Chapter 7

Endocrine-Disrupting Pollutants in Industrial Wastewater and Their Degradation and Detoxification Approaches

Izharul Haq and Abhay Raj

Abstract Endocrine-disrupting chemicals (EDCs), a group of chemicals that alter the normal function of the endocrine system of humans and wildlife, are a matter of great concern. These compounds are widely distributed in respective environments such as water, wastewater, sediments, soils, and atmosphere. Chemicals like pesticides, pharmaceuticals and personal care products, flame retardants, natural hormones, heavy metals, and chemicals derived from basic compounds (such as plasticizers and catalysts) are major endocrine disruptors. EDCs emerging from industries such as pulp and paper, tannery, distillery, textile, pharma, etc. have been considered as major source of contamination. Alkylphenol ethoxylates (APEOs), bisphenol A (BPA), phthalates, chlorophenols, norethindrone, triclosan, gonadotropin compounds, pesticides, etc. are generally escaped during wastewater treatment and contaminate the environment. Endocrine-disrupting activity of these compounds is well documented to have adverse effect on human-animal health. Globally, efforts are being approached for their efficient removal from sewage/wastewaters. Thus, this chapter provides updated overview on EDC generation, characteristics, and toxicity as well as removal/degradation techniques including physical, chemical, and biological methods. This chapter also reviews the current knowledge of the potential impacts of EDCs on human health so that the effects can be known and remedies applied for the problem as soon as possible.

Keywords Endocrine disruptors · Industrial wastewaters · Human-animal health · Biological treatment

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1 Introduction

Thousands of anthropogenic toxic chemicals currently are released from industries into the environment (Vandenberg et al. 2009; Eskinazi et al. 2003; Markey et al. 2003). Endocrine systems control a huge number of metabolic, developmental, and reproductive processes including embryonic development, gonadal formation, sex differentiation, growth, and digestion. Chemicals that interfere normal function of the endocrine system at certain doses are referred as endocrine-disrupting chemicals (EDCs). EDCs may affect these processes by either binding to or blocking hormone receptors, thereby triggering or preventing hormonal response (Pedersen et al. 1999; Markey et al. 2003; Witorsch 2002; Hotchkiss et al. 2008).

EDCs are highly diverse that includes synthetic chemicals used as industrial solvents/lubricants and their by-products (polychlorinated biphenyls, polybrominated biphenyls, dioxins), plastics, plasticizers, pesticides, fungicides, phytoestrogens, pharmaceutical agents, and certain industrial or commercial products (Bharagava and Mishra 2018; Diamanti-Kandarakis et al. 2009; Falconer et al. 2006). These chemicals can enter the aquatic environment through effluent discharge or storm-water runoff. The major transport of EDCs to the aquatic environment is through industrial and municipal wastewater discharge into the rivers, streams, and surface waters (Clara et al. 2005; Falconer et al. 2006; Zhang et al. 2015) (Fig. 7.1). Potable water resources, including both surface water and groundwater, can become contaminated through surface water discharge or deep-well injection of wastewater treatment plant effluent (Mompelat et al. 2009; Bharagava and Chandra 2010b; Kumari et al. 2016).

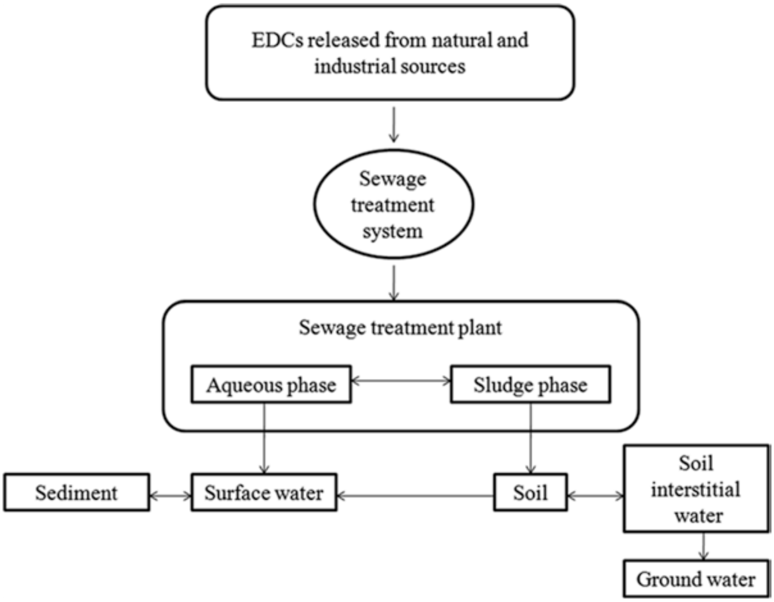

Fig. 7.1 Distribution of EDCs in environment

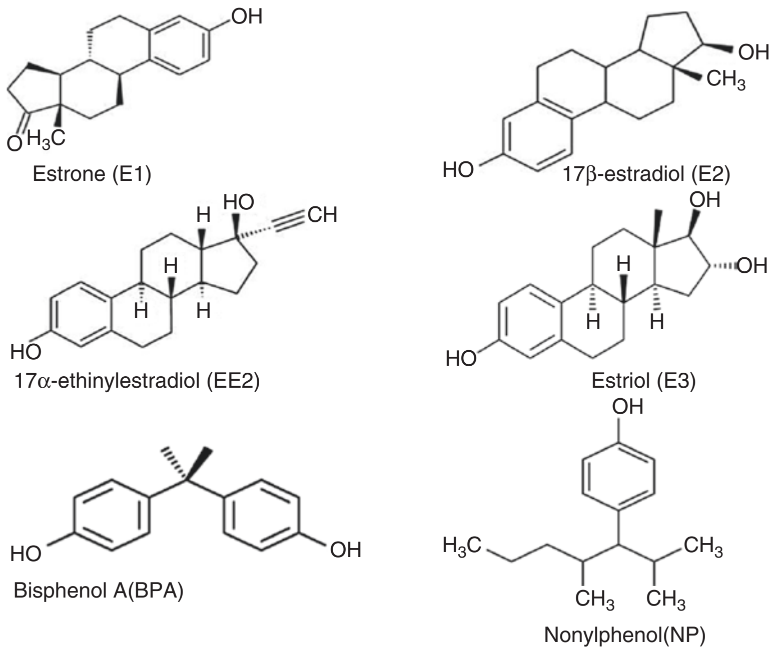

Fig. 7.2 Structure of common synthetic and industrial estrogenic compounds

Effects of EDCs on wildlife (invertebrates, fish, amphibians, reptiles, birds, and mammals) include abnormal blood hormone levels, altered gonadal development (e.g., imposex and intersex), induction of vitellogenin gene and protein expression in juveniles and males, masculinization/feminization, hermaphroditism, and decreased fertility and fecundity (Jobling et al. 2004; Kidd et al. 2007; Janex-Habibi et al. 2009; Nadzialek et al. 2010).

Estrogenic compounds specifically target estrogen signaling including steroidal estrogens, synthetic estrogens, and industrial compounds which mimic estrogen. 17β-Estradiol (E2) is the main natural estrogen, which has greatest potency than estrone (E1), a metabolite of E2 and estriol (E3), considered to be the final metabolite. 17α-Ethinylestradiol (EE2) is the synthetic steroidal estrogen component of contraceptives (Spencer et al. 2009). Bisphenol A (BPA) is a chemical used in industry to produce lacquers, food-can liners, and thermal paper (Danzl et al. 2009). Nonylphenol (NP) is the persistent and estrogenic final product of the biodegradation of the nonionic surfactant nonylphenol ethoxylate (NPEO) (Gultekin and Ince 2007; Chandra et al. 2011). β-Sitosterol, a plant sterol, emerged from pulping industry and discharged into stream waters. The structures of some commonly used synthetic and industrial EDCs are given in Fig. 7.2.

The emissions of natural and synthetic EDCs from industries and its effects in receiving aquatic flora and fauna are not well regulated. However, monitoring of chemical compounds clearly indicates that conventional wastewater treatment processes, e.g., activated sludge process (ASP) and aerated lagoon, are unable to remove most EDCs. Consequently, wastewater treatment plants are one of the most crucial point sources for the contamination of receiving waters by EDCs. Various physicochemical (ozonation, chlorination, adsorption, membrane filtration, advance oxidation process, activated carbon treatment, electrochemical method, etc.) treatment processes are currently proposed for the removal of EDCs (Hu et al. 2003; Huber et al. 2004; Nakamura et al. 2006; Bila et al. 2007). Alternatively, bioremediation is a cost-effective and environmental friendly approach which employs microorganisms for detoxification of environmental pollutants (Zhang et al. 2013; Yang et al. 2014; Bharagava et al. 2017a, b). Among the different microorganisms, fungi and bacteria have been well documented for the degradation of EDCs. During the last decade, considerable amounts of scientific and financial resources have been employed to identify the EDC sources, distribution, potential risk to human and animal health, and its remedial approaches. The present book chapter provides updated information on the EDCs including source, distribution, human and animal health risk, and their remedial strategies for environmental safety.

2 Sources of EDCs

Industrial effluents have been considered as the key source of EDCs to the aquatic environment (Ying et al. 2002; Saxena et al. 2017; Voutsa et al. 2006). The exposure of EDCs is usually diverse and widely distributed all over the environment and society of the world. But the situation is neither constant nor can be predicted easily since there is a significant usage difference of these substances among the countries. Globally, it can be seen that EDCs have notable toxicities in human as well as in wildlife (Diamanti-Kandarakis et al. 2009; Saxena and Bharagava 2017). Among the EDCs, alkylphenol ethoxylates (APEOs), BPA, phthalates, chlorophenols, norethindrone, triclosan, gonadotropin compounds, pesticides, etc. are most concerned because of their widespread in the environment from various sources (Liu et al. 2011; Staples et al. 2000). EDCs are discharges from various industries including pulp and paper, tannery, distillery, etc. Pulp and paper mill effluents are major source of various types of EDCs (Yadav et al. 2016; Chowdhary et al. 2017).

Alkylphenol polyethoxylates (APEs) and related compounds recently have been reported to be estrogenic, because they have similar effects to estradiol both in vitro and in vivo. Alkylphenols are degradation by-products of APEs, which are used in paper industry as defoamers, cleaners, and emulsifiers. β-Sitosterol, a plant sterol found in higher plants, emerged from pulp and paper industry. Phthalates are known EDCs, which originate in paper production process mostly as softeners in additives, glues, and printing inks. Besides, EDCs like NP; 4-aminobiphenyl, hexachlorobenzene, and benzidine have also been detected in effluents emerging from tannery industries (Kumar et al. 2008). Phthalates compounds like bis(2-ethylhexyl) phthalate (DHEP), dibutyl phthalate (DBP), and bis(2-methoxyethyl) phthalate have also been found in tannery effluents (Bharagava et al. 2017a, b; Alam et al. 2010).

EDCs are also present in distillery effluents such as (butanedioic acid bis(TMS) ester; 2-hydroxyisocaproic acid; di-n-octyl phthalate; dibutyl phthalate (Chowdhary et al. 2018; 2017; Chandra et al. 2012; Chandra and Kumar 2017) benzenepropanoic acid, α-[(TMS)oxy], TMS ester; vanillylpropionic acid, bis(TMS)), and other recalcitrant organic pollutants (2-furancarboxylic acid, 5-[(TMS)oxy] methyl], TMS ester; benzoic acid 3-methoxy- 4-[(TMS)oxy], TMS ester; and tricarballylic acid 3TMS) (Chandra and Kumar 2017). The major EDCs found in different industrial effluents are alkylphenol ethoxylates (APEOs), a group of nonionic surfactants, are widely used in domestic and industrial applications. APs, NPs, 4-tert-octylphenol (4-t-OP), and 4-tert-butylphenol (4-t-BP) are part of APEOs. APs are more toxic and stable than their parent compounds in the environment.

Several countries have been reduced the use of APEOs and APs due to adverse effects to humans and wildlife. BPA is a polar monomer of polycarbonate plastic, used in polycarbonate, epoxy resins, unsaturated polyester-styrene resins, flame retardants, and many other products. Some final products from the BPA are used as coatings on cans, additives in thermal paper, and antioxidants in plastics (Kang et al. 2006). Due to large-scale production and extensive use of BPA, it has become an essential part of industrial wastewater streams (Staples et al. 2002; Latorre et al. 2003). BPA has also been shown to be estrogenic via in vivo screenings (Brotons et al. 1995; Laws et al. 2000). A low-dose effect of BPA was observed in rats (Kobayashi et al. 2005), whereas in fish, it affects the progression of spermatogenesis (Sohoni and Sumpter 1998). Furthermore, BPA has also been found to have the paradoxical effect to block the beneficial effects of estradiol on neuronal synapse formation and the potential to disrupt thyroid hormone action (MacLusky et al. 2005; Zoeller et al. 2005; Mohapatra et al. 2010).

Phthalates or phthalic acid esters (PAEs) have been widely used as plasticizers for polyvinyl chloride (PVC) resin, cellulose film coating, styrene, adhesives, cosmetics, and paper manufacturing (Olujimi et al. 2010; Patnaik 2007). The PAEs such as dimethyl phthalate (DMP) and diethyl phthalate (DEP) are among the most frequently detected in diverse environmental samples including surface marine waters, freshwaters, and sediments (Jing et al. 2011; Zeng et al. 2011). The estrogenic activity of PAEs such as dibutyl phthalate (DBP), butylbenzyl phthalate (BBP), dihexyl phthalate (DHP), diisoheptyl phthalate, di-n-octyl phthalate, diisononyl phthalate, and diisodecyl phthalate is observed by Zacharewski et al. (1998). Further studies in fish have shown that both BBP and DEP induced vitellogenin (VTG) at an exposure to low concentration in the range of $\mu g\ L^{-1}$ via the water (Harries et al. 2000; Barse et al. 2007). Numerous in vivo screens and tests have demonstrated that PAEs mediated their effects through binding to the estrogen receptor (Andersen et al. 1999; Zacharewski et al. 1998).

In addition to these estrogenic effects, some PAEs are also considered to be toxic to microorganisms, aquatic life, and human beings (Bajt et al. 2001). Recent studies have indicated that phthalate metabolites such as monoethyl phthalate (MEP), mono-(2-ethylhexyl) phthalate (MEHP), mono-n-butyl phthalate (MBP), and monobenzyl phthalate (MBzP) can induce DNA damage in human sperm (Duty et al.

2003; Hauser et al. 2007). Norethindrone is one of synthetic progestogens which is used in contraceptive treatments for the promotion of menstrual cycles and correction of abnormal uterine bleeding (Chang et al. 2011). Norethindrone is released into the environment through humans' and animals' urines and feces (Fent 2015). Triclosan is an important bactericide used in various personal care and consumer products (Ying and Kookana 2007). Natural or synthetic steroids are excreted by mammals, and eventually, they occur in domestic effluents and in livestock waste (Ying et al. 2002). The previous reports revealed that the existing industrial wastewater treatment processes could not completely remove EDCs (Ifelebuegu 2011; Samaras et al. 2013) and discharge into receiving water bodies.

3 Impact of EDCs on Human and Animal Health

As mentioned in earlier sections, our daily life is surrounded by a wide range of EDCs. Thus, from a physiological perspective, natural or synthetic EDCs alter the hormonal and homeostatic systems of an organism. EDCs tend to be relatively bioaccumulative and persistent and produce adverse developmental, reproductive, neurological, hormonal, metabolic, and immune effects in both humans and wildlife (Fig. 7.3).

Studies conducted on animal, clinical observations, and also the epidemiological studies have indicated its potential role in affecting reproductive systems, prostate, breast, lung, liver, thyroid, metabolism, and obesity (Polyzos et al. 2012). EDCs can disrupt sperm production and development while exposed to the testicle of an organism (Aly et al. 2009). In the recent study, it has been suggested that high concentration of DDT/DDE has negative impact on sperm motility, morphology, count, and semen volume (Aneck-Hahn et al. 2007). Other studies also observed that exposure of DDT adversely affects sperm quality, mainly through decreased motility (Rignell-

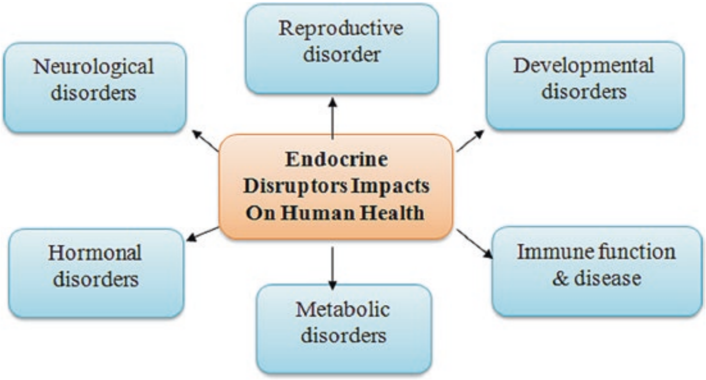

Fig. 7.3 Impact of endocrine disruptors on human health

Hydbom et al. 2004; Toft et al. 2006). Several studies have been reported carcinogenesis of environmental EDCs which leads to development of prostate cancer, breast cancer, and testicular cancer (Høyer et al. 1998; Ritchie et al. 2005; Hardell et al. 2006; McGlynn et al. 2008).

Thyroid hormones play an important role in the maintenance of body homeostasis. The unusual changes in thyroid status may lead to altered basal metabolic rate, lipid metabolism, as well as cardiovascular, gastrointestinal, neurological, and muscle function. Thyroid hormones are especially important during growth and development such as the maturation of the brain. A number of environmental agents can alter thyroid hormone levels in humans and animals. Hypothyroidism in rodents has been observed after exposure to PCB and chlorinated pesticides (Crisp et al. 1998). Developmental neurotoxicity involving cognitive and neurobehavioral disturbances has been implicated following perinatal exposure to environmental pollutants (Jacobson and Jacobson 1996). There are several studies which show that EDCs produce a wide spectrum of neurochemical and neuroendocrine effects in humans and animals (Koopman-Esseboom et al. 1996; Tilson and Kodavanti 1997; Eriksson et al. 1998).

EDCs not only affect human health but also terrestrial and aquatic organisms. There are large numbers of evidences available which indicated that endocrine disruptors are also responsible for different wildlife crises. However, wildlife is not exposed to single contaminants, instead to a mixture of chemicals, some of them acting through a common pathway. The exposure route of wildlife to EDCs is also very critical and crucial. This is because many of the endocrine disruptors do not persist in the environment and organism. Most of the EDCs are either degraded in the environment by sunlight, bacteria, and chemical processes or persist for different time ranges. The organism may follow the same uptake route as the humanlike ingestion or by absorption through the skin (Kidd et al. 2012).

For better understanding of the EDCs exposure to wildlife, studies have been conducted on animals of distant and local places. The fishes are expected to be exposed to a high level of EDCs because of their existence near the localized area. The water source as well as the low-level land is continuously being exposed to EDCs due to sewage treatment process and industrial effluent discharges. Although EDCs cannot persist in the water for longer periods of time, regular disposal of these chemicals into the water makes the aquatic wildlife be in contact with EDCs continuously. However, in spite of being in remote places, highest levels of perfluorooctanesulfonate (PFOS), polychlorinated bisphenols (PCBs), and organochlorine pesticides have been also detected in the polar bears which may be due to the long-distance transport of these chemicals into those areas (Kidd et al. 2012). However, for animal lives, primary source of EDC exposure can be thought to be the water source. The fish uptake these contaminants from water through gills, while wild birds and mammals may uptake these through drinking water. After EDC exposure to animals, it travels across the body and is metabolized. While some of the EDCs accumulate in fat tissues, or they are eliminated from the body by taking a variety of routes. They can also be end in eggs of fishes and birds exposed to them. In this way, EDCs travel from the body through lactation and pass into the offspring.

However, in the animal body, the liver is the main site of their metabolism, after which they are eliminated through urine and feces (Kidd et al. 2012). Various studies have been done previously to understand the effects of EDCs on wildlife. These studies have provided a relationship between these chemicals and their effects on animals. However, some EDCs have shorter half-lives, so sometimes the tasks are difficult to be performed. The persistent organic pollutants (POPs) have been intensively studied in wildlife including dichlorodiphenyltrichloroethane (DDT), chlordanes, dieldrin, PCBs, dioxins, polybrominated diphenyl ethers (PBDEs), etc. Therefore, it is clear that the monitoring of EDC evaluation on wildlife will take several years. Available information is still not enough to provide a visible indication of the level of EDC exposure in many areas, especially in tropical and subtropical areas. At this point, it is crucial to conduct more studies on wildlife to save our endangered wildlife as well as our precious ecological systems.

4 Wastewater Treatment Techniques and EDC Removal

4.1 Physical and Chemical Treatment

4.1.1 Membrane Technology

There are two basic types of membrane separation processes: pressure-driven and electrically driven. Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) are the pressure-driven filtration processes which use hydraulic pressure to force water molecule through the membrane (Adams et al. 2002; Walha et al. 2007). In the electrically driven membrane process, electric current is used to move ions across the membrane leaving purified water behind. Membrane treatment, applied to the end of conventional wastewater treatment system, is a viable method of achieving desired effluent quality level. Membranes are commonly used for the removal of dissolved solids, color, and hardness in drinking water. Published scientific literatures show removal of several types of EDCs present in wastewaters (Bodzek 2015). The RO and NF processes can effectively remove phytoestrogens (70–93%), PAH (85–99%), and surfactants (92–99%) from industrial wastewaters. In wastewater contaminated with greater pollutants, UF process can be used for the surfactant removal. Plant protection products such as pesticides, herbicides, and insecticides present in surface and groundwaters can be effectively removed from water during NF or by integrated systems of MF or NF and activated carbon adsorption (powdered or granulated) (Bodzek 2015; Mavrov et al. 1992).

The concentration of phthalates in different parts of the environment, especially in water, should also be controlled by these processes, as high retention of phthalates was observed during both RO and NF processes (initial concentration 40 μg/L) (Bodzek et al. 2004). Retention rates achieved for diethyl phthalate, di-n-butyl, and di2-ethylhexyl were very high and amounted from 89.7% (UF) to 99.9% (RO and NF). Phenolic xenoestrogens such as APs, NPs, BPA, and BPF can be removed from water by means of NF (Bodzek and Możliwości 2013). Recently, an increase in concentrations of the synthetic hormone such as α-ethinylestradiol, mestranol, and diethylstilbestrol which emerged from the large amounts of expired pharmaceuticals, both from households and from wastewater and hospital wastes as well as pharmaceutical plants, has been observed. It was shown that elimination of this type of pollutants from water could be performed by means of membrane processes (Bodzek and Dudziak 2006). Considering relatively low molecular weight of those pollutants RO or NF must be applied. It has been found that RO membranes totally eliminate particular hormones, while retention coefficients obtained for NF and UF membranes were lower (Bodzek and Dudziak 2006).

4.1.2 Absorption by Activated Carbon

Use of activated carbon (AC) is a well-known process for removing various organic contaminants and organic carbon in general. AC is most commonly used as a powdered form (powder activated carbon, PAC) or in a granular form (granular activated carbon, GAC) in packed bed filters. Granular activated carbon (GAC) is used at many water treatment plants in the United States and Canada. The GAC can be used as a replacement for anthracite media in conventional filters, thus providing both adsorption and filtration. Alternatively, GAC can be applied post-conventional filtration as an adsorbent bed (Snydera et al. 2007). Several authors have confirmed the effectiveness of AC (PAC and GAC) for the removal of trace organic pollutants from water (Matsui et al. 2002; Asada et al. 2004; Westerhoff et al. 2005; Zhou et al. 2007). Also, several studies have found that AC can remove broad range of EDCs for artificial and real wastewater in the laboratory and pilot and full-scale plants (Choi et al. 2005; Fukuhara et al. 2006). In a study by Abe (1999), absorbability of about 70 EDCs by AC was estimated from their chemical structures, and then their adsorption by AC was proven effective for their removal from wastewater. Representative endocrine substances such as estrone (E1) and17β-estradiol (E2) were also used to evaluate the removal performance on absorption. Compared to artificial EDC wastewater, a great difference in EDC removal by AC adsorption between simulated wastewater and real wastewater was observed.

4.1.3 Advanced Oxidation

Advanced oxidation processes (AOPs), through the use of cheap chemicals, have been demonstrated as a viable alternative for removing trace estrogens. Few installations are in operation today because of their high operating costs on large-scale installations (Madsen et al. 2006). Ozone and hydrogen peroxide are the most diffused oxidants. Ozone has long been used for disinfection of drinking waters, and this was the origin of its application to the degradation of several organic micropollutants. Ozone can react selectively, as an oxidant in its molecular form (O_3), thus leading to typical ozonation reactions with, for example, double bonds, amines, and phenol derivatives, or nonselectively, after formation of hydroxyl radicals (HO•) (Von Gunten 2003; Haag and Yao 1992). In all cases, depending on the structure of the organic substrate, steroid derivatives can be degraded to lower molecular weight compounds (by-products) of unknown estrogenicity. The synergistic use of ozone with other oxidants (e.g., H_2O_2) or in association with physical means such as UV radiation is justified to reduce the selectivity of action in the hope of amplifying the destruction of trace organics (Von Gunten 2003). Fenton process is based on the use of ferrous ions in association with hydrogen peroxide in acidic media (pH = 3). This process couples synergistically with the oxidation process to coagulation/flocculation, with the latter process occurring later when the pH is raised to neutralize the final effluent (Petruzzelli et al. 2007). Kunde et al. (2009) explored the oxidation reaction of BPA and tetrabromobisphenol A (TBBPA), among the most heavily used polymer plasticizers and flame retardants, respectively (Haag and Yao 1992; Kunde et al. 2009). This treatment appears to be promising in the degradation of estrogens because of the self-regenerating cycle operated by MnO_2, thus proving to be cost-effective in the long run. Ferrate ion was investigated as a viable oxidant and coagulant alternative.

Although the oxidation potential of ferrate ions is greater than that of ozone, the acidic conditions of operation strongly limit its use in full-scale installations (Lee et al. 2005; Jiang et al. 2005). Chlorine and chloramines both have the potential to react with various EDCs and personal care compounds. Chlorine gas is a fast-reacting and efficient chemical for phenols and amino functional derivatives but is nearly nonreactive with ketones and alcohols, which are only partially oxidized. It is also important to note that chlorine oxidation is pH-dependent, with the best performance obtained in acidic media. In a study recently carried out by the American Water Works Association, it was confirmed that the antibiotics sulfamethoxazole, trimethoprim, and erythromycin are among the compounds that exhibit better removal efficiency upon the use of chlorine gas as compared to chloramines (Synder et al. 2011).

4.1.4 Electrochemical Methods

Electrochemical process involves chemical reactions caused by interaction between electrode and chemicals. It is an emerging technology applied in water and wastewater treatment. Most focus has been given to electrodeposition, electrocoagulation (EC), electroflotation (EF), and electrooxidation. Anodic degradation (oxidation) of EDCs, namely, 17β-estradiol (E2) and bisphenol A (BPA), by the use of a boron-doped diamond electrode (BDDE) was investigated at the laboratory scale. Cyclic voltammetry experiments were carried out to evaluate the electrochemical process response to E2 and BPA degradation as a function of the applied voltammetry cycles. Apparently, electrooxidation reaction was controlled by the applied current density, which was evaluated and discussed at three different levels. Electrolysis at high anodic potential caused quantitative oxidation of EDCs with formation of CO_2. The effects of operating conditions in the electrolytic bath [e.g., pH, background solutions (Na_2SO_4, $NaNO_3$, and NaCl)] were discussed in terms of electro-generated inorganic oxidants such as $S_2O_8^{2-}$, H_2O_2, and ClO^-. Better performance of the BDDE anode was found on a comparative basis with respect to, for example, Pt and amorphous graphite under similar experimental conditions (Yoshihara and Muruganathan 2009).

4.2 Biological Treatment

Physicochemical methods were shown to be effective in removing EDCs from water and wastewaters (Hu et al. 2003; Bila et al. 2007). However, use of these processes is cost-effective and energy-consuming, and the efficiency of these processes under field conditions is limited by the low concentrations of the contaminants. Biological treatment is a particularly attractive and cost-effective approach, as it represents natural and economically feasible processes for detoxification of environmental pollutants under environmental conditions. An environmental friendly process alternative for the elimination of EDCs may be the use of fungi, bacteria, algae, and plants. Among the different microorganisms, white rot fungi have been studied for their ability not only to eliminate EDCs but also to reduce their estrogenic activity. EDC-reducing ability of fungi is usually related to the production and secretion of lignin-modifying enzymes.

4.2.1 Conventional Wastewater Treatment Plants

Wastewater treatment plants receive raw wastewater from domestic and/or industrial discharges. The main objective of a wastewater treatment system is to remove only phosphorus, nitrogen, and organic substances. The wastewater treatment system mainly used activated sludge process (ASP) which is most widely used in the entire world because of its high efficiency and cost-effectiveness. In the ASP, effluent received after primary treatment is treated by the biological process using microbes for the removal of organic contaminants from wastewater. Currently, under optimized conditions, more than 90–95% of substances can be eliminated by conventional biological-based methods used in wastewater treatment plants (Li et al. 2000; Cases et al. 2011). However, ASP is energy intensive due to the high aeration requirement, and it also produces large quantity of sludge (about 0.4 g dry weight/g COD removed) that has to be treated and disposed of. As a result, the operation and maintenance cost of the ASP is considerably high. Anaerobic process for domestic wastewater treatment is an alternative that is potentially more cost-effective, particularly in the subtropical and tropical regions where the climate is warm consistently throughout the year. Anaerobic wastewater purification processes have been increasingly used in the last few decades. These processes are important because they have positive effects: removal of higher organic loading, low sludge production, high pathogen removal, methane gas production, and low energy consumption (Nykova et al. 2002). Researchers have shown that anaerobic systems such as the upflow anaerobic sludge blanket (UASB), the anaerobic sequencing batch reactor (AnSBR), and the anaerobic filter (AN) can successfully treat high-strength industrial wastewater as well as low-strength synthetic wastewater. However, the existing conventional wastewater treatment plants are not able to remove pollutants specially EDCs (Bolong et al. 2009; Liu et al. 2009; Berge´ et al. 2012).

The conventional aerobic and anaerobic treatment system cannot degrade all compounds completely or convert into biomass. For instance, the EDCs found in effluent are the products of incomplete breakdown of their respective parent compounds (Johnson and Sumpter 2001). Wintgens et al. (2002) and Gallenkemper et al. (2003), using toxicological evaluations, also indicated that wastewater treatment plants were not able to remove these novel substances sufficiently before disposing effluent into the environment. Liu et al. (2009) reported that APs, BPA, etc. are not completely removed by existing wastewater treatment plants and remained at fluctuating concentrations in effluent, so discharge of such effluent may be the main reason for the wide distribution and occurrence of EDCs in surface waters, groundwaters, and even in drinking waters.

4.2.2 Membrane Bioreactors (MBRs)

MBR technology is considered as new and promising biological approaches in wastewater treatment system, thus integrating biological degradation with membrane filtration, with specific reference to biopersistent organic substrates. The main advantages of using MBR systems are (a) improved quality of the treated wastewater, (b) more compact plant size, (c) less sludge production, and (d) higher flexibility of plant operations for improved EDC removal by the adoption of variable solid retention time (SRTs) and/or hydraulic retention time (HRTs). In these systems, the quality of the final effluent depends strongly on the settling characteristics of the sludge and the hydrodynamic conditions in the sedimentation tank. Accordingly, large-volume sedimentation tanks, offering residence times of several hours, and strict control of the biological unit are necessary to favor sludge settling (granulation), thus minimizing bulking phenomena. The final objective is to obtain adequate solid/liquid separations for optimal performance of the membrane separation to follow. Very often, site-specific and economic constraints limit such options.

4.2.3 Fungal Treatment of EDCs

The white rot fungus *Pleurotus ostreatus* HK 35 has been tested in the degradation of typical representatives of EDCs (BPA, estrone, 17β-estradiol, estriol, 17α-ethinylestradiol, triclosan and 4-nnonylphenol), and degradation efficiency under laboratory conditions was greater than 90% within 12 days (Kˇresinov'a et al. 2017). Castellana and Loffredo (2014) indicated that in a period of 20 days, *Trametes versicolor* growing on the various substrates removed almost 100% of BPA, EE2, NP, and linuron and from 59% to 97% of dimethoate and *Stereum hirsutum* showed a marked degrading activity only toward NP, which was totally removed after 20 days or less with any substrate and, to a lesser extent, linuron. Pezzella et al. (2017) have investigated degradative capabilities of *Trametes versicolor*, *Pleurotus ostreatus*, and *Phanerochaete chrysosporium* to act on five EDCs, which represent different classes of chemicals (phenols, parabens, and phthalate). *T. versicolor* was able to efficiently remove all compounds during each cycle converting up to 21 mg L^{-1} day^{-1} of the tested EDCs. In a study of Sei et al. (2007), five highly laccase producible fungal strains, *Trametes hirsute* 1674, *T. orientalis* 1071, *T. versicolor* IFO 30340, *T. versicolor* IFO 30338, and *Pycnoporus coccineus* 866, were used for the removal of various EDCs. The result found that bis(4-hydroxyphenyl) sulfone, diethylhexylphthalate (DEHP), pyrene (PY), anthracene, 3,5-dichlorophenol, and pentachlorophenol could not be removed by laccase. DEHP and PY could not be removed even with mediators. In another study of Macellaro et al. (2014), different strains of *Pleurotus ostreatus* producing laccases were also used for EDC enzymatic treatment. The use of fungi for the removal of EDCs is well documented. However, use of fungi in industrial scale is not feasible due to their reduced enzymatic activity in real effluent condition. The use of bacterial system for the removal of EDCs from wastewaters has been undertaken because the enzymatic system of bacteria is stable in harsh environmental and physiological conditions.

4.2.4 Bacterial Treatment of EDCs

Biodegradation of EDCs had been well reported by bacterial cultures in wastewater treatment plants (Federle et al. 2002; Singer et al. 2002; Bharagava et al. 2009; Thompson et al. 2005). Among biological processes, bacterial degradation is particularly an easy and prominent way to remove various EDCs present in aquatic environment (Husain and Qayyum 2012; Bharagava and Chandra 2010a). Singer et al. (2002) had reported that approximately 79% of triclosan in wastewater was biodegraded; 15% was sorbed into biosolids, and 6% was released into the receiving water bodies. Several studies have reported removal of triclosan by different biological treatment processes (Kanda et al. 2003), including activated sludge (Federle et al. 2002; Thompson et al. 2005), rotating biological contactors, and trickling filters (Thompson et al. 2005). Unlike triclosan, greater than 90% of removal has been reported for BPA (Staples et al. 1998) and ibuprofen and its metabolites (Buser et al. 1999). Recently, several estrogen-degrading bacteria were isolated from activated sludge, including *Novosphingobium tardaugens* (ARI-1) (Fujii et al. 2002), *Rhodococcus zopfii* and *Rhodococcus equi* (Yoshimoto et al. 2004), and *Achromobacter xylosoxidans* and *Ralstonia* sp. (Weber et al. 2005). *Pseudomonas nitroreducens* strain LBQSKN1, *Pseudomonas putida* strain LBQSKN2, *Stenotrophomonas* sp. LBQSKN3, *Enterobacter asburiae* strain LBQSKN4, *Pseudomonas* sp. LBQSKN5, and *Pseudomonas* sp. LBQSKN6 isolated from soil sample were able to degrade NPs (Qhanya et al. 2017).

The study of Roh et al. (2009) showed *Nitrosomonas europaea* and mixed ammonia-oxidizing bacteria could degrade triclosan, BPA, and ibuprofen in nitrifying activated sludge. In the study of De Gusseme et al. (2009), microbial consortium was used for the removal of 17α-ethinylestradiol (EE2) and was found to remove EE2 from both a synthetic minimal medium and industrial effluent with >94% removal efficiency. Villemur et al. (2013) isolated bacterial cultures from three enrichment cultures adapted to a solid-liquid two-phase partitioning system using Hytrel as the immiscible water phase and loaded with estrone, estradiol, estriol, ethinylestradiol, NP, and BPA. All molecules except ethinylestradiol were degraded in the enrichment cultures. In study of Chen et al. (2007), di-2-ethylhexyl phthalate (DEHP) degradation strain CQ0110Y was isolated from activated sludge and identified as *Microbacterium* sp. The results of this study showed the optimal pH value and temperature, which influenced the degradation rate in wastewater: pH 6.5–7.5, 25–35 °C. The efficacy of two rhizobacteria (*Sphingobium fuliginis* TIK1 and *Sphingobium* sp. IT4) of *Phragmites australis* for the sustainable treatment of water polluted with phenolic endocrine-disrupting chemicals (EDCs) was investigated (Toyama et al. 2013). Strains TIK1 and IT4 have been isolated from *Phragmites rhizosphere* and shown to degrade various 4-alkylphenols–TIK1 via phenolic ring hydroxylation and meta-cleavage and IT4 via ipso-hydroxylation.

The two strains also degraded BPA, BPB, BPE, BPF, BPP, and BPS. Yu et al. (2007) isolated 17α-estradiol-degrading bacteria (strains KC1-14) from activated sludge of a wastewater treatment plant. These isolates were widely distributed among eight different genera – *Aminobacter* (strains KC6 and KC7), *Brevundimonas* (strain KC12), *Escherichia* (strain KC13), *Flavobacterium* (strain KC1), *Microbacterium* (strain KC5), *Nocardioides* (strain KC3), *Rhodococcus* (strain KC4), and *Sphingomonas* (strains KC8, KC11, and KC14) – of three phyla, *Proteobacteria*, *Actinobacteria*, and *Bacteroidetes*. All 14 isolates were capable of converting 17α-estradiol to estrone, but only 3 strains (strains KC6, KC7, and KC8) showed the ability to degrade estrone. Only strain KC8 could use 17α-estradiol as a sole carbon source. In the study of Fernández et al. (2016), five bacteria isolated from enrichment cultures of sediments of mud volcanoes of the Gulf of Cadiz (Moroccan-Iberian margin) were identified as aerobic E2 biodegraders, which produce low amounts of biotransformed estrone (E1). An analysis of 16S rDNA gene sequences identified three of them as *Virgibacillus halotolerans*, *Bacillus flexus*, and *Bacillus licheniformis*. Among the set of strains, *Bacillus licheniformis* showed also ability to biodegrade E2 under anaerobic conditions.

5 Conclusion

This chapter provided detailed information about EDCs releasing into the environment as a result of poor treatment by industrial wastewater treatment plant which leads to environmental pollution and toxicity to human and wildlife health. The conventional wastewater treatment process such as activated sludge process is not designed to remove these micropollutants. Although available treatment technologies, such as adsorption processes, AOPs and membrane processes, and electrochemical methods as promising alternatives are efficient for efficient removal of EDCs, it is not feasible at large scale because of its high operation costs and formation of by-products. Recently, biological treatment process as a promising approach has gained popularity to remove EDCs from industrial wastewaters. The biological system which employed fungi and bacteria having ligninolytic enzymatic system is a significantly useful procedure for targeting a number of EDCs of diversified properties and structures. On the basis of available literature on the effect of EDCs on human and wildlife health and their treatment/degradation process, it seems that there is a need of attention to address the limitation in existing treatment process and provide effective solution on it. Thus, this chapter covers all EDC-associated problems and treatment technology for the sustainable development of environment.

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