



Review article

Removal of endocrine disrupters from the contaminated environment: public health concerns, treatment strategies and future perspectives - A review

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ABSTRACT

Endocrine-disrupting compounds (EDCs) are emerging contaminants of concern (ECC) that disturb endocrine hormones and system functionality even at very low concentrations (i.e. $\mu\text{g/L}$ or ng/L levels). Hence, EDCs are found in all components of the environment including surface and groundwater, wastewater, soil, outdoor and indoor air and in the contaminated foods from a variety of sources (run off from agricultural activities, sewage treatment plants, leakage from septic tanks etc.), and the effects are more severe as the majority of EDCs do not have standard regulations. The environmental mobility of EDCs is higher as conventional wastewater treatment does not degrade efficiently and the development of effective and sustainable removal technologies specifically designed for the removal of those emerging micropollutants is essential. Accordingly, EDCs cause various public health diseases such as reproductive abnormalities, obesity, various cancer types, cardiovascular risks, metabolic disorders, epigenetic alterations, autism, etc. This paper reviews the existing and emerging treatment technologies for the removal of phenolic based EDCs, such as natural estrogens (estrone (E1), 17 β -estradiol (E2), estriol (E3)), synthetic estrogen 17 α -ethinylestradiol (EE2) and phenolic xenoestrogens (4-nonyl phenols (4-NP) and bisphenol-A (BPA)) from the contaminated environment. These includes advanced oxidation processes (AOP), adsorption processes, membrane based filtration, bioremediation, phytoremediation and other integrated approaches. The sustainability of EDCs removal can be assured through the use of combined processes (i.e. low-cost - biological and adsorption methods with efficient and costly - AOPs) techniques through system integration to achieve better removal efficiency than using a single treatment technique. Besides, the public health concerns and future research perspectives of EDCs are also highlighted.

1. Introduction

Endocrine disrupting compounds (EDCs) are emerging environmental micropollutants that cause serious environmental pollution and consequently affect wildlife and public health concerns due to their hormone-like behaviors (Guo et al., 2019). EDCs are defined as "agents that interfere with the synthesis, secretion, transport, binding, action, or elimination of natural hormones in the body that are responsible for the maintenance of homeostasis, reproduction, development, and/or behavior". The Endocrine Society defines EDCs as "an exogenous (non-natural) chemical, or a mixture of chemicals, that interfere with any aspect of hormone action"; while the IPCS also defines the endocrine disruptors as "as exogenous substance or mixture that alters function(s)

of the endocrine system and consequently causes adverse health effects in an intact organism, or its progeny, or (sub) populations" (IPCS, 2002). They are contaminants having xenobiotic and exogenous origins with estrogenic or androgenic activity (Gadupudi et al., 2019) that inhibit the natural/normal actions of the endocrine system in humans and animals (Li and Zhang 2014). Besides estrogenic or androgenic activity, EDCs may have other activities, such as progestagenic, anti-estrogenic, anti-androgenic activities, etc. Among the huge number of EDCs, the steroid estrogens (see Supplementary Information, Figure S1) (estrone (E1), 17 β -estradiol (E2), 17 α -ethinylestradiol (EE2), estriol (E3)) and phenolic xenoestrogens (4-nonyl phenols (4-NP) and bisphenol A (BPA)) are the most potent disruptions of the endocrine system. They originate from both anthropogenic and natural sources in a large variety of by/products

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and materials manufacturing and composition of wastewater, or are subjected to environmental and biological transformations that may form other EDCs intermediate metabolites (see section 4) (Zhang et al., 2015; Combarous 2017; Liu et al., 2019; Regkouzas and Diamadopoulou 2019; Dong et al., 2019; Guo et al., 2019). The interference of disruptor chemicals alters the functions of an organism's normal hormones and endocrine systems for two reasons (Figure 1): (i) by blocking or mimicking endogenous hormones (Jagne et al., 2016; Ahmed et al., 2018), and (ii) because of their endocrine-modulating behaviors/characteristics within the body (Dong et al., 2018). EDCs, for example, cause birth defects, neurological or developmental disorders, sexual organ malformation, adverse immune effects, breast cancer, cancerous tumors, and other acute and chronic effects even at low EDC concentrations ranging from ng/L to g/L (Jung et al., 2015; B-Moon et al., 2019).

During biological wastewater treatment, the EDCs can be transferred from the sewage to activated sludge because of their hydrophobic properties. For example, the concentration of E1, E2, EE2, E3 and BPA in waste activated sludge (WAS) was from 2–100 ng/g, 5–49 ng/g, 2–17 ng/g, 272–406 ng/g and 620 ng/g, respectively (Li and Zhang 2014). The average composition of estrogen compounds in the effluent of sewage treatment plants (STP) is from 15–70 ng/L, and is high enough to pose public health problems (Ahmed et al., 2018). Human exposure to EDCs in the environment is a life-threatening concern with unknown long-term consequences, particularly when EDC-laden sludge has been used as a fertilizer or effluent reused in agricultural practices. In addition, the principal source of these contaminants in the environment is through wastewater treatment plants (WWTP). As a result, the wastewater treatment scheme was not designed to remove these chemicals, and they are discharged alongside the effluents, causing various environmental contaminations (Kabir et al., 2015; Jung et al., 2015). EDCs have been detected as trace level organic micropollutants in the aquatic environment (such as surface waters, groundwater, marine ecosystems, industrial and municipal effluents, runoff, landfill leachates, and in natural water) (Jung et al., 2013; Tapia-Orozco et al., 2016; Dong et al., 2019; Regkouzas and Diamadopoulou 2019). The extensive occurrence, bio-accumulation, persistence of EDCs and their corresponding potential effects on human health and ecosystem functioning (Abargues et al., 2013) have created a huge global concern. Accordingly, this paper reviews the state of the art removal strategies of the main potent phenolic EDCs from the contaminated environmental sources. Besides, the recent advances, challenges and perspectives are also highlighted.

2. EDCs mechanism of actions on human health

The human endocrine system produces and secretes several endogenous hormones (such as the pituitary gland, thyroid gland, hypothalamus, ovaries and testes, adrenals and pancreas glands) (Combarous 2017). Endogenous hormones operate as a chemical signal to control biological functions (Khetan, 2014), to communicate with our body tissues, and to regulate development, reproduction and growth (Combarous, 2017). Endocrine disruptors are anthropogenic and natural agents with diverse chemical structures (Supplementary Information, Figure S1) that interfere with endogenous hormones and pose a risk to public health. EDCs alter hormone balance through a variety of mechanisms, including disrupting hormone synthesis/breakdown, mimicking hormones, altering hormone binding or acting as hormone antagonists, and altering the development of hormone receptors, resulting in a variety of disorders and public health issues (Combarous and Nguyen, 2019). This includes the increasing prevalence of "hormone-sensitive cancers", reproductive and developmental abnormalities, alterations in the immune system functions, children's abnormal growth patterns and neurodevelopmental delays (Kumar et al., 2020). The EDCs target nuclear receptors (i.e. "hormone dependent transcription factors that exert long-term control of their target phenotype cells"), whereas membrane receptor signaling can also be affected by the EDCs, resulting in short-term effects due to their signaling pathways exerting more acute

effects on the target cells. Table 1 and Figure 1 illustrate the main mechanisms of action (Mechanism 1–9) that EDCs can exhibit. Mechanism 1 is shared by all hormone-related mechanisms of action, whereas mechanisms 2–9 show an imbalance in endocrine homeostasis and are not related to the direct hormonal type mechanisms of action by EDCs (Combarous and Nguyen, 2019).

3. Removal techniques

3.1. Adsorption techniques

The adsorption process is the most effective and low-cost removal technique (B-Moon et al., 2019). However, searching and the development of novel adsorbents has been a thriving research area in the advancement of adsorption science and technology. The activated carbon (AC) adsorption process has been applied to removing several EDCs, mainly due to numerous advantages, such as high surface area and pore structure, the possibility of regeneration and reuse, low capital cost, applicability to removing them at very low concentrations, suitability for batch and continuous systems, hydrophobic nature of surface properties, easily enhanced by various activation techniques (Delgado et al., 2012). The biochar produced by the pyrolysis of biomass in an oxygen-limited environment (Regkouzas and Diamadopoulou 2019; Liu et al., 2019) has gained increasing attention for the removal of EDCs (Jung et al., 2013; Kim et al., 2016; B-Moon et al., 2019), because of its low cost, being environmentally benign and easy availability, the surface density of functional group chemistry, higher condensed structure, volume properties and others (Jung et al., 2013; Dong et al., 2019). Besides, the biochar system plays a significant role in the adsorption capacity and removal mechanism due to the existence of "surface oxygen-containing functional groups" such as carboxyl, lactone, quinone, lactol and their corresponding level of concentrations (Delgado et al., 2012; Jung et al., 2013; Kim et al., 2016; B-Moon et al., 2019).

Kamarehie et al. (2018) achieved an adsorption capacity of BPA of 9.2 mg/g and 22.28 mg/g with powdered activated carbon (PAC) and PAC coated with MgO, respectively. Table 2 shows the advantages and limitations of various EDCs removal technologies, while Table 3 presents the removal efficiencies of phenolic-based EDCs using various techniques. The adsorption capability of adsorbents can be improved by using different surface modifying agents (Kamarehie et al., 2018; B-Moon et al., 2019). Guo et al. (2019) immobilized EE2 (2 mg/L initial concentration) in a biochar derived from corn straw and achieved 1.148 $\mu\text{g/g}$ of EE2 q_e from the contaminated sediment. Biochar made of sewage sludge has been utilized and removed 67–99% of mixed EDCs from the water table, while a 35–97% range of removal efficiency was achieved from environmental wastewater samples (Regkouzas and Diamadopoulou 2019). The electrostatic interactions and synergetic effects of hydrophobicity play an important role in explaining the adsorption mechanism in the pore-filling processes (Regkouzas and Diamadopoulou 2019; B-Moon et al., 2019). Following KOH activation, Liu et al. (2020) achieved 100.6 mg/g of q_e for E2 adsorption using a biochar made of lotus seedpod (LSP). The enhancement of adsorption capability, magnetic sensitivity and fast magnetic separation of E2 was investigated by modified biochar using magnetic nanoparticles (Mag-BCNPs) and achieved 50.24 mg/g of q_e from the polluted water (Dong et al., 2018). The removal efficiency of E2 decreases as its ionic strength increases, with possible adsorption mechanisms including π -interactions, chemisorption, electrostatic interactions, and monolayer adsorption (Dong et al., 2018; Liu et al., 2020). Dong et al. (2019) also investigated achieving an 85% degradation efficiency of 4-nonylphenol (4-NP) using Fe_3O_4 modified bamboo biochar (BBC) for the treatment of real river sediment. The Fe_3O_4 -BB has a low cytotoxic potency and is suitable for contaminated sediment remediation. Ahmed et al. (2018) also achieved 100% removal efficiency of E1, E2, E3, EE2, and BPA using functionalized biochar (fBC) with initial concentrations of 500 g/L and doses of 400 mg/L, where π -interactions and hydrogen bonding play a role in the sorption mechanism. The

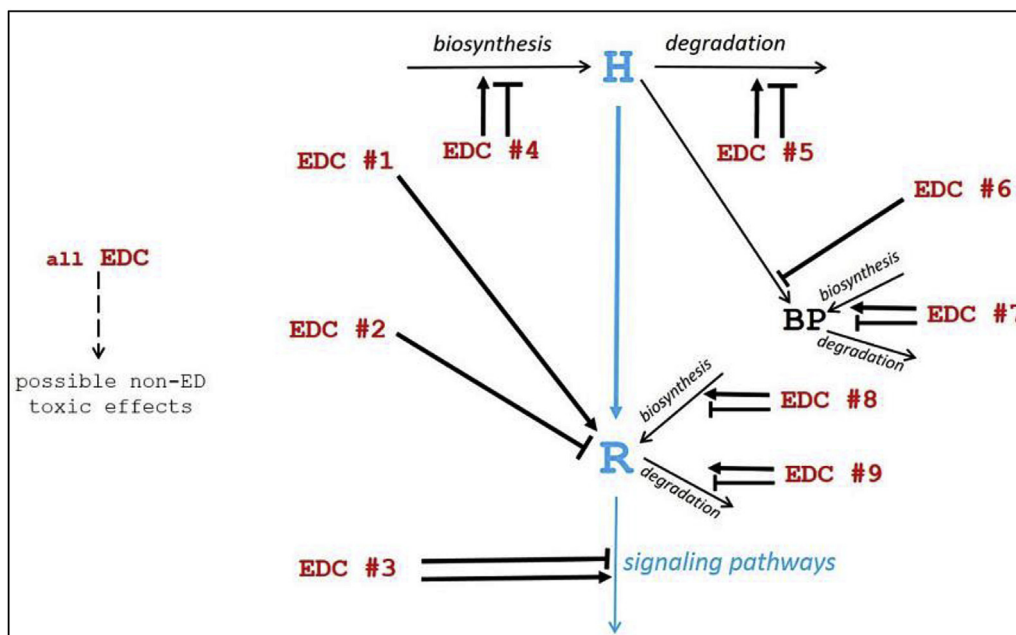


Figure 1. Schematics showing the EDCs mechanism of action on human health: Blue – physiological hormonal mechanisms, Red – diverse EDCs mechanisms of action (EDC 1–9) shown by the black arrows directing to their site of action (→ simulation and ⊥inhibition) (Combarnous and Nguyen, 2019).

enhanced removal of BPA was also carried out using biochar supported magnetic CuZnFe₂O₄ composite (CZF–biochar) and achieved a higher *q_e* of 263 mg/g (Heo et al., 2019) with charge-assisted hydrogen bonding, π-π electron donor-acceptor interactions and hydrophobicity governed

the adsorption of BPA from the simulated wastewater. Hence, the CZF-biochar could be a benign, efficient and stable adsorbent for the removal of BPA and other emerging EDCs (Heo et al., 2019). From other perspectives, a metal-organic framework (MOFs) as an adsorbent has

Table 1. EDCs mechanisms of action on human health (adapted from Combarnous and Nguyen, 2019).

Type of mechanism	EDCs mechanisms of action in human health
Mechanism 1: the EDCs directly exhibiting hormonal activity through the receptor binding that leads to the activation of its signaling pathways.	<ul style="list-style-type: none"> - Most EDCs exhibit a hormone with this type of mechanism of action (i.e. able to bind to and activate a hormone receptor. EDCs have structures that differ from those of hormones, allowing them to enter their binding sites and interfere with their mechanisms of action. - EDCs can act as hormones by directly interacting with and activating hormone receptors (HR), and they may be less efficiently degraded than natural hormones, but they are more active in vivo due to their longer half-life in blood cells.
Mechanism 2: EDCs bind to the HR, leading to directly inhibiting the endogenous hormone action through receptor occupation of its signaling pathways.	<ul style="list-style-type: none"> - The new molecules resemble hormones and are able to bind to receptors that lead to 1) freezing the receptors' conformation in their inactive state, and 2) thus antagonizing the endogenous hormone actions, and the exogenous molecules can clearly exert endocrine disruption. The EDCs are able to bind to the receptors as hormones by exerting an antagonistic effect in contrast to the hormones.
Mechanism 3: EDCs interact with the components of the hormone signaling pathways downstream of the receptor.	<ul style="list-style-type: none"> - The EDCs do not interfere with the HR at several possible sites downstream of them, which can be difficult to identify. This mechanism can lead to both direct, non-endocrine, and toxic effects.
Mechanism 4: Stimulation	<ul style="list-style-type: none"> - Many EDCs exert endocrine disruptions not by interfering directly with the HR, but by affecting, negatively or positively, endogenous hormone biosynthesis (Mechanism 4) or degradation (Mechanism 5). - These EDCs have structures that differ from those of hormones, and they do not compete with hormones at the receptor level.
Mechanism 5: Inhibition of endogenous hormone biosynthesis	<ul style="list-style-type: none"> - The EDCs do not interfere with the hormone receptors but, by affecting the endogenous hormone concentration, impact on either their degradation or biosynthesis.
Mechanism 6: Binding to the circulating hormone binding protein	<ul style="list-style-type: none"> - The hydrophobic EDCs compete with small hydrophobic hormones (such as thyroids and steroids) for these transport proteins in the blood. - Hence, the EDCs directly interfere with hormone-binding transport proteins, thereby competing with the endogenous hormone concentration in the blood and exerting their effect through this mechanism. This means that the EDCs do not compete with the hormones at the receptor level, but at the level of circulating binding proteins.
Mechanism 7: Stimulation or inhibition of hormone-binding protein degradation or synthesis.	<ul style="list-style-type: none"> - The EDCs have an effect on the biosynthesis/degradation of hormone-binding transport proteins, which may affect both the "total hormone concentration"/or "and its associated free active fraction." - In this way, the EDCs exhibit the chemical structure that is different from those of hormones. Then, the hormone-binding transport proteins are often degraded or synthesized by the liver, which, as a degrading organ, is the main target of toxicants.
Mechanism 8: Simulation	<ul style="list-style-type: none"> - The simulation of endogenous HR is a way in which a number of EDCs interfere with endocrine homeostasis.
Mechanism 9: Inhibition of HR expression	<ul style="list-style-type: none"> - The inhibition of HR expression is also a mechanism responsible for the alteration of EDCs of the endocrine system. In this mechanism, the EDCs do not need to resemble hormones to exert their adverse effect by the availability of a modifying receptor, whereas the synthesis or/and degradation of the receptor is frequently controlled by its cognate hormones. - The structural similarity of EDCs with hormones could be responsible for such type of effect.

Table 2. Advantages and limitations of various EDCs removal technologies (Liu et al., 2019; Jung et al., 2013; Si et al., 2019).

Technology	Advantages	Limitations
Adsorption process	<ul style="list-style-type: none"> - Regeneration/reuse possibility at a lower capital cost - Having higher surface area and pore structure as well as hydrophobic surface properties - Robust, simple to install and maintain - Thermal regeneration of the carbon to destroy the adsorbed solute waste - Highly flexible, allows the rapid start-up and shutdown when enquired. 	<ul style="list-style-type: none"> - The pollutants are removed from water, but unable to destroy (i.e. regeneration difficulties). - The system did not tolerate suspended solids (SS) in the effluent stream due to clogging. - The disposal of sludge loaded is challenging, if there is no possibility to regenerate. - Development of a novel adsorbents is a prosperous area of research
Ultrafiltration (UF)	<ul style="list-style-type: none"> - Consumes lower energy and constitutes a cost effective method of treatment - No chemicals used in the treatment processes and no environmental pollution caused due to effluents discharge. - Requires lower operational pressure 	<ul style="list-style-type: none"> - Possible to remove EDCs at lower operated pressure membrane process - Inadequate water quality of the effluents - Unable to filter soluble materials from the water - Causes membrane damage at > 3 bar pressure
Nanofiltration (NF)	<ul style="list-style-type: none"> - Its physical nature of separation and the membrane modular design makes NF makes an alternative separation technique. - The process is free from chemicals used (i.e. environmentally benign) 	<ul style="list-style-type: none"> - Higher energy consumption (0.3–1.0 kWh/m³) - Higher installation cost - More expensive than RO membranes - Pretreatment (i.e. 0.1–20 microns) is needed for highly polluted water
Microfiltration (MF)	<ul style="list-style-type: none"> - No energy consuming phase transfer is required (for evaporation, etc.). - Requires lower operational pressure - Relatively cheap 	<ul style="list-style-type: none"> - Removal at lower pressure is limited because of its large pore size. - Causes membrane damage at >1 bar of pressure. - Inadequate quality of treated wastewater
Bioremediation techniques	<ul style="list-style-type: none"> - Solely natural process with no harmful side effects - No dangerous transport within the <i>in situ</i> treatment - Consumes little energy than incineration and landfilling techniques 	<ul style="list-style-type: none"> - Lower biodegradability for micropollutants and chlorinated compounds - Microbial metabolism of contaminants may generate toxic intermediate metabolites.
Phytoremediation techniques	<ul style="list-style-type: none"> - Cost effective technology (it does not require specialized equipment and costly biosorbents) - Apply at onsite to remediate contaminated shallow soil, surface and groundwater - Does no cause environmental impacts (improved soil ecosystem) - Used in large scale environmental apartment operations - Used for treatment of mixed pollutants 	<ul style="list-style-type: none"> - The generated intermediate may cause plant cytotoxicity - Harvesting and disposal of the accumulated pollutants requires regulatory standards - Requires large area (land) than other remedial methods - Achievement of the phytoremediation can be influenced by the tolerance of the plants species used to treat various micropollutants
Membrane filtration	<ul style="list-style-type: none"> - The process saves energy consumption - Possible to couple with other operations and processes - Produces higher quality products with variable operational parameters - No chemicals and additives used, therefore, the process is environmentally friendly. 	<ul style="list-style-type: none"> - Concentrated the produced sludge - Influenced by the physical/chemical properties of the pollutants to be treated - Causing fouling problems - Low selectivity and short membrane life-time
Advanced Oxidation	<ul style="list-style-type: none"> - Can be used as pre/post treatment of biological system - Suited for higher COD industrial pollutants - Contributes for 50% sludge reduction and provides a complete mineralization of pollutants into CO₂ and H₂O. - No further treatments are required and consumes less energy supply - Does not generate large amounts of hazardous sludge 	<ul style="list-style-type: none"> - The complete degradation of pollutants into non-hazardous compounds are not cost effective - Comparatively higher operational, capital and maintenance costs - Process limitations can be influenced by aggregation of particles, pH changes and modification of surface characteristics of the heterogeneous catalysts
Reverse Osmosis (RO)	<ul style="list-style-type: none"> - Provide for removal of all mineral salts and chemical auxiliaries - Common in wastewater treatment for the removal of organic micropollutants 	<ul style="list-style-type: none"> - The removal rate depends on the magnitude of the EDCs and properties of the membrane - Requires higher pressure

been developed and achieved a 138.4 mg/g and 200.4 mg/g adsorption capacity (q_e) of BPA and EE2, respectively (B-Moon et al., 2019). From these studies, it could be possible to conclude that the EDCs adsorption processes are robust, simple and efficiently employed in large scale applications without forming an intermediate byproduct in the environment. The EDC adsorption performance of other adsorbents is summarized in Table 3.

3.2. Membrane based filtrations

Membrane filtration technologies such as nano-filtration (NF), ultra-filtration (UF) and reverse osmosis (RO) are a promising wastewater treatment for removing various micropollutants (Bolong et al., 2010; Escalona et al., 2014) due to their lower energy requirement. The large pore size of the UF membrane limits the effective removal of EDCs, while NF is popular because of its effective physical nature of separation and membrane design. Numerous studies have shown the effectiveness of

EDC removal by RO, NF and UF supported membrane filtration (Cases et al., 2011; Yüksel et al., 2013; Si et al., 2019). Si et al. (2019) investigated the effect of effluent organic matter (EfOMs), humic acid (HA), sodium salt (NaAH), bovine serum albumin (BSA) using ozonation and UF-MBR were investigated in the secondary treatment effluent. Over 90% cumulative removal efficiency of all EDCs (BPA, E1, E2, EE2 and E3) was achieved. The removal of BPA, alkylphenol compounds (4OP and 4tOP) and nonylphenols (NP) in combined conventional activated sludge with tertiary treatments (CAS-TT), and two membrane bioreactor pilot plants installed with flat sheet (MBR-FS) and hollow fiber membrane (MBR-HF) modules were studied by Cases et al. (2011). The MBR or CAS-TT demonstrated efficient removal of 4tOP and BPA, while the MBR-HF demonstrated adequate removal efficiency of NP. The removal efficiency of EDCs in all treatment schemes is illustrated in Table 3. In another study, the removal efficiency of BPA from a simulated wastewater (50 mg/L) was $\geq 98\%$, 10–40% and 80%, respectively, with polyamide and cellulose acetate based reverse osmoses (PARO and

Table 3. Removal efficiencies of phenolic based EDCs from the contaminated environment.

No.	Type of EDC	Source	Techniques/Process	Conditions	RE	qe	Reference	
1	Bisphenol A (BPA)	X	Adsorption using Al -organic based process	2 mg/L concentration, 313 K	-	70.2	B-Moon et al. (2019)	
		Water	Photo-Fenton process	4.4×10^{-5} mol/L concentration	59	-	Jiang et al. (2014)	
		WAS	UV/H ₂ O ₂ oxidation processes	pH 3, UV fluence rate 0.069 Mw/cm ² , H ₂ O ₂ dosage 0.5 mol/L, time 2 min	89	-	Zhang and Li (2014)	
		Municipal wastewater	Freshwater green alga (<i>Nannochloris sp.</i>)	7 d of incubation through ultrafiltration effluent using <i>Nannochloris sp.</i>	46	-	Bai and Acharya (2019)	
		Municipal wastewater	Conventional activated sludge processes		Flow rate 59, 010 m ³ /d, MLSS 8 gss/L with a capacity of 75,000 m ³ /d	96		Cases et al. (2011)
				Flat sheet (MBR-FS)	Flow rate 4.8 m ³ /d, MLSS 8 gss/L with 20 flat sheet (MBR-FS), area 16 m, flow rate 400 L/h.	92		Cases et al. (2011)
				Hollow fibre (MBR-HF) modules	Flow rate 4.8 m ³ /d, MLSS 8 gss/L with total area of 20 m ² and a flow rate of 400 L/h	97		Cases et al. (2011)
		X		Photo Fenton process	UV between 100 λ <math>< 280</math> nm, pH 3, 2.7 mmol/L Fe(II) and 5 mmol/L H ₂ O ₂ after 50 min	100		Jamil et al. (2017)
		X	PAC		20–100 mg/L of BPA, PAC (2–6 g), pH (3–11) and contact time (10–60 min)		9.2	Kamarehie et al. (2018)
				MgO-PAC crystals	20–100 mg/L of BPA, MgO-PAC (2–6 g), pH (3–11) and contact time (10–60 min)		22.28	Kamarehie et al. (2018)
		X		CuZnFe ₂ O ₄ composite (CZF–biochar)	250 mL of BPA and SMX solutions, adsorbents dose 0.05 g of CZF, temp. 25 °C, time 24 h	-	263	Heo et al. (2019)
		X		Biochar composite	250 mL of BPA and SMX solutions, adsorbents dose 0.05 g of CZF, temp. 25 °C, time 24 h	-	185	Heo et al. (2019)
		Secondary effluent		EfOM supported Ultrafiltration and ozonation	20 mg/L of sodium alginate (NaAg) with 100 µg/L of EDC concentration	90	-	Si et al. (2019)
		Tap water		Pilot-scale SPBB using <i>P. ostreatus</i>	60 mg/L of each EDC concentration within 2 h incubation, pH 6.5 flow of 0.3 L/h	78	-	Kresinová et al. (2018)
		X		Pilot-scale TBR using <i>P. ostreatus</i>	10 d, 200 mg/L TRC concentration	76	-	Kresinová et al. (2018)
		Urban wastewater		Pilot-scale SPBB using <i>P. ostreatus</i>	After 24 h and 60 mg/L of each EDC concentration	97	-	Kresinová et al. (2018)
		Water		Adsorption on cellulose fiber	30 mg/L of BPA and 1 g of fiber in 100 mL of water, pH 5, 20 °C temperature, 5 min contact time	70	-	Tursi et al. (2018)
		Domestic Wastewater		Constructed Wetlands	25 mg/L BPA concentration in Heliconia-CW	73	-	Toro-Vélez et al. (2017)
		WWTP		Nitrifying activated sludge (NAS)	Batch experiments with 1 mg/L BPA concentration and at 96 h contact time	100	-	Kassotaki et al. (2019)
		WWTP		Ammonia oxidizing bacteria (AOB)	15 mg/L BPA concentration, 2 h batch experiments	39	-	Kassotaki et al. (2019)
		X		enzyme polymerization using NF membranes	180 min, pH 7, MR contained 20 mL of 20 mg/L of BPA	95	-	Escalona et al. (2014)
		Wastewater		functionalized biochar (fBC)	BPA concentration 500 µg/L, 110 rpm, 25 °C for 48 h at pH 3.25, 400 mg/L dose of fBC	100	-	Ahmed et al. (2018)
		X		Polyamide RO membranes	Model solution of 50 mg/L at 10 bar pressure	≥98	-	Yüksel et al. (2013)
X		BWRO membrane	Model solution of 50 mg/L at 10 bar pressure	40	-	Yüksel et al. (2013)		
X		NF membranes	Model solution of 50 mg/L at 10 bar pressure, 6 h	98	-	Yüksel et al. (2013)		
AMBR effluent		Microalgae culture reactor	Aerated experiments after the first 22.5 h	91	-	Abarguesa et al. (2013)		

(continued on next page)

Table 3 (continued)

No.	Type of EDC	Source	Techniques/Process	Conditions	RE	qe	Reference
		AMBR effluent	Microalgae culture reactor	The non-aerated experiment after 22.5 h	80	-	Abarguesa et al. (2013)
		WAS	Calcium peroxide (CaO ₂) oxidation	At neutral pH with CaO ₂ dosage 0.34 g/g TS, EDC 69 mg/g TS BPA concentration	99	-	Zhang et al. (2015)
2	17 α -ethynylestradiol (EE2)	X	Al-based metal-organic	2.0 mg/L BPA concentration and 313 K tem	-	87.0	B-Moon et al. (2019)
		WAS	UV/H ₂ O ₂ oxidation processes	pH 3, UV irradiation rate 0.069 Mw/cm ² , H ₂ O ₂ dosage 0.5 mol/L, 2 min time	95	-	Zhang and Li (2014)
		X	Photo-Fenton process	8.5 mg/L H ₂ O ₂ under irradiation of 0.1 g catalyst and 8.5 mg/L H ₂ O ₂ with UV light at 60 min	98	-	Baycan and Puma (2018)
		MWWE	Freshwater green alga (<i>Nannochloris sp.</i>)	7 d of incubation through ultrafiltration effluent using <i>Nannochloris sp.</i>	60	-	Bai and Acharya (2019)
		Sediment –water interface	Adsorption on corn straw biochar	48 h, 150 rpm, temp 25 °C, pH 7, with initial PFOs and EE2 concentrations of 2 mg/L, pore volume 0.201 cm ³ /g	-	1,148 μ g/g	Guo et al. (2019)
		Tap water	Pilot-scale SPBB using <i>P. ostreatus</i>	60 mg/L of EE2 concentration, 2 h incubation, pH 6.5 flow of 0.3 L/h	78	-	Kresinová et al. (2018)
		X	Pilot-scale TBR using <i>P. ostreatus</i>	10 d, 200 mg/L TRC concentration	76	-	Kresinová et al. (2018)
		Urban wastewater	Pilot-scale SPBB using <i>P. ostreatus</i>	After 24 h and 60 mg/L EE2 concentration	97	-	Kresinová et al. (2018)
		X	ultrafiltration and ozonation	In sodium alginate (NaAg) 20 mg/L and 100 μ g/L EE2 concentration	90	-	Si et al. (2019)
		X	Algae pond	9.71 ng/l of EE2 concentration with 128 mg/L algae in TSS, 90 strokes per min for 3 h	86.8	-	Shi et al. (2010)
		X	Duckweed pond	9.71 ng/L of concentration seeded with 5,000 mg fresh duckweed (TSS about 380 mg), 90 strokes per min for over 3 h.	93.9	-	Shi et al. (2010)
		WWTP	Nitrifying activated sludge (NAS)	At 1 mg/L on initial concentration in batch experiments with 96 h contact time	100	-	Kassotaki et al. (2019)
		WWTP	Ammonia oxidizing bacteria (AOB)	15 mg/L EE2 concentration, 2 h batch experiments	34	-	Kassotaki et al. (2019)
		WAS	Fenton oxidation	H ₂ O ₂ dosage 15.62 mmol/g, pH 3, time 60 min, Fe(II) to H ₂ O ₂ molar ratio of 0.167	84	-	Li and Zhang (2014)
		WAS	Calcium peroxide (CaO ₂) oxidation	At neutral pH and CaO ₂ dosage of 0.34 g/g TS, EDC with 69 mg/g TS concentration	94	-	Zhang et al. (2015)
Wastewater	oH ₃ PO ₄ activated	500 μ g/L of EE2 concentration, 110 rpm, 25 °C for 48 h at pH 3.25, 400 mg/L dose of fBC	100	-	Ahmed et al. (2018)		
X	Photo oxidation process	8.5 mg/L H ₂ O ₂ under irradiation of 0.1 g catalyst and 8.5 mg/L H ₂ O ₂ with UV light at 60 min	95	-	Baycan and Puma (2018)		
3	Estrone (E1)	X	Ultrafiltration (UF) and ozonation	20 mg/L of sodium alginate (NaAg) and 100 μ g/L E1 concentrations	90	-	Si et al. (2019)
		MWWTP	Conventional activated sludge (CAS) processes	Flow rate 59, 010 m ³ /d, MLSS 8 gss/L with a capacity of 75,000 m ³ /d and treats wastewater of 420,000 inhabitants	99	-	Cases et al. (2011)
		MWWTP	Flat sheet (MBR-FS)	Flow rate 4.8 m ³ /d, MLSS 8 gss/L with 20 flat sheet (MBR-FS), area of 16 m ² and flow rate of 400 L/h.	99	-	Cases et al. (2011)
		MWWTP	Hollow fibre (MBR-HF) modules	Flow rate 4.8 m ³ /d, MLSS 8 gss/L with total area of 20 m ² and a flow rate of 400 L/h	99	-	Cases et al. (2011)
		MWWE	Freshwater green alga (<i>Nannochloris sp.</i>)	7 d of incubation through ultrafiltration effluent using <i>Nannochloris sp.</i>	29	-	Bai and Acharya (2019)

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Table 3 (continued)

No.	Type of EDC	Source	Techniques/Process	Conditions	RE	qe	Reference
		WAS	UV/H ₂ O ₂ oxidation processes	pH 3, UV wavelength 253.7 nm, UV fluence rate 0.069 Mw/cm ² , H ₂ O ₂ dosage 0.5 mol/L, 2 min time	97	-	Zhang and Li (2014)
		Tap water	Pilot-scale SPBB using <i>P. ostreatus</i>	60 mg/L E1 concentration, 2 h incubation, pH 6.5, flow rate of 0.3 L/h	78	-	Kresinová et al. (2018)
		X	Pilot-scale TBR using <i>P. ostreatus</i>	10 d, 200 mg/L TRC concentration	76	-	Kresinová et al. (2018)
		Urban wastewater	Pilot-scale SPBB using <i>P. ostreatus</i>	After 24 h and 60 mg/L of each EDC concentration	97	-	Kresinová et al. (2018)
		X	Algae pond	43.5 ng/L of E1 concentration, 128 mg/L algae in TSS, 90 strokes per min for 3 h	83.9	-	Shi et al. (2010)
		X	Duckweed pond	43.5 ng/L of concentration, 5,000 mg fresh duckweed (TSS about 380 mg), 90 strokes per min for over 3 h.	94.4	-	Shi et al. (2010)
		Wastewater	functionalized biochar (fBC)	460.5 µg/L E1 concentration, 110 rpm, 25 °C for 48 h at pH 3.25, and for 400 mg/L dose of fBC	100	-	Ahmed et al. (2018)
		WWTE	Nitrifying activated sludge (NAS)	15 mg/L E1 concentration, 2 h batch experiments	100	-	Kassotaki et al. (2019)
		WWTE	Ammonia oxidizing bacteria (AOB)	15 mg/L E1 concentration, 2 h batch experiments and at 18.2 mg/L/h of acetate consumption	38	-	Kassotaki et al. (2019)
		WAS	Fenton oxidation	H ₂ O ₂ dosage 15.62 mmol/g, pH 3, time 60 min, Fe(II) to H ₂ O ₂ molar ratio 0.167	70	-	Li and Zhang (2014)
		WAS	Calcium peroxide (CaO ₂) oxidation	At neutral pH and CaO ₂ dosage of 0.34 g/g TS, E1 concentration of 69 mg/g TS	92	-	Zhang et al., (2015)
		X	Biochar	At 25 °C, 200 rpm, 30 µL of magnetic biochar nanoparticle with 200 mg/mL influent, pH 4	-	50.24	Dong et al. (2018)
4	17β-estradiol (E2)	X	Adsorption KOH treated lotus seedpod biochar	7 mg/L E2 concentration, 27 °C temp, 20 h time, pH 10.0, adsorbent dose 3 mg, 160 r/min	-	100.6	Liu et al. (2020)
		X	Graphene oxide based biochar	6 mg/L E2 concentration, 298 K and pH 7.0.	-	46.22	Liu et al. (2019)
		X	Photo-Fenton process	8.5 mg/L H ₂ O ₂ under irradiation of 0.1 g catalyst and 8.5 mg/L H ₂ O ₂ with UV light at 60 min	92	-	Baycan and Puma (2018)
		WAS	Calcium peroxide (CaO ₂) oxidation	At neutral pH and CaO ₂ dosage of 0.34 g/g TS, 69 mg/g TS E2 concentration	100	-	Zhang et al. (2015)
		WAS	UV/H ₂ O ₂ oxidation processes	pH 3, UV wavelength 253.7 nm, UV fluence rate 0.069 Mw/cm ² , H ₂ O ₂ dosage 0.5 mol/L, 2 min time	92	-	Zhang and Li (2014)
		X	Algae pond	29.7 ng/L of E2 concentration, 128 mg/L algae in TSS, 90 strokes per min for 3 h	91.2	-	Shi et al. (2010)
		X	Duckweed pond	29.7 ng/L of concentration, seeded with 5,000 mg fresh duckweed (TSS about 380 mg), 90 strokes per min for over 3 h.	95.4	-	Shi et al. (2010)
		Tap water	Pilot-scale SPBB using <i>P. ostreatus</i>	60 mg/L E2 concentration, 2 h incubation, pH 6.5 flow of 0.3 L/h	78	-	Kresinová et al. (2018)
		X	Pilot-scale TBR using <i>P. ostreatus</i>	10 d, 200 mg/L E2 concentration	76	-	Kresinová et al. (2018)
		Urban wastewater	Pilot-scale SPBB using <i>P. ostreatus</i>	24 h and 60 mg/L of E2 concentration	97	-	Kresinová et al. (2018)
		X	ultrafiltration and ozonation	In sodium alginate (NaAg) 20 mg/L and 100 µg/L E2 concentration	90%	-	Si et al. (2019)
		MWWE	Freshwater green alga (<i>Nannochloris sp.</i>)	7 d of incubation from the algal culture in UF effluent using <i>Nannochloris sp.</i>	60	-	Bai and Acharya (2019)

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Table 3 (continued)

No.	Type of EDC	Source	Techniques/Process	Conditions	RE	qe	Reference
		WWTE	Nitrifying activated sludge (NAS)	At 1 mg/L on E2 concentration, 96 h contact time	100	-	Kassotaki et al. (2019)
		WWTE	Ammonia oxidizing bacteria (AOB)	15 mg/L E2 concentration, 2 h batch experiments	100	-	Kassotaki et al. (2019)
		WAS	oH ₃ PO ₄ activated biochar	500 µg/L E2 concentration, 110 rpm, 25 °C for 48 h at pH 3.25, 400 mg/L dose of fBC	100	-	Ahmed et al. (2018)
		WAS	Fenton oxidation	H ₂ O ₂ dosage, 15.62 mmol/g, initial pH 3, time 60 min, Fe(II) to H ₂ O ₂ molar ratio 0.167	90	-	Li and Zhang (2014)
		WWTE	Nitrifying activated sludge (NAS)	At 1 mg/L E2 concentration, 96 h contact time	100	-	Kassotaki et al. (2019)
		WWTE	Ammonia oxidizing bacteria (AOB)	15 mg/L E2 concentration, 2 h batch experiments	78	-	Kassotaki et al. (2019)
5	Estriol (E3)	Tap water	Pilot-scale SPBB using <i>P. ostreatus</i>	60 mg/L E3 concentration, 2 h incubation, pH 6.5 flow of 0.3 L/h	78	-	Kresinová et al. (2018)
		X	Pilot-scale TBR using <i>P. ostreatus</i>	10 d, 200 mg/L E3 concentration	76	-	Kresinová et al. (2018)
		Wastewater	functionalized biochar (fBC)	500 µg/L E3 concentration, 110 rpm, 25 °C for 48 h at pH 3.25, 400 mg/L dose of fBC	97	-	Ahmed et al. (2018)
		Urban wastewater	Pilot-scale SPBB using <i>P. ostreatus</i>	24 h and 60 mg/L of E3 concentrations	97	-	Kresinová et al. (2018)
		X	Ultrafiltration and ozonation	In sodium alginate (NaAg) 20 mg/L and 100 µg/L E3 concentration	90%	-	Si et al. (2019)
		WAS	Fenton oxidation	H ₂ O ₂ dosage 15.62 mmol/g, initial pH 3, time 60 min, Fe(II) to H ₂ O ₂ molar ratio 0.167	98	-	Li and Zhang (2014)
		WAS	Calcium peroxide (CaO ₂) oxidation	At neutral pH and CaO ₂ dosage of 0.34 g/g TS, 69 mg/g TS E3 concentration	93	-	Zhang et al. (2015)
		WAS	UV/H ₂ O ₂ oxidation processes	pH 3, UV wavelength 253.7 nm, UV fluence rate 0.069 Mw/cm ² , H ₂ O ₂ dosage 0.5 mol/L, 2 min time	94	-	Zhang and Li (2014)
		X	Photo-Fenton process	8.5 mg/L H ₂ O ₂ under irradiation of 0.1 g catalyst and with UV light at 60 min	70%	-	Baycan and Puma (2018)
		Water	Fungus <i>Umbelopsis isabellina</i>	12 h of incubation, 25 mg/L E3 concentration	90	-	Janicki et al. (2016)
6	Nonylphenols (NP)	Wastewater	<i>U. isabellina</i> fungus	25 mg/L of NP and 10% of fungal culture	15.2	-	Janicki et al. (2018)
		Domestic Wastewater	Constructed Wetlands	25 mg/L NP concentration in Heliconia-CW	63	-	Toro-Vélez et al. (2017)
		Tap water	Pilot-scale SPBB using <i>P. ostreatus</i>	60 mg/L of NP concentration within 2 h incubation, pH 6.5 flow of 0.3 L/h	78	-	Kresinová et al. (2018)
		X	Pilot-scale TBR using <i>P. ostreatus</i>	10 d, 200 mg/L NP concentration	76	-	Kresinová et al. (2018)
7	4-n-nonylphenol (4-NP)	Urban wastewater	Pilot-scale SPBB using <i>P. ostreatus</i>	24 h and 60 mg/L of 4-NP concentration	97	-	Kresinová et al. (2018)
		X	UV/H ₂ O ₂ oxidation processes	At pH 3, UV fluence rate 0.069 Mw/cm ² , H ₂ O ₂ dosage 0.5 mol/L, 2 min time	67	-	Zhang and Li (2014)
		River marine sediments	Biochars	pH 3, dosage of 3.33 g/L Fe ₃ O ₄ -bamboo biochar (BB) and 2.3 × 10 ⁻⁵ M persulfate (PS) in a biochar-sediment system.	85	-	Dong et al. (2019)
		AMBR effluent	Microalgae culture reactor	Aerated experiments after the first 22.5 h	91	-	Abargues et al. (2013)
		AMBR effluent	Microalgae culture reactor	For non-aerated conditions	100	-	Abargues et al. (2013)
8	technical-nonylphenol (t-NP)	X	Adsorption on α-cellulose with KOH activation	120 min time, 150 mg/L of t-NP concentration	82.8	1072.9	He et al. (2016)
		AMBR effluent	Microalgae culture reactor	Aerated experiments after the first 22.5 h	91	-	Abargues et al. (2013)

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Table 3 (continued)

No.	Type of EDC	Source	Techniques/Process	Conditions	RE	q _e	Reference
		AMBR effluent	Microalgae culture reactor	For non-aerated conditions	80	-	Abarquesa et al. (2013)
		MWWE	Freshwater green alga (<i>Nannochloris sp.</i>)	7 d of incubation from the algal culture in UF effluent using <i>Nannochloris sp.</i>	100	-	Bai and Acharya (2019)
11	4-tert-octylphenol (4-t-OP)	Wastewater	oH ₃ PO ₄ activated	500 µg/L concentration, 110 rpm, 25 °C for 48 h at pH 3.25, 400 mg/L dose of fBC	97	-	Ahmed et al. (2018)
12	4-cumylphenol (4-CP)	Wastewater	<i>U. isabellina</i> fungus	25 mg/L of concentration and 10% of fungal cultures	93.1	-	Janicki et al. (2018)
13	4-tert-butylphenol (4tBP)	Wastewater	<i>U. isabellina</i> fungus	25 mg/L concentration and 10% of fungal culture	65.3	-	Janicki et al. (2018)
14	4-cumylphenol (4-CP)	AMBR effluent	Microalgae culture reactor	Aerated experiments after the first 22.5 h	91	-	Abarquesa et al. (2013)
15	4-tert-octylphenol (4-t-OP)	AMBR effluent	Microalgae culture reactor	For non-aerated conditions with 22.5 h	80	-	Abarquesa et al. (2013)

Note: X - synthetic wastewater, q_e - equilibrium adsorption capacity (mg/g), RE-removal efficiency, SPBB-static packed-bed bioreactor, TBR - trickle-bed reactor, BWRO - Brackish water reverse osmosis, PAC - Powdered activated carbon, RO - reverse osmosis, NF - nanofiltration, AMBR - Anaerobic membrane bioreactor, MWWT - municipal wastewater treatment plant, WWTE - Wastewater treatment effluent, WAS - Waste activated sludge, ROC - Reverse osmosis concentrate, PPCPW - Pharmaceutical and personal care products wastewater.

CARO) and NF membranes (Yüksel et al., 2013). This confirms that the PARO membrane exhibited better removal performance of BPA than the CARO membrane. In addition, over 90% and 70% BPA (10 mg/L initial concentrations) removal efficiency was achieved at pH 8 in a lab scale fabricated NF hollow fiber membrane (PES-PEGHBS) from wastewater and deionized water, respectively (Bolong et al., 2010). The main reason for achieving a higher removal efficiency of BPA is due to the solute under the influence of the alkaline solution (pH 8) feed water matrix and the negatively charged properties of the hollow fiber membrane. Escalona et al. (2014) facilitated using enzymatic BPA degradation, operated in a NF membrane coupled to recycling mode using two enzymatic (i.e. peroxidase and laccase) treatment and achieved over 95% removal efficiency of BPA within 180 min and at pH 7.

3.3. Advanced oxidation processes (AOPs)

Advanced oxidation processes (AOPs) are a promising and robust technology that is capable of mineralizing organic micropollutants (i.e. transforming them into CO₂ and H₂O and other inorganic salts) (Cong et al., 2014; Yang 2015; Tasca et al., 2019; Cunha et al., 2019). With this approach, the removal of EDCs can be improved by optimizing the existing technology, specifically through activated sludge treatment or by upgrading the prevailing WWTPs with novel and end pipe treatment technologies. The EDCs biodegradation has commenced by various chemical reaction mechanisms illustrated in Supplementary Information, Table S1. The oxidation process in H₂O₂-based degradation involves: (i) the generation of hydroxyl radicals in water; followed by the reaction between free radicals and organic pollutants, resulting in the complete degradation of complex structured EDCs and conversion to simple chemical structured compounds, such as carbon dioxide and methane (Cesaro and Belgiorno 2016). While in ozonation pursued for wastewater treatment (WWT), the oxidation process relies on the direct action of ozone itself (i.e. selective), where the self-degradation of ozone molecules acts as a dipole (as electrophilic agents and as nucleophilic agents that occurs at an acidic pH). However, the indirect ozonation mechanism involves the indirect action of the hydroxyl radicals formed by the decomposition of ozone in water at basic conditions. It is influenced by the presence of chemical species that can pledge (e.g. hydroxyl and perhydroxyl ions), promote (like methanol and ozone) and inhibit (e.g. carbonate ion) the decomposition of ozone (Cesaro and Belgiorno 2016), where both direct and indirect reactions are taking place concurrently. However, the ozonation efficiency depends on the compounds to be removed and the optimization level of operating conditions.

Accordingly, the removal efficiency of EE2 was increased from 20% to 90% by 15 mg/L of H₂O₂ addition by the photolysis process in water (Rosenfeldt and Linden 2004), while 98% removal efficiency of E1 and E2 was achieved in 1h using UV irradiation; and the degradation rate increased with the H₂O₂ addition (Zhang et al., 2007). The combined UV/H₂O₂ technique is also found to be very effective in the degradation of organic micropollutants in water; hence, the H₂O₂ photolysis destroys the EDCs completely (Zhang and Li 2014). In research done by Zhang and Li (2014), the removal efficiency of E1, E2, EE2, E3, BPA and 4-NP in 2 min was 97%, 92%, 95%, 94%, 89%, and 67%, respectively, using the combined H₂O₂ process in waste activated sludge (WAS) treatment. The oxidation process of EDCs was governed by the hydroxyl radical (.OH) (Zhang and Li 2014). In Fenton processes, oxidation of EDCs has occurred by hydroxyl radicals, generated by the presence of ferrous or ferric ions (Fe²⁺ or Fe³⁺) in its catalytic decomposition process. However, the performance of the Fenton process depends on H₂O₂ and Fe²⁺ concentrations and dosage, the properties of the EOM and the acidic nature of the solution. The oxidation rates of EDCs are strongly increased when the UV system is combined with the Fenton process UV/(H₂O₂/Fe²⁺) (Li and Zhang 2014). Accordingly, the removal of E1, E2, EE2 and E3 in WAS treatment with combined Fenton/H₂O₂ was 70%, 90%, 84% and 98%, respectively (Li and Zhang 2014), while Zhang et al. (2015) described the removal efficiency of E1, E2, EE2 and E3 from WAS using calcium peroxide (CaO₂) as 90%, 92%, 100%, and 93%, respectively. When comparing the oxidation potential of CaO₂ with other oxidative agents (like H₂O₂), the EDCs products during CaO₂ oxidation had less estrogenic activity than the original EDCs.

Heterogeneous photocatalysis is an AOP method in which the heterogeneous catalyst occurs at different phases than the reaction medium. Examples are titanium dioxide (TiO₂), zinc oxide (ZnO), zinc sulfide (ZnS), cadmium sulfide (CdS), tin oxide (SnO₂) and ferric oxide (Fe₂O₃). Amongst them, TiO₂ is the most common and widely used photocatalyst because of its high stability, considerable activity, low cost and environmentally benign (Cesaro and Belgiorno 2016). UV irradiation induces electron-hole pair formation in this process, and the charge carriers have reacted with chemical species (e.g., H₂O₂ molecules and molecular O₂ in the air) to generate hydroxyl radicals, which contribute to the degradation of EDCs at the photocatalyst surface (Cesaro and Belgiorno 2016). Accordingly, Zhang and Li (2014) described the combined UV oxidation with the immobilization of iron on perfluorosulfonic polymer (Nafion) has been examined as a carrier for the oxidation of EDCs estrogens and resulted in over 90% removal efficiency of E1, E2, EE2 and E3 with the only addition of 8.5 mg/L of H₂O₂ at 60 min. Hence, the photo-Fenton

reaction catalysis by the composite and the presence of iron through the catalyst led to the rapid degradation of the estrogenic compounds. The degradation of E2 (10 µg/L initial concentrations) in water using TiO₂ photocatalysis was investigated by Ohko et al. (2002) and E2 was completely mineralized into CO₂ at a 1 g/L dose of the catalysts for 3h. This showed that TiO₂ photocatalysis effectively removed the natural and simulated estrogen pollutants without generating any biologically active intermediate byproducts in its degradation pathways (see Supplementary Information, Figure S2).

Electrochemical (EC) is one of the AOPs developed recently for water and wastewater treatments (Yang 2015). The performance of this technique in WWT depends mainly on the anode properties and on the characteristics of organic pollutants involved in the oxidation process (Cong et al., 2014; Yang 2015). In EDCs removal of the WWT, the hydroxyl radical generated by the electrode in the EC processes is consumed by the co-existing pollutants, thereby requiring and consuming a high energy supply (Cong et al., 2014). The demonstration of the EC process has achieved complete mineralization of 100 µg/L of EE2 within 7 min (Frontistis et al., 2011), and 250–500 µg/L of E2 in 30–40 min (Murugananthan et al., 2007) using boron doped diamond electrode reactors. 96% of EE2 was able to be removed in 15 min using a mixed Ti/SnO₂ electrode (Feng et al., 2010). About 98% of E1, E2 and EE2 (each 1 µg/L initial concentrations) were removed in continuous treatment systems using an electrolytic reactor containing carbon electrodes, while 74–88% removal efficiency of E1 has been found at 100 µg/L loading rate (LR) within 21 d of operation (Cong et al., 2014). In this approach, the removal of estrogen was recognized through estrogen polymerization, while the degradation of the polymer formed (regeneration of the electrode) was done using the hydroxyl radicals generated during the ozone reduction in the process (Cong et al., 2014).

Yang (2015) performed the degradation of BPA using sulfate radicals in an electrochemical assistant Fe²⁺-activated PDS process and achieved 97% of removal efficiency in 120 min at 2.5 mg/L of Na₂S₂O₈ from the simulated wastewater. In a study by Cunha et al. (2019), the performance of a carbon nanotube (CNT) electrochemical filter was applied to remove estrogenic EDCs at 2.5 V and achieved up to 99.1% removal efficiency of both E2 and EE2 in ultrapure water and 96.3% in urban wastewater. In this process, the oxidation of estrogenic compounds has adhered to the CNT surface and may cause electrode polymerization. Besides, no formation of estrogen intermediates was found, while confirming the presence of remaining E2 and EE2 concentration effluents (Cunha et al., 2019).

3.4. Bioremediation techniques

The bioremediation technique is an effective and the most economic means of water treatment for removing micropollutants and produces high biomass energy using photo/bioreactor systems (Shi et al., 2010; Kresinová et al., 2018; Bai and Acharya 2019; Kassotaki et al., 2019). The removal of E2 and EE2 from two spiked UF and ozonation wastewater effluents cultivated with green algae *Nannochloris* sp. and found 60% removal efficiency of the EDCs in 7 d of operation in the UF effluents. In a study by Shi et al. (2010), 95.4%, 94.4% and 93.9% removal efficiency of E1, E2 and EE2, respectively, were achieved using duckweed ponds from the treatment of sewage plant effluents, while 83.9%, 91.2% and 86.8% removal efficiency of E1, E2 and EE2 were found with algae ponds, respectively. The process of estrogenic removal for simulated wastewater can be rapidly adsorbed on algae or duckweed, followed by degradation by the biological biota using microorganisms, duckweed or algae in the WWT system. Abarguesa et al. (2013) achieved 91% removal efficiency for BPA, 4-NP, t-NP and 4-cumylphenol (4-CP) at 22.5 h HRT using the aerated microalgae culture reactor from the membrane effluents. In a study by Kassotaki et al. (2019), another batch of experiments were conducted with lab-scale cultivated enriched biomasses (Nitrifying activated sludge (NAS), Ammonia oxidizing bacteria (AOB), and conventional activated sludge (CAS)) for the removal of E1, E2, E3, EE2 and BPA

in an effluent withdrawn from the municipal wastewater treatment plant (WWTP). In this regard, the heterotrophic fraction of CAS performed better attenuation than AOB and NAS for removal of 100% E2 and 78% E3, while the removal of E1, EE2 and BPA was persistent biologically with a removal efficiency ranging from 10–46% for E1 and 10–39% for EE2 and BPA. This showed that the degradation capability of EDCs was exhibited among the compounds (Kassotaki et al., 2019). The white rot fungus *P. ostreatus* HK 35 was able to remove over 90% of BPA, E1, E2, EE2, E3 and 4-NP within 12 d of lab-scale operation, while above 95% of EDCs removal efficiency was achieved in a pilot-scale trickle bed bioreactor comprising substrates of spent mushroom with the suppression of estrogenic activity. After 10 d of operation, the pilot-scale trickle bed bioreactor was able to remove over 76% of EDCs successfully from WWTP (Kresinová et al., 2018). The *U. isabellina* culture was able to remove t-NP and 4t-NP (each 25 mg/L initial concentration) with 90% removal efficiency at 12 h of operations, while under the presence of metal ion competition, 15.2% of t-NP and 65.3% of 4t-NP removal efficiency had been achieved (Janicki et al., 2018). This demonstrates the ability of the single culture, *U. Isabellina*, to remove both heavy metals and EDCs at the same time, and confirms its suitability for the development of strategies for removing xenobiotic pollutants from co-contaminated environments. The *T. versicolor* pellets were able to degrade the EDCs with 83.2% removal efficiency in a batch fluidized bed bioreactor under the non-sterile conditions of real mixed hospital wastewater treatment (Cruz-Morató et al., 2014).

3.5. Phytoremediation techniques

Constructed wetlands (CWs) based phytoremediation process applied to the concurrent removal of nutrients, COD and trace level organic pollutants from the contaminated ecosystem (Dai et al., 2017). In developing countries, the phytoremediation technique is an area chosen by many researchers for treating xenobiotic pollutants due to its low cost of operation, lower energy consumption, and reduced energy consumption. In a study by Toro-Vélez et al. (2017), tropical horizontal sub-surface CWs were investigated using *Heliconia* to achieve 73% of BPA and 63% of NP, while *Phragmites* CW accomplished 70% BPA and 52% NP removals and, in an unplanted CW system, performed at 62% BPA and 25% NP removal efficiencies. The higher removal performance of *Heliconia* CW suggests that a native plant from the tropics can achieve better removal efficiency of EDCs. The capability of a stacked constructed wetland (SCW) involving horizontal and vertical flows of CWs and an assembled biofilter (ABF) was investigated to remove phenolic EDCs (PEDCs) from the sewage treatment plant (STP) at higher loading rates (HLRs) of 0.5–2.0 m/d. The SCW and ABF revealed different PEDCs removal performances with 58 and 51% of 4-t-octyl phenol (4-t-OP), 48 and 44% of E1, 45 and 32% of BPA, respectively, due to their process structural differences (Dai et al., 2017). This showed that process optimization by increasing the number of substrate layers, by considering ABF design flexibility and through integrating approach, a better removal performance of EDCs can be achieved than single CWs systems.

4. Removal mechanisms of some EDCs

EDCs are ubiquitous in the aqueous ecosystem, and are an emerging concern for water quality deterioration as multiple EDCs have been detected in surface waters and wastewater effluents (Neamtu and Frimmel 2006). Supplementary Information, Figure S2 to Figure S4 expresses the degradation mechanisms of some phenolic EDCs (E2, BPA) in typical AOPs, while Figure S5 and Figure S6) shows the metabolic biodegradation mechanisms of EE2 and BPA, respectively. Figure S3 illustrates the degradation mechanism of E2 into intermediate byproducts such as 10 ϵ -17 β -dihydroxy-1,4-estradieno-3-one (m/z 287) was formed, while byproduct 2 (m/z 278) was proposed by the ozonolysis of the phenolic moiety (Pereira et al., 2011). Katsumata et al. (2004) also investigated the treatment of BPA using Fenton reagent in water under UV-irradiation

and achieved more than 90% mineralization after 36 h. Figure S4 illustrates the photo-Fenton decomposition of BPA and produces 6 intermediate byproducts: phenol, 4-hydroxyacetophenone, 4-isopropylphenol, p-hydroquinone, 4-isopropenyl-phenol, methyl benzofurans and some oxidative ring-opened products (i.e. formic acid, acetic acid and acetaldehyde and then finally converted into carbon dioxide and water (Katsumata et al., 2004). Figure S5 also shows the biodegradation of BPA using environmentally distributed soil bacteria *Sphingomonas* sp. strain AO1 and the cytochrome P450 monooxygenase system was also involved (Sasaki et al., 2005). The BPA biodegradation pathway involves two primary intermediate metabolites, namely 4-hydroxybenzoic acid, 4-hydroxyacetophenone, and two primary metabolites (2,3-bis(4-hydroxyphenyl)-1,2-propanediol and 2,2-bis(4-hydroxyphenyl)-1-propanol). The 4-hydroxybenzoic acid was made from 4-hydroxyacetophenone through oxidative rearrangements (Figure S5) (Suzuki et al., 2004). EE2 (30 mg/L) was metabolized up to 87% within 10 d using bacterial strain *Sphingobacterium* sp. JCR5 (Haiyan et al., 2007) from a WWTP of the oral contraceptive factory. Figure S6 shows the biodegradation pathways of EE2 undergoing subsequent ring cleavages. An intermediate metabolite of 3,4-dihydroxy-9,10-secoandrosta-1,3,5(10)-triene-9,17-dione was formed. Furthermore, two intermediate metabolites, namely, 2-hydroxy-2,4-dienevaleric acid and 2-hydroxy-2,4-diene-1,6-dioic acid are formed and mineralized into CO₂ and H₂O (Figure S6) (Haiyan et al., 2007).

5. Future research perspectives

The EDCs, identified as emerging micropollutants, are known as a problematic issue due to their bioaccumulation nature and owing to their accidental discharge in the environment (Kassotaki et al., 2019; Bai and Acharya 2019). The majority of the negative health effects of EDCs are caused by the poor and incomplete degradation of emerging EDCs by a single treatment technique in water and wastewater. As a result, the existing conventional treatment system has reverted to using EDCs as a point source of ecological contamination by producing toxic sludge containing EDCs. Therefore, the mitigation of EDCs toxicity should be based on the degradation processes through mineralization pathways into nontoxic and stable products. The complete removal of EDCs is challenging because of their persistency and complexity in water, wastewater and sediment. As a result, more research will be needed on: (i) the concurrent adsorption of various EDCs using the same adsorbent materials in both simulated and real wastewater to achieve better removal efficiency; (ii) most reports of EDCs removal are from water or wastewater, but food is identified as one of the most common EDC exposures to humans, therefore, further study will be needed on the removal of EDCs from the contaminated food; and (iii) the upgrading, installing and redesigning of conventional WWTPs with the available convenient technologies aiming for the complete removal (degradation or mineralization) of the EDCs from the urban WWTPs as a post treatment, and (iv) the research on emerging EDCs detection and removal should be prolonged in the developing countries, which will benefit the water industry and policy makers to pick alternative and appropriate advanced wastewater treatment techniques.

6. Conclusion

This review illustrates the recent advances in the main phenolic-based endocrine disruptors and their metabolites from the contaminated environment. EDCs are an emerging contaminant of concern even at very concentrations (in µg or ng levels). The conventional WWTP does not entirely remove EDCs and part of the removal is attained by transferring the pollutants into a high volume of sewage sludge, which becomes a source of other secondary pollution that causes various risks to human and environmental health. Due to the lack of the most convenient tertiary WWT technologies, the problem is more severe in developing countries. The majority of research has been conducted on simulated waste containing a single

compound to be degraded, with only a few studies on real waste and endocrine disruptors existing in mixtures of various components. The performance of the techniques depends on the operating conditions of the wastewater treatment (such as initial concentrations, pH, temperature, nature of the EDCs in the aquatic environment, etc.). The sustainability of EDCs removal can be assured through the use of combined wastewater treatment processes (combination of AOPs with adsorption/biological techniques).

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