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Review article

Occurrence and fate of emerging contaminants in water environment: A review

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ABSTRACT

Emerging contaminants (ECs), such as personal care products (PCPs), endocrine disrupting compounds (EDCs), pharmaceuticals (PhACs) and their transformation products,whose occurrence at trace levels in treated wastewater is of concern for human health and the aquatic ecosystem. Due to the relatively new introduction or detection of these pollutants, there exists a gap in the knowledge on their fate, behaviors and effects, as well as on treatment technologies for their efficient removal. Furthermore, despite the advances in treatment technologies, the design of existing treatment plants are not suited to remove these ECs, in addition to there being a lack of published health standards that provide guideline in treating these pollutants. Many new ECs are being introduced into the environment without detection. In these context, this paper reviews existing research that provide reliable and quantitative information on pharmaceuticals, PCPs and EDCs and their concentrations in surface water, ground water, drinking water and treated wastewater and the removal efficiency of different treatment processes for different emerging pollutants, with a focus on recent studies regarding the fate and behavior of the contaminants in wastewater treatment plants and in the environment as well. The paper also highlights various biological and chemical treatment techniques and their drawbacks. Also, this review discusses the scope of future research on ECs.

1. Introduction

In the recent decades, studies on wastewater characteristics have drawn attention towards the environmental occurrence of a variety of newly identified compounds of anthropogenic origin. The occurrence of such trace compounds (mostly organic), known as the "*emerging pollutants*" and their harmful impact on both aquatic and terrestrial life forms as well as on human health is now an issue of concern among the scientists, engineers, and the general public as well. The non-regulated organic trace pollutants, known as emerging micro-pollutants, have been recently introduced or newly detected with the help of advanced analytical technologies (Richardson, 2007). A contaminant whose new origin, alternate route to humans or new treatment techniques has been innovated is termed as "emerging". They are categorized by apprehensible, probable or actual risk to human health and environment (DoD, 2011; US EPA, 2012). They may be industrial in origin or may originate from municipal (domestic), agricultural, hospital or laboratory wastewater. In large part, the compounds in question are derived from three broad categories, viz.

a) Pharmaceuticals (PhACs)
b) Personal Care Products (PCPs) and
c) Endocrine Disrupting Compounds (EDCs).

But they are not confined to the above and may comprise of nanomaterials (NMs), metabolites of ECs, illegal drugs, engineered genes, etc. NMs affect the bacterial biomass during waste water treatment and thereby decrease their biological activity leading to decrease in EC removal efficiency (Wang et al., 2012). ECs are present and have been found in surface water, ground water as well as drinking water and in

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wastewater treatment plants (WWTP) discharge (Samaras et al., 2013; Yang et al., 2014; Cabeza et al., 2012). Municipal wastewater is viewed as one of the principle discharge sources for the emanation of emerging contaminants like non-point and point sources, industries and storm water, wastewater from households and water treatment facilities into the environment (Ternes et al., 2004). Also there is a growing concern of sludge management due to high levels of ECs in them (Wu et al., 2010). The design of the current WWTP could not restrict the elimination of the emerging contaminants and their metabolites where it is released into rivers or streams having high biodiversity as sewage effluents. So far, considerable work have been done in regard to the performance of wastewater technologies in case of nutrient removal (Molinos-Senante et al., 2012; Daughton and Ternes, 1999; Heberer, 2002; Barcelo, 2003; Daughton, 2004; Petrovic et al., 2009), while there is an absence of data on the ability to removal of ECs, and additionally on the adverse ecotoxicological impacts of these compounds on surface water bodies.

Pharmaceutical molecules identified in the wastewater belong to several classes of human and veterinary antibiotics, human prescription and non-prescription drugs, and some sex and steroid hormones as well. Personal care products (PCPs) include chemicals found in consumer products (e.g. galaxolide, tonalide). Endocrine disrupting compounds (EDCs) can elicit adverse effects on endocrine systems as they have androgenic or estrogenic activities even at low concentrations (Fig. 1). Potential concerns from the environmental presence of these emerging contaminants include abnormal physiological processes and reproductive impairment, increased incidences of cancer, the development of antibiotic-resistant bacteria, and the potential increased toxicity of chemical mixtures.

ECs are generally not monitored in our environment and they are not regulated in our drinking water (Oviedo and Aga, 2016). The present gap of knowledge holds specifically for incessant impacts that have just seldom been explored. Whilst significant amount of human medicines/ pharmaceuticals are discharged into the environment, succinct controls for ecological risk assessment are to a great extent missing. Prior to their discharge, efficient treatment of wastewaters is required. Thus, the occurrence of emerging contaminants at trace level in wastewaters, their behavior during wastewater treatment and drinking water production are the key issues that require further study. Therefore, this review presents an overview on the state-of-art as regards ECs of concern such as PhACs, PCPs and EDCs. Our objective is to obtain reliable and quantitative information on PhACs, PCPs and EDCs concentrations and their removal efficiency. We also reviewed an account for removal efficiency of the different treatment processes for different PhACs, PCPs and EDCs. Focus has been given on recent studies regarding the fate and behavior of the contaminants in WWTP and in the environment as well.

1.1. Emerging contaminants

1.1.1. Pharmaceuticals (PhACs)

PhACs are a set of developing ecological contaminants that are broadly and progressively being utilized as a part of human and veterinary medication. They include compounds of environmental concern like antibiotics, legal and illicit drugs, analgesics, steroids, beta-blockers, etc. (Fatta-Kassinos et al., 2010). Their persistence in the body occurs due to their specific mode of action. They have been detected in WWTP effluents, sludge, sediments, natural waters, drinking water and groundwater. They are supposed to provoke the development of antibiotic resistant genes in soil bacteria. Nowadays, active pharmaceutical ingredients (APIs) and their biotransformation products, which are largely unstudied, are bioaccumulating and causing significant consequences to ecosystem (Arnold et al., 2013). Although these compounds have been entering the environment for many years, but investigation on their adverse effects on aquatic organisms have started only recently. They are considered as pseudo-persistent pollutants, which continually enter the environment at very low concentrations. A large number of more than 160 different pharmaceuticals have as of now been detected in aquatic systems in very low concentrations of ng L^{-1} to low μg L^{-1} range (Kummerer, 2010). There is very little knowledge about the eco-toxicological impacts of pharmaceuticals on terrestrial and aquatic life forms and a complete analysis eco-toxicological impact is inadequate. One of the critical targets is the aquatic organisms, as they are subjected to wastewater remnants over their entire life (Fent, 2003).

1.1.2. Personal care products (PCPs)

PCPs are yet another class of emerging contaminants that

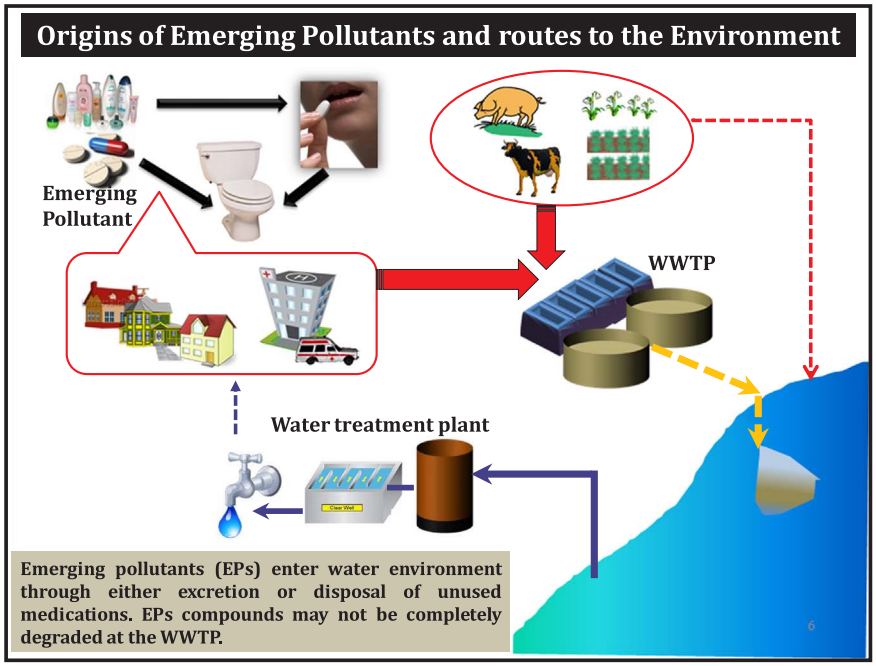

Fig. 1. Conceptual depiction of the origin of emerging pollutants (EPs) and their route to the environment.

incorporate prescribed and non prescribed veterinary and human pharmaceuticals and the agile and inert elements for individual care purposes. A few PCPs to name are cosmetic products, engineered hormones, steroids, perfumes, shampoos, etc. UV filters, known to have estrogenic activity, are reported to be one of the most commonly found PCPs in ground water and other aquatic environment (Jurado et al., 2014; Oviedo and Aga, 2016). PCPs are released into wastewater and advance toward WWTPs, in their native or biologically transformed structures. In WWTPs, the likely fates of PCPs and their metabolites are conversion to CO_2 and water; mixing with the receiving water bodies either as the original or mineralized product; and sorption by the solids like sludge/biosolids, mainly if the compound or the biologically moderated transformation product is lipophilic.

1.1.3. Endocrine disruptors (EDC_S)

EDCs are characterized as the artificial chemicals that, when ingested into the body can either copy or obstruct hormones and effect body's normal functioning. The Environmental Protection Agency (EPA) characterizes EDCs as external agents that meddle with the formation, release, transport, attachment, activity, or displacement of body's natural hormones that maintains homeostasis, development, reproduction and behavior (United States Environmental Protection Agency (USEPA), 1997). It is by and large acknowledged that the three main classes of EDCs are estrogenic i.e. they mimic or alters the functioning of natural estrogens, androgenic (copy or obstruct natural testosterone) and thyroidal (causes immediate or oblique effects to the thyroid) (Snyder et al., 2003). Natural and engineered EDCs are discharged into the environment by human activities, creatures and industries; essentially through sewage treatment systems before finally going to soil, surface water, silt and ground water. Preponderance research has concentrated just on estrogenic compounds. EDCs are present in immensely low concentrations (ng L^{-1} or μg L^{-1}) in wastewater. These compounds are of profound concern as their long term exposure and adverse impact on human are unknown.

2. Environmental/health issues and regulations related to emerging contaminants

Due to the absence of relevant data on the impacts, fate and concentration levels of emerging contaminants make it troublesome for governments to control their utilization and also manage the levels that are already persisting in the environment. There are as of now no laws or mandates illustrating the upper limits of concentrations of emerging contaminants in wastewater discharge, drinking water, or the environment. In United States, a preparatory observing technique was organized and an archive was communicated illustrating the preparatory way to deal with EDCs and to decrease its intrusion in people and wildlife (European Commission, 2011). This document concentrates on the diminishment of the utilization of EDCs in consumer products, sustenance added substances, and beauty care products however does not set any proposals for uttermost permissible concentrations in drinking water, wastewater and in nature (European Commission, 2011). The European Union Water Framework Directive (EC, 2013) listed 45 priority compounds with environmental quality standard (EQS) to be respected in aquatic environments and listed 10 others on contemporary watch list (Decision 2015/495, published on 24th march 2015). Similar regulations were followed by Switzerland for several ECs. In 1995, the European Union (EU) set 10 ng L^{-1} and 10 μg kg^{-1} as the concentration of PhACs and PCP_S in surface water and soil. The U.S. Food and Drug Administration (FDA, USA) publicized directions for the evaluation of human drugs. Environmental assessment has reported the expected introduction concentration of pharmaceuticals in the aquatic environment as ≥1 μg L^{-1} (US FDA, 1998). EAWAG Institute, Switzerland proposed similar to EQS environmental quality criteria for several ECs such as pharmaceuticals and hormones (PPHs) and pesticides (Kase et al., 2011). Many disinfection byproducts that are transformation products of ECs after treatment are regulated by the US, European Union and World Health Organization (WHO) (Oviedo and Aga, 2016). But in Canada and India, there is no such regulation for ECs.

The Water Framework Directive included anti-inflammatory diclofenac or the synthetic hormones Ethynylestradiol (EE2) in the supposed 'watch list' of priority compounds to address the risk posed by these substances (Collado et al., 2014; European Commision, 2013). Various PhACs and EDCs, were enlisted in the Drinking Water Contaminant Candidate List (US EPA, 2012). Different PhACs, for example, carbamazepine, naproxen, sulfamethoxazole, ibuprofen, gemfibrozil, atenolol, diclofenac, erythromycin and bezafibrate have been rated prime concern pharmaceuticals to the water cycle by the Global Water Research Coalition (Global Water Research Coalition, 2008).

Contaminants from PhACs, PCPs and EDCs enter water bodies and can exceed and persist beyond acceptable levels. The widespread occurrence of ECs in water has high probability of their incorporation in crops irrigated with contaminated water and posses risk to human health upon consumption (Hurtado et al., 2016). ECs can cause harmful impacts on aquatic and terrestrial wildlife and human communities. Table 1 summarizes the adverse effects of ECs in the environment. Endocrine disrupting chemicals cause a number of reproductive and sexual abnormalities in wildlife and humans. Subjection to these chemicals amid pre and postnatal life, can impair the development and signaling of the endocrine system. The effects during development are permanent and sometimes irreversible. Managing ECs in water resources is a critical issue that requires attention especially in sensitive ecosystems and in rapidly developing areas. However, the ecological effects of ECs in natural environment are different from laboratory tests. When present in the environment many factors like pH, soil or water type, ionizable compounds, etc. can influence the bioavailability of the ECs (Nichols et al., 2015; Du et al., 2015). There is a need for a comprehensive framework that aims at system-wide abatement (source-transfer-fate levels) using both structural and non-structural approaches.

3. Emerging contaminants in wastewater

3.1. Occurrence (sources) and concentration

Municipal wastewater treatment plants (WWTPs) are by and large not furnished to manage complex pharmaceuticals, as they were constructed and updated chiefly with the intension to eliminate effortlessly or modestly biodegradable carbon, phosphorus and nitrogeneous substances and microbes, which consistently appear at the WWTP in μg L^{-1} levels (Verlicchi et al., 2012). Parent human pharmaceuticals or their metabolites enter the aquatic systems through WWTPs and PCPs (e.g. perfumes) are released through shower waste.

Countries like USA (Boyd et al., 2004), Japan (Nakada et al., 2006), United Kingdom (Ashton et al., 2004), Finland (Lindqvist et al., 2005) and Spain (Carballa et al., 2004) had documented the presence of PhACs and PCPs in concentrations of ng L^{-1} to μg L^{-1} in WWTPs. Nowadays engineered nanomaterials from consumer products are entering the environment which is seldom detected but can have inimical affects (Bour et al., 2015). In March 2015 a total of 1814 consumer products with nanomaterials were included in an inventory (Vance

Table 1
Environmental effects of pharmaceuticals, PCPs and EDCs.

Chemical	Adverse effect	Reference
Penicillin, sulfonamides, tetracylines (Antibiotics)	Cause resistance among bacterial pathogens, that lead to altered microbial community structure in the nature and affect higher food chain	Witte (1998) Daughton and Ternes (1999)
Roxithromycin, clarithromycin, tylosin (Antibiotics)	Growth inhibition of algae (*Pseudokirchneriellasubcapitata*)	Yang et al. (2008)
Caffeine (Stimulant drug)	Endocrine disruption of goldfish (*Carassiusauratus*)	Li et al. (2012)
Diclofenac (Nonsteroidal anti-inflammatory drug)	Renal lesions and gill alterations of rainbow trout (*Oncorhynchusmykiss*)	Schwaiger et al. (2004)
Carbamazepine (Antiepileptic drug)	Oxidation stress of rainbow trout (*Oncorhynchusmykiss*)	Li et al. (2010)
Gemfibrozil (Blood lipid regulator)	Growth inhibition of algae (*Anabaena sp.*)	Rosal et al. (2009)
Propranolol (β-blocker)	Reduction of viable eggs of Japanese medaka (*Oryiaslatipes*)	Huggett et al. (2002)
HHCB (Synthetic musk)	Oxidation stress of goldfish (*Carassiusauratus*)	Chen et al. (2012)
Fragrances (musk)	carcinogenic to rodents, easily absorbed by human skin and may damage the nervous system	Bronaugh et al. (1998) Kirschner (1997)
Triclosan and triclocarban (Antimicrobial agents)	Growth inhibition of algae (*Pseudokirchneriellasubcapitata*)	Yang et al. (2008)
Bisphenol A (BPA) (Endocrine disrupting compound)	Proven to have estrogenic effects in rats and hormonal effects which increase breast cancer risk in human Reported to act as anti-androgen that causes feminising side-effects in men.	Dodds and Lawson (1938) Krishnan et al. (1993) Sohoni and Sumpter (1998)
Estrone and 17-β estradiol (steroidal estrogens) and 17-α ethynylestradiol (synthetic contraceptive) – contained in contraceptive pills	Feminisation in fishes, mimics as estrogen hormone to non-targets	Witte (1998)
Preservatives, i.e., parabens (alkyl-phdroxybenzoate) – used for anti-microbiological preservatives in cosmetics, toiletteries and even foods	Shows weak estrogenic activity	Routledge et al. (1998)
Disinfectants/antiseptics,.i.e., (triclosan – used in toothpaste, hand soaps, acne cream)	Acts as toxic or biocidic agent and cause of microbial resistance	Okumura and Nishikawa (1996) McMurry et al. (1998)

et al., 2015). Table 2 describes the concentrations of some emerging contaminants in various environmental media. Studies also report that the effectiveness of removal of ECs by traditional wastewater treatment techniques such as sedimentation, flocculation, and active sludge treatment is low (Castiglioni et al., 2006; Lishman et al., 2006; Paxeus, 2004; Santos et al., 2007).

The primary origin of steroidal hormones in aquatic environment is human and animal defecation. In the long run the natural and engineered hormones and their metabolites finally reach WWTP. Hormone replacement therapy (HRTh) along with oral dose of progesterone, estrogens and at times testosterone, can add to the absolute estrogenicity of municipal wastewater. The conceivable removal course of the hormones from various treatment techniques can be categorized under four procedures, viz. abiotic degradation, biological degeneration, volatilization and adsorption onto solids (Hamid and Eskicioglu, 2012).

3.2. Fate of ECs during wastewater treatment

The traditional wastewater treatment systems for the most part comprises of an primary treatment, secondary treatment and occasionally a tertiary step, with various biological and physicochemical procedures accessible for every phase of the treatment. In the primary treatment the solid waste substances of the wastewater such as settleable solids, plastics, oils and fats, sand and grit, etc., are separated. This method is common for almost all Urban wastewater treatment plant (UWTP) and is accomplished mechanically by filtration and sedimentation. Nonetheless, the secondary treatment, which normally depends on the biological (aerobic or anaerobic) degradation of organic substances or nutrients, can vary significantly. Among the various biological treatment techniques e.g. fixed bed bioreactors (FBR), Membrane bioreactors (MBR), moving bed biofilm reactor (MBBR) etc., used in UWTPs, the most well-known technique is conventional activated sludge(CAS). Organic substances and nitrogen are removed under certain conditions by activated sludge plants through the formation of biological floc utilizing dissolved oxygen. Lastly in the tertiary treatment phosphorous can be removed by precipitation and filtration (Batt et al., 2007). Also some UWTPs disinfection of the effluent is done by UV irradiation or chlorination, before discharging them in the environment. But these treatments alone do not ensure complete removal of the ECs. The most common activated sludge technique which is used worldwide cannot remove all PPCPs efficiently and entirely e.g. diclofenac and carbamazepine that are resistant to biodegradation (Celiz et al., 2009). Moreover, various processes like biological and chemical degradation and photolysis may transform ECs into forms that can be more toxic than their parent compound. Partial oxidation of PhACs leads to generation of transformation products (TPs) with more ecotoxicity such as N-(4-carbamoyl-2-imino-5-oxoimidazolidin)-formamido-N-methoxyacetic acid(COFA) and carboxy-acyclovir are the two TPs of the antiviral drug acyclovir that are more harmful than

Table 2
Concentrations of some emerging contaminants in various environmental samples.

Emerging contaminants	Treated wastewater	Surface water	Ground water	Drinking water
Analgesics and anti-inflammatory	60 μg/L[a]	5 μg/L[o]	–	0.12 μg/L[h]
Lipid regulators	5 μg/L[p]	0.2 μg/L[o]	7.5 μg/L[e]	0.17 μg/L[h]
β blockers	9 μg/L[i]	2 μg/L[o]	–	0.27 μg/L[m]
Antibiotics	6 μg/L[l]	1.9 μg/L[j]	0.2 μg/L[e]	–
Antiepileptic drugs	22 μg/L[d]	1.8 μg/L[j]	1.1 μg/L[p]	0.05 μg/L[m]
Estrone (E1)	< 0.1–19 ng/L[b]	< 0.1–17 ng/L[f]		0.20–0.60 ng/L[o]
17β-Estradiol (E2)	< 0.1–650 ng/L[k]	< 0.1–6.0 ng/L[f]	13–80 ng/L[e]	0.20–2.1 ng/L[e]
Estriol (E3)	5.0–7.3 ng/L[g]	1.0–2.5 ng/L[f]		
Bisphenol A	4.8–258 ng/L[c]	0.5–250 ng/L[n]	3–1410 ng/L[m] Drinking water well	0.50–44 ng/L[e] Groundwater well

[a] Alder et al. (2006).
[b] Karthikeyan and Meyer (2006).
[c] Loraine and Pettigrove (2006).
[d] Klaus Kummerer (2001).
[e] Wicks et al. (2004).
[f] Dorabawila and Gupta (2005).
[g] Kolodziej et al. (2004).
[h] Kuch and Ballschmiter (2001).
[i] Braga et al. (2005).
[j] Cargouet et al. (2004).
[k] Heisterkamp et al. (2004).
[l] Korner et al. (2000).
[m] Pawlowski et al. (2003).
[n] Suzuki et al. (2004).
[o] Williams et al. (2003).
[p] Rudel et al. (1998).

acyclovir (Schlüter-Vorberg et al., 2015). Disinfection byproducts (DBPs) are yet another type of TP that is formed when organic matter present in water reacts with disinfection agents like ozone, chlorine, etc. Today more than 600 DBPs are reported and the number is ever rising (Richardson, 2011).

4. Effect of treatment technologies on the removal of emerging pollutants

4.1. Aerobic processes

4.1.1. Activated sludge process

Table 3, summarizes the removal efficiencies of PhACs, PCPs and EDCs by different treatment technologies in environmental media. In 1999, Germany, Canada and Brazil studied the removal of EDCs by activated sludge process in which they were analyzed by GC-MS/MS. The removal efficiency for estrone (E1), 17β-estradiol (E2), and 17α-ethinylestradiol (EE2) was 83%, 99.9%, and 78%, respectively (Ternes et al., 1999). A mean removal rate of 81% was found for estrogenic EDCs in case of activated sludge process using Yeast estrogen screen (YES) bioassay in Sweden indicating a high efficiency of the process over other treatment processes (Svenson et al., 2003). An overall degradation of E1 and E2 has been reported to be about 90% in activated sludge process in Andersen et al. (2003) while EE2 removal was found to be slower. The removal efficiency of EDCs in aerobic conditions was reported to be more than in anaerobic condition (Furuichi et al., 2006). 90- > 99% efficiency of removal of EDCs by activated sludge process have been reported by Leusch et al. (2006). Results obtained from Suzuki and Maruyama (2006) suggests that E1 and E2 were initially absorbed by activated sludge at a high rate and then within a few hours they were biodegraded by the same. The removal of estrogens by activated sludge is independent of the dissolved organic carbon (DOC) decomposition and nitrification. This study also revealed that the activity of activated sludge noticeably decreases at lower temperature. E1 concentration in WWTP effluent was found to be greater than that in influent in some instances, which has been explained as a result of biotransformation of E2 into E1 (Johnson and Sumpter, 2001). Layton et al. (2000) reported that 70–80% of added E2 was mineralized to CO_2 by activated sludge obtained from WWTPsin 24 h. While the mineralization of EE2 was 25–75 fold less than that of E2. EE2 was also recorded to be degeneratedentirely within 6 days by nitrifying activated sludge (Vader et al., 2000). In Paris, France a promising technique of tertiary treatment using fluidized bed micro-grain activated carbon (μGAC) was done and tested in large pilot scale. μGAC have been found to reduce BOD (38–45%), COD (21–48%), DOC (13–44%) and removes PPHs (60–80%), PCPs, artificial sweeteners, pesticides, etc (50- > 90%). The technique has many operational advantages as well as produces better wastewater quality as compared to powdered activated carbon (PAC) (Mailler et al., 2016).

4.1.2. Membrane bioreactor (MBR) and Sequencing batch reactor (SBRs)

The removal efficiency of emerging micropollutants (EPs) under anaerobic conditions combined with MBR was reported to be very lessas compared to conventional activated sludge systems (Joss et al., 2004). During anaerobic digestion of sludge under thermophilic and mesophilic conditions the removal efficiencies of sulfamethoxazole (SMX) were above 99% (Carballa et al., 2007), while Hai et al. (2011) reported the removal efficiencies of SMX under anoxic and aerobic conditions to be 65%. When cellulose was used as a primary substrate under anaerobic conditions, it was seen that ethinylestradiol, progesterone and metoprolol tartrate was significantly removed faster (Musson et al., 2010). With unified process of anaerobic pre-treatment followed by aerobic treatment might be a good choice for enhancement of EP removal. Removal efficiencies of more than 70% have been observed for some EPs such as 4-nonylphenol or caffeine and trimethoprim by the amalgamation of aerobic and anaerobic processes (Xue et al., 2010; Reyes-Contreras et al., 2011). Racz et al. (2012) had reported the removal of 17b-estradiol (E2) and 17a-ethinyl estradiol (EE2) with removal efficiencies of 99.3% for E2 and 95.7% for EE2 in the first 2 h in SBR 1% and 98.5% and 96.4% respectively within the first 2 h in SBR 2.

4.2. Natural aerobic processes

4.2.1. Waste stabilization ponds (WSP)

Li et al. (2013) reported that the WSP technology's removal

Table 3
Removal efficiencies (%) of Pharmaceuticals, PCPs and EDCs in environment media by different biological treatment processes.

TreatmentProcesses	Matrix	Compounds detected	Removal efficiency (%)	References
Activated Sludge process	Waste water	Cephalein	96, 38–99.8	Costanzo et al. (2005)
		Tetracycline	66–90	Lin et al. (2009)
		Ciprofloxacin	90	Zorita et al. (2009)
Membrane bioreactor (MBR)	Waste water	Nonylphenol	80	Wintgens et al. (2003)
		Ketoprofen	90	Urase et al. (2005)
		Pharmaceuticals	99	Lesjean et al. (2005)
		Steriods	80	Carballa et al. (2007)
		Sulfamethoxazole	99	Hai et al. (2011)
		Trimethoprim, 4-nonylphenol, Caffeine	65	Xue et al. (2010)
			70	Reyes-Contreras et al. (2011)
Upflow anaerobic sludge blanket (UASB)	Waste water	Berberine	95.2	Qiu et al. (2013)
Sequencing batch reactor (SBRs)	Waste water	Tetracycline	86.4	Kim et al. (2005)
		17b-estradiol	99.3	Racz et al. (2012)
		17a-ethinyl estradiol	95.7	
		Ibuprofen	63–90	Hasan et al. (2016)
		Ketoprofen	13–92	
Waste stabilization ponds	Waste water	Caffeine, Naproxen	99	Matamoros et al. (2016)
		Ibuprofen	92	
		Triclosan	97	
		Galaxolide	96	
		Tonalide	83	
Constructed wetlands (CWs)	Waste water	Caffeine	99	Matamoros et al. (2007)
		Enrofloxacin	94	Matamoros et al. (2009)
		Tetracycline	98	Carvalho et al. (2013)
		Acetaminophen (Paracetamol)	> 99	Conkle et al. (2008)
		Diclofenac	> 95	Avila et al. (2013)
		Diclofenac, 3-hydroxycarbamazepine (3-OH-CBZ), Venlafaxine (VLX), Odesmethylvenlafaxine (O-DM-VLX), Tramadol (TMD), Trimethoprim,	78–87	Hijosa-Valsero et al. (2010)
		Erythromycin, Clarithromycin, Metoprolol, Atenolol, Bezafibrate, Acyclovir and Codeine	> 70	Ruhmland et al. (2015)
Anaerobic treatment	Activated Sludge	Sulfomethoxazole	99	Carballa et al. (2006)
		Estrone	96/68	Paterakis et al. (2012)
		Nonylphenol	50/100	
		Ibuprofen, Naproxen	> 80	Samaras et al. (2013)

efficiencies for the identified pharmaceuticals and personal care products were relatively high with the exception for carbamazepine and a general removal efficiency extending from 88% to 100%. WSP technology had the greatest overall removal efficiency with an average removal efficiency of 82% and overall mass weight removal of 96%. The high effectiveness of removal accomplished by this technique may be elucidated by the conjunction of various removal techniques, such as biodegradation, sorption and photodegradation, as well as the technology's high hydraulic retention time which is around 20–30 days.

4.2.2. Constructed wetlands (CWs)

Removal efficiencies of certain ECs such as ciprofloxacin HCl, oxytetracycline HCl, nadolol, cotinine and enrofloxacin in CWs are reported to be as high as 70%. On the other hand removal efficiencies of monensin, narasin and salinomycin are comparatively less in the range of 20–50% (Li et al., 2014). It is observed that for various other pharmaceuticals, the greater part of their removal efficiencies in CWs seem, by all accounts, to be nearly or better than those in traditional WWTPs. Thus it can be inferred that CWs have great capability of being utilized as an option for the removal of pharmaceuticals from wastewater in secondary WWTP.

4.3. Anaerobic treatment

Bench scale upflow anaerobic sludge blanket (UASB) reactor study along with aerobic stirred tank reactor has been conducted for the degradation of sulfonamide sulfamerazine (Sponza and Demirden, 2007). In spite of the detected removal efficiencies up to 97%, the concentration used in the study which was 90 mg L^{-1} are not virtually similar with those found in agro-based wastewaters and therefore may not be useful in mirroring the real removal mechanism and therefore might not be practical. Samaras et al. (2013) proclaimed the consequential removal of more than 80% of Ibuprofen and Naproxen from sludge through anaerobic digestion. In a former report Carballa et al. (2006), listed the removal for these compounds to be 40% and 87% using laboratory-scale mesophilic anaerobic digesters. Muller et al. (2010) studied the removal of three natural hormones viz. E1, E2 and E3 in a full-scale anaerobic digester with akin feed (65:35, v/v, primary sludge: secondary sludge) and found no critical removal efficiency (~ 30–40%). The conjunction of various wastewater treatment technologies exhibited more noteworthy efficiency in the removal of phthalate esters than individual treatment procedures, e.g., the combination of anaerobic digester and a membrane bioreactor increment the efficiency of phthalate ester removal from 65–71% to 95–97%.

Table 4
Removal efficiencies (%) of Pharmaceuticals, PCPs and EDCs in environment media by different physico-chemical treatment processes.

Treatmentprocesses	Matrix	Compounds detected	Removal efficiency (%)	References
Activated carbon adsorption	Wastewater water	Bisphenol-A, Diclofenac, Carbamazepine, Sulfamethoxazole	50–100, 80–100 50–100 40–100	Nguyen et al. (2016)
Coagulation/flocculation	Drinking water	Acetaminophen, Diclofenac, Erythromycin 17α-estradiol, Estrone, Ethynylestradiol Musk Ketone, Triclocarban, Oxybenzone	< 20, < 20 < 40 < 40, < 20 < 20 < 20, < 40 < 20	Westerhoff et al. (2005)
Advanced oxidation process	Wastewater water	Acetaminophen, Diclofenac,	> 90, > 90	Rosario-Ortiz et al. (2010)
		Sulfamethoxazole	> 90	Reungoat et al. (2011)
		Galaxolide, Musk Ketone	20–90, 40–70	
Ozonation	River water	Estrone, Estradiol, Estriol, 17α-ethynylestradiol	98–99	Westerhoff et al. (2005)
	Drinking water	Estrone, Estradiol, Estriol,	96	Broseus et al. (2009)
		17α-ethynylestradiol	99.1–99.8	Bila et al. (2007)
		Estradiol	> 90	Gerrity et al. (2011)
		Acetaminophen, Diclofenac, Sulfamethoxazole	> 90	
	Wastewater	Estrone, Estradiol,	> 90	Hashimoto et al. (2006)
		17α-ethynylestradiol	~100, 95–99	
		Estrone, Estradiol, 17α-ethynylestradiol		
Chlorination	River water	Estrone, Estradiol, Estriol, 17α-ethynylestradiol	~100	Westerhoff et al. (2005)
	Drinking water	Acetaminophen, Diclofenac, Sulfamethoxazole	> 90, > 70 > 40	
		17α-estradiol, Estriol, Ethynylestradiol	> 90, > 90 > 70	
		Oxybenzone, Triclosan	> 90, > 70	
Ultraviolet irradiation	Secondary effluent of a sewage treatment plant	Sulfonamides	45–65	Kim et al. (2009)
		Macrolides	20–45	
		Tetracyclines	80–95, > 95	
		Fluoroquinolones	80–95, > 95	
		Trimethoprim	45–65	
	Surface water	Sulfonamides	80–95	Adams et al. (2002)
		Trimethoprim	45–65	
Nano filtration	Wastewater	Diclofenac, Ibuprofen, Metronidazole, Moxifloxacin, Telmisartan, Tramadol	~ 100	Beier et al. (2010)
		Roxithromycin, Azithromycin	> 87	Liu et al. (2014)

4.4. Physical-chemical treatment

Table 4 summarizes the removal efficiencies of PhACs, PCPs and EDCs by different treatment technologies in environmental media.

4.4.1. Activated carbon adsorption

Numerous hydrophobic and charged pharmaceuticals in water can be removed by activated carbon adsorption (Le-Minh et al., 2010). The adsorption process predominantly consists of the subsequent steps: (i) the solute is transported by the static fluid film encompassing the adsorbent by the bulk-adsorbate movement, (ii) film diffusion-adsorbate transport across the film, (iii) pores diffusion-adsorbate diffusion through the permeable system to the active sites, (iv) the interaction between the porous fabric and the adsorbate (Homem and Santos, 2011). The properties of the adsorbent such as specific surface area, permeability, polarity etc., removal efficiency of the activated carbon adsorptive treatment framework. In addition, the inceptive concentrations of the chosen compounds e.g. temperature, pH, existence of various other species may considerably amend the sorption effectiveness of antibiotic agents (Aksu and Tunç, 2005). Non-particular dispersive interactions like the van der Waals interaction between molecules are the prevailing means of removal of antibiotics and other organic compounds by activated carbon. While the non polar antibiotics with $\log K_{OW} > 2$ are eliminated. Furthermore, ionic/polar antibiotics can be removed by the electrostatic interaction of it with the surface charge group of activated carbon (Snyder et al., 2003).

Powdered activated carbon (PAC) and granular activated carbon (GAC) both posses pronounced prospective for the adsorption of smidgen ECs, especially the non-polar contaminants with $\text{Log } K_{OW} > 2$. PAC measurements or GAC recovery/substitution are necessary for better removal rates (Snyder et al., 2006; Bolong et al., 2009). Schafer et al. (2003) proclaimed that with 5 mg L^{-1} and contact time of 4 h the removal efficiency of PAC may reach up to 90% for endocrine disrupting compounds; Snyder et al. (2006) analyzed removal efficiency at 5 mg L^{-1} concentration and 5 h contact time of PAC for 66 PPCPs and just nine of them were removed with less than 50%. It is vital to consider the inevitable carbon redemption/disposition issue. PAC must be discarded through land filling or different solid management technique. Whilst used GAC should either is discarded or recovered. Thermal recovery used for GAC requires an enormous amount of energy, which may lead in a roundabout way to more prominent ecological dangers than the presence of ECs.

4.4.2. Hydrothermal carbonization (HTC) of biomass

In HTC biomass is heated in temperatures (180–250 °C) under saturated pressure in water medium for several hours. Unlike carbonization by pyrolysis which requires very high temperatures, HTC is simple process and reduces energy and economical cost (Fernandez et al., 2015). The other processes applied to the biomass are hydrolysis, decarboxylation, dehydration, polymerization, etc. but their detailed

reaction mechanisms are not known (Funke and Ziegler, 2010). The resultant biochar developed contains functional groups on their surface that increases their chemical reactivity and they serve as potent adsorbent (Roman et al., 2013). Kitchen and food wastes that are dumped can be used as the starting material for preparing carbonaceous adsorbent. Many ubiquitously available agro industrial wastes have been utilized as precursor to produce biochar such as sunflower stem, olives and walnut shells (Roman et al., 2012), orange peels (Fernandez et al., 2015), corn silage and poultry waste (Oliveira et al., 2013), empty bunches of palm (Parshetti et al., 2013).

4.4.3. Coagulation-flocculation

Coagulation is the alteration of colloidal particles causing aggregation by chemicals allowing them to flump over time. In a study it is reported that certain pharmaceuticals such as betaxolol, warfarin and hydrochlorothiazide are removed (80% removal efficiency) using aluminum sulfate ($Al_2(SO_4)_3$) by coagulation flocculation combined with sand filtration (Huerta-Fontela et al., 2011). Few antibiotics have been observed to be removed fairly adequately by this process (Choi et al., 2008). Musk compounds (personal care products), in particular celestolide galaxolide, and tonalide from hospital wastewater were reported to be removed with notable removal rates of 83%, 79% and 78% respectively (Suarez et al., 2009).

Many aspects such as pH, temperature, coagulant type and quantity, etc., govern the efficacy of the coagulation process. The utilization of aluminum sulfate as a coagulant was turned out to be profoundly powerful in eliminating certain hydrophobic pharmaceuticals, specifically doxazosin, warfarin, chlordiazepoxide, bromazepam, betaxolol, etc., (Huerta-Fontela et al., 2011), on the other hand for estrone and estradiol the efficiency was found to be less than 5% (Bundy et al., 2007; Le-Minh et al., 2010). It was also found that coagulants that are pre-hydrolyzed (polyferroussulphate (PFS), polyaluminium chloride (PACl), polyferric chloride (PFCl), etc.) can remove soluble dyes more effectively than hydrolyzing metal salts (Verma et al., 2012). PCPs or musk were reported to be removed by ferric chloride (Suarez et al., 2009). The media pH likewise impacts the level of dissociation of the contaminants, and thus shows definite removal operation for specific kind of coagulant (Zhao et al., 2008).

4.4.4. Advanced oxidation process (AOP_S)

Biological oxidation process has many advantages but some of the biorefractory compounds are not effectively removed (Lor et al., 2012). AOPs can be applied pre and/or post biological treatments. Oxidative debasement can happen either by direct response with the administered oxidant, or by means of formation of reactive secondary species such as hydroxyl radicals (OH^-)(Metcalf and Eddy, 2007). AOPs refer to the processes that assist the elevated generation of hydroxyl radicals. The generation of hydroxyl radicals usually starts with UV radiation and can be attained by various methods e.g. photo-catalysis using titanium dioxide (TiO_2) (Egerton et al., 2006; Murray and Parsons, 2006)or electrooxidation using boron doped diamond electrodes (Barrios et al., 2015) or by direct reaction of hydrogen peroxide (H_2O_2) (Rosenfeldt and Linden, 2004). Other AOPs are Ozone/H_2O_2, UV/Ozone, etc., that enhances hydroxyl radical production. AOPs are very proficient novel strategies for water and wastewater treatment (Legrini et al., 1993; Klavarioti et al., 2009; Malato et al., 2009). HO^- is intense oxidizing agent that oxidizes organic matter (Litter, 2005), but it is non-specific catalysis. This property is of great significance in wastewater treatment since radicals strafe oxidizable organic compounds with rate of 106–109 M^{-1} S^{-1}(Andreozzi et al., 2003).

The adaptability of the AOPs is improved by the reality there are diverse methods for creating hydroxyl radicals, expediting assent with particular treatment necessities. The main drawback of this process is the economic constraint. Therefore, nowadays environment friendly energy saving techniques using solar energy for oxidation is used. An investigation using solar heterogeneous photocatalysis with TiO_2, ozonation and solar photo Fenton (using Fe (III)) was done and it was found that photocatalytic ozonation offers a higher degradation rate than photocatalytic oxidation (Gimeno et al., 2016). The most well-known AOPs that have been utilized and assessed chiefly at a bench scale and also at pilot scale are: ozone (O_3), hydrogen peroxide (H_2O_2), photolysis using UV, Fenton reagent (homogeneous), semiconductors (heterogeneous) and ultrasound (sonolysis).

4.4.5. Ozonation

Ozonation treatment technology is based on the use of ozone gas which is a strong oxidizing agent and also as a disinfectant. Ozone after the reaction with microbes and their products and other chemicals is transformed into oxygen. About > 90% removal efficiency was noted by ozonation for aromatic compounds that are abundant in electron (e.g. sulfamethoxazoles) and additionally those compounds with deprotonated amine groups (e.g. trimethoprim) and also non-aromatic alkenes as they are profoundly amendable to oxidative reactions (Dickenson et al., 2009). Amid wastewater ozonation, numerous antibiotics e.g. sulfonamides, fluoroquinolones and macrolides have been appeared to be prevalently changed by ozone (Dodd et al., 2006) while N4-acetyl sulfamethoxazole, cephalexin and penicillin were changed to a huge degree by hydroxyl groups (Dodd et al., 2006). The bactericidal characteristics of anti-microbials are deactivated by ozone by altering the functional groups for example, dimethylamino groups and N-etheroxime (Lange et al., 2006), aniline part of sulfonamides (Huber et al., 2005), etc.

4.4.6. Chlorination

Chemical compounds can be made non-functional by inactivating the functional groups by chlorine substitution or addition (Crain and Gottlieb, 1935). On the other hand, compounds like antibiotics having active properties may be oxidized/disintegrated by chlorine (Crain and Gottlieb, 1935). The efficacious removal of antibiotic agents by chlorination from drinking water needs adequate free chlorine and enough contact time. A chlorine concentration of 1.2 mg L^{-1}, contact time of 24 h diminution of antibiotics in drinking water viz., > 99% for tetracyclines, 42% for trimethoprim, 50–80% for sulfonamides, 30–40% for fluoroquinolones, and less than 10% for macrolides, were accomplished and these were entirely removed after the duration of 10 days (Gibs et al., 2007). At chlorine concentrations of 3.5–3.8 mg L^{-1} and time interval of one day, expulsions of 90% to > 99% were accomplished for sulfamethoxazole, trimethoprim and erythromycin in river water (Westerhoff et al., 2005).

Albeit a few antibiotics might be more impervious to chlorination than others, they appear to slowly degenerate when subjected to free chlorine. Moreover, with increase in organic matters in the water the ideal concentration and contact time should be increased. Reactivity of sulfonamides with HOCl decreases in the subsequent manner: sulfadiamethoxine > sulfathiazole > sulfamethazine > sulfamerazine > sulfamethoxazole > sulfamethizole (Chamberlain and Adams, 2006). A pH greater than 8 (alkaline) restrains the expulsion of sulfonamides by chlorine oxidation (Chamberlain and Adams, 2006; Gibs et al., 2007). Huber et al. (2005) detected that in water the reaction between ClO_2 and antibiotics e.g., sulfonamides and macrolides, are rapid. Chlorine reacts with organic matter to produce toxic byproducts hence chlorination technique for treatment is of concern.

4.4.7. Ultraviolet (UV) irradiation

UV radiation can be utilized to debase the organic matter in water (Rosenfeldt and Linden, 2004). Degradation is administered by quantum yield of the compounds and the amount of UV imbibed (Kim et al., 2009). The contact time, dosage and DOC concentration are additionally imperative variables regulating the removal efficiency. The radiation emitted from UV light has germicidal properties and is used as a wastewater disinfectant. Appropriate dose of UV radiation has ended up being a successful bactericide and virucide for wastewater, not

adding to build up of byproducts. UV rays are utilized generally for microbial purification of wastewater. Besides, they have been utilized for removal of micropollutants. UV rays are absorbed by various pharmaceuticals, EDCs and PCPs that contains chromophores, which are transformed amid UV treatment. It was accounted for that UV light delivered by mercury lamps oxidizes PCPs and with the introduction of hydrogen peroxide shows elevated transformation rates. Nevertheless, UV illumination is to some degree costlier when analogized with most other traditional treatment techniques.

4.4.8. Nano filtration (NF)

Pharmaceuticals can be removed by NF films by means of three methods: adsorption, electrostatic repulsion and sieving (Dolara et al., 2012). Depending on various compounds the removal efficiencies vary, and is stringently associated to: (a) the micro-pollutants' physicochemical properties viz., size, solvency, hydrophobicity, charge, polarity, etc., (b) film properties (porousness, pore size, hydrophobicity and surface charge), and (c) membrane functioning conditions such as flux, transmembrane pressure, dismissals/recuperation and water supply quality. NF has been shown to be a propitious option for dispensing with pharmaceuticals, as it can accomplish more than 90% removal efficiency (Bolong et al., 2009).

5. Future research perspectives

Relatively little data is available with respect to the adequacy of recent treatment techniques including chemical and biological treatments, membrane filtration, adsorption processes, etc. Nevertheless, with the guide of a couple of essential molecular and physicochemical properties, it has been conceivable to subjectively describe the sensitivity of different antibiotic agents to some of the removal processes. While it seems to be no processes with good efficiency for the removal of every single EC under regular operational conditions, inferring that prudent treatment techniques should be applied for the efficacious removal. With the development of advanced analytical instruments it is possible to quantify the emerging contaminants. Approach should also be made to detect new ECs in our complex environment and to find their efficient removal strategies.

A large number of new emerging contaminants are emanating that are not reported or detected due to the lack of knowledge. The knowledge about the fate of persistent transformation products, after the treatment of ECs that can be more harmful and toxic to the environment is still lacking. The absence of ECs does not guarantee lack of TPs. Also, the ECs behave differently in natural environment and shows different ecotoxicological effects that cannot be predicted by conventional tests in labs. Therefore, it is required to develop new protocols for ecotoxicity test and measure different effects by using different organisms with suitable endpoints. It is suggested that more exhaustive studies be led to fill knowledge gaps in the conduct of ECs under traditional sewage treatment and advanced treatment techniques. Future research ought to incorporate a keen emphasis on the treatment techniques and fate of these contaminants into the sewage biomass and their conversion into more toxic or pharmacologically active metabolites during the treatment. Combination of treatment methods should be applied for removal of ECs that is more efficient in removing, rather than treatment applying single technique or conventional methods. Besides, future research ought to govern and delineate all fundamental treatment plant operational variables since these are crucial for later correlation or analysis.

In summary, the appearance of emerging contaminants in the nature has become more evident as persistently enhancing assay methods have brought down the limit of detection for a broad cluster of ECs in natural specimens. ECs are commonly found in the water environment since at standard temperatures and pressures ECs dissolve nimbly without being evaporated. Although individual EC parameters occurring usually at ng L^{-1} levels, their impact on the environment and human health is unknown, particularly the additive effects have still not been considered. Also, it is important to direct research on the prevalence, fate and treatment of human originated metabolites in WWTPs. Majority of antibiotics and their byproducts are discharged into the municipal sewage by men; notwithstanding, there is very few knowledge about their behavior and biodegradability in water. In future, to predict the sources, fate and behaviors of ECs in the water environment researchers should concentrate on the development of a risk based screening models and framework.

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