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Review

Endocrine Disrupting Compounds Removal Methods from Wastewater in the United Kingdom: A Review

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Abstract: Endocrine disrupting compounds (EDCs) are contaminants with estrogenic or androgenic activity that negatively impact human and animal communities. These compounds have become one of the most significant concerns for wastewater treatment in recent decades. Several studies have evaluated EDC removal methods from wastewater across the globe, including the United Kingdom (UK). Accordingly, the current study reviews EDC removal methods from municipal/domestic wastewater in the United Kingdom (UK) for the period of 2010–2017. The current study analysed original research articles (250), review articles (52), short communication (43), and other associated documents via the ScienceDirect.com database. A total of 25 published articles, which covered EDC removal methods from UK wastewaters, were reviewed rigorously. The research highlights that despite the relative efficacy of existing chemical and physical methods for removing certain EDCs from wastewater, there is emerging evidence supporting the need for more widespread application of nature-based and biological approaches, particularly the use of biofilms. The analysis reveals that there have been relatively few research studies on EDC removal methods carried out in the UK in the 2010–2017 period. Only four papers addressed the removal of specific endocrine disrupting compounds from UK municipal wastewater, and none of the studies addressed EDC removal by using direct biofilms. Finally, this review suggests that more research is needed to remove EDCs, particularly through the application of biofilms, from municipal wastewater in current scenarios.

Keywords: wastewater treatment; temperatures; systematic review; biofilms; endocrine disrupting compounds

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1. Introduction

The occurrence of endocrine disrupting compounds (EDCs) in wastewaters, aquatic systems, and drinking water is considered one of the main environmental problems globally. Endocrine disrupting compounds (EDCs) have gained significant interest in recent decades in the academic press because of the many, serious diseases related to them. Exposure to EDCs is interlinked with decreased fertility, changed sexual behaviour, and amplification of abnormalities and cancers in humans and laboratory animals [1–3]. Kavlock et al. [4] defines EDC as "an exogenous agent that interferes with the synthesis, secretion, transport, binding, action, or elimination of natural hormones in the body which are responsible for the maintenance of homeostasis, reproduction, development and or behaviour". Several hundreds of chemicals may have endocrine disrupting properties [5,6].

More than 100 substances are categorised as potential endocrine disrupters such as: Carbon disulfide, o-phenylphenol, tetrabrominated diphenyl ether, 4-chloro-3-methylphenol, 2,4-dichlorophenol, resorcinol, 4-nitrotoluene, 2,2′-bis(4-(2,3-epoxypropoxy) phenyl) propane, 4-octylphenol, estrone (E1), 17α-ethinylestradiol (EE2), and 17β-estradiol (βE2 [7]. The main EDCs include the following pharmaceuticals: Triclosan, nonylphenol, nonylphenol ethoxylates, octylphenol, octylphenol ethoxylates, bisphenol A, phytoestrogens, and steroid hormones have been identified in wastewater [8], and these compounds also frequently occur in domestic wastewater, industrial wastewaters, and livestock wastes [9–12]. These and other associated EDCs such as: Bisphenol A (BPA), Polychlorinated biphenyls (PCBs), and phthalates, have been identified in human serum, fat, and umbilical cord blood [13–16]. The four important categories of EDCs are: Natural steroidal oestrogens, synthetic oestrogens, phytoestrogens, and various industrial chemicals (xenoestrogens) contaminants have a strong estrogenic strength.

Natural and synthetic oestrogens have greater oestrogenic effects than phyto- and xenoestrogens [17–20]. Nevertheless, the concentrations of phyto- and xenoestrogens in aquatic environments are usually higher. Recent research indicates that several sewage treatment plant effluents and rivers in the UK comprise sufficient quantities of oestrogenic compounds to induce harmful effects on aquatic species [21–26]. The occurrence of EDC pollutants in wastewater creates a huge concern to humans and animals and therefore, the current paper provides an overview of the adverse effects of EDC contaminants and removal methods which were used in the UK for the period of 2010–2017. The research highlights that despite the relative efficacy of existing chemical and physical methods for removing certain EDCs from wastewater, there is emerging evidence supporting the need for more widespread application of nature-based and biological approaches, particularly the use of biofilms. This review helps water and environmental engineers, biologists, chemical engineers, chemists, civil engineers, and microbiologists gain up-to-date knowledge on EDCs in wastewater and removal methods in the UK.

2. Endocrine Disrupting Compounds and Their Impacts

Endocrine disrupting compounds are a type of exogenous endocrine disruptors and encompass natural hormones and synthetic compounds secreted by humans and animals. The majority of EDCs have not been studied completely, analytical approaches for many of the recognised EDCs have yet to be established, and the stages of toxicological consequences or impacts are yet to be conducted. Since 1940, there has been a rapid increase in the number and usage of chemicals for various purposes, and some of those chemicals have been discharged into the environment [15]. This chemical upsurge has changed the environment ecosystems and caused damage to human health and wildlife (Table 1). The first publication on EDCs came out in 1962, which emphasised that Dichlorodiphenyltrichloroethane (DDT) could be accountable for the reduction of bird populations owing to reproductive failure triggered by DDT and other associated harmful chemicals. It is predicted that more than 24% of human disorders and diseases are caused by environmental factors across the globe [27]. While the percentage of urogenital malformations in male babies were raised amongst those exposed to DDT in the developing countries [28,29]. The constant exposure to the DDT in the infanthood and adolescence stages has been identified in the results of substantial reproductive irregularities, cancers, early adolescence, and low pregnancy rates in females [30,31]. Endocrine related disease rates have risen due to the sharp increase in use and application of manufactured chemicals. Since 1970, production of plastics has risen from 50 million to 300 million tonnes today, and the chemical industry has experienced a huge growth in global sales, which has increased from £126 billion in 1970 to over £2.9 trillion in 2013 [32].

Table 1. EDC (endocrine disrupting compound) paths of exposure in humans. Source: Adapted from Gore et al. [15].

How Humans Are Exposed to EDCs	EDC Source	Examples of EDC
Consumption of contaminated water or food	Industrial wastewater or ground water	Polychlorinated biphenyls (PCBs), dioxins, perfluorinated compounds, Dichlorodiphenyltrichloroethane (DDT)
Consumption of contaminated water or food	Discharge of chemicals from food or beverages	Bisphenol A (BPA), phthalates, chlorpyrifos, DDT
Contact with skin or inhalation	Household furniture treated with flame retardants	Brominated flame retardant (BFR)
Contact with skin and/or inhalation	Pesticides used in agriculture, homes, or for public disease	DDT, chlorpyrifos, vinclozolin, pyrethroids
Application to skin	Vector control, certain cosmetics, and personal care products	Parabens, phthalates, insect repellents triclosan
Biological transfer from mother's milk	Maternal body burden due to past and or/current exposures	Several EDCs are found in breast milk

Snyder (2003) [18] found that oestrogenic compounds in drinking water are not responsible (highly likely) for adverse human health effects because of the relatively small amounts of oestrogenic content in water compared to the quantity in foods. However, exposure to EDCs for humans is distinct from aquatic organisms such as fish, therefore, an equivalent hormonal response might not be anticipated [33,34]. Some of the EDC have a long life span, and these will be harmful to humans and wildlife. The substances of these EDC do not decay over a certain period, and there is a chance they fragment into more complex toxic elements than the initial particles. Moreover, some of the EDC substances which were banned several decades ago still remain in significant amounts in the environment, and these particles are not easily detectable in aquatic animals and other organisms [35,36]. EDCs also cause abnormalities in fish reproductive systems and stimulation of vitellogenin synthesis in fish plasma, and can also significantly reduce (nearly 90%) the production of fertile eggs of sand goby fish [37]. In summary, various natural and synthetic chemical compounds have been identified that induce estrogenic responses, and several studies specify that EDCs are ubiquitous in diverse media, and their probable risks to humans and lab animals are very high. Therefore, EDCs are of concern worldwide because of their wide range of negative impacts on the environment. EDCs have complex molecular structures and distinctive biological systematic mechanisms and as a result, cannot be effectively removed by sewage treatment plants (STPs) [38], but may effectively be processed in wastewater treatment plants.

3. Wastewater Treatments and EDCs Removal Procedures

Several types of wastewater treatment plants (WTPs) have been proven to eliminate significant quantities of several EDCs from the wastewater. However, low concentrations of EDCs in wastewater can still lead to in-stream concentrations that are of significant quantities to harm aquatic organisms [39,40]. The toxic consequences of many EDCs are not fully known and require further scrutiny. The actual amount experienced by aquatic organisms is dependent upon the amount of water available for dilution in the receiving stream. Based on EDCs physicochemical characteristics, EDCs can be removed from waste water via various methods such as: Absorption, adsorption, chemical degradation, biological degradation, transformation, and volatilisation [41,42]. Several studies reported that elimination efficiency via wastewater treatment differs significantly depending on the kind of element and removal procedure [43–45]. Cutting-edge research specifies that endocrine active substances, which are primarily found in domestic sources, were more vulnerable to dissolution and removal. With other types of contaminants, very little reduction may occur through the WTP. If these harder chemicals must be eliminated, application of advanced wastewater treatment technologies may be needed [46,47]. Thus, the technology applied at any given plant must be based on a thorough understanding of wastewater constituents.

Table 2 specifies that some of the EDC compounds undergo substantial degradation as a result of biological treatment methods, predominantly nitrifying structures with longer SRTs. While sand filtration or microfiltration act to eliminate 17β-estradiol and/or 17α-ethinylestradiol with significant efficiency; more advanced treatments like reverse osmosis, offer considerably greater removal rates.

Table 2. Kinds of treatment methods and removal efficiencies for selected EDCs. Source: Adapted from Birkett and Lester (2003) [41].

Compound	Kinds of Method	Removal Efficacy
Polychlorinated biphenyls (PCB)	Biofiltration Activated sludge Biofiltration/activated sludge	90% 90% 99%
Nonylphenol (NP)	High loading/non-nitrifying Low loading/nitrifying	37% 77%
NP1EO **	High loading/non-nitrifying Low loading/nitrifying	−3% degradation product produced 31%
NP2EO **	High loading/non-nitrifying Low loading/nitrifying	−5% produced as degradation product 91%
17β-oestradiol/17α-Ethinylestradiol	Filtration—Sand/microfiltration Advanced treatment—Reverse osmosis	70% 95%
Organotins	Primary effluent Secondary effluent Tertiary effluent	73% 90% 98%

Several researchers investigated the removal of EDCs from wastewater through biological wastewater treatment procedures. Findings from research efforts across the globe such as: [10,48–57]; explored a range of removal outcomes for different kind of chemicals. However, it is important to note the methodological limitations with the above research. Firstly, treatment circumstances and objectives, such as SRT, temperature, pH, nitrification, denitrification, and bio-P, are often not adequately defined by investigators. These issues can have an important influence on EDC removal rate at wastewater treatment plants. Secondly, the sampling collection approach and analysis processes may impact the results significantly (Table 3) [39].

Table 3. EDCs removal methods and explanation. Adapted from Johnson and Sumpter (2001) [39].

Removal Method	Explanation
Activated Carbon Adsorption	Activated carbon cost-effectively removes hydrophobic organic compounds. Activated carbon is usually applied in one of two methods: (1) Powdered activated carbon (PAC) and (2) granular activated carbon (GAC).
Ozonation	Ozone is the dominant oxidant. Ozonation removes trace elements: However, this method will not work efficiently in some circumstances.
Advanced Oxidation Processes (AOPs)	AOPs have strong oxidants, degrade strong organic pollutants, and remove certain inorganic pollutants in wastewater.
Reverse Osmosis (RO)	RO can remove EDCs based on compound magnitude and membrane properties. However, RO is a less appropriate option for wastewater treatments for drinking and riverine waters.

Ternes et al. (1999) and Korner et al. (2001) [58,59] revealed that trickling filters (TF) were less effective in the removal of the oestrogenic content from influent wastewater than activated sludge, as evidenced from two waste water treatment plants in the U.S.

Activated carbon arborisation is also a very expensive process because of the cost of raw materials, costly distillation, and significant capital costs [60]. However, in activated sludge procedures, hydraulic residence time (HRT) and SRT are particularly significant factors in the EDC removal process. In European activated sludge systems, the HRT is between four and fourteen hours [39], and this method provided better results than trickling filters. Several scientists have noticed that EDC removal with increased SRT is more efficient [61–65]. While microfiltration membranes on their own would not support an efficient removal of EDC, it was proposed that EDC adsorption of particulate matter, that is preserved by the membrane, would condense EDC intensity in the effluent. Other researchers [66–69] revealed that EDC removal was mainly due to biodegradation or membrane biofilms. Usually, biological procedures are the most cost effective in removal of organic EDS from wastewater, however, when these organics are contaminated or non-biodegradable, physical and chemical techniques must be implemented [70–73]. These removal approaches comprise adsorption, chemical oxidation, and membrane processes. Research on advanced EDC removal procedures is being studied across the world. The following approaches are examples of innovative and traditional technologies that are appropriate for full-scale application if ultra-low EDC concentration limits are required.

4. Methodology

Recently published literature on EDCs and wastewater treatment allows the examination of trends in EDCs removal methods from wastewater subjects, particularly with more advanced and innovative solutions in recent years. Literature searches also help to examine the current developments in a particular research field and assist in recognising research gaps and new challenges. Current research articles about the EDCs and their effects on human and animals and EDC removal methods from the UK wastewater were reviewed. Accordingly, Sciencedirect.com, a research article search engine, has been used to evaluate the research articles which were published between 2000–2017 on EDC removal from municipal/domestic wastewater in the United Kingdom. Therefore, suitable keywords such as: EDC and impacts, EDC in wastewater, and EDC removal from municipal wastewater in the UK were searched for using Sciencedirect.com; narrowing the searching period for the 2000–2017 years for critical analysis. Subsequently, the current review analysed original research articles, book chapters, conference papers, abstracts, editorial notes, short communication, and other associated documents. In the first stage of the review, the current study identified almost 250 original research articles, 52 review articles, 43 short communications, and 11 technical reports. From those, a total of 47 published articles, which covered EDC removal methods from wastewaters/municipal waters were reviewed rigorously. From those, a total of 25 published articles, which covered EDC removal methods from UK wastewaters were reviewed more thoroughly. Subsequent analysis established the number of different methods for EDC removal in the UK for the period of 2010–2017. The current review also evaluated how many studies directly addressed EDC removal from wastewater treatments through the use of biofilms.

5. Results and Discussion

Significant research results obtained through the analysis of diverse types of published articles were scrutinised via the ScienceDirect.com database. There were a very limited number of original research articles (24) published for the period of 2010–2017 on EDC removal methods from wastewater in the UK. Only four papers addressed the removal of specific endocrine disrupting compounds from UK municipal wastewater, and none of the studies addressed EDC removal by using direct biofilms. Below are the two main papers which have been published in the UK for the period of 2010–2017, which are now examined in more detail.

5.1. Research Article-1

This research examined the role of polyamide-6 for the removal and recovery of the oestrogenic endocrine disruptors estrone, 17β-oestradiol, 17α-ethinylestradiol, and the oxidation product 2-hydroxyestradiol in water. Tizaoui et al. (2017) [74] described the removal of endocrine disrupting compounds such as oestrone (E1), 17β-oestradiol (E2), and 17α-ethinylestradiol (EE2), and the oxidation product 2-hydroxyestradiol (2OHE2) from secondary treated wastewater in Wales (United Kingdom) using polyamide 6 as a sorbent material in their study. Results explored that PA6 is an effective sorbent material for the removal and recovery of EDCs from different water environments. Hydrogen bonding is the main tool compelling the adsorption of these EDCs on PA6 at pHs less than the EDCs pKas (~10.5) (Figure 1), and their adsorption was not disturbed by the water matrix. In addition, PA6 was also an efficient solid phase extraction sorbent.

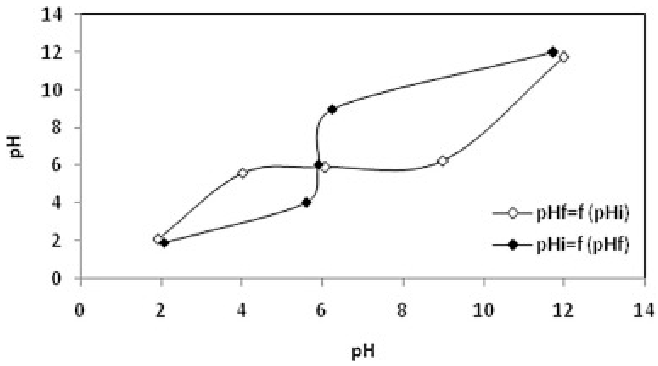

Figure 1. Correlation of the PH to the point of 0 charge of PA6.

5.2. Research Article-2

This research examined the effectiveness of anaerobic digestion in removing oestrogens and nonylphenol ethoxylates. Paterakis et al. (2011) [75] investigated the two groups of endocrine disrupting compounds such as steroid oestrogens and nonylphenol ethoxylates, appraised under mesophilic and thermophilic circumstances during the anaerobic digestion of primary and mixed sewage sludge (Table 4) where digestion occurred more than six retention times. The research is based on the treatment of sludges in United Kingdom wastewater treatments plants. Results explored sludge intensities of both groups and proved that temporal variants and concentrations in mixed sludge are impacted by the occurrence of waste activated sludge. The biodegradation of steroid oestrogens is more than 50% during initial sludge digestion. It is evident that anaerobic digestion diminishes the intensity of these compounds. This study also highlighted that anaerobic digestion shields the reuse environment from steroid oestrogens and Nonylphenol Ethoxylates (NPEO'S), and that eliminations of NPEOs are greater in the absorption of mixed sludge.

Table 4. Sludge features, operational circumstances and digester enactment at mesophilic and thermophilic temperatures at six retention spells. Adapted from Paterakis et al. (2011) [75].

	Mesophilic		Thermophilic	
	Initial Sludge	**Mixed Sludge**	**Initial Sludge**	**Mixed Sludge**
Influent Sludges				
TS (g L^{-1})	51.1 ± 3.7	57.1 ± 4.3	39.5 ± 0.1	49.7 ± 0.1
VS (g L^{-1})	36.5 ± 2.6	44.0 ± 3.0	29.2 ± 0.1	38.1 ± 0.1
VFA (mg acetic acid L^{-1})	1314 ± 68	1592 ± 44	1168 ± 98	1470 ± 52
Estrone (E1) (μg kg^{-1} dw)	158 ± 14	90 ± 21	64.3 ± 2.5	32.3 ± 2
17β-Oestradiol (E2)	9 ± 1	6 ± 1	6 ± 3	3 ± 2
Estriol (E3)	9 ± 1	8 ± 1	6 ± 1.5	5 ± 1
Estrone-3-sulfate (E1-3S)	7.6 ± 1.5	7 ± 1.5	4 ± 1	4 ± 1
17α-Ethinyl oestradiol (EE2)	18 ± 4	10 ± 2	9 ± 1	10 ± 2
4-Nonylphenol (NP) (mg kg^{-1} dw)	0.3 ± 0.1	0.23 ± 0.1	0.23 ± 0.1	0.1 ± 0.1
Nonylphenoxy acetic acids ($NP_{1-3}EC$) (mg kg^{-1} dw)	26.5 ± 0.1	241.5 ± 0.1	0.1 ± 0.1	0.08 ± 0.1
Nonylphenol monoethoxylate and diethoxylate ($NP_{1-2}EO$) (mg kg^{-1} dw)	2.1 ± 0.5	1.7 ± 0.5	15 ± 0.1	90 ± 0.1
Nonylphenol polyethoxylates ($NP_{3-12}EO$) (mg kg^{-1} dw)	1.5 ± 0.4	0.7 ± 0.4	1.3 ± 0.25	0.7 ± 0.25
Operating Circumstances				
T (°C)	35 ± 0.2	35 ± 0.2	55 ± 0.2	55 ± 0.2
SRT (d)	30	30	15	15
OLR (kg VS m^{-3} d^{-1})	1.3 ± 0.1	1.5 ± 0.1	1.9 ± 0.0	2.5 ± 0.0
TS (g L^{-1})	26.7 ± 2.3	38.5 ± 1.3	22.7 ± 1.8	33.9 ± 1.3
VS (g L^{-1})	19.5 ± 1.6	23.9 ± 2.0	11.5 ± 4.5	22.0 ± 2.2
pH	7.1 ± 0.1	7.5 ± 0.1	7.2 ± 0.0	7.6 ± 0.1
ORP (mV)	−320.8 ± 12.8	−380.6 ± 29.8	−411.6 ± 36.9	−419.0 ± 34.9
VFA (mg acetic acid L^{-1})	176.4 ± 7.3	132.9 ± 17.3	1098.5 ± 189.6	829.3 ± 145.9
Total alkalinity (mg L^{-1})	2399 ± 37	5362 ± 63	4000 ± 453	4770 ± 85
Biogas				
Daily production (l d^{-1})	0.8 ± 0.0	0.8 ± 0.1	1.0 ± 0.1	1.6 ± 0.1
GRP (m^3 m^{-3} d^{-1})	0.51 ± 0.0	0.52 ± 0.0	0.67 ± 0.0	1.08 ± 0.0
SGP (m^3 CH_4 kg^{-1} $VS_{removed}$)	0.7 ± 0.1	0.6 ± 0.1	0.4 ± 0.1	0.7 ± 0.1
Biogas yield (m^3 kg^{-1} $VS_{removed}$)	0.95 ± 0.2	0.80 ± 0.1	0.60 ± 0.1	1.02 ± 0.1
Elimination efficiencies (%)				
VS	53.5 ± 6.9	40.1 ± 2.1	43.2 ± 3.0	32.4 ± 1.0
TS	47.3 ± 8.5	33.7 ± 4.6	37.0 ± 4.4	29.8 ± 2.6
g $VS_{removed}$ d^{-1}	1.07 ± 0.1	0.98 ± 0.1	2.24 ± 0.2	1.84 ± 0.1

These are the two main papers written by various researchers on the removal of EDC compounds in wastewater in the UK. Furthermore, several articles addressed EDCs, but not in relation to the removal of UK wastewater.

6. Conclusions

The current review presents endocrine disrupter removal methods which were established for the period of 2010–2017 from wastewater in the United Kingdom. Accordingly, the current study analysed original research articles (250), review articles (52), short communication (43), and other associated documents via the ScienceDirect.com database. A total of 25 published articles, which covered EDC removal methods from UK wastewaters, were reviewed rigorously. From those articles, two key research articles have been discussed in more depth. Analysis results explored that research on EDCs removal methods in the UK is extremely sparse and existing research addressed only limited endocrine disrupters such as bisphenol A and 4-tert-octylphenol. Most of the existing research evaluated physical and chemical methods rather than biological methods. Several published articles that explored the removal rate efficiency (70–90%) of EDCs by using chemical and physical methods are good, but concluded that biofilms might be the better opportunity. Some other researchers identified that some advanced treatments such as Advanced Oxidation Processes (AOPs), Reverse Osmosis (RO), and Activated Carbon Adsorption (ACA) are better methods for the removal of EDCs from wastewater however they are expensive. Finally, this research concluded that very limited research had been undertaken on EDCs removal from wastewater in the UK, and more research is needed in the near future.

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