Treatment strategies for enhancing the removal of endocrine-disrupting chemicals in water and wastewater systems

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Abstract

The emergence of endocrine-disrupting chemicals (EDC) in water and wastewater systems has high-risk implications for the environment. This manuscript discusses the treatment strategies for the removal of EDC in water and wastewater systems. The reviewed treatment outlines for EDC removal are classified into physical, biological, and chemical treatments. The application of EDC treatments is discussed based on the removal and degradation process to eliminate the EDC compounds. Interestingly, the physical treatment of membrane filtration processes has been an efficient method for EDC removal without using chemical disinfection in a treatment system. Nevertheless, like other EDC treatment methods, the membrane filtrations are not able to remove emerging contaminants completely. Thus, the overall factor of limitations and challenges in EDC treatment methods such as solubility, hydrophilicity, degradability, and polarity has also been discussed. Besides, alternative approaches, such as sequential and hybrid treatments that enhanced the considerable removal of EDC have also been included. Finally, this article gathered each treatment approach's effectiveness and limitations, providing a potential outlook of EDC treatment strategies in water and wastewater treatment systems.

Keywords: Endocrine-disrupting chemicals, emerging contaminants, nano-organic molecules removal, and treatment persistence.

1. Introduction

During this decade, increased awareness of the risks posed by emerging contaminants to human health has raised concerns for water quality improvement. Many researchers have focused on exploring treatments for removing emerging contaminants [1, 2]. At present, endocrine-disrupting chemicals (EDC) are emerging contaminants that have continuously elicited interest in water and wastewater treatment studies [2, 3]. Such interest is primarily due to the diverse threats that EDC can pose to the environment and living organisms [4]. As reported by the World Health Organization (WHO) and the United Nations Environment Program (UNEP) in 2013, EDC can induce endocrine disorders among wildlife and humans [5]. Moreover, EDC is believed to cause neurological disorders, cardiovascular diseases, various types of cancer, and human reproductive system problems [6]. Li *et al.* [6] asserted that strong evidence is available for the impact of EDC exposure on human health.

EDC refers to a mixture of chemical agents that interfere with human body systems' processes, including the synthesis, secretion, transport, metabolism, and binding action of natural blood-borne hormones[7]. They work as agents of functional changes that can disrupt the control system and chemical release of human hormones [8]. The disruptor chemicals of the endocrine system originate from natural and synthetic sources. Natural EDC sources, which occur in living organisms, are classified into groups of estrogens, androgens, progestogens, and phytoestrogens, as illustrated in **Figure 1** [9]. Meanwhile, synthetic EDC is divided into six groups: phthalates, pesticides, phenolic compounds, polyhalogenated compounds, drugs, and pharmaceutical and personal care products (PPCPs)[10]. Estrone (E1), 17β-estradiol (E2), ethinylestradiol (EE2), estriol (E3), bisphenol A (BPA), nonylphenol (NP), nonylphenol ethoxylates (NPnEO), octylphenol, and triclosan are among the most common EDC that has been investigated [11-14]. Most EDC emergence is initiated from various manufacturing, usage, disposal, and discharge of chemicals and pharmaceutical products, which finally significantly impact the environment and living organism[4, 15].

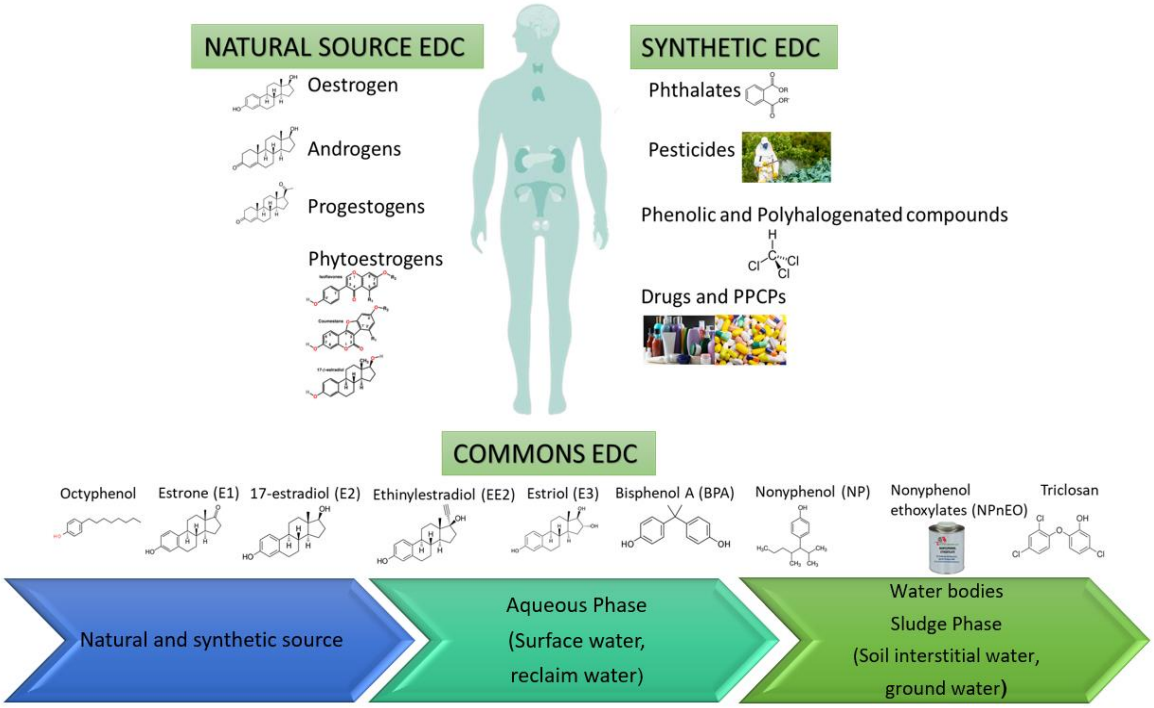


Figure 1. The diagram of EDC sources and path of distributions

There are diverse potential sources that lead to EDC contaminations in the environment. EDC's widespread exposure mainly originates from wastewater discharge from household sewerage channel, industrial operation, medical waste system, agricultural processing waste, and permanent waste disposal, such as landfill and dumpsite [16]. This concentrated EDC is entered different medium and area routes that used to be the discharge control system, such as wastewater treatment facilities. In particular, the EDC contaminants in wastewater effluent will follow a wastewater treatment system or open flow of runoff system where the effluent directly entered the groundwater and surface waterbody. As the report by WHO in 2015 [17], the extent of EDC contamination exposure varies considerably among the species, individual, and localities, depending on the type of medium exposure such as air, soil, water, food, and other types of consumer products. Thus, the effects of EDC exposure to the environments have various outcomes to the living species and habitats that dependent on sources intake.

The effects of EDC have spread diversely and are mostly observed in fish, birds, mammals, reptiles, agricultural plants, and humans. Low concentrations (ng/L) of EDC in surface water can alter the gender of some fish species and disrupt their reproductive system[18, 19]. Meanwhile, the impacts of EDC on wildlife are reproductive dysfunctions, egg thinning, and delays of sexual maturation, which affect the growth factors and populations of wildlife[20]. In agricultural plants, the exposure of estrogen, 17β-estradiol (E2), and androstenedione has caused the removal of atmospheric CO_2, inhibited the algal growth and increasing the rehydration of plants [21]. The human body's intake of EDC through consumption, inhalation, and exposure also significantly affects body systems, causing major damage in the central nervous system (i.e., nonreproductive neural and neurogenesis processes) and the reproductive system [22]. Moreover, the effects of EDC on the human reproductive system can include ovulation disorders, endometriosis, breast cancer, uterine fibroids, and pregnancy and fertility problems [23, 24]. EDC has also been examined to determine if it can cause other health problems, such as cardiovascular diseases, obesity, early puberty, and mental retardation [25]. The effects of EDC on living organisms and human health are summarized in **Table 1**.

Table 1. List of EDC impact on living organism and human health.

1 Living Organism	Source/Type of EDC	Impacts	Ref.
Fish	Natural and synthetic EDC; e.g. Estradiol,17β Estradiol and Bisphenol A (BPA)	Fish Feminism, lower reproductive fitness, lower sperm quantity alter the reproductive characteristics	[26]
Birds	Natural and synthetic EDC; e.g. Catecholamines, and Gonadotropin	Eggshell thinning, functional alterations that contribute to decreased fitness and populations, reproductive and growth factors, stress axes	[27]

	Reptiles	Pesticides; e.g Dichlorodiphenyldichloroethylene (DDE), Dichlorodiphenyldichloroethane (DDD), Dichlorodiphenyltrichloroethane (DDT)	Reproductive dysfunctions and abnormalities	[28]
	Agricultures	Estrogens and Androgens; e.g. E1-Estradiol, E2- 17β-Estradiol; 17α-Estradiol, E3- Estriol, EE2-Ethinyl Estradiol. and Androstenedione	Reduced root growth and photosynthesis rate, remove atmospheric CO_2 and inhibiting algal growth, rehydration of plants	[21]
2	**Human Health**	**Source/Type of EDC**	**Impacts**	
	Nervous System	Natural and synthetic EDC	Brain injury, nonreproductive neural effects and neurogenesis effects	[29]
	Reproductive System	Natural and synthetic EDC, e.g.; BPA, Phytoestrogens, Triclosan	Reproductive system damage. ovulation disorders, Breast cancer, endometriosis, uterine fibroids, pregnancy and fertility problems	[30]
	Metabolic and Cell Disruptors	Natural and synthetic EDC, e.g.; BPA, 17β-Estradiol	Cardiovascular diseases, obesity, affect the sex and growth hormones, abnormal cell proliferation, prostate cancer cells	[31, 32]
	Growth and Development System	Natural and synthetic EDC, e.g.: BPA, Pesticides and PPCPs	Growth and mental retardation, early puberty	[25]

Considering the various harmful effects of EDC, the remediation of water and wastewater systems involves various treatment methods, such as biological, physical, and chemical treatments, as illustrated in **Figure 2**. The application of biological treatments has successfully degraded a large proportion of EDC molecules. However, the biological degradation process fails to eliminate nonorganic EDC molecules [33]. Comparatively, physical treatment methods can remove recalcitrant nonorganic molecules. Several membrane filtration treatment methods such as ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) have been utilized in EDC removal [34, 35]. However, membrane filtration processes are commonly known to suffer from fouling problems due to pore blockage[36]. Chemical treatment methods, such as the advanced oxidation process (AOP), can also be used for EDC removal. However, the AOP treatment method can potentially form by-product residuals. These limitations are attributed to the challenges posed by the biological persistence and also physical-chemical characteristics of the targeted compounds such as hydrophilicity, pH, acid solubility (pKa), water partition coefficient (Kow), degradability, and polarity of EDC

[37, 38]. Therefore, the goal of this review is to the treatment efficiencies and evaluate strategies for EDC removal.

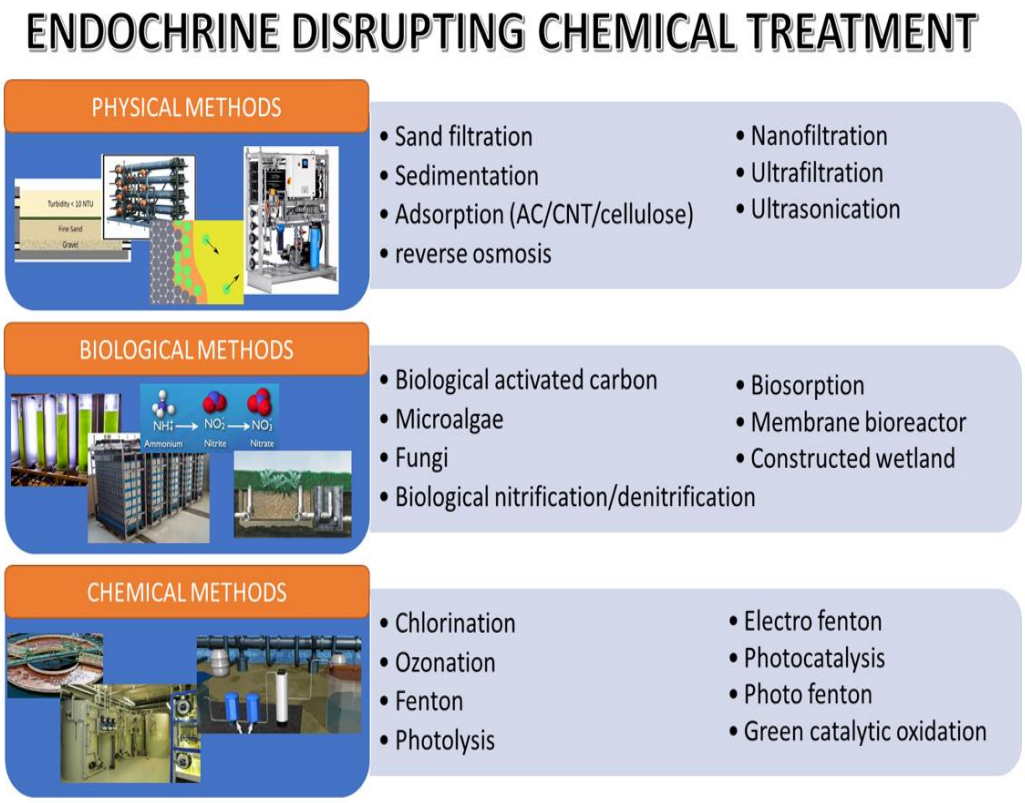


Figure 2. The lists of EDC treatment method

2. Physical treatments

2.1 Sedimentation

Sedimentation refers to the process of particle settlement under the effect of gravity. This technique is commonly applied during the primary treatment stage of sewage treatment plants (STPs) and wastewater treatment plants (WWTPs) prior to the filtration and disinfection processes [39]. At present, the sedimentation method is ineffective in eliminating EDC. Behera *et al.* [40] reported that the removal efficiency for diclofenac and E3 (estriol) using the sedimentation method is less than 28%. Moreover, significant removals have not been reported for estrone, ibuprofen, and sulfamethoxazole by using the sedimentation method[41, 42]. In particular, the ineffectiveness of the sedimentation method implies the challenges posed by contaminants in water with high hydrophilicity and solubility[43]. The types of EDC, such as hormone (e.g. Estriol), PPCPs (e.g. ibuprofen), and pesticides, have different physical-chemical properties. As reported by Kim *et al.* [44] mechanistic prediction model for compounds removal is based on the compounds molecular weight, acid solubility (pKa) and octanol water partition properties (Kwo). pKa coefficient indicates the compound charge at a given pH. In contrast, Kow coefficient is the ratio of the concentration of a compound in two phases of a mixture of two immiscible solvents. In general, the high Kow (>2) value indicates the potential of the EDC compounds that could accumulate in sediments. Relatively, moderate and high removal efficiency might be observed for the compounds with lower pKa and Kow. Nevertheless, due to the diverse physical-chemical and molecular properties, none of the treatment methods effectively removes all types of EDC. Therefore, EDC treatment methods need to be assessed on each compound basis.

Besides, such difficulty requires an advanced secondary treatment to remove the remaining proportion of EDC entirely in the aqueous phase post sedimentation process. In this condition, the high EDC solubility compounds require a hydrophobic interaction treatment medium for removal. Lin *et al.* [45] proposed advanced treatments, such as granular activated carbon (GAC) adsorption combined with sedimentation and filtration, which were the major removal mechanisms in advanced water treatment plants (WTPs). PPCPs in raw water are completely removed through the combination of treatments in advanced WTPs[45]. Therefore, the physical and chemical properties such as the Kow value is a good indicator to evaluate the suitable EDC removal interaction. Evidently, sedimentation treatment alone is insufficient for efficiently removing EDC.

2.2 Adsorption

2.2.1 Activated carbon (AC) adsorption

AC adsorption processes by using GAC and powder-activated carbon (PAC) have been extensively used in water and wastewater treatments. This method has elicited considerable attention due to its simple operation, absorbent regeneration potential, and suitability for batch and continuous processes [46]. In general, the adsorption processes are applied at the early filtration stage of the water and wastewater treatment process. The performance of AC adsorption to remove EDC compounds depend on the adsorbent dose and contact time and the physical and chemical properties of the target compounds [47]. The AC removes the water's absorbance through the hydrophobic interactions between the compound and the absorbent surface. The water system that contains a dissolved organic compound is required to be removed prior to adsorption. Predominately, the removal of the compounds before adsorption will provide longer periods between regenerations [48]. The reduction in dissolved compounds reduces the absorbent loading and probably avoids interference during the adsorption process[49]. Moreover, as reported by Kennedy *et al.* [50] in a full and pilot scale of organic micropollutants by AC adsorption, an increase in background dissolved organic matter resulted in more and earlier micropollutants breakthrough.

In terms of characteristics, the well-developed pore structure and surface chemistry properties of AC contribute to its specific interactions with EDC-adsorbed compounds [16]. Vidal *et al.*[51] reported that incorporating sulfur into the carbon structure results in a positive effect on dynamic adsorption capacity by increasing the total amount of adsorbed trimethoprim from 195 mg/g to 240 mg/g. In addition, the adequate contact time of adsorption and the dosage of AC affect the performance of EDC removal [52]. As demonstrated by Noutsopoulus *et al.*[53], high removal of triclosan (84%), naproxen (91%), ibuprofen (95%), and ketoprofen (93%) compounds was recorded at a high AC dose of 100000 μg L^{-1} at 60 min contact time.

In practice, AC adsorption treatment is highly effective in removing all target chemicals. As studied by Jiang *et al.* [54], the powdered AC recorded the highest adsorption capacity (132.73 mg/g) compared to other carbon nanotubes (103.81 mg/g), graphene oxide (77.86 mg/g) and biochar (9.19 mg/g) adsorbents for estrogen removal. Besides, Fu *et al.* [55] reported the high hydrophobicity of PPCPs contaminants could be effectively eliminated (>75%) by incorporation with secondary filtration. In their study, acebutolol, diazepam, and diltiazem compounds were removed entirely from the treatment process. Moreover, Rao *et al.* [56], studied the removal of PPCPs residues from treated effluents and further emphasized the capability of AC adsorption treatments. Their results demonstrated that 90%–98% of PPCPs

were removed from low-concentration compounds, exhibiting an evident relationship of dose response between compound concentration and adsorbent dosage. A long contact time can significantly increase compound removal[56]. Nevertheless, removal efficiency for EDC varies depending on the compound type. Several compounds, such as carbamazepine and propranolol, have exhibited relatively low removal. In this regard, understanding the specific properties of targeted compound conditions should be emphasized to achieve optimum adsorbent capability.

2.2.2 Carbon nanotubes (CNT)

Since their discovery, CNTs have received considerable attention in various research areas. The unique characteristics of the mechanical and electronic properties of multi-walled CNTs (MWCNTs) have broadened their practical applications [57].To date, CNTs have received extensive research attention as a new type of adsorbents because of their potential applications in removing various natural and synthetic EDC [58]. In contrast with AC adsorbents, CNT structures contain rolled graphite sheet layers; they are called single-walled carbon nanotubes (SWCNTs) when they have a single rolled graphite sheet layer and MWCNTs when they have double or multiple rolled graphite sheet layers [59]. Moreover, CNT adsorption properties considerably differ from AC properties. Regardless of the pore properties and surface areas, the overall adsorption properties of CNTs depend on the adsorption site, purity, and surface functional groups of nanometer-thick layered carbon [60]. Moreover, process parameters, which include pH, ionic strength, initial solute concentration, and temperature, are major factors that affect the sorption rate of EDC [61].

The application of CNTs as absorbent exhibit effective adsorption capacities in removing a wide range of EDC, such as BPA (92 mg/g), E2 (27.2 mg/g), diuron (40.37mg/g) and tetracycline (175 mg/g) [58, 62-65]. The mechanisms of the donor-acceptor system π–π bond formed between EDC and CNTs have been suggested to be significant forces that affect the adsorption performance of CNTs [58]. In addition, significant EDC adsorption is attributed to the larger van der Waals interactions and multilayer adsorption properties of MWCNTs [66]. Comparatively, the compactness of an SWCNT bundle eliminates groove areas and interstitial spaces, reducing adsorption capability [54]. In this regard, the compactness of the bundles requires the rapid dispersion of particles to provide additional adsorption sites for SWCNTs [60]. The modification of surface functional groups can further enhance the adsorption capability of SWCNTs [67]. However, although the adsorbents of MWCNTs and SWCNTs have demonstrated adequate performance, investigations on synthetic water samples remain limited[68]. In contrast with AC adsorbents, applying CNTs to real treatment plants has not been well established. In addition, the toxic effects of CNT adsorbents are still debatable [69]. In this regard, additional research on real water systems and investigations on the effects of CNTs is required to improve the understanding of CNT adsorbents' application to EDC removal.

2.2.3 Graphene oxide (GO) nanosheets adsorption

GO nanosheets are new promising adsorbents for water contaminants because of their excellent hydrophilicity properties, high surface area, and abundant surface oxygen-containing groups [70]. The use of GO nanosheets as absorbents has demonstrated outstanding removal performance for various contaminants, including divalent metal ions, aromatic organic compounds, and various dyes from aqueous solutions [71]. In addition, contaminants are easily

and rapidly extracted from water through the magnetic attraction of the hybrid GO nanosheet adsorbents [71]. Interestingly, Jiang *et al.* [72] reported the strong potential of GO nanosheet adsorbents with a maximum adsorption capacity (Qm) of 149.4 mg/g to remove EE2 hormones in an aqueous solution. The exothermic and spontaneous adsorption processes were claimed among the highest EE2 adsorption values compared with other adsorbents[73]. Nevertheless, the investigation of GO nanosheet absorbents for EDC removal in actual water and wastewater systems remains limited at present, and performance is unknown. Therefore, this significant research finding can be the basis for further investigations in the future.

2.2.4 Cellulose adsorption

Cellulose is considered one of the most promising areas of scientific and technological development in the field of plant products. Interestingly, cellulose-based adsorbents have made significant contributions compared with other available synthetic material adsorbents for water treatment [74]. However, native cellulose is less effective in eliminating EDC in aqueous solutions and having an undesirable adsorbent capacity for emerging compounds due to the low reproducibility of the process and the adsorbate's full capacity [74, 75]. To date, various modified cellulose-based adsorbent methods have been investigated for water treatment applications. In particular, the chemical modification of cellulose-based adsorbents has been found to improve the adsorption capacity of cellulose adsorbents for EDC removal [76]. As reported by Hu *et al.* [77], cellulose grafting modification that utilizes quaternary ammonium salt has achieved considerable improvement in removing amoxicillin with a maximum adsorption capacity of 183.14 mg/g. Moreover, the findings that recorded superior removal performance are the adsorption capacities (Qm) of 1072.86 mg/g and 786.18 mg/g for tetracycline and sulfamethazine, respectively, by using a sustainable α-cellulose absorbent activated by KOH [78]. Consequently, the effectiveness of cellulose-based adsorbents indicates the importance of using natural polymers as low cost, sustainable, and effective adsorbents for removing EDC in water and wastewater systems. Nevertheless, although many studies have investigated cellulose-based adsorbents in water treatment, most of these studies have focused on general water contaminants. Research on the application of cellulose-based adsorbents to EDC removal is still emerging. Further exploration and utilization of innovative methods are required to develop efficient cellulose-based adsorbents for EDC removal.

2.3 Membrane filtration

2.3.1 RO

RO is a membrane filtration method that has been effectively used for the removal of dissolved micropollutants from drinking water systems [79]. In the process, dissolved contaminants are mainly rejected through the small molecular weight cut-off (MWCO) and membrane pore sizes and the electrostatic charge's repulsion between the dissolved contaminants and the membrane [80]. Besides, the contaminants are effectively separated at low pKa and Kow value (<2) of compound properties and at low solvent permeability of RO treatment [81]. Predominately, thin-film composite membrane (TFC) is the most outstanding membrane that provides significant selectivity, compaction resistance, and chemical stability in RO treatment compare to other membrane polymers such as polyamide, polybenzimidazoline, and poly(piperazine-amide)[82]. As reported by Kassim *et al.*[83] TFC membrane was fabricated from a thin layer of hybrid membrane formulated from a blend of polyvinyl alcohol (PVA)/chitosan and cross linked with tetraethylorthosilicate (TEOS), which

was layered on the polysulfone (PSF) membrane. Besides, the TFC membrane are being further chemically modified to improve its rejection capacity for EDC removal through graft polymerization and crosslinking modification[84]. The significant characteristic of TFC that shows membrane performance has extensively attracted its application in RO treatment.

In general, RO membrane filtration has been extensively used to separate EDC compounds in WTPs [85]. In particular, emerging compounds, such as PPCPs, pesticides, and BPAs, are effectively separated via RO membrane filtration [86]. Wang *et al.* [87]found that the overall removal efficiency in the final treated water is greater than 95% (at concentrations lower than 10 µg/L) for most PPCPs compounds. In addition, Katibi *et al.* [36], reported that nearly complete rejection (≥98%) was achieved for BPA separation with the application of polyamide-based RO membranes. Nevertheless, in contrast with other membrane filtration approaches (e.g., NF and UF), concentrates (brine) from RO treatment are primarily discharged to the surface water. These RO concentrates are almost 20 % of the influent concentration of rejected contaminants [88]. Thus, one challenge in RO is the management of brine generated from the filtration process that exerts harmful effects on the environment.

Predominately, the brine concentration is vary depending on feed water quality, effluent quality, type of treatment, and the nature of chemicals used [89]. In current findings, the effort of providing the environmentally friendly of brine treatment are still limited to be implemented. Although AOPs have been the most widely investigated method for brine treatment, the energy-intensive and high cost in a single treatment have limited its application**.** Interestingly, Xiang *et al.*[90] has reviewed and proposed treatment options for concentrates produced by RO membrane processes, and a hybrid treatment approach was recommended as a solution. The proposed integrated hybrid are comprising FO, pre-coagulation, AO, and post-biological treatment as a better option for brine treatment at lower cost and energy.

In the first stage, the post-treatment for the RO brine compound is to separate the dissolved compound from the concentrated brine. The dissolved organic matter except for hydrophobic brine is removed using different separation processes such as forward osmosis and coagulation. Then, the oxidation method is used to eliminate the concentrated hydrophobic compound separated at the early stage. Finally, as a supplementary approach, the post-biological treatment provides an efficient degradation of the hydrophilic intermediates produced at the secondary stage, eliminating the brine contamination from the RO water treatment process. This hybrid approached showing significant potential in improving the operation cost and usage of energy in the treatment system. However, this significant approached are very depending on the RO brine characteristics in selecting the material and method of the treatment process.

2.3.2 Nanofiltration

The NF membrane method has been recognized as a promising treatment method for micropollutants, such as hormones and pharmaceutical contaminants, in water and wastewater system [50,96]. In principle, NF membranes function with water pressure forced through nano-sized pores (between 0.2 nm and 0.4 nm), and contaminants are adsorbed on the membrane via charge and size interactions [97,98]. In addition, NF membranes exhibit charge selectivity for dissolved components [99]. Monovalent ions and water can permeate, whereas divalent and multivalent ions are retained [100]. In practice, the performance of NF membrane treatment is generally known to achieve high quality and produce effluents with low organic

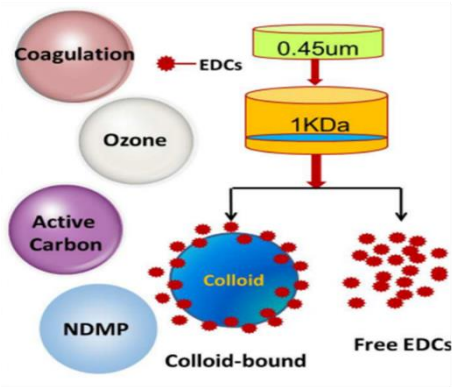
concentrations; moreover, the removal of microbes and viruses does not require adding chemical disinfectants [6,41]. However, Liu *et al.* [41] reported that the variety of membrane adsorbent types (membrane supports, such as polysulfone, ceramic acetate, polyacrylonitrile, and polyethersulfone) and the different size exclusion and charge repulsion properties of various EDC compounds can change the removal range (10% to 99.9%). Consequently, understanding the NF membrane mechanisms involved in rejecting target contaminant compounds is important. Semiao *et al.* [96] found that the pore radius of the active layer is a determining factor for the removal of adsorbing contaminants (i.e., estrone and estradiol) in NF membranes. Semiao *et al.* [96] added that a combination of partitioning effects and internal surface area access plays a role in the adsorption and retention of hormones (i.e., estrone and estradiol) by NF membranes. In addition, Semiao *et al.* [101] suggested that convection and diffusion are adequate transport modes for adsorbing hormones (i.e., estrone and estradiol) of NF membranes, with convection mechanisms significantly contributing to the transport of hormones at pressures higher than 11 bar. In this regard, the hormone removal mechanisms that use NF membranes have been investigated extensively. Nevertheless, many removal mechanisms for various contaminant types remain unknown. Numerous EDC contaminants in water are still difficult to remove using single treatment methods. In the actual applications of NF membranes to WTPs, multiple or hybrid treatments have been proposed to overcome the weaknesses of NF membranes. Therefore, NF membranes are commonly combined with AOP, and concentrated residuals require further treatment [50].

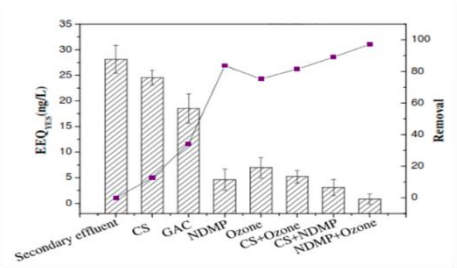
2.3.3 Ultrafiltration (UF)

UF is a low-pressure cross-flow membrane separation process with pore sizes ranging from 0.01 μm to 0.1 μm [91]. UF membranes are effective in eliminating macromolecules and particles, but their effectiveness is highly dependent on the type of material that constitutes the membrane [92]. UF membranes have elicited considerable attention due to their extensive applications to advanced secondary effluent treatment. However, the application of UF membranes to the removal of EDC is less effective. UF membranes have been reported to reject extremely few target EDC compounds compared with the NF and RO methods [93]. A comparative review of Patel *et al.* [49]concluded that the removal of selected pharmaceuticals (e.g., amoxicillin, naproxen, metoprolol, and phenacetin) via UF is moderately successful compared with NF processes. In addition, Ojajuni *et al.* [94] found that hydrophobic adsorption and size exclusion are the dominant mechanisms that retain EDC on NF membranes; meanwhile, UF membranes retain typically hydrophobic EDC. Consequently, UF is rarely used as a single treatment for the removal of EDC. Its limited use is attributed to the fact that the MWCO range (10–100 kDa) of UF membranes is higher compared with the molecular weight (<1 kDa) of most micropollutants[95].

Meanwhile, Huang *et al.* [96]studied the ultrafiltration process on effluents through 1kDa cross-flow into two phases, colloidal phase (0.45 μm-1kDa) and soluble phase (<1 kDa). They compared the estrogenic activity with the other processes, which are coagulation sedimentation (CS), GAC adsorption, magnetic ion exchange resin (NDMP), and ozone processes, as shown in **Figure 3**. The EDC with lower Kow values has higher removal activity by NDMP while the ozone process successfully removes both colloidal and soluble phases EDC. Therefore, the colloid-bound EDC has a good performance which suggested that the combination of NDMP and ozonation processes achieved a higher reducing estrogenic activity with satisfied the predicted no-effect concentration [97, 98]. In this regard, although UF allows the passage of

low-molecular-weight organic solutes through its membrane, it can be used as a pre-treatment for RO because it can remove high concentred effluent from passed RO membrane. Sun *et al.* [99] reported that compounds larger than pore is rejected by membrane surface and remain on the feed or concentrate side. Meanwhile, the smaller compounds than the pore can pass through the membrane to filtrate the compounds with a high fouling tendency (leachate effluents) to RO membranes [99, 100].





	pH	Dissolved oxygen (mg/L)	Conductivity (ms/cm)	SS (mg/L)	TOC (mg/L)
Secondary effluent	7.8	3.7	1.19	14.5	12.8
Coagulation sedimentation	7.5	4.1	1.17	10.2	10.9
Granular activated carbon	7.9	4.2	1.08	5.4	8.2
NDMP resin	7.4	4.3	0.97	11.2	7.5
Ozone	7.3	5.2	1.11	13.6	9.2

Figure 3. Estrogenic activity of effluent and removal by advance treatment through an ultrafiltration process. Huang *et al.* [108]

2.4 Ultrasonication (US)

US is an advanced treatment process for removing low-concentration (ng/L to µg/L) and complex contaminants in wastewater systems [101, 102]. It is fast, clean, and related to the wave degradation process, which does not produce secondary products [101]. In accordance with Chadi *et al.*[103] the wave degradation process of US treatment operates on the basis of the nucleation/growth/collapse of cavitation bubbles in water due to the high pressure and temperature caused by ultrasound waves. Complex contaminants exposed to ultrasound waves will undergo thermal and chemical reactions that promote the degradation of solutes in gaseous and aqueous solutions [104]. In general, the removal of EDC via US treatment has achieved efficient performance. For example, Im *et al.* [105]found that the degradation of PPCPs compounds (acetaminophen and naproxen) via US treatment demonstrated high removal efficiency for naproxen (>99%) and acetaminophen (86.1%) at 1000 kHz. Nevertheless, Naddeo *et al.* [106]indicated that the specific removal rate depends on the chemical structure of the analyzed compound. At a treatment time of 180 min, triclosan is nearly completely degraded (95%). However, other pharmaceuticals, such as erythromycin and iopromide, are only partially removed (50%) using the same ultrasound power at the same frequency. In particular, the degradation and removal of EDC via US treatment are influenced by several factors. Chu *et al.* [107]found that the removal efficiency of EDC varies and is influenced by the following: (i) water quality and sonication conditions (pH, temperature, background ions, promoters, US frequency, power, and reactor type); (ii) catalysts (non-carbon-based and carbon-based catalysts), and (iii) compound properties. To maximize the effectiveness of EDC degradation through US treatment, a comprehensive assessment is necessary to establish

standard conditions for each environmental matrix. Thus, the evaluation of US treatment for various types of water and wastewater sources should be explored.

3. Biological treatment

3.1.Biological active carbon (BAC)

BAC filtration is a combination of GAC adsorbents under cover of a biofilm. In principle, BAC treatments are dominated by adsorption and microbial degradation mechanisms [108]. Under this dual mechanism, contaminants and dissolved oxygen in the treated solution interact with granular activated particles and microorganisms [109]. In practice, the combination of this dual mechanism process has been found to remove low-level EDC successfully during the treatment of drinking water [49]. Li *et al.* [110]determined that the presence of a readily biodegradable carbon source is beneficial for E2 removal in a BAC reactor, and an E2 removal ratio higher than 99% was maintained regardless of the primary carbon source type, as shown in **Figure 4**. In particular, BAC filtration was implemented as a complementary treatment for solutions with low-level concentrations. Chuang *et al.* [111] found that treatment via BAC filtration after ozonation can reduce the concentrations of most remaining estrogenic compounds by up to 95%. Consequently, BAC filtration is likely to be used as a tertiary treatment process[112]. Moreover, the combination of advanced treatments with BAC filtration under recycled carbon can provide considerable reductions in the capital costs of operation [113]tras

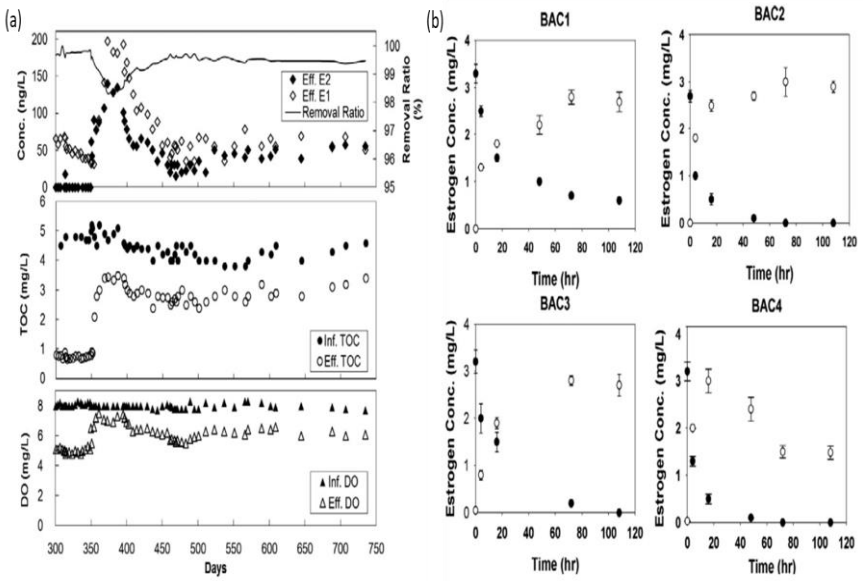


Figure 4. (a) Response of BAC reactor performance when additional carbon source was switched from acetic acid to humic acid on Day 350. BAC reactor was operated at a constant EBCT of 30 minutes from Day 300 to Day 735. (b) Profile of Estrogen degradation for four E2 degrading isolates (BAC1-BAC4). Solid and open circles represent E2 (o) and E1 (•) concentration, respectively [110].

3.2.Biological nitrification and denitrification (BND)

BND is a biological process of oxidation and reduction that supports the high removal of organics through heterotrophic and slow-growing nitrifying microorganisms [114]. At present, BND treatments have been applied as secondary treatments after performing other advanced biological treatments, such as the application of MBRs or anaerobic/aerobic treatment systems

[115]. Under this type of treatment, EDC removal performance is largely effective for estrogenic and several pharmaceutical compounds. Ting *et al.* [116] reported that secondary activated sludge treatment, followed by nitrification/denitrification, effectively stopped more than 95% of estrogenic activities. However, removal performance for other EDC compounds, such as carbamazepine, diclofenac, clofibricacid, gemfibrozil, erythromycin, and pesticides (atrazine and fenoprop), exhibited lower efficiency [117]. In this regard, the performance of a BND treatment alone is less effective compared with dual or hybrid treatment processes. Wigginton *et al.* [118] indicated that the major differences in ammonia-oxidizing and nitrous oxide-reducing community composition and structure between centralized and decentralized BND wastewater treatment systems. Therefore, the challenge posed by the BND process can be effectively managed by combining it with other advanced biological treatments.

3.3.Microalgae

At present, microalgae-based wastewater treatments have received considerable attention for removing EDC effluents because they provide high quality treated effluents [33, 119, 120]. Moreover, microalgae treatments are considered economical practices due to the multiple uses of microalgae, such as in pollutant removal and energy resources [119]. Young *et al.* [121] reported that shallow raceway reactors produce high-rate algal and consume oxygen via microalgal photosynthesis. Thus, microalgae-based treatment systems do not require external oxygen aeration in the system [122]. In practice, microalgae-based treatments have been proven to remove EDCs effectively from effluents through evaporation, photodegradation, and biodegradation mechanisms [33]. Wang *et al.* [120] evaluated algae-mediated biotransformation as possible mechanisms for removing 17α-ethinylestradiol (75.3% removal) and 17β-estradiol (95% removal). In addition, Ruksrithong *et al.* [119] proved that biodegradation and adsorption were the predominant mechanisms for removing E1 and E2. Consequently, the effectiveness of microalgae-based biodegradation has successfully eliminated most estrogenic and pharmaceutical effluents in wastewater systems. Nevertheless, the removal performance for pesticide compounds is relatively low (32%–89%) compared with other EDC compounds [123]. In this regard, the culture and growth of microorganisms can be further examined to understand the effectiveness of microalgae in removing pesticides. In particular, the integrated biological treatment process with microalgae is among the potential options for further improving performance.

3.4.Fungi

The fungal treatment method utilizes the enzymatic biodegradation of micropollutants that have been alternatively used in EDC removal. The capability of fungi to degrade EDCs in synthetic media and real wastewater has been widely explored. Among conventional methods used in practice, fungal treatments can be considered efficient biological treatments for removing pharmaceutical compounds [37]. Naghdi *et al.* [124] reviewed that literature found the stable fungal reactor requires systematic investigation on the contribution of biosorption and biodegradation during removal of PhACs and micropollutants [125]. In addition, Mir-Tutusaus *et al.* [126] found that a fungal operation successfully removed analgesics and anti-inflammatories and even eliminated the most recalcitrant pharmaceutical families, such as antibiotics and psychiatric drugs, from wastewater. In practice, the degradation efficiency of fungal treatments is dependent on several factors, such as sterilization processes, fungal nutrient additions, and aeration rates [127]. Moreover, the wastewater type can affect the

survival of fungi during the treatment process. Highly concentrated wastewater can reduce the number of surviving fungi due to bacterial competition. Therefore, the application of fungal biological treatments is reliable for secondary treatment or post-treatment processes. The pretreated effluents obtained using physical methods (i.e., RO and coagulation–flocculation) are the most suitable for the fungal treatment process [126].

3.5.Biosorption

El–Naggar *et al.*[128] described biosorption as a dual mechanism (i.e., bio-oxidation and sorption) process that occurs when microorganisms are immobilized onto an adsorbent. In practice, the removal of EDCs using biosorption treatments is effective for highly hydrophobic compounds [138]. Dhangar *et al.* [86] reviewed the removal of 17β-estradiol, 17α-acetate, pentachlorophenol, 4tert-octylphenol, and triclosan achieved using a biosorption treatment process. Similar observations indicated that the soluble concentrations of target compounds decreased rapidly for selected microconstituents [129]. In particular, soluble or hydrophobic EDC compounds are primarily targeted to be removed using biosorption treatments. Nevertheless, in slow decrement processes, most extremely hydrophobic compounds are also removed by a biodegradation mechanism. The theory of the biosorption process in wastewater treatment can be deeply investigated using sufficient isotherm data, mass transfer coefficients, biological growth, and destruction activity values estimated from independent measurements [130, 131]. Therefore, the efficiency of biosorption treatments in removing EDCs should be compared with other biological treatments. In addition, regardless of hydrophobic properties, the efficiency of removing EDCs using biosorption treatments can be further enhanced via dual or hybrid treatment processes.

3.6. Membrane bioreactors (MBRs)

The use of MBRs is a superior biological treatment method that has elicited substantial attention due to its technical innovations and high quality treated effluents compared with other conventional biological treatments (i.e., activated sludge treatment) [132, 133]. In particular, the efficiency of MBRs is attributed to the retention of sludge on the membrane surface, which promotes extensive microbial degradation and physical retention of all molecules [134]. In accordance with Hai *et al.* [133], the effectiveness of MBR treatment is dependent on sludge age, concentration, the existence of anoxic and anaerobic compartments, wastewater composition, operating temperature, pH, and conductivity factors. For example, Park *et al.* [135] reported that considering the slow degradation of pharmaceuticals, removal by using MBRs is better due to the relatively old ages of sludge, leading to the development of distinct microbial communities in MBRs compared with that in activated sludge plants. In addition, Arca-Ramos *et al.* [136] determined that one stage and two stages enzymatic MBR is performed for the removal of micropollutants from secondary effluents. In practice, MBR treatments have demonstrated effective performance for the removal of EDCs. Approximately 81% to 99% of various types of EDC compounds (e.g., pharmaceuticals, pesticides, and phenolic and estrogenic hormones) have been removed via MBR treatment [117, 132, 137]. In addition, several PPCPs, such as salicylic acid and propylparaben, have been eliminated (100%removal) using MBR treatment. Nevertheless, some pharmaceuticals (ofloxacin, sulfamethoxazole, lorazepam and propranolol) are poorly degraded via MBR treatment and has been another approach on membrane fouling and current density [138]. In addition, membrane fouling rates decrease the removal efficiencies. In this regard, alternative solutions

should be explored to investigate the optimum configuration for high MBR treatment performance.

3.7.Constructed wetlands (CW)

A CW is a natural treatment method that can be operated simply and has a low environmental impact. CWs are defined as an integrated engineered biological treatment system that can replicate the natural wetland concept through a combination of biological (biodegradation), physicochemical (sorption), and chemical (oxidation) interactions among plants, substrates, and soil [139]. In general, wetland types are classified into two basic systems: vertical flow CWs (VFCWs) and horizontal subsurface flow CWs (HSSFCWs) [140, 141]. Thalla *et al.* [142] defined VFCWs as highly aerobic systems that drain wastewater vertically through the planted matrix, allowing excellent oxygen transfer that favors aerobic microbial processes. Meanwhile, HFCW systems are considered anoxic systems that favor anoxic (oxidized and reduced zones) microbial processes, such as traditional denitrification [142]. At present, CW treatments have demonstrated effective performance in removing EDCs. For example, Chen *et al.* [143] reviewed that a constructed wetland that utilized lightweight expanded clay aggregate (LECA) substrate and macrophyte species which is *Iris Sibirica*, effectively treated wastewater contaminated with carbamazepine with higher than 90% removal efficiency. The removal mechanism is illustrated in **Figure 5**. In addition, the monitoring results of Tatoulis *et al.* [144] indicated that HSSF is a promising technology design with many innovations such as reducing area, minimized clogging risk and various plastic media to remove contaminants as well as suspended solids from municipal sewage due to its high treatment efficiency [144, 145]. Technically, the efficiency of CW treatment is predominated by the sorption processes (composting materials) of plant materials (resistant to the biodegradation of organic materials). The hydrophobic properties of the contaminants further enhance the sorption process for the support matrix (i.e., plant materials) of the treatment process [146]. Nevertheless, the effectiveness of CW treatment is highly limited to the availability of land in a community area; moreover, the treatment requires city communities to find a suitable location for treatment areas [141, 147]. Constructed wetland microcosms (CWMs) also another approach on artificially designed ecosystem which utilizes both complex and ordinary interactions between supporting media, macrophytes, and microorganisms. This design able to treat almost all types of wastewater, green and sustainable technology that consists of lower energy input, less operational cost and flood control [148]. In this regard, further investigations are necessary to extend the benefits of CW treatment.

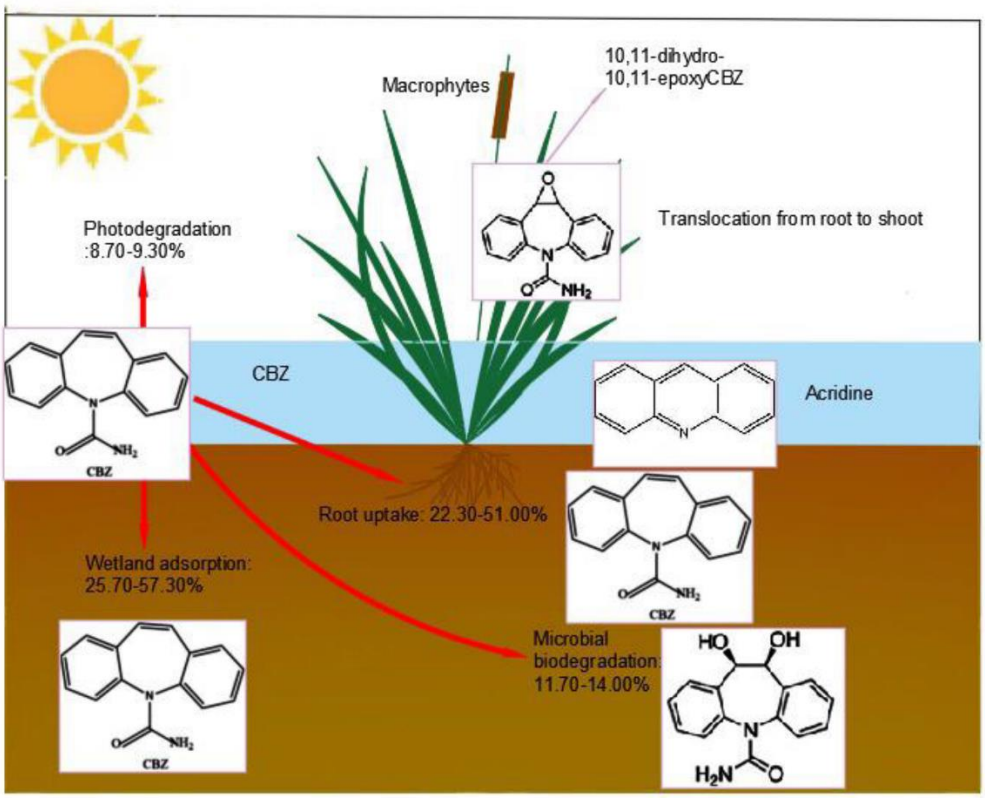


Figure 5. The contribution of different degradation pathways to CBZ degradation in constructed wetlands [143]

4. Chemical treatment

4.1.Chlorination

Chlorination is the most common conventional method used in drinking water and wastewater treatment processes [149, 150]. In principle, chlorination is a chemical disinfection method used to eliminate microorganisms by disrupting the activity of cell membrane respiration. In water treatment systems, chlorination methods are located in either the primary or secondary treatment steps. This method is used to control the biological growth and ensure that the appropriate chlorine residual levels are consistent throughout the distribution system [151]. In general, the performance of chlorination treatment is less effective for removing EDCs. Chlorination treatment is most effective for removing estrogenic compounds (>98%) compared with other synthetic EDCs [152]. Most pharmaceutical compounds were not completely degraded by chlorination and needed other process stages to increase removal efficiency [153-155]. In addition, Matsushita *et al.* [156] reported that pesticides with strong mutagenicity after chlorination could be degraded after the chlorination process by PAC adsorption through hydrophobic interaction. Du *et al.* [157] indicated that toxic and harmful by-products had been identified after the treatment process. In particular, chlorination treatment requires an advanced process for secondary treatment or post-treatment. A single treatment via chlorination is ineffective in eliminating various types of EDC compounds. Moreover, chlorine dose, contact time, and pH conditions during the process can differ for each EDC compound [158]. Therefore, standard effective conditions for chlorination treatments can be developed to eliminate EDCs. Simultaneously, the chlorination conditions for removing EDCs can be optimized.

4.2.Ozonation

Ozone (O_3) treatment is among the methods that have received continuous attention in wastewater treatment. As part of the oxidation process, ozone is a highly reactive gas that can oxidize bacteria, organic materials, and micropollutants in water and wastewater systems [159]. In practice, the efficiency of ozonation mostly depends on the reaction rate of the process. Leresche *et al.* [160] found that OH radicals in the ozone process create fast reactions with the compounds that contain electron-rich centres with electron-donating substituents to degrade the organic and inorganic compounds. In addition, the optimization of ozone treatments depends on factors such as ozone dosage, energy input density, catalyst type, temperature, airflow rate and pressure [161, 162]. In general, ozone has been demonstrated to remove EDCs effectively. The removal efficiency of EDCs in water and wastewater systems by using the ozonation process ranges from 40% to 100%. Si *et al.* [163] reported that complete removal of EDC in wastewater is done through ozonation treatment. Moreover, ozonation treatment recorded good performance in several pesticide and PPCPs compounds with higher removal efficiency [164, 165]. Moreover, the treatment of EDCs by using ozonation can also produce additional by-product or transformation product (TP) residuals. Soltermann *et al.* [166] observed a remarkable negative effect on the toxic oxidation by-product bromate in bromide-wastewater after ozonation treatment. Predominately, the transformation products (TP) is a low concentration by-products that produce through incomplete mineralization of micropollutants in ozonation process[109].

As reported by Kharel *et al.*[167] , the maximum yield of TP are occurred at the same specific ozone dose (Z = 0.55 mg O_3/mg DOC) from any individual WWTP. The concentration of the TP is largely dependent on the ozone dose implemented in the process and the elements of wastewater matrix as well as the types of EDC. The major constituent such natural organic matter (NOM) in wastewater matrix influencing the efficiency of the treatment process that competing with the targeted compounds[168]. In practice prefiltration could use to remove the non-targeted compound from the wastewater matrix. In current practice, the biological and physical post treatment methods are introduced in eliminates the TP residuals that danger the environment. These effluents polishing treatment such as sand filtration, moving bed, fixed bed and granular GAC filtration, AOP are introduced to stabilized TP from its parent compounds[169, 170]. Nevertheless, the current conducted analysis is still limited to the specific targeted compounds, there are many nontargeted substances that are potentially relevant to be identified. Therefore, further analysis and optimization studies should be conducted to reduces and eliminates the TP of reaction intermediates.

Fenton processes

4.2.1. Fenton

Fenton processes are economical treatment methods that work through the degradation of contaminants from the oxidation reaction of hydroxyl radicals. In practice, organic pollutants are attacked by hydroxyl radicals, resulting in the complete breakdown of contaminants into CO_2, water, and inorganic salts as end products [171]. Hydroxyl radicals remove the electrons from the present contaminants to form hydroxide anions and gain a hydrogen atom to replace the atom that disappears during the process [172]. In particular, the performances of Fenton treatments are primarily controlled by reaction process parameters. Mirzaie *et al.* [173] asserted that the parameters of reaction concentration, catalyst type, pH, radiation intensity, water

matrix, substrate salinity, feeding mode, temperature, and reaction time are significant for the performance of Fenton treatment processes. In general, these processes are largely involved in the removal of EDCs in water and wastewater treatment systems. In Sun *et al.* [174], Fenton oxidation techniques were successfully utilized to remove E3, BPA, diethylstilbestrol (DES), E2, and EE2 in cow manure wastewater, achieving a removal efficiency of 84.9%, 99.5%, 99.1%, 97.8%, and 84.5%, respectively as shown in **Figure 6**. In addition, Amin *et al.* [175] achieved the removal of carbamazepine by using Fenton-like oxidation with Fe@Fe_2O_3 nanowires while Dwiwedi *et al.* [176] removed using granulated activated carbon (GAC) filtration. Nevertheless, Fenton processes suffer from certain limitations that prevent them from being used as effective treatment processes for EDC removal. Mirzaie *et al.* [173] determined that the regeneration of iron ions is infeasible, and the final effluent should be treated to meet the discharge standards for iron concentrations. In addition, the consumption of greenhouse gas emissions is a critical aspect that should be focused on. Therefore, further investigations should be conducted to develop a sustainable contaminant removal process.

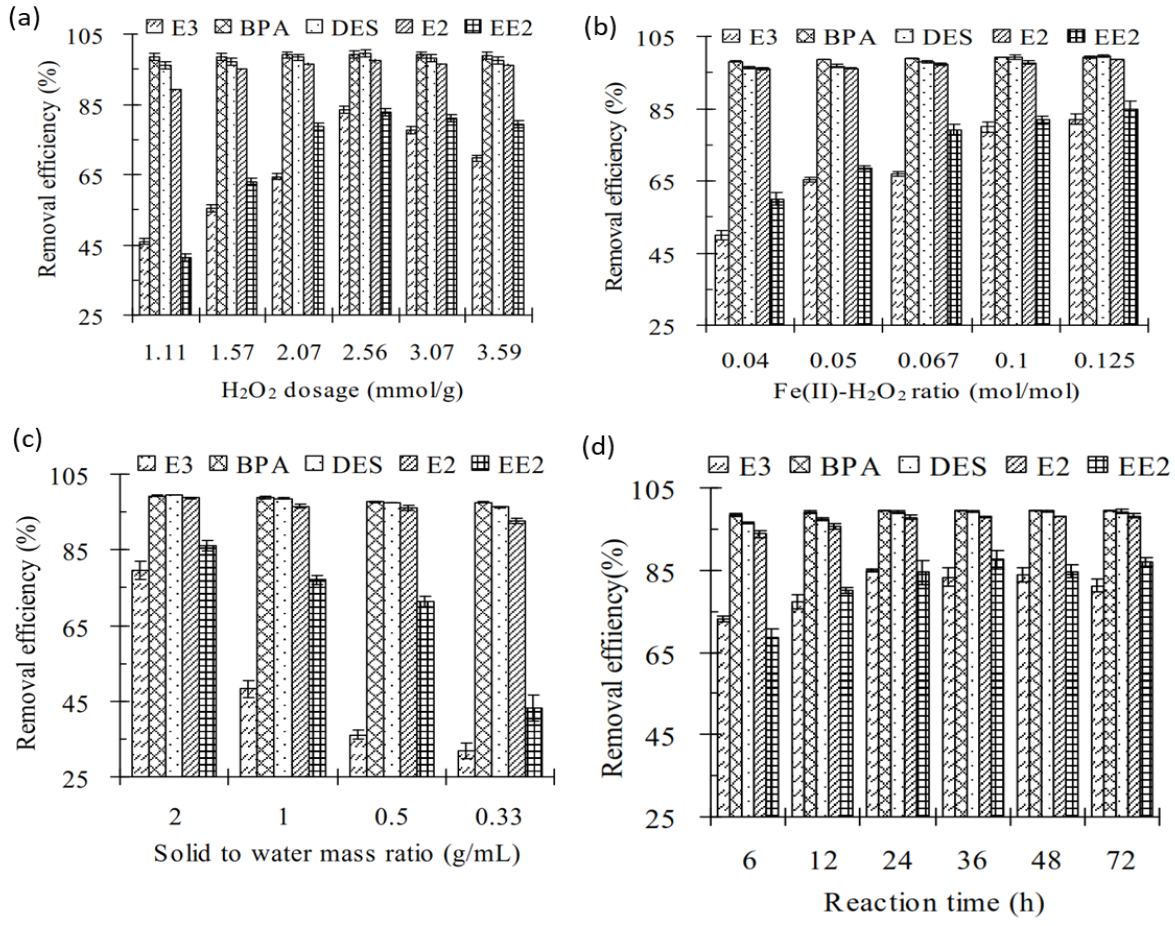


Figure 6. (a) Effect of the H_2O_2 dosage, (b) Effect of the Fe (II) to H_2O_2 molar ratio, (c) Effect of the solid to water mass ratio, (d) Effect of reaction time on the removal efficiency of estrogens from cow manure by the Fenton oxaidation process [174].

4.2.2. Photo-Fenton

In photo-Fenton treatment, Fe(III) is widely used as a catalytic agent to produce hydroxyl radicals through the reaction with H_2O_2 under ultraviolet (UV) light [177]. In this reaction

(preferably under acidic condition), hydroxyl complexes, such as $Fe(OH)^{2+}$ and $Fe(OH)_2^{4+}$, which absorb light in the UV/visible region, undergo photoreduction to generate hydroxyl radicals and Fe^{2+} [178]. In particular, an acidic condition is preferable to achieve optimum performance; nevertheless, neutralization processes are required before discharge[179]. At present, photo-Fenton processes have been effectively used to remove many types of hormones, phenolic, pesticide, and PPCPs compounds. Pharmaceutical compounds have been effectively removed via photo-Fenton treatment, with removal efficiency ranging from 95% to 100% [180-182]. For antibiotics removal such as ciprofloxacin, amoxicillin, sulfathiazole, and sulfamethazine, the efficiency achieved in range of 80- 95%. The photo-Fenton process under UV and solar radiation reduced total coliforms and E. coli after 90 min [183]. An interesting finding of Silva *et al.* [184] is the complete reduction of estrogenicity (E2) and seven endocrine disruptors (EDs) through LED irradiation as an alternative to solar photo-Fenton in case solar radiation is not available, thus reducing hazards associated with WWTP effluents reuse or discharge. In this regard, photo-Fenton processes seem promising methods for EDC removal. Under certain conditions of photo-Fenton modification, catalyst design and surface modification on the catalyst can improve the oxidation efficiency to remove the micropollutants [179, 185]. Therefore, investigating and extending knowledge on the formation of highly reactive ions will be beneficial for determining the fate of reactions.

4.2.3. Electro-Fenton

Electro-Fenton is an approach used to enhance the generation of hydroxyl radicals by combining Fenton and electrocoagulation processes. Under these conditions, the oxidizing power of H_2O_2 increases with the electrical assistance of the electro-Fenton process [186]. In general, electro-Fenton treatments operate under two configuration conditions, which are through (i) the high catalytic activity of inert electrodes used as anode material, with Fenton reagents added to the reactor from the outside; or (ii) hydrogen peroxide is added from the outside and Fe^{2+} is provided from sacrificial cast iron anodes [187, 188]. During this process, H_2O_2 that is electrochemically generated from the process is used to increase the degradation of high-strength organic pollutants in wastewater systems [189]. In particular, electro-Fenton is highly effective in eliminating high concentrations of EDC pollutants in wastewater treatment. Moreira *et al.* [190] reported that electro-Fenton eliminates pharmaceutical and pesticide compounds at concentrations of 100000 $\mu g L^{-1}$. In addition, other types of electro-Fenton-based treatments, such as bio-electro-Fenton, photo-electro-Fenton, and solar electro-Fenton, have demonstrated highly effective performance in removing EDCs [191-193]. Nevertheless, this advanced treatment process requires high operation and maintenance costs. The costs of conventional treatment methods are considerably lower than the costs of the electro-Fenton process. In this regard, the cost-effectiveness of the proposed treatment plant should be reviewed and evaluated before wastewater treatment operation is conducted.

4.3.Photolysis

Photolysis is a photodegradation process that results from the irradiation and adsorption of a UV light photon that is conditionally processed by either direct absorption of the UV light photon or indirectly by using a photon sensitizer (i.e., H_2O_2) [194]. In practice, photolysis is a less effective process for the treatment of low EDC concentrations (5–10 $\mu g\ L^{-1}$) under UV light absorption with slow degradation rates. However, the artificial light treatment process can successfully eliminate most EDCs at high concentrations (0.7 to 2.5 $mg\ L^{-1}$) with a removal

efficiency of 80 – 97 % [3]. Nevertheless, under the indirect influence of H_2O_2/UV, the influence of UV is greater than the typical disinfection practice and the standard UV/AOP applications [195]. Apart from UV light, gamma radiation also exhibits potential in the degradation of EDCs through photolysis. Rozsa *et al.* [196] found that photolysis through gamma radiation successfully transformed atrazine at a concentration of 1×10^{-4} mol L^{-1} and 4.6×10^{-7} mol L^{-1}. However, gamma radiation requires high maintenance and operation costs. In addition, further investigations are required to evaluate the residual effects on the environment. In this regard, comparisons of photolysis are necessary to provide a clear assessment for selecting an efficient treatment.

4.4.Photocatalysis

Photocatalysis is a photoactivation process that transforms chemicals through the irradiation of semiconductor metal oxides (as catalysts). Photocatalysis is among the most favored treatments due to its environmental compatibility [197]. Gopinath *et al.* [198] reported that photocatalytic oxidation had been demonstrated to be a promising technique because of its nontoxicity, relatively low cost, lack of mass transfer limitations, chemical stability, and possible operation at ambient temperatures. To date, various types of photocatalysts have been used for this treatment process. However, TiO_2 and ZnO are the most frequently used catalysts for photocatalytic processes in water treatment due to their high photochemical stability and piezoelectric characteristics [198, 199]. In particular, the application of ZnO exhibits higher EDC removal efficiency compared with the application of TiO_2. In principle, the efficiency of the EDC removal process through photocatalyst treatment depends on process parameters, such as catalyst dosage, substrate concentration, pH, and photocatalyst modifications [200]. TiO_2 catalyst will give the best performance compared with other catalysts in term of various design such as carbonaceous composites [197, 201]. In particular, although a modified approach of photocatalysis and photosensitized oxidation has been widely studied to enhance the use of commercial photocatalysts (TiO_2), the application of photocatalytic technology with TiO_2 to wastewater treatment has been limited within the wavelength range of radiation (λ<388 nm). Nevertheless, the photocatalysis process of UV/TiO_2 has completely (100%) eliminated pharmaceutical compounds in wastewater treatment regardless of this limitation [201]. In this regard, economic aspects and operational costs are major evaluation points in defining a suitable photocatalyst approach for wastewater treatment.

4.5.Green catalytic oxidation

Since introduced by Collins in 2002 [202], the applications of iron tetra-amido macrocyclic ligand (Fe-TAML) green catalysts in oxidation processes have received substantial attention from many researchers. Fe-TAML functions as a peroxide activator and provides a robust, broad, and green oxidation process [203]. In practice, the catalytic activation of H_2O_2 through Fe-TAML catalysts has significantly contributed to enhancing the oxidation process in water treatment. Collins *et al.* [204] determined that the slow kinetics of the H_2O_2 oxidation process is activated by the Fe-TAML catalyst and results in the complete degradation of water contaminants. In contrast with other catalysts, Fe-TAML can significantly produce an effective mild oxidizing condition within a shorter period, i.e., low-concentration usage, neutral pH, and ambient temperature conditions [202]. In addition, no residual toxicity product is produced from catalytic oxidation treatment. In practice, Fe-TAML green catalysts have exhibited effective performance in EDC removal. Wang *et al.* [205] reported that the degradation of

natural and synthetic estrogens is rapid, with 95% of the original estrogens degraded after 15 min of treatment. Moreover, the total degradation of phosphorothioate pesticides, such as fenitrothion, parathion, and chlorpyrifos, by using Fe-TAML/H_2O_2 oxidation has been observed [206]. In particular, the high degradation rates of Fe-TAML catalysts are attributed to the head and tail ligand sections that considerably affect the lifetime and rates of hydrolytic degradation [204]. Such potential makes a remarkable contribution to the removal of emerging contaminants in wastewater treatment. Nevertheless, data on the performance of Fe-TAML catalysts in real WTPs remain limited. In this regard, further investigations are necessary to develop a successful green oxidation treatment for water and wastewater plants.

5. Alternative treatment approached

Combinations of EDC treatment methods have been used to overcome the limitations of single treatment methods. A variety of treatment methods have been proposed through the integration of treatments into sequential and hybrid approaches. These methods have successfully reduced the high concentrations of EDCs and have eliminated the low concentrations of emerging contaminants in water and wastewater systems. At present, EDC hybrid treatment methods have been mostly developed through the combination of chemical treatment with physical or biological treatment methods [207, 208]. In addition, EDC hybrid treatment methods have also been developed using a combination of physical and biological treatment methods [209-211]. Examples of remediation approaches for EDC removal through the integration of sequential and hybrid treatments are presented in Table 2.

Sequential treatment approaches for EDCs integrate treatment in accordance with the categories, followed by another treatment. As indicated in **Table 2**, two types of physical treatment methods are integrated to improve EDC removal performance. UF treatments are mostly considered to combine with other physical treatments, such as adsorption treatment methods (AC and CNT). Li *et al.* [212] reported that the removal rate of EE2 increased dramatically from 7.01% to 80.03% by using a PAC/UF hybrid process compared with a single UF treatment, as shown in **Figure 7**. Constructed wetland models have also been developed for sequential treatment to increase removal efficiency for EDCs [213, 214]. The integration of advanced chemical processes through photocatalysis and ozonation [O_3/TiO_2/ Fe(III)/UV] has successfully eliminated high BPA concentrations (100 µg/L) in water. Alternatively, the integration of similar treatment processes is suitable for implementation in pre-treatment and post-treatment systems.

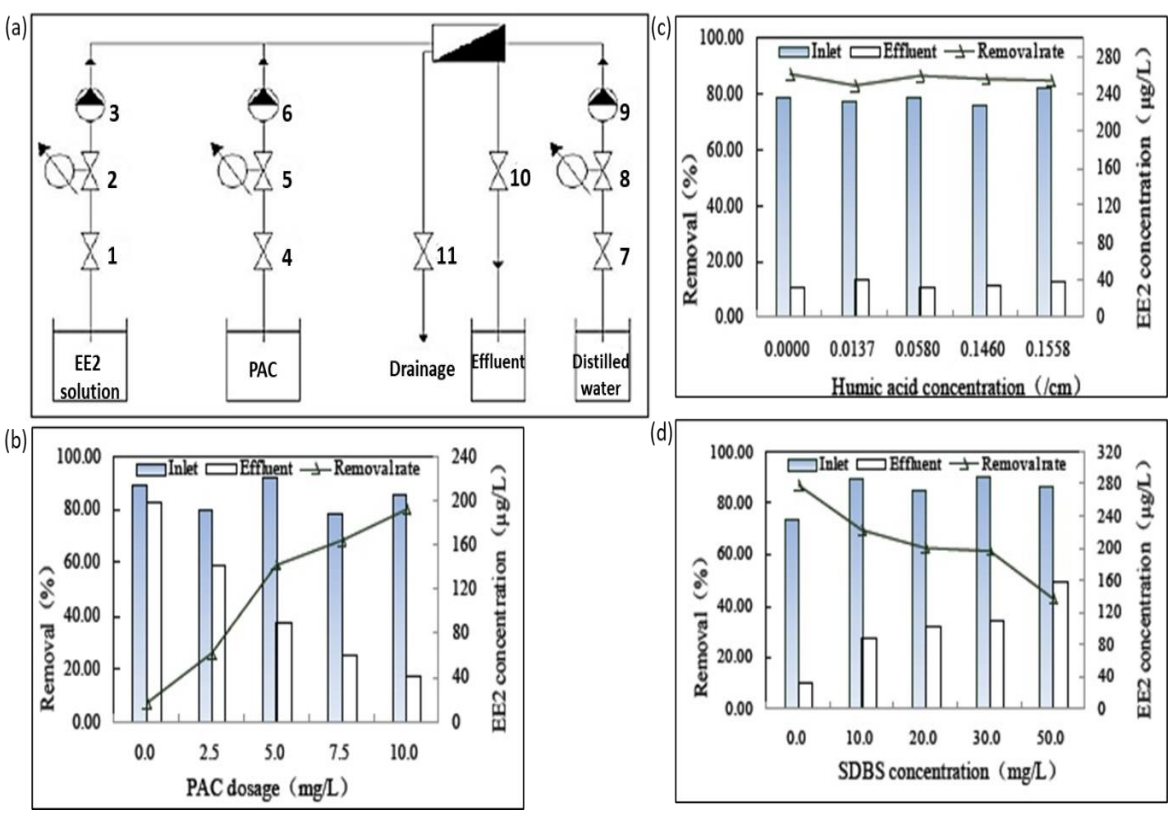


Figure 7. (a) Schematic diagram of the PAC/UF system: (1) valve, (2) pressure gauge, (3) pump, (4) valve, (5) pressure gauge, (6) pump, (7) valve, (8) pressure gauge, (9) pump, (10) valve, (11) valve. (b) Effect of PAC dosage, (c) Effect of natural organic matter (NOM), (d) Effect of anionic synthetic detergent on the removal efficiency of EE2 [212].

Meanwhile, hybrid treatment processes combine two or more treatments and formulate efficient removal strategies for EDCs. Oxidation chemical-based treatment processes, such as ozonation, have been combined with other physical and biological treatment processes. Combinations include ozonation with GAC, BAC, biofiltration, and anaerobic treatment [112, 215, 216]. In addition, other chemical oxidation-based treatments, such as photocatalysis, photo-Fenton, and electrochemical oxidation, have also been combined with physical-based treatment processes [217-219]. These hybrid treatment processes have successfully eliminated emerging contaminants at high concentrations of up to 200 µg/L. Eliminations are comparative depending on the types of EDCs removed.

Meanwhile, MBR-based hybrid treatments are effective for eliminating large amounts of EDC compounds in water and wastewater systems. A few studies have applied hybrid MBR processes, followed by functionalized biochar, GAC, RO, NF, and UF; these processes have successfully eliminated most EDCs in wastewater samples [220]. For example, 500 µg/L of EDC compounds (E1, E2, EE2, E3, BPA and 4-tert-butylphenol) are completely eliminated through the hybrid treatment of MBR with fBC [221]. Nghiem *et al.* [222] also reported the GAC post-treatment was observed to significantly complement MBR treatments to obtain high overall removal efficiencies of less hydrophobic and biologically persistent trace organics. The BPA contaminants found more effectively removed through the hybrid ozonation-based treatment. The hybrid ozonation treatment with adsorption and catalytic ozonation has successfully recorded high removal of BPA [223]. Hooper *et al.* [224] observed approximately

15% blend of advanced treated reclaimed water met potable water quality criteria. They conducted the ozonation treatments without the use of RO, where nitrate is below the MCL of 10 mg nitrogen per liter, and total dissolved solids are below the SMCL of 500 mg per liter. Overall, the performance in removing EDC contaminants is improved through the MBR and hybrid ozonation-based treatment system. Meanwhile, an AOP exhibits better removal performance as a hybrid-based treatment.

Table 2. EDC removal by integrated sequential and hybrid treatment methods.

Specification	Treatment types	Target compounds	Influent Concentration (% removal)	Ref.
Sequential treatment	Activated Carbon with Ultrafiltration (PAC/ UF)	17α-ethynylestradiol (EE2)	200 µg/L (80 %)	[212]
	Constructed Wetlands (VFCWs / HFCWs / FWCWs)	Triclosan	0.15 µg/L (79 %)	[208]
	Oxidation, ozonation-electrodegradation	Nonylphenol	50 ppm (70%)	[208]
Chemical/Physical hybrid treatment	Adsorption and catalytic ozonation (MWCNTs/Fe_3O_4)	BPA	25 to 75 ppm (98%)	[207]
	Fenton and Phanerochaete chrysosporium	BPA	1 g/L (58.23%)	[171]
	Constructed wetlands: Effective plant-bacteria	Pharmaceuticals	17.52 µM (66%)	[146]
	Fenton and GAC	Carbamazepine	8.5 g/L (49.39 ± 0.93%)	[176]
	UV/H_2O_2	Nonylphenol deca-ethoxylate	100 mg/L (99%)	[218]
Chemical/Biological hybrid treatment	Multi-stage biofiltration	PPCPs	Not mentioned (79%)	[55]
	Biological treatment and photo-fenton	Pharmaceuticals	(80-95%)	[183]

Biological/Physical hybrid treatment	MBR and UF	E2, E3, BPA, Triclosan	5 μg/L (99.5%, 98.3%, 98.6%, 99.2%)	[209]
	MBR and NF	17β-estradiol-17-acetate, 4-tert-butylphenol, Triclosan	5 μg/L (99.3%, 95.7%, 98.7%)	[209]
	MBR and RO	E1, E2, 17β-estradiol-17-acetate, 4-n-nonylphenol, Triclosan	5 μg/L (99.4%,99.6%, 100%, 100%,99.2%)	[209]
	MBR and GAC	E1, E2, EE2, E3, BPA, 4-n-nonylphenol, 4-tert-butylphenol, Triclosan, Ibuprofen Diclofenac	5 μg/L (100%)	[209]

6. Challenges and potentials

The effects of EDC on health and environmental issues have raised public concern due to their active capabilities at trace levels. The trace levels of EDC require accurate and sensitive quantification and detection for the broad determination of EDC in environmental matrices [225]. Interestingly, various interferences of EDC sources can form new and unknown emerging contaminants that can continuously expand [226]. The degradation of EDC compounds can extensively lead to the formation of various matrices of by-products [33]. At present, more studies should explore the effects of existing contaminants on the formation of intermediate reactions in water and wastewater treatment systems. In addition, the optimum process parameters, reaction mechanisms, and removal kinetics of the treated samples should be studied to ensure performance efficiency.

In the biological treatment method, the factor of suitability, capacity, reliability, and safety of using this biological water treatment technology is commonly raised [227]. Besides, the uncontrolled growth of microorganisms during the treatment process could also affect the process stability in term of cleanliness and material durability [228]. As reported by Abu Hassan *et al.* [227] the selectivity of microorganisms towards specific contaminants could overcome the inadequacy of the water treatment. To be precise, the biological treatment method requires full monitoring of the living organism applied. As the process started, the changes in microbial communities and operating condition will affect the performances of treated water quality. Therefore, the process sensitivity and stability of biological treatment are highly requiring a segregation of microbial community that meet the specific treatment condition of EDC removal.

Meanwhile, physical treatment methods have been extensively used in EDC treatment processes, particularly for drinking water treatment. The applications of adsorption treatments

in the removal of EDC are expanding with a variety of adsorbent materials. The effectiveness of AC and CNT are comparable with the effectiveness of other adsorbent materials, such as cellulose and graphite. However, the current study is mainly focused on the batch system adsorption approached. The evaluation for continuous scale implementation is still limited. Besides, the membrane filtration that shows the effectiveness is limited to a few membranes such as RO, NF, or UF[44]. The study of membrane filtration is still limited under the selected number of compounds and process condition.

In addition, Hybrid MBR treatments have resulted in the considerable removal of EDC in water and wastewater samples. AOP of chemical treatments are found to be the effective method for EDC removal. The application of chemical oxidation methods, such as ozonation, UV photocatalysis, and photo-Fenton processes, has been found to be the best EDC removal process for most PPCPs compounds. However, the potentials of AOP and chemical treatments remain open to be studied. A substantial consideration should be taken in identifying an effective treatment process that contributes efficiently. Besides, a more advanced oxidation approached should be extensively studied in real industrial-scale applications.

As a matter of fact, optimum and standard EDC treatment methods have not been well established in providing the solutions for the clean and safe conditions of water and wastewater systems. The formidable challenge begins with the identification of reliable analytical techniques and EDC treatment that cover complex environmental samples. In addition, the different conditions of various WWTPs, STPs, and drinking water treatment plants (DWTPs) require specific operational protocols. Therefore, the tremendous challenges when moving toward EDC remediation initiatives have resulted in the exploration of a wide potential scope to identify the improvements for EDC removal.

In principle, an actual treatment solution for the removal of specific EDC should be explored by integrating and optimizing advanced physical, chemical, and biological treatment methods. The exploration and investigation of each advanced treatment process are necessary to determine unknown potential limitations. The route of EDC treatment is potentially exploring the importance of reproducibility efficiency toward a green environment. In addition, the most important aspect that potential to be focused on is the effectiveness of reducing the effects of treated EDC samples on humans and other living organisms. The establishment of specific treatment mechanisms is highly valuable in relation to human health and living organisms' importance.

7. Conclusions

EDC has been proven to pose various potential threats to the environment and to living organisms. Given the various concentrations and complexity of EDC compounds, the treatment of EDC contaminants requires accurate and appropriate sampling, determination, extraction, quantification, storage, and preservation procedures. In some cases, each EDC contaminant has different treatment procedures. The appropriate selectivity of the treatment method that matched with EDC characteristics is essential to ensure the accuracy and efficiency of the treatment process. In practice, the hybrid treatment method through treatments combination is the most viable approach in eliminating various types of EDC. Besides, the AOP show promising performance for application in the water treatment plant. Nevertheless, the

disadvantages of other available treatment are not limiting the application for water and wastewater treatment. A continuous improvement of the limitations could extend the EDC treatment and removal implementation's performance and applicability.

Acknowledgements Data Availability Statement

No data, models, or code were generated or used during the study.

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