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A Review on Emerging Contaminants in Indian Waters and Their Treatment Technologies

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ABSTRACT

Emerging contaminants (ECs) have been detected recently in many water bodies across India. Studies have found the presence of ECs in surface water, groundwater, stormwater, treated wastewater, treated industrial effluent, bottled water and snow from glaciers in Indo-Chinese border which contaminate water bodies. The surface water recharges the groundwater, thereby the ECs make their way to deep water aquifers. The soil also gets contaminated and plants can uptake ECs. These micropollutants can cause adverse ecological and human health effects. Antimicrobial resistance of bacteria to commonly used antibiotics has been observed in India. An exhaustive review of emerging contaminants in Indian waters and their treatment technologies has been carried out. Antibiotic-resistant genes can be easily transferred resulting in a plethora of antimicrobial-resistant bacteria which can cause devastating effects on human health. Conventional biological treatment is not capable of removing ECs completely. Advanced oxidation processes using ozonation and visible light active photocatalyst are a sustainable solution for the removal of most ECs. Hence, it is of utmost importance to monitor the presence of ECs in the water environment and develop treatment technologies for its removal.

INTRODUCTION

United States Geological Survey (USGS) defines an Emerging Contaminant (EC) as any synthetic or naturally occurring chemical or microorganism that is not commonly monitored in the environment but has the potential to enter the environment and cause unknown or suspected adverse ecological and human health effects. Sources of ECs are compounds such as pharmaceuticals and personal care products (PPCPs), contrast media, plasticizers, food additives, wood preservatives, laundry detergents, surfactants, disinfectants, flame retardants, pesticides, natural and synthetic hormones, and a few disinfection by-products (DBPs) (La Farré et al. 2008) that enters through several pathways (Fig. 1). Balakrishnan and Mohan (2016) identified the ECs in stormwater in Chennai. Several aromatic and aliphatic hydrocarbons such as decane, p-xylene, octanol, eicosane etc. were detected in the stormwater. Some of them are used in fuels while others are plasticizers or used as precursors for perfumes and flavouring agents.

Due to its persistent, bioaccumulative and toxic nature, it poses a great risk on the environment and human health. Hence, extensive ecotoxicological studies must be carried out to establish toxicity endpoints. The conventional biological treatment plant is not capable of removing the ECs and requires advanced treatment technologies. This review is intended to present an overview of the presence of ECs in Indian water matrices along with the remediation technologies used for some predominant ECs.

OCCURRENCE OF ECS IN INDIAN WATERS

In India, the contribution of emerging contaminants in aquatic environment comprises of 57% pesticides, 17% pharmaceuticals, 15% surfactants, 7% personal care products and 5% phthalates (Gani & Khazmi 2016). Several studies ascertained the presence of Active pharmaceutical ingredients (API), personal care products (PCP), pesticides, endocrine-disrupting chemicals (EDC) and artificial sweeteners (ASW) in surface water bodies.

Pharmaceuticals

The Delhi-based non-profit Centre for Science and Environment (CSE) reports that 78 percent of the sewage generated in India remains untreated and is discharged in rivers, groundwater or lakes (CSE 2016). This could explain the reason for high levels of API in water matrices.

William et al. (2019) investigated the presence of pharmaceuticals in Ahar River in Udaipur city and found 19 pharmaceuticals including 4 antibiotics. The river originates as a potable water source which gets contaminated as it

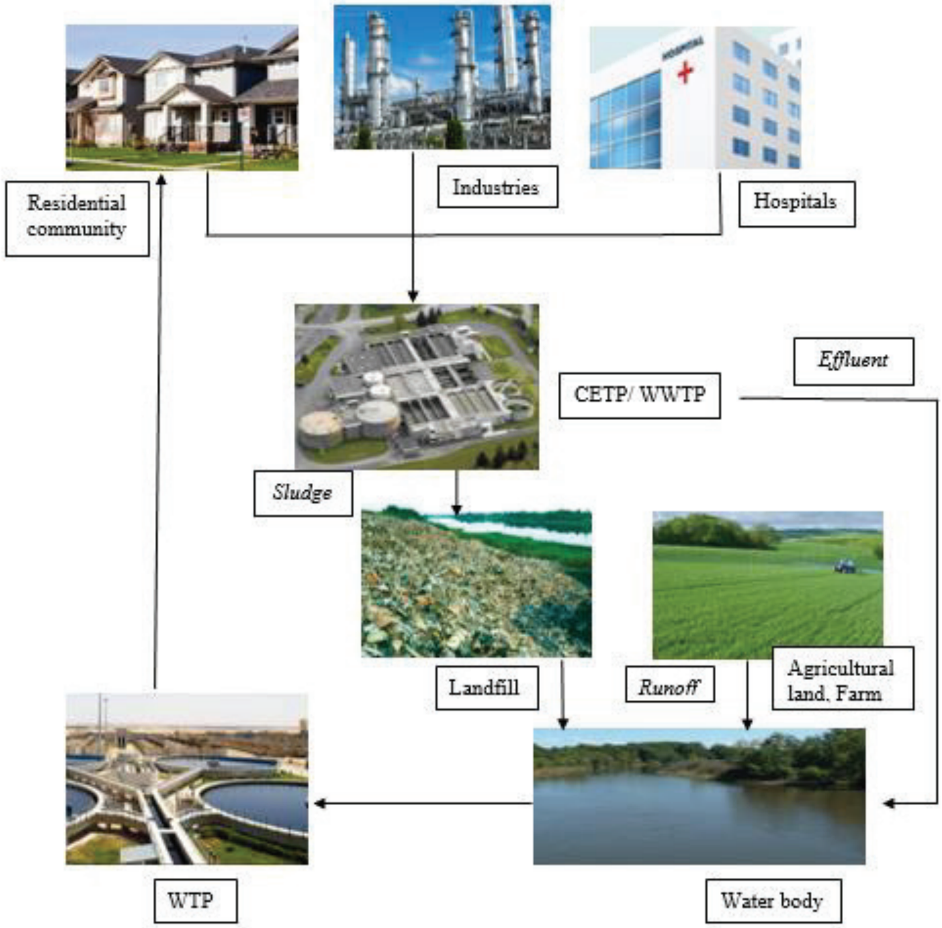

Fig. 1: Pathway for ECs into water environment.

moves through urban areas. It was observed that the concentration of emerging contaminants increased as it passed through densely populated regions indicating the potential discharge of domestic wastewater into the river. Mutiyar & Mittal (2014) studied the occurrence of antibiotics in influent and effluent of STP and Yamuna river in Delhi. Ampicillin concentration in untreated and treated wastewater was 104.2 µg/L and 12.7 µg/L. Fick et al. (2009) found extremely high concentrations of Ciprofloxacin and Cetirizine in the lake water, Isakavagu-Nakkavagu rivers and well water sample from Hyderabad. Lubbert et al. (2017) reported a high concentration of Fluconazole, an antifungal, to be 236,950 µg/L from a sewer in the industrial area apart from several classes of antibiotics. The effluent after preliminary treatment is carried through sewers to the nearby Sewage Treatment Plant (STPs) and treated effluents are finally discharged into the Musi river. The area is home to 30 pharmaceutical drug manufacturers. Similar studies by Larsson et al. (2007) in the water bodies and wells near the pharmaceutical manufacturing industrial zone found the highest reported concentrations of API in the world. Antimicrobial resistance has also been observed in bacteria from this region. Antibiotic resistance caused by sulfamethoxazole (SMX) has been determined in River Ganga and groundwater in Varanasi (Lapworth et al. 2018). Philip et al. (2017) presented a comprehensive review of the presence of antibiotic-resistant genes (ARG) in India.

Ramaswamy et al. (2011) studied the occurrence of carbamazepine (CBZ), antiepileptic in water samples from the Kaveri, Vellar, Tamiraparani rivers and Pichavaram mangrove in Tamil Nadu (TN). This was the first study to report the presence of a pharmaceutical in south Indian rivers. Lapworth et al. (2018) determined the presence of SMX and CBZ in River Ganga. It was interesting to note that the CBZ concentration in River Ganga and groundwater from deep wells at a depth of even 140 meters in Varanasi had a concentration of 0.02 µg/L. Similarly, it was observed that the concentration of CBZ and SMX were much higher in the groundwater compared to Ganga river water (Sharma et al. 2019). The major reason for the presence of API in groundwater is bank infiltration and the accumulation of API in groundwater due to limited flow as against surface water. Anumol et al. (2016) investigated several pharmaceuticals in the influent and effluent of three wastewater treatment plants (WWTPs) in Chennai and found several API namely CBZ, SMX, naproxen and atenolol in all the effluent samples. Iohexol used as a contrast agent during X-rays was found in high concentrations in the influent and effluent of WWTPs and had very less removal. It was found that the major classes of contributors were X-ray contrast media, pharmaceuticals and stimulant, caffeine. The average concentration of CBZ, SMX and naproxen in the influent of WWTP was 0.92 µg/L. Caffeine presented a high concentration of 65 µg/L in the incoming wastewater. Gemfibrozil, the anti-cholesterol drug was less than 0.1 µg/L. API such as CBZ, SMX, gemfibrozil, caffeine, naproxen, ibuprofen and diclofenac (DCF), Non-Steroidal Anti-inflammatory drug (NSAID), are found to be the chief API in Indian waters. Table 1 represents the details of 42 predominant API found across Indian water bodies.

Personal Care Products

Triclosan and Triclocarban are the commonly used antibacterials in soaps, shampoos, hand sanitizers, etc. The occurrence of triclosan, triclocarban and parabens (preservative in cosmetics), commonly used in personal care products (PCP), in water samples from the Kaveri, Vellar, Tamiraparani rivers and the Pichavaram mangrove in South India (Ramaswamy et al. 2011, Vimalkumar et al. 2018). Triclosan and triclocarban were detected in the influent and effluent of WWTP in Chennai (Anumol et al. 2016).

Table 1: Details of API commonly encountered in Indian waters.

API	Class of drug	Concentration (µg/L)	Location of sampling	Reference
Carbamazepine	Antiepileptic	13.00	Kaveri, TN	Ramaswamy et al. (2011)
		5.14	Tamiraparani, TN	Ramaswamy et al. (2011)
		2.67	Vellar, TN	Ramaswamy et al. (2011)
		6.65	Pichavarom Mangrove, TN	Ramaswamy et al. (2011)
		0.113	Ahar River, Udaipur	William et al. (2019)
		0.09	Groundwater, Varanasi, UP	Lapworth et al. (2018)
		0.02	Groundwater, Ramnagar, UP	Lapworth et al. (2018)
		0.02	River Ganga, UP	Lapworth et al. (2018)
		0.016	Stretch of River Ganga	Sharma et al. (2019)
Fluconazole	Antifungal	236950	Sewer, Patancheru Pashamylaram Industrial area, Hyderabad	Lubbert et al. (2017)
Voriconazole	Antifungal	2500	Sewer, Patancheru Pashamylaram Industrial area, Hyderabad	Lubbert et al. (2017)
Acetaminophen	Analgesic	0.004	Stretch of River Ganga	Sharma et al. (2019)
		0.002	Groundwater near Ganga basin	Sharma et al. (2019)
Atenolol	Beta-blocker	0.001	Stretch of River Ganga	Sharma et al. (2019)
		6.9	Influent, WWTP, Chennai	Anumol et al. (2016)
Ketoprofen	NSAID	0.11	Stretch of River Ganga	Sharma et al. (2019)
		0.023	Groundwater near Ganga basin	Sharma et al. (2019)
Ibuprofen	NSAID	1.89	Ahar River, Udaipur	William et al. (2019)
		0.023	Stretch of River Ganga	Sharma et al. (2019)
		0.049	Groundwater near Ganga basin	Sharma et al. (2019)
Diclofenac	NSAID	0.41	Ahar River, Udaipur	William et al. (2019)
		0.041	Stretch of River Ganga	Sharma et al. (2019)
		0.002	Groundwater near Ganga basin	Sharma et al. (2019)
		5.3	Influent, WWTP, Chennai	Anumol et al. (2016)

Table cont....

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API	Class of drug	Concentration (µg/L)	Location of sampling	Reference
Naproxen	NSAID	0.24	Ahar River, Udaipur	William et al. (2019)
		0.003	Stretch of River Ganga	Sharma et al. (2019)
		0.002	Groundwater near Ganga basin	Sharma et al. (2019)
Iohexol	X-ray contrast media	9.6	Influent, WWTP, Chennai	Anumol et al. (2016)
		8.7	Effluent, WWTP, Chennai	Anumol et al. (2016)
Caffeine	Cosmetic, Psychoactive drug	37.5	Ahar river, Udaipur	Williams et al. (2019)
		0.743	Stretch of River Ganga	Sharma et al. (2019)
		0.262	Groundwater near Ganga basin	Sharma et al. (2019)
		65	Influent, WWTP, Chennai	Anumol et al. (2016)
		5.4	Effluent, WWTP, Chennai	Anumol et al. (2016)
Hydrochlorothiazide	Diuretics	0.004	Stretch of River Ganga	Sharma et al. (2019)
		0.001	Groundwater near Ganga basin	Sharma et al. (2019)
		0.39	Ahar River, Udaipur	William et al. (2019)
Trimethoprim	Antibiotic	0.05	Ahar River, Udaipur	William et al. (2019)
Azithromycin	Antibiotic	0.99	Ahar River, Udaipur	William et al. (2019)
Moxifloxacin	Antibiotic, fluoroquinolone	694.1	Sewer, Patancheru industrial area	Lubbert et al. (2017)
Linezolid	Antibiotic	37	Sewage storage, Patancheru Kazipally Industrial area, Hyderabad	Lubbert et al. (2017)
Clarithromycin	Antibiotic	27.7	Weir in Musi River, Hyderabad	Lubbert et al. (2017)
		0.05	Ahar River, Udaipur	William et al. (2019)
Levofloxacin	Antibiotic, fluoroquinolone	12.8	Musi River, upstream of Amberpet treatment plant, Hyderabad	Lubbert et al. (2017)
Ciprofloxacin	Antibiotic, fluoroquinolone	14000	Effluent-Patancheru Enviro Tech Ltd. (PETL), Hyderabad	Fick et al. (2009)
		2500	Isakavagu-Nakkavagu River, Hyderabad	Fick et al. (2009)
		6500	Lake, upstream of PETL, Hyderabad	Fick et al. (2009)
		14	Well, near lake, upstream of PETL, Hyderabad	Fick et al. (2009)
		28000-31000	Effluent-CETP, Hyderabad	Larsson et al. (2007)
		0.029	Stretch of River Ganga	Sharma et al. (2019)
		0.005	Groundwater near Ganga basin	Sharma et al. (2019)
Cetirizine	Antihistamine	2100	Effluent- CETP, Hyderabad	Fick et al. (2009)
		1200	Lake, upstream of PETL, Hyderabad	Fick et al. (2009)
Norfloxacin	Antibiotic, fluoroquinolone	25	Effluent-PETL, Hyderabad	Fick et al. (2009)
		520	Lake, upstream of PETL, Hyderabad	Fick et al. (2009)
		390 - 420	Effluent-CETP, Hyderabad	Larsson et al. (2007)
Glibenclamide	Hypoglycemic drug	0.80	Ahar River, Udaipur	William et al. (2019)
Citalopram	Selective serotonin reuptake inhibitor	430	Effluent-PETL, Hyderabad	Fick et al. (2009)
		8	Lake, upstream of PETL, Hyderabad	Fick et al. (2009)

Table cont....

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API	Class of drug	Concentration (µg/L)	Location of sampling	Reference
Enalapril	Angiotensin-converting enzyme inhibitor	2.2	Effluent-PETL, Hyderabad	Fick et al. (2009)
		1	Lake, upstream of PETL, Hyderabad	Fick et al. (2009)
Enrofloxacin	Antibiotic, fluoroquinolone	210	Effluent-CETP, Hyderabad	Fick et al. (2009)
		25	Lake, upstream of PETL, Hyderabad	Fick et al. (2009)
Metoprolol	Beta-adrenoreceptor antagonist	4	Effluent-PETL, Hyderabad	Fick et al. (2009)
		7	Lake, upstream of PETL, Hyderabad	Fick et al. (2009)
		0.07	Ahar River, Udaipur	William et al. (2019)
Ofloxacin	Antibiotic, fluoroquinolone	55	Effluent-PETL, Hyderabad	Fick et al. (2009)
		11	Lake, upstream of PETL, Hyderabad	Fick et al. (2009)
		150–160	Effluent-CETP, Hyderabad	Larsson et al. (2007)
Terbinafine	Antimycotic	120	Effluent-PETL, Hyderabad	Fick et al. (2009)
		15	Lake, upstream of PETL, Hyderabad	Fick et al. (2009)
		10.6	Weir in Musi River, downstream of Amberpet treatment plant, Hyderabad	Lubbert et al. (2017)
Sulfamethox-azole	Antibiotic	0.82	Ahar River, Udaipur	William et al. (2019)
		0.1	River Ganga, UP	Lapworth et al. (2018)
		0.04	Groundwater, Varanasi, UP	Lapworth et al. (2018)
		0.02	Groundwater, Ramnagar, UP	Lapworth et al. (2018)
		0.028	Stretch of River Ganga	Sharma et al. (2019)
		0.004	Groundwater near Ganga basin	Sharma et al. (2019)
Sulfanilamide	Antibiotic	0.12	Groundwater, Varanasi, UP	Lapworth et al. (2018)
		0.12	Groundwater, Ramnagar, UP	Lapworth et al. (2018)
Ampicillin	Antibiotic, Penicillin	29.1	Sewer, Patancheru industrial area	Lubbert et al. (2017)
		12.7	Treated wastewater, STP, Delhi	Mutiyar & Mittal (2014)
		13.75	Yamuna River, Delhi	Mutiyar & Mittal (2014)
Doxycycline	Antibiotic	14.9	Sewer, Patancheru Pashamylaram Industrial area	Lubbert et al. (2017)
Sparfloxacin	Antibiotic, fluoroquinolone	2.09	Yamuna River, Delhi	Mutiyar & Mittal (2014)
Cefuroxime	Antibiotic	1.7	Yamuna River, Delhi	Mutiyar & Mittal (2014)
Gatifloxacin	Antibiotic, fluoroquinolone	0.48	Yamuna River, Delhi	Mutiyar & Mittal (2014)
Desipramine	Antidepressant	0.075	Ahar River, Udaipur	William et al. (2019)
Tramadol	Analgesic	0.09	Ahar River, Udaipur	William et al. (2019)
Cyclophospha-mide	Antineoplastics	0.32	Ahar River, Udaipur	William et al. (2019)
Diltiazem	Calcium channel blocker	0.016	Ahar River, Udaipur	William et al. (2019)
Propranolol	Beta-blocker	0.016	Ahar River, Udaipur	William et al. (2019)
Venlafaxine desmethyl	Antidepressant	1.5	Ahar River, Udaipur	William et al. (2019)
Verapamil	Calcium channel blocker	0.02	Ahar River, Udaipur	William et al. (2019)

Table 2: Details of PCP commonly encountered in Indian waters.

PCPs	Class of PCPs	Concentration (µg/L)	Sampling Location	Reference
Triclosan	Antibacterial	0.04	Kaveri	Ramaswamy et al. (2011)
		0.14	Tamiraparani	Ramaswamy et al. (2011)
		0.009	Vellar	Ramaswamy et al. (2011)
		0.02	Pichavarom Mangrove	Ramaswamy et al. (2011)
		0.005	Stretch of River Ganga	Sharma et al. (2019)
		0.01	Groundwater near Ganga basin	Sharma et al. (2019)
Triclocarban	A[[ntibacterial	1.12	Kaveri	Vimalkumar et al. (2018)
		0.17	Tamiraparani	Vimalkumar et al. (2018)
		0.10	Vellar	Vimalkumar et al. (2018)
		0.003	Stretch of River Ganga	Sharma et al. (2019)
		0.002	Groundwater near Ganga basin	Sharma et al. (2019)
UV-9	BUVSs	0.027	Kaveri	Vimalkumar et al. (2018)
		0.028	Tamiraparani	Vimalkumar et al. (2018)
		0.004	Vellar	Vimalkumar et al. (2018)
UV-329	BUVSs	0.03	Kaveri	Vimalkumar et al. (2018)
		0.03	Tamiraparani	Vimalkumar et al. (2018)
		0.016	Vellar	Vimalkumar et al. (2018)
DEET	Mosquito repellant	0.39	Ahar river, Udaipur	Williams et al. (2019)
		0.022	Stretch of River Ganga	Sharma et al. (2019)
		0.015	Groundwater near Ganga basin	Sharma et al. (2019)
Benzotriazole	UV stabilisers	0.526	Ahar river, Udaipur	Williams et al. (2019)

DEET used in mosquito repellents has been detected in Ahar River, Udaipur (William et al. 2019). This indicates the contamination of river water by domestic wastewater. It was found that the concentration of PCP increased with an increase in population density. The presence of benzotriazole ultraviolet stabilizers (BUVSs) which are used extensively in personal care products such as soaps, shampoos, sunscreen lotions, shaving creams etc. has been detected in the three major rivers in south India - Kaveri, Tamiraparani and Vellar (Vimalkumar et al. 2018). Among the BUVSs, UV-329 and UV-9 were predominant. Benzotriazole has also been reported to be present in Ahar River (William et al. 2019). Table 2 shows PCPs commonly encountered in Indian water bodies.

Perfluorinated Compounds

Recent studies have reported the presence of perfluorinated compounds (PFCs) in groundwater, surface water, drinking water, snow and precipitation water. Of the PFCs reported, short-chain (C<5) PFCs were predominant. PFOS is one of the prominent PFCs present in Ganga river and groundwater samples from Varanasi and Ramnagar besides PFDA and PFNA. The highest concentration reported in the groundwater and River Ganga were 0.033 µg/L and 0.025 µg/L respectively. Interestingly, the PFAS concentration in groundwater in Varanasi was much higher than in river Ganga (Lapworth et al. 2018). It indicates the contamination of ECs in water through the recharge pathway and its mobility through the substrata. Yeung et al. (2009) studied the occurrence of PFCs in surface water bodies in several cities in India including the stretch of River Ganga. High concentrations of PFCs were reported in Chennai followed by Allahabad, Varanasi, Patna and Kanpur. Generally, the PFOS was dominant over PFOA while PFOA was predominant in Chennai. 23.1 ng/L of PFOA was found in Cooum River, Chennai. The concentration of PFOS was 3.91 ng/L and 12 ng/L in Cooum River and untreated sewage respectively. High concentrations of PFOS were reported at Ganges-Yamuna confluence, Allahabad. Sharma et al. (2016) studied the PFAS contamination in river Ganges and groundwater samples from the river basin. The river water had the highest concentration for PFHxA (4.7 ng/L) and PFBS (10.2 ng/L) among PFCAs and PFSAs. In groundwater, PFBA (9.2 ng/L) and PFBS (4.9 ng/L) had the highest concentrations among PFCAs and PFSAs respectively. The daily PFAS exposure was below the safety threshold for oral non-cancer risk for all age groups. The contamination trend in river water and groundwater are similar indicating that the aquifer is recharged by the river.

Sunantha & Vasudevan (2016) investigated the PFOA and PFOS in Velachery Lake, Porur Lake, Chitlapakkam Lake, Pallavaram Lake, Korattur Lake, Ambattur Lake (Chennai), Cauvery River (Erode), and Noyyal River (Tiruppur). PFOA was high in the range 4 to 93 ng/L and PFOS varied from 3 to 29 ng/L. Kwok et al. (2010) studied the occurrence of PFCs in precipitation samples from several countries including India. PFAS concentration was found to be 0.04 ng/L in samples from Patna. Perfluoro-n-pentanoic acid (PFPeA) accounted for more than 25% of PFCs. Wang et al. (2019) reported the highest PFAS concentration in surface snow sample from glaciers in west China to be 3.97 ng/L. The two predominant atmospheric circulation patterns over western China are Westerly winds and the Indian monsoon, 70 % being contributed by Indian monsoon, indicating the presence of PFC in the atmosphere. The previous studies have reported the presence of short-chain PFAS and other PFC in the Indian water environment.

Endocrine Disrupting Chemicals

Endocrine Disrupting Chemicals (EDCs) have received wide attention recently due to its ability to disrupt the endocrine system of humans, special vulnerable groups include pregnant mothers, children and elderly. The occurrence of phthalates, polychlorinated biphenyls (PCBs), plasticizers and perchlorate have been reported in several studies. Phthalate intake has been estimated by analysing phthalate concentration in urine samples in India, the second highest concentration (389 ng/mL) was found (Guo et al. 2011). PCBs have been reported to affect the reproductive system. Chakraborty et al. (2016) determined the total PCBs in River Hooghly and River Brahmaputra at concentrations of 0.233 μg/L and 0.161 μg/L respectively. PCBs can make their way into the environment through leaching from open dumpsites, poorly managed landfills and improperly discarded electronic waste. William et al. (2019) found six steroid hormones in Ahar river, Udaipur such as estrone, 17αestradiol, 17αestradiol, androsterone, dihydrotestosterone, testosterone. The concentration of estrone and androsterone were 1.55 μg/L and 0.15 μg/L respectively. In addition, bisphenol A, plasticizer, was also found at a concentration of 0.3 μg/L.

Yamazaki et al. (2015) studied the presence of Bisphenol A and bisphenol (BP) analogues including BPS and BPF in surface water samples from Japan, China, Korea and India. In India, the highest BPA concentrations found in Buckingham and Cooum were 1.39 μg/L and 0.423 μg/L respectively. The highest BPS concentrations were 1.08 μg/L and 6.84 μg/L in the Buckingham canal and Adyar river respectively which was higher than in other countries. BP was not detected in Puzhal lake. Selvaraj et al. (2014a) analysed Kaveri, Vellar and Tamaraparani river waters for octylphenol (OP), nonylphenol (NP), and bisphenol A (BPA) and the concentrations of OP, NP and BPA ranged from ND (not detected) to 16.3 ng/L, ND to 2200 ng/L, and 2.8 to 136 ng/L respectively. High concentrations were observed in the Kaveri river. NP was much higher than BPA followed by OP. Srivastava et al. (2010) reported a high concentration of phthalic acid esters (PAE), plasticizer, in the sediments of Gomti river and the mean concentrations of DMP, DEP, DBP, DEHP and DOP were 10.54, 4.57, 10.41, 31.61 and 5.16 μg/kg respectively. DEHP was the most frequently detected PAE. Such high concentration in the sediments from Gomti river indicates the high contaminant load of PAE in the river, which deposit on sediments due to their hydrophobicity. Selvaraj et al. (2014b) studied the presence of phthalates in the stretch of Kaveri river and found the highest concentration of DMP, DEP, DBP, BBP, DEHP and DOP as 94, 520, 372, 145, 822 and 85 ng/L. DEHP and DEP accounted for 57% and 22% of phthalates. The high concentration was observed in Bhavani town sampling location at the confluence of Bhavani and Noyyal rivers. The concentration of phthalates in the sediments of the Kaveri river was lower than the Gomti river.

Das et al. (2014) measured 15 phthalates in drinking water from Jawaharlal Nehru University and the Okhla industrial area. The sum concentration of all phthalates was found to be 0.39 and 3.804 μg/L. DEHP was the most predominant phthalate and the estimated total daily intake level was 70 mg/kg/d. Phthalates such as DEP, DBP, BBP and DEHP have been reported in the three wastewater treatment plants. The highest mean concentration of DEP, DBP and DEHP was 7, 14 and 19 μg/L in summer. The main removal process of phthalates is biotransformation (Gani & Kazmi 2016). Phthalates have also been reported to be present in bottled water at a maximum total phthalate concentration of 7.82 μg/L. Similar to previous studies the highest phthalate contributor was DEHP. The phthalate concentration was found to increase with an increase in shelf life (Selvaraj et al. 2016).

Perchlorate is used extensively in fireworks, matches, arms and ammunition industries. It also finds its application in lubricating oils, aluminium refining, paint and rubber manufacturing, leather tanning, paper and pulp processing (used in bleaching powder) and as a dye mordant. Sodium hypochlorite used as a disinfectant in chlorination during water and wastewater treatment has been identified as a source of perchlorate contamination (USEPA 2007). It can interfere with iodine transport, thereby reducing thyroid production (Fisher & McLanahan 2008). It can cause haemolytic anaemia, delayed development, affect pregnant women, foetus and children (Pearce et al. 2007). The USEPA drinking water standard for perchlorate is 15 μg/L. Nadaraja et al. (2015) analysed surface and groundwater samples from 27 locations in Kerala. Perchlorate was detected in 58% of groundwater samples and the highest concentration was 7270 μg/L. All the surface water sample analysed were reported to be contaminated with perchlorate and the highest observed concentration was 355 μg/L, while all the five bottled drink-

Table 3: Details of the pesticide concentration in Indian waters.

Pesticides	Concentration (μg/L)	Location	Reference
HCH	4.403	Surface water, Dibrugarh, ASM	Mishra & Sharma (2011)
	5.168	Groundwater, Dibrugarh, ASM	Mishra & Sharma (2011)
	4.911	Surface water, Nagaon, ASM	Mishra & Sharma (2011)
	5.574	Groundwater, Nagaon, ASM	Mishra & Sharma (2011)
	1.95	Unnao district, Madya Pradesh	Singh et al. (2007)
	0.022	River Brahmaputra	Chakraborty et al. (2016)
	0.114	River Hooghly	Chakraborty et al. (2016)
DDT	5.402	Surface water, Dibrugarh, ASM	Mishra & Sharma (2011)
	6.549	Groundwater, Dibrugarh, ASM	Mishra & Sharma (2011)
	6.121	Surface water, Nagaon, ASM	Mishra & Sharma (2011)
	6.904	Groundwater, Nagaon, ASM	Mishra & Sharma (2011)
	0.23	Unnao district, MP	Singh et al. (2007)
	0.225	River Brahmaputra	Chakraborty et al. (2016)
	0.026	River Hooghly	Chakraborty et al. (2016)
Total OCP	3.72	Unnao district, MP	Singh et al. (2007)
	0.245	River Brahmaputra	Chakraborty et al. (2016)
	0.154	River Hooghly	Chakraborty et al. (2016)
Endosulfan	0.13	Unnao district, MP	Singh et al. (2007)
	0.053	River Brahmaputra	Chakraborty et al. (2016)
	0.01	River Hooghly	Chakraborty et al. (2016)
Endrin	0.01	Unnao district, MP	Singh et al. (2007)
Aldrin	1.88	Unnao district, MP	Singh et al. (2007)
	0.019	River Brahmaputra	Chakraborty et al. (2016)
	0.009	River Hooghly	Chakraborty et al. (2016)
Chlorpyrifos	0.004	River Ganga, UP	Lapworth et al. (2018)
	0.004	Groundwater, Ramnagar, UP	Lapworth et al. (2018)
	0.003	Groundwater, Varanasi, UP	Lapworth et al. (2018)
Phenoxyacetic acid	0.7	River Ganga, UP	Lapworth et al. (2018)
	0.05	Groundwater, Ramnagar, UP	Lapworth et al. (2018)
	0.2	Groundwater, Varanasi, UP	Lapworth et al. (2018)
Diuron	0.04	River Ganga, Uttar Pradesh	Lapworth et al. (2018)
Atrazine	0.03	River Ganga, Uttar Pradesh	Lapworth et al. (2018)
Heptachlor	0.01	River Brahmaputra	Chakraborty et al. (2016)
	0.026	River Hooghly	Chakraborty et al. (2016)

ing water was free of perchlorate. Raj & Muruganandam (2014) found very high perchlorate concentration of 93,500 μg/L in the real industrial effluent sample from Vellore, Tamil Nadu. The mean perchlorate concentrations in drinking water, groundwater, surface water and effluent water were 126, 831, 1110 and 15754 μg/L respectively. Isobe et al. (2013) studied the perchlorate contamination in Sivakasi and Madurai, which accounts for 90% of the global production of fireworks. The high concentration of 7690 μg/L, 30 μg/L and 0.39 μg/L were reported for groundwater, surface water and drinking water respectively. Firework industries are the source of perchlorate in water.

Pesticides

Lapworth et al. (2018) determined the concentration of pesticides such as Chlorpyrifos, phenoxy acetic acid, diuron and atrazine in Ganga river, groundwater samples in shallow and deep abstraction wells from Varanasi and Ramnagar, Uttar Pradesh (UP). The high concentration of phenoxy acetic acid was observed. Chakraborty et al. (2016) analysed the presence of organochlorine pesticides (OCPs) such as HCH, DDT, endosulfan, heptachlor and aldrin in river Hooghly (RH) and river Brahmaputra (RB). It was found that in RH, HCH was predominant while in RB, DDT was more predominant. Studies have also found organochlorine pesticide residues in water samples from Unnao district, Madya Pradesh (MP) and Dibrugarh and Nagaon, Assam (ASM) (Singh et al. 2007, Mishra & Sharma 2011) (Table 3).

Artificial Sweeteners

Several studies have established the presence of Artificial Sweeteners (ASWs) in Indian water matrices. ASWs such as sucralose are widely used in India in food and beverages. The sucralose concentration in river Ganga was reported as 0.05 μg/L. Besides, the groundwater in Varanasi and Ramnagar were also found to be contaminated with sucralose even at a depth greater than 140 meters (Lapworth et al. 2018). The contribution of ASWs in three WWTPs in Chennai, India was in the range 9.7% to 17.1%, unlike USA WWTPs which accounted for 49.8%. The removal of artificial sweeteners in WWTP was low where Hydraulic Retention Time (HRT) was less while additional removal was observed with increase in HRT. The removal might be due to physical adsorption and flocculation process. The highest sucralose concentrations in the influent and effluent of WWTPs were 1.9 μg/L and 1.87 μg/L indicating very low removal. The concentration of acesulfame was found as 0.89 μg/L in the influent of WWTPs (Anumol et al. 2016). Sharma et al. (2019) investigated the presence of ASWs in river Ganga and groundwater in the vicinity. Saccharine was found at 0.085 μg/L and sucralose had 100% detection frequency in River Ganga and groundwater at concentrations of 0.023 μg/L and 0.024 μg/L respectively. The concentration in the river water and groundwater were nearly equal. It could be due to the high mobility and load of sucralose which accumulates in the aquifer. Acesulfame and cyclamate were also detected in the river and groundwater. From the previous studies, it can be inferred that sucralose, saccharin and acesulfame are predominant ASWs. Fig. 2 shows the prime ECs observed in Indian water.

REGULATORY STANDARDS

To ascertain the risk of ECs, toxicity endpoints are required. Hence, extensive toxicological studies need to be carried out. There is a paucity of regulatory standards on various classes of ECs. In many countries, there are no proper regulations on ECs due to shortage of toxicological data. In India, the drinking water standards include the regulatory standards for only a few pesticides in the Indian Standards (IS 10500). The water treatment plants and WWTPs are not monitored for the presence of ECs. World Health Organisation (WHO) has published extensive regulatory standards for several classes of ECs. The European Union (EU) Water Framework Directive has listed 45 priority compounds with environmental quality standard to be adhered in aquatic environments. The United States Environmental Protection Agency (USEPA), under the Safe Drinking Water Act regulates the ECs listed on the Contaminant Candidate List (CCL) which have been detected in the public water system and has not yet been regulated. The USEPA regulates at least five contaminants from the CCL every five years. It is also required to issue a new list of not more than 30 unregulated contaminants to be monitored by public water systems every five years. CCL 4

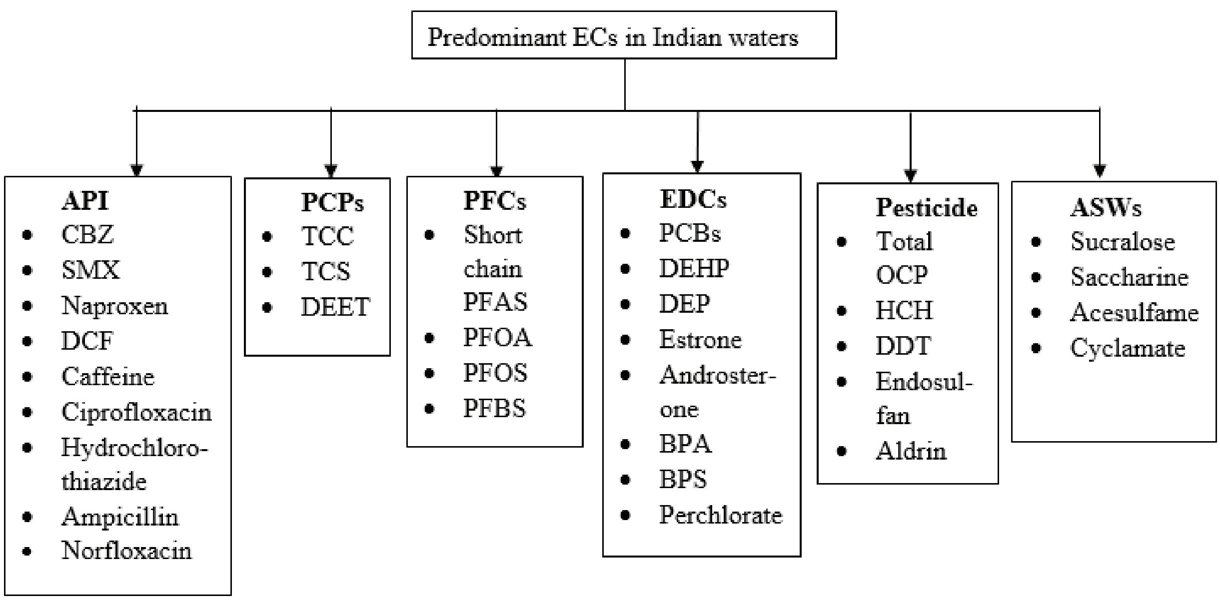

Fig. 2: Predominant ECs from different classes found in the Indian water environment.

Table 4: Regulation of ECs as per WHO, USEPA, EU and IS 10500 (µg/L).

ECs	WHO	USEPA	EU	IS 10500
Pesticides				
DBCP	1	0.2		
2,4-D	30	70		30
Chlordane	0.2	2		
Lindane	2	0.2		2
Chloropyrifos	30	10	0.1	30
DDT	1		0.025	1
Mecoprop	10			
Pentachlorophenol	9	1	1	
Endrin	0.6	2	0.01	
Alachlor	20	2	0.7	
Terbuthylazine	7			
Other Chemicals				
Perchlorate	70	15		
Toluene	700	1000		
1, 4-Dioxane	50			
Benzo (a) pyrene	0.7	0.2	0.1	0.1
Styrene	20	100		
Tetrachloroethene	40			
Vinyl chloride	0.3	2		
Xylenes	500	10,000		
Trichloroethene	20			
Polychlorinated Biphenyls		0.5		0.5
PFOA/ PFOS		0.07	36	
Dichloromethane	20		20	
Pharmaceuticals				
Edetic acid	600			
Disinfection By-products				
Bromate	10			
Bromodichloromethane	60			60
Bromoform	100	80		100
Chlorate	700			
Dibromoacetonitrile	70			
Dibromochloromethane	100	80		100
Dichloroacetonitrile	20			
Dichloroacetate	50	60		
Chloroform	300	80		200
N-Nitrosodimethylamine	0.1			
Trichloroacetate	200			
2,4,6-Trichlorophenol	200			

is the latest list which includes 97 chemicals or chemical groups and 12 microbial contaminants. Table 4 shows the regulation of ECs by WHO, USEPA, EU and IS 10500 (μg/L). The regulation on ECs is critical as it is important in monitoring and mitigating the ill effects of ECs on human health and the environment.

TREATMENT TECHNOLOGIES

Improvement in lifestyle, urbanisation, agricultural and industrial practises has resulted in an increase in the load of ECs into the water environment. The control of ECs is possible through the incorporation of efficient and cost-effective treatment technologies for the removal of ECs. Previous studies employing adsorption, biological treatment and advanced oxidation processes (AOPs) for the abatement of some priority ECs are discussed.

Adsorption

Adsorption of ECs onto the adsorbents is a way to separate the ECs from the water matrix while it presents no significant treatment and reusability of adsorbents is a challenge. Several studies have investigated the various adsorbents for the adsorption of ECs (Sophia & Lima 2017). The commonly employed adsorbent is activated carbon. Sharipova et al. (2016) studied the adsorption of triclosan using activated carbon sorbents. The adsorption capacity is 85 mg/g. Mestre et al. (2014) studied the adsorption of ibuprofen, paracetamol, acetylsalicylic acid, clofibric acid, caffeine and iopamidol using activated carbon from industrial pre-treated cork. Ibuprofen had high sorption capacity of 174.4 mg/g. Removal efficiencies were between 40 and 90%. Salman (2014) studied the use of activated carbon from palm oil fronds as adsorbents for the adsorption of Pesticides - Bentazon, Carbofuran, 2,4-Dichlorophenoxyacetic acid (2,4-D), the removal percentages were 8.2, 1.3 and 9.2% respectively. Suriyanon et al. (2013) studied the adsorption of two pharmaceuticals DCF, NSAID, and CBZ, antiepileptic using silica-based porous material and powdered activated carbon. Low sorption capacities of 31.93 μg/g (DCF) and 27.59 μg/g (CBZ) were reported. Rodríguez-Liébana et al. (2016) studied the adsorption of fungicides, Metalaxyl and Fludioxonil, using natural clay as adsorbent. It was found that Metalaxyl had a high adsorption capacity of 284±4 μg/g, while Fludioxonil had an adsorption capacity of 176±1 μg/g. Han et al. (2012) studied the adsorption of bisphenol-A on lignin from black liquor of paper mill industry. The adsorption capacity observed was 237.07 mg/g. Rajapaksha et al. (2014) studied the adsorption of sulfamethazine on tea waste biochar. The sorption capacity determined was quite low (33.81 mg/g).

Biological Treatment

Biological treatment using constructed wetlands and activated sludge processes have been evaluated by several studies. Ramprasad & Philip (2015) studied the removal of surfactants and personal care products such as sodium dodecyl sulphate (SDS), propylene glycol (PG) and trimethylamine (TMA) from greywater using a hybrid constructed wetland (HYCW). The pilot-scale HYCW system planted with *Phragmites australis* was fed continuously with greywater at a flow rate of 2.5 cu.m/day. It was found that HYCW was capable of removing SDS (94-98%), PG (95-98%) and TMA (95-99%). Kruglova et al. (2014) studied the biodegradation of ibuprofen, DCF and CBZ using nitrifying bacteria activated in sequential batch reactors at 12^0 with the initial concentration of pharmaceuticals as 20±10 μg/L and solids retention time was 10-12 days. It was found that ibuprofen and DCF were biologically degraded while carbamazepine was not biodegraded. Tiwari et al. (2014) studied the biotransformation of chlorpyrifos (CPF), organophosphate insecticide, in aqueous medium under 15 days aerobic and 60 days anaerobic batch experiments. At the end of the batch experiments, 2.78 ± 0.11 mM of CPF was degraded by 82% aerobic and 66% anaerobic aqueous environments.

Advanced Oxidation Processes

AOPs are a promising technique in the degradation of ECs such as pesticides and API. Glaze et al. (1995) are one of the early studies for the removal of 1,2-dibromo-3-chloropropane (DBCP), pesticide using H_2O_2/UV. DBCP is spiked at 0.3 mg/L in distilled water, initial concentration of H_2O_2 was 1.8mg/L and 3.3 mg/L, pH 8-8.2 and UV lamp (254 nm) was used. It was observed that nitrate and bicarbonate/carbonate alkalinity have a detrimental effect on the rate of oxidation of DBCP, the former due to UV shielding and the latter due to hydroxyl radical scavenging. Carbonate Alkalinity (C_T) 0.1 mM and reaction time 19 minutes resulted in 95% removal. Though the H_2O_2/UV system is powerful in oxidising by the generation of hydroxyl radical, residual H_2O_2 increased chlorine demand. It can be quenched by treatment using Granular Activated Carbon (GAC). Smaller size GAC would be more effective (Huang et al. 2018). Studies have been carried on the effectiveness of AOPs in wastewater and drinking water treatment. Ternes et al. (2003) investigated the ozonation in the removal of pharmaceuticals, contrast media and musk fragrance from effluents of a German municipal WWTP. By applying 10-15 mg/L ozone for contact time 18 minutes, all the pharmaceuticals were completely removed. For effective degradation, the ozone concentration should be equal to the dissolved organic carbon value. Asha & Kumar (2015) investigated sulfamethoxazole (SMX) removal from the poultry wastewater using a continuous-mode membrane-photocatalytic slurry reactor. SMX concentration in poultry wastewater varied from 0 to 2.3 mg/L. The optimized condition for complete SMX removal was 125 minutes of hydraulic retention time and GAC-TiO_2 catalyst dosage 529.3 mg/L.

Few studies have reported the use of Tungsten Trioxide (WO_3) as a visible light photocatalyst. It can provide sustainable solutions in water treatment due to its low cost, low toxicity, ease of availability and visible light activity owing to its low band gap. Nishimoto et al. (2010) studied the degradation of phenol under different combinations such as O_2/vis/WO_3, O_3/dark/WO_3, O_3/vis, O_3/ vis/WO_3 and O_3/dark systems. The study was carried out at ozone dose of 0.45 g/hr, xenon lamp of wavelength > 420nm, 0.6 g of WO_3 and 250 mL of phenol solution (200 ppm). It was found that O_3/vis/WO_3 system removed about 100% TOC after 120 min of treatment and the removal rate of phenol within 40 min was similar to O_3/dark/WO_3, O_3/vis and O_3/dark systems. Rey et al. (2014) investigated photocatalytic ozonation using 0.25 g of composite catalyst TiO_2 and WO_3 (4%) for the removal of ibuprofen, metoprolol and caffeine. The study was carried at an ozone concentration of 10 mg/L and ozone flow rate of 0.34 litres per minute (lpm) by spiking 4 mg/L ECs, the irradiation time of 2 hours and absorbance at the upper visible region (300-350 nm). It was observed that TiO_2-WO_3 had higher efficiency than TiO_2 alone. Ozonation alone was equally effective. The parent ECs were completely removed in 40 mins and 64% TOC removal was achieved in 2 hours. Zhu et al. (2018) studied the degradation of SMX, a veterinary drug, using visible light WO_3-CNT photocatalyst. The light source was solar simulator 300 W Xe Arc lamp. The concentrations of SMX and WO_3 were 10 mg/L and 0.5 g/L respectively. WO_3 has a lower band gap of 2.4-2.8 eV. It was observed that bare WO_3 shows light absorption around 450 nm and incorporation of CNT shifts the wavelength to the visible range. Compared to pure WO_3, all three WO_3-CNT composites showed enhanced photocatalytic activity.

Several studies have shown that integration of ozone and photocatalysis as in case of photocatalytic ozonation can enhance the removal efficiency. Rivas et al. (2012) studied the removal of nine pharmaceuticals using TiO_2 photocatalytic ozonation and in different combination. The study was carried out at an airflow rate of 0.5 lpm, initial concentration of ECs at 10 mg/L, TiO_2 dose of 0.25g/L and UV (313 nm) as the light source. It was found that TOC removal after 120 minutes was 95% for TiO_2 photocatalytic ozonation (PCO), 85% for UV and ozone, 60% for UV and TiO_2, 25% for photolytic (UV) and 30% for ozonation. PCO had the highest removal rate. The higher dose of TiO_2 causes shielding of UV rays.

Garc´ıa-Araya et al. (2010) investigated TiO_2 PCO and its combinations for DCF, NSAID, at ozone concentration of 10 mg/L, initial DCF concentration of 10^{-4} mol/L and TiO_2 concentration of 1.5 g/L. It was found that TOC removal was 50% and 80% for O_3 and TiO_2 photocatalytic systems. O_3 achieves complete removal in less than 7 mins (5% TOC removal). The O_3/UV-A/TiO_2, pH 7, removed DCF (90-95% in 10 mins) and TOC (90% in 15 min).

Rajeswari & Kanmani (2009) studied the TiO_2 PCO of carbaryl pesticide. Optimum values for maximum degradation were 40 mg/L of carbaryl (500 mL) at pH 6 with 0.28 g/h of ozone and 1 g/L of TiO_2. The mineralization rate constant in the photocatalytic ozonation process was 0.09/min while the sum of the mineralization rate constant of photocatalysis and ozonation was only 0.05/min. 92% COD reduction and 76.5% TOC removal were achieved in 180 min. The BOD_5/COD ratio was increased to 0.38.

CONCLUSION

The past studies have established the presence of ECs such as API, PCP, ASW, pesticides and EDC across the Indian water environment. It was observed that certain ECs are predominant and ubiquitous. It includes carbamazepine, sulfamethoxazole, caffeine, triclosan, triclocarban, perchlorate, sucralose and organochlorine pesticides. The regions near the bulk drug manufacturing hub in Hyderabad was found to contain high levels of antibiotics, API and ARG. Several studies have shown ECs in surface water, groundwater, drinking water, treated wastewater from STP and CETP. EDCs such as plasticisers, PCB, steroid hormones were found in water. DEHP was the commonly observed phthalate. ECs can cause harmful impacts on aquatic, terrestrial wildlife, vegetation and human communities. It can persist, bioaccumulate and impart toxicity. Several reports show that biological treatment and adsorption imparts low removal of ECs. Advanced oxidation processes using visible light-driven photocatalytic ozonation is found very effective for the removal of most ECs except PFCs which can be easily removed by electrochemical methods. Hence, the review concludes that incorporation of AOPs in STP, CETP and WTP will aid in the removal of ECs.

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