Removal of antibiotic from the water environment by the adsorption technologies: a review

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ABSTRACT

Antibiotics are known as emergent pollutants because of their toxicological properties. Due to continuous discharge and persistence in the aquatic environment, antibiotics are detected almost in every environmental matrix. Therefore antibiotics that are polluting the aquatic environment have gained significant research interest for their removal. Several techniques have been used to remove pollutants, but appropriate technology is still to be found. This review addresses the use of modified and cheap materials for antibiotic removal from the environment.

Key words | antibiotic, effect, environment, removal, source, water

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INTRODUCTION

Antibiotics attenuate or inhibit the proliferation and growth of microorganisms (Bilal *et al.* 2020) and are useful in the treatment of human infectious disease, livestock industry and aquaculture (Anchordoquy *et al.* 2019; Ben *et al.* 2019; Cycoń *et al.* 2019). The estimated consumption of antibiotics worldwide lies between 1×10^5 and 2×10^5 tons annually (Mala & Dutta 2018; Wang *et al.* 2019). Antibiotics after administration are only partially metabolized in the body, while the rest of the antibiotics are excreted via feces and urine (50–80%) into the ecosystems (Duan *et al.* 2019; Bilal *et al.* 2020). Sewage treatment plants of the municipal systems and pharmaceutical businesses are not efficient enough to remove these antimicrobials. The effluents containing antibiotics then get access to the aquatic matrix (Balakrishna *et al.* 2017; Azanu *et al.* 2018; Binh *et al.* 2018). Studies have confirmed the occurrence of pharmaceutical products in sludge, soil, sediments, surface water, underground water, loams, overland running water, aquatic and terrestrial animals and plants (Bilal *et al.* 2020).

Antibiotic accumulation has become the major cause of concern to scientists and environmentalists (Azhar *et al.* 2016). Nowadays special attention has been given to the effects these potentially harmful antibiotic residues (micro-contaminants) on the living environment (Muhammad *et al.* 2020) as these biologically active residues, even in small traces, induce the multi-resistance in bacteria, which poses serious issues for aquaculture, humans, agriculture and livestock (Li *et al.* 2018; Bilal *et al.* 2020). The presence of antibiotics in the environment is associated with chronic toxicity to some organisms (Muhammad *et al.* 2020). Their presence in the aquatic environment has grabbed attention for the potential toxicity on non-target species also (Machado & Soares 2019). The long time exposure to low-level antibiotics produces toxic effects which include hypersensitive reactions, abnormalities in digestive system functioning and allergic reactions to the microorganisms (Azhar *et al.* 2016; Azanu *et al.* 2018).

The removal of antibiotics from the aquatic environment remains the main focus to reduce the toxicity on the aquatic flora and fauna. For that conventional systems are being widely used for the removal of antibiotics from wastewater systems (Al Aukidy *et al.* 2014). Several methods have been reported for the removal of antibiotics (advanced oxidation, ozonation, reverse osmosis, membrane filtration, electrochemical methods and biological treatments) (Sri *et al.* 2018; Liu *et al.* 2020). Most of these techniques are costly and result in by-product formation or they are less effective. The high-cost production, maintenance and not being environment-friendly make it important to look for a more appropriate and convenient method to remove antibiotics.

Among the available methods, adsorption technologies features of simple design, ease of operation and comparatively low cost of application and thus are considered to

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be one of the highly efficient methods for removal of different pollutants from the aquatic bodies (Liu *et al.* 2020).

DIFFERENT SOURCES OF ANTIBIOTICS: AQUATIC POLLUTION

Overuse and inappropriate use of antibiotics and negligent disposal are a major concern for the environment. Because of the chemical nature of the antibiotics, they remain unmetabolized in the system either completely or partially. Due to this, the ecosystem gets polluted and the antibiotics pose a health hazard to the flora and fauna. Figure 1 depicts the primary sources through which antibiotics enter the water environment and then in the human food chain.

Production and consumption

Antibiotics are widely used for agricultural purposes, animal farming and human therapy (Mala & Dutta 2018). Gradually, new and revised versions of antibiotics are being added to tackle various ailments. The approximate production of antibiotics in India during 2006–2007 was more than 2,332 Mt and production increases 10% each year (Mutiyar & Mittal 2013). The antibiotic production for use in farming in the United States in 2003 was about 9,200 tons (Hu *et al.* 2010). According to the European Federation of Animal Health (FEDESA 2001), 13,288 tons of antibiotics were used in Switzerland and the European Union, and 65% was used as human medicine (Al- Ahmad *et al.* 2009). In China, 210,000 tons of antibiotics were produced each year, 48% of which is applied in the agriculture and livestock industry (Zhou *et al.* 2013). Globally, the consumption of antibiotics grew more than 30% from 50 billion to 70 billion standard units in 71 different countries and most of these antibiotics that are used are either recommended by health care providers or procured directly by consumers (Van Boeckel *et al.* 2014). In New Zealand (2002), around 93 tons of antibiotics were used for the veterinary purpose (Hou *et al.* 2015). There has been about 41%

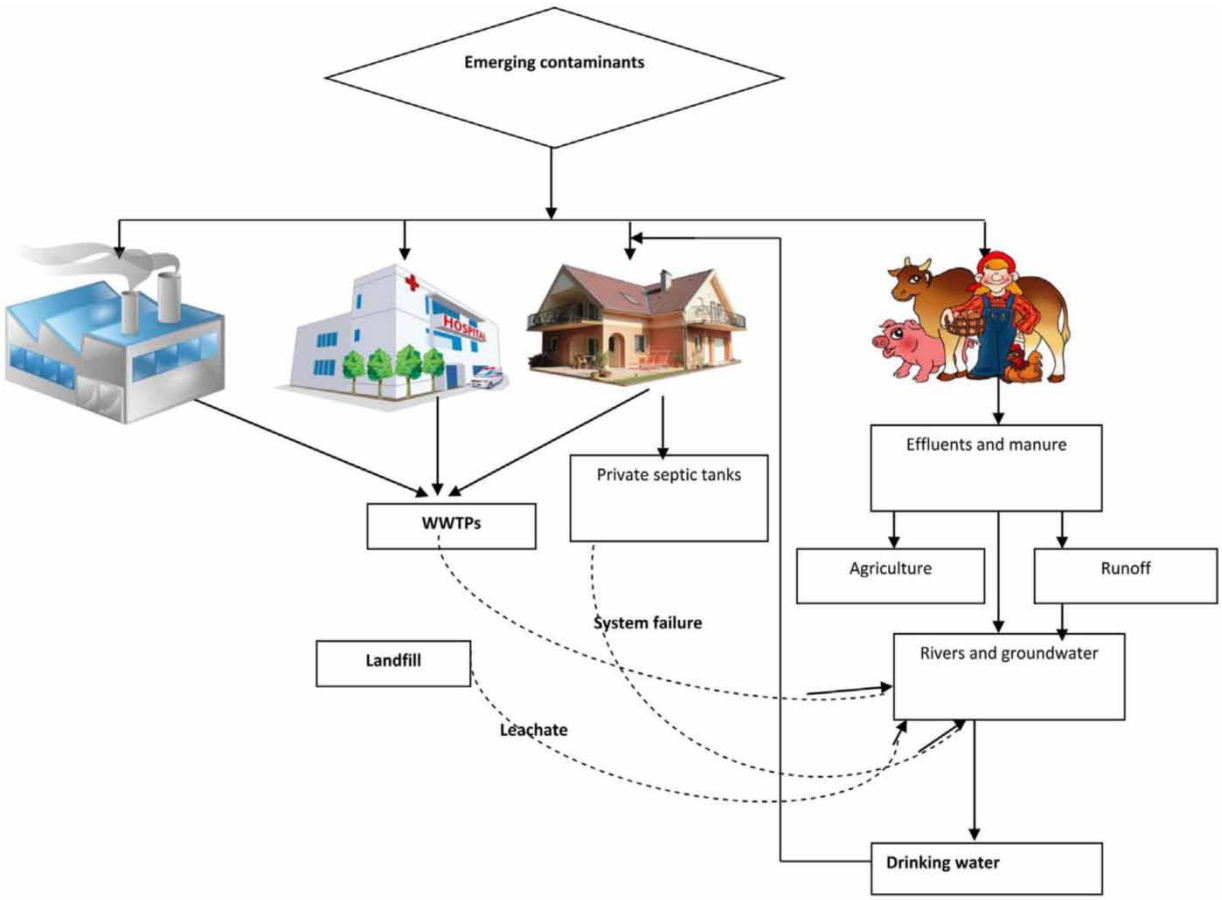

Figure 1 | Different pharmaceutical sources into the environment (Sophia & Lima 2018).

increase in penicillin and cephalosporin consumption in the first decade of this century. Van Boeckel *et al.* (2014) estimated that the 10-yearly antibiotic consumption in livestock may increase by 120% in China, 60% in the USA and 100% in India, to treat common infections. The countries which consumed most antibiotics overall in 2010 were India, 13 billion standard units; China, 10 billion standard units; and US 7, billion standard units. Emission of antibiotics from production units has found importance, as antibiotic concentrations of several mg/L have been detected in effluents coming out from these production units (Larsson *et al.* 2007).

Domestic waste (hospital, veterinary, municipal waste) in the environment

Antibiotics are used to treat infections and are foundational components of modern medicine. They are used for the treatment of diseases caused by bacteria. Antibiotics are broadly used in human, veterinary medicines and in aquaculture practice to promote growth. However, indiscriminate use, improper handling, and disposal have resulted in their existence in the environment. Municipal and hospital waste, and human and animal excretions consistently are also contributing the partially metabolized drug to the ecosystem. The following are the sources of leakage and addition of antibiotics in the aquatic ecosystem, posing current threats to the world through the development of antibiotic resistance, which is a serious concern.

Hospital effluents

Hospital effluents are considered as a chief source of antibiotics in the environment, and are the main source in terms of generated pharmaceutical load (Diwan *et al.* 2009). The wastewater of the hospital is composed of effluents of different services: laboratories, radiology department, outpatient departments, transfusion centers and wards (Verlicchi *et al.* 2012). Along with health care establishments, the hospital wastes are discharged directly into the urban sewer systems (Sharma *et al.* 2013). The hospitals discard about 1,250 tons of pharmaceuticals per annum as waste, which are either used or unused (Brain *et al.* 2009). Pharmaceuticals that are discharged in hospital wastewater are found to be in concentrations higher than in municipal sewage (Ashfaq *et al.* 2016). Many of the chemical substances used by hospitals, such as pharmaceuticals, disinfectants and solvents for medical purposes, are being found to be resistant to wastewater treatment plant (WWTP) processes (Sharma *et al.* 2013).

Veterinary wastes

The veterinary antibiotics used in livestock farms are used to increase livestock productivity, suppress parasites and prevent disease caused by bacteria (Awad *et al.* 2014). They are regarded as emerging contaminants (Rodríguez-Navas *et al.* 2013), widely in use worldwide, which are released incompletely metabolized into the environment (Kołodziejska *et al.* 2013). They enter the environment either by animal excretion, by discharge of wastewater, or by way of manure application in aquaculture (Pan & Chu 2017). The consumption of veterinary antibiotic consumption globally up to 2010 was at least 63,000 tons and is expected to reach 106,600 tons by 2030 (Pan & Chu 2017). About 30–80% of antibiotics are defecated into the environment via manure (Zhang *et al.* 2014).

Municipal wastes

The household pharmaceuticals enter municipal wastewaters through three major ways, which are: (1) human excreta, (2) household wastes, and (3) wash-off of the human body. Generally, the expired or remainder medicines are disposed of down household drains. In a study in Germany, one-third of total pharmaceuticals and 25% in Austria (Kümmerer 2009) are disposed as wastes down the drain. Therefore, the disposal of unused pharmaceuticals, either down the toilet or in household waste, is the prominent route that needs a greater attention. After administration of these useful drugs, the incompletely metabolized drugs are excreted and transported to the sewage treatment plant (Gao *et al.* 2012). As 10–25% of antibiotics consumed by patients come directly from hospitals and as the municipal system is receiving these hospital wastes, the municipal wastewater becomes an important source of antibiotics (Valitalo *et al.* 2017).

Landfill leachates may be responsible for the presence of pharmaceuticals for human medical care in groundwater (Heberer 2002). An increasing amount of proof indicates that the leaching of antibiotic compounds in groundwater and deeper soil profiles. There are reports of municipal landfill leaching which contains antibiotics that end up in water bodies (Li 2014). Sulfadimethoxine and sulfamethazine are

reported from six private wells, which are used as sources for drinking water by the residents of Washington County, USA. In the US Geological Survey, sulfamethoxazole is detected at a high relevance ratio of 23%, and from 23 European countries, 24% in 164 individual groundwater samples (Tong *et al.* 2014). There are numerous other different important classes of antibiotics that have been reported in water bodies (Martinez 2009). The input of antibiotics in the aquatic system from agricultural fields is generally via surface runoff (Kümmerer 2009).

AGRICULTURAL WASTES

The use of antimicrobials in food production is divided into livestock production, fish farming and crop growing (O'Neill 2015). The practice of antibiotic use in aquaculture is a growing concern as residues of antibiotics have been detected in the fish products (O'Neill 2015). The aquatic environment is a fast medium for spreading of antibiotics. It is suggested that 70–80% of antibiotics given to fish are excreted into water (Burridge *et al.* 2010). Since the 1950s, control of diseases of vegetables, high-value fruits and ornamental plants has been in practice (Kümmerer 2009). The frequent use of antibiotics for agricultural purposes results in their continuous release and detection in the environment (Zhou *et al.* 2016). Antibiotics reported in wastewater and through irrigation can be introduced into the agricultural lands (Gulkowska *et al.* 2008). Manure and sludge which are used as fertilizers for agricultural land are often contaminated with antibiotics (Cheng *et al.* 2019). The WWTPs incompletely remove antibiotics (Escher *et al.* 2011), which then find their way into surface waters (Brown *et al.* 2006).

PRESENCE OF ANTIBIOTICS IN WATER BODIES

Most of the countries are facing water quality issues, as every source of water has been reported to be contaminated with different contaminants including antibiotics (Gothwal & Shashidhar 2015). The production, consumption and unsafe disposal of antibiotics are responsible for their frequent detection in every environmental matrix (Ashfaq *et al.* 2016). Several investigators have reported the antibiotic presence in different water compartments, including WWTP effluents (Guerra *et al.* 2014), sediments (Awad *et al.* 2014), surface water (He *et al.* 2015), groundwater (Ma *et al.* 2015) and seawater (Zhang *et al.* 2013). An overview of the different works published in different journals in this area is presented in Table 1.

ANTIBIOTIC EFFECTS ON AQUATIC ORGANISMS

Antibiotics are synthetic, semi-synthetic or natural compounds, which are able to kill or inhibit growth or metabolic activity of microorganisms. These compounds are biologically active molecules with antibacterial activities and these properties of antibiotics have become the cause of concern, due to potential impact on non-target organisms (Machado & Soares 2019). Long-term change of the composition of the bacterial community can lead to variations in aquatic ecosystems (Xiong *et al.* 2019).

Development of antibiotic resistance: a major concern

The presence of antibiotics in water exposes the pathogens for a longer period and therefore spreads resistance among the microorganisms (Agerstrand *et al.* 2015). However, recent reports have suggested that these bacteria have gained resistance even to common antimicrobials. According to the World Health Organization, in 2014 *Escherichia coli*, *Klebsiella pneumoniae* and *Staphylococcus aureus* were the greatest agents of hospital and community-acquired infections (Gelband *et al.* 2015). In a study reported by Arslan *et al.* (2005), the prevalence of infections is increasing due to antibiotic-resistant bacteria which make urinary infection treatment more difficult. Fluoroquinolones like ciprofloxacin, norfloxacin, ofloxacin and lomefloxacin are the major contributors to developing resistant microorganisms. Many genes are identified to develop fluoroquinolone resistance and are thought to develop from water dwelling-bacteria and have been transferred to humans as well (Suzuki & Hoa 2012). Resistance due to other drugs has been reported, such as sulfonamides which are required to treat bacterial and protozoal infections (Blahna *et al.* 2006). Tetracycline is also a broad-spectrum antibiotic and it is reported that there is a high occurrence of resistance among microorganisms against tetracycline (Vo *et al.* 2010).

The metabolized and unmetabolized antibiotics in the environment result in development of antibiotic resistance by producing destructive enzymes, mutation (drugs cannot recognize them), reducing the permeability to antibiotics and creating bypasses (Gelband *et al.* 2015). Antibiotic-resistant organisms are disseminated among humans and animal populations by water (Baquero *et al.* 2008). Resistant

Table 1 | Detection of different classes of antibiotics in aquatic system

Antibiotics	Concentration (μg/L)	Reference
Fluoroquinolones		
Ofloxacin	1,119.0, 66.0	Ma *et al.* (2015); Ashfaq *et al.* (2016)
Enrofloxacin	4.6, 49.0	Chen & Zhou. (2014); Ma *et al.* (2015)
Ciprofloxacin	185.14, 18.0	Bai *et al.* (2014); Ashfaq *et al.* (2016)
Norfloxacin	703.0, 9.6	Ma *et al.* (2015)
Lomefloxacin	9.3, 159.0	Guerra *et al.* (2014); Ma *et al.* (2015)
Difloxacin	23.0	Ma *et al.* (2015)
Sparfloxacin	0.58	Ashfaq *et al.* (2016)
Moxifloxacin	224.0	Ashfaq *et al.* (2016)
Gemifloxacin	0.2	Ashfaq *et al.* (2016)
Tetracyclines		
Minocycline	531.7	Pena *et al.* (2010)
Epitetracycline	18.9	Pena *et al.* (2010)
Tetracycline	48.0	Ma *et al.* (2015)
Doxycycline	191.0	Chen & Zhou (2014)
Chlorotetracycline	46.7, 76.0	Chen & Zhou (2014); Ma *et al.* (2015)
Oxytetracycline	4.5, 80.0	Tong *et al.* (2014); Ma *et al.* (2015)
Sulfonamides		
Sulfamerazine	9.3	Guerra *et al.* (2014)
Sulfamethoxazole	8.3	Zhang *et al.* (2013)
Sulfathiazole	10.57	Awad *et al.* (2014)
Lincomycin	0.045	Bernot *et al.* (2013)
Cefotiam	2.4	Tong *et al.* (2014)
Sulfaquinoxaline	23.5	Yan *et al.* (2013)
Sulfadimethoxine	0.16	Bai *et al.* (2014)
Sulfamethazine	9.60	Awad *et al.* (2014)
Sulfachloropyridizine	58.29	Jiang *et al.* (2011)
Sulfadiazine	37.4,	Bai *et al.* (2014)
Streptozocin	0.5	Jiang *et al.* (2011)
Sulfapyridine	103.1	Chen & Zhou (2014)
Sulfaquinoxaline	64.2	Chen & Zhou (2014)
Sulfacetamide	12.25	Bai *et al.* (2014)
Sulfadimidine	5.91	Bai *et al.* (2014)
Sulfamethoxypyridazine	8.04	Bai *et al.* (2014)
Sulfaquinoxaline	14.59	Bai *et al.* (2014)
Macrolides		
Erythromycin	0.30	Guerra *et al.* (2014)
Roxithromycin	4.1	Chen & Zhou (2014)
Azithromycin	153.0	Collado *et al.* (2014)
Clarithromycin	100	Collado *et al.* (2014)
Tylosin	0.0004	Gao *et al.* (2012)
Lincosamides		
Lincomycin	110	Guerra *et al.* (2014)

bacteria might be transferred to humans through alternative pathways such as environmental exposure, person to person transfer and direct exposure to animals (food chain) (Mathur & Singh 2005).

Selective pressure and development of resistance genes are two basic factors for the development of antibiotic bacterial resistance (Mathur & Singh 2005). Antibiotics as growth promoters have become responsible for the development of resistant bacterial strains in animals (Salisbury *et al.* 2002), and have been reported to affect soil bacterial community structure, and enzymatic and respiratory activities (Chen & Zhang 2013). Bacterial populations isolated from animal gut were found to be more resistant microorganisms (Sarmah *et al.* 2006). The other possible mechanism through which the microorganisms became resistant is the sharing of extrachromosomal antibiotic resistance plasmids (R-plasmids) (Sarmah *et al.* 2006). Resistance to antibiotics makes the treatment of infections extremely difficult and costly and in many cases results in high morbidity and mortality (Alanis 2005). Antibiotic resistance has contributed to 25,000–700,000 deaths per year (Martinez 2009). Antibiotics could contribute to the increase in allergies in humans (Zuccato *et al.* 2010). Due to potential toxicity effects and antibiotic resistance, these antibiotics in the environment possibly increased human health risks (Brown *et al.* 2006).

Toxic effect of waterborne antibiotics

The absence of adequate knowledge about the disposal of antibiotics and waste treatment paves the way for these chemicals to enter the environment especially the aquatic compartment. Toxicity studies have reported that antibiotics in water result in the development of resistance and are toxic to many microorganisms. Results of the experiments have shown that long-term exposure of microorganisms to antibiotics causes a toxic effect (Kümmerer *et al.* 2000). Studies on the toxic effect of antibiotics have clearly stated that antibiotics are potential genotoxic agents, which has been proved through animal and microbial assays (Cavas & Gozukara 2005). Various tests have been designed to assess the toxicity effects of antibiotics, such as the SOS chromotest on *E. coli* and the point mutation test (Ames test) on *Salmonella* species (Isidori *et al.* 2005). Higher plants also have been used as the model to test the genotoxicity of antibiotics through micronucleus formation, root tip chromosomal aberration assay and sister chromatid exchange (Liu *et al.* 2012). The toxic levels of antibiotics for microorganisms, bacteria and micro-algae existing in the environment are 2–3 times the magnitude below the toxic values for greater trophic levels (Wollenberger *et al.* 2000). In recent times, effects on aquatic organisms and plant species have been studied under controlled laboratory conditions (Bauger *et al.* 2000). Table 2 depicts the extensive work done on the toxicity effect of the antibiotics which may reach the aquatic compartment of the environment.

ANTIBIOTIC REMOVAL PROCESS: THE CHALLENGE

Conventional technologies such as filtration, coagulation/flocculation/sedimentation, oxidation, ion-exchange, activated sludge, oxidation process, carbon filtering, membrane distillation and reverse osmosis are used for the removal of pollutants. The conventional treatment plants are mostly not able to remove antibiotics efficiently (Al Aukidy *et al.* 2014). The high costs of the removal of antibiotics demands that more appropriate, convenient and cost-effective removal technologies of antibiotics should be found, for a safer environmental system, thereby reducing the harmful effect of the leftover antibiotics and their metabolites on aquatic organisms.

Adsorption technology: the future scenario

Adsorption is widely acknowledged as an efficient, effective and economical method for water contamination applications for the removal of several pollutants from aqueous solutions because of its easy operation, simple design, flexibility, suitability for batch and continuous processes, low capital cost, and possibility of regeneration and reuse (Amouzgar & Salamatinia 2015). The chances to reach high removal percentage of the pollutants are more than for other wastewater treatment methods. Until now, a variety of adsorbents have been developed; among available adsorbents, some low-cost adsorbents which have already been used include sawdust (Bajpai *et al.* 2012), biochars (Zheng *et al.* 2013), cellulose (Rathod *et al.* 2015), date palm leaflets (El-Shafey *et al.* 2012) and corn bracts (Yu *et al.* 2017). Some other adsorbents – agricultural wastes (Ahmed & Theydan 2013), lotus stalk (Liu *et al.* 2011) and rice husk (Chen *et al.* 2016) – are being used as low-cost alternatives to expensive adsorbents. Some of these adsorbents require a previous activation treatment (such as chemical or thermal activation) to increase their surface areas and consequently the adsorption efficiency. The various materials used for the removal of antibiotics are presented in Table 3.

Table 2 | Toxicity data of investigated antibiotics from literature

Compound	Species	Critical effect	Reference
Florfenicol	*Lemna minor* *Scenedesmus vacuolatus*	Growth inhibition	Kołodziejska *et al.* (2013)
Oxytetracycline	*Lemna minor* DT40 cell line *Labeo rohita*	Genotoxicity Mortality Growth inhibition	Liu *et al.* (2012), Ambili *et al.* (2013), Kołodziejska *et al.* (2013)
Tetracycline	*Eisenia fetifa*	Genotoxicity/oxidative stress	Dong *et al.* (2012)
Doxycycline	Human peripheral blood lymphocytes (HPBL)	Genotoxic/Cytotoxic effect	Sekeroglu *et al.* (2012)
Chlorotetracycline	*Eisenia fetifa* *Vibrio fischeri* *Pseudomonas putida* Human lymphocyte culture	Genotoxicity/oxidative stress Chronic ecotoxicology Chronic ecotoxicology Cytogenetic toxicity	Dong *et al.* (2012), Vasquez *et al.* (2013), Vasquez *et al.* (2013), Şekeroğlu *et al.* (2017)
Dirithromycin	Cultured human lymphocytes	Genotoxicity	Kayraldız *et al.* (2015)
Cefazolin	*Salmonella typhimurium*	Genotoxicity	Li *et al.* (2013)
Trimethoprim	*Mytilus edulis*	Genotoxicology	Lacaze *et al.* (2015)
Chloramphenicol	Human stem cell	Delayed cell growth	Turani *et al.* (2015)
Rifampicin	Human stem cell	Delayed cell growth	Turani *et al.* (2015)
Roxithromycin	HPBL	Cytogenetic stress	Arslan *et al.* (2017)
Ceftriaxone	HPBL	Cytogenetic stress	Metovic *et al.* (2013)
Fluoroquinolone	*Vicia faba*	Genotoxicity	Li & Zhang (2013)
Ciprofloxacin	Hep G2 cells *Phragmites australis* *E. coli* PQ37 *S. typhimurium*	Genotoxicity Root activity/leaf activity Genotoxicity Geno/cytotoxicity	Garcia-käufer *et al.* (2012), Liu *et al.* (2013), Toolaram *et al.* (2016), Bos *et al.* (2015)
Amoxicillin	*Vibrio fischeri*	Luminescence	Ji *et al.* (2013)
Sulfamethoxime	*Phragmites australis*	Root activity/leaf activity	Liu *et al.* (2013)
Erythromycin	Mice pups *Oncorhynchus mykiss* Mice	Genotoxicity Genotoxicity/oxidative stress Lipid metabolism	Singh *et al.* (2014), Rodrigues *et al.* (2016), Jin *et al.* (2016)
Penicillin	Mice	Lipid metabolism dysfunction	Jin *et al.* (2016)
Balofloxacin	*S. typhimurium*	Genotoxicity	Li *et al.* (2016)

ADSORPTION MECHANISM OF ANTIBIOTICS

The phenomenon of adsorption involves the following steps: (i) solute transportation in the bulk, (ii) film diffusion of adsorbate, (iii) pore diffusion of adsorbate, (iv) interaction between adsorbate and adsorbents. These interactions are strong and specific than physical adsorption (Ahmed *et al.* 2017). The main mechanism for the adsorption of antibiotics onto the carbon-based adsorbents is electrostatic interaction, hydrogen bonds, pore filling, hydrophobic effect and other processes (Tan *et al.* 2015).

The sulfamethoxazole interacted with the polarized aromatic rings on the surface of carbon nanotubes (CNTs) via the mechanism of π–π electron coupling (Ji *et al.* 2009). In another study, sulfamethoxazole sorption on biochar at pH between 1.0 and 6.0 was dominated by π–π electron-donor-acceptor (EDA) interaction of the protonated aniline ring with the π–electron-rich graphite surface, and

Table 3 | Removal of antibiotics by using various adsorbents

Antibiotics	Material used	Results	Reference
Norfloxacin	Lotus stalk/iron doped activated alumina (LAC)	- Average pore size for LAC was observed to be 3.41 nm, and for Al_2O_3/Fe was 5.33 nm. - At room temperature and at pH 6.5, maximum adsorption capacity of Al_2O_3/Fe of 21.58 μmol/g occurred at pH 6.5 and maximum adsorption capacity of LAC of 992.70 μmol/g occurred at pH 5.5. - Followed pseudo-second-order. - The point of zero charge value (i.e., pHpzc) of Al_2O_3/Fe and LAC was reported to be 6.53 and 5.48, respectively.	Liu *et al.* (2011)
Ciprofloxacin	Sawdust	- At adsorbent dosage 40 mg/20 mL, 64% of antibiotic was adsorbed, which remained constant even after increasing dosage of adsorbent. - Maximum adsorption 11.6 mg/g observed at pH 5.8. - Followed pseudo-second-order kinetic model. - Pore diameter of adsorbents was nearly 10 μm. - The pHpzc of the adsorbent was reported to be 5.4. - Intraparticle diffusion process involved in adsorption mechanism.	Bajpai *et al.* (2012)
Ciprofloxacin	Date palm leaflets	- Maximum adsorption capacity observed on dry adsorbent was 133.3 mg/g and on wet adsorbent 125 mg/g at temperature 45 °C. - Followed pseudo-second-order kinetics. - Maximum adsorption observed at pH 6.0. - Followed Langmuir isotherm model. - Adsorption endothermic and spontaneous. - The presence of hydroxyl, carboxyl and other carbon-oxygen species detected. - With increase in ionic strength adsorption capacity decreased.	El-shafey *et al.* (2012)
Gatifloxacin	Sludge biochar	- Antibiotic readily adsorbed onto sludge biochar. - Highest adsorption observed on biochar was 19.80 mg/g. - Followed Freundlich isotherm model.	Yao *et al.* (2013)
Gatifloxacin	Nanoparticles	- More than 90% of antibiotic removed within 15 minutes of adsorption process on g-BN (graphene-like layered hexagonal boron nitride), while only 10% was adsorbed on commercial BN adsorbent. - Adsorption pH independent. - Followed Langmuir isotherm model with adsorption capacity of 88.5 mg/g at 288 K. - Exothermic and spontaneous adsorption process. - Electrostatic and π–π interaction govern adsorption process between gatifloxacin and g-BN. - With the increase in ionic strength adsorption capacity decreased.	Chao *et al.* (2014)
Ciprofloxacin (CIP) Norfloxacin (NOR)	Activated carbon	- Activated adsorbent surface area was 1,824.88 m^2/g, micropore volume 0.645 cm^3/g and mesopore volume 0.137 respectively.	Ahmed & Theydan (2014)

(*continued*)

Table 3 | continued

Antibiotics	Material used	Results	Reference
		- At initial antibiotic concentration of 20 mg/L, contact time of 90 min and pH of 9.0, 96.12% of CIP was removed, and 98.13% of NOR was removed at pH of 5.0, 20 mg/L initial NOR concentration and 60 min contact time. - Followed pseudo-second-order kinetic model. - Ciprofloxacin adsorption was endothermic, norfloxacin exothermic. - Followed Langmuir isotherm model with maximum adsorption capacities of 131.14 and 166.99 mg/g for CIP and NOR, respectively. - Followed pseudo-second-order model for both antibiotics. - Equilibrium time observed for CIP was 90 min, and for NOR was 60 min. - The pH value of 8.7 was considered best for maximum removal of antibiotics as adsorption increased with increasing pH, reached a plateau when solution pH was approaching its p*K*a2 (8.7) and then decreased at higher pH.	
Ciprofloxacin (CIP)	Kandira stone	- At 12 g/L adsorbent dosage the removal percentage of CIP was 68.5%. - Equilibrium adsorption achieved at 30 min. - Removal of antibiotic decreased with the increase in pH. - Followed Freundlich isotherm model. - Followed pseudo-second-order. - Maximum removal observed at temperature 20 °C. - Exothermic adsorption.	Genc (2015)
Ciprofloxacin	Biochar (herbal residue)	- Best adsorption capacity observed on biochar produced at 800 °C was 37.6 ± 0.87 mg/g. - Maximum adsorption of ciprofloxacin was obtained at pH 7.0. - Followed Freundlich isotherm model. - Followed pseudo-second-order kinetics.	Shang *et al.* (2016)
Ciprofloxacin (CIP) Norfloxacin (NOR) Ofloxacin (OFL)	Graphene oxide	- Maximum removal observed at pH 7 for NOR and CIP. - Adsorption was maximum at pH 4 for OFL. - Followed pseudo-second-order model. - Followed Langmuir isotherm model. - Adsorption equilibrium obtained at 50 minutes.	Yadav *et al.* (2018)
Ciprofloxacin	Magnetic biochar	- Maximum adsorption capacity 68.9 ± 3.23 mg/g was observed at pH 6.0. - With the increase in adsorbent dosage, adsorption capacity decreased. - Followed pseudo-second-order kinetic model. - Followed Langmuir isotherm model.	Kong *et al.* (2017)
Levofloxacin	Corn bracts (CBs)	- Zr-modified CBs showed maximum adsorption capacity (Qmax = 73 mg/g).	Yu *et al.* (2017)

(*continued*)

Table 3 | continued

Antibiotics	Material used	Results	Reference
		- Maximum adsorption occurs at pH 7.0 - Followed pseudo-second-order kinetics and Freundlich isotherm model.	
Ciprofloxacin	Activated carbon	- The adsorbent had mesoporous structure, displayed surface area up to 1,435 m^2/g. - At 40 °C, maximum adsorption capacity of antibiotic was 335.8 mg/g. - Equilibrium data obeyed the Liu isotherm equation. - Followed Avrami kinetic model. - Adsorption spontaneous and endothermic.	De Oliveira *et al.* (2019)
Cephalexin	Mesoporous silica	- The pore diameter of MCM-41 was 2.0 nm, surface area was 1,097 m^2/g and mean crystallite size was 75 nm. - Fourier transform infrared (FTIR) analysis results revealed the H-O-H, Si-OH and Si-O-Si bonds are formed. - At initial pH 3.0, adsorbent dosage 800 mg/L, initial antibiotic concentration 50.0 mg/L, temperature of 40.0 °C and adsorption time period of 30.0 min, the removal percentage of antibiotic was 90.3%.	Panahi *et al.* (2019)
Ciprofloxacin	ICDW-APTES (inorganic sludge from construction and demolition wastes grafted with 3-aminopropyltriethoxysilane)	- The removal percentage of two synthetic effluents by ICDW-APTES was 71.0 and 69.0% respectively. - Liu kinetic model showed better fitness. - With the increase in temperature, adsorption capacity (Qmax value of 138.3 mg g^{-1}) of antibiotic also increased. - Mechanism involves electrostatic interaction.	Caicedo *et al.* (2020)
Tetracycline Chloramphenicol	Bamboo charcoal (BC)	- Adsorption capacity of chloramphenicol and tetracycline on BC increased with increasing bed height and decreased with increase in flow rate and influent concentration. - The adsorption mechanism involved π-π electro-donor–acceptor, cation-π bond and hydrogen bonding interactions. - Followed pseudo-first-order kinetic model. - Followed Langmuir isotherm and Dubinin–Radushkevich model.	Liao *et al.* (2013)
Tetracycline Ciprofloxacin	Graphene-soy protein (GS) aerogel	- Specific surface area of GS was 30.07 m^2/g with abundant microspores. - Adsorption capacity of GS was 500 mg/g, observed for both antibiotics, which was relatively higher than other adsorbents. - Followed both Langmuir and Freundlich isotherm model.	Zhuang *et al.* (2015)
Amoxicillin	Organobentonite	- The adsorption percentage on DK1, DK1N, DK2 and bentonite was 97.9, 61.5, 53.1 and 13.8% respectively (DK: bentonite modified with hexadecyl trimethyl ammonium).	Xing *et al.* (2013)

(*continued*)

Table 3 | continued

Antibiotics	Material used	Results	Reference
		- Followed second-order kinetics. - At pH 6 about 99% amoxicillin absorbed. - Followed both Langmuir and Freundlich isotherm model. - Adsorption mechanism involves both ion-exchange and partition.	
Amoxicillin (AMX)	NH_4Cl-induced activated carbon (NAC) Standard activated carbon (SAC)	- The prepared adsorbent had surface area of 1,029 m^2/g and a mean pore volume of 2.46 nm. - At pH 6.0, 50 mg/L initial antibiotic concentration and 0.4 g NAC/L adsorbent, over 99% amoxicillin was adsorbed, while at similar experimental conditions, SAC could only adsorb around 55%. - Increase in temperature increased adsorption. - Followed pseudo-second-order kinetics and Langmuir isotherm model. - Maximum adsorption capacity of AMX onto SAC and NAC was 262 and 437 mg/g, respectively.	Moussavi *et al.* (2013)
Tetracycline	Activated carbon	- At pH 7.0, maximum adsorption capacity on Cu-13X was 455.33 mg/g. - Followed pseudo-second-order kinetics and Langmuir isotherm model. - Adsorption depends on the strong complexation of Cu(II) with NH_2 radical of amide group of tetracycline.	Lv *et al.* (2015)
Amoxicillin, Tetracycline Penicillin G Cephalexin	Activated carbon (vine wood)	- The adsorbent had area and pore volume of 13.397 m^2/g and 54.79 cm^3/g, respectively. - At pH 2.0, equilibrium time of 8 h and adsorbent of 0.4 g/L, 20 mg/g of antibiotics was successfully removed. - Followed second-order reaction kinetics and Freundlich isotherm model.	Pouretedal & Sadegh (2014)
Penicillin G	Fe^{+3}-TiO_2/UV-A process	- Maximum removal rate of penicillin G occurred at pH 3.0. - With the increase in initial concentration of penicillin G, removal rate of antibiotic decreased. - Significant increase observed in the rate of antibiotic removal due to the effect of UV on catalyst activation in Fe^{+3}-TiO_2/UV-A process.	Dehghani *et al.* (2014)
Tetracycline	Activated carbon from macadamia nutshell (ACM)	- ACM composed of micropores (78.2%), surface area of 1,524 m^2/g and pHpzc value of 8.74. - The maximum adsorption capacity achieved was 455.33 mg/g. - Followed Elovich kinetic and Temkin isotherm model.	Martins *et al.* (2015)
Amoxicillin	Wheat grains	- At adsorbent particle size of 150 µg, temperature 25 °C, pH 7, antibiotic concentration of 4 mg/L, 5 min contact time, and 0.24 g/L adsorbent concentration, maximum adsorption was found to be 84.0%.	Boukhelkhal *et al.* (2015)

(*continued*)

Table 3 | continued

Antibiotics	Material used	Results	Reference
		- Followed second-order reaction kinetics and Temkin isotherm model.	
Tetracycline (TC)	Multi-walled carbon nanotubes (MWCNTs)	- Antibiotic adsorption was significantly decreased by Fe^{3+} ions onto the MWCNT. - Other cations and anions showed no effect on TC adsorption. - Maximum adsorption capacity of 253.38 mg/g on MWCNT was observed. - FTIR analysis of oxidized MWCNT confirmed oxygen-containing functional groups beads as a good choice for adsorption process. - Followed Avrami fractionary-order. - Followed Liu isotherm model with the maximum adsorption capacity of 253.38 mg/g. - Optimum pH for adsorption was 5–7 and equilibrium achieved in 120 minutes.	Babaei *et al.* (2016)
Tetracycline (TC)	Zinc chloride impregnated activated carbon (Zn-AC)	- Followed general order kinetic and Redlich–Peterson model. - The adsorption of TC onto the adsorbent reached equilibrium after 120 min. - The pH had no significant effect on TC adsorption. - Adsorbent specific surface area was 224 m^2/g. - Cations and anions significantly decreased TC adsorption onto the Zn-AC. - The amount of TC adsorbed onto the Zn-AC (Qmax) was 282.06 mg/g.	Takdastan *et al.* (2016)
Amoxicillin (AMX)	Magnetic activated carbon (MAC)	- The maximum sorption capacity of AMX obtained for MAC-1 and MAC-2, respectively, was 280.9 and 444.2 mg/g. - Followed general order kinetic and Liu isotherm model.	Saucier *et al.* (2017)
Oxytetracycline	NaOH-activated carbon (guava seeds)	- At 2,000 mg/L the initial adsorption rate was 100 mg/(g·h) - Followed pseudo-second-order. - Oxytetracycline adsorption was affected by presence of Cu^{2+}. - Adsorption mechanism involves surface complexation, cation-π interactions, cation exchange and metal bridging. - Maximum adsorption observed at pH 5.0.	Xiancai *et al.* (2017)
Chlortetracycline	Biochar (pine wood)	- Adsorption capacity of raw and activated biochar was 2.1 and 208.3 mg/g. - Maximum adsorption occurred at pH 1.0. - Followed Langmuir isotherm model. - The mean particle size of the raw biochar was 25.7 μm and that of activated biochar was 19.1 μm. - Optimum temperature selected for adsorption was 298 K.	Taheran *et al.* (2016)

(*continued*)

Table 3 | continued

Antibiotics	Material used	Results	Reference
Ciprofloxacin Doxycycline hydrochloride Tetracycline hydrochloride	Rice husk biochar	- Biochars produced at high temperature (i.e., 700 °C) showed maximum adsorption. - Maximum adsorption capacities of tetracycline hydrochloride, doxycycline hydrochloride and ciprofloxacin were 80.9, 85.2 and 36.1 mg/g, respectively. - Adsorption mechanism involves hydrogen bonding and π-π electron donor-acceptor interaction.	Zeng *et al.* (2018)
Amoxicillin (AMX)	Activated sludge (palm bark biomass) (PB)	- Under optimum conditions of time of 90 min, initial AMX concentration 10 mg/L, adsorbent dose 3 g/L, contact time 90 min and temperature 25 °C, 98.1% of antibiotic was removed. - Followed Langmuir isotherm model with adsorption capacity to be 35.92 mg/g. - PB biomass maximum surface area was 124.36 m^2/g and lowest particle size was 1.18 mm. - Cu ions enhance adsorption ability. - Adsorption process highly pH dependent.	Balarak *et al.* (2017a)
Amoxicillin (AMX)	Activated carbon (AC) (*Azolla filiculoides*)	- With the increase in adsorbent concentration from 0.15 to 0.60 g/L and at 100 mg/L initial AMX concentration, removal of AMX increased from 49% to 90%. - Followed pseudo-second-order kinetic and Langmuir isotherm model, with adsorption capacity 265.2 mg/g. - Pore volume and specific surface area of AC were found to be 0.47 cm^3/g and 484 m^2/g.	Balarak *et al.* (2017b)
Oxytetracycline (OTC)	Willow residue	- Adsorption of OTC onto raw willow residue reached equilibrium in about 12 hours and on desugared willow in 24 hours. - Adsorption followed pseudo-first-order model. - Followed both Freundlich and Langmuir isotherm models.	Wang *et al.* (2017)
Tetracycline	High surface area porous carbon material	- The maximum antibiotic adsorption observed at temperature 30, 40, and 50 °C was 128.52, 162.62, and 210.18 mg/g respectively. - Followed pseudo-first-order model.	Ahmed *et al.* (2017)
Tetracycline	Rice straw (biochar)	- The rice straw samples pyrolyzed at 500 °C and 700 °C (R700) showed high removal efficiency. - The maximum adsorption on R700 at 35 °C was between 92.8% and 96.7%. - Maximum adsorption occurred at pH 5.5.	Wang *et al.* (2017)
Tetracycline	Graphene oxide/calcium alginate (GO/CA)	- Maximum adsorption capacity of GO/CA composite fibers reached 131.6 mg/g. - Followed pseudo-first-order. - Followed both Freundlich and Langmuir isotherm models. - Maximum adsorption observed at pH 6.0.	Zhu *et al.* (2018)

(*continued*)

Table 3 | continued

Antibiotics	Material used	Results	Reference
		- Adsorption mechanism involves hydrogen bonding and π-π interaction.	
Amoxicillin Ibuprofen	Activated carbon (cocoa cob waste)	- At pH 6, the removal rate observed was 77.4%. - Followed pseudo-first-order and Elovich model.	Tovar *et al.* (2018)
Amoxicillin	Activated carbon	- Total pore volume and surface area of adsorbents were 1,457 m^2/g and 0.275 cm^3/g (capsules of Para cashew pyrolyzed at 600 °C; CCP.600) and 1,419 m^2/g and 0.285 cm^3/g capsules of Para cashew pyrolyzed at 700 °C; CCP.700). - At 45 °C maximum adsorption capacities of 451.0 (CCP.600) and 454.7 mg/g (CCP.700) were obtained for both adsorbents. - Followed Avrami fractional kinetic model and Liu isotherm model.	Lima *et al.* (2019)
Cefradine Cephalexin Ceftazidime Cefixime	Alga-activated sludge combined system	- Removal rate of antibiotics was cefradine (89.9%), cephalexin (94.9%), ceftazidime (89.7%) and cefixime (100%). - Individual activated sludge treatment removal rate was lower than 23.0%. - The acclimated activated sludge removed 46.3% antibiotic after a 2-week adaptation. - Combined system removal rate was 97.91%.	Guo & Chen (2015)
Cephalexin Cefradine	Activated carbon (herbal residue)	- About 84% of both antibiotics was removed from wastewater. - Maximum adsorption of cefradine and cephalexin by activated carbon was 7.1 mg/g. - Intraparticle diffusion observed as rate limiting factor. - Followed pseudo-second-order kinetic and Langmuir and Temkin isotherm models.	Li *et al.* (2017b)
Metronidazole	Nano-scale zero-valent iron particles	- Metronidazole completely removed within 90 min. - Removal efficiency increased with the decrease in pH. - Precipitation and co-precipitation were not the main processes for removing metronidazole. - Primary removal mechanism involves reduction process.	Chen *et al.* (2012)
Metronidazole	Agricultural wastes activated carbon	- Precursors have surface area 1,824.88 m^2/g and 0.782 cm^3/g total pore volume. - Maximum adsorption capacity of 196.31 mg/g reported at pH 7. - Followed pseudo-second-order and Langmuir isotherm model.	Ahmed & Theydan (2013)
Metronidazole	Rice husk	- Antibiotic removal increased with the increase in temperature. - Process endothermic and spontaneous. - At 3 g of biomass and temperature of 25 °C, 99% of antibiotic was removed.	Azarpira & Balarak (2016)

(*continued*)

Table 3 | continued

Antibiotics	Material used	Results	Reference
		- Higher adsorption percentages obtained within 90 minutes at lowest concentration. - Follows Langmuir isotherm models with maximum adsorption capacity obtained of 21.42 mg/g.	
Metronidazole	Canola stalk	- Maximum adsorption capacity 21.42 mg/g. - Maximum adsorption of antibiotic obtained within 90 minutes. - Followed both Langmuir and Temkin isotherm models.	Balarak & Mostafapour (2016)
Dimetridazole Metronidazole	Biochar	- The removal efficiencies of dimetridazole and metronidazole were 98.0% and 88.8% respectively, at biochar dosage of 4.0 g/L and temperature of 30 °C. - Followed Brunauer–Emmett–Teller isotherm model with maximum adsorption capacities 72.17 and 23.61 mg/g for dimetridazole and metronidazole, respectively. - Follows pseudo-second-order kinetics. - Adsorption mechanisms involves hydrogen bonding and π-π interaction.	Sun *et al.* (2018)
Linezolid	MgO nanoparticles ZnO–MgO nanocomposites	- At initial adsorbent concentration of 0.5 g/L, temperature 308 K and pH 10.0, MgO nanoparticles removed 123.45 mg/g and ZnO–MgO 140.28 m/g of antibiotic respectively. - Follows pseudo-second-order and Langmuir isotherm model.	Fakhri & Behrouz (2015)
Amoxicillin cephalexin Tetracycline Penicillin G	Carbon nanoparticles	- With the pH from 3 to 5, the biosorption capacity of TC on to nanocellulose increased from 1.94 to 4.51 mg/g. - Maximum biosorption occurred at pH 5. - Followed pseudo-second-order model.	Rathod *et al.* (2015)
Sulfadiazine (SDZ) Norfloxacin (NOR) Metronidazole (MDE) Tetracycline (TC)	Granular activated carbon (maize straw)	- Maximum sorption capacity followed the order SDZ (147.12) > NOR (112.86) > MDE (110.64) > TC (85.29) mg/g respectively. - Followed pseudo-second-order kinetic and Freundlich model.	Liu *et al.* (2017)
Tylosin	Porous resins	- pH dependent adsorption observed. - Showed different adsorption rate on four adsorbents. - Mechanism involved intermolecular interactions. - Followed pseudo-second-order kinetic and Langmuir isotherm model.	Lu *et al.* (2013)
Chloramphenicol	Activated carbon (AC)	- AC had surface area of 794.8 m^2/g. - The acidic and basic functional groups on AC were 2.078 and 0.995 mmol/g. - Adsorption mechanism involves π-π EDA interaction and hydrophobic interaction, in conjunction with hydrogen bonding interaction.	Li *et al.* (2016)

(*continued*)

Table 3 | continued

Antibiotics	Material used	Results	Reference
		- Followed pseudo-second-order and Freundlich isotherm model. - Maximum antibiotic adsorption capacity observed was 0.424 mmol/g. - pH and ionic strength had minimal effects on CAP adsorption.	
Chloramphenicol	Activated carbon	- Most efficiently adsorbed on F-300 (94.7%) from solution with pH 7.0 and on activated carbon ROW 08 supra (94.7%) from solution with pH 2.0. - Adsorption kinetics followed the pseudo-second-order kinetic model and Langmuir isotherm model.	Lach & Ociepa-Kubicka (2017)
Sulfamethoxazole	Biochar (rice straw)	- Cadmium presence promoted the sorption on rice straw biochar. - Maximum adsorption observed at pH 3.0.	Han *et al.* (2013)
Sulfamethoxazole	Biochars	- Followed Langmuir isotherm model with adsorption capacity on rice straw (RS) of 3,650 mg/kg and 1,963 mg/kg on alligator flag (AF). - Temperature had no effect on adsorption. - At pH greater then 7.0, RS showed larger adsorption of antibiotic than AF adsorbent. - Presence of ions increased the adsorption of antibiotic on both RS and AF. - Specific surface area and total pore volume of RS was 29.6 m^2/g and 0.069 cm^3/g and of AF was 7.1 m^2/g and 0.039 cm^3/g.	Li *et al.* (2015)
Sulfamethyldiazine (SM2) Sulfamethazine (SMT) Sulfathiazole (ST) Sulfamethoxazole (SMX)	Spent mushroom substrate (SMS)	- Adsorption capacity increased with the increase in initial antibiotics concentration. - The equilibrium time required for adsorption on SMS had a range of 150–180 min. - Adsorption capacity followed the order: ST 1.8103 (SM2), 2.1072 (SMT), 2.2133 (SMX), and 2.2991 (ST) mg/g. - Followed pseudo-first and second-order kinetic models. - Followed Langmuir isotherm model.	Zhou *et al.* (2016)
Sulfamethazine (SMT)	Biochar	- At pH 3.0 activated biochar showed highest sorption capacity, 37.7 mg/g, compared to non-activated biochar. - Followed Freundlich and Temkin isotherm model. - Mechanism involves electrostatic interaction between SMT and biochar.	Rajapaksha *et al.* (2015)
Sulfonamide	Reduced graphene oxide ferrite hybrids	- Highest extraction efficiency obtained at pH 6.0. - Extraction efficiency increased with the increase in time from 5 to 15 min.	Wu *et al.* (2016)
Sulfamethoxazole Tylosin	Triazine frameworks	- Adsorption coefficient for sulfamethoxazole decreased slightly between pH 3 and 5, and increased when the pH increased from 5 to 10 for tylosin.	Liu *et al.* (2017)

charge-assisted hydrogen bonds regulated anionic sulfamethoxazole species sorption above pH 7.0. (Zheng *et al.* 2013). Chloramphenicol adsorption was observed to be affected by the charge distribution in aromatic rings via substituent effects. The different hydrophobic or heterocyclic substituents might contribute to the adsorption affinity of sulfonamides (Zhao *et al.* 2016). It was reported that the adsorption of sulfamethazine on mesoporous cellulose biochar adsorbents involves an interfacial interaction mechanism. Electrostatic and π-π interactions both contributed to the high adsorption of sulfamethoxazole on Ni@CNFs (carboxylic-functionalized carbon nanofibers-encapsulated Ni magnetic nanoparticles) at pH 7.0 (Lan *et al.* 2016). The main reason for adsorption of antibiotics was observed to be π-π interactions in the adsorption process (Peng *et al.* 2016). Zhou *et al.* (2016), reported that π-electron interaction, not the electrostatic interactions, was the main interfacial interaction for the adsorption of sulfamethazine on mesoporous cellulose biochar. Ji *et al.* (2009) proposed π–π EDA interaction during adsorption between π electron acceptor compounds and the π electron-rich regions of the graphene surface of CNTs and graphite, and a similar mechanism was reported by Ji *et al.* (2010) for KOH activated CNTs for the adsorption of sulfamethoxazole, tetracycline and tyrosine. The high adsorption capacity of copper-based metal framework for sulfonamide antibiotic was attributed mainly to π-π stacking, hydrogen bonding and electrostatic interactions (Azhar *et al.* 2016). The adsorption of Cu(II) and ciprofloxacin onto nitrilotriacetic acid-functionalized magnetic graphene oxide (NDMGO) was enhanced through Cu(II) bridging effect (Li *et al.* 2017a) by hydrogen bonds, amidation reaction, and electrostatic and π-π interaction (Figure 2).

The adsorption of quinolone antibiotics by activated carbon observed at different pH showed that hydrophobic interaction, electrostatic interaction and π–π dispersion force were the main mechanisms of adsorption (Fu *et al.* 2017). The antibiotic ciprofloxacin was adsorbed by using magnetic graphene oxide/nitrilotriacetic acid nanocomposite.

The adsorption process observed was mainly determined by the π–π adsorbate–adsorbent dispersion interactions (Rivera-Utrilla *et al.* 2009). An adsorption experiment using metal-organic frameworks modified with urea or melamine for nitroimidazole antibiotics from the water was performed. Based on the results, a plausible adsorption mechanism was suggested for the improved performance: H-bonding between $-NH_2$ of the metal-organic frameworks and $-NO_2$ of the nitroimidazole antibiotics (Seo *et al.* 2017).

The adsorption of gatifloxacin was governed by π–π dispersion interaction between gatifloxacin and g-BN, and electrostatic interaction may also exist in the adsorption process (Chao *et al.* 2014). The adsorption rate of antibiotics diatrizoate, metronidazole and dimetridazole on activated carbons was governed by intraparticle diffusion, and surface diffusion was the main mechanism controlling the intraparticle diffusion (Flores-Cano *et al.* 2016). The

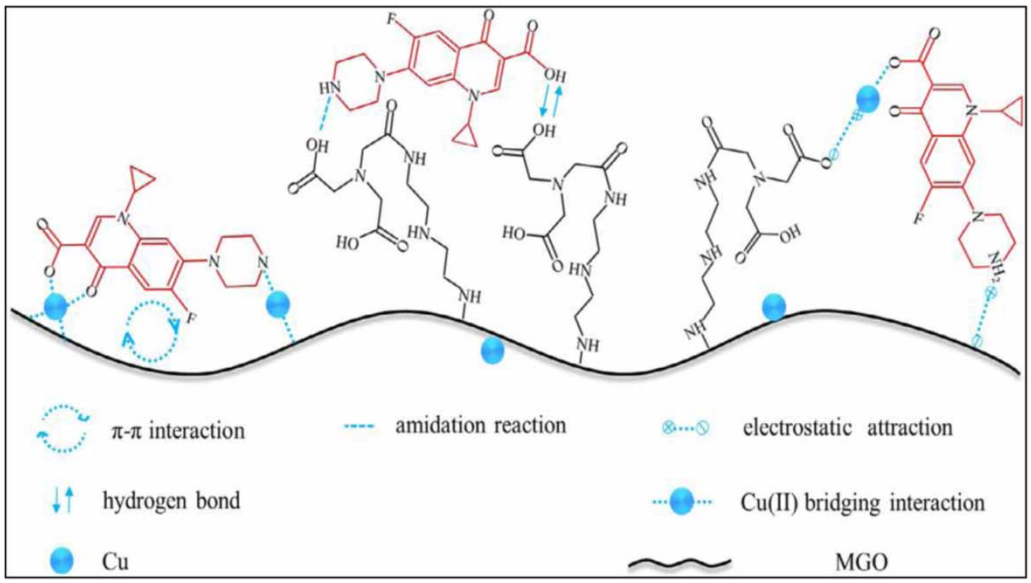

Figure 2 | Schematic illustration of ciprofloxacin adsorption mechanism by NDMGO (Li *et al.* 2017a).

EDA interaction, hydrophobic interaction and cation exchange were likely important mechanisms for the sorption on activated carbon prepared from lotus stalk, and complexation and cation bridging seem to be the main mechanisms for norfloxacin sorption on Al_2O_3/Fe (Liu *et al.* 2011). Bentonite has different mechanisms for adsorptive removal of contaminants. The dominant mechanism for adsorption of ciprofloxacin on bentonite was cation exchange (Putra *et al.* 2009). The adsorption mechanism of levofloxacin included the complexation between the ketone/carboxyl groups of levofloxacin and the zirconium atoms and the π-π EDA interaction (Yu *et al.* 2017). The adsorption mechanism of amoxicillin on organobantonite was proposed as being both partition and ion-exchange (Xing *et al.* 2013). Activated carbon modified with iron oxalate and cobalt presented a better method for adsorption of amoxicillin and paracetamol for removal from wastewater, due to positional entropy which in turn was the result of the special arrangement of the two pharmaceuticals on the adsorbent (Sellaoui *et al.* 2017).

Fluoroquinolone molecules were better suited for cation exchange than for cation bridging. The cation exchange mechanism occurred between the protonated heterocyclic (N atom) of positively charged ciprofloxacin and montmorillonite surface (Genc 2015). In the removal of ciprofloxacin from water, cation exchange was confirmed as the major mechanism for ciprofloxacin uptake by birnessite (Jiang *et al.* 2013). The uptake mechanism for ciprofloxacin hydrochloride was found to be attractive non-electrostatic interactions, involving H-bonding interactions between H atoms and other electronegative species (Bajpai *et al.* 2012). The adsorption mechanism in the case of ciprofloxacin adsorption was mainly related to cation exchange and hydrogen bonding (El-Shafey *et al.* 2012). The adsorption of ciprofloxacin on birnessite was studied under varying conditions. Experimental results showed that surface adsorption instead of cation exchange was responsible for the uptake of ciprofloxacin on birnessite. In a study, Kandira stone was used for removal of antibiotic ciprofloxacin hydrochloride from water. The adsorption behavior followed the pseudo-second-order kinetic model, indicating that the adsorption process can be expressed with the chemisorption mechanism (Genc 2015). The mechanism for the adsorption of tetracycline on graphene oxide involves cation–π bonding and π–π interaction (Gao *et al.* 2012). The adsorption mechanism of tetracycline on Fe_3O_4@SiO_2-chitosan/graphene oxide nanocomposite was investigated, by a pH impact study and XPS analysis. Besides electrostatic interaction and π–π interactions, the Cu(II) acts as a bridge between tetracycline and Fe_3O_4@SiO_2-chitosan/graphene oxide nanocomposite, which significantly improved the adsorption of tetracycline (Huang *et al.* 2017). The mechanism of sulfamethazine adsorption by mesoporous cellulose biochar at the electronic level in the V-shape configuration is mainly through π-π EDA interactions (Chen *et al.* 2019) (Figure 3).

CONCLUSION

The antibiotics' presence and fate have received special attention over the last two decades. These pollutants are persistent, hardly degraded and accumulate into the environment. For these reasons, several degradation/removal processes have been studied to solve environmental contamination issues. Various sources are contributing to the increasing antibiotics in surface water, groundwater, drinking water, seawater, plants, soil, and aquatic organisms. Both human and agricultural sources are responsible for discharging of antibiotics into the environment. The biological and physico-chemical properties of antibiotics have become a cause of concern, about their potential for the impact to non-target species. Antibiotics are responsible for acute and chronic toxic effects on humans and wildlife.

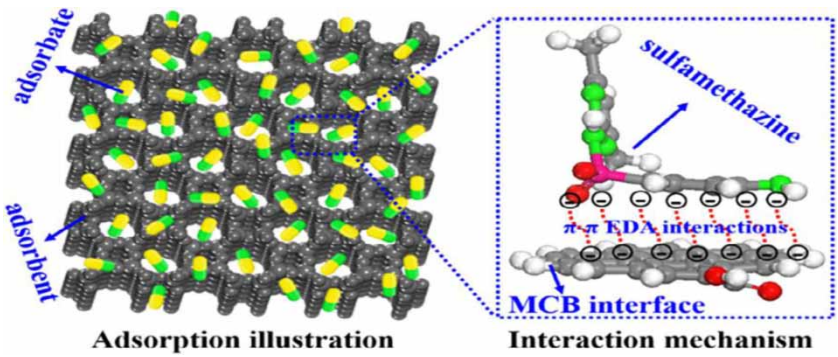

Figure 3 | Illustration of SMT adsorption onto mesoporous cellulose biochar (MCB) and interfacial interaction mechanism (Chen *et al.* 2019).

Several effects on growth, body weight and survival of microflora and microfauna, effect on primary productivity of an aquatic ecosystem, allergic reactions, potential development of antibiotic-resistant bacteria and toxic effects have been studied.

Most conventional treatments applied in WWTPs and drinking water treatment plants (such as coagulation, flocculation, sedimentation and filtration) are unsuccessful in the removal of these compounds. Ozonation has the advantage of being applicable in fluctuating flow rates and compositions but the high cost of equipment, the production of metabolites that are more toxic than the parent compounds and the energy required to supply the process constitute major drawbacks. Microfiltration or ultrafiltration (UF) membranes are utilized for a high-quality final effluent. Passing the wastewater through these types membranes ensures efficient elimination of suspended contaminants, but they are generally not able to remove pharmaceuticals pollutants. The majority of pharmaceuticals were not rejected when passing through a UF system. Although the technical feasibility of membranes has been proven, high investment and operational costs keep their application very limited. Currently, membrane filtration, nanofiltration and reverse osmosis are almost exclusively applied in drinking water treatment facilities, whereas their application during wastewater treatment is scarce. The main disadvantage of electrochemical oxidation is its high operating costs. Fenton's oxidation is one of the most studied advanced oxidation processes. When applied as a homogeneous process, the production of oxyhydroxides precipitates and the necessity to recover the dissolved catalyst constitute disadvantages. The combined methods are not a very common practice; these are one of the most powerful processes for antibiotics removal from the environment, reducing drastically the toxicity of treated effluents. An advanced oxidation process followed by biological treatment or by a membrane or even by an adsorption process is the most usual combined process. These methods are not usually used due to their complexity, high operating costs and most of the time due to their impracticability of being used in a continuous mode.

Adsorption is widely recognized as an effective, efficient and economic method for water contamination applications, and for separation, analytical purposes have been reported as an alternative to oxidation techniques, though not widely applied to the more prescribed antibiotics. The most common adsorbents in wastewater treatment are activated carbon, clays, zeolite and agricultural wastes. This technique has the advantage of removing the analytes instead of producing potentially more dangerous metabolites. The possibility to reach high removal percentage of the contaminant is more than with other water treatment techniques. Most of the studies used activated carbon, a high cost adsorbent material. Despite the effectiveness of activated carbon, there are major drawbacks of its application. Since the activation stage requires very high temperature, a lot of energy is required to provide sufficient heat for such high temperature. As a result, the production cost will be very high. Furthermore, the production of activated carbon is also not environmentally friendly as activated carbon dust might be emitted to the environment during production and the chemicals used will contaminate the water source and affect the aquatic ecosystem. From the author's point of view, the combined treatment process including cheap adsorption material would provide the best result for the treatment of effluents containing antibiotics.

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