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Case Report

A review on emerging water contaminants and the application of sustainable removal technologies

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A R T I C L E I N F O

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A B S T R A C T

Emerging contaminants (ECs) are synthetic or naturally occurring chemicals or any microorganisms that are not commonly monitored in the environment but have the potential to enter the environment and cause known or suspected adverse ecological or human health effects. The issue of ECs persistent in the environment and can disrupt the physiology of target receptors, they are recognized as Contaminants of emerging environmental concerns. The prominent classes of ECs include pharmaceuticals and personal care products (PPCPs), plasticizers, surfactants, fire retardants, nanomaterials, and pesticides. Several ECs have been recognized as endocrine disruptive compounds (EDCs) due to their deleterious effects on endocrine systems (EDCs). The contaminants present in the aquatic environment resources are a major cause of concern for human health and the environment and safety concern. These contaminations have risen into a major threat to the water distribution system. The impact of emerging contaminants (ECs) such as medicines, x-ray media, endocrine disruptors, insecticides, and personal care items has been reported in surface water, wastewater, and groundwater sources worldwide in recent years. Various techniques have been explored for ECs degradation and removal to mitigate their harmful effect. Numerous prior or continuing investigations have focused on the degradation and removal of contaminants using a variety of treatment techniques, including (1) physical, (2) chemical, and (3) biological. However, experimental data is insufficient to provide precise predictions regarding the mechanistic degradation and removal fate of ECs across various in-practice systems. The membrane technology can remove particles as fine as 10 μm and colloidal particles, It can be effectively eliminated by up to 99% through the use of MBR and treatment technologies such as reverse osmosis, ultrafiltration, or nanofiltration at concentrations up to 5 g/liter. In this paper, the emerging contaminants overview, their sources, and their removal by application of various treatments based on recent studies have been presented.

1. Introduction

Water pollution, a severe issue, requires appropriate policy and techniques to monitor and execute strategies and policies for attaining solutions. The annual wastewater discharge is reported to be in the range of 1500 km^3 [1]. The excess rise in water demand caused by increased population, industrial, and agricultural expansion may be satisfied by preventing contamination of freshwater supplies and improving wastewater treatment. A multitude of new substances have been identified as completely anthropogenic or naturally occurring compounds in the aquatic environment in recent decades which has caused increasing concern regarding the world's environment and

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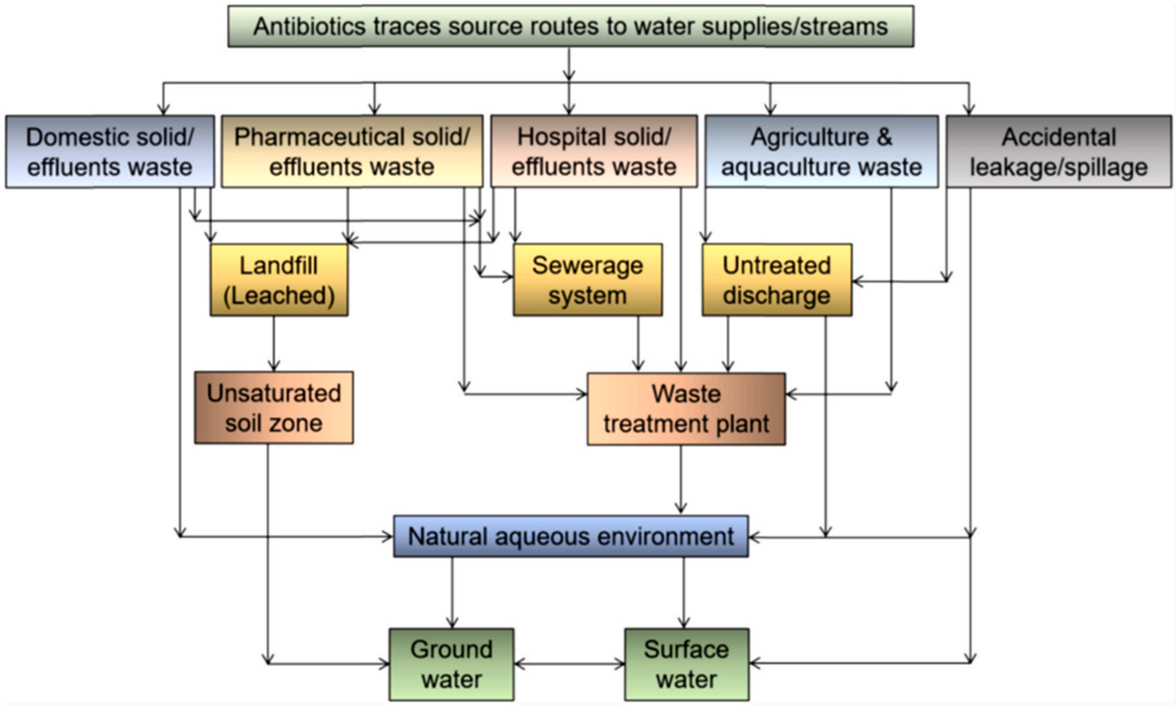

Fig. 1. Diagram showing pathways of emerging pollutants from sources to environment (Source: Bilal et al. [35]).

surface waters. In nature, most of these contaminants are organic in nature and typically occur in traces in the range of parts per trillion (ppt or ng/L) to parts per billion (ppb or μg/L) [2]. These compounds are named as "emerging contaminants (ECs)", micropollutants (MPs), emerging pollutants (EPs), contaminants of emerging concern (CEC), or trace organic compounds (TrOCs). As defined by the United States Geological Survey, ECs are chemicals of synthetic or natural origin, or microorganisms that are not commonly monitored in the environment, but that may have adverse effects on the environment or human health. In recent years, a diverse variety of micropollutants, also known as emerging contaminants (ECs), have been studied in surface water, drinking water, subsurface water, and effluent/wastewater, including common household chemicals and industrial additives. "Emerging contaminants are synthetic or naturally occurring chemicals or any microorganisms that are not commonly monitored in the environment but can potentially enter the environment and cause known or suspected adverse ecological or human health effects" [3]. It is crucial to note that the bulk of emerging contaminants are not new or recently introduced pollutants into the environment. However, most newly discovered contaminants are already well-known pollutants with a newly discovered harmful impact or mode of action (MOA). Therefore, the term "emerging" describes both the contaminant and the emerging concern about the contaminant as a threat. In this way, emerging contaminants are often called contaminants of emerging concern"or "chemicals of emerging concern" [4].

Contaminants are characterized as "emerging" when they have a new source, an alternate route to people, or novel treatment approaches [5]. Contaminants of emerging concern have been identified throughout the hydrological cycle, including groundwater, surface waterways, and wastewater treatment plant effluents their negative impact on terrestrial and aquatic life forms and human health is becoming a source of concern for scientists, engineers, and the general public. The emergence of such chemical compounds in environmental media is not a new phenomenon; it can be dated back to 2000 years ago, when the oldest global contaminant, lead, emerged due to Roman and Greek overexploitation of lead mines [6]. After then, the trend progressively spreads from conventional pollutants to modern nanomaterials, medicines, personal care items, and so on. There are different chemicals contaminants in surface and groundwater that have been found in recent studies. These include pharmaceuticals and hormones, pesticides, illicit drugs, artificial sweeteners, personal care products, disinfection byproducts, perfluorinated compounds, and UV filters, as well as other industrial chemicals that have been found in the ng/L–g/L range [7]. Emerging contaminants (ECs) have been found in wastewater, groundwater, and surface waters, including pharmaceuticals, X-ray contrast media, cosmetics, and personal care products [8–25]. After invading the environment via leaky sewage pipes and septic systems, these chemicals penetrate groundwater, travel through wastewater treatment facilities, and finally discharge into receiving rivers. ECs enter the aquatic environment through different routes, including direct discharge of treated or raw wastewater from municipal, industrial wastewater treatment plants (WWTPs), hospitals, sewer overflow/sewer leakage, landfill leachate, and surface runoff from agricultural or urban areas where treated sludge/wastewater or manure Refuse is applied for irrigation activities. Many ECs are often associated with discharges from WWTPs due to the ubiquitous usage of many of these compounds and a lack of methods with appropriate removal effectiveness, such as adsorption, ozonation, and their combinations [26]. In reality, many of these compounds are not yet included in current wastewater treatment legislation (Directive 2000/60/EC, Directive 2008/56/EC, Directive 2013/39/EU), so WWTPs are not explicitly intended to eradicate them. As a result, it has been shown that WWTPs only remove a portion of numerous ECs, such as diclofenac or carbamazepine (<25%). Continual discharges provide various aquatic ecosystems at sublethal levels that might attain chronic levels (low g/L range) of many CEC [27–29]. Other ECs include remains of recreational drugs and their metabolites, as well as non-commonly monitored agricultural chemicals such as different herbicides, insecticides, and medicines used in animal husbandry [30–34]. Fig. 1 shows a schematic representation of the pathways by which ECs reach the environment.

ECs are usually bio-active and bio-accumulative. It is possible to occur on a wide scale and have a persistent presence. A rise in the global human population, particularly in high-density places, is predicted to result in an increase in the concentration of ECs in the environment, as

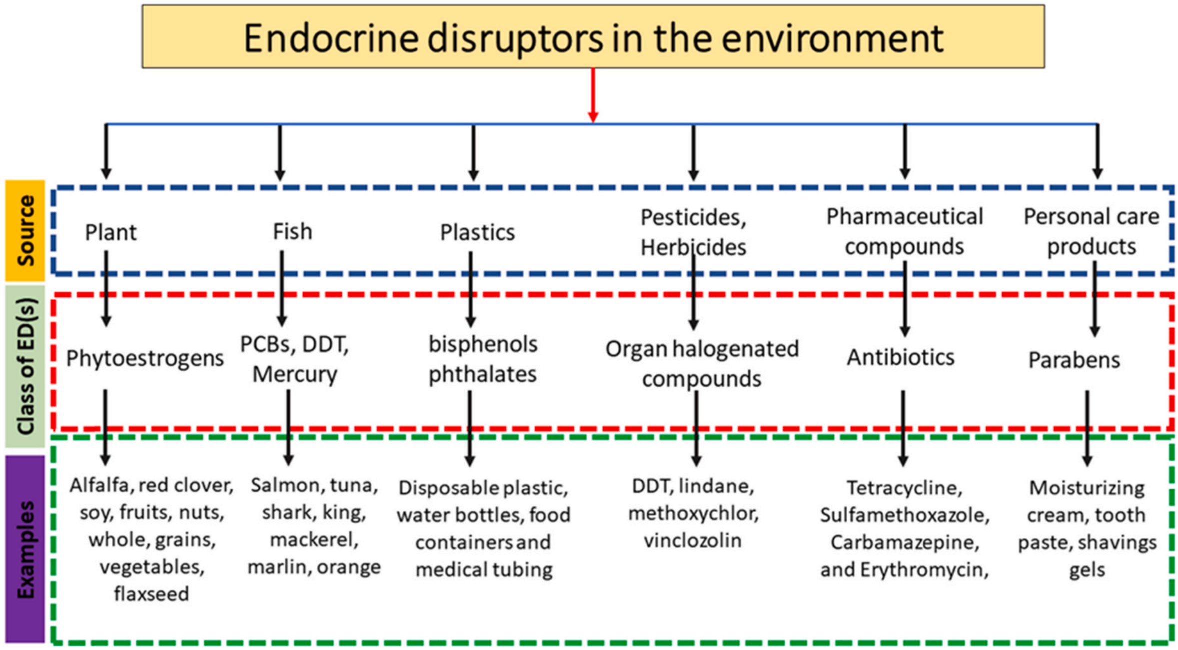

Fig. 2. Various sources of endocrine disruptor compounds (EDCs).

well as the number of ecosystems contaminated. The immense numbers of ECs are present; there is no regulation; hence, it is critical to monitor regular examination and report on their likely existence in water supplies and wastewater discharges and their potential toxicity. Contamination of the environment (water, soils, wastewater, and river sediments) by these pollutants is challenging due to the low concentration levels needed, the complex nature of the samples and the difficulty in separating these compounds [36]. Although EC values in aquatic sources have been found in the range of ng/L to g/L, long-term exposure to these chemicals can be harmful [17,37,38]. Knowledge of the identification of developing pollutants in water and technology trends for their removal is a requirement that must be addressed to enlighten about the adoption of optimum treatment procedures for assuring the use of water that is safe to drink for the general public. This review's objective is to offer a fundamental overview of developing toxins and their environmental origins, particularly wastewaters, and a discussion of the many treatment technologies implemented to eradicate ECs from drinking water resources.

Various techniques of membrane are presently used for removal of contamination of water treatment. Various treatment technology with a wide range of applications is contributing to reducing contamination to mitigate water requirement.

2. Emerging contaminants and sources

The presence of emerging contaminants (ECs), including pesticides, personal care items, x-ray contrast media, endocrine disruptors, and medicines in wastewater, groundwater, and surface waters, has been widely documented in recent years [15,39–42]. A particular class of EC that has received much attention recently is known as disruptive endocrine chemicals (EDCs). The Endocrine Society defines EDCs as follows: "an exogenous (non-natural) chemical, or a mixture of chemicals, that interferes with any aspect of hormone action. "These compounds affect the body's hormonal balance by various mechanisms; they may disrupt hormone production, mimic hormones, influence the development of hormone receptors, function as hormone antagonists, or modify hormone binding. Endocrine disruptors are a diverse group of molecules that include Pharmaceuticals and personal care products (PPCPs), synthetic chemicals used as industrial solvents/lubricants and their byproducts, plastics [bisphenol A (BPA)], [polybrominated biphenyls (PBBs), dioxins], plasticizers (phthalates) [43]. At concentrations as low as a few nanograms per liter of solution, EDCs have been proven to be physiologically active. EDCs get accumulated into the environment, especially waters, via various paths, which can be point sources (such as municipal sewage, industrial wastewaters, landfill) and non-point sources (such as agricultural runoff underground contamination), as shown in Fig. 2.

Biological hormones are imitated by these natural or synthetic substances, connected to major changes in the natural processes of species, including wildlife and fish [44,45]. Among the growing, contaminant class, Pharmaceutical and personal care products (PPCPs) are receiving increasing attention from the scientific community. Analgesics, antiseptics, antibiotics, and a range of other chemicals are among them. Since polar functional groups are common in this class of contaminants, identifying and removing them is more challenging [40]. According to Muzamil and Ahmad [46], a total of 42 articles in India reported the measured concentrations of emerging pollutants in their respective countries. Fifty-seven % of the studies verified the presence of pesticides, 17% of the studies verified the presence of medicines, 15% of the studies verified personal-care products, and 5% of the studies verified the presence of phthalates, respectively. For example, engineered nanoparticles are employed in PCPs.

ECs are found in the environment of 14 countries, according to a study by Jiang et al. [47], and examined for their occurrence, destiny, and movement. More than 80 distinct types of PCPs, EDCs, and pharmaceuticals were found in both treated and untreated sewage, streams, lake, oceans, sediments, and even tap water, according to the results of Jiang et al. [47]. Surface water, groundwater, and wastewater have all been reported to contain ECs. However, only a handful have been discovered in the environment. Their content is typically higher when directly observed at the outflow of wastewater and sewage treatment plants due to volatilization, photolysis, biodegradation, sorption, or a combination of these processes.

In contrast, it is typically lower when measured in surface waters due to photolysis, biodegradation, volatilization, sorption, or a combination of these processes [48,49]. Surface waters, on the other hand, are the primary recipients of effluents from wastewater treatment plants. Because surface water residence times are shorter than groundwater

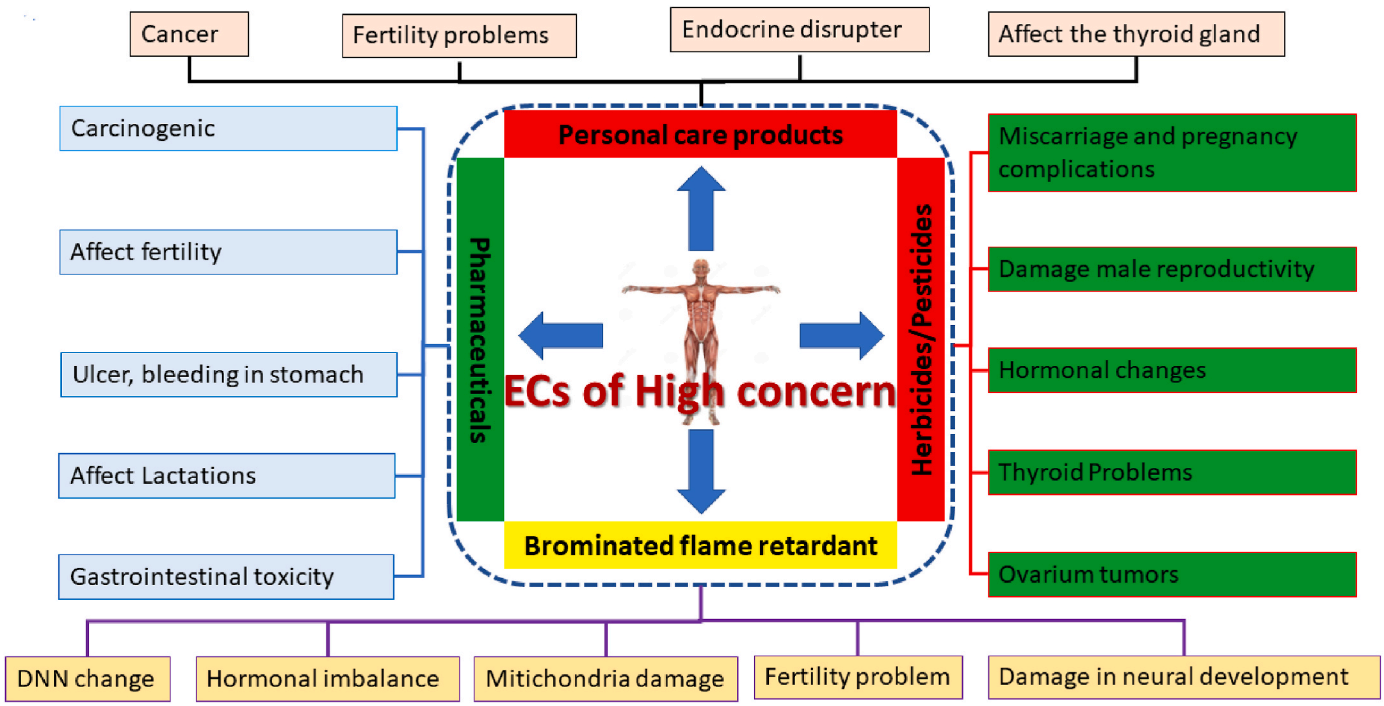

Fig. 3. Major impacts and harmful effects on human health of emerging contaminants.

residence times, the magnitude and concentration levels measured in surface waters are typically higher than in groundwaters [19]. Some of the most commonly detected groundwater ECs in the environment include ibuprofen, sulfamethoxazole, caffeine, bisphenol A diclofenac, and carbamazepine [50].

The source of the ECs influences the characteristics of the substance present. The transport and conversion of ECs also depend on physico-chemical characteristics and environmental parameters, including water solubility, temperature, polarity, volatility, organic matter content, pH, precipitation, altitude, and latitude. These factors must be considered while determining the life expectancy of an EC in the environment. There are a variety of ECs sources, both in terms of number and character. Point pollution sources and diffuse pollution sources are the two types [50].

These include unwanted flushing pharmaceuticals, human and animal excrement, manufactured goods in the home, landfill sites, industrial plants, mining operations, and food processing plants. The most important source of pollution is processed municipal and industrial wastewater from wastewater treatment plants in the urban, industrial, and agricultural sectors, as well as other locations [49–54]. PPCPs (natural hormones and synthetic steroids) are key point sources of many environmental contaminants. According to current estimations, over 30% of medications sold in Germany and 25% of pharmaceuticals sold in Austria are disposed of as expired or unused drugs [55–57]. Some of the compounds found in hospital wastewater (diatrizoate, iopamidol, iopromide, for example) are highly tenacious in the aquatic environment, having been discovered in both groundwater and surface water [58–61]. Landfill sites also serve as a significant source of ECs (polychlorinated compounds), particularly prevalent in groundwater due to their toxicity. Many nations, including Croatia [62,63], Denmark [64], and the United States [62], have reported groundwater pollution with pharmaceutical chemicals in landfill regions [65]. Farms that raise livestock are known to be point sources of estrogenic chemicals in the environment. Soils and river sediments serve as a source and sink for many biologically active compounds, including steroid hormones and veterinary pharmaceuticals.

Diffuse sources, such as stormwater runoff, terrestrial runoff from roads, metropolitan areas, highways, and agricultural land, often discharge fewer amounts of pollutants into the environment than concentrated sources [50]. Pesticides are usually recognized as one of the most significant contributors to contamination in agriculture. Herbicides (mecocrop and bentazone) and pesticides (two DDT metabolites) were discovered in the German lakes of Tegel and Wannsee, as well as flame retardant compounds [Tris(2-chloroethyl)-phosphate and Tris (2-chloroisopropyl)-9-phosphate], which were used for artificial recharge of the local aquifer [66]. Chemical contaminants (ECs) found in biosolids from wastewater treatment plants utilized for land application can reach and affect groundwater sources [67–69].

3. Effect of emerging contaminants on animal and human health

The impact of emerging contaminants on the animal is well documented. On the other hand, the direct impact on humans is still being explored. In Fig. 3, ECs pose a substantial risk to humans, even in a trace amount. Human exposure to endocrine-disrupting chemicals (EDCs) occurs mainly from the ingestion of foods and beverages contaminated with microbes, soil, water, plants, and animals. This can manifest as biomagnification and bioaccumulation, particularly for those species at the top level of the food chain. However, there is insufficient information on the toxicity and consequences of heavy metal ions, EDCs, including bisphenol-A (BPA), primarily found in WWTPs, landfills, surface runoff, and seepage [70,71]. Several studies investing the chronic impacts of ECs compounds have revealed substantial impact. For example, Female *Danio rerio* was exposed to a pharmaceutical cocktail (carbamazepine, acetaminophen, gemfibrozil, and venlafaxine) along with WWTPs effluent demonstrated a considerable decline in embryo development over six weeks [72].

It is impossible to dissociate pharmaceuticals in an aqueous solution to a large extent since they are water-soluble. Because pharmaceuticals are designed to carry out various physiological and biochemical functions, they can penetrate biological barriers and remain stable in the human body. Pharmacologically active compounds' ability to accumulate and have harmful effects on species other than those intended for use raises severe concerns. Such substances harm the physiology of

Table 1
Details of ECs on aquatic life forms and human health as per USEPA, EU and WHO (μg/L) [86,87].

ECs	USEPA	EU	WHO
Dichloromethane		20	20
DBCP	0.2		1
Chlordane	2		0.2
Polychlorinated Biphenyls	0.5		
Lindane	0.2		0.2
Toluene	1000		700
Styrene	100		20
Vinyl Chloride	2		0.3
Xylenes	10,000		500
Chlorpyriphos	10	0.1	30
2,4-D	70		30
Pentachlorophenol	1	1	9
Alachlor	2	0.7	20
Malathion	70	0.1′	900
Endrin	2	0.01	0.6
Perchlorate	15		70
Terbuthylazine			7
1,4-Dioxane			50
DDT		0.025	1
Trichloroethene			20
Tetrachloroethane			40
PFOA/PFOS	0.07	36	
Benzo (a) pyrene	0.2	0.1	0.7
Diuron		701.8	
Benzophenone-3	152		
TCEP		4	
TnBP		0.1	
Bisphenol-A	77	.24	.1
Diazinon		12.1	
Mecoprop		3.6	10
Bromate			10
2,4,6-Trichlorophenol			200
Chloroform	80		300
Dibromoacetonitrile			70
Edetic acid			600
Bromodichloromethane			60
Dibromoacetonitrile			70
Dibromochloromethane	80		100
Dichloroacetate	60		50
Bromoform	80		100
Chlorate			700
Trichloroacetate			200
N-Nitrosodimethylamine			0.1
Ibuprofen		0.011	
Erythromycin		0.2	0.103
Ciprofloxacin		0.1	
17 β -estradiol	0.175		1
Carbamazepine		2	
Trimethoprim		12	
Atenolol		150	

animals that are exposed to them. The ecotoxic effects of drug orthologs (human) on 16 different species were studied. It was discovered that 86%, 61%, and 35% of the orthologs were found in zebrafish, daphnids, and a green alga [73]. Antibiotics used in food (milk, meat, eggs, fruits, vegetables, and fish) as growth promoters, therapeutics, and prophylactics can pose health risks. Thus thiam-phenicol, erythromycin, sarafloxacin, and oxytetracycline have been identified in aquatic products. Additional enrofloxacin and ciprofloxacin concentrations in vegetables ranged from 2.0 to 32.3 and 2.5–27.5 mg/kg, respectively [74]. Plants have also been shown to transmit antibiotics from root to stem to leaf. Biological organisms may absorb pesticides in various ways because they are usually highly soluble. These chemicals are quickly transported throughout the body after absorption, undergoing biotransformation processes [75,76]. It has already been reported by the US environmental protection agency in a document that some of these chemicals have the potential to act asan endocrine disrupters, altering hormone levels [77,78].

According to Buzea et al. [79], the detrimental effects of

Table 2
Removal efficiencies of ECs by Biological treatments.

Treatment technique	Contaminant	Removal efficiency	Reference
Activated Sludge Process	Triclosan	94%	[97]
	Caffeine	79%	[98]
	Galaxolide	98%	[99]
	Ciprofloxacin	83%	[100]
	Tetracycline	66–90%	[101]
Pilot plant–3-stage moving bed biofilm reactors (MBBRs)	Ibuprofen	>90%	[102]
	Acetylsulfadiazine	>90%	[102]
	Propranolol	>90%	[102]
Biofilm reactor	Diclofenac	41%	[103]
	Propranolol	94%	[103]
	Iopromide	58%	[103]
	Iohexol	57%	[103]
	Iomeprol	85%	[103]
Mixed liquor-activated sludge	Acetaminophen	90%	[104]
	Caffeine	90%	[104]
	Carbamazepine	90%	[104]
	Digoxigenin	95%	[104]
Membrane bioreactor (MBR)	Steroids	80%	[105]
	Naproxen	86–89%	[106]
	Octylphenol	70.2%	[107]
	2,4-D	99%	[108]
	Diclofenac	49%	[109]
	Azithromycin	74%	[110]
	Sulfamethoxazole	97%	[111]
	Ciprofloxacin	76%	[112]
	Ceftriaxone	47%	[113]
	Cefoperazone	79%	[113]
	Triclosan	90%	[107]
	Androstenedio	99%	[113]
MBR with submerged hollow-fiber ultrafiltration membrane	Diclofenac	80%	[114]
	Metoprolol	90%	[114]
	Clarithromycin	100%	[114]
	Erythromycin	100%	[114]
	Atenolol	100%	[114]
	Codeine	100%	[114]
Nitrification and denitrification	Acetaminophen	99%	[115]
	Naproxen	60%	[115]
	Caffeine	94%	[115]
	Atrazine	8–32%	[116]
	Pentachlorophenol	88–98%	[116]
Microorganism based treatment	Bisphenol A	85%	[117]
	Diazinon	63%	[118]
	Diclofenac	60%	[118]
	Sulfamethazine	91%	[119]
	Hydrocinnamicacid	99%	[117]
	Hydrochlorothiazide	83%	[120]

Nanoparticles on human health are determined by various factors, including pre-existing illness and heredity, size, shape, exposure, and the chemistry, electromagnetic characteristics, and aggregation state of the NPs. Recent epidemiological research has shown a strong relationship between particulate air pollution levels and cardiovascular and respiratory diseases. Recent epidemiological research has shown a strong relationship between respiratory and cardiovascular disorders, particle air pollution levels, cancers, and death. Several nanomaterials can produce reactive oxygen species and in vitro cytotoxicity. They can also penetrate cell membranes and biological hurdles such as blood-brain barriers [80–83].

4. Treatment of emerging contaminants

The presence of micropollutants in the aquatic ecosystem can cause major ecological vulnerabilities, including interaction with high-level species' endocrine systems, reduced microbiological resistance, and accumulation in soil, plants, and animals, among many other concerns [84]. Because of ECs in the ecosystem, naturally occurring water sources, their ecosystems, and aquatic life are at greater risk than human health, which is a concern for many people [85]. The detail of aquatic life forms and human health WHO guide line and regulation of ECs by

WHO, EPA, EU and IS 10500 (g/L) is shown in Table 1.

It is vital for the environment and health to have regulation over ECs in order to monitor and mitigate their adverse effects. Due to the fact that standard wastewater treatment procedures do not entirely eradicate ECs [88,89], a variety of technologies have been used to eliminate ECs during the last several decades, including physical, chemical, and biological methods [90,91]. A blend of two or more treatment technologies, like membrane treatment with micro-filtration and reverse osmosis [92], simultaneous adoption of membrane ultra-filtration, and activated carbon adsorption. Ultrasound irradiation [93], or a combination of two or more treatment technologies such as enzymatic catalysis and electrocoagulation, is presumed to be more effective in eliminating ECs as well as the utilization of ultraviolet light and hydrogen peroxide during advanced oxidation before membrane bio-filtration [94–96].

4.1. Biological treatment technologies

Aerobic processes and anaerobic processes are the two types of biological treatment technologies available. Active sludge, membrane bioreactors, and a sequencing batch reactor are among the effective aerobic treatments. Anaerobic technologies include anaerobic sludge reactors and anaerobic film reactors, both of which are environmentally friendly. The many research investigations on eliminating different developing pollutants utilizing biological treatments are depicted in Table 2, which may be found here.

For decades, bio-trickling filters are being employed in wastewater treatment facilities (WWTPs) for pathogen decontamination, to remove biochemical oxygen demand (BOD) and chemical oxygen demand (COD), for smell control, and air pollution, but their usage to remove ECs has yet to achieve broad recognition. While trickling filter beds effectively remove organic micropollutants, the activated sludge treatment method is perhaps more efficacious [121]. Trickling filter beds in a WWTP resulted in average removal efficiency of less than 70% for all 55 PPCPs investigated. Half of them were not removed.

In contrast, the use of activated sludge treatment resulted in average removal efficiency of more than 85% for all 55 PPCPs studied [121,122]. It has been increasingly recognized that the ability of biological processes to remove micropollutants through biodegradation has been investigated [123,124]. Anaerobic/anoxic/aerobic-membrane bioreactors removed certain environmental contaminants (EDCs), personal care products (PPCPs), and pharmaceuticals [125]. With influent variables ranging from ng/liter to mg/liter, these methods removed approximately 70% of the target EDCs and PPCPs [126]. Biodegradation is hindered in the case of some ECs that are poisonous and resistant to microbial development. When a growth substrate is necessary to support microbial growth for biodegradation, cometabolism is used [127].

For a wide variety of ECs, the denitrification and nitrification processes have poor removal efficiency. Still, they may be combined with MBR and other processes to increase removal efficiencies [128–131]. At a g/liter influent concentration, denitrification may remove EDCs such estrone (E1), 17-ethinylestradiol (EE2), 17-estradiol (E2), estriol (E3), bisphenol A, 4-start-butyl-phenol, and 4-*tert*-octylphenol, as well as PPCPs including benzophenone, galaxolide, oxybenze, salicylic acid [116]. It is most commonly used after ozonation to remove pollutants; however, because it is excellent at removing nitrogen and organic carbon from water streams, it may also be used as part of a tertiary treatment process for reclamation [132,133]. The biological activated carbon method exhibited lower effectiveness in removing certain EDCs, such as E3, octylphenol, and bisphenol A, according to Gerrity et al. [132]. Still, it was 99% efficient in the removal of E1.

The biological method of pollutant breakdown through oxidoreductase enzymes (like peroxidases) is a relatively young and promising area of research. To effectively break down a variety of organic contaminants, enzyme systems have been used that can oxidize and degrade them into smaller intermediates. Enzyme-based treatments have many benefits, like working at both high and low levels of pollution, low energy input, less sludge production, and more. They can also treat a wide range of pollutants [134–136]. Laccase and peroxidases are two enzymes primarily employed in the bioremediation of contaminated wastewater [137,138]. These are widely used enzymes in enzymatic -remediation studies because of their high capacity to degrade various contaminants [139]. These enzymes accelerate the oxidative-reductive biodegradation of various contaminants, including phenols, cresols, herbicides, chlorinated phenols, pesticides, synthetic textile dyes, dioxins, pharmaceuticals, and personal care products (PPCPs), among others [42,136,140]. Oxidoreductases include peroxidases, oxidases, oxygenases, and dehydrogenases.

Laccases (Lac) belong to the class of multi-copper oxidases found mostly in Various plants, bacteria, insects, and fungi. Laccases that originates from microbial sources, such as wood-decaying fungi, have increased interest because of their capacity to oxidize a wide range of compounds and a wide range of substrate specificity [141]. Laccases have been successfully employed to degrade various ECs and different classes of aromatic compounds. For instance, research conducted by Morsi et al. [142] proved the laccase's ability to breakdown various estrogen hormones like 17β -estradiol (E2), estrone (E1), and 17α -ethinylestradiol (EE2) into products that have lower or no estrogenic activity efficiently. Peroxidases are heme-containing antioxidant proteins found in plants, fungi, bacteria, and animals. They catalyze the oxidation of various chemical substrates by using H_2O_2 or organic hydroperoxides as a co-substrate [143]. Due to their excellent specificity, these enzymes can degrade contaminants effectively [144]. Several studies have demonstrated peroxidases' potential to bio-remediate many emerging pollutants. HRP, CPO, MnP, and SBP are the most frequently used peroxidases for wastewater treatment [142].

Microorganisms (bacteria, algae, and fungus) employed in biological wastewater treatment can emulate natural ecosystems' ability to decrease contaminants in water cost-effectively and justifiably. A variety of drugs such as pharmaceutical beta-blockers (sotalol, propranolol, and atendol), anticancer and gastroesophagealdrugs (famotidine, crimetidine, citalopram, acridoneandranitidine), anti-inflammatory drugs (acetaminophen, including the stimulant butalbital), and antibiotics (sulfathazole, sulfamethazime, sulfapyridine, azithromycin, and erythromycin) could be eliminated by 100% through fungal generators. When employed in an algae-based polishing pond treatment process at a concentration level of 1 g/liter, environmental pollutants (ECs) such as E1, E2, and EE2 may be removed by more than 95% [117]. Although the activated sludge process is the most widely used and adapted in so many ECs removal applications worldwide, the proportion of ECs removed by primary setting, chemical precipitation, aerating volatilization, and sludge absorption is negligible. The majority of ECs in wastewater are separated by biodegradation, the most common method of ECs removal [35,145]. The activated sludge technique is also quite efficient in removing EDCs, with the removal rate ranging from 75 to 100% [115,146–148].

4.2. Physio-chemical treatment technology

Physio-chemical treatment technologies used to remove various ECs as reviewed from literature are summarized and available in supplementary data S1.

4.2.1. Coagulation-flocculation

Coagulation is a chemical change in colloidal particles that causes molecules to aggregate and settle over time. When utilized in conjunction with coagulation-flocculation paired with sand filtering, Huerta-Fontela et al. [149] discovered that aluminum sulfate $(Al_2SO_4)_3$ was efficient in removing medicines such as hydrochlorothiazide, warfarin, and betaxolol (with an 80% removal efficiency). Musk compounds (personal care products) were discovered to be removed in high quantities from hospital wastewater, particularly celestolide, galaxolide, and tonalide, with significant removal rates of 83%, 79%, and 78%, respectively [150]. Chemical treatment, such as coagulation, flocculation, or lime softening, was shown to be ineffective for removing EDCs and PPCPs, with the compounds tested (carbadox, sulfadimethoxine, and trimethoprim) not being removed by metal salt coagulants (aluminum sulfate and ferric sulfate) [151].

Electrocoagulation, when a current of electricity is passed through water, coagulant precursors are produced by electrolytic oxidation of anode material, typically aluminum or iron. Electrocoagulation technology decreases pollutant levels by transmitting an electrical current through water and producing coagulant precursors by electrolytic oxidation of anode material (typically aluminum or iron) [152]. In his study, he used an electrocoagulation (EC) device with aluminum blades to remove six estrogenic EDCs (estrone (E1), 17β-estradiol (E2), estriol (E3), 17α-ethinylestradiol (EE2), bisphenol-A (BPA), and nonylphenol) from breast milk (NP). Sixty-two %, 60%, 68%, 53%, 42%, and 98% were found to be removed (estrone (E1), 17β-estradiol (E2), estriol (E3), 17α-ethinylestradiol (EE2), bisphenol-A (BPA), and nonylphenol respectively) from the body by these six endocrine-disrupting chemicals. During the research, it was discovered that aluminum EC can significantly lower EDC concentrations in municipal wastewater influent and effluent streams.

4.2.2. Activated carbon adsorption

It is possible to eliminate several hydrophobic and charged medicines, EDCs, and PPCPs by utilizing activated carbon adsorption as an adsorption approach [153–155]. An activated carbon adsorption system is useful in that it can eliminate most organic molecules due to its hydrophobic interactions, particularly non-polar chemicals (compounds with $K_{ow}>2$) [156]. Schafer at al [157] stated several studies have found that powdered activated carbon (PAC) has the potential to remove EDC up to 90%, while Snyder et al. [158] investigated the removal efficiency of powdered activated carbon (PAC) at 5 mg/liter concentration and contact time of 5 hours for 66 Pharmaceuticals and personal care products (PPCPs), finding that only nine of them were removed with less than 50%.

4.2.3. Conventional oxidation processes

Chemical treatment technologies are applied alternative to the other treatment methods for the further removal ECs. These techniques, generally referred to as aqueous phase oxidation methods, depend on highly reactive chemical species as an intermediary in the process [159]. Oxidation reactions have mainly been used to enhance the treatment of ECs rather than the replacement of conventional systems [160].

Oxidation is an efficient EC removal procedure, particularly by using chlorine or ozone. Oxidation effectively degrades EDCs/PPCPs present in low dissolved organic carbon (DOC) because the application of the ozonation process has a strong impact on DOC [161]. Noutsopoulos et al. [162] used 1000 ng/liter of each EC pollutant to assess the effect of chlorine on the removal of some ECs to determine the effect of chlorine on the removal of some ECs. Before being removed, the first chlorine dosage of 11 mg L1 was exposed for 60 minutes. As the maximum removal rates, Naproxen and diclofenac had the highest removal efficiency, with 95% and 100%, respectively. Another investigation demonstrated that the elimination of EE2 by chlorination was seen to be up to 100% effective within 10 minutes [163]. Some ECs can be eliminated faster by raising the chlorine dosage, extending the contact duration, or changing the pH of the solution [164]. Ozone oxidizes substrates either directly or indirectly by producing hydroxyl radicals, which react with other substances and produce further oxidation [165]. Compared to a normal wastewater treatment plant, the energy consumption of an ozone treatment system might increase by 40–50% [129]. Researchers have discovered that the process of ozonation may remove all forms of environmental contaminants (ECs) by 90–100%, and this approach has been demonstrated as being more beneficial in a wide range of situations.

Furthermore, while eliminating ECs such as 2-phenoxyethanol, methyl salicylate, and amitriptyline hydrochloride, it was observed that the reaction rate of the chlorination method was three orders of magnitude lower than the reaction rate of the ozonation process [166]. When it comes to the elimination of EDCs (5–10 μg/liter) and pesticides (80%–100%), the photolytic procedure is quite effective [145,167]. With this, it is possible to totally remove some medications, such as ketoprofen, tetracycline, iopamidol, diclofenac, oxytetracycline, and mefanamic acid procedure. According to Ahmed et al. [129], when EC concentrations were mg/liter, the UV photolysis/H_2O_2 process can successfully eradicatemost ECs up to100%. Some ECs such as lincomycin and diclofenac are exceptionsas they can be removed by about 80%.

4.2.4. Advanced oxidation processes

Ikehata et al. [168,169] studied that in advanced oxidation processes (AOP), the formation of free radicals, particularly hydroxyl radicals, allows the pollutants to get converted into less hazardous and more biodegradable compounds. Advanced oxidation processes (AOP) convert pollutants into less harmful and more biodegradable compounds [168,169]. AOPs are extremely effective techniques for water and wastewater treatment [170,171]. Photocatalysis, also known as accelerated oxidation, is the chemical change that occurs when a catalyst gets activated due to light availability, which supplies sufficient energy for the process to occur [172,173]. Photocatalysts are semiconductor metal oxides with a small energy band gap, making them ideal for use as photocatalysis. Titania is one of the most studied heterogeneous photocatalysts, because of its photostability, inert nature, and low cost, among many other things [174]. Alternatively, photocatalysis in the presence of hydrogen peroxide may be used to remove pesticides such as aldrin, diazinon, malathion, and certain antibiotics such as amoxicillin, ampicillin, and chloxacclin with high removal efficiency (99–100%) [175].

In Ozone oxidation technology Organic compounds are rarely degraded completely when oxidized by ozone alone, since the effect is relatively weak, and a significant amount of ozone is required to achieve this. Therefore, ozone oxidation technology is often used in combination with other technologies. In combination with ultraviolet radiation (O_3/UV), ozone oxidation technology provides good results for removing complex organic matter. Its oxidation technology works as follows:

$$\mathbf{O_3 + H_2O + hv \rightarrow O_2 + H_2O_2}$$

$$\mathbf{H_2O_2 + hv \rightarrow 2 \cdot OH}$$

UV/O_3 methods are capable of increasing the variety of organic compounds that can be degraded by oxidation and accelerating the rate of degradation. Oxidizing free radicals produced by ozone can be created under the influence of ultraviolet light. The combination of ultrasound and ozone can also enhance the ozone oxidation process, aside from using UV and ozone separately. When ozone molecules are decomposed, many free radicals are produced that have strong oxidative properties. The ultrasonic waves can also be used in the reaction process to increase the surface area of ozone contact with liquid water, which improves the mass transfer rate of ozone, and is of high efficiency and environmental protection. The ozonation process is extensively applied to water and wastewater treatment, for example, to eliminate organic pollutants and disinfect water and wastewater. It is possible to address the shortcomings of ozonation by adding a catalyst to it, or combining it with some other AOPs.

Fenton oxidation is a type of oxidation during which hydrogen peroxide reacts with iron to form hydroxyl radicals when the iron is present [176]. Because of their speed and efficiency, Fenton reactions are a potential alternative for wastewater treatment.

To address the inadequacies of the conventional Fenton process and enhance the efficacy of pollutant removal, the Electro-Fenton method was recently developed [177]. With an efficiency of 88–93% [178], the solar photoelectron Fenton technique has effectively eliminated beta blockers such as metoprolol tartrate propranolol hydrochloride and cendol. Photo-Fenton reactions are frequently utilized as alternate operating techniques for the removal of ECs from wastewater. These procedures entail the use of ultraviolet light to generate radicals due to interactions between hydrogen peroxide and iron in the presence of iron. Except for penicillin G, many types of medicines have been reported to have better removal efficiency (95–100%) when subjected to the photo-Fenton method [14,176,179]. Based on these data, we may conclude that the photo-Fenton procedure based on UV radiations can eliminate more beta-blockers and pharmaceuticals than the solar-based photo-Fenton techniques [129]. Gimeno et al. [180] investigated heterogeneous solar photocatalysis using solar photo Fenton, ozonation, and TiO_2. They discovered that photocatalytic ozonation has a greater degradation rate than photocatalytic oxidation, which they ascribed to the presence of oxygen.

In Electro-Oxidative Advanced Oxidation Processes for eliminating organic contaminants contained in wastewater is electrooxidation, also known as anodic oxidation or electrochemical incineration. Electric currents of 2–20 A are applied between two electrodes in water. The result is the production of hydrogen peroxide and hydroxyl radicals between the electrodes.

$$\mathbf{H_2O \rightarrow OH + H^+ + e^-}$$

$$\mathbf{O_2 + 2H^+ + 2e^- \rightarrow H_2O_2}$$

The global reaction is:

$$2\mathbf{H_2O} + \mathbf{O_2} \rightarrow \mathbf{H_2O_2} + 2\ \mathbf{OH}$$

This process, also known as anodic oxidation or electrochemical incineration, is one of the most widely used electrochemical advanced oxidation methods for the reduction of organic contaminants found in wastewater.

5. Advanced and hybrid treatments

Conventional wastewater treatment techniques are not enough to effectively remove many ECs. As a result, several sophisticated and hybrid therapy approaches for considerably eliminating ECs have been reported in the literature. Ultraviolet (UV) photolysis, ion exchange, ultrasonic irradiation, and membrane filtration are examples of advanced forms of treatment. During the last few years, several hybrid treatment methods have been described that have resulted in substantial improvements in the avoidance of EC release into the aquatic environment through effluent discharge [129]. In general, hybrid treatment technology combines biological remedies with suitable physical or chemical approaches to treatment.

To treat wastewater, the most often used hybrid technique is a chemical oxidation-based treatment, such as ozonation, combined with a biological process. Combinations such as ozonation followed by biological activated carbon, MBR-reverse osmosis/filtration/ozonation/microfiltration/ultrafiltration/activated sludge followed by ultrafiltration, and MBR-reverse osmosis followed by biological activated carbon are a few examples. Microfiltration and ultrafiltration are becoming increasingly popular as a substitute for traditional granular media filtration methods [181].

Ultrasound irradiation, also known as sonochemical irradiation, has been demonstrated to successfully remove ECs from wastewater treatment plants (WWTPs). When using ultrasound to remove EDCs such as E1, E2, E3, and equaling from aqueous solution at a concentration of 10 μg/liter, it was discovered that up to 80–90% of the EDCs were eliminated [182,183]. It is frequently necessary to use additional therapies in conjunction with the ultrasonic irradiation technique to effectively remove ECs. In addition to enhancing the flow of pollutants and controlling fouling of the membrane [184–186], ultrasound also promotes the adsorption of contaminants by adsorbents [187–189]. A hybrid technique, developed by Secondes et al. [93], was tested to determine the efficacy of removing ECs using membrane-activated carbon adsorption, ultrafiltration, and ultrasound irradiation all at the same time to remove the contaminants. In this study, the removal of diclofenac, carbamazepine, and amoxicillin was tested at ultrasound frequencies of 35 kHz and 130 kHz. The outcomes indicated that the clearance of these drugs ranged from 99 to 100%. Ultrasonic irradiation greatly improved the removal of the least hydrophobic EC in the feed by increasing its hydrophobicity.

In ECs removal, membrane techniques, which employ membranes prepared from various materials with particular filtering characteristics (hydrophobicity, size of pores, and surface charge) that govern the type of contaminant which can be collected, are a sort of phase-shifting process having diverse applications [157,190]. There are several types of membrane filtration available, including gnanofiltration (NF), microfiltration (MF), ultrafiltration (UF), reverse osmosis (RO), and forward osmosis (FO); Ultrafiltration is the most common type of membrane filtration (RO). In the case of micropollutant removal, membrane filtration techniques like reverse osmosis and nanofiltration are proven to be a safe and effective alternative [191,192]. The FO process, which employs the concentration gradient to produce a net flow of water over the membrane, unlike the RO process, which uses hydraulic pressure to separate contaminants, is used to separate contaminants. Because it can remove particles as fine as 10 μm and colloidal particles, RO is more efficient than other methods [2]. Several EDCs, including E1, E2, EE2, E3, 17-estradiol 17-acetate, bisphenol A, 4-*n*-nonylphenol, and 4-*tert*-butyl-phenol, have been reported to be effectively eliminated by up to 99% through the use of MBR and treatment technologies such as reverse osmosis, ultrafiltration, or nanofiltration at concentrations up to 5 g/liter [167,193,194]. It has been proven that NF is more successful than UF in the elimination of several ECs.

5.1. Design criteria for advanced and hybrid treatments

An effectiveness implementation of advanced and hybrid treatments takes a dual focus a in assessing effectiveness and implementation. Three hybrid types: (1) testing effects (2) dual testing of and implementation interventions/strategies; and (3) testing of an implementation strategy while observing and gathering information on the clinical intervention's impact on relevant outcomes. Photolysis has potential of mineralizing which can be enhanced by Photo process with environmental pollutants, it can be processed via oxidation or reduction routes, called advanced oxidation processes.

Dolar et al. [110] evaluated the removal efficiency of pharmaceuticals present in municipal wastewater from a coastal WWTP in Spain, utilizing an integrated pilot-scale membrane system (MBR–RO) connected with a membrane bioreactor. When MBR and RO treatment were combined, the findings revealed that the combination demonstrated outstanding removal of emergent pollutants, with removal rates exceeding 99% for all of them. MBR technology has shown high removal effectiveness for several chemicals such as metronidazole, hydrocodone, codeine, and ranitidine, with up to 95% removal efficiency for some of these compounds. Furthermore, the removal rates as indicated by RO membrane were always higher than 99%. A study was also carried out by Wang et al. [195] to design a modified ultra-filtration membrane to remove steroidal estrogens, which was published in 2016. Polyvinylidene fluoride (PVDF)-polyvinylpyrrolidone (PVP)–TiO_2nanoinorganic modified ultrafiltration membrane was designed to increase the ultra-effect filtration as well as to tackle problems associated with the recycling of nanomaterials in water. The results showed that the removal efficiencies of E1 and E2 by the PVDF-PVP-TiO_2 ultrafiltration membrane under UV photocatalysis were higher than those achieved by the PVDF-PVP membrane under UV photolysis, indicating that the PVDF-PVP membrane was superior to the PVDF-PVP membrane under UV photocatalysis. The equilibrium for removal of E1 and E2 by the PVDF-PVP-TiO_2 membrane was reached in around 90 minutes, with removal efficiencies of approximately 93.4% and 73.1%, respectively,

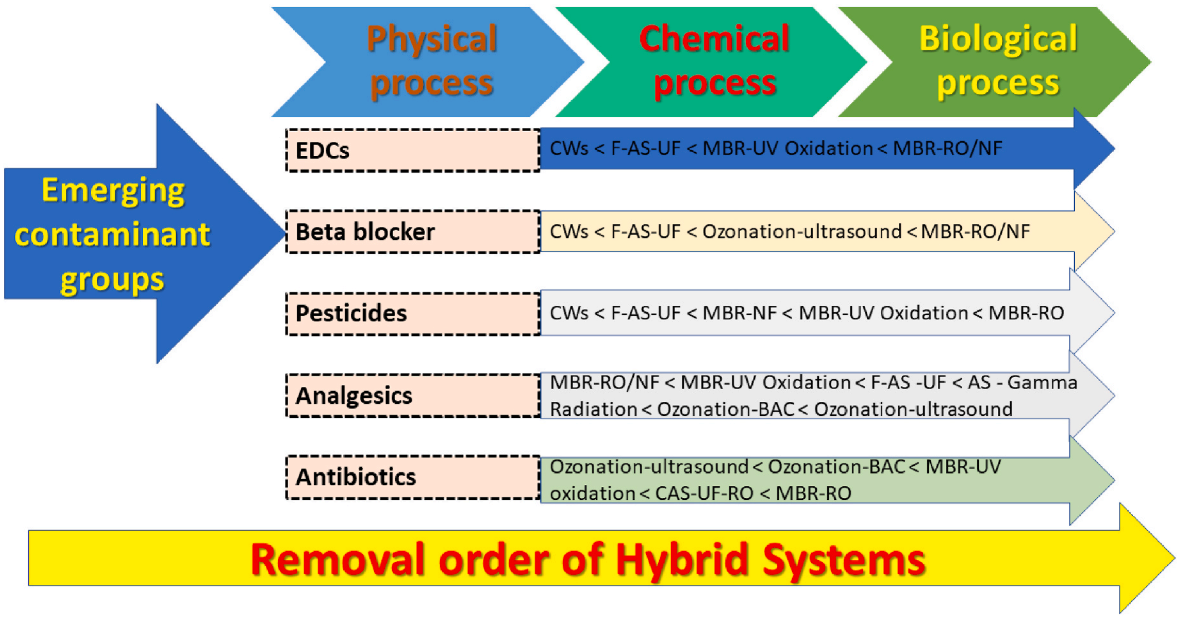

Fig. 4. Removal order of hybrid treatment systems for various classes of ECs.

after the membrane was exposed to the chemicals. Fig. 4 summarizes the removal order of hybrid systems for different classes of wastewater treatment systems. Hybrid systems offer several options for wastewater treatment according to personal preference and consideration.

6. Conclusions

ECs present and redistributed in various water resources have toxic effects on the living organisms disrupting their natural endocrine activity. Several harmful compounds are not efficiently removed from WWTPs. Hence, their residual concentrations reach the surface and groundwater resources, thus, posing a risk to the environment. The review gives a brief highlight of the significant research work conducted in recent years about the progress and advancement of emerging contaminant removal technologies from polluted water sources. To completely remove ECs from water, traditional wastewater treatment techniques are insufficient. As a result, technologies for physical, chemical, and biological treatment are required to ensure that all ECs are completely removed from the water. Chemical treatment techniques (activated sludge, activated carbon, membrane bioreactors (MBRs), and treatment based on microorganisms) have successfully removed ECs such as EDCCs, PPCPs, surfactants, and pesticides analgesics, antibiotics, beta-blockers, and pharmaceuticals from wastewater with high removal efficiencies. Advanced oxidation processes are highly capable of treatment of wastewater. The membrane technology can remove particles as fine as 10 μm and colloidal particles, It can be effectively eliminated by up to 99% through the use of MBR and treatment technologies such as reverse osmosis, ultrafiltration, or nanofiltration at concentrations up to 5 g/liter.

Similarly, a photo-Fenton method based on ultraviolet radiation may efficiently remove many ECs such as beta-blockers and pharmaceuticals. Therefore, it is necessary to adopt advanced and hybrid treatments for removing of ECs efficiently. A combination of treatments employed to eliminate ECs is much more effective in eliminating ECs than applying a single technique or conventional methods. Finally, the existing treatment techniques could be modified through various approaches like nanotechnology and genetic engineering to enhance their removal capacities and efficiencies.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cscee.2022.100219.

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