performance was quite diverse for each targeted ECs. The highest removal was obtained for caffeine (94-100%), followed by atenolol (89-96%) and atrazine (16-40%). Atrazine removal was related to redox and microbial condition on the system.

Martínez *et al.*, (2013) removed ECs (i.e., nicotine, hydrochlorothiazide 4-acetamido antipyrine, sulfamethoxazole, ranitidine hydrochloride, nicotine) by a combination of photocatalytic oxidation and membrane filtration. The photocatalytic oxidation process was facilitated by the TiO_2 photocatalysis and Fe_2O_3/SBA-15 in H_2O_2 photo-Fenton. The membrane types were RO and NF. NF membrane showed a better choice for the hybrid system than the RO due to its lower energy footprint and higher flux while still offering good EC rejection. TiO_2 photocatalysis and Fe_2O_3/SBA-15 with H_2O_2 photo-Fenton showed ECs removal of 80-100%. Among all targeted ECs, nicotine has the lowest removal efficiency. However, Fe_2O_3/SBA-15 in H_2O_2 photo-Fenton showed better performance on nicotine removal than the TiO_2 photocatalysis system.

Chen *et al.*, (2019) assessed the hybrid process between UF and magnetic ion exchange resin (MIEX) for carbamazepine removal. This hybrid system exploited the UF advantage of turbidity treatment and MIEX advantage in EC removal. MIEX was used as pre-treatment. The system could reduce the secondary contaminant of resin and increase the membrane lifespan.

The removal efficiencies of this system were 25-79%, depending on the water turbidity. Data in **Table 8** suggest that membrane filtration-based hybrid systems offered a wide range of ECs removals. Some of them can achieve complete removal, while only achieved as low as 16%.

5.3. Membrane Filtration / Adsorption Hybrid Process

Membrane filtration and adsorption can be combined in many configurations. They include adsorption pre-treatment, integrated adsorption/ membrane systems (IAMPs), and adsorption post-treatment. The integrated adsorption/ membrane process can be done through (1) a low-pressure membrane combined with adsorption, (2) membrane adsorption bioreactor, and (3) membrane adsorption. Several studies assessed the performance of those configurations for ECs removal.

Wang *et al.*, (2020) conducted the experiments on a hybrid system of NF/UF+adsorption, in which PAC was used for the pre-treatment to ease the membrane fouling. The result showed that PAC pre-treatment could reduce membrane fouling caused by organic foulant in UF and NF membranes.

Table 8. Application of membrane filtration-based hybrid system for ECs removal.

Hybrid system	ECs	Removal efficiency	Reference
Osmotic membrane bioreactor + microfiltration	Caffein, atenolol, and atrazine	Atenolol (89–96%), Caffeine (94–100%), Atrazine (16–40%)	(Pathak *et al.*, 2018)
Nanofiltration + TiO_2 and Fe_2O_3/SBA-15 photo-Fenton	Sulfamethoxazole, diclofenac, hydrochlorothiazide 4-acetamidoantipyrine, nicotine, ranitinide hydrochloride	80-100%	(Martínez *et al.*, 2013)
magnetic ion exchange resin + UF	carbamazepine (CBZ)	35-79%	(Chen *et al.*, 2019)
Catalytic ozonation + membrane filtration (catalytic ceramic membranes, CCMs)	clofibricacid(CA), bisphenolA(BPA), benzotriazole(BTZ)	38%	(Lee *et al.*, 2019)
metal-organic frameworks + UF	Ibuprofen, 17α-ethinyl estradiol	53.2%	(Kim *et al.*, 2020)
advanced oxidation processes + UF	oxytetracycline	49%.	(Espíndola *et al.*, 2019)

Huang *et al.,* (2019) investigated a combination between activated carbon adsorption and NF to remove octyl phenol, diclofenac, and caffeine by involving coagulation as the pre-treatment. They compared the performances between adsorption+NF and NF+ adsorption configurations. The result shows the hybrid system performance is better than the single systems. The adsorption+NF had better performance than the NF+adsorption configuration with removals of targeted ECs of ≥ 95%.

Ivancev-tumbas & Hobby (2010) assessed the adsorption+UF system for carbamazepine and p-nitrophenol removals by employing the integrated configuration. They compared three types of adsorbents with different properties. The found the best adsorbent was the one with the smallest particle size. The removal efficiency was influenced by the density and the particle size of the adsorbent. The presence of coagulant improved the removal efficiency of the treatment system. The maximum removals of carbamazepine and p-nitrophenol were 40.0 and 30.7%, respectively.

5.4. Application of Hybrid System Combining Membrane Filtration and Adsorption for Emerging Contaminants Removal

The application of the hybrid system for ECs removal has great attention because it combines the advantages of each technology. There are several configurations of hybrid adsorption-membrane filtration (AD+M), as summarized in **Table 9.** Most of ECs treated by AD+M hybrid system were pharmaceuticals. The common adsorbent and membrane processes were activated carbon and UF, respectively. Generally, the processes were applied with pressure and stirring under various adsorbent loadings. The removal efficiencies were quite diverse, depending on ECs, adsorbent characteristics, membrane properties, operating conditions, and the process configurations.

Sharma *et al*., (2017) investigated M+AD for antibiotic removals**.** The hybrid system used the adsorption pre-treatment followed by the membrane filtration. The adsorbent was synthesized through co-precipitation resulting in modified layered adsorbent material. The membrane material was a low-cost MF ceramic with an average pore size of 1 µm and a porosity of 47%. The result showed that the optimum conditions were pH (7), ECs concentration (of 10 mg/L), and adsorbent dosage (1 g/L). This system could achieve the removal efficiency of 98.7% for norfloxacin and 94.6% for ofloxacin. PH conditions highly influenced the performance.

Sheng *et al*., (2016) compared adsorption and coagulation as the pre-treatment of UF. It was used to remove 16 ECs, namely bezafibrate, acetaminophen, gemfibrozil, sulfamethazine, triclosan, naproxen, acetaminophen, sulfamethoxazole, cotinine, caffeine, diclofenac, ibuprofen, metoprolol, sulfadimethoxine, trimethoprim, and carbamazepine. These ECs were collected from many WWTPs in the USA. The adsorbent was the PAC, and the coagulant was poly-aluminum chloride. The result showed that the hybrid systems (adsorption+UF and coagulation+UF) had better performances than the single systems (adsorption, coagulation, or UF only). Adsorption has better performance than coagulation pre-treatment. The average removal efficiencies of coagulation, UF, adsorption, coagulation+UF, and adsorption+UF systems were 7, 29, 50, 33, and 90.3%, respectively. These results showed that the hybrid system combining adsorption and UF, in which adsorption acted as the pre-treatment, had an excellent prospect for ECs removal.

Table 9. Application of hybrid system combining membrane filtration and adsorption for ECs removal.

ECs	Adsorbent	Membrane	Operating condition of adsorbent	Operating condition of the membrane	Removal efficiency	References
Ofloxacin (OFL), norfloxacin (NOR)	Ni-Al layered double hydroxide (LDH)	MF (ceramic membrane)	Adsorbent dose = 0.1 g antibiotic concentration 10 mg/L	Pressure = 34.47–172.36 kPa.	NOR (98.7%), OFL (94.6%)	(Sharma *et al.*, 2017)
Octylphenol, caffeine, diclofenac,	Activated carbon F400	NF-270	Adsorbent dose = 10 mg/L	transmembrane pressure= 100 psi; cross flow velocity= 0.27 mg/s.	> 95%	(Huang *et al.*, 2019a)
Ibuprofen (IBP), carbamazepine (CBM), 17 α-ethinyl estradiol (EE2)	Activated biochar	A commercial flat sheet polyamide UF membrane	Adsorbent dose = 10 mg/L initial concentration of ECs= 10 Mm.	stirring speed (300 rpm), trans membrane pressure (520 kPa (75 psi)	45.2%	(Kim *et al.*, 2019)
Bezafibrate, Gemfibrozil, Sulfamethazine, Naproxen, Sulfamethoxazolem, Caffeine, Diclofenac, Ibuprofen, Metoprolol, Sulfadimethoxine, Sulfathiazole, Triclosan, Trimethoprim, Acetaminophen, Carbamazepine, Cotinine,	Powdered activated carbon	UF membrane with MWCO 100 kDa	Adsorbent dosage = 100 ppm. Configuration: adsorption pre-treatment	Pressure = 6.9×103 kPa (1000 Psi). maximum flow rate is 22.7 L/min	90.3%	(Sheng *et al.*, 2016)
Sulphametoxazol, Carbamazepine, Diclofenac, Diuron, Erythromycin	Powdered activated carbon	UF with tight poly-ether sulfone	Adsorbent dose = 20 mg/l	Feed pump pressure regulates permeate low	> 81±13%	(Echevarría *et al.*, 2020)
Phenol	Cross-linked macronet polymer adsorbents	UF: AZW-1 model (GE)	Adsorbent dose = 1, 2, 3, and 4 g/L	The flow rate = 4 L/min	90%	(İpek *et al.*, 2012)

Table 9 (Continue). Application of hybrid system combining membrane filtration and adsorption for ECs removal.

ECs	Adsorbent	Membrane	Operating condition of adsorbent	Operating condition of the membrane	Removal efficiency	References
Octyl-phenol, diclofenac, caffeine	Activated carbon	NF-270	Adsorbent dosage = 10 mg/L	Trans membrane pressure (100 psi); crossflow velocity (0.27m/s)	>95%	(Huang *et al.*, 2019)
Carbamazepine, p-nitrophenol	Powdered activated carbon	UF membrane (non-ionic hydrophilic membrane)	Adsorbent dosage for p-nitrophenol removal= 5 or 10 mg/L Adsorbent dosage for carbamazepine removal is 0.3 mg/L	Flux = 20 L/m2/h Pressure = 10 bar	Carbama-zepine (40%) and p-nitrophenol (30.7%)	(Ivancev-tumbas & Hobby, 2010)
Phenol	Hypercrosslinked macronet polymer (Purolite MN 202 and MN 200)	UF	Adsorbent dosage for purolite MN 202 and MN 200 is 0.2 g/50mL solution and 0.1 g/50 ml solution, respectively. Stirring = 250 rpm	Air flow rate = 4 L/min	90%	(İpek *et al.*, 2012)
Naproxen, triclosan, paracetamol, amitriptyline, clozapine, caffeine, verapamil, DEET, gemfibrozil, sulfamethoxazole, atenolol, ketoprofen, simazine, trimethoprim, fluoxetine, primidone, triclocarban, carbamazepine, diclofenac	granular activated carbon	MF	Adsorbent dosage = 10 mg/L with replacement of 10%/day	Flux: 10 L/m2	80%	(Shanmuganathan *et al.*, 2017)

Shanmuganathan *et al*., (2017) investigated a combination of submerged membrane filtration-adsorption (M+AD) for removals of 19 ECs. They were naproxen, triclosan, paracetamol, amitriptyline, clozapine, caffeine, verapamil, DEET, gemfibrozil, sulfamethoxazole, atenolol, ketoprofen, simazine, trimethoprim, fluoxetine, primidone, triclocarban, carbamazepine, and diclofenac. These ECs were obtained from RO concentrate of water reclamation plant in Australia. The membrane-type was hydrophilic polyacrylonitrile MF. The adsorbent was GAC with a size of 300–600 µm. Most ECs were removed with a removal efficiency of 80% on day 1, except for sulfamethoxazole and DEET. The excellent performance was attributed to the electrostatic interaction between hydrophobic or neutral/positive charge ECs with the negative charge of GAC.

Generally, a combination between adsorption and membrane filtration shows high removal efficiency. However, some investigations still showed unsatisfied removal efficiencies of <50%. Further studies are still required to unravel the ECs removal mechanism and the most ideal process layout, including strategies for scale-up.

6. CONCLUSION

ECs need to be removed due to their potential adverse effects on the environment, human health, and other organisms. There are several available treatment technologies for ECs removal classified into physical, biological, and chemical treatments. Adsorption and membrane filtration are physical treatments that can be used for ECs removal as a single/ standalone or in hybrid systems. As a single system, adsorption or membrane filtration had shown diverse removal efficiencies. Adsorption or membrane filtration can also be combined with various other technologies. Hybrid systems are of great interest due to their potential to overcome the shortcomings of standalone technology. The combinations between adsorption and membrane filtration have been done in three configurations: adsorption as the pre-treatment, integrated adsorption/membrane filtration, and adsorption as the post-treatment. Most of the hybrid systems offered better performance than the standalone systems due to the synergistic effects. However, further investigations are needed to assess the large-scale application and remove a wide range of ECs.

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8. AUTHORS’ NOTE

The authors declare that there is no conflict of interest regarding the publication of this article. The authors confirmed that the paper was free of plagiarism.

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