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Review

Adsorption of emerging contaminants from water and wastewater by modified biochar: A review[☆]Ning Cheng^a, Bing Wang^{a, b, c, *}, Pan Wu^{a, b, c}, Xinqing Lee^d, Ying Xing^e, Miao Chen^{a, b, c}, Bin Gao^f^a College of Resources and Environment Engineering, Guizhou University, Guiyang, Guizhou, 550025, China^b Key Laboratory of Karst Georesources and Environment, Ministry of Education, Guiyang, Guizhou, 550025, China^c Guizhou Karst Environmental Ecosystems Observation and Research Station, Ministry of Education, Guiyang, Guizhou, 550025, China^d State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550081, China^e School of Chemistry and Materials Science, Guizhou Normal University, Guiyang, 550001, China^f Department of Agricultural and Biological Engineering, University of Florida, Gainesville, FL, 32611, USA

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ABSTRACT

Emerging contaminants (ECs), a group of relatively low-concentration but high-toxicity pollutants in the environment, have attracted widespread attention in recent years. These trace pollutants can be enriched in organisms and finally transferred to human bodies, posing a potential hazard to public health. Biochar, a low-cost and high-efficiency adsorbent, has been used to treat ECs in water. However, due to certain limitations of pristine biochar, such as poor adsorption capacity, narrow adsorption range, and other shortcomings, it is necessary to modify biochar to improve its applications in water treatment for ECs. Currently, there are a lot of reports on the removal of ECs from water by modified biochar. These studies explored different modification methods to functionalize biochar with various physicochemical properties, which resulted in distinct adsorption effects, behaviors and mechanisms of modified biochar on different ECs. There is a need to systematically review and digest the knowledge on the adsorption of ECs on modified biochar. In this review, recent biochar modification methods used in ECs removal are firstly summarized, and the adsorption performance and mechanisms of modified biochar on typical ECs are then systematically reviewed. Finally, the main research directions and trends, as well as recommendations and suggestions for future development are pointed out.

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1. Introduction

With the continuous increase of humans' needs for the environment, many pollutants with low contents but great harm in the environment are gradually attracting attention, such as antibiotics, endocrine disruptors, etc. They are called emerging contaminants (ECs). ECs are a group of chemical pollutants that have potential threats to the human health and ecological environment. They are very complex organic matters, generally exist in water. ECs usually come from pharmaceuticals, personal care products, endocrine-disrupting chemicals, antibiotics, persistent organic pollutants,

disinfection by-products, and other industrial chemicals (Bo et al., 2015). These ECs persistent in the environment and last for a long time. Previous studies have found more than thirty kinds of ECs in untreated wastewater, treated wastewater, urban rainwater, agricultural rainwater, and freshwater. Among them, artificial sweeteners, pharmaceuticals, and personal care products were detected in various water samples (Tran et al., 2019). ECs are constantly circulating, migrating and transforming in environmental media. Although the concentrations of these ECs are relatively low in water, they may have potential impacts on the environment and human health through the food chain after being accumulated by organisms (Gomes et al., 2017). Therefore, how to effectively remove ECs from water has attracted widespread attention.

Table 1 summarizes the types, impacts and general treatment methods of some typical aqueous ECs. At present, the methods commonly used to remove ECs from water mainly include microbial method (Ferreira et al., 2016), electrochemical method (Barrios

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Table 1
Types, impacts, and treatment methods of typical ECs.

Types	Impacts	Treatment methods	References
Antibiotics	Antibiotics can cause antibiotic resistance in other people, animals, and the environment, making antibiotics used by one patient ineffective for many other patients	Ozonation, Fungal treatment, Activated carbons, Biochar	(Ahmed, 2017; Carlet and Waaar, 2015; Greiner et al., 2018; Lucas et al., 2016; Xu et al., 2010)
Endocrine-disrupting chemicals	EDC may interfere with other endocrine axes, including the testes, adrenal glands, and thyroid hormone systems. These chemicals can have harmful effects on reproductive tract development and function	Electrocoagulation, Ozonation, Photodegradation, Activated carbon, Biochar	(Acerini and Hughes, 2006; de Leon-Condés et al., 2017; Govindaraj et al., 2012; Jeirani et al., 2017; Kim et al., 2016; Zhang and Zhou, 2008)
Caffeine	The adverse effects of caffeine on neurophysiological health include anxiety and panic attacks, as well as hallucinations caused by caffeine above a medium dose. Besides, caffeine may also impair learning and memory	Oxidative, Photo-Fenton, Activated carbon, Biochar	(Lins et al., 2019; Persad, 2011; Sotelo et al., 2012; Souza and Feris, 2015; Trovo et al., 2013)
Polychlorinated biphenyls	PCBs are easily absorbed, distributed and accumulated in the body, and can cause genotoxic effects	Aerobic degradation, Phytoremediation	(Lauby-Secretan et al., 2013; Pieper, 2005; Van Aken et al., 2010)

et al., 2016), adsorption method, membrane process, and chemical oxidation process (Acero et al., 2015). Among them, adsorption is widely accepted because of its advantages such as low cost, high efficiency, and wide processing range. Commonly used adsorbents include activated carbon (Esmaeeli et al., 2017; Leite et al., 2018; Wong et al., 2018), silica gel (Qu et al., 2018; Wu et al., 2019c), alumina (Zhao et al., 2016), polyacrylamide (Peng et al., 2019), absorbent resin (Wang et al., 2019a), and zeolite (Jiang et al., 2020). Because each adsorbent has different physicochemical properties, it shows different adsorption properties and mechanisms. Although activated carbon has the advantages of large specific surface area (SSA) and strong adsorption capacity, its production cost is relatively high, while other adsorbents like silica gel, alumina, adsorption resin, etc. Have the disadvantage of narrow adsorption surface and polyacrylamide is difficult to be produced. Therefore, it is urgent to seek low-cost, high efficiency and environmentally friendly adsorbents to remove ECs.

As a novel adsorbent, biochar has been widely used in the removal of organic and inorganic pollutants in the environment (Oliveira et al., 2017; Wang et al., 2017). It was found that the persistent free radicals in biochar could activate $S_2O_8^{2-}$ or H_2O_2 to generate active oxygen, thereby effectively degrading organic and inorganic pollutants (Ruan et al., 2019; Yang et al., 2016). In recent years, there have been increasing studies on the adsorption of ECs with biochar. For instance, antibiotics, as one group of the ECs, have gained increasing attention (Peiris et al., 2017). Data can be found by searching for the keywords “biochar” and “antibiotic” in the Web of Science (Fig. 1). There were only eleven literature published in 2015, while ten times more literature were published in 2019 than 2015. This rapid growth trend indicates that more and more people are paying attention to the research of using biochar to remove ECs. This review is based on more than one hundred papers published, with keywords such as modified biochar, antibiotics, tetracycline (TC), sulfonamide and bisphenol A (BPA). Previous studies have systematically reviewed various treatment technologies for ECs (Rodriguez-Narvaez et al., 2017). However, compared with activated carbon, the newly prepared biochar has limited adsorption capacity (Wang et al., 2017). Previous studies compared the adsorption capacity between activated carbon and biochar on the removal of phenol from water and found that the adsorption rate of activated carbon is $20.575 \text{ g mg}^{-1} \text{ min}^{-1}$, while that of biochar was only $0.040 \text{ g mg}^{-1} \text{ min}^{-1}$ (Hao et al., 2018). Therefore, the modification of biochar to give it different physicochemical properties and adsorption capacities has become an inevitable trend (Wang et al., 2017).

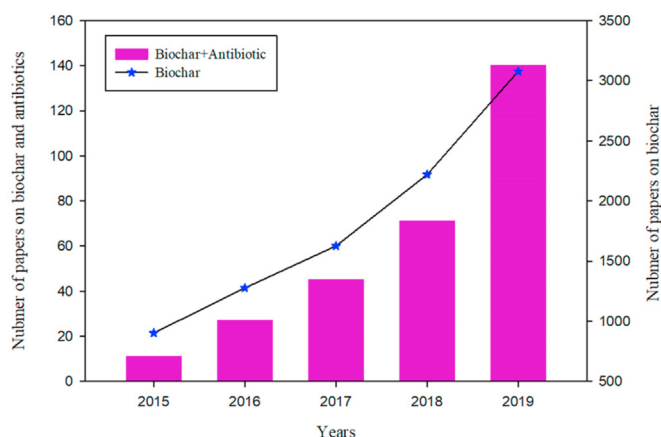


Fig. 1. Number of recent research literatures on biochar adsorption of ECs (Data from Web of Science).

At present, there are many studies on the adsorption of different pollutants by modified biochar. Different modification methods are used to screen efficient biochar to treat ECs, such as antibiotics and endocrine disruptors (Sophia and Lima, 2018). A large number of preparation methods and adsorption behaviors and mechanisms of different modified biochar for different ECs have been carried out (Tan et al., 2016). However, due to the different physicochemical properties of different modified biochars, the adsorption effects and mechanisms of different pollutants are quite different. Although numerous studies have been conducted, the results are inconsistent. Especially, the studies of some ECs still in their infancy which have not been attracted enough attention, and the explanation of the adsorption mechanism is not clear (Lins et al., 2019; Wu et al., 2018). So it is necessary to systematically review the current research status. Based on the above reasons, this paper systematically summarizes the recent progress on the adsorption of ECs by modified biochars from several aspects, such as their modification methods, the adsorption capacity, and mechanisms, etc. The review also puts forward suggestions and recommendations for future research. Prospects are finally provided as a reference for subsequent research work.

2. Biochar modification for ECs

Different modification methods may have different effects on

the physicochemical characteristics of biochar. Modification of biochar can not only affect its SSA and surface functional groups, but also alter the structures and size distributions of the pores. Differences in the treatment process and the modifier used can directly affect the adsorption performance and mechanism of biochar to pollutants. Therefore, it is important to select the appropriate biochar modification methods for the adsorption of ECs according to their physicochemical characteristics. At present, the modification methods commonly used to improve the adsorption capacity of biochar to ECs include acid-base modification, metal oxide and metal salt modification, clay mineral modification, and ball milling modification (Fig. S1).

2.1. Acid-base modification

The use of acid or alkali modification is the most commonly used method to change the surface characteristics of the adsorbent, mainly by increasing the SSA and pore structure of the biochars, which can affect the physical adsorption of ECs. Besides, the C—OH and C—H functional groups formed by acid-base modification also play a major role in the chemical adsorption process, thus change the adsorption capacity of biochars (Hu et al., 2018). For example, phosphoric acid pretreatment is a method of biochar modification that treats biomass with H_3PO_4 before pyrolysis. Due to acid catalysis and cross-linking, the micropores of the pretreated biochar particles are much larger than those without H_3PO_4 pretreatment, resulting in a significant increase in the SSA of the pretreated biochar (Chu et al., 2018). It was found that the SSA of the biochar modified by acid and alkali had been increased by 10 times and 14 times, respectively. The equilibrium adsorption capacity of enrofloxacin on acid and alkali modified biochar increased by 27.80% and 54.08%, respectively (Wang et al., 2019b). The acid-base modified biochar has been used to remove a variety of ECs, such as TC (Huang et al., 2017; Jang and Kan, 2019), ciprofloxacin (CIP) (Li et al., 2018), BPA (Zhao et al., 2017) and so on.

2.2. Metal oxide and metal salt modification

Metal oxide and metal salt modification can improve the adsorption efficiency, and make biochar have different adsorption capacities for pollutants according to different metal characteristics, mainly including metal salts and metal oxides. The biochar after metal oxide and metal salt modification has good electrostatic attraction ability, precipitation, and anion exchange capacity, thereby improving its adsorption capacity for ECs (Wu et al., 2019b). Studies show that the allocation of sulfurized nanoscale zero-valent iron (nZVI) to phosphorus-functionalized biochar (pBC-S-nZVI) could enhance its ability to remove florfenicol. The removal rate of pBC-S-nZVI to florfenicol is 4.3 times that of without modification (Xu et al., 2019). Other studies showed that the adsorption process of levofloxacin on Fe/Mn-biochar was highly pH-dependent, and Fe/Mn-biochar could maintain a certain levofloxacin adsorption capacity after 5 cycles (Xiang et al., 2019). Biochar-supported Ag/Fe nanoparticles (Ag/Fe/MB) could remove cephalixin from aqueous solution. It was found that the removal rate of cephalixin was over 86% in 90 min through adsorption and reduction (Wu et al., 2017). Cu-biochar also had an adsorption capacity of 93.22% for doxycycline hydrochloride in aqueous solution, which was twice as high as that of unmodified biochar. Comprehensive consideration of cost, efficiency and its application in real water, Cu-biochar had great potential to enhance the efficiency of removing doxycycline hydrochloride from water (Liu et al., 2017). In another study, Fe—Cu layered double hydroxide Fe—Cu-LDH/biochar nanocomposite was successfully synthesized by hydrothermal method. The biocatalytic efficiency for degrading cefazolin sodium

(CFZ) from water reached 97.6% (Gholami et al., 2020a). In general, metal oxide and metal salt modification enhance the chemical adsorption capacity of modified biochar by giving it more different functional groups to produce more chemical bonds.

2.3. Clay mineral modification

The research of biochar-clay composite (CM-BC) has gradually become more extensive in recent years. There are basically two preparation methods, one is to mix the raw materials with clay minerals for pyrolysis, and the other is to mix the pyrolyzed biochar with clay minerals. After modification, clay minerals can not only increase the porosity of CM-BC, but also improve its interface compatibility with ECs, thereby enhancing the adsorption capacity of CM-BC to ECs (Fu et al., 2020). The mixture of wheat straw and montmorillonite was heated at 400 °C to prepare montmorillonite-biochar composite (MT-BC), and the effects of pH, dissolved humic acids and Cu^{2+} on norfloxacin adsorption with MT-BC were studied. Compared with the pristine biochar, MT-BC had higher adsorption capacity for norfloxacin. The adsorption capacity of TC increased by 2.41 times (Zhang et al., 2018b). Corn cob biochar and montmorillonite composites were used for single and co-adsorption of Pb^{2+} and atenolol. The maximum equilibrium adsorption capacities for Pb^{2+} and atenolol in a single adsorption system were 139.78 and 86.86 mg/g, respectively (Fu et al., 2020). This composite is more functional and generally plays a different role in the treatment of different ECs (Ashiq et al., 2019).

2.4. Ball milling

Ball milling is a method of grinding samples including biochar to nanometer size (Wang et al., 2017). Ball milling can increase the SSA, surface functional groups and pore structure of biochar. The increase of SSA can increase the physical adsorption capacity of biochar to ECs. Meanwhile, the use of grinding media in the grinding process can make biochar have new functional groups to improve the chemical adsorption capacity of biochar to ECs (Qin et al., 2019). The weight ratio of spheres of yttria stabilized zirconia to biochar and the quality of solvents used in wet milling directly affect the physicochemical properties of biochar after ball milling (Peterson et al., 2012). By measuring Boehm titration, the results show that ball milling of biochar to agate balls increases the oxygen-containing functional groups (e.g., carboxyl, lactone, and hydroxyl) of biochar (Lyu et al., 2018). Singh et al. found that by choosing different grinding media (de-ionized water, ethanol, isopropanol and dimethylformamide), the structure and optical parameters can be customized (Singh et al., 2019). Even the shape of the grinding media could cause the raw material to produce different effects (Simba and Moys, 2014). Some studies prepared ultrafine magnetic Fe_3O_4 -biochar by ball milling to absorb ECs. After grinding for 2 h, the prepared hybrid adsorbent showed a high removal rate of carbamazepine (CBZ) and easy magnetic separation (Shan et al., 2016). Similarly, another research also proved that the nano-biochar produced by the green method was expected to remove trace pollutants by removing up to 95% of carbamazepine from water (Naghdi et al., 2017). Other studies also used ball-milled biochar to remove two sulfonamide antibiotics (SAs), sulfamethoxazole (SMX) and sulfa pyridine (SPY) from wastewater (Huang et al., 2019). Currently, although ball milling technology is a green modification method, few experiments have been carried out on the adsorption of ECs by ball-milled biochar. Ball-milled biochar may become an important research direction and hotspot in the future because of its green and low cost.

3. Adsorption mechanisms of modified biochar to ECs

ECs mainly contain antibiotics and environmental hormones. Antibiotics, as a major kind of ECs, have attracted wide attention. There have been many reports on the use of biochar to treat antibiotics in water. The effect of biochar on the environmental risk of antibiotic resistance genes during anaerobic digestion of cow dung wastewater had been studied. Adsorption performance of different modified biochar on antibiotics is shown in Table 2. It was showed that biochar significantly reduced the overall relative abundance of antibiotic resistance genes indigestion products (Sun et al., 2018b). At present, many studies have been conducted on the adsorption of single antibiotic pollution by biochar (Ben et al., 2019). However, the real water generally contains multiple antibiotics. Some studies

began to use modified biochar to remove several types of antibiotics from water simultaneously. Luo et al. studied the competitive adsorption mechanisms of three antibiotics (norfloxacin, sulfadiazine, and oxytetracycline) in water by modified biochar, and found that modified biochar significantly increased the adsorption of norfloxacin. The adsorption rate of oxytetracycline also increased slightly, while the adsorption rate of sulfadiazine decreased, mainly due to the effects of cation bridging and surface complexation (Luo et al., 2018). There are also some studies on the adsorption performance of modified biochar and the effect of functional groups. Modified biochar usually has multiple functional groups, such as -COOH, alcohol/phenol-OH, and -CHO, which shows excellent performance in removing toxic metals or organic pollutants (Wu et al., 2019a).

Table 2
Adsorption performance of modified biochar to antibiotics.

Raw material	ECs	Modification method	pH	Pyrolysis temperature (°C)	Adsorption capacity (mg/g)	Removal rate (%)	References
Rice husk	TC	MeOH	2.0	—	18.53	—	Jing et al. (2014)
Rice straw	TC	Alkali-acid combined, magnetization	9.0	300	98.33	—	Dai et al. (2020)
Coffee grounds	TC	Magnetite	2.0	250	—	96	Nguyen et al. (2019)
Municipal sewage sludge	TC	Alkali-acid combined method	7.0	800	286.913	—	Tang et al. (2018)
Aerobic granular sludge	TC	ZnCl ₂	7.0	700	93.44	—	Yan et al. (2019)
Poplar	TC	KOH	3.3	300	21.17	—	Huang et al. (2017)
Yak dung	TC	—	10.52	700	—	77.78–91.14	Wu et al. (2019a)
Municipal sludge	TC	Alkali-acid binding method	7.0	800	—	86	Zhou et al. (2019b)
Swine manure	TC	H ₃ PO ₄	9.0	700	365.4	—	Chen et al. (2018)
Wasted sludge	TC	Chitosan and Fe/S (BCFe/S)	9.0	750	183.01	—	Liu et al. (2019)
Pinus taeda	TC	NaOH	6.0	—	274.8	—	Jang et al. (2018)
Coconut, pine nuts and walnut shells	TC	Fe ₃ O ₄	6.0	500	94.2	—	Shan et al. (2016)
Coconut, pine nuts and walnut shells	CBZ	Fe ₃ O ₄	6.0	500	62.7	—	Shan et al. (2016)
Alfalfa hays	TC	NaOH	10.0	300	302.37	—	Jang and Kan (2019)
Wheat straw	TC	Ball milling	7.26	700	268.3	—	Li et al. (2020)
Bamboo	TC	Steam-activated	5.0	500	—	95.75	Wang et al. (2020b)
Date palm waste	CTC	—	5.0	600	28.19	—	Ahmad et al. (2019)
Date palm waste	CTC	Zeolite	5.0	600	30.42	—	Ahmad et al. (2019)
Date palm waste	CTC	Silica	5.0	600	45.57	—	Ahmad et al. (2019)
Date palm waste	CTC	Nano-zerovalent iron	5.0	600	89.05	—	Ahmad et al. (2019)
Shredded cotton stalks	SDM	H ₂ O ₂	9.0	350	—	71	Sun et al. (2018a)
Shredded cotton stalks	SMT	H ₂ O ₂	9.0	350	—	74	Sun et al. (2018a)
Shredded cotton stalks	NSMX	H ₂ O ₂	9.0	350	—	68	Sun et al. (2018a)
Shredded cotton stalks	SMX	H ₂ O ₂	9.0	350	—	49	Sun et al. (2018a)
Shredded cotton stalks	SDZ	H ₂ O ₂	9.0	350	—	39	Sun et al. (2018a)
Reed straw	SMX	Zn–TiO ₂	5.03	500	—	81.21	Xie et al. (2019)
Thalia dealbata	SMX	MgCl ₂	7.0	500	—	86.1	Tao et al. (2019b)
Bamboo	SPY	Ball milling	6.0	450	—	89.6	Huang et al. (2019)
Hickory chips	SMX	Ball milling	6.0	450	—	83.3	Huang et al. (2019)
Pristine hickory	SPY	Carbon nanotube	—	600	—	86	Inyang et al. (2015)
Bagasse	SPY	Carbon nanotube	—	600	—	56	Inyang et al. (2015)
Bagasse	SMX	Anaerobic digestion	2.6	600	54.38	—	Yao et al. (2018)
Bagasse	SPY	Anaerobic digestion	9.0	600	8.6	—	Yao et al. (2018)
Bagasse	SMX	Fe ³⁺	5.0	800	187.31	—	Zhang et al. (2020)
Hickory chips	SMX	Ball milling	—	450	—	83.3	Huang et al. (2020)

Table 2 (continued)

Raw material	ECs	Modification method	pH	Pyrolysis temperature (°C)	Adsorption capacity (mg/g)	Removal rate (%)	References
Bamboo	SPY	Ball milling	–	450	–	89.6	Huang et al. (2020)
Cassava	NOR	KOH	10.6	500	–	86.8–96.5	Luo et al. (2018)
Cassava	OTC	KOH	10.6	500	–	65.5–96.2	Luo et al. (2018)
Cassava	SMR	KOH	10.6	500	–	12.2–27.5	Luo et al. (2018)
Wood-processing residues	TYL	Nano-hydroxyapatite	6.0	700	118.835	–	Li et al. (2019b)
Wood-processing residues	SMX	–	6.0	700	149.254	–	Li et al. (2019b)
Palm	TYL	Goethite	7.0	600	8132.89L/kg	–	Guo et al. (2016)
Straw	NOR	Amontmorillonite	7.0	400	25.53	–	Zhang et al. (2018b)
Corn stalks	OTC	MnO ₂	4.0	350	39.92	–	Gao et al. (2018)
Potato stems and leaves	CIP	KOH	–	500	23.36	–	Li et al. (2018)
Peanut shells	DOX	CuNO ₃	8.0	450	–	93.22	Liu et al. (2017)
Camphor leaf	CIP	ZnO	4.0	650	449.40	–	Hu et al. (2019)
Astragalus residue	CIP	Fe ³⁺ /Fe ²⁺	6.0	700	68.9	–	Kong et al. (2017)
Vinasse	CIP	Fe/Mn	5.0	800	133	–	Xiang et al. (2020)
Vinasse	PEF	Fe/Mn	5.0	800	253	–	Xiang et al. (2020)
Grapefruit	CIP	Chitosan	3.0	450	36.72	–	Afzal et al. (2018)
Vinasse wastes	LEV	Fe/Mn	5.0	800	212	–	Xiang et al. (2019)
Sawdust	Cefotaxime	Co/Fe	5.0	500	–	99.23	Wu et al. (2018)

TC: Tetracycline; CBZ: Carbamazepine; TYL: Tylosin; CTC: Chlortetracycline; SMX: Sulfamethoxazole; SDM: Sulfadimethoxine; SMT: Sulfamethazine; NSMX: N₄-acetyl-sulfamethoxazole; CIP: Ciprofloxacin; DOX: Doxycycline; LEV: Levofloxacin; OTC: Oxytetracycline; NOR: Norfloxacin.

Environmental hormones are another type of ECs that has attracted much attention in recent years. The main endocrine disruptors include estrogen, BPA, etc. Phenolic organic compounds are harmful to the environment and affect the growth and development of animals and plants. The most studied phenolic compound is BPA. It is a suspected endocrine-disrupting compound and a phenolic chemical. It is widely used in the manufacture of paints and polycarbonate plastic. The widespread demand and use of BPA in important industries and commercial products have led to a large number of continuous discharges into the environment (Liu et al., 2009). BPA is frequently detected in a variety of aquatic environments, including surface water, groundwater, and even drinking water. The main concern about the widespread distribution of BPA in the water system is that it may limit or completely inhibit gene expression by mimicking the activity of natural hormones or hormone receptors in the body (Jung et al., 2019). For example, BPA can affect the quality and maturity of oocytes, decrease and quality of sperm production, damage to testicular cells in animals, disturbance of hormone levels, and damage to ovarian function and uterine shape (Siracusa et al., 2018). Besides, bisphenol S is another type of phenolic compound that may damage the human digestive and genital systems (Fang et al., 2020). The results of adsorption experiments showed that both biochar and dissolved organic matter could improve the adsorption capacity of bisphenol S in Fe–Al (Zhang et al., 2019b). At present, people have characterized the general heating and hydrothermal biochar, and studied the difference in adsorption characteristics between the two biochars through the adsorption of BPA, 17 α -ethynyl estradiol (EE2) and phenanthrene. The results showed that hydrothermal biochar could adsorb a wider spectrum of polar and non-polar organic pollutants (Sun et al., 2011). Some people studied the adsorption mechanisms of phenolic endocrine disruptors by functionalized biochar. EDC adsorption was highly pH-dependent with maximum adsorption occurring at pH 3.0–3.5. In terms of the adsorption mechanisms, EDC adsorption mainly occurred through the π - π bond and the formation of different hydrogen bonds (Ahmed et al., 2018). Adsorption performance of different modified biochar on EDCs is shown in Table 3.

The adsorption of ECs onto modified biochar is closely related to its surface properties, including SSA, surface charge, functional groups, pore volume and distribution (Rajapaksha et al., 2016;

Zhang et al., 2018a). The chemical modification of biochar can enhance its adsorption capacity by increasing the generation of more and abundant adsorption sites, so that the surface of biochar is more conducive to electrostatic attraction, surface complexation, and surface precipitation (Rajapaksha et al., 2016). Physical modification can affect the micropore structure and SSA of biochar, thereby affecting its adsorption mechanisms such as pore filling and intra-particle diffusion. The adsorption of modified biochar is affected by many influencing factors (Fig. 2). From most studies, the adsorption kinetic model of modified biochar for ECs is more in line with the pseudo-second-order kinetic model, and some adsorption processes also conform to the Elovich model (Jang et al., 2018). This indicates that the adsorption of ECs is mostly chemical adsorption and intra-particle diffusion adsorption, while adsorption thermodynamics show that these adsorption processes are spontaneous endothermic reactions. The adsorption mechanisms of ECs depend on the physicochemical properties of biochar, including the SSA, functional groups, and special properties of biochar feedstocks (Zheng et al., 2013). Modified biochar produces both physical and chemical adsorption during the adsorption processes, but the dominant adsorption mechanisms may not be the same when adsorbing different pollutants. In general, there is a strong electron donor-acceptor (EDA) interaction between the aromatic ring of π -donor on the modified biochar surface and the π -protonated adsorbate. Besides, electrostatic interactions, hydrogen bonding, surface complexation, cation exchange and non-specific van der Waals interactions may also occur during the adsorption (Peiris et al., 2017) (Fig. 3).

3.1. π - π bond interaction

π - π bond interaction is a weak interaction that often occurs among aromatic rings (Wang and Wu, 2008). The adsorbent and adsorbate are bound together by electron transfer between the electron donor and acceptor. This adsorption mechanism plays a leading role in the adsorption process of many carbonaceous materials on pollutants. When biochar is applied to adsorb ECs, the typical ECs mainly affected by this mechanism include TC, CIP, SAS, EDCs, and CBZ (Jing et al., 2014).

First of all, TC is one of the most widely used antibiotics, ranking the top three in clinical prescriptions in the United States in 2010

Table 3
Adsorption performance of modified biochar to EDCs.

Raw material	Endocrine Disrupting Chemicals	Modification method	pH	Pyrolysis temperature (°C)	Adsorption capacity (mg/g)	Removal rate (%)	References
Banana peel	BPA	FeSO ₄ ·7H ₂ O (0.1 g)	6.28	600	—	90	Rong et al. (2019)
Bamboo	BPA	CuZnFe ₂ O ₄	7.0	550	263.2	—	Heo et al. (2019)
Spruce	BPA	FeSO ₄ ·7H ₂ O	7.6	1200	77.4	—	Baldikova et al. (2019)
Corn stalks	BPA	H ₃ PO ₄	7.0	300	476.19	—	Zhao et al. (2017)
Sawdust	BPA	PMS/Fe	8.2	700	66.02	—	Jiang et al. (2019)
Kenaf bar	BPA	nZVI	3.0	600	—	98	Liu et al. (2018a)
Bamboo	4-NP	Fe ₃ O ₄	3.0	800	—	85	Dong et al. (2019)
Rice	NP	nZVI	7.0	400	—	96.2	Hussain et al. (2017)
Cotton straw	E2	NH ₂ Cl	9.0	350	—	87	Wang et al. (2019c)
Cotton straw	EE2	NH ₂ Cl	9.0	350	—	75	Wang et al. (2019c)
Rice husk	E2	NaOH/NaHCO ₃	7.0	500	44.9	—	Zhang et al. (2019a)
Bagasse	E2	Fe ₃ O ₄	4.0	400	50.24	—	Dong et al. (2018)
Bagasse	E2	Fe ₃ O ₄	4.0	600	41.71	—	Dong et al. (2018)
Bagasse	E2	Fe ₃ O ₄	4.0	800	34.06	—	Dong et al. (2018)
Lotus seeds	E2	KOH	7.0	650	100.6	—	Liu et al. (2020)
Litchi	E1	Fe/Mn	3.0	650	4.18	—	Tao et al. (2019a)

BPA: Bisphenol A; NP: Nonylphenol; E2: Estradiol; E1: Estrone; EE2: Ethinyl estradiol.

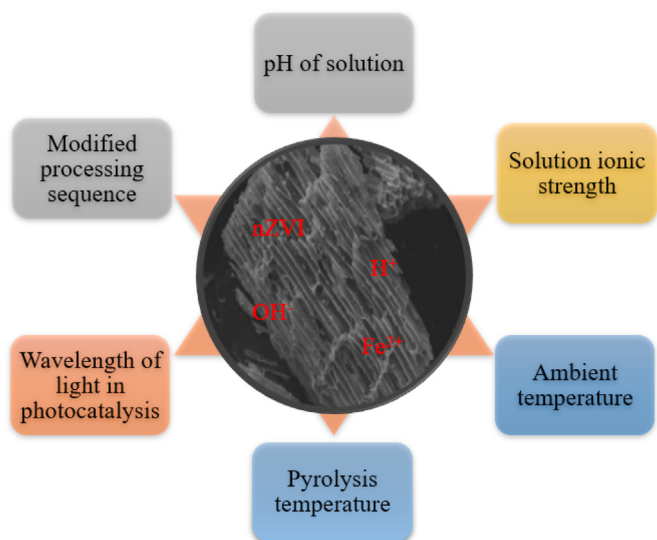


Fig. 2. Factors of EDCs adsorption on modified biochar.

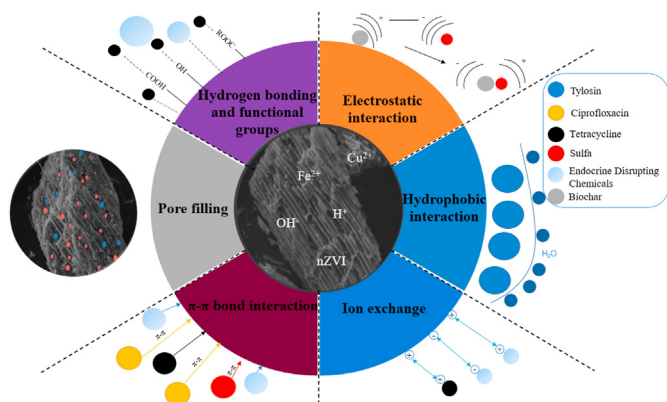


Fig. 3. Adsorption mechanisms of EDCs on modified biochar.

and global sales of animals in 2009 (Park et al., 2017). There have been a lot of research on the use of modified biochar to remove TC from water (Nguyen et al., 2019; Tang et al., 2018; Zhou et al., 2019b). Methanol modification has the effect of increasing the TC adsorption capacity and reducing the organic content in the biochar. XPS showed that the main reason was that the change in oxygen-containing functional groups in the modified biochar affected the π-π EDA interaction between biochar and TC (Jing et al., 2014). Some studies have explored the adsorption of TC on pig manure-derived biochar. The results showed that chemical adsorption (including hydrogen bond and π-π EDA interactions) might be the main adsorption mechanisms (Chen et al., 2018). Liu et al. showed through experiments that π-π stacking, pore filling, silicate bonds, and hydrogen bonds were the main mechanisms for biochar removal of TC (Liu et al., 2019). Meanwhile, steam-activated biochar was also prepared to study co-adsorption. The adsorption capacity and the interaction between TC and Cu²⁺ were studied through mono- and binary systems and adsorption isotherms. When TC coexisted with Cu²⁺, the adsorption capacity of steam-activated biochar for both pollutants was significantly enhanced. Synergistic removal of TC and Cu²⁺ involved bridge enhancement and site competition (Wang et al., 2020b). Most studies have shown that the adsorption of TC onto modified biochar is closely related to the interaction of π-π EDA, which indicates that the π-π EDA interaction plays a leading role (Li et al., 2020; Li et al., 2019a; Yan et al., 2019). Therefore, providing more π-π EDA interactions on the surface of biochar could be an effective method to increase the adsorption efficiency.

Some studies modified potato stem and leaf biochar with KOH and found that the modified biochar had better performance in removing CIP. The maximum adsorption capacity increased by 2.3 times. FTIR analysis showed that its adsorption mechanism mainly included π-π interaction EDA, hydrogen bonding interaction, and electrostatic interaction (Li et al., 2018). In terms of other studies on compound pollution containing sulfa antibiotics, pine sawdust was used to prepare biochar and then magnetized it to adsorb SMX. Studies at different pH, ionic strength, natural organic matter (NOM) and 17α ethinyl estradiol revealed that the adsorption mechanisms of SMX and EE2 could be attributed to π-π EDA and hydrophobic interaction (Reguayal and Sarmah, 2018). Although many people have studied the adsorption of SAs onto modified

biochar, in-depth study of the adsorption mechanism remains to be done. The most common adsorption mechanism is believed to be the interaction between the electron moiety and the EDA on the surface aromatic ring. Due to the strong EDA interaction between a large number of π -donor aromatic hydrocarbon rings and π -protonated adsorbates on the surface of biochar, high-temperature production of biochar shows high adsorption under moderately acidic conditions (Peiris et al., 2017). In another study on the co-adsorption of Cr^{6+} and BPA, the results of infrared spectroscopy and molecular simulation studies showed that Cr^{6+} was mainly adsorbed on the surface of biochar by chemical complexation, while BPA was adsorbed by π - π EDA interaction (Zhao et al., 2017). By using zeolite, silica or nano-zero valent iron, the biochar derived from the date palm waste was modified to adsorb chlortetracycline. The results showed that the main adsorption mechanisms of chlortetracycline on biochar, silica modified biochar and zeolite modified biochar included chemical adsorption, hydrogen bonding and interparticle diffusion. The difference is that chlortetracycline is easier to be adsorbed by nZVI modified biochar due to the π - π EDA interaction and redox reaction. Therefore, nZVI-DBC can be used as an efficient green adsorbent to remove chlortetracycline from aqueous solution and reduce the pollution of surface date palm waste (Ahmad et al., 2019).

In addition to the above-mentioned ECs, carbamazepine (CBZ) is a class of anti-epileptic drugs, and it can affect the embryonic development and larval behavior of fish in aquatic environment (Qiang et al., 2016). Some studies have prepared ultrafine magnetic biochar/ Fe_3O_4 modified with coconut, pine nuts, and walnut shells. The hybrid adsorbent prepared after grinding for 2 h had a high carbamazepine removal rate and was easy to magnetically separate. Rapid adsorption of CBZ and TC was shown within the first hour. Grind the spent adsorbent with CBZ and TC could degrade the adsorbed pollutants. It was found that the addition of quartz sand could improve the mechanochemical degradation of CBZ on biochar/ Fe_3O_4 , and the degradation percentage was as high as 98.4% at the dosage of 0.3 g quartz sand/g adsorbent (Shan et al., 2016). At the same time, there are also researchers who combine biochar with photocatalytic technology. A magnetically separable heterojunction photocatalyst $\text{Fe}_3\text{O}_4/\text{BiOBr}$ was prepared by a hydrolysis method, which was stacked on a reed straw biochar ($\text{Fe}_3\text{O}_4/\text{BiOBr}/\text{BC}$) with visible light response. The photocatalytic activity of $\text{Fe}_3\text{O}_4/\text{BiOBr}/\text{BC}$ on the target pollutant carbamazepine was further studied. The results show that under 50 W visible light, the $\text{Fe}_3\text{O}_4/\text{BiOBr}/\text{BC}$ has good CBZ photodegradation activity (95.51%). The pH had little effect on the photodegradation of CBZ using $\text{Fe}_3\text{O}_4/\text{BiOBr}/\text{BC}$, which meant wider range of practical applications. Good reusability and stability proved that the $\text{Fe}_3\text{O}_4/\text{BiOBr}/\text{BC}$ has superior practicality and feasibility in removing organic pollutants in water (Li et al., 2019a). Activated biochar was prepared by carbonizing and using pomelo peel as a precursor for further KOH activation. It was found that the adsorption process was spontaneous and exothermic, mainly physical adsorption. The pH analysis and the surface analysis of the biochar show that the adsorption of CBZ on activated biochar was mainly controlled by π - π EDA interaction (Chen et al., 2017).

3.2. Hydrogen bonding interaction and functional groups

Functional groups are atoms or groups of atoms that determine the chemical properties of organic compounds. Many functional groups can react with each other, and some functional groups can also form hydrogen bonds (Vijay et al., 2008). Through the formation of hydrogen bonds, they have strong bond energy and are not easy to separate. Many studies have shown that the ECs most sensitive to this adsorption mechanism are some environmental

hormones, such as E2, EE2, BPA and nonylphenol.

Hydrogen bonding and hydrophobicity are the main mechanisms that affect the adsorption of E2 by biochar (Dong et al., 2018; Zhang et al., 2019a). Some people have made magnetic modified biochar to adsorb E2, and the magnetic biochar produced had a SSA nearly 14.5 times larger than that of the original biochar, which helped improve the adsorption capacity of E2. The synthesized magnetic biochar had excellent E2 adsorption capacity. As the pyrolysis temperature increased, the main adsorption interaction between E2 and magnetic biochar changed from hydrogen bonding to π - π interaction. The magnetic biochar regenerated through odorization could be repeatedly subjected to E2 adsorption at least 5 times, while the adsorption capacity did not reduce too much (Dong et al., 2018). Others have studied the effects on pre- and post-treatment of biochar. The studies found that the sequence of KOH activation and activation steps had a great impact on the material. After KOH treatment, SSA, pore size, and pore volume were significantly improved compared with the original biochar. The adsorption capacity of post-treated biochar to E2 was better than that of direct-treated biochar, pre-treated biochar, and original biochar. Higher pyrolysis temperature would be more helpful for removing E2. External environmental factors (temperature, pH, and ionic strength) might greatly affect the E2 adsorption capacity of biochar (Liu et al., 2020). In one study, biochar was prepared from litchi and used as an adsorbent to control the mobility of estrone in water and soil environments. Modified biochar (Ca-BC and Fe-Mn-BC) could effectively control estrone diffusion, especially Fe-Mn-BC. These findings were essential for controlling the transport of estrogen compounds in the water and soil environment (Tao et al., 2019a). Some people have studied the activation effect of biochar on monochloramine (NH_2Cl), and about 95% of the parent E2 and EE2 were removed in cotton buffer (Cot350) at 350 °C by Cot350/ NH_2Cl , and 87% of E2 and 75% of EE2 were removed from the urine within 24 h. Electron paramagnetic resonance analysis and free radical quenching experiments display that biochar activated by NH_2Cl mainly produced NO free radicals, which was helpful for the degradation of EDC (Wang et al., 2019c).

In the adsorption process of biochar to phenolic compounds, people have also found the influence of this mechanism. For the removal of BPA, a magnetic biochar (γ - $\text{Fe}_2\text{O}_3/\text{BC}$) with high SSA was prepared by a simple and effective hydrothermal method. It was found that γ - $\text{Fe}_2\text{O}_3/\text{BC}$ was very effective in the degradation of BPA without the pH adjustment, and it could be completely removed within 20 min (Rong et al., 2019). In order to remove the compound pollutants (BPA and SMX) from aqueous solution, Heo et al. synthesized a new type of biochar supported magnetic $\text{CuZnFe}_2\text{O}_4$ composite (CZF-biochar) by a simple hydrothermal method. It was found that the main mechanisms included charge-assisted hydrogen bonding, hydrophobicity, and π - π EDA interactions (Heo et al., 2019). In order to facilitate the separation of the modified biochar from water, magnetic sorbents attracted much attention due to their advantages such as economic convenience, high adsorption efficiency, and easy operation. Some people have used magnetic modification to make a very simple spruce sheet-shaped modified biochar, and found that its efficiency of removing BPA was very high. The adsorption process was a spontaneous exothermic process and could be well described by a pseudo-second-order kinetic model (Baldikova et al., 2019). For the simultaneous removal of heavy metals and BPA, a new type of biochar-supported zero-valent iron (BC-nZVI) was synthesized by a green method, and BC-nZVI and persulfate (BC-nZVI/PS) combination to remove Cu^{2+} and BPA simultaneously. The results showed that SO_4^{2-} was the main free radical responsible for BPA degradation (Liu et al., 2018a).

Nonylphenol is an endocrine-disrupting chemical that can

interfere with the hormone system of various organisms in the environment. nZVI was loaded on biochar made from low-cost rice husks to produce nanocomposites (nZVI/BC). The effects of initial pH, persulfate concentration, and amount of nZVI/BC nanocomposites were studied. Due to the existence of large-area oxygen molecular functional groups on the biochar surface, nZVI/BC nanocomposites increased the generation of SO_4^{2-} , thereby enhancing the degradation of nonylphenol (Hussain et al., 2017). Some studies have synthesized two types of MnO_2 biochar composites with three-dimensional structure through hydrothermal processes with duration control and evaluated their ultrasound-assisted heterogeneous Fenton-like catalytic activity. The results exhibited that in the ultrasound-assisted heterogeneous Fenton-like process, the sea urchin-like modified biochar was expected to show excellent catalytic performance, thereby making it widely applicable to the environmental remediation of various organic pollutants (Jung et al., 2019). For the research of such phenolic compounds, more works need to be carried out in terms of adsorption behavior and mechanisms in the future.

3.3. Hydrophobic interaction

When non-polar groups (such as C–H) exist in the molecule, there is mutual repulsion between water molecules. The repulsion is called hydrophobic interaction (Xie et al., 2020). This adsorption mechanism is also common in the adsorption process of ECs by biochar, such as estrogen, E2, tylosin (TYL) and some SAs. Previous studies have investigated the effect of microwave-assisted chemical modification methods on the properties of biochar and applied it to remove 17-estradiol (E2) from water. The results exhibited that the alkali-modified biochar had higher adsorption capacity for E2 than other modified biochars, which was attributed to its higher SSA and excellent hydrophobicity (Zhang et al., 2019a). Many studies on the adsorption of SAs by different modified biochars have been carried out. Modified biochar derived from cotton stalks was used to adsorb SAs. It was found that van der Waals and hydrophobicity played major roles in the adsorption behavior of the SAs (Sun et al., 2018a). Studies performed characterization of nano Co_9S_8 and CoO coated with nitrogen-sulfur-doped biochar ($\text{CoO}/\text{Co}_9\text{S}_8@N\text{-S-BC}$) and used them to activate peroxymonosulfate (PMS) to remove SMX. The result showed that $\text{CoO}/\text{Co}_9\text{S}_8@N\text{-S-BC}$ could effectively activate PMS to degrade sulfamethoxazole, and 0.08 mM SMX could be completely degraded within 10 min (Wang and Wang, 2020). Ball-milled biochar has been used to remove two SAs (SMX and SPY) from wastewater (Huang et al., 2019). Some studies have also applied self-nitrogen-doped biochar to decorate the nickel foam electrode and used it to electro-Fenton process to remove SMX (Deng et al., 2019)). In addition, photochemical degradation method was also used to deal with it, that is, the TiO_2 doped with zinc element was stacked on the biochar of the reeds straw, and the pretreatment was carried out with acid ($\text{Zn-TiO}_2/\text{pBC}$). The photocatalytic activity of $\text{Zn-TiO}_2/\text{pBC}$ was further studied through the photodegradation of SMX (Xie et al., 2019).

TYL is a multi-component antibiotic. It is a complex mixture of closely related components produced by a streptomyces strain. It is used in veterinary disease treatment and to promote poultry growth (Hamidian et al., 2018). The enhanced TYL removal effect of new goethite biochar composites from water has been reported. Scanning electron microscope image characterization showed that goethite nanoparticles were well dispersed on the surface of biochar. The adsorbent had fast and efficient adsorption performance for TYL. The adsorption mechanism was attributed to hydrophobicity, static electricity, hydrogen bonding, cation exchange, and EDA interaction (Guo et al., 2016). Regarding the researches on TYL adsorption, the current biochar modification methods mainly use

metal oxides or clay minerals. The main reason is that the two modification methods are easier to enhance surface adsorption. After clay mineral modification, it can make the pore size of biochar larger, and the hydrophobicity is stronger.

3.4. Electrostatic interaction

Electrostatic interaction is the essence of ionic bond formation, which includes electrostatic attraction and repulsion. Ionic bond is a chemical bond formed by electrostatic interaction between anions and cations generated after atoms gaining and losing electrons (Verliefde et al., 2008). Previous studies have shown that the electrostatic interaction plays a dominant role during the adsorption of TC, SAs, BPA, and NPX onto biochar (Chen et al., 2018; Paunovic et al., 2019).

The adsorption of SAs onto modified biochar shows high adsorption performance under moderately acidic conditions. Electrostatic interaction may occur during the adsorption process (Peiris et al., 2017). Chen et al. found that when the pH was increased from 5.0 to 9.0, the adsorption capacity of modified biochar for TC increased, which was largely due to the increased electrostatic attraction between TC and biochar (Chen et al., 2018). Others have successfully prepared a Z-type photocatalyst based on $\text{biochar}@CoFe_2O_4/Ag_3PO_4$ by a simple method. When BPA was selected as the target pollutant, its degradation efficiency and mineralization rate could reach 91.12% and 80.23%, respectively (Zhai et al., 2020). Due to the difficult degradation of PMS, a Fe-functional biochar composite, porous carbon with rich functional groups and nanofibers (Fe-BC-700) was prepared to activate PMS and effectively remove BPA. The results exhibited that the electron transfer of nanofiber-mesoporous carbon structure and the effects of persistent free radicals inherent in biochar could help improve the performance of biomass (Jiang et al., 2019).

For other ECs, some studies used wild plums as raw materials to make raw biochar, and then immersed the raw biochar in KOH solution. Then microwave functionalization was performed at 700 W for 12 min to prepare functional biochar (WpOH). It was applied to the removal of the ionizable drug naproxen (NPX). The main adsorption mechanisms were studied by FTIR, XRD and other analytical methods, indicating that the electrostatic attraction between the negatively charged NPX and the positively charged WpOH functional group was the main interaction mechanism (Paunovic et al., 2019).

3.5. Pore filling

In addition to the above-mentioned mechanisms, most adsorption processes also involve pore filling and intra-particle diffusion. Some studies used alfalfa hay as a raw material to prepare biochar by activating with NaOH to remove TC in water. The results showed that the activated biochar had higher adsorption capacity (302.37 mg/g) for TC than the pristine biochar, which was comparable to commercially available activated carbon. The adsorption kinetics and isotherm studies showed that strong chemisorption interaction occurred between TC and biochar, in which intra-particle diffusion was considered to be the main mechanism of TC adsorption onto biochar (Jang and Kan, 2019). This result is also verified in another study (Huang et al., 2017). Since there is not only a single antibiotic in the real water environment, but also a large number of heavy metals and environmental hormones, therefore, people used nano-hydroxyapatite modified biochar to adsorb Cu^{2+} and sulfamethoxazole/TYL in water. The adsorption capacity was positively correlated with specific surface area and pore size. Surface adsorption was one of the main mechanisms (Li et al., 2019b). Another research showed

that the removal mechanism of TYL included surface complexation, electrostatic interaction, hydrogen bond, and hydrophobic interaction (Yin et al., 2018). For the adsorption of BPA, some studies believed that this was related to the surface oxidation of biochar (Zhou et al., 2018). Besides, pore filling is a common adsorption mechanism in SAs due to the small size of modified biochar (Peiris et al., 2017). Some studies investigated the adsorption mechanism of TC onto activated biochar derived from pinus taeda, and found that the Elovich kinetic model and Freundlich isothermal model were well-fitted, indicating that the surface hydrogen bonding and π - π interaction could be the main mechanism, and intra-particle diffusion would be the main limitation of TC adsorption on activated biochar (Jang et al., 2018). It can be deduced from the above research that pore filling and intra-particle diffusion play an important role in the adsorption process of many ECs.

3.6. Ion exchange

Ion exchange is a reversible reaction between ions in the liquid phase and ions in the solid phase. When some ions in the liquid phase are adsorbed by the ion exchange solid, the ion exchange solid will release equivalent ions back into the solution to maintain the electrical neutrality of the aqueous solution. Since ECs are organic compounds, they are not easily converted into ionic forms. Therefore, ion exchange seldom occurs when biochar adsorbs ECs. However, some studies have also shown that ion exchange may exist in the adsorption process of ECs on biochar (Guo et al., 2016; Wu et al., 2019b). For instance, waste sludge biochar modified by chitosan and Fe/S (BCFe/S) has been prepared for TC removal in water. The results showed that electrostatic attraction, π - π stacking, pore filling, silicate bonds, and hydrogen bonds were the main mechanisms for biochar removal of TC. Besides, chelation and ion exchange were also the adsorption mechanisms of BCFe/S-4 for TC (Liu et al., 2019). More further works are highly needed to verify this viewpoint.

3.7. Adsorption mechanisms of other ECs

In addition to the common antibiotics mentioned above, there are still many antibiotics in the environment, such as oxytetracycline (Ramanayaka et al., 2020), chlortetracycline, etc. Some of them have not attracted much attention because of their low concentrations, but they also could cause irreversible consequences. Some studies have synthesized nano-manganese dioxide ($n\text{MnO}_2$) modified biochar to improve the adsorption capacity of biochar on di-n-butyl phthalate and oxytetracycline (Gao et al., 2018). It is reported that the preparation of a highly efficient novel adsorbent based on Co/Fe bimetal nanoparticles loaded with alkali-modified biochar (Co/Fe/MB) could remove cefotaxime from aqueous solutions. The results of high-performance liquid chromatography-electrospray ionization mass spectrometry showed that the opening and cleavage of the β -lactam ring was the

first step in the degradation of cefotaxime. It was found that the Co/Fe/MB composite was an effective material for removing cefotaxime from solution (Wu et al., 2018). At present, there are few studies on the adsorption of these antibiotics on biochar, and many studies are still in their infancy. Research on the adsorption effect and mechanism of modified biochar on these antibiotics will be the future research direction.

Some people have conducted research on two types of activated biochar (N-/O-biochar), which target non-steroidal anti-inflammatory drugs (diclofenac, naproxen, ibuprofen), and explained binding energy by modeling binding molecules. Weak adsorption of ibuprofen was observed in the presence of adsorption competitors. Due to the lower binding energy, polarity, and π - π bond energy, binding to the adsorbent, and electrostatic repulsion from the solute occupying the adsorption site, the presence of the competitive adsorption resulted in lower binding energy (Jung et al., 2015). Some people used K_2FeO_4 as an activator to treat biochar, and modified biochar (Fe@BC) was used to remove diclofenac sodium in aqueous solution. The maximum adsorption capacity reached 123.45 mg/g. The possible mechanisms of the adsorption process of diclofenac sodium on Fe@BC might involve chemisorption with hydrogen bonding and π - π interaction (Tam et al., 2020). Zn-Co-LDH@BC showed high photocatalytic efficiency due to inhibiting the agglomeration of LDH nanostructures by incorporation of biochar. The results show that in the presence of Zn-Co-LDH catalyst, 92.7% of gemifloxacin was photocatalytic degraded (Gholami et al., 2020b). Caffeine is becoming more and more harmful in water. People begin to study how to adsorb caffeine in water. Studies have synthesized a biochar composite using bovine bone activated carbon as the carrier of layered double hydroxide particles and used it as an adsorbent for removing caffeine from water. Kinetic studies found that equilibrium could be achieved only by contacting the adsorbent with the adsorbed substance for 20 min. The study proved that the composite was satisfactorily synthesized (26.219 mg/g), its use in decaffeination was very attractive, and it was a potential adsorbent in water treatment applications (Lins et al., 2019). Studies have prepared activated biochar, characterized and examined the feasibility of removing ethyl para-hydroxybenzoate from aqueous solutions. After five consecutive adsorption/desorption cycles, the adsorption capacity remained 90.82% (Zhou et al., 2019a). Adsorption performance of different modified biochar on other ECs is shown in Table 4. Currently, researches on these ECs are of less concern, so the scope of research needs to be broadened in the future.

4. The cost and regeneration performance of biochar

Another important reason why biochar becomes popular is its low cost, most of the raw materials for biochar are solid waste or agricultural waste, which is one of the reasons why the cost of biochar is lower. Some studies have found that the price of modified biochar is almost half of activated carbon, and its adsorption

Table 4
Adsorption performance of modified biochar on other ECs.

Raw material	ECs	Modification method	pH	Pyrolysis temperature (°C)	Adsorption capacity (mg/g)	Removal rate (%)	References
Bovine bone	Caffeine	MgAl-LDH	12.0	—	26.219	—	Lins et al. (2019)
Pomelo peel	CBZ	KOH	4.4	400	286.5	—	Chen et al. (2017)
Reed straw	CBZ	Fe_3O_4	6.0	500	—	95.51	Li et al. (2019a)
Wild plum kernels	NPX	KOH	6.0	500	73.14	—	Paunovic et al. (2019)
Oil palm fibre	EP	KOH	6.0	500	349.65	—	Zhou et al. (2019a)
Husks and paper sludge	GMF	Zn-Co-LDH	5.5	500	—	92.7	Gholami et al. (2020b)
Pitch pine sawdust	DCF	K_2FeO_4	6.5	900	123.45	—	Tam et al. (2020)

CBZ: Carbamazepine; NPX: Naproxen; EP: Ethyl hydroxybenzoate; GMF: Gefifloxacin; DCF: Diclofenac sodium.

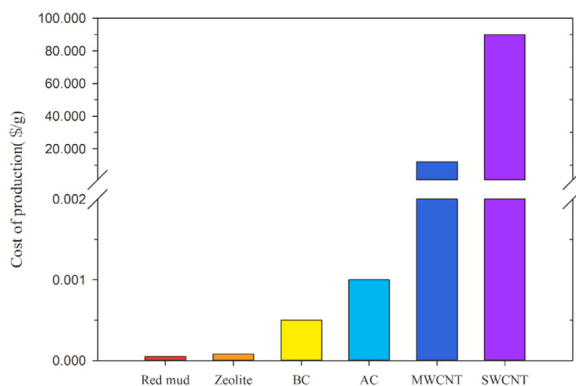


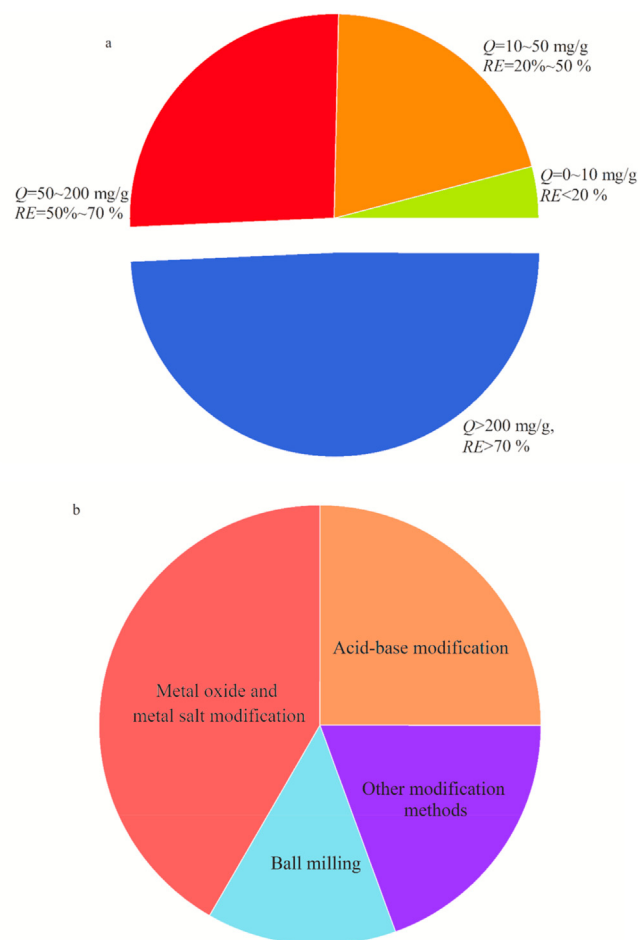
Fig. 4. Cost of different adsorbent materials (Lin and Juang, 2009; Lu et al., 2008; Marousek, 2014).

capacity is nearly equivalent to activated carbon (Fdez-Sanroman et al., 2020). Furthermore, its adsorption capacity is much higher compared with other cheap adsorbents (Lin and Juang, 2009) (Fig. 4). Currently, most of the feedstocks for biochar preparation are agricultural and forestry solid wastes, sewage sludge and other organic solid wastes. Therefore, the use of solid wastes to prepare biochar can not only reduce its production cost, but also realize the resource utilization of solid waste. In addition, during the production and preparation of biochar, bio-oil and biogas are produced, both of which can be used for energy recovery and utilization, thereby reducing its production cost to some extent. With the continuous optimization of pyrolysis equipment and processes, the bio-oil and biogas produced during the pyrolysis process can also be recycled for the pyrolysis process (Frank et al., 2020).

Regarding the regeneration performance of biochar, many studies conduct regeneration experiments. The methods of regeneration include pyrolysis, elution with eluent, microwave elution (de Lima et al., 2019). The regeneration method of modified biochar with adsorbed antibiotics is mostly elution with eluent. Afzal et al. conducted a study to evaluate the regeneration of chitosan biochar hydrogel beads and found that it could be adsorbed and desorbed for 6 cycles, maintaining a high level in the first three cycles. Adsorption performance starting from the fourth cycle, the adsorption performance began to decline (Afzal et al., 2018), ZnCl₂ modified biochar could also maintain a constant adsorption capacity in three cycles (Yan et al., 2020). While other studies found that acid-base modified biochar slightly decreased after five adsorption cycles (Tang et al., 2018), and wheat straw montmorillonite modified biochar could remain stable within five cycles (Zhang et al., 2018b). In another experiment, the magnetic modified biochar was able to maintain stable adsorption in five cycles, and a higher adsorption capacity was observed in the first three cycles (Dai et al., 2020a). In summary, most of the modified biochar has a good regeneration capacity and can maintain a stable adsorption capacity or adsorption efficiency within three to five cycles. The high-quality renewability gives biochar great potential for practical applications.

5. Conclusions and future perspectives

In recent years, the research scope of different ECs adsorption onto modified biochar is gradually expanding. This paper systematically reviews the modification methods of biochar and the related adsorption effects, behaviors, and mechanisms for several highly concerned ECs. Modified biochar generally shows a better adsorption effect on ECs. According to statistical analysis, metal oxide and metal salt modification are considered to be a better



Q: Adsorption capacity; RE: Removal rate

Fig. 5. (a) Distributions of ECs adsorption effects by different modified biochars (b) Distributions of different modification methods with high ECs adsorption capacity (Q > 200 mg/g; RE > 70%) (Data from 76 literatures).

modification method (Fig. 5). With the researches on biochar adsorption of ECs, the degradation efficiency of modified biochar can be greatly improved. How to screen out effective methods to modify biochar for enhanced adsorption capacity of ECs is the focus of future research. With the continuous development of modern treatment technology, more methods will be used for the modification of biochar and its removal of ECs in the environment. Recent advances in nanotechnology provide ample opportunities to develop next-generation water treatment processes (Liu et al., 2018b). Compared to traditional materials in bulk or large particles, due to their unique size, some nanomaterials have shown better potential for removing ECs in laboratory-scale studies (Zhao et al., 2018).

In the future, it is also possible to combine biochar with other treatment methods, such as combining biochar with biofilter to expand the strong adsorption performance of biochar for environmental remediation (Lu and Chen, 2018). More studies on the combination of photocatalytic technology and ball milling technology with biochar applied to the removal of ECs in the environment, this technology provides new sights for future study in this field (Lyu et al., 2020). It has been found that biochar can reduce the environmental risk of antibiotic resistance genes in some cases (Sun et al., 2018b). Therefore, improving the removal efficiency and

reducing the resistance of ECs by biochar modification may be a new research direction in the future.

With the development of biochar research, new problems and challenges inevitably will come out. Therefore, the following issues should be addressed in future studies (Fig. S2):

The first issue is related to regeneration. How to effectively desorb the pollutants adsorbed on the biochar for subsequent safety treatment and to recycle the adsorbed biochar well are the issues that need to be considered (Wang et al., 2015). At present, heat treatment and modification of biochar maybe two feasible methods to solve the problem. Heat treatment regeneration is to heat the biochar under low temperature (100–300 °C) to desorb pollutants (Zeng et al., 2021), recover its surface activity and adsorption performance. Since the heat treatment regeneration is under really low temperature, it has been widely used due to its low energy consumption and easy operation heat regeneration could enhance the economic and environmental sustainability of biochar adsorbents (Greiner et al., 2018). It was found that the SMX adsorption amount of heat-treated biochar was about 20% of activated carbon after heat treatment regeneration. However, the gas pollutants generated during the heat treatment regeneration process may affect the quality of ambient air. Meanwhile, the adsorption efficiency of biochar may decrease after heat treatment regeneration. Therefore, developing more environmentally friendly treatment methods is highly in need in the future (Wu et al., 2019a).

The second is the issue of environmental risks. The stability and environmental risk of modified biochar have become the focus of current research. Is there any possibility of secondary pollution to the environment for modified biochar? For example, can the metal-modified biochar react with other compounds in the environment after being applied to water or soil? Whether it has an antagonistic effect with other target compounds and how to separate the treated biochar from the water are the key issues which need to be urgently solved in the future.

The third is the improvement of biochar modification. Two aspects could be focused on in the future, mainly including adsorption capacity improvement and cost reduction. In terms of improvement of modification methods, the key point is to prepare different modified biochar according to actual application needs. The first is to produce modified biochar with high adsorption capacity to absorb pollutants as many as possible while ensuring its adsorption effect. Secondly, modified biochar with selective adsorption function needs to be developed to adsorb target pollutants. In the aspect of cost reduction, although the modified biochar has better adsorption efficiency, some modification methods are costly (Wu et al., 2017). In the future, the use of environmentally friendly, green and low-cost modifiers such as some industrial solid wastes could be a future development direction (Lian et al., 2019; Wang et al., 2020a). Industrial solid wastes include red mud, fly ash, phosphogypsum, coal gangue, tailings, etc. The pyrolysis of industrial solid wastes and biochar raw materials together could not only reduce the toxicity of industrial solid wastes, but also realize stabilization and resource utilization, thus reducing the harm of industrial solid wastes to the environment (Qiu et al., 2018).

The fourth is on the adsorption mechanism of modified biochar. How the pollutants are adsorbed on modified biochar still needs more in-depth research. Many existing studies have explained this as a result of several interacting mechanisms. However, it is still unclear which mechanism plays a leading role and how much it contributes. Clarifying these issues is of great significance for improving the adsorption capacity and environmental applications of modified biochar. At the same time, the solution to these problems is also the key to future industrialization applications. In terms of research methods, biochar adsorption experiments should be

gradually approached to actual dynamic operations. The simulation experiments performed in the laboratory are different from the actual ones. Future research should focus on solving these problems mentioned above. In summary, the research and development prospects of modified biochar in the treatment of ECs in water are promising.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2021.116448>.

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