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REVIEW ARTICLE

Degradations of endocrine-disrupting chemicals and pharmaceutical compounds in wastewater with carbon-based nanomaterials: a critical review

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Abstract

Although water occupies 75% of the earth's surface, only 0.0067% of the total water is available for human activities. These statistics further decline with the population growth and consequent multiplication in the amount of annual waste produced. The demand for clean and safe drinking water has always been a prime concern in the global scenario. Among various types of waste materials, endocrine-disrupting chemicals (EDCs) and pharmaceutical effluents have become a constant threat to the aquatic ecosystem and possess challenges worldwide. Endocrine-disrupting chemicals (EDCs) are a mixed group of emerging concern chemicals with the ability to mimic the mechanisms of biosynthesis, transport, and metabolism of hormones. These chemicals pose various health threats such as early puberty, infertility, obesity, diabetes, reproductive disorders, cancerous tumors, and related disorders (immune cells, hormones' activity, and various organs). On the other hand, pharmaceutical compounds such as antibiotics also harm the natural environment, human health, and soil microbiology. Their low concentration, ranging from a few ng/L to μg/L, gives rise to a micro-pollution phenomenon, which makes it difficult to detect, analyze, and degrade in wastewater treatment plants. Activated carbons (ACs) and other adsorbents, including naturally occurring materials (wood, keratin) are considered as nanomaterials (NMs) reference for the separation of organic pollutants. It is generally acknowledged that mass-transfer phenomena control sorption kinetics at the liquid/solid interface, with retention controlled by the sorbent/sorbate properties. Therefore, the type of interaction (strong or weak van der Waals forces) and the hydrophilic/hydrophobic properties of the adsorbent are two crucial factors. Besides, EDCs and pharmaceutical compound sorption on such kinds of nanoporous solids depend on both the molecule size and charge density. The applications of nanomaterials on non-conservative methods, like advanced oxidation processes or AOPs (e.g., photocatalysis and Fenton reaction), are contemplated as more apt in comparison to conservative technology like reverse osmosis nanofiltration, and adsorption, etc. One of the reasons is that AOPs generate free radicals (hydroxyls), which are strong oxidants for the demineralization of organic compounds and the extreme case that hydroxyl radicals can attack any kinds of pollutants with the generation of only water and carbon dioxide as final products. AOPs may imply the use of NMs as either catalysts or photocatalysts, which improve the selective removal of the target pollutant. Therefore, various literature reviews have revealed that there is a timely need to upgrade the efficiency of the remediation approaches to protect the environment against EDCs and pharmaceuticals adequately. There is currently a lack of definitive risk assessment tools due to their complicated detection and associated insufficiency in the health risk database. Hence, our present review focuses on applying carbon-based nanomaterials to remove EDCs and pharmaceuticals from aqueous systems. The paper covers the effect of these pollutants and photocatalytic methods for treating these compounds in wastewater, along with their limitations and challenges, plausible solutions, and prospects of such techniques.

Keywords Endocrine-disrupting compound · Pharmaceutical waste · Carbon-based materials, Advance oxidation processes

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Abbreviations

EDCs	Endocrine disruptor chemical
PPCPs	Pharmaceuticals and personal care products
AOPs	Advanced oxidation processes
WHO	World Health Organization
U.F.	Ultrafiltration
POPs	Persistent organic pollutants
G.O.	Graphene oxide
CNT	Carbon nanotube
A.C.	Activated carbon
MWCNT	Multi-walled carbon nanotubes
SWCNT	Single-walled carbon nanotubes
PANI	Polyaniline
CBM	Carbon-based nanomaterial
GR	Reduced graphene oxide
BPA	Bisphenol A
DOMs	Dissolved organic materials
ROS	Reactive oxygen species
CBZ	Carbazipame
NSDC	Nitrogen and sulfur-doped porous carbon
DMP	Dimethyl paraben

Formulae

Titania	TiO_2
Zinc Oxide	ZnO
Copper oxide	Cu_2O
Silver Orthophosphate	Ag_3PO_4
Bismuth Vanadate	$BiVO_4$
Graphitic Carbon Nitride	g-C_3N_4
Zinc Chloride	$ZnCl_2$

Introduction

Water, one of the essential requirements for the survival of all life forms and the most valuable natural resource for human civilization since ancient times, is the most widely discussed topic globally. Constant access to clean and safe water is one of the primary concerns of the modern-day world. With the limited sources of potable water, the world faces a significant challenge in providing the same global mass. The necessity of good-quality and potable drinking water cannot be undermined; otherwise, human health would be at risk and compromised with safe drinking water (Zubair et al. 2020). Safe and pure water is being endangered due to various anthropogenic activities that include primarily the disposal of effluents containing emerging micro-pollutants known to persist in different water sources. Human health, industries, agriculture, and the economy are highly dependent on water for their sustainability (Dhote and Ingole 2012; Hutton et al. 2007).

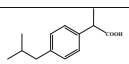
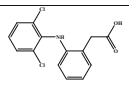
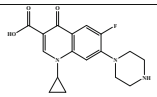
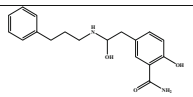
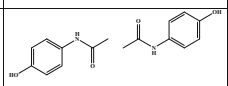
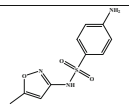
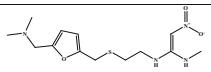
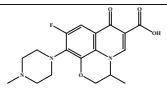
On the other hand, various factors like population growth, industrialization, and urbanization are fiercely creating stress on hydrological resources (UNESCO 2015). Eutrophication of lakes and rivers, biomagnification of pollutants, and dead zones in water bodies with the ever-increasing use of fertilizers and other chemical substances further add up to deteriorate the present situation of water scarcity (UN-Water 2016). Besides, the negligence of recycling wastewater and the absence of Government policies or non-implementation of existing policies create hurdles in achieving goals (Corcoran et al. 2010). The lack of clean water reflects on human health and hygiene, environment, economy, and society. According to the WHO (2017) reports, 785 million people still lack basic drinking-water service, and 144 million population of this is mainly dependent on surface water. More than 8 million people every year die because of diarrhea caught through contaminated drinking water. Reports also showed that nearly half of the population in this world would be living in "water-stressed" regions by the end of 2025.

The various sources of water pollution range from industrial wastes, domestic, agricultural to health-based effluents usually contain dyes, paints, oil spills from ships, industrial runoff, mining, and metallurgical leaches, fertilizers, pesticides, pharmaceutical wastes, cosmetic wastes, etc. have been adding to water and making it unsuitable for drinking and other applications. The steady rise in waste produced by anthropogenic activities and subsequent release of these into the environment negatively impacts the overall scenario. The advancement in analytical techniques for detecting these chemicals present even at trace amounts helps remove the pollutants from water bodies. A significant class of pollutants at low concentrations includes endocrine disruptor chemicals (EDCs) and pharmaceuticals and personal care products (PPCPs). Disposal of pharmaceutical wastes was earlier limited for preventing accidental exposure to children or animals. Hence, pharmaceuticals were being washed away into the sewage systems or disposing of them off to municipal trash. Besides, due to the overuse of common antibiotics like quinolones, the drug and their metabolites are emerging at the toxic level in all types of water bodies, soils, sediments, and sewages (Du et al. 2018). The gradual introduction of these wastes into the environment began to surface. Various routes (such as flushing off body wastes, disposal of expired products, leaching off chemicals from landfills, agricultural wastes, and chemicals runoff) of penetration into the environment have been studied. Hignite and Azarnoff (1977) reviewed the detection of several drug metabolites, such as salicylic acid, in sewage treatment plants (influent and effluent systems). There have been many such studies since that time for the presence of drugs and their metabolites in water cited in the references (Wilkinson et al. 2017; Yang et al. 2017). Highly sensitive analytical techniques (ppb and ppt level) for detection and quantitative analysis of pharmaceutical products present in biological systems and the environment have been developed since then, thereby eradicating these wastes from the ground. The term "PPCPs" has been coined by Daughton and Ternes (1999) for such classes of chemicals that are

entering into the system through medications and products of daily use such as cosmetics. PPCPs include chemicals such as acetaminophen, acetylsalicylic acid, carbamazepine, fenoprofen, musk xylene, musk ketone, triclosan, butyl acetate, diethanolamine, 1,4-dioxane, etc. which are directly or indirectly have become an indispensable part of our regular used products and are consequently being added to the environment through various means (Fénichel and Chevalier 2017). Sewage treatment methods cannot altogether remove environmental persistent pharmaceutical pollutants (EPPP). They can further enter into the aquatic systems by discharge from sewage treatment units (Hirsch et al. 1998; Anderson et al. 2004; Combarnous 2017). Some of the common pharmaceuticals that have been found in wastewater and their effect on human health have been listed in Table 1 (Cerro-Lopez and Mendez-Rojas 2017; Hirsch et al. 1998).

Endocrine-disrupting chemicals (EDCs), on the other hand, belong to a class of chemicals that mimic the hormonal activities and therefore interfere in the proper functioning of endocrine systems (synthesis and regulation of hormones). These interferences can lead to tumors (cancerous), birth defects, a developmental disorder such as learning disabilities in kids, severe attention deficit disorder, cognitive child development, brain development problems; deformation in the body parts such as limbs; cancer (breast cancer, cervical cancer, prostate cancer, thyroid cancer, etc.); issues in sexual development of human beings such as de-masculinization of male gonads or de-feminization of female; etc. (Lucacioni et al. 2020). EDCs have been introduced into the environment through either natural or anthropogenic pathways or both. These mainly include pharmaceuticals products such as dioxins and their derivatives, polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT), and few other pesticides plasticizers (bisphenol A), Di(2-ethylhexyl) phthalate (DEHP), and phytoestrogens (plant-based hormones). Recently, experiments have been performed to investigate the cytotoxicity due to the photodegradation by-products of 17β-estradiol, bisphenol A, and 17-α ethynilestradiol; there have been cases of inhibition of estrogenic activities of the cellular cultures (Saggioro et al. 2019). These chemicals are continuously being added to the ecosystem in various forms: plastic containers, metal cans, soaps and detergents, fire retardants, foods, toys, cosmetics, and pesticides. Literature reflects that EDCs may pose a high risk during organ and neural system

Table 1 Common pharmaceuticals present in wastewater and their health hazard (Cortés Muñoz et al. 2013)

Examples of common Pharmaceuticals in Wastewater	Structure	Effects
Ibuprofen		Hypertension, myocardial infarction, dizziness
Diclofenac		headache, dizziness, drowsiness
Ciprofloxacin		Tendinitis, seizures, headache, long-term carcinogenicity
Metoprolol		Bradycardia, hypotension, arrhythmias
Acetaminophen		Liver damage, Asthma
Sulfamethoxazole		Nausea, vomiting, skin rashes, anorexia
Ranitidine		Dizziness, somnolence, insomnia, vertigo, arrhythmia, liver failure
Ofloxacin		Nausea, headache, insomnia, neurological disorders

development, i.e., prenatal and early postnatal development (NIEHS). Some of the common EDCs and their adverse effects are discussed in summarized form in Table 2 (Dodson et al. 2012; Fucic et al. 2018; Homes n.d.; Barrios-estrada et al. 2018).

A wide range of techniques have been studied and experimentally applied to treat EDCs and pharmaceutical wastes, from conventional methods such as adsorption, micro- and ultrafiltration, ozonization, coagulation-flocculation chlorination, etc., to the modern techniques such as advanced oxidation processes (AOPs), photocatalysis, and biodegradation (Vieira et al. 2020). With the help of modern technologies, such waste treatment is gaining popularity due to cost-effectiveness and the absence of side product generation. However, none of these techniques is useful in the complete removal of waste. It has been reported that ozonation results in the production of some by-products, which may be even more toxic than the parent/target molecule (Klavarioti et al. 2009; Esplugas et al. 2007; Iervolino et al. 2020). Activated carbon and biochar are common adsorbents that have been extensively explored to remove these chemicals. Their application is limited to laboratory scale, and implication on a large scale has not been achieved (Jung et al. 2013).

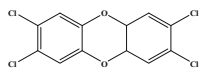
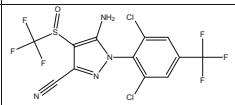
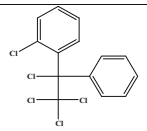
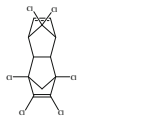
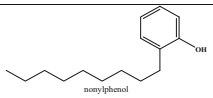
On the other hand, newly developed nano-based adsorbents need extensive knowledge in interaction with the pollutant moiety and interfacial chemistry of the system (Rodriguez-Narvaez et al. 2017). Membrane separation techniques, such as ultrafiltration (U.F.), depend primarily on the chemicals' polarity that needs to be removed. Mostly polar and highly water-soluble compounds are effectively removed by membrane separation techniques than the non-polar and less water-soluble compounds. Some female hormone-based drugs like EE2, estrone, E2, and ketoprofen are removed efficiently, while phthalate esters can be withdrawn at a much slower rate (Vieira et al. 2020). It can be observed that treatment techniques for EDCs and PPCPs are still lagging in several areas. However, AOPs are the most promising treatment methods with assured complete demineralization of these compounds. Heterogenous photooxidation has been established as one of the best techniques for the removal and treatment of many recalcitrant organic compounds with high efficiency (Singh et al. 2016).

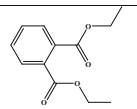
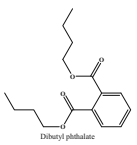
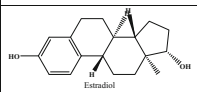
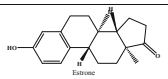
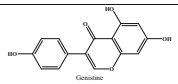
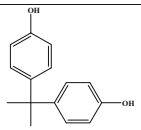
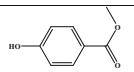
Photocatalysis has emerged as another most advanced and highly reliable technique for pollution abatement in recent years. Extensive research has been carried out to develop catalysts to demineralize a wide range of organic pollutants (dyes, paints, drugs, petrochemicals, aromatic compounds, etc.) in the presence of light (U.V./Visible). This method has been considered a "clean technology" for dealing with pollutants without using any toxic chemicals but with many satisfactory results compared to other conventional pollution tackling techniques (Koe et al. 2020). Even though biodegradation has been effective in the case of organic pollutants and domestic waste treatment, EDCs and PPCPs are not biodegradable due to their low solubility and high cytotoxicity on the cellular level (Lei et al. 2017; van Drooge et al. 2017). The highly stable structure of these chemicals establishes them as persistent organic pollutants (POPs). The need for sustainable development and a "Green World," there is an utmost requirement of eco-friendly, cost-effective, advanced, and alternative conventional methods for treating pollutants, with less energy consumption and minimum chemical use. AOPs serve as highly reliable and quite promising techniques for treating and removing POPs from aqueous systems. Their robustness and high demineralizing capability of organic compounds lay a firm ground for proving their role in curbing such stable chemicals. Transition metal oxides (single or mixed), e.g., Titania (TiO_2), Zinc Oxide (ZnO), and Copper Oxide (Cu_2O) or in combination with carbon-based nanomaterials, i.e., graphene oxide (G.O.) or carbon nanotube (CNT) combined with metal oxide-based components (TiO_2, WO_3, Nb_2O_5, Co_3O_4, and Cu_2O) form a significant class of photocatalysts. Carbon-based nanomaterials are extensively discussed in this review for the photocatalytic degradation of organic pollutants, which are difficult to be treated otherwise (Zhao et al. 2018; Canle et al. 2012; Canle et al. 2013; Swaminathan et al. 2013).

Over the past few decades, the discovery and development of carbon-based nanostructures are found to be versatile and crucial in different technological applications. Apart from enormous natural abundance, various allotropic forms of carbon and their structures like activated carbons, charcoal, Fullerene, graphene oxide, graphene, and carbon nanotubes (CNTs), etc., exhibit different physical and chemical properties. Recently, they have emerged as essential classes of nanomaterials in wastewater treatment due to their large surface areas, high electron mobility, thermal conductivity, and mechanical strength, and also high charge carrier mobility (Santhosh et al. 2016; Ye et al. 2019). Their application has three major stages: adsorption, ultrafiltration, and photocatalysis (Kurwadkar et al. 2019). In recent times, such nanocomposites and semiconductor materials have emerged as active photocatalysts in pollution abatement. The low toxicity and high surface area promote the application of these nanomaterials in environmental remediation. CNTs-based nanomaterials involving TiO_2 as semiconductor metal oxide, cadmium sulfide (CdS), silver orthophosphate (Ag_3PO_4), bismuth vanadate ($BiVO_4$), stannate (SnO_2), and polymer-CNTs nanocomposites have also played an essential role in photocatalytic degradation of POPs (Kim and Park 2011; Xie et al. 2015; Ye et al. 2019; Wang et al. 2012).

Interestingly, binary (polymer-CNTs) and ternary composites (polymer-CNTs-metal/metal oxide/metal sulfides) have shown efficient photocatalytic activity against harmful chemicals present in water. In this report, all kinds of nanocomposites and their conventional application methods will be

Table 2 Adverse effects on human health of common EDCs

Class	Chemical example(s)	Sources/Origin	Effects
Polychlorinated Compounds	2,3,7,8-Tetrachlorodibenzodioxin 	Incineration, Leaches	Diabetes, effects on the immune system, Central nervous system damage, Thyroid disorders
Insecticides	 Fipronil	Agricultural chemicals	Possible Human Carcinogen (Thyroid Tumors)
Organochlorine Pesticides	 Dichlorodiphenyltrichloroethane ,  1,2,3,4,10,10-Hexachloro-1,4,4a,5,8,8a-hexahydro-1,4:5,8-dimethanonaphthalene	Agricultural runoff / Atmospheric transport	Neurotoxicity Hepatocarcinogenity
Alkylphenols	 nonylphenol	Surfactants and washings of industries	Mimics female hormones and affects pregnancy

Class	Chemical example(s)	Sources/Origin	Effects
Phthalates	 Diethyl phthalate ,  Dibutyl phthalate	Industrial effluent	Increases Quantity of damaged DNA in sperm decreases sperm motility, reduces male fertility
Natural Hormones (Produced naturally by animals)	 Estradiol	Municipal effluents	Breast cancer, ovarian cancer, and endometrial cancer
Synthetic steroids	 Estrone	Contraceptive pills (added through municipal wastes or industrial effluents from the pharmaceutical industry)	Abnormal sexual development in teenagers and functioning of the prostate
Phytoestrogens (found in plant material)	 Genistine	Pulp mill effluents	Rashes, itching, and anaphylaxis, tiredness, nausea
Plastics (bisphenol A)	 4,4'-(propane-2,2-diyl)diphenol	Plasticizers in plastic products	Infertility
Cosmetics, personal products	 Methylparaben	Through domestic wastes and sewage systems	Disrupts hormonal function, increases the risk of breast cancer and reproductive toxicity.

reviewed and are believed to provide deep insight into the application of carbon-based nanocomposites in combating such toxic organic pollutants. Various applications of carbon-based nanocomposite are shown in Fig. 1.

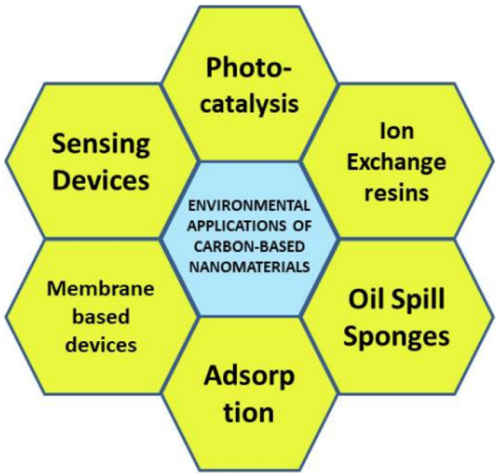


Fig. 1 Environmental application of carbon-based nanocomposites (Sarkar et al. 2018)

Removal of EDCs and pharmaceutical wastes from aqueous systems

Many conventional techniques have been developed to remove these pollutants at the laboratory and the industrial level. The most common PPCPs, i.e., ibuprofen and diclofenac, are non-biodegradable and challenging to eliminate from water in conventional wastewater treatment plants. Their polar structure and low solubility in water make it difficult to remove them from the system. Thus, alternative methods for treatments such as ultrasonic treatment, degradation techniques, solid-phase extraction via molecular recognition, solid extraction, electrochemical methods, etc were used. However, these methods do not provide a guaranteed removal of wastes (Bhadra et al. 2017).

Carbon-based nanocomposites: a new-breakthrough

Carbon is one of the most abundant elements on the earth's crust. For the past 25 years, carbon-based nanomaterials have been extensively used in pollution remediation. Carbon-based nanomaterials such as graphene, fullerenes (Buckminsterfullerene), CNTs (multi-walled and single-walled), and graphitic carbon nitride (g-C3N4) have emerged as major thrust areas for research. Their extending application varies from Li-ion batteries to semiconductors, sensors to molecular imaging, and catalysis to energy storage, resulting in pollution control (Kumar et al. 2017; Lu and Astruc 2018). Carbon-based nanomaterials have been preferred for their low or non-toxicity and hence, are considered environmentally benign. Due to high sorption capacities, their role in removing toxic metal ions from wastewater has been immensely focused (Mauter and Elimelech 2008; Tofighy and Mohammadi 2011). The enhanced photochemical activity of carbon nanomaterials combined with semiconductor photocatalysts is designed using conduction band electrons or valence band holes of a photocatalyst (Ng et al. 2012). Significant attention is being paid toward the utilization of these carbon nanomaterial-based semiconductor photocatalysts in environmental remediation. Their incorporation in photocatalysts as composite creates a hydrophobic microenvironment that localizes the reactants and increases concentration. This process prohibits the recombination of charges and promotes photocatalytic radical generation (Robel et al. 2005). The graphitic carbon acts as a sensitizer on forming an effective complex with TiO_2 at the interface (Wang et al. 2005) or the addition of TiO_2 as an impurity to create Ti–O–C bonds that enhance the light absorption region (Kaur et al. 2016). An increment in carbon nanotube performance was reported up to 90% for the photodegradation of gas-phase benzene (Tang et al. 2011). CdS nanoparticles loaded on graphene sheets (Zhang et al. 2011a), TiO_2, and ZnO nanoparticles incorporated on graphitic sheets were studied for their photocatalytic activity the microbial inactivation (Akhavan and Ghaderi 2009; Akhavan et al. 2011). The d-p interaction leads to electron coupling where the d-orbital of TiO_2 and p-orbital of carbon overlap and reduces the energy of absorption, and hence absorption occurs at a higher wavelength. According to Ng et al. (2012), shuttling of the electrons occurs between the graphitic carbon and semiconductor photocatalysts (electron cycle for Z-scheme water splitting).

Given below is a report (Table 3) originally compiled by Smith and Rodrigues 2015showing the pollutant removal capacities of different types of carbon-based nanomaterials and composites along with the respective mode of removal. In the present review, the table has been modified by adding more recent studies.

From the above table, it can be briefly known that organic functionalization of carbon-based nanomaterials can enhance these nanomaterials' adsorption capacities (Minella et al. 2017b). For example, G.O. and carbon nanotubes properties can be escalated by functionalizing them with –O.H. and –COOH groups, respectively. This process can result in increased dispersing quality, thereby increasing the available surface area. More surface exposure of these nanocomposites means more space open for better removal activity of microbial and chemical contaminants (Ji et al. 2012; Inyang et al. 2015). As shown from the table, the amalgamation of metal or metal oxides in carbon-based nanocomposites enhances the disinfecting capability and adsorption properties of nanomaterials. According to the target contaminants, carbon-based materials' surface charge can be redesigned to improve the electrostatic interaction and sorption capacity. The surface charge property of these materials can also be modified based on the target pollutant to enhance qualities like electrostatic interaction and sorption capacity (Zhang et al. 2013b; Zhang et al. 2011a; Ahmmad et al. 2014). It has also been reported that the mesopores are more efficient than the micropores for accommodating varied sizes of pollutants removal. In

Table 3 Modified reports of Smith and Rodrigues (2015) on remediation capabilities of various carbon-based nanomaterials and composites for common types of water contaminants

Nanomaterials/nanocomposites	Chemical and biological removal	Capacities	References
SWNT	NOM, metals, bacteria, viruses	22–26 mg/g NOM, 35 mg/g Pb^{2+} 3.18×10^{12} CFU/mL	Canle et al. 2012, 2013; Santhosh et al. 2016
MWNT	Synthetic organics, metals	20 mg/g Pb^{2+}	Kurwadkar et al. 2019; Esplugas et al. 2007
G	Bacteria, metals, synthetic organics	85% inactivation, 22–35 mg/g Pb^{2+} 154–204 mg/g methylene blue	Jung et al. 2015; Kanel et al. 2015; Joseph et al. 2011
G.O.	Biomolecules, metals, bacteria, synthetic organics	350 mg/g lysozyme, 365–659 mg/g Pb^{2+}, 70–95% inactivation, 351–1939 mg/g methylene blue	Jung et al. 2015; Kanel et al. 2015; Hu and Cheng 2015; Ahmed et al. 2015; Tian et al. 2013; Zhang et al. 2011b; Ma et al. 2015
Poly-N-vinyl carbazole- SWNT (OVK-SWNT)	Bacteria	> 80% inactivation	Inyang et al. 2015
PVK-G	Bacteria	75–80% inactivation	Kanel et al. 2015; Joseph et al. 2011
PVK-GO	Metals, bacteria	888 mg/g Pb^{2+} 90–95% inactivation	Kanel et al. 2015; Joseph et al. 2011; Tian et al. 2013
GO-TiO2	Metals	66 mg/g Pb^{2+}	Jung et al. 2015
GO-EDTA	Metals, bacteria	455–525 mg/g Pb^{2+} 60–95% inactivation	Kanel et al. 2016; Zhang et al. 2011c; Ma et al. 2015
Carbon nanotubes/Ce-TiO_2 (CNT/Ce-TiO_2)	Organics	94% phenol degradation	Staff 2019
CNT/TiO_2	Synthetic organics	40% of degradation methylene blue	Wang et al. 2015b
CNT/Fe-Ni/TiO_2	Synthetic organics	90 % degradation of methylene blue	Peng et al. 2012
G-Fe_3O_4	Synthetic organics	89 mg/g fuchsine	Kumar et al. 2017; Jung et al. 2015
MWNT/ZnO	Organics	100 % degradation acetaldehyde	Solomon et al. 2012
MWNT-Al_2O_3	Metals	37.5 mg/g Pb^{2+}	Esplugas et al. 2007
CS/MWNT	Metals, synthetic organics	450 mg/g Pb^{2+} 450 mg/g Congo Red	Carré et al. 2014
Chitosan/G.O. (CS/GO)	Metals	77 mg/g Pb^{2+}	Kabra et al. 2004
Poly(vinyl alcohol)-MWNT (PVA-MWNT)	Organics	98.8% organic solute rejection	Li et al. 2012
Polysulphone-MWNT	Cd^{2+}	78% improved	Singh et al. 2013
MWNT-Ag	Bacteria	96% inactivation	Fujishima and Honda 1972; Trapalis et al. 2016
G-Ag	Bacteria	99.5% inactivation	Bhadra et al. 2017
GO-Ag	Bacteria	88–100% inactivation	Da Silva and Dos Santos 2017
PVA/PAA/TiO_2/G.O.	Synthetic organics	100% degradation methylene blue	Minella et al. 2017a
GO-based Fe-Mg (Hydr)oxide	Metals	617.3 mg/g Pb; 432.9 mg/g Cu; 142.2 mg/g Au; 121.7 mg/g Zn	Huang et al. 2018a; Ahmad et al. 2020
Polyethyleneimine cross-linked GO	Metals	436.3 mg/g Cr (VI)	Geng et al. 2019; Ahmad et al. 2020
Me-GO and porous carbon (PC)	Organic	288 mg/g methylene blue; 232 mg/g acid orange 7; 110 mg/g bisphenol A; 68 mg/g phenol	Mahmoud 2020

aggregated carbon, mesopores are found to be higher than conventional carbons like granular activated carbon (Wang et al. 2015a; Ji et al. 2012).

Additionally, the incorporation of the Fe-Mg hydroxide group into oxygen-containing GO can work synergistically and increases the removal capacities of heavy metal due to the synergistic effect between Fe-Mg and oxygen (Huang et al. 2018a; Ahmad et al. 2020). Mahmoud (2020) reported that the mild oxidation of Me-GO with carboxyl and lactone functional groups contributes to the high removal efficiencies and capacities of organic dye like Methylene Blue (99.7%), acid orange 7(93.6%), bisphenol A, (99.4%), and phenol (83.9). Me-GO adsorbent exhibits numerous advantages over common adsorbents as a small dose can apply over a wide range of pH and micropollutant concentration. Besides, nitrogen atoms in the adsorbents cause competitive interaction between hydroxyl ions and CrO_4^{2-} ions for adsorption. The electron pair of nitrogen from amine acted as an active site for the chelation of Cr (III) ions into adsorbent complexes. Furthermore, amine and hydrogen bonding in polyethyleneimine modified GO successfully stabilized the adsorbents and prevented GO aggregation (Geng et al. 2019; Ahmad et al. 2020).

Adsorption onto carbon-based nanomaterials

These carbon-based nanomaterials have been distinguished for their large specific surface area and active sites for adsorption. They show high selectivity and more adsorption sites, reduced intra-particle diffusion distance, easier reprocessing and reuse, and many more properties make them one of the most demanding and efficient (Kurwadkar et al. 2019). Although these carbon-based nanomaterials have been said to have such unique properties that serve to recommend their application as superior adsorbents and disinfectants over conventional methods, however, systematic review and research work on carbon-based composites at large for EDCs and PPCPs, have not been reported yet. Jung et al. 2015 have reported only CNT for its efficient adsorption mechanism for a few selected EDCs and PPCPs, as shown in Table 4. It summarizes the removal information by CNTs on selected representative classes of EDCs and PPCPs, based on literature reports. The removal efficiency was found to vary by compound and described in turn for each combination below. From the table, it can also be known that the surface area and pore volume of carbon-based compounds are essential for the removal of EDCs and pharmaceutical products. Also, the characteristics of EDCs and PPCPs play a crucial role in their removal rate in aqueous solutions. CNTs have also been reported to have hollow-cylindrical structures with a larger specific surface area, higher pore volume, and hydrophobic walls that may readily adsorb non-polar molecules, high electrical conductivity, and stable system, and better chemical properties. These qualities make them an excellent adsorbent for removing EDCs and PPCPs (Jung et al. 2015; Kanel et al. 2016). Their adsorption applicability can be used for various substrates: heavy metals, phenols, dyes, petrochemicals, etc. It is interesting to note that EDCs and PPCPs can also be removed using CNTs based nanomaterials (Jung et al. 2015). The easier regeneration of adsorbents without loss of adsorption efficiency and high absorptivity of CNTs make them excellent adsorbents (Tian et al. 2013). Pharmaceuticals such as diclofenac, amoxicillin, sulfamethoxazole, and EDCs such as bisphenol A have been effectively removed by adsorption technique using CNT-based adsorbents (Joseph et al. 2011; Hu and Cheng 2015; Ahmed et al. 2015). Many antibiotics can also be removed by application of adsorbents based on CNTs, e.g., removal of tetracycline by modified CNT adsorbents such as Pristine-MWCNT (Zhang et al. 2011c), CNTs–C@Fe–chitosan composite (Ma et al. 2015); sulphapyridine by CNT-modified biochars (Inyang et al. 2015), functionalized MWCNTs (Tian et al. 2013); sulfamethoxazole by $Co_2Fe_2O_4$-based CNTs (Wang et al. 2015a); fluoroquinolone by Graphitised multi-walled CNTs, carboxylated multi-walled CNTs, hydroxylated multi-walled CNTs (Peng et al. 2012). Metal-organic frameworks (MOFs) have also been realized as great adsorbents in recent times. Among these newly developed materials, porous carbon-derived from MOFs (PCDMs) is well recognized for the effective removal of ibuprofen and diclofenac from their aqueous solution (Bhadra et al. 2017). Bhadra and Jhung (2018) were successful in removing several pharmaceuticals and personal care products (PPCPs) *viz.* atenolol; neutral *N*,*N*-diethyl-3-methylbenzamide; chloroxylenol; and clofibric acid by adsorbing them onto Bio-MOF-derived carbons (BMDCs) at pH ranging from acidic to primary.

On the other hand, the hydrophobicity of graphene sheets allows the interaction of solutes having π-electrons with the π-electron cloud in the sheet structure. This phenomenon makes them very useful adsorbents for a wide range of aromatic compounds. A similar type of π-π coupling between π electrons of phenanthrene and graphene surfaces (Gotovac et al. 2007) and π-π electron donor-acceptor interaction between 17β-ethinylestradiol (EE2) and bisphenol A (BPA) on SWCN (Fujishima and Honda 1972) T, MWCNTs, and fullerene promoted adsorption on the surface of these carbon-based materials (Pan et al. 2008a), which are commonly observed. Ji et al. (2009a) proposed that Van der Waals-type of forces, π-π electron donor-acceptor, and cation-π bonding interactions were responsible for adsorption onto CNTs graphene surfaces of some broad-spectrum antibiotics such as tetracyclines.

Table 4 shows the modified and upgraded review work from Jung et al., where he had accumulated different studies on the adsorption of selected EDC and PPCPs. This compilation can be considered one of the pioneering reviews based on CNTs and EDCs, and PPCPs.

Table 4 Modified report of Jung et al. on the summary of selected EDCs and PPCP removal by CNT

EDC/PPCP class	Adsorbent	Surface area (m^2/g)	C_0 (μg/L)	Source water	Q_m (mg/g)	References
Analgesics						
Ibuprofen	SWNTs, MWNTs, MWNTs-O	1020, 283, 287	50–2000	Synthetic water	232, 81, 19	Cho et al. 2011
Diclofenac	MWNTs	162	NA	Pure water W.W. effluent	41, 1, 22, 3	Sotelo et al. 2012
Antibiotics						
Oxytetracycline	MWNTs	58-357	2500	Synthetic water	41-7,910[a]	(Oleszczuk et al. 2009)
Asulfapyridine	MWNTs	300	210–7300	Synthetic water	103–104[b]	Ji et al. 2009a
	MWNTs	174	100,000		600–800[b]	Xia et al. 2013
Sulfadimethoxine	MWNTs	174	100,000	Synthetic water	1300–1500[b]	Xia et al. 2013
Sulfamethoxazole	MWNTs	300	380–2500	Synthetic water	102–103[b]	Ji et al. 2009b
	SWNTs, MWNTs	410-653, 157–422	810–48,000	Synthetic water	102–103[b]	Ji et al. 2010
Tetracycline	SWNTs, MWNTs	410–653, 157–422	1400–84,000	Synthetic water	34,100–55,500 12,500–45,300[b]	Ji et al. 2010
Tylosin	MWNTs	157–422	2900–174,000	Synthetic water	11,300–33,900[b]	Ji et al. 2010
	MWNTs-H	228	20,000–100,000	Synthetic water	85	Lu et al. 2013
Antiepileptics						
Carbamazepine	MWNTs-COOH	200-400	90,000	Synthetic water	13.9c	Cai and Larese-Casanova 2016
	SWNTs	380	10–20,000	Synthetic water, including DOM	130	Lerman et al. 2012
	MWNTs	58–357	2500	Synthetic water	30-190	[162]
	MWNTs/Al_2O_3	237	3658	Synthetic water	37.2	Wei et al. 2013
Antiseptics						
Triclosan	SWNTs, MWNTs, MWNTs-O	1020, 283, 287	50–2000	Synthetic water	558, 435, 106 155-166	Cho et al. 2011; Zhou et al. 2013
	MWNTs	281	1000–12,000	Synthetic water	58.7, 18.7, 21.4 22.6–44.8	Bohdziewicz and Kamińska 2013; Joseph et al. 2011a
Bisphenols						
Bisphenol A	SWNTs, MWNTs, MWNTs-COOH	467, 456, 338	1000	Synthetic WW	58.7, 18.7, 21.4	Bohdziewicz and Kamińska 2013
	SWNTs	407	2280	Synthetic Landfill Leachate	22.6–44.8	Joseph et al. 2011a
	SWNTs, acid-treated SWNTs	407	228	Synthetic water	52.8, 41.4	Zaib et al. 2012
	SWNTs	407	228	Synthetic seawater, brackish water	13.4–16.1	Joseph et al. 2011b
	SWNTs	380	10–20,000	Synthetic water	359	Lerman et al. 2013
Bisphenol AP	MWNTs, MWNTs-OH MWNTs-COOH	> 500	20,000	Synthetic water	136-162	(Zhang et al., 2013a)
Hormones 17a-ethinyl estradiol	SWNTs	407, 233	2960	Synthetic Landfill Leachate	24.9–120	Joseph et al. 2011a
	SWNTs, acid-treated SWNTs	407	296	Synthetic water	115, 101	Zaib et al. 2012
	SWNTs	407	296	Synthetic seawater, brackish water	35.6–35.7	Joseph et al. 2011b
	MWNTs	84.3	5000	Synthetic water	0.472	Al-Khateeb et al. 2014
	SWNTs, MWNTs	541, 174	300–3,300	Synthetic water	276, 119	Pan et al. 2008b
Perchlorate	SWNTs, MWNTs, DWNTs	418, 176, 619	1000	Synthetic water	1.13, 0.29, 1.50	Fang and Chen 2012
	SWNTs	364	20,000	Synthetic water	10.0–13.6	Lou et al. 2014
Pesticides/herbicides						
Atrazine	SWNTs	407	100	Synthetic water	4.97[c]	Brooks et al. 2012
	MWNTs-O	167, 178, 185	500–20,000	Synthetic water including Cu, Pb or Cd	47.6, 36.1, 20.1	

Table 4 (continued)

EDC/PPCP class	Adsorbent	Surface area (m^2/g)	C_0 (μg/L)	Source water	Q_m (mg/g)	References
	(0.85, 2.16, 7.07%)					
	MWNTs-O (0.85, 2.16, 7.07%)	167, 178, 185	1000–8000	Synthetic water	60.3, 33.3, 24.0	Chen et al. 2009
	MWNTs	189	1000–30,000	Synthetic water	61–67	Rambabu et al. 2012
	Magnetic MWNTs	163	5000	Synthetic water	42	Tang et al. 2012
	SWNTs, MWNTs	167, 300	30,000	Synthetic water	33, 110	Yan et al. 2008
Diuron	MWNTs-O (1.52, 2.66, 7.58%)	159, 157, 161	600–22,000	Synthetic water	50.3, 48.0, 29.8	Chen et al. 2011
Dichlobenil	MWNTs-O (1.52, 2.66, 7.58%)	159, 157, 161	600–13,000	Synthetic water	39.4, 37.2, 23.5	Chen et al. 2011
Isoproturon	MWNTs	162	NA	Pure water W.W. effluents	16.3, 8.1	Sotelo et al. 2012
Stimulant						
Caffeine	MWNTs	162	NA	Pure water	41.6	Sotelo et al. 2012
Fipronil	g-C3N4/H-ZSM-5	172	600	Synthetic water	4	Aanchal et al. 2020
Ciprofloxacin	MWCT	9 cm^2	150000	CPX solutions	209.6	Yu et al. 2016
Sulfapyridine	MWCNTs	NA	50–1200	NA	2.0–10.0	Wei et al. 2016
Ofloxacin	BNNS	50.02	20000	NA	15–36	Singla et al. 2018
Tetracycline, Ofloxacin and Cephalexin	BNNS	1801.9	20000–50000	Aqueous solution	46.66 (TC); 72.50 (OFL); 225.0 (CFX)	Bangari and Sinha 2019
Cephalexin	SnO_2/Ni@NCNT	155 to 217 m	50000	NA	> 70%	Duan et al. 2020

C_0 EDC/PPCP initial concentration, *WW* wastewater, *NA* not available, q_m maximum sorption capacity

[a] Sorption capacity from Polanyi-Manes

[b] Distribution coefficient (L/kg) calculated from the Freundlich model

[c] K_p = capacity factor (μg/g)(m^3 /mg) for the Freundlich model

Based on the table above and explanations that have been done in the previous sections, the adsorption of two of the EDCs, namely Diuron and Dichlobenil, escalated with larger surface areas and pore volumes of MWNTs. It is elucidated by the differences in the van der Waals interaction and logs K_{ow} values. Diuron (log K_{OW} = 2.85) with more excellent value and greater surface area is removed more efficiently than dichlobenil (log KOW = 2.74) due to the relatively more significant dipolar moment (López-Ramón et al. 2007). Again, among PPCPs studied, sulfapyridine exhibited higher adsorption nonlinearity than sulfamethoxazole for the MWNTs. It indicates a more heterogeneous dispersal of the adsorption active points for sulfapyridine (Ji et al. 2009b). Again, by their chemical and anatomical orientations, they seem to have more affinity to MWNTs. Both sulfapyridine and sulfamethoxazole have one benzene ring and one aromatic heterocyclic group, which likely interact with the polarized aromatic rings on the surface of MWNTs through the mechanism of π-π electron coupling (Chen et al. 2007). Kurwadkar et al. have also mentioned that the tubular cylinders of carbon atoms in CNT can better absorb and remediate PPCPs and EDCs due to their large specific surface area and developed pore structure. In another study, the adsorption amount of sulfapyridine was shown to decrease with increasing oxidation degree of CNTs. In general, the presence of oxygen-containing functional groups inhibited the adsorption of it, and the inhibited effect followed the order of –COO- > –C=O > –OH > –COC– (Wei et al. 2016). Again, Wu et al. 2017 reported that the duplex-structured of Ti/SnO_2-Sb/SnO_2-Sb-CNT shows synergistic adsorption and electrochemical oxidation effect for bisphenol A degradation. On the other hand, Singla et al. (2018) synthesized morphologically different boron nitride nanomaterials: BNNPs (nano-particles) and BNNS (nanosheets) as adsorbents toward the adsorption of toxic and adverse water pollutants, i.e., antibiotics (Ofloxacin and Moxifloxacin). In the study, the pH range of 4–6 was found as an optimum pH for the adsorption of antibiotics by BN nanoparticles and followed the pseudo-second-order kinetic model. In 2020, Duan et al. showed that electro persulfate oxidation could be developed into an efficient technology for the elimination of recalcitrant pollutants. On the other hand, Zhong et al. (2020) proposed new insight on persulfate activation for tetracycline degradation. The study shows that in N-Cu/ biochar/ persulfate system, the main degradation reaction was the free radical pathway and $HO^{\cdot}$ Played a crucial role in tetracycline degradation. In another study, Bangari and Sinha (2019) reported that the large surface area of BNNSs has maximum adsorption capacities of 346.66 mg g^{-1} tetracycline, 72.50 mg g^{-1} ofloxacin, and 225.0 mg g^{-1} cephalexin. This superior adsorption behavior of BNNs demonstrates the potential for commercial applications for potable and wastewater treatment.

Membrane filtration

Nanocomposite membranes containing SWCNT and MWCNT were fabricated to remove specific pollutants such as triclosan, acetaminophen, and ibuprofen. It was observed that with an increasing number of aromatic rings, the organic pollutant removal efficiency increased from 10 to 95%. It also depends on the surface area (SWCNT>MWCNT) and oxygen content of membrane-based filters. The increase in pH from 4 to 10 augmented the PPCP removal ability by almost 70%. This observation proves to be a more uncomplicated technique for the removal of neutral molecules such as tetracycline in comparison to the one bonded by electrostatic interactions (ibuprofen) or hydrogen bonding (acetaminophen) (Wang et al. 2015b).

Photocatalysis: a highly promising technique

The idea of exploiting sunlight as a highly sustainable source of energy generation and its simultaneous application in environmental pollution treatment, photocatalytic splitting of water has been popularized in pollution remediation. Global energy crisis and environmental protection are two major concerned areas on which tedious research is going on. Photocatalysis has been proved to be an eco-friendly pollution control method that utilizes only two primary resources, i.e., sunlight and water, for its operation.

CBNs as photocatalysis

CNTs, being the most common class of carbon-based nanocomposites, have been modified into CNT/TiO_2 photocatalysts hybrid composites. TiO_2 nanoparticles deposited on CNTs (Dechakiatkrai et al. 2007), SWCNTs coated on TiO_2 nanoparticles (Yao et al. 2008), TiO_2-coated CNTs (Kedem et al. 2009), and CNTs grew within TiO_2-nanotubes arrays (Yang et al. 2008) are some of the basic variations that have been achieved for their applications in photocatalysis. Besides, many non-TiO_2-based photocatalysts have also been fabricated using metal oxides such as ZnO (Jiang and Gao 2005), WO_3 (Wang et al. 2009), and SnO_2 (Wang et al. 2011b). The loading amount of CNTs in these composites has a significant influence on photocatalytic activity. Electrochemical deposition of flower-like ZnO nanoparticles on CNTs shows good photocatalytic activity in visible regions (Kou et al. 2012). SnO_2 nanoparticles coated on nitrogen-doped CNTs (Wang et al. 2012), Ag_3PO_4 nanoparticles on MWCNTs (MWCNTs/Ag_3PO_4) (Wang et al. 2013), CNT/$BiVO_4$ (Zhou et al. 2011), V_2O_3/CNTs/TiO_2 using vanadyl acetylacetonate and titanium-butoxide as precursors (Chen and Oh 2010), cadmium sulfide/CNT nanocomposites with photo corrosion phenomenon (Huang and Gao 2004), CNT/ZnS heterostructures prepared by microwave irradiation (Wu

et al. 2008), and CNT/$Cd_{0.8}Zn_{0.2}S$ nanocomposites with extraordinary high photocatalytic activity (Liu et al. 2012) are some of the notable examples of carbon-based nanomaterials that have been applied for photocatalytic reactions in recent years. Very recent research has mentioned the MnO_2 with modified graphitic oxide, where manganese is in a mixed oxidation state. The role of the carbonaceous phase in the photodegradation of bisphenol A was also determined (Saroyan et al. 2019).

As already mentioned, carbon nanotubes (CNTs) have been reported to possess the unique quality of tubular cylinders consisting of carbon atoms. This property corresponds to their substantial and discrete surface area, with advanced pore structure, resulting in incompetency to adsorb and remediate PPCPs and EDCs efficiently. More extensive active surface area availability is beneficial for photocatalytic activity, like in adsorption mechanization, filtration, etc. Fortunately, among the carbon-based materials and composites, CNT has also been shown to possess the excellent photocatalytic activity and high mechanical strength. If it is co-ordinated with other removal techniques like filtration, CNTs show highly synergistic results. At the optimum situation, it can remove PPCPs and EDCs up to ~ 95% (Jung et al. 2015). A pulse discharged plasma (PDP) system coupled with activated carbon has been applied for the photodegradation of bisphenol A has been investigated (Guo et al. 2018). N-doped carbon dots have been studied by Hou et al. (2018) for the photodegradation of bisphenol F, and the effect of peroxydisulfate was reported. Peroxymonosulfate effects on the photodegradation of bisphenol A by sludge-based biochar has been performed with a mineralization efficiency of 80% (Huang et al. 2018). Table 4 shows the rate at which PPCPs and EDCs are degraded photo-catalytically using CNTs. Mechanistically, when exposed to appropriate light, it generates reactive oxygen species. These reactive species are well known for oxidizing contaminants into environmentally friendly products, namely CO_2 and H_2O. CoFe-based magnetic carbon xerogels for the photodegradation of bisphenol A was studied, taking into consideration the role of persulfate has been carried out, and the role of humic acid and chlorides has also been analyzed (Outsiou et al. 2017). Recently, a novel O-linked band and porous defect co-modified C_3N_4 was synthesized and applied in the photodegradation of bisphenol A and 2-mercaptobezothiazole was done (Jing et al. 2020). Novel $CeTi_2O_6$/g-C_3N_5 heterojunction photocatalyst with an outstanding photoactivity has used the photodegradation of 2,4-dichlorophenol which was synthesized very recently and the process involved photodegradation via superoxide and hydroxide radicals. This has also opened opportunities for the development of such low bandgap g-C3N4 type semiconductors for pollutant treatments (Vadivel et al. 2020). Again, CNT is hybridized with suitable semiconductor like titanium dioxide (TiO_2), and if CNTs is used as a substrate, an advanced level of utilizing visible light can also occur. This activity can be achieved by moderating the bandgap. Hence, not only the commonly used U.V. but also visible light can also be applied to the photocatalysis process.

Below is an example showing the degradation mechanism of a compound, ciprofloxacin (CIP), using molecularly imprinted carbon nanosheets supported TiO_2 (MICT). The carbon-based nanocatalyst showed selective identification and intense synergy of adsorption-photocatalysis process toward CIP removal. Li et al. 2020 had also identified its degradation intermediates and its pathways with the help of HPLC-MS technology. As shown in Fig. 2, two distinct pathways were proposed. The first part is a stepwise cleavage of the piperazine ring (CIP, A, C, E, and F path). The lone pair electrons in the N11 and N14 (CIP (m/z = 332) as seen in the figure can be easily oxidized by reactive radicals like hydroxyl through electron transfer, producing cation radicals. Then, through deprotonation action, the α-carbon of the cation radicals generates carbon-centered radicals, which interact with

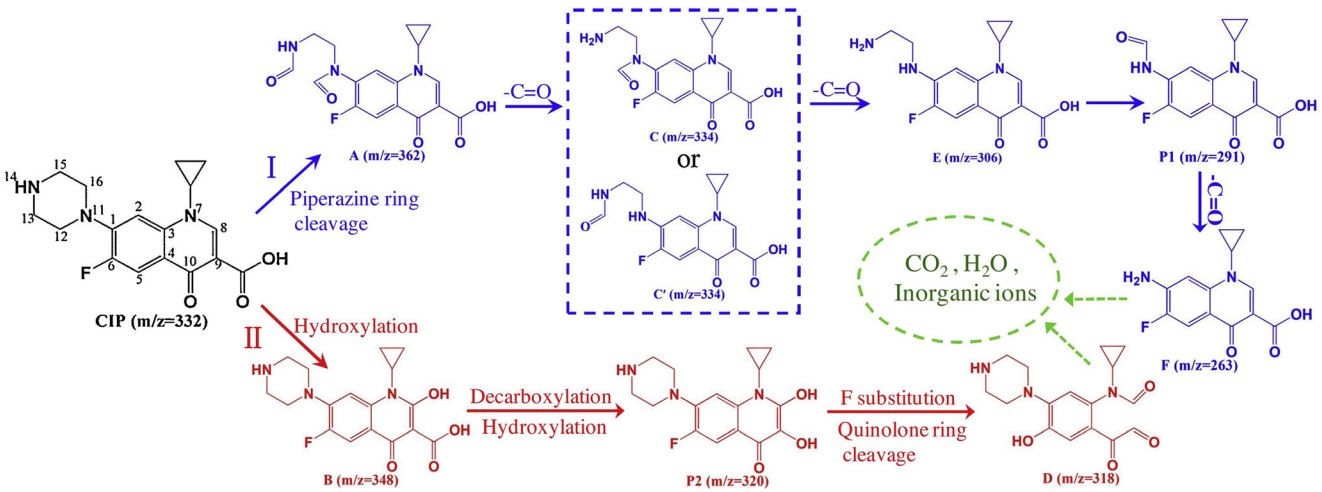


Fig. 2 The main degradation pathways of CIP over MICT

dissolved oxygen to give peroxyl radicals. This step of breaking piperazine ring leads to the creation of dialdehyde derivative A. Increased oxidation of compound A results in chronological depletion of formaldehyde groups. The process steers to the evolution of monoaldehyde isomers of C or C ′, and then E. The final product of this step, F, is formed through amine extraction, oxidation, and finally, loss of C=O. The second pathway (CIP, B, D) is the breakage of quinolone ring. Firstly, C8 of CIP will be attacked by hydroxyl radicals (produced through photocatalysis in the present context), giving the intermediate compound B. It will be accompanied by decarboxylation and hydroxylation reactions. The final product D of the second pathway is obtained by hydroxyl substitution of F. The sequential steps of the degradation process are shown in the figure. However, as explained for AOP, the final products are always expected to be CO_2, H_2O, and in some cases, inorganic ions (Li et al. 2020).

Basic principle and application of CBNs

In the photocatalytic treatment, organic pollutants are broken down with U.V./visible radiation, absorbed by a semiconductor material (i.e., photocatalyst). When the absorbed light is of energy higher than that of the semiconductor bandgap, it excites an electron from its valence band of the semiconductor to the conduction band, thereby generating an electron-hole (e^--h^+) pair. These holes are electron-deficient and tend to oxidize molecules that are adsorbed on the surface of the photocatalyst. These holes oxidize the organic molecules (R) or H_2O to produce R^+ and •OH radicals, respectively, as shown in Fig. 3. On the other hand, electrons reduce R or react with O_2 to make the superoxide radical anion, i.e., O_2•. Among all the species generated in this process, •OH radicals play a significant role in the oxidation of organic compounds to simpler units (CO_2, H_2O, or/and simple salts) in a process called demineralization. The basic features that must be exhibited by a semiconductor material for being a photocatalyst (as suggested by Da Silva and Dos Santos 2017) can be summarized as below:

- Active absorption and photoactivity in the UV-visible range.
- Conduction band with high redox potentials to efficiently stimulate the demineralization process.
- High oxygen reduction efficiency.
- Low toxicity, cost-effective and eco-friendly.

Semiconductor photocatalysis has been proved to be a promising technology for complete demineralization of the organic pollutants (Solomon et al. 2012; Carré et al. 2014; Kabra et al. 2004) and removal of toxic metal ions (Li et al. 2012; Singh et al. 2013). TiO_2 based semiconductor materials have been exploited for the last two decades in the splitting process of water molecules (Fujishima and Honda 1972; Singh et al. 2016). Numerous combinations of semiconductor materials have been explored to attain maximum efficacy of photocatalytic activity. Since then, many research works have been performed in the field of heterogeneous photocatalysis, devising their mechanism of action, effects of various parameters on their activity, and thereby applying them in wastewater treatment. The function of photocatalysts primarily depends upon the capability of semiconductors to produce charge carriers (electrons and holes) upon irradiation with light. Upon interaction with light, the generation of free radicals such as •OH takes place, responsible for initiating the degradation process and finally ending up in demineralization of organic pollutants (Trapalis et al. 2016; Singh et al. 2016; Herrmann 1999; Tan et al. 2013; Morales-torres and Pastrana-martínez 2012). Some other metal oxide-based nanomaterials

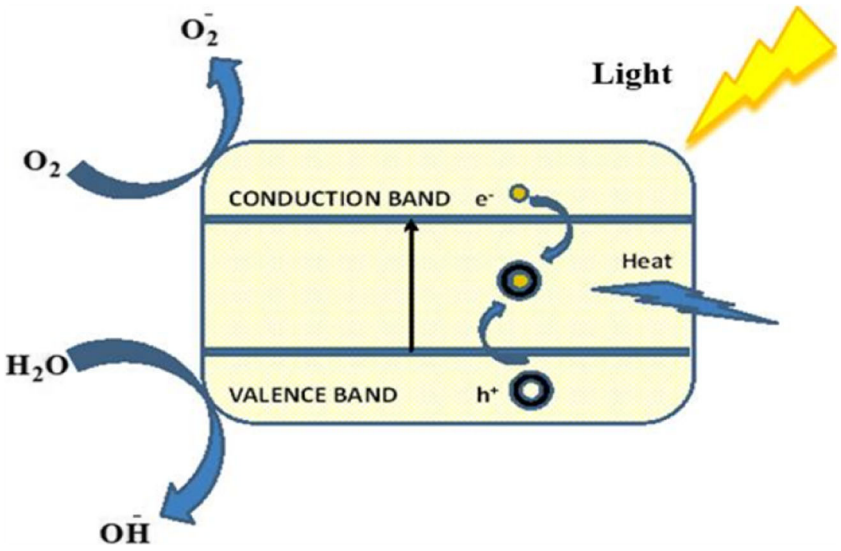


Fig. 3 Semiconductor photoactivation by light (Cincinelli et al. 2015)

utilized for the degradation of EDCs and PPCPs are MnO_2, Cu_2O, and CdS; however, their efficiency was much lesser than TiO_2-based materials (Hasanpour and Hatami 2020). It has been observed that the combination of single phases of two or more nanomaterials imparts a synergistic effect rather than additive effects on their properties. Those nanomaterials exhibit an additional advantage in terms of gradient potential at the interface than in the individual components, leading to a larger e^--h^+ pair separation. Hence, the adsorption properties can be easily enhanced by bringing the bandgap energy within the range of visible light, which eventually increases the selectivity of the material (Kim and Park 2011; Zhang et al. 2015; Sridhar et al. 2020; Tan et al. 2018). Major categories of the hybrid materials that have been applied in photocatalytic materials can be classified as below (Minella et al. 2017a).

- Homo or hetero hybrid materials are having two or more inorganic phases.
- Inorganic semiconductors coupled with carbon-based materials (graphene, G.O., reduced-GO or G.R., SWCNTs, MWCNTs, Quantum dots, etc.).
- Hybrid inorganic semiconductors such as g-C_3N_4 or boron nitride.
- Inorganic nano-hybrid semiconductors and polymers such as Polyaniline (PANI).
- Surface functionalized inorganic semiconductors (silica or graphene) with organic molecules that could be easily sensitized in the visible region and generate charge carriers, improve selectivity or lead to an increment in the adsorption toward a particular species group of molecules.

TiO_2/CBM hybrid materials successfully draw special attention because of their higher competence over other inorganic semiconductor materials. TiO_2/CBM hybrid materials have been tested diversely for photodegradation of pharmaceutical products and EDCs. However, it was observed that efficiency and performance depend largely on the type of carbon material used and the nature of the substrate being degraded. More than 20 pharmaceutical products have been satisfactorily photodegraded using TiO_2/SWCNT. Drugs such as ketoprofen, paracetamol, diclofenac, and carbamazepine were found to be unaffected in the process of photocatalysis using TiO_2/MWCNT hybrid. TiO_2/G.R. can photo-catalytically degrade antibiotics such as acetaminophen, risperidone, diphenhydramine, atenolol, diclofenac, sulfamethoxazole, carbamazepine, and ibuprofen. A conclusion can be deduced from the above observation that the introduction of graphene-based materials enhances the photocatalytic activity of TiO_2, as shown in Fig. 4 (Minella et al. 2017b). The combination of Al_2O_3 and SiO_2 and CNTs further enhances the photocatalytic efficiency of TiO_2 (Jitianu et al. 2004). Out of nine different varieties of MWCNTs/TiO_2/SiO_2 nanocomposites, most of them were found to be highly effective for successful degradation of carbamazepine (CBZ) at a faster rate when irradiated by UV-A (U.V. spectra near to the visible region) light when as compared to that by Degussa P25 photocatalyst. The nanocomposites were having a lesser percentage of showed less activity than the rest of the combinations. Photo-degradation of bisphenol A (BPA) by nanocomposites having 3.5 wt% MWCNTs was more effective than that with pure P25 photocatalyst (Czech and Buda 2015). Fabrication of pre-coated MoS_2 as a sacrificial layer on CNTs to immobilize Ag nanocrystals has developed a new composite material that displays excellent and higher catalytic performances under visible light irradiation (Liu et al. 2016). Sulfanilamide photodegradation occurring under irradiation with the help of carbon-sensitized and nitrogen-doped TiO_2 semiconductor material has also been reported (Wang et al. 2011a). Fipronil degradation using g-C3N4/H-ZSM-5 nanocomposites with an efficiency of around 89% has been achieved in recent times, which is much higher than the other metal-based nanocomposites (Aanchal et al. 2020). Sludge-based carbon (SBC) for the removal of Cu/Ni and its investigations for the estrogens removal has also been investigated (Ai et al. 2019).

A series of experiments have proved that CNTs-based photocatalysts such as SWCNT/TiO_2, MWCNTs/TiO_2, and MWCNTs/TiO_2/SiO_2 were very much useful for the photodegradation of POPs. The photoactivity of such functionalized CNTs, primarily hydroxylated MWCNTs (MWCNTs-OH), enhances due to the hydrophilic nature of CNTs (Ji et al. 2012). Graphene-based composites with γ-Fe_2O_3 nanoparticles were synthesized and studied for high-performance photocatalytic degradation of dibutyl phthalate, bisphenol A, 1-naphthol, and atrazine. Due to the presence of magnetic Fe_2O_3 components, recovery of the catalyst from the system after the degradation process becomes easy. The graphene-graphene interaction is partially inhibited by γ-Fe_2O_3 but promotes graphene dispersion, making more of the graphene surface available for reaction (Sinha and Jana 2013). ZnS/NSDC nanocomposites were designed with chitosan-thiourea resin and $ZnCl_2$, with an extensive specific surface area (642.24 m^2/g), the higher percentage of C/N/S co-doping, to reduce recombination of charge carriers at the time of photocatalytic degradation of BPA in visible light. The photodegradation efficiency was reported to be around 88.0%, and it follows a simple first-order kinetics model with the recyclability of catalyst to be about seven times (Al-Kahtani et al. 2019). BPA photodegradation over CNT-COCl/TiO_2 composites (Yuan et al. 2016) and $FeCo_2O_4$/TiO_2/G.O. (Bai et al. 2016) has been studied over a range of pH, and their effect on the photocatalytic efficiency has been determined extensively. In another report, bismuth ferrite/reduced graphene oxide ($BiFeO_3$/rGO) nanocomposites have been found to show 100% degradation of BPA invisible regions. This may be attributed to the combined effects of the reduction in bandgap and particle size that retard the recombination of e^--h^+ pairs and π-π interaction between BPA and

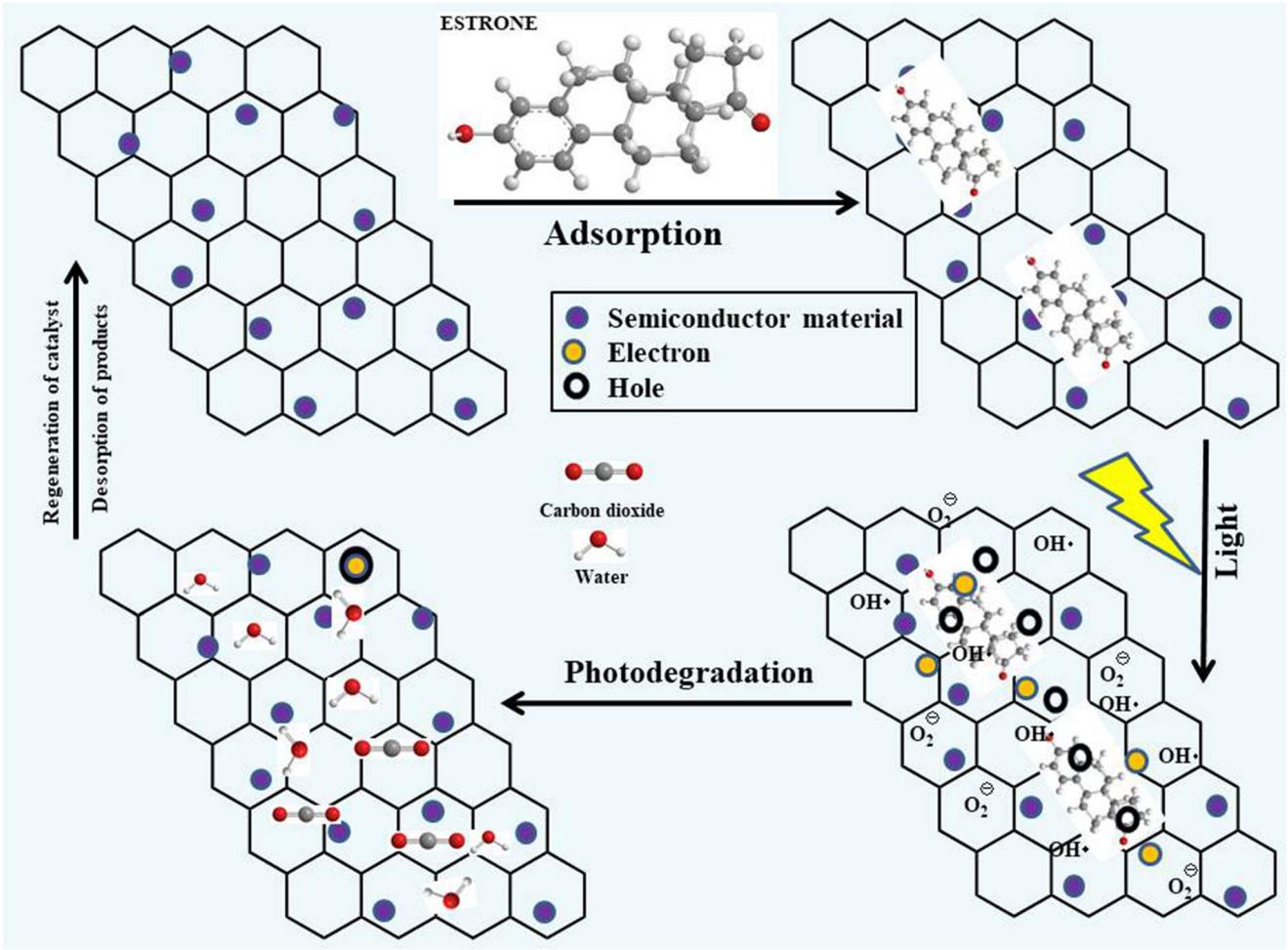


Fig. 4 Graphene-based nanocomposites: photodegradation (A schematic representation)

rGO due to the conjugation plane of graphene (Soltani and Lee 2016). Among several other catalysts for BPA photodegradation, G.O., or G.R.-based aluminosilicate mineral mullite with $Bi_2Fe_4O_9$ shows a much higher photocatalytic activity as compared to that of pure $Bi_2Fe_4O_9$ (Hu et al. 2015). 17a-ethinylestradiol (EE2) and other estrogen-based compounds were degraded over GO-TiO_2 photocatalysts (Mboula et al. 2015). Effective degradation by porous carbon-based composites of TiO_2; CdS on porous carbon; PANI/CNT immobilized on CNTs have been reported to show high activity for DMP (Cui et al. 2016; Wang et al. 2016; Hung et al. 2017). Studies have found that g-C_3N_4/$Bi_2O_2CO_3$ and g-C_3N_4/BiOCl to be highly effective for the photodegradation of DMP (Shan et al. 2016). Bi_2O_3/BiOCl immobilized on graphene sand composite, and BiOCl-doped over graphene sand composites were synthesized and analyzed for photocatalytic degradation of drug molecules such as ampicillin (semi-synthetic penicillin) and oxytetracycline (antibiotic) (Priya et al. 2016a, b). The novel cobalt-to-oxygen-doped g-C_3N_4 used in the photodegradation of bisphenol A also established a metal-ligand charge transfer process for the photoactivity assisted by nitrogen vacancies in the graphitic carbon nitride sheets (Yu et al. 2020a, b). Tunable V2O5/B-doped g-C3N4 has been newly added to the list of these carbon-based nanomaterials, which effectively worked for the photodegradation of diclofenac. The efficiency was reported to be 100% which was achieved in less than 105 min. The high photocatalytic efficiency was due to the charge separation assisted by Born, and V2O5 showed a synergistic enhancement in the photoactivity (Oliveros et al. 2021). The adsorption of these drugs on the catalyst surface followed second-order kinetics. Solar light was used as the source of radiation with the catalyst recyclability of around ten times. BiOCl holds extra advantages of being non-toxic and corrosion-resistant. Ruthenium (Ru) impregnated in MWCNTs using various Ruthenium precursors to prepare catalyst which oxidizes aniline (100% conversion) in wastewater was studied by Garcia et al. (2006).

Factors affecting photodegradation

Photocatalytic reactions mostly depend on temperature and atmospheric pressure, which holds an additional advantage in

water treatment methods (Lee et al. 2016). The optimum reaction temperature for photocatalysis lies between 20 and 80 °C (Herrmann 1999). Diffusion/recombination rates and adsorption of pollutants on the photocatalyst surface are highly influenced by temperature (Sarkar et al. 2014). Temperature higher than 80 °C reduces the adsorption on the surface (Chong et al. 2010). It enhances the recombination of charge carriers (Gaya and Abdullah 2008), reducing the photocatalytic efficiency of the CNB photocatalysts. Low temperature affects the energy required for the reaction as it needs higher activation energy for the reaction to start; this is responsible for lower catalytic efficiency (Herrmann 1999). Dissolved oxygen (D.O.) is another critical parameter that alters the phenomenon of photocatalysis. It accepts electrons during the photocatalytic reaction to generate free radicals (Hoffmann et al. 1995), and therefore, a higher amount of dissolved oxygen promotes the rate of photocatalytic reactions. Investigations were made to study the influence of nitrate ions (NO_3^-), Ferric ions (Fe^{3+}), and dissolved organic materials (DOMs) (humic acid fulvic acid) on photocatalysis of atenolol by MWCNTs-OH. Although atenolol was resistant to direct photolysis, it was easily degraded in the presence of these moieties (Ji et al. 2012). It was later proposed that the presence of DOMs, NO_3^-, and Fe^{3+} modified the photochemical properties of organic contaminants, e.g., PPCPs/EDCs in aqueous systems through ionization, oxidation, or energy transfer and thereby creating photo-generated reactive oxygen species (ROS) (Zhang et al. 2013a). Inorganic species in wastewater affect the photocatalytic remediation of PPCP and EDCs to a greater extent. Species such as peroxides, persulfates, bromates, and sulfites favor the photocatalytic reaction attributed to their electron scavenger nature and therefore increase the generation of hydroxyl radicals some other oxidizing species (Lee et al. 2016). But an excess of inorganic species (chlorides, ferric ions, calcium aluminium, sulfates, carbonates, etc.) opposes the photodegradation process by competing for hydroxyl radicals and cause a variation in the pH of the solution as well as modifies the active site for adsorption (Rioja et al. 2016). In recent times, graphene-based heterojunctions have attracted many researchers' interests due to the high conductivity offered by graphene sheets with consequent enhancement in photocatalytic activity of the heterogeneous catalysts. The highly conjugated π-π electrons over the graphene surface promote the adsorption of different types of pollutants during the photocatalytic reaction. But it was observed that if the composition of graphene was more than one wt %, it creates a light-shielding effect and reduces the semiconductor photoactivity (Low et al. 2017).

Limitations, feasibility, and possible solutions

Even though carbon-based nanomaterials have emerged as an excellent photocatalytic material for the degradation of POPs, there are few limitations, as discussed below:

- Gibbs free energy for the splitting of water is beneficial, i.e., thermodynamic unfavorable. Therefore, cost-effective and eco-friendly catalyst is higher in demand for preventing radicals-recombination and regenerating of water molecules.
- Graphene-based nanocomposites are challenging to work with as the surface is easily functionalized and modified. It becomes quite a challenge to control the surface structure for effective performance (Kumar et al. 2017).
- Most of the carbon-based nanocomposites are doped with metal and their oxides. However, new methods are being developed for the metal-free carbonaceous material for the green photodegradation of pollutants. One such recent research has focused on N-doped hydrochar for the photodegradation of endocrine disruptors. In this composite, graphitic nitrogen was found to allow radical as well as a non-radical pathway for the degradation of the organic compound (Yu et al. 2020a).
- The photocatalytic degradation of atenolol by MWCNTs-OH was unfavorable and negatively affected the environment (Zhang et al. 2011a). Therefore, it becomes crucial to trace the toxicity, potential human health hazards, and ecological risks that may arise because of the use of CNTs. The toxicity profile varies with CBN used as well as the substrate undergoing photodegradation. For example, photodegradation of atenolol by MWCNTs-OH was much more potent but environmentally non-viable than that of BPA and CBZ (Czech and Buda 2015).
- Cost, toxic effect on the aquatic flora and fauna, and bench-scale to pilot-scale feasibility have to be examined before bringing CNTs into practice in water purification and wastewater treatment. The use of low-cost materials in devising CBNs for their application in photocatalytic degradation of organic wastes is a vital aspect to be considered (Song et al. 2018). While SWCNT is the most expensive among CBNs, biochars are still much cheaper but less efficient (Ahmed et al. 2015). Studies on nano-sized TiO_2/SWCNT showed that photocatalysts can be recovered quickly by physical processes such as centrifugation or filtration and can be reused for several cycles (Ahmmad et al. 2014). The versatile and flexible nature of CBNs can be explained in terms of their easy recovery, regeneration, and reusability in photocatalytic and other treatment processes of wastewater.
- The present scenario demands a treatment method that can quickly, cost-effectively eliminate targeted contaminants. Therefore, it becomes much necessary to create such systems that are made by integrating two or more properties or hybrid materials (Ahmed et al. 2015).

Conclusion

A complete system integrating all relevant researches in monitoring, detection, and control for multi-barrier approach management is projected for EDCs. It has a preventive measure for the risks associated with EDCs in the environment (e.g., water, wastewater, soil, and another natural ecosystem). EDCs cause various adverse effects on human health, and the risk inferences are analyzed using the Needs, Approaches, Benefits, and Challenges (NABC) method. A compassionate tool is essential for decision-making for management purposes. A better understanding of general principles regarding the mode of action in different risk assessments has become the need of the hour based on rapid, selective, and sensitive detection of EDCs. For such purpose, the use of nanostructured materials is a fascinating choice owing to their numerous advantages (e.g., fast response, automation, high selectivity/sensitivity, and decent accuracy) for precise monitoring of EDCs. Since the effects and reactions of EDCs are potentially higher even in trace concentrations, the rapid detection and monitoring of EDCs by developing various multi-tiered analytical approaches (especially sensor-based) would be of more significant concern in the coming years. Integrated with advanced monitoring techniques of treatment, the necessary decision making can be accelerated to facilitate their removal from the environment. The present review will also be organized to describe the existing removal methodologies and the discussion on the further scope of improvement in terms of their efficiency and deployment. The mesoporosity of CNTs is much higher than activated carbon (A.C.). Therefore, they show more adsorbing capacity for organic compounds. EDCs and pharmaceutical drugs have been analyzed and reported to be easily confiscated by applying these CNTs. The high cost of production of CNTs poses a challenge for their large-scale commercial application as photocatalyst in wastewater treatment units. Most of the experiments have been done at a bench-scale level that restricts their role in large systems. The combination of CNTs with systems to attain more desirable properties is still needed to be addressed for their practical application in pollution control.

Future prospects

The literature available on the toxicity of carbon-based nanomaterials to living beings is still inadequate and inconsistent. Such loopholes invite more research and hence, the addition of more detailed insight. There is an additional need for deeper exploration in the material properties and their applications, like conjunction with other materials to enhance the efficacy without compromising the negative aspects of toxicity (Wang et al. 2015a). For example, carbon-based contemporary compounds like fullerenes are yet to traverse intensely for their utility in water treatment applications. Again, a robust risk assessment protocol and calibrated/standardized toxicity analysis is a critical need of the hour to determine the potential health hazard and environmental damage caused by these materials (Song et al. 2018). As for the removal of EDCs and pharmaceuticals using the mentioned materials, though the adsorption mechanism has been successfully effectuated, however, most of the carbon-based materials are still commercial. Besides the discussed compounds being categorized among the emerging contaminants, it is recommended to consider superior augmentable options like surface modifications. Designing and developing authentic physical and chemical features with minimum health and ecological impacts, giving assurance to sustainability, is being suggested. Overall, the cost is one of the most significant drawbacks while using advanced carbon-based materials. Hence, rigorous research in *modus viventi* with quality and the viable cost is highly endorsed.

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