

Review

Nanocarbons as Potential Tool for the Removal of Pharmaceutical Compounds from Water Resources: An Updated Review

Deepak Sharma ¹, Pritha Chakraborty ², Jithin Thomas ³, Mridul Umesh ⁴, Basheer Thazeem ⁵, Subhrangsu Sundar Maitra ⁶, Kanchan Kumari ⁷ and Vinay Kumar ^{8,*}

¹ Department of General Surgery, Saveetha Medical College and Hospital, Saveetha Institute of Medical and Technical Sciences (SIMATS), Chennai, 602105, India

² School of Allied Healthcare and Sciences, JAIN (Deemed to be University), Bangalore 560066, India

³ Department of Biotechnology, Mar Athanasius College, Kothamangalam 686666, India

⁴ Department of Life Sciences, Christ University, Bangalore 560029, India

⁵ Postgraduate and Research Department of Zoology, Maharaja's College, Ernakulam 682011, India

⁶ Metagenomics and Bioprocess design laboratory, School of Biotechnology, Jawaharlal Nehru University, New Delhi 110067, India

⁷ Department of Physics & Materials Science, Jaypee University of Information Technology, Waknaghat 173234, India

⁸ Biomaterials & Tissue Engineering Laboratory (BITE LAB), Department of Community Medicine, Saveetha Medical College and Hospital, Saveetha Institute of Medical and Technical Sciences (SIMATS), Chennai 602105, India

* Correspondence: vinayiplrd@gmail.com or vinaykumar.smc@saveetha.com

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Abstract: Nanomaterials have been an area of great research for the pollution control in recent decades due to their beneficial properties such as higher efficiency, low cost, easier fabrication, and higher surface area. Nanocarbons have been considered as a promising nanomaterial in remediation of emerging pollutants. Pharmaceutical compounds are one of the major products of healthcare which are present in different environments including water resources. These compounds include several categories of chemicals and medicines such as antibiotics, analgesics, antidepressants, hormones, and anticonvulsants. These pharmaceutical compounds enter the environment through sources such as pharmaceutical effluents, hospital waste, livestock farming, landfill leachate, and aquaculture industry. Considering the environmental and human health concerns associated with these pharmaceutical contaminants, the presented review is focused on understanding the mechanisms of pharmaceuticals removal through carbon-based nanomaterials by the processes such as adsorption and catalysis. In addition, it explores the factors which affect the removal efficiency of the pharmaceuticals by nanocarbons. Moreover, recent advances and emerging technologies such as hybrid materials and composites, smart and responsive nanocarbon systems, membrane technologies and artificial intelligence for predictive performance have been discussed. The review article also provides the information on critical challenges and future perspectives in the research area.

Keywords: nanocarbon; pharmaceutical waste; toxicity; pollution; health problems

1. Introduction

Water is valued as the most essential natural resources for all living forms. In modern day world, clean and safe to drink water is not available to a considerable number of populations due to limited ground water resources. Safe and potable drinking water is important for human health. In this scenario, the available ground water



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resources are being polluted by constant disposal of effluents containing persistent micro and macro-pollutants [1]. According to the data from World Health organization (WHO), clean drinking water is not available to approximately 785 million of people and 145 million people only survive with the help of surface water bodies. Nearly 8 million people die annually due to water borne diseases. It is also predicted that by 2025, half of the world population will live in water stressed area [2]. There are various sources of water pollution like agricultural, pharmaceutical, domestic and industrial wastes. The slow but steady increase of anthropogenic wastes and their release in environment leads to harmful impact overall [3]. Pharmaceutical wastes include endocrine disruptors, personal care products and undigested parts of drugs in low to moderate concentration. Pharmaceutical wastes get dumped or washed away and enter municipal trash and sewage system. The common antibiotics and their metabolites are found in high concentration in the sewages, all water bodies, soil and sediments [4]. Several studies report different analytical techniques identifying the presence of pharmaceutical products at higher concentration ranging from ng/L to mg/L. Wastewater treatment plant and sewage treatment plant are not well equipped to filter out these persistent pharmaceutical contaminants and are considered as hotspot for pharmaceutical compounds pollution [5]. A study conducted by U.S. Geological Survey in 139 streams around 30 states across the country showed 80% of the streams have high concentration of antibiotics and non-prescription drugs. More than 100 different pharmaceutically active compounds are found in tap water, surface water and ground water in USA and European countries at higher concentration. Approximately 30 different pharmaceutically active compounds are found different water resources across South Asian countries [6].

With variable structure and target specificity, these pharmaceutically active compounds exert toxic effects on plants, terrestrial and aquatic species, livestock and humans. Accumulation of pharmaceuticals from soil to root vegetables, leaves, fruits and grains are reported by several studies [7]. Additionally, pharmaceuticals can be categorized on the basis of the structure, mode of action and target groups. Analgesics and anti-inflammatory drugs are found high concentrations. Paracetamols, alcohol, non-steroidal-anti-inflammatory drugs (NSAIDs), cyclooxygenase inhibitors, morphine, ibuprofen, diclofenac are some of the widely used analgesics and anti-inflammatory drugs [8]. NSAIDs are reportedly found at a very low concentration (nano to micro gram) in soil, sediment, ground water and surface water as well as drinking water and arctic ice. NSAIDs can exert toxic effects on environment despite of their negligible concentration [9]. Ingestion of these drugs in negligible quality for a longer period though untreated water can cause organ damage, metabolic and mitochondrial dysfunction, reduced membrane stability in vertebrates and invertebrates by inducing oxidative stress. In freshwater fishes, diclofenac and ketoprofen can cause cardiac damage [10]. According to [11], NSAIDs can cause DNA damage alongside endocrine disruption. Antibiotics are widely used pharmaceutical compounds to safeguard human health preventing contagious diseases. Spreading of antibiotic resistance in microbes is encouraged by wide usage of antibiotics [12]. Different health risks are associated with antibiotics like immune function and hepatic dysfunction, bone marrow defect and cardiac arrhythmia. Antidepressants are neuroactive pharmaceuticals which can be persistent in environment with 60 to 70% bioavailability [13]. These antidepressants along with their metabolites are found in sewage, effluent of wastewater treatment plants and surface water. The toxic side effects of these compounds include fertility dysfunction, growth inhibition and hypoglycaemia [14].

There are several conventional techniques for the removal of pharmaceutical pollutants from wastewater such as ozonation, photocatalysis, and activated carbon adsorption. These techniques are reported to have limited efficiency suggesting partial removal of pollutants [15]. Some pharmaceutically active compounds like bezafibrate and clofibric acid are only removed up to 34–52% and 50–90% respectively [16]. Activated carbon works as an adsorbent for removing these pollutants in aqueous phase by van der Waals interactions or ionic interactions mostly as post treatment for drinking water treatment. High energy demand of this method makes it unsuitable for upscaling [17]. Reverse osmosis, nanofiltration are various membrane filtration techniques for pharmaceutical pollutants removal. Efficiency of this process depends on membrane properties and physico-chemical parameters of pollutants. Effective usage of this process is restricted due to membrane fouling, affecting its application in wastewater treatment [18]. Ozonation is a secondary treatment method which requires high energy and yields unusual end products with partial removal of pollutants. The adsorption and diffusion of pharmaceutical pollutants also have limitations. UV radiation or photocatalysis also require high energy and it is time consuming. In recent years, novel approaches were proposed to replace the conventional methods improving the removal percentage and lowering the costs [19].

Nanotechnology offers several innovative opportunities to achieve complete removal of pharmaceutical compounds. Nanomaterials with unique characteristics like size, surface area, photosensitivity, electrochemical properties, quantum effect and catalytic activity promises great potential for pharmaceutical pollutants removal [20]. Nanocarbons are carbon-based nanomaterials having dimensions at the nanoscale (below 100 nm in at least one dimension). The nanomaterials exhibit unique physicochemical properties arising from their size, structure, and

surface chemistry. Current review article discusses carbon nanomaterials such as fullerenes, carbon dots, carbon nanotubes, carbon nanofibers, graphene, graphene oxide, reduced graphene oxide and porous nanocarbons. In recent years, carbon-based nanostructures with versatile features have found applications in pollutants removal strategies. Features like large surface area, thermal conductivity, electron mobility and mechanical strength makes nanocarbons essential in wastewater treatment. There are five possible interactions between nanocarbons and pharmaceutical pollutants like covalent and electrostatic interactions, hydrophobic interactions, hydrogen bonding and stacking. These interactions help in the adsorptive removal of pharmaceutical compounds [21].

In this review, recent developments in removal of pharmaceutical pollutants using nanocarbons and nanotechnology are summarized. This review focuses on different types of pharmaceutical pollutants in water. It offers an overview of carbon nanomaterials and discusses the possible mechanisms involved in removal strategies. The recent advances along with challenges and future direction of research are discussed to complete the knowledge gaps.

2. Pharmaceutical Contaminants in Water

Pharmaceutical contaminants in water belong to the class of chemicals of emerging concern and necessitates rapid attention. They are usually produced from sources like domestic, industrial and agricultural applications and their unregulated or inadequately regulated use pose a threat to humans and the environment.

2.1. Sources of Pharmaceutical Contamination

Pharmaceutical contamination can be broadly classified into two categories based on their sources. This includes point sources of pharmaceutical pollution in which the contaminants may originate from a single source and whose attribute can be precisely identified through mathematical modelling. The second type includes a diffuse source of pharmaceutical pollution which includes a discrete geographical location and is often difficult to trace and identify accurately [22]. Major point sources of pharmaceutical contamination include pharma manufacturing industries, hospitals/medical facilities, sewage waste water and water comes from the domestic septic tanks. Landfills are also regarded as principal sources of contamination by pharmaceutical compounds as the leachate coming from these sources often has pharmaceutical compounds that contaminates the groundwater and surface water [23]. Non-point sources of pharmaceutical contaminants are otherwise regarded as diffuse sources of pollution and are often difficult to trace. Entry of the pharmaceutical contaminants from sewage sludge to soil and then eventually into water constitutes one of the major non-point sources of pharmaceuticals pollution [24]. Agricultural runoff and veterinary pharmaceutical residues mixing with water or presence of these contaminants in faecal matter that mixes with water also constitutes a major non-point source of pharmaceutical pollution. The groundwater often gets contaminated with the pharmaceutical compounds from the surface water through the groundwater—surface interface as a result of lateral and vertical hydraulic exchange [25]. The potential routes through which pharmaceutical contaminants can infiltrate water sources is represented in Figure 1.

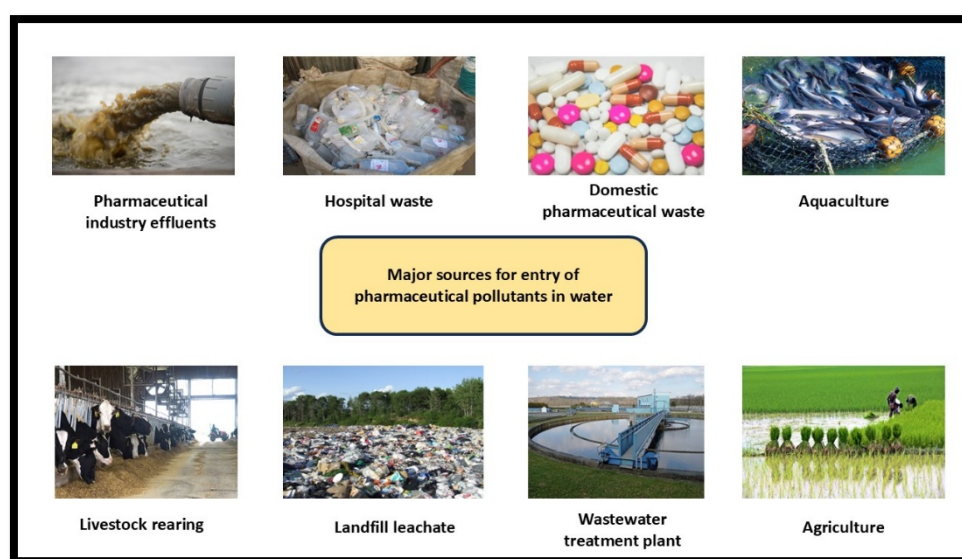


Figure 1. Routes of pharmaceutical contaminants entry into the water resources.

2.2. Classification of Pharmaceutical Pollutants

The major class of pharmaceutical contaminants in water include antibiotics, analgesics, antidepressants, psycho stimulants, hormonal compounds and antiepileptic drugs.

2.2.1. Antibiotics

Antibiotics are substances produced from an organism that can kill or inhibit the growth of a pathogen and thus have had a pivotal role in treatment of infectious diseases since their discovery. A significant proportion of the antibiotics (70–90%) is excreted from the animals or human body and eventually enters the water bodies. The major sources of entry of antibiotics includes from hospital wastes, livestock and aquaculture farms [26]. The limitations of existing procedures in the wastewater treatment plants (WWTPs) to effectively remove pharmaceutical residues has also resulted in the enhanced persistence of these antibiotics in the environment. The presence of antibiotics in the marine ecosystem is linked to the evidence of the spread of antibiotic resistant genes that leads to serious environmental threats as well [27]. Antibiotics being incompletely metabolised by living organisms and having high degree of mobility offers a serious challenge for bioremediation procedures. The concentration of antibiotics detected in the water shows a considerable variation between 0.0005–0.0214 µg/mL in drinking water, 0.0013 to 0.0125 µg/mL in wastewater, and 0.0003–0.0039 µg/mL in river water with the predominant antibiotic types being tetracycline, sulfonamides, quinolones, nitroimidazoles, and fluoroquinolones [23].

2.2.2. Analgesics

Analgesics represents a notable amount of persistent pharmaceutical contaminants in wastewater. They are produced in large quantities globally (about 100 tonnes annually) and used widely for the treatment of inflammations, pain, fever, arthritis and osteoarthritis. Analgesics are often used in combination with NSAIDs, nonnarcotic medications, and opioids for various treatment procedures [28]. The major class of analgesic and NSAIDs include ibuprofen, ketoprofen, acetaminophen, and diclofenac. Their complex structure and high degree of stability make them resistant to degradation and removal in treatment plants. The presence of analgesics in water is often very low making them difficult to trace and remove during treatment procedures [29].

2.2.3. Antidepressants

Antidepressants are a class of pharmaceutical compounds employed for treating anxiety, depressions, chronic pain and addiction associated issues. They are used to regulate and correct the imbalance associated with neurotransmitters and are recorded to possess a high degree of bioavailability more than 60% [30]. The clinically approved classification of antidepressants includes selective serotonin reuptake inhibitors (SSRIs), serotonin modulator and stimulators (SMSs), serotonin-norepinephrine reuptake inhibitors (SNRIs), serotonin antagonists and reuptake inhibitors (SARIs), tetracyclic antidepressants (TeCAs), monoamine oxidase inhibitors (MAOIs), norepinephrine reuptake inhibitors (NRIs), norepinephrine-dopamine reuptake inhibitors (NDRI) and tricyclic antidepressants (TCAs). Presence and persistence of these antidepressants in water is linked to the evidence of acute and chronic toxicity to aquatic organisms [31].

2.2.4. Hormonal Compounds

Hormonal compounds are a class of endocrine disrupting compounds (EDCs) as they have the potential to mimic the action of estrogen on the target cells. Minimizing the exposure to these EDCs through water has attained significant research attention as it is linked to the onset of several types of cancers, neurological issues and reproductive disorders in humans. Among the most common hormonal compounds persistent in water, estrogens, glucocorticoids, mineralocorticoids, androgens and progesterones were found to be the major contributors [32].

2.2.5. Anticonvulsants

Anticonvulsants also regarded as antiepileptic drugs (AEDs) are chemical compounds used for treatment/prevention of depression, mania and migraines. These are routinely used as mood stabilizers for patients suffering from bipolar disorder and alcohol withdrawal. The major class of persistent anticonvulsants are carbamazepine, lamotrigine and gabapentin [33]. AEDs are known to cause selective modification of excitability of neurons even in non-target organisms when these compounds enter their system through water. These compounds were found to cause biochemical and metabolic changes in aquatic animals including fishes and were reported to have high levels of adverse effects once they enter the food chain [34].

2.2.6. Other Types

Other types of pharmaceutical contaminants include anticancer drugs and antiviral medications. As there is a significant increase in the use of antiviral drugs post Covid 19 pandemic their persistence in the environment has also increased exponentially and need more elaborate studies to understand its acute and chronic effects on humans and animals [35]. The increase in the number of cancer cases globally accelerated the use of anticancer drugs used in chemotherapy. Most of these anticancer drugs like cyclophosphamide, tamoxifen, and methotrexate (MTX) are not completely metabolised in humans and are reported to be persistent in aquatic environments as it is difficult to remove these compounds using the existing water treatment methods [36].

2.3. Physicochemical Properties Affecting Environmental Persistence

Environmental persistence of pharmaceutical components in water is greatly influenced by factors like pH, temperature, dissolved oxygen (DO), salinity, solubility, hydrophobicity, molecular weight, turbidity and presence of organic matter. These factors along with other physical factors like intensity of sunlight, depth of the water body and the microbial composition in water bodies determine the nature and persistence of pharmaceutical contaminants in water [37]. The effect of various physicochemical parameters on persistence of pharmaceutical contaminants is summarized in Table 1.

Table 1. Physical factors and their role in determining the persistence of pharmaceutical pollutants in water.

Physical Factors	Significance	Reference
pH	Changes in pH of water can alter the solubility and distribution of pharmaceuticals. It also has a significant impact on their degradation in aquatic environment	[38]
Temperature	Photolysis and hydrolysis of pharmaceuticals due to the influence of temperature and the microbial enzymatic activity is affected by increase in temperature	[39]
DO	High level of DO accelerates the oxidative degradation of pharmaceuticals that can be degraded through oxidation reduction reactions	[40]
Molecular weight	Low molecular weight pharmaceutical compounds undergo fast biodegradation where as high molecular weight compounds are recalcitrant to biodegradation and persist in aquatic environment	[41]
Salinity	Poorly soluble drugs undergo precipitation in hyper saline conditions and thus have an effect on their bioavailability	[42]
Solubility	Highly soluble pharmaceutical compounds to have high mobility in aquatic environment	[43]
Turbidity	High level of turbidity in water reduces light penetration and effects the enzymatic degradation of pharmaceuticals by microorganisms	[44]
Presence of organic matter (OM)	Presence of organic matter in the water can have an impact on the biodegradation and thereby persistence of pharmaceutical compounds as OM is supposed to have variety of interaction with the pharmaceutical compounds	[45]

2.4. Conventional Removal Methods and Their Limitations

Conventional methods for removal of pharmaceutical contaminants from water majorly involve techniques like coagulation, flocculation, sedimentation, filtration and disinfection. There are several demerits associated with these methods that question their efficiency in removing pharmaceutical pollutants from water [46]. Coagulation and flocculation employ use of large quantities of chemicals like ferric chloride or ammonium sulfate thereby making it non ecofriendly. Nevertheless, this method has limited application when the pharmaceutical compounds are present in trace amounts or in dissolved state [47]. Sedimentation and filtration also face the drawbacks due to the heterogenous nature of pharmaceutical contaminants in water because of their type, size and abundance, making it difficult to optimize the removal process [48]. Disinfection of water through ozonization and chlorination that works on the basics of oxidative breakdown of pharmaceutical contaminants may not be effective against the pharmaceuticals that are resistant to oxidation [49]. Generation of toxic byproducts due to oxidative breakdown of pharmaceutical contaminants can have serious deleterious effects on the environment. More advanced techniques like advanced oxidation process, membrane filtration and photocatalysis although successful in removing other major contaminants from water have serious limitations in removing pharmaceutical components due to their energy intensive nature, problems associated with membrane fouling and variation due to the effect of other environmental factors [50].

3. Nanocarbons

Nanocarbons exists in various forms such as fullerenes, carbon nanotubes (CNTs), carbon nanofibers, nanodiamonds, graphene, graphite, carbon nanofibers and carbon hybrids [51,52]. Each type carbon has distinctive features in nanoscale. Based on the dimensional these nanocarbon materials can be in zero-dimensional, one-dimensional, two-dimensional, and three-dimensional structure (Table 2) [53].

Table 2. Summarised information of carbon nanomaterials based on their dimensions along with examples.

Carbon Nanomaterial Dimension	Carbon Nanomaterial Example	References
Zero-dimensional	Fullerenes, carbon dots, and nanodiamonds.	[54]
One-dimensional	CNTs (single-walled carbon nanotubes, double-walled carbon nanotubes and multi-walled carbon nanotubes) and carbon nanofibers.	[54,55]
Two-dimensional	Graphene, graphene oxide, reduced graphene oxide and graphdiyne.	[56,57]
Three-dimensional structure	Carbon aerogels and foams of CNTs, graphene, and carbon nanofibers.	[58–60]

3.1. CNTs

CNTs comes under two-dimensional nanomaterial category having hexagonal carbon atoms lattice and also called as bucky tube. The objects in CNTs are bend in a particular direction form a hollow cylindrical configuration structure. CNTs were first time discovered in year 1991 [61]. CNTs generally ranges from 1 to 100 μm in lengths and try to agglomerate or form bundles because of higher surface area. The bundles of CNTs are gripped by van der Waals forces of interactions [62]. CNTs formed by sp^2 bonds, similar to graphite. They are generally categories into two categories single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). The synthesis of SWCNTs having diameter of 1nm was first time reported in 1993 [63]. SWCNTs can be synthesized by vertical floating technique [61]. These SWCNTs are longer in wavelength as compared to typical CNT. SWCNT have different physical and chemical properties in its two separate regions i.e., the sidewall (first) and the end cap (second) of the tube. Furthermore, the properties of SWCNTs are also different from MWCNTs. MWCNTs structurally are concentric SWCNTs. In addition to morphology and structure MWCNTs also have unique texture [62]. Figure 2 represents the structure of SWCNTs and MWCTs and CNTs structure based on chirality.

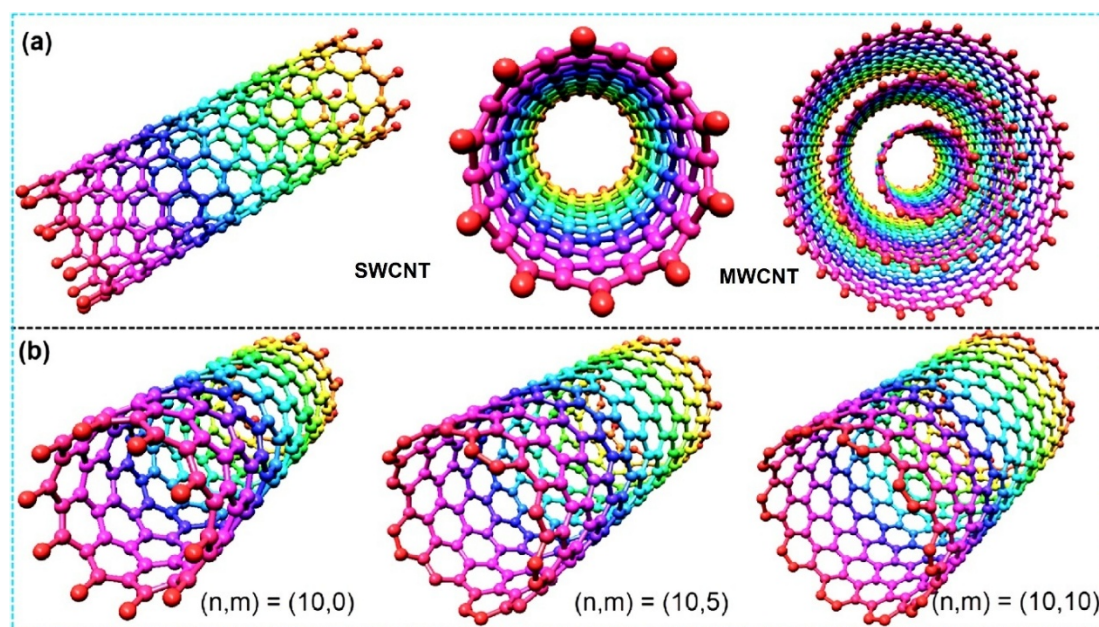


Figure 2. Pictorial representation of CNTs (a) SWCNTs and MWCNTs and (b) CNT structure based on charity. Figure is adapted from [64] and is permitted to use under a Creative Commons license.

3.2. Graphene and Graphene Oxide

Graphene is recognised as worlds thinnest material. Graphene is a two-dimensional single atomic layer structure of carbon atoms having sp^2 bonding. These carbon atoms are organised in a hexagonal lattice which is very similar

to ubiquitous graphite [65–67]. The six carbon atoms of graphene have a molecular bond of ~ 0.142 nm [68]. Graphene is also used for the production of graphitic materials such as zero-dimensional fullerenes, one-dimensional CNTs, and three-dimensional graphite's [69]. Furthermore, graphene can be transformed into quantum dots, nanoribbons, nano mesh and sheets, and graphene oxide (GO). Graphene exhibits good thermal and electrical conductivity at room temperature along with higher surface area which is well suited for catalytic activity [70]. Graphene can be produced by top-down and bottom-up synthesis approaches. Top-down approach involves transformation of graphite into graphene while bottom approach utilizes carbon as precursor for the synthesis [71,72]. Graphite can be converted GO by oxidation to form a single-atomic layer of carbon, hydrogen, and oxygen (Figure 3). However, these GO can be converted into reduced GO (rGO) by removal of oxygen (Figure 3) [73,74]. It is interesting to note that nanoforms of GO and rGO has been reported to show better adsorption potential for the removal of pharmaceutical active compounds as compared to activated carbon, carbon nanotubes, clays, and montmorillonite in a number of reports [75–77].

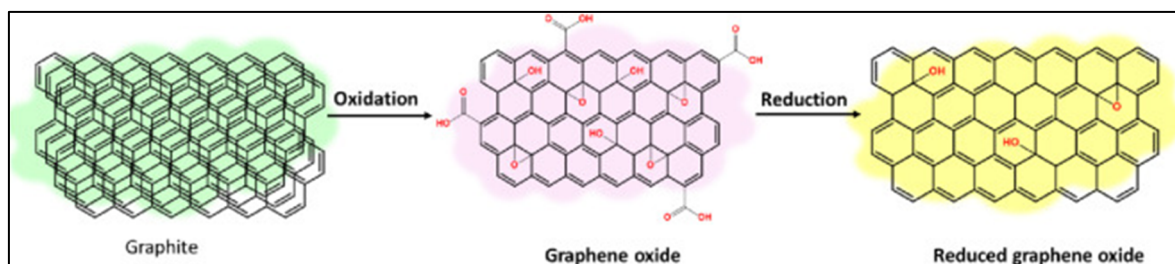


Figure 3. Schematic presentation of transformation of graphite into graphene oxide and reduced graphene oxide. Figure is adapted from [74] and is permitted to use under a Creative Commons license.

3.3. Activated Carbon

Activated carbon is a porous material having higher surface area suitable for adsorption phenomena [78]. The production of activated carbon is generally achieved from a range of materials such as coal, residual agricultural biomass, coconut shells etc through physical and chemical methods [79]. The physical activation process of carbon employs carbonization at high-temperature followed by gasification either through steam or CO_2 . In contrast the chemical activation process employs phosphoric acid and potassium hydroxide but at lower temperatures [80]. However, the adsorption potential of activated carbon depends on distribution of pore size and surface chemistry [81]. The hydrophobic property of activated carbon and its ability to interact with pharmaceutical compounds through π - π interactions makes them useful in water purification technology [82,83]. There are many studies reports on the utilization of activated carbon for the removal of pharmaceutical compounds with higher removal efficiencies [84,85]. Activated carbon can be subjected to surface modifications to improve its efficiencies for specific pollutant removal. In a nutshell activated carbon are exciting cost-effective and sustainable material for the elimination of pharmaceutical compounds from contaminated water.

3.4. Fullerenes and Carbon Quantum Dots

Fullerenes which are commonly known as bucky balls are allotropic form of carbon. Harold W Kroto, Robert F Curl and Richard E Smalley discovered them in 1985 [86]. These exists in a closed cage structure of hexagonal (20) and pentagonal (12) rings in icosahedral. All the carbons atoms are bonded to each other in sp^2 hybridization. Fullerene have a total of 60 carbons in their structure and have two different bond lengths i.e., 6:6 bonds and 6:5 bonds [87]. Fullerenes in composite and nanoforms has been reported for the removal of pollutants from pharmaceutical wastewater and other wastewater. Fullerenes produce active oxygen species which are useful in photocatalytic activity in oxidation processes for the wastewater treatment [88]. Fullerenes also exhibit adsorption properties which are useful in pollutant removal. In a study, functionalized magnetic fullerene nanocomposites have been reported the removal of ciprofloxacin due to their adsorption properties [89].

Carbon quantum dots which are also known as fluorescent carbon nanoparticles have been emerged as new carbon material. Carbon quantum dots have size less than 10nm. They have good optical property, low toxicity and low production cost [90]. The functionalization advantage of carbon quantum dots helps them for multiple application such as sensing, bioimaging, medicine, photocatalysis and electrocatalysis [91,92].

3.5. Nano-Carbons as Adsorbent

Carbon-based adsorbents have been extensively utilized in the separation and purification technologies including the removal of pharmaceutical contaminants. Activated carbons, biochar, and carbon nanotubes are the most widely investigated and utilized materials for the adsorption [93]. Activated carbon is especially valued in water and wastewater treatment owing to its high surface area, porosity, and availability. Adsorption, which harnesses the high specific surface area of adsorbents, is considered one of the most effective and economical approach [94]. Adsorption performance, however, can be influenced by several factors, including contaminant size, concentration, molecular weight, temperature, and physicochemical properties. Compared with other carbon-based adsorbents activated carbon generally exhibit higher efficiency for removing pharmaceutical pollutants and are often integrated with complementary treatment methods to further enhance removal efficiency [95]. Furthermore, activated carbon-based adsorption is advantageous in terms of reusability, operational simplicity, and overall effectiveness.

4. Mechanism of Pharmaceutical Removal by Nanocarbons

Several nanocarbons including graphene and CNTs are most used for pharmaceutical waste removal. Among the mechanisms adopted by nanocarbons for drug degradation, adsorption and photocatalytic degradation are the two most prominent techniques.

4.1. Adsorption (Physical and Chemical)

A surface phenomenon, in which solutes get adhered to solid material surface is known as adsorption. Nanocarbons absorb pharmaceutical waste materials into porous structure or onto the surface, through various chemical and physical interactions. Nanocarbons are highly efficient contaminant removers, due to their high surface to volume ratio, that in turn provides many active sites for adsorption [96]. Various mechanisms are involved in adsorption of pharmaceutical wastes on to the surface of nanocarbon and has been schematically presented in the Figure 4.

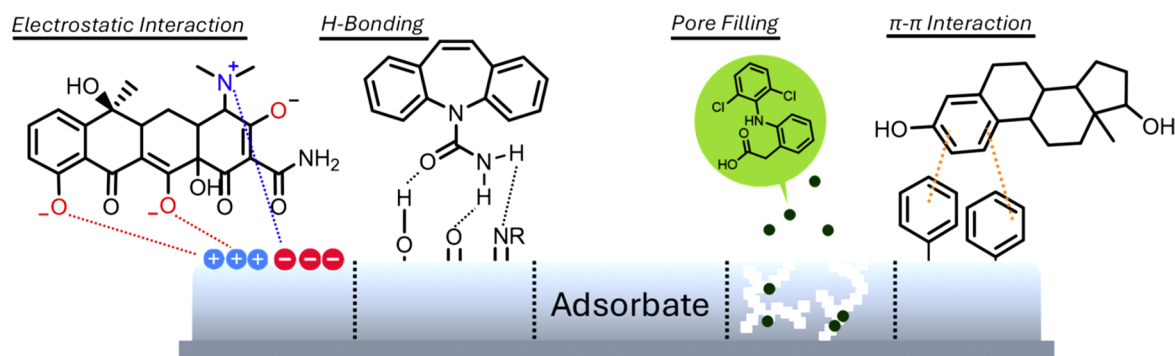


Figure 4. Schematic presentation of adsorption mechanisms of pharmaceutical pollutants by various adsorbent (Image adapted from [97] and is permitted to use as the article is under a Creative Commons license <http://creativecommons.org/licenses/bync-nd/4.0/>).

4.1.1. Hydrophobic Interactions

Most of the pharmaceutical compounds are hydrophobic or partially hydrophobic in nature. Certain nanocarbons such as pristine graphene and CNTs possess hydrophobic surfaces which helps in the adsorption through π - π stacking interaction and van der Waals forces (Figure 4). This kind of non-covalent interaction helps in the strong binding of aromatic pharmaceutical materials to conjugated carbon structures [98].

4.1.2. π - π Electron Donor-Acceptor Interactions

Delocalized π -electron systems present in the nanocarbons interact with electron deficient or π -electron-rich, pharmaceutical compounds. For instance, aromatic rings present in the pharmaceutical compounds forms π - π stacking interactions along with graphene layers, which enhance absorption efficiency (Figure 4). This kind of mechanism is crucial for the absorption of pharmaceutical compounds like ibuprofen, diclofenac and naproxen, which are aromatic and non-polar in nature [99].

4.1.3. Electrostatic Interactions

There is an electrostatic interaction among positively charged pharmaceutical ions and negatively charged nanocarbon (Figure 4). This is mainly due to the presence of oxygen containing groups (such as epoxy, hydroxyl, carboxyl) that imparts negative charge to the surface. This facilitates electrostatic attraction between pharmaceutical ions (such as analgesics and antibiotics) and nanocarbon under appropriate pH [100].

4.1.4. Hydrogen Bonding

Functional groups present on the surface of nanocarbons (after surface modification) forms hydrogen bonds with various pharmaceutical compounds. For example, carboxyl or hydroxyl groups present on the surface of graphene oxide interacts with hydroxyl or amine group of pharmaceutical molecules, which enhance adsorption (Figure 4) [101].

4.1.5. Pore Filling Mechanisms

Activated nanocarbon structures possess a high degree of micro-porosity, which can physically trap minute pharmaceutical particles by diffusion driven adsorption or capillary condensation (Figure 4). This mechanism plays a significant role in total adsorption. Nanocarbons possess three types of pores, such as micropores (favors high adsorption capacity), mesopores (facilitates transport of pharmaceutical molecules towards micropores) and macropores (helps in faster diffusion) [102].

4.2. Catalytic Degradation

Catalytic degradation is a process in which harmful pharmaceutical molecules are transformed into less toxic materials mainly by redox reaction, which is catalyzed by nanomaterials. When compared to adsorption, catalytic degradation is more beneficial as it leads to partial or complete breakdown of the pharmaceutical compounds [103]. Nanocarbons hybridized with metal nanoparticles or in certain scenarios doped along with heteroatoms, may act as catalysts that facilitate degradation reactions.

4.2.1. Types of Catalytic Process

Fenton-like Reactions

Nanocarbons possess the capability to catalyze Fenton or Fenton—like reactions when it is coupled with $\text{Fe}^{2+}/\text{Fe}^{3+}$ or any other transition metals. These Fe-based nanocarbon compounds generate hydroxyl radicals in the presence of hydrogen peroxide, which in turn is capable of degrading pharmaceutical compounds. Graphene oxide (GO) possesses oxygen functional groups which can enhance Fe ion distribution and stability, that boosts radical generations and efficiency of degradation [104].

Persulfate Activation

Persulfates in the activated form generates sulfate radicals ($\text{SO}_4^{\bullet-}$) which are strong oxidants. When nanocarbons are combined with transition metals such as Mn and Co, or doped with nitrogen, have the capability to activate persulfates indirectly. This is mainly due to the formation of reactive sites, ability to transfer electrons and enhanced redox property. These radicals have the potential to oxidize pharmaceutical compounds through electron transfer reactions, which break down complex structures [105].

Photocatalysis

Photocatalysis is light controlled mechanism, in which a semiconductor absorbs photons, which in turn produces electron-hole pairs that initiate redox reaction. This reaction gradually breakdown complex pharmaceutical compounds into less toxic products such as water and carbon dioxide. Nanocarbons such as carbon dots, carbon nanotubes (CNTs) and graphene, play a crucial role in pharmaceutical compound degradation through photocatalysis [106].

The mechanism of photocatalysis begins when a photocatalyst such as zinc oxide or titanium dioxide is irradiated with visible or ultraviolet light. This source of energy leads to excitement of electrons from the valence band towards the conduction band, that generates hole in valence band. These photo initiated electrons and holes often migrates towards the surface and reacts with oxygen and water molecules for producing ROS (Reactive Oxygen

Species). Some of the commonly produced ROS are superoxide anions and hydroxyl radicals. These kinds of ROS molecules are highly reactive in nature and attacks pharmaceutical compounds by breaking chemical bonds [107].

Nanocarbons plays a crucial role in the process of photocatalysis, by serving as electron acceptors or conductors, that facilitates rapid charge separation and reduces the electron hole pair recombination. For example, titanium dioxide is combined with graphene, the latter acts as a electron sink, that accepts electrons and transports it away from the photocatalyst surface, which in turn enhances the ROS generation efficiency [108].

Also, nanocarbons increases the surface area and provides more active sites that favours pharmaceutical compound adsorption and degradation. The π - π interactions among aromatic pharmaceutical molecule and nanocarbon, increases adsorption, by bringing pollutant molecules closer towards the reaction zones. Finally, nanocarbons have the capability to extend adsorption of light into visible spectrum, especially when doped or modified along with non-metal or metal elements. This facilitates the photocatalyst to use large area of solar energy, which in turn makes the process more energy efficient [99].

Furthermore, nanocarbons such as carbon dots produces ROS independently, thus acting like a standalone photocatalyst. These nanocarbons possess the quantum confinement and tuneable photo-luminescence, which enables them to strong interaction with pharmaceutical compounds and facilitates degradation. Some of the nanocarbons such as carbon nanotubes, graphene quantum dots and carbon dots possess photo-luminescence properties due to their chemical stability, optical property, low toxic behaviour and biocompatibility [109].

Nanocarbon's ability to increase surface area, generate ROS independently, enhance charge separation and extend absorption of light, makes it ideal for pharmaceutical waste removal [110]. More research studies should be conducted for optimizing nanocarbon based photocatalysts for real world applications. Thus, the photocatalytic mechanism in nanocarbon based composites include these key steps. Light absorption with the help of UV or visible light leads to electron excitement, which in turn creates electron hole pairs. The next step is the charge separation and transfer in which nanocarbons such as graphene accepts the excited electrons, which minimizes recombination as it acts as electron sinks. In the next the dissolved oxygen is reduced to superoxide radicals by electrons. Hydroxide ions are also oxidised by holes into hydroxyl radicals. CODs may act as photosensitizers, thus generating ROS in visible light. Finally, generated ROS attacks pharmaceutical compounds, gradually breaking them into intermediates and finally into non-toxic substances like water, carbon di oxide and inorganic ions (Figure 5) [111].

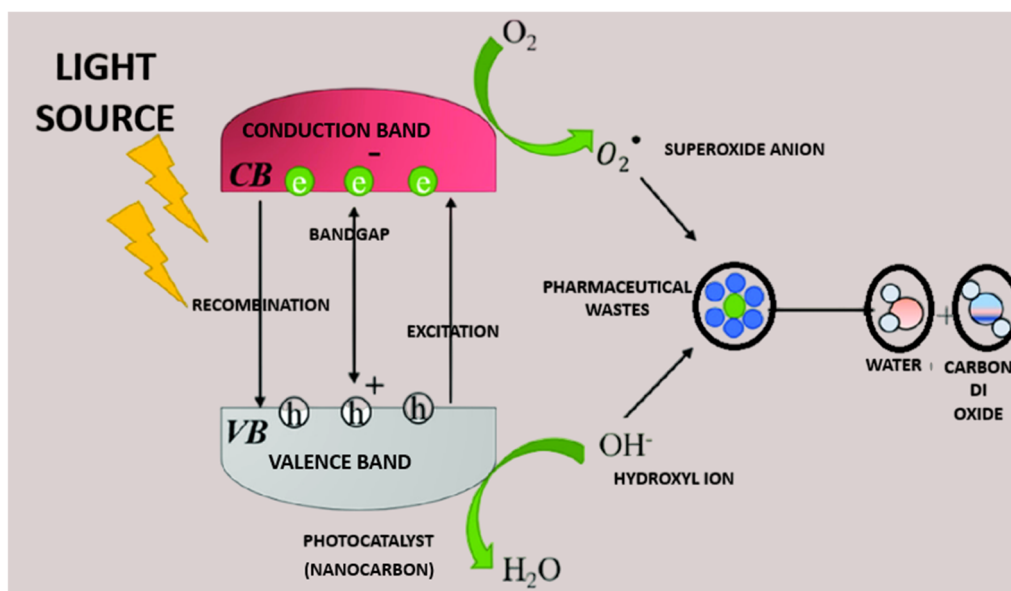


Figure 5. Schematic diagram of photocatalytic degradation of pharmaceutical compounds using nanocarbons.

4.3. Factors Influencing Removal Efficiency

The efficiency of pharmaceutical molecule removal by nanocarbons is dependent upon various environmental, operational and physio-chemical factors.

4.3.1. Physio-Chemical Properties of the Pharmaceutical Compounds

Hydrophobicity, molecular structure, charge and solubility of the pharmaceutical compound play a crucial role in increasing efficiency of the removal mechanism by nanocarbons. Functional groups and aromatic rings

influence the interacting strength with nanocarbons. Also, non-polar carbon surfaces (e.g., Pristine carbon) always prefer hydrophobic nanocarbons to adsorb, indicating the relevance of hydrophobicity in degradation mechanism. Apart from these, the solubility and ionization state of the pharmaceutical compound also affect adsorption, which in turn affects efficiency [112,113].

4.3.2. pH and Temperature

Kinetic effects play a crucial role in efficiency of nanocarbons. High temperatures normally increase the reaction rates and diffusion, but it destabilizes the nanocarbon structure and leads to desorption of the drugs. Both the drug and nanocarbon may lose or gain charge which depends on the pH, this in turn affects electrostatic interactions. At optimum pH, adsorption will be maximum because of favorable charge alignment among the pharmaceutical molecules and nanocarbon [112,114].

4.3.3. Doping and Surface Functionalization

Hetero-atom doping of nanocarbons introduces active sites that increases conductivity and extends response of visible-light. Also, coupling nanocarbons along with ZnO, Ag or TiO₂, expands photocatalytic activity mainly by charge separation and ROS generation [115].

4.3.4. Contact Time and Dosage

Sufficient contact time is required for the proper interaction of pharmaceutical compounds and nanocarbons. Higher doses of nanocarbons improve drug removal, and beyond a particular limit there would be reductions in surface area and penetration of light [116].

4.3.5. Occurrence of Co-Contaminants

Various co-contaminants occur along with pharmaceutical compounds, which usually compete for catalytic sites. Thus, the presence of other similar co-contaminants reduces the specificity of nanocarbons to the targeted drug. This competition alters thermodynamics and kinetics of adsorption, which makes selection more challenging [113].

5. Recent Advances and Emerging Trends

5.1. Hybrid Materials and Composites

Hybrid nanosystems are noticeably well-known for the removal of pharmaceuticals from water and wastewater, compared to that of the traditional biological techniques, photocatalysis, oxidation and membrane separations [117]. Encouraging redox potentials, highly reactive surface area and sufficient charge carrier separations of metal oxide based nanospheres (such as Bismuth Oxide, Titanium Dioxide, Zinc Oxide) have gained significance in pharmaceutical drug removal and simultaneous degradation [118]. Nanocarbon dots from graphite remove tetracycline, from citric acid removes ciprofloxacin, tetracycline and gemfibrozil, from L-ascorbic acid removes levofloxacin [119]. On the other hand, less explored hybrids like Ferric oxyhydroxide based nanocarbon dots were researched to fully remove tetracycline by 40 min [120]. Just recently, blends of nanomaterials and biochars is a subject of critical focus due to their photocatalytic and adsorption properties respectively. Functionalized biochar based nanoblends have shown to remove acetaminophen more effectively than ciprofloxacin [121]. According to [122], the removal of phenol from pharmaceutical wastewater was 92.9% with silver-doped magnetic biochar [Ag-Fe₃O₄/BC] than the unmodified biochar (78.6%), enabling the modified one as an advanced adsorbent. Such biochar-based nanocomposites are of special emphasis nowadays. Leao et al., compared carbon nanomaterials having different dimensions and oxidation levels for atenolol adsorption [123]. The results from the study showed that nanocarbons of higher dimensionality and oxidation have higher sorption capacity for the atenolol. Furthermore, three-dimensional oxidized reduced graphene oxide exhibited the highest removal efficiency and reusability, making it a promising material for water treatment. Zhao et al., developed carboxylated nano carbon skeleton microspheres and modified with octadecylamine to achieve superhydrophobicity [124]. The results from the study showed the high adsorption of toluene (509.43 mg/g) and other benzenes by developed carbon skeleton microspheres. Recently, Makhseed et al., have reported phenothiazine derived nanocarbon (PTZ-C) with a high surface area (3041 m²/g), uniform pores (1.89 nm), and good adsorption capacity for diclofenac (588.2 mg/g) and caffeine (333.3 mg/g) [125]. Furthermore, the PTZ-C also showed the iodine uptake up to 6.322 g/g in vapour with good retention. PTZ-C were reusable for multiple cycles.

5.2. Smart and Responsive Nanocarbon Systems

Due to huge drug intake, active pharmaceutical materials are unavoidable water contaminants. Surface structures' complexities make such drugs' recovery harder for which advanced nanocarbon based materials pave a speedy way for removal [126,127]. Studies on integrating nanocarbon based scavengers to develop smart filters for the effective removal of pharmaceutical materials showed about 99% removal of gemfibrozil and triclocarban followed by 63% removal of carbamazepine, enabling out performance of the existing scavengers [128]. Smart nanosystems involve formulations of hybrid liquid (ionic supramolecular) gels doped up with nanocarbon materials, using which maximal removal efficiency of about 51% in 3 h was obtained for ciprofloxacin, with a relatively slow removal efficiency of around 88% throughout a day. Such gels are regenerative and recyclable. Coupling such nanogels to dialysis membranes would be a realistic approach to this finding, suggesting them as challenging candidates for pharmaceutical drug removal from water [129]. Graphene based carbon nanotubes (multi-walled) blended with titanium dioxide based nanostructures (single-walled) were impregnated on textile to attain effectual photocatalysis, where a dye (methylene blue) and a drug (ciprofloxacin) were tested with irradiation [130]. About 95.7% drug degradation within a time span of 120 h was achieved. This evidenced the safe removal of pharmaceutical material using nanoblends. Waste tea residue derived carbon dots were examined for the removal of organic contaminants that turned successful [131].

5.3. Integration with Filtration and Membrane Technologies

Graphene based nanosheets and/or functionalized membranes [126] are appreciated for their hydrophilicity due to which their incorporation into polymeric membranes and ceramic membranes through membrane fabrication, drop coating, vacuum filtration, impregnation into electrospun polymeric layers or through spin casting finds great attention for the ease and less rough nature of the same [132]. Carbon nanofibers, graphite, graphene oxide and carbon nanotubes, activated carbon, were investigated by researchers as promising platforms for membrane technology and nanofiltration. In such systems, nanopores play a vital role in enhancing the flux, wettability, mechanical strength, interaction with drugs and hydrophilicity of pharmaceutical drugs contaminated water [133]. Some novel particles like MXene are at the beginning of their comprehensive research findings. Carbon nanoparticles in membrane systems are in with the advancements of gradient-porous, doped nanostructures, especially nanotubes [134]. Further expansions in terms of ball-milled and/or heat-treated carbon nanomaterials in such technologies need extensive experiments [135]. Previous studies also include some state-of-the-art alterations like plasma functionalization in these models that may further activate the gaseous levels in water during drug removal. Engineered nanocarbons have been significantly tested for alternative wastewater/water disinfection techniques [136].

5.4. Artificial Intelligence and Modelling for Predictive Performance

Traditional nanocarbon evaluation methods are adversely affected with lack of sophistication, accuracy and technicality [137]. In this context, artificial intelligence (AI) with mining attributes is aiding as a supreme weapon in nanocarbon technology for nano-scale-structures-designing via data image processing and programmed algorithms [138]. AI driven technologies utilize data-driven models, algorithms (machine learning), and decision-making systems to optimize, analyze, and predict the processes. AI modelling technique is particularly more popular in adsorption processes hence it can be utilized in the nanocarbon based adsorption processes for the removal pharmaceutical contaminants [139]. It has been reported that AI's integration into the adsorption process improved the efficiency by 20% as revealed by the preliminary tests. Furthermore, AI models also permit the adjustment of process parameters (adsorbent concentration and contact time) to achieve maximum pollutant removal [140]. Machine learning has also been employed in the adsorption modelling to analyse large datasets and the predict the adsorption capacities, isotherms analysis, and kinetics calculations [141]. Some of the most common algorithms are SVM, decision trees, random forests, and neural networks [142]. However, the choice of algorithm highly depends on the nature of task, complexity of dataset, and the desired interpretability. Researchers reported the support of Convolutional Neural Networks (CNN) in keenly investigating the mechanisms of nanocarbon augmentations and microstructural characterization; providing a larger scale image results and meticulous interpretations to gain a deeper understanding about the formulated nanomaterials and mechanisms of drug removal [143]. An intense knowledge about the cracking patterns of the nanocarbons, drug removal mechanisms, their strength, shelf life, bonds' breakages in atomic level, performance and time period is crucial for the nanocarbon synthesis and programmed actions to occur, which could be accurately predicted by the modern AI tools and programmes [144].

6. Challenges and Future Perspectives

Although nanocarbons have remarkable potential for the pharmaceutical pollutant removal but still there are a number of challenges that need to be addressed for its global acceptance and industrial application. The aggregation of carbon-based materials is a big challenge in aqueous remediation process of pharmaceutical compounds. The aggregation and poor dispersion of carbon nanomaterial in water reduces their effective accessible surface area hence therefore reduces the adsorption efficiency. The varying pH due to the nature of drug molecules and other by products some time causes the destabilization and aggregation of nanomaterials. Functionalization of carbon nanomaterial is also an important aspect for its application in pharmaceutical contaminant removal. Although introducing oxygenated, polar, or other active functional groups in carbon nanomaterials can increase affinity for pharmaceuticals but at the same time it can also reduce its structural stability [145]. The lower concentrations of many pharmaceutical compounds in waters resources makes it difficult to remove and translate it to next level [146]. Furthermore, the repeated adsorption/desorption cycles may degrade the material and reduce its adsorption capacity and stability in realistic conditions [147]. The coexisting drug species or chemicals and other organic matter in the wastewater can interfere with adsorption and degradation performances of nanomaterials [137]. The scalability is also a concern that need to be addressed [148]. Highly functionalized and defect-engineered nanocarbons may be expensive and sometimes uses hazardous reagents which could be responsible for their toxicity [149]. Furthermore, selectivity of carbon nanomaterials can also affect the pharmaceutical pollutant removal process. Therefore, a regulatory framework for utilization and disposal of carbon nanomaterials is needed. The development of green approach for the production of carbon nanomaterials can help in the reduction of toxic chemicals release and environmental toxicities. The designing multifunctional carbon nanomaterials could be an effective approach e.g., catalytic as well as adsorption potential. Surface engineering of nanocarbon can help in increasing their specificity, selectivity and stability. The integration of nanocarbon as adsorbents and catalytic agent into membranes, packed bed reactors, solar-driven systems and demonstration of their potential at pilot-scale can be pivotal to validate their feasibility and transition from bench to field.

7. Conclusions

Nanocarbon-based materials such as activated carbon, biochar, carbon nanotubes, and graphene and their derivatives have emerged as promising alternative for the removal of pharmaceutical pollutants from water resources. The distinctive feature of these nanomaterials such as high surface area, porous structure, and versatile surface modifications are advantageous for the pharmaceutical contaminant removal. The adsorption efficiency of carbon nanomaterials is generally governed by π - π interactions, electrostatic forces, and hydrophobic effects, making them suitable for hydrophobic pharmaceuticals. Activated carbon remains the most widely applied due to its cost-effectiveness and reusability. CNTs and graphene's are commendable in performance due to the surface modification luxury. However, there are certain challenges for the global acceptance and industrial scale application such as stability, selectivity and toxicity. Therefore, future research should be focused on the optimizing green methods for the production of carbon nanomaterials, functionalization methods, integration of nanocarbons with advanced treatment methods, and assessment through pilot-scale studies to ensure its practical and sustainable implementation.

Author Contributions

D.S.: Conceptualization, Data curation, Formal analysis, Investigation, Validation, Visualization, Writing—original draft, Writing—review & editing; P.C.: Data curation, Visualization, Writing—original draft; J.T.: Data curation, Visualization, Writing—original draft; M.U.: Data curation, Visualization, Writing—original draft; B.T.: Data curation, Visualization, Writing—original draft; S.S.M.: Data curation, Visualization, Writing—original draft; K.K.: Data curation, Visualization, Writing—original draft; V.K.: Conceptualization, Validation, Writing—review & editing, Supervision. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest

The authors declare no conflict of interest.

Use of AI and AI-Assisted Technologies

No AI tools were utilized for this paper.

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