Review

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Biochar for environmental management: mitigating greenhouse gas emissions, contaminant treatment, and potential negative impacts

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Abstract

With increased interest in scientific investigations and large-scale applications of lignocellulosic biomass derived biochar for environmental management, a comprehensive understanding of its function in the fragile earth environment is urgently needed. The study on the relationship between biochar properties and potential applications requires continuing research. This review provides new insights into the state-of-the-art accomplishments in the utilization of biochar in environmental management and covers three perspectives: firstly, mitigation of greenhouse gas (GHG) emissions, such as sequestration of CO_2 and CH_4 in global carbon pools and mitigation of N_2O emissions; secondly, pollution control, including adsorptive removal and reactive removal of inorganic and organic contaminants; thirdly, potential negative aspects of biochar applications, including contaminations originated from biochar, negative alterations to soil properties and soil biota, negative impacts of biochar on GHG emissions and negative impacts of biochar migration. From a unique and comprehensive environmental perspective, this article aims to provide a critical review of updated knowledge on both positive and negative impacts of biochar for environmental management, based on an exponentially increased number of publications on the topic over the past decade.

Key words: Biochar; Carbon sequestration; Contaminant management; Adsorption; Reaction

1. Introduction

To address and meet the serious global environmental issues, food security and energy shortage with an ever-increasing human population in a still generally fossil fuel-based society, it is inevitable to seek innovative, efficient, sustainable and economically attractive solutions [1]. Biomass is a biological material originally derived from the reactions between readily available atmospheric carbon dioxide, water and sunlight, via photosynthesis, while its sustainability as a feedstock is a great source for biofuels and/or chemicals production without carbon emission [2, 3]. In the past decade, biomass has gained great attention for its sustainable and convertible properties. The total annual available biomass is 220 billion dry tons (equivalent to ca. 4500 EJ of energy content) and the theoretically harvestable bioenergy potential is estimated to be 2900 EJ (270 EJ could be considered technically available on a sustainable basis) [4]. Biological (e.g., hydrolysis, fermentation, anaerobic digestion) and thermal (e.g., pyrolysis, gasification, combustion) methods have been used for biomass conversion into fuel (bio-oils and gases) and byproducts [5, 6]. Many literature reviews have extensively described the pyrolysis and gasification technologies of lignocellulosic biomass for bio-oils, syngas and process heat [7-10]. However, few reviews [11-13] have focused on the black byproduct (biochar) from the perspective of global sustainability, especially the role in environmental management.

For biochar (short for bio-charcoal), Lehmann and Joseph defined it as "*a carbon (C)-rich product when biomass such as wood, manure or leaves is heated in a closed*

container with little or unavailable air" and distinguished it operationally from charcoal based on their end-use application (charcoal for producing energy and fuel; biochar for carbon sequestration and environmental management) [14]. The original application of biochar in microclimate control in China can be traced back to 206 BC-220 AD (Han Dynasty). Tons of biochar (amorphous carbon and graphite-like structure) were used to build isolation strips to block the matter exchange with external environment, preserving the body and funerary objects fresh for about two thousand years (Fig. 1), and the production method in that era was still an enigma [15]. Biochar utilized in agricultural production is connected to the ancient Amerindian in the Amazon region, where rich black earth was created by using of slash-and-char techniques [16, 17].

Fig. 1. Relics from Mawangdui Han Dynasty Tombs, P.R. China. (a) Painted pottery figurines and T-shaped painting on silk. (b) Figurines of musicians. (c) Coffin of the third layer with painted design on vermilion lacquer coating (Length 230 cm, width 92 cm, height 89 cm). Copyright

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The basis for recent interest in biochar is multi-fold (Fig. 2). Biochar-based strategies are mainly for food security, energy production and global environmental issues. Firstly, biochar-type substances are the explanation for high amounts of organic carbon and sustained fertility in *terra preta* soils [18]. Justifiably or not, biochar has, as a consequence, been frequently connected to agricultural production since it can increase base saturation, porosity, water-holding capacity, cation exchange capacity and nutrients holding capacity of the soil [19, 20], and undoubtedly has a significant impact on soil organism communities and their functions [21, 22], which depends on the properties of both biochar and soil.

Secondly, capturing energy during the production process of biochar and, conversely, using biochar as soil amendment are mutually beneficial for generating the biomass, and reducing greenhouse gas (GHG) emissions. Biochar amendment instead of using it as a fuel is particularly effective in offering global environmental solutions rather than solely producing energy [14, 23]. Converting waste biomass to biochar has great potential for managing agricultural wastes originated from plants or animals and thus contributes to the mitigation of the associated environmental issues. The long-term stability of biochar is a critical factor in decreasing GHG (CO_2, N_2O, and CH_4) emissions into the atmosphere by both abiotic and biotic mechanisms [24-26]. Furthermore, the specific properties of biochar make it possible to remediate contaminated soil and water [27-29]. Compared with other sustainable carbon materials [30-32], biochar is low-cost and effective for treatment of organic and

inorganic contaminants [33, 34].

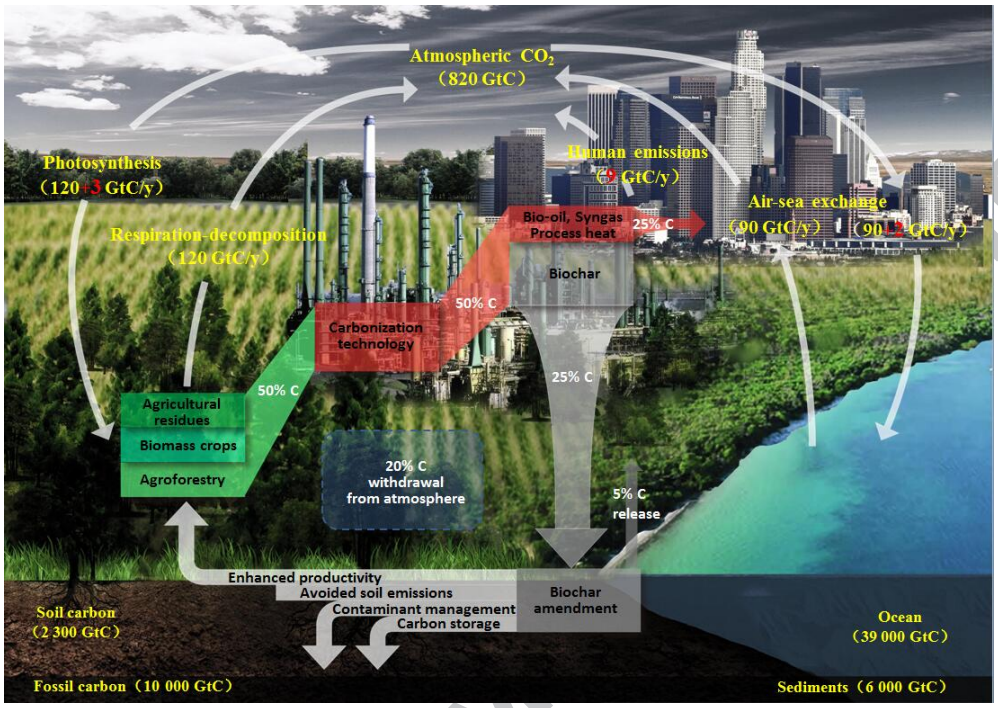


Fig. 2. Overview of the sustainable biochar applications and the global carbon cycle and biomass carbonization technology concept for bio-oil, syngas, process heat and biochar (yellow numbers are natural fluxes and red numbers are human contributions; GtC = Gigatons of Carbon). The numerical data are from Yi et al. [35] and Lehmann [36].

The strong interest can be demonstrated by the numerous publications on biochar for environmental management over the past years (Fig. 3), since conceptual paper "*A handful of carbon*" was published on *Nature* by Johannes Lehmann in 2007 [36]. With the increased interest in scientific investigations and large-scale applications of the black gold "biochar" for environmental management, a comprehensive understanding of its function in the fragile environment is necessary. The available books and papers about biochar are mainly concerning carbon storage [37, 38],

enhancement of crop yield [39, 40], soil amendment [41, 42], mitigation of climate change [23, 43], energy production [44, 45], soil biota [46, 47], etc. [48, 49]. Until recently, only one brief review [50] about its negative responses in soil has been published. There are few reviews focused on biochar from the perspective of environmental management. Throughout the history of human civilization, understanding the whole environmental system and identifying how proposed technologies affect interconnected, complex and managed natural environmental systems are indispensable, especially to solve multiple challenges simultaneously with complementary solutions. This review aims to provide up-to-date knowledge about GHG emissions, contaminant management, and relevant negative aspects. Although biochar can be used as an energy carrier and for agronomical benefits, this paper focuses on biochar for environmental management.

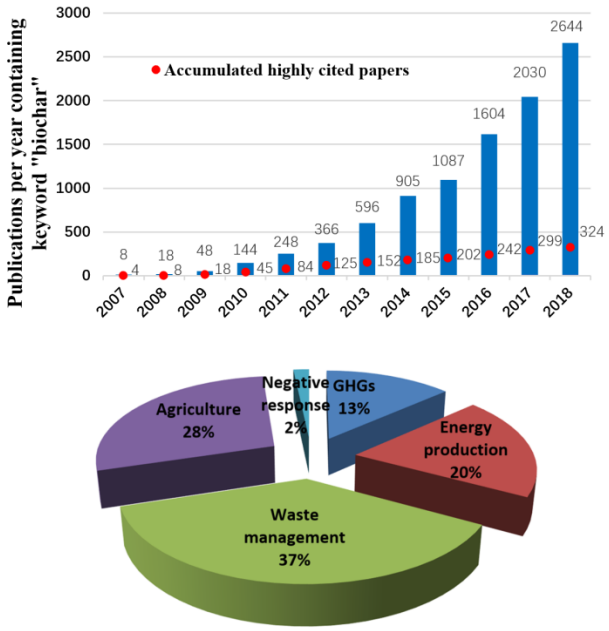


Fig. 3. Publications per year and the accumulated numbers of highly cited papers containing keyword "biochar" on indexed journals between 2007 and 2018. The

percentage of motivation in biochar application. The data are based on the search results from Web of Science (2018).

2. Biochar for mitigating GHG emissions

Biochar and its storage in soils have been heralded as a solution to mitigate GHG emissions by sequestering carbon (C) and simultaneously providing environmental and agricultural benefits [51, 52]. It has been recognized that the decrease of GHG emissions for mitigating climate changes is globally necessary. The Kyoto protocol and Paris Agreement were made aiming at controlling GHG emissions under the UN framework convention on climate change, and nitrous oxide (N_2O), carbon dioxide (CO_2), methane (CH_4), fluorohydrocarbons (HFCs), perfliorocarbons (PFCs), and sulfur hexafluoride (SF_6) were listed as the main GHGs to be mitigated. The contributions of these GHGs to the greenhouse effect are influenced by the properties and abundance of that gas, and potential indirect effects it may cause. HFCs, PFCs, and SF_6 are GHGs with global warming potential many thousands of times greater than CO_2, while CH_4 and N_2O with global warming potential 25 and 298 times greater than CO_2. However, the contribution percentage of CO_2 is ca. 55% for its large amount [53, 54]. Management strategies that avoid CO_2, CH_4, and N_2O emissions can significantly contribute to the mitigation of climate changes.

2.1. Sequestration of CO_2 in global carbon pools

Over the past centuries, the burning of fossil fuels has raised the level of CO_2, which accounts for about forty percent of the total anthropogenic carbon emissions in

the atmosphere [55]. The concentration of CO_2 in the earth's atmosphere was 280 ppm in the 1760s, but climbed to 410.6 ppm in Feb. 2019 [56, 57]. The Intergovernmental Panel on Climate Change (IPCC) forecasted that by the end of twenty-first century the CO_2 concentration would reach 590 ppm and the average global temperature could rise by 1.9 °C [58].

Biochar has been reported as one possible material for reducing the concentration of atmospheric CO_2 [23, 51]. The climate mitigation potential of biochar is based on its excellent stability, which decelerates the rate at which photosynthetically fixed C is recovered. As the possible strategy for removing CO_2 from atmosphere, biochar solution is not the only one, but notable in this regard. Woolf et al. analyzed that sustainable global implementation of biochar proposal can decrease ca. 12 % of current anthropogenic CO_2-C equivalent (CO_2-C_e) emissions, which means 1.8 Pg CO_2-C_e per year of the annual emissions (15.4 Pg CO_2-C_e), and over this course of one century, the net offset of biochar solution would be 130 Pg CO_2-C_e [23]. Furthermore, the conversion of all obtained biomass to maximize bioenergy would offset a maximum of 10 % current anthropogenic CO_2-C_e emissions. Also note that the climate-mitigation potential of biochar and bioenergy depends on the fertility of the soil amended, the C intensity of the fuel being offset and the biomass feedstocks. Higher soil fertility and coal as the fuel being offset are suitable for bioenergy production. And for all other situations the climate-mitigation potential of biochar is higher.

Significant mitigation of GHG emissions by biochar have been demonstrated in

161 short-term laboratory incubations or field conditions [59-61]. However, long-time
162 field trials are still needed. Case et al. investigated a 2-year amendment in a
163 miscanthus bioenergy crop field [62]. Biochar amendment decreased ca. 33% soil
164 CO_2 emissions and annual net soil CO_2 equivalent emissions by 37%, which was
165 lower than the data obtained in laboratory. The reduced enzymatic activity, increased
166 carbon-use efficiency and the adsorption of CO_2 onto the biochar surface may be the
167 dominant mechanisms. Compared with biomass feedstock, the biochar had contrasting
168 effects on CO_2 emissions. Hu et al. studied the impact of wheat straw and biochar
169 addition on GHG emissions in two growing seasons (15 months) [63]. Their results
170 indicated that application of biochar, as opposed to the application of biomass, would
171 decrease CO_2 and N_2O emissions. Recently, the interactions between aged biochar,
172 fresh low molecular weight carbon and soil organic carbon after 3.5 year soil-biochar
173 incubations have been reported [64]. The long term observations indicated that the
174 decline in soil C sequestration was related with the attenuated adsorption of aged
175 biochar.

176 Recent observations of the decrease of GHG emissions from biochar amended
177 soils have been utilized to prove the proposed environmental management. The
178 duration of the GHG reductions is of great importance for the inclusion of biochar
179 into future environmental management. Spokas evaluated the influences of natural
180 aging on GHG production/consumption in biochar amended soil [65]. The selected
181 biochars were aged in an agricultural field in Rosemount, MN (2008–2011). The
182 results revealed that weathering enhanced CO_2 production (3 to 10-fold compared

with fresh biochars), indicating the enhancement of microbial mineralization rate of the aged biochars. Moreover, aged biochars had no remarkable impact on the tested soil methanotrophic activity. For the three selected biochars, field aging significantly alters the GHG emissions of the biochar amended soil systems. Another interesting phenomenon was reported that potassium doped biochar increased the carbon sequestration potential by 45%. It can be translated to an increase in the estimated global biochar carbon sequestration potential to over 2.6 Gt CO_2-C(eq) yr^{-1} [66].

However, increases in CO_2 emissions after biochar addition have also been reported [67]. Sagrilo et al. studied possible interactions between native soil organic carbon (SOC) and biochar [68]. The results of 46 studies about CO_2 emissions of biochar amended soils have been combined in a meta-analysis. A statistically significant increase of 28% in CO_2 emissions has been found from biochar amended soils, indicating that the interactions between biochar and SOC accelerated the loss of SOC, thereby reducing biochar C sequestration potential. When the ratio of biochar C and SOC is greater than 2, a significant increase in CO_2 emissions was observed. However, those with a ratio less than 2 showed no significant influence of biochar application on CO_2 emissions. The ratio of biochar C and SOC provides the best predictor of CO_2 emissions after biochar addition to soil. Moreover, the albedo impact caused by biochar in the overall climate impact assessment has been studied for assessing biochar amendment comprehensively. Mayer et al. studied the impact of albedo on the suitability of biochar amendment for GHG emission mitigation [69]. A modeled biochar system using the global warming potential (GWP) characterization

factors revealed a decrease of the overall GHG mitigation benefits by 13−22% for the changing of albedo. Another interesting study was published by Gupta et al., who used biochar as carbon sequestering additive in cement mortar [70]. Compared with biochar saturated with carbon dioxide, incorporating fresh biochar in mortar mixture, the mechanical strength of the mixture can be enhanced and the permeability can be improved.

2.2. Mitigation of N_2O emissions

N_2O is the most important ozone depleting compound in atmosphere [71]. The atmospheric concentration of N_2O prior to industrialization was 270 parts per billion by volume, and its current atmospheric abundance is ~324 parts per billion by volume [72]. The main source of global anthropogenic N_2O emissions is owing to the extensive utilization of nitrogenous fertilizers. N_2O emissions are predominantly generated via N transformations in soils, while N_2O can be produced by the reaction of abiotic redox [73-75]. Rondon first reported the decrease in the emission of N_2O after biochar amendment. The emission of N_2O decreased ca. 50% for soybean and ca. 80% for grass, which grew in a barren oxisol at Colombian savanna. The proposed hypothesis elucidated that the impact of biochar on the emission of N_2O included abiotic mechanisms and biotic mechanisms (e.g., biochar liming effect [76], interaction with N [77], interaction with dissolved organic C [75, 78], impact on soil aeration [79], release of toxic/inhibitory compounds [80, 81], interactions with the soil biota [82]).

Cayuela et al. conducted a meta-analysis using 261 experimental treatments from 2007 to 2013 [73]. They found 54% reduction of soil N_2O emissions in both laboratory studies and field tests. The feedstocks, pyrolysis parameters and C/N ratios are proved to be the critical factors affecting N_2O emissions. Borchard et al. also carried out a meta-analysis. Data were compiled from 88 publications obtained from 608 observations up to May 2016. The overall reduction of N_2O was about 38%, but the N_2O emission reductions tended to be negligible after one year. Moreover, biochar amendment had the highest N_2O emission reducing effect in sandy soils and paddy soils [83].

Cayuela et al. deeply investigated the quantity of reduced N_2O emissions and the reduction mechanisms [84]. They found that biochar significantly affects denitrification, with a reduction in N_2O emissions by 10-90% in 14 different soils. By ^{15}N gas-flux method, they found a consistent reduction of the $N_2O/(N_2+N_2O)$ ratio, indicating that biochar facilitated the last step of denitrification. A pH shift in soil caused by biochar acid buffer capacity was an important aspect for the mitigation of N_2O emissions. Furthermore, biochar promotes the transfer of electrons to denitrifying microorganisms, which accelerates the N_2O reduction together with the liming effect. Ameloot et al. ran a field experiment with biochar for 7 months [85]. They found that biochar additions decreased N_2O and N_2 emissions in all cases but did not reduce the $N_2O/(N_2+N_2O)$ ratio.

N cycling can be affected when biochar is used as the soil conditioner. Addition of activated switchgrass biochar increased the abundance of microbial nitrogen

cycling gene in an aridic subsoil [86]. Similar results have been reported by Xu et al. via high-throughput sequencing of biochar amended soil microbial community [87]. The results revealed that biochar application enhanced the α-diversity and altered the relative abundances of C and N cycling related microbes. Biochar addition stimulated both nitrification and denitrification. Via redundancy analysis, the soil chemical properties were changed and they resulted in the shift of soil microbial community, thus regulating soil N_2O emissions and N cycling. Harter and co-workers also found that biochar amendment could shape the composition of N_2O-reducing microbial communities [88]. In addition to the N_2O reduction from amended soil, biochar could reduce N_2O emissions as well in composting, especially in the later stages [89]. Small amount of biochar incorporated in composting could improve N cycling by increasing the content of NO_3^--N, which indicated a higher nitrifying activity [90]. However, the opposite result was also obtained. Anderson et al. investigated the alterations of seasonal bacterial community in two years observation of N-transformations under bovine urine patches in Canterbury, New Zealand [91]. No obvious influence on the structure of microbial community over the two years was found. The proportion of denitrifiers and nitrifiers increased for large influxes of urine derived N. This is related to different N_2O production pathways in control soils, where biochars affect these processes differently. Moreover, in biochar field aging experiments, weathering negated the suppression of N_2O emission, which was originally found from the fresh biochar in laboratory incubations [65]. The cause of these phenomena and actual duration of the mitigation effect should be further investigated.

2.3. Sequestration of CH_4 in global carbon pools

The earth's atmospheric CH_4 concentration has increased by ca. 150% since 1750, and it accounts for 20% of the anthropogenic warming effect [92]. CH_4 is emitted via natural sources such as wetlands and human activities. To evaluate the actual benefits of biochar for mitigating GHG emissions, it is necessary to quantify the effect of biochar on CH_4 production from amended soils, especially in wetland, where soils are routinely drained and flooded, thus accelerating the CH_4 and N_2O emissions.

Dong et al. compared the responses of CH_4 emissions of biochar and straw applications in a paddy field experiments [93]. In a 2-year amendment, the results revealed that rice straw derived biochar was more efficient than bamboo derived biochar in the reduction of CH_4 emissions from a paddy field. Compared with direct return of rice straw, incorporating rice straw derived biochar into paddy field was able to decrease CH_4 emissions by 47.30%–86.43% during the rice growing cycle. Another biochar amendment in paddy soil revealed that a significant reduction (112.2-185.4 mg kg^{-1} dry weight soil, dws $season^{-1}$) has been observed. And this was attributed to the decrease of methanogens activity along with the increase of the pmoA gene abundance of methanotrophs and the activity of CH_4 oxidation [94]. In China, a 4-year field experiment was conducted to study the effects of biochar amendment on GHG emissions [95]. Straw-derived biochar amendment decreased annual total CH_4 emissions by 20-51% in four years. The results indicated that biochar amendment at 24 t ha^{-1} could be a consistently effective and economic measure for mitigating GHG emissions. Similar results were obtained by Proyuth et al. [96] and Chen et al. [97].

However, increases in CH_4 emissions after biochar addition have also been reported [98, 99].

Mohammadi and co-workers calculated the climate change effects of residues open burning and converting to biochars using Life Cycle Assessment. The largest contributor to the C footprint of rice was CH_4 emissions in the two systems. Compared with open burning of residues, biochar amendment decreased the C footprint of summer rice and spring rice by 14% and 26%, respectively. And the values would increase to 38% and 49% after eight years of biochar amendment [100].

Another interesting study was published by Thomazini et al., who improved the predictability for the same biochar on the GHG impact [101]. They studied the impact of hardwood biochar to reveal driving variables which affect CO_2, N_2O, and CH_4 emissions across ten different soils in US. Biochar prominently impacted the emission of N_2O (P=0.03) and CO_2 (P=0.04) in all tested soils, however no differences had been found in production/oxidation rates of CH_4 (P = 0.90). The biochar evoked changes were strongly related to the original GHG emissions in the control soils, indicating a general correspondence across different soils to the same biochar. There is no obvious change in the CO_2 mineralization rate, without regard to the effect of CO_2 released from biochar (24 μg C g_{BC}^{-1} d^{-1}). These evidences revealed the increase of CO_2 emissions was individually attributed to the release of abiotic CO_2 from biochar. The average suppression of N_2O production was 63% across all the biochar amended soils, which was also related to the initial N_2O production. This biochar has predictable impacts on GHG emissions despite the differences of soil types.

3. Biochar for contaminant management

Biochar is a versatile carbonaceous material and widely used in contaminant management [28, 102]. The efficacy of biochar in contaminant management depends on its large specific surface area, surface functional groups, and pore size distribution [103, 104]. The available peer-reviewed scientific literatures are largely focused on the adsorptive removal of contaminants. However, the effectiveness of biochars on management of different organic/inorganic pollutants needs further investigation because the soil-water system is highly complex. In this section, the latest scientific findings about the efficiency and mechanism of the contaminants adsorption and degradation by biochars in water and soil are reviewed.

As summarized and discussed in our previous work, the key parameters (e.g., feedstock type, pyrolysis temperature, and residence time) affect the physicochemical characteristics of biochars and influence the efficiency and mechanisms of contaminants removal [28]. Compared with activated carbon, the surface area of biochar is not high, which limits their application as sorbents for removal of contaminants. This is counterbalanced by its large amounts of surface functional groups [105, 106]. These multi-functional characteristics make biochar an attractive environmental adsorbent for inorganic and organic contaminants in water and soil. Meanwhile, recent publications indicate that biochar can mediate certain reactions under ambient conditions [107]. Thus, this section discusses the adsorptive removal and reactive removal of contaminants by biochar and suggests avenues for further research.

3.1. Biochar for adsorptive removal of inorganic contaminants

Inorganic contaminants, particularly heavy metals [108-111], nitrogen (N) and phosphorus (P) [112, 113], are the most urgent need for governance. According to our previous review, nearly 46 % of literatures are about the removal capacity of biochar for heavy metal ions, 13% for N and P, and 39% for organic pollutants [28]. Heavy metals originate mostly from anthropogenic sources (e.g., smelting, mining, metal finishing, leaded gasoline, battery manufacture, etc.) and pose serious health threats even at trace amounts [114]. Heavy metals are non-biodegradable and cumulative poisons [115, 116]. They appear among the main pollutants in industrial society. Sorption studies have been investigated with Pb, Hg, Cr, Cd, Cu, Al and Fe ions. Activated carbon has been deliberately applied for in situ remediation of metal contaminated soils and waters, but its sorption efficiency was not satisfactory. Moreover, in wastewater treatment, there exist regeneration problems and high expense problems. Solid biomass derived biochar may be an alternative solution.

3.1.1. Adsorptive removal of heavy metals

In recent years, literatures regarding metals removal with biochar have been published to elucidate the sorption capacities and sorption mechanisms. Table 1 summarizes the recent investigations on biochar applications for removal of metals in water and soil. Dong et al. studied the mechanisms of Hg removal by biochars obtained from Brazilian pepper at 300, 450, and 600 °C [117]. In low-temperature biochars (300 and 450 °C), Hg was irreversibly adsorbed by complexation with

carboxylic and phenolic hydroxyl groups (XPS analysis revealed that 77–69% and 23–31% of Hg was adsorbed associated with phenolic hydroxyl and carboxylic groups). For biochar obtained at 600 °C, Hg was adsorbed with a graphite-like domain on an aromatic structure. Another in-situ amendment of biochars and activated carbons for mercury and methylmercury (produced from inorganic mercury and always present at some level in mercury contaminated sediments) has been evaluated [118]. The results revealed that steam activated carbons were more efficient than biochars in the sorption of mercury and translated to modeled porewater mercury reduction of 31−73% for sediments with high native K_d values and 94−98% with low native K_d values for mercury. For the sorption of methylmercury, biochars were as effective as steam activated carbons. Cui and co-workers studied cadmium removal by biochars produced at different temperatures and their quantitative contributions [119]. The maximum sorption ability of biochar prepared at 500 °C was 188.8 mg g^{-1}. With increasing pyrolysis temperature, the contribution of metal ion exchange and surface complexation decreased from 43.3% and 24.5% to 4.7% and 0.7%, respectively. The contribution of Cd^{2+}-π interaction and precipitation significantly increased from 2.5% and 29.7% to 5.1% and 89.5%, respectively. Beesley and Marmiroli investigated the ability of biochars to immobilize and retain cadmium, zinc and arsenic from a multi-element contaminated sediment-derived soil [120]. Surface sorption of zinc and cadmium onto biochars reduces the concentrations in leachates from a polluted soil 45 and 300-fold, respectively. As for Cr^{6+}, biochar acted as both electron donor and electron shuttle for the reduction transformation of Cr^{6+} during the

379 sorption progress. -C-O and -C=O groups of biochar were the dominated electron
380 donors for the reduction of Cr^{6+}, while semiquinone-type radicals were the electron
381 shuttle for enhancing the reduction of Cr^{6+} by lactate [121].

382 As for manure-derived biochar (high-mineral biochar) and lignocellulose-derived
383 biochar (low-mineral biochar), they had different impacts on cadmium adsorption in
384 soil. The maximum adsorption capacity of the swine-manure-derived biochars was
385 10-15 times higher than that of the wheat-straw-derived biochars for the high polarity
386 and ash content of the swine-manure-derived biochars [122]. Inyang et al. studied the
387 removal of Cd^{2+}, Ni^{2+}, Cu^{2+} and Pb^{2+} by biochars derived from anaerobically digested
388 biomass [123]. Jiang and co-workers investigated the sorption of Pb^{2+} on variable
389 charge soils amended with rice-straw biochar [124]. The mobility/immobility of Cu^{2+}
390 is highly affected by the organic C content of biochar. Normally, biochars obtained at
391 500 °C or greater have a higher level of dissolved organic C content facilitating the
392 generation of soluble Cu complexes with dissolved organic C [125]. Moreover, a high
393 level of dissolved organic C would block the porous structure of biochars, decreasing
394 the sorption capacity.

395 Competitive sorption of mono and multimetal heavy metals by sesame straw
396 derived biochar was evaluated [126]. The results indicated that the maximum sorption
397 capacities (mg g^{-1}) of metal ions by sesame straw biochar were in the order: Zn (34) <
398 Cu (55) < Cr (65) < Cd (86) < Pb (102) in the isotherms of mono-metal sorption, and
399 Cd (5) < Zn (7) < Cr (21) < Cu (40) < Pb (88) in the isotherms of multi-metal sorption.
400 Another interesting phenomenon was that Cd was easily exchanged by other metals in

401 multi-metal sorption.

402 **Table 1. Biochars for remediation of metals contaminated soil and water**

Biochar type	Contaminants	Matrix	pH	Q_{max} (mg g^{-1})	Isotherm	Dominated sorption mechanisms	References
Brazilian pepper (300 °C, 2h)	Mercury	Water	6.0	24.2	Langmuir	Complexation by phenolic hydroxyl (77-69%) and carboxylic (23-31%) groups	[117]
Brazilian pepper (600 °C, 2h)	Mercury	Water	6.0	15.1	Langmuir	91% of sorbed mercury was due to the graphite-like domain on an aromatic structure	
Pine dust and phragmites (600 °C, 2h)	Mercury	Soil				Complexation with organic matter	[118]
Sesame straw (700 °C, 4h)	Lead	Water	7.0	102	Langmuir	Surface complexation with function groups, and ion exchange	[126]
Sugar beet (600 °C, 2h)	Lead	Water	5.0	40.8	Langmuir	Surface precipitation	[123]
Rice straw (300 °C, 4h)	Lead	Soil	-	-	-	Non-electrostatic mechanism via the generation of surface complexes between surface functional groups and Pb^{2+}	[124]
Bamboo (700 °C, 1h)	Cadmium	Water	5.0	154.16	Langmuir	Cd^{2+}-π interaction (81.55%), functional groups complexation (8.56%) and precipitation or cation exchange (9.89%)	[127]
Canna indica (300 °C, 2h)	Cadmium	Water	5.0	63.3	Langmuir	Increasing pyrolysis temperature, the contribution of metal ion exchange and surface complexation reduced from 43.3% and 24.5% to 4.7% and 0.7%, while the contribution of Cd^{2+}-π interaction and precipitation significantly enhanced from 29.7% and 2.5% to 89.5% and 5.1%, respectively. The co-precipitation and metal ion exchange dominated the sorption of Cd^{2+} (73-94%), and co-precipitation was the dominated mechanism of Cd^{2+} sorption on biochars derived at high temperatures (accounted for 86-90%)	[119]
Canna indica (400 °C, 2h)	Cadmium	Water	5.0	105.8	Langmuir		
Canna indica (500 °C, 2h)	Cadmium	Water	5.0	188.8	Langmuir		
Canna indica (600 °C, 2h)	Cadmium	Water	5.0	140.0	Langmuir		

Wheat straw (300 °C, 1h)	Cadmium	Soil	8.6	9.59(5% biochar)	Langmuir-Langmuir	Adsorption onto inorganic fraction of biochar and influenced by the polarity of biochars	[122]
Hardwoods (400 °C)	Cadmium Zinc	Soil	6.2	-	-	Surface sorption, enhanced pH	[120]
Ramie (300 °C, 2h)	Hexavalent chromium	Water	2.0	82.2	Langmuir	Electrostatic interactions via film and intraparticle diffusions and chemical binding by polar surface functional groups	[128]
Ramie (600 °C, 2h)	Hexavalent Chromium	Water	2.0	61.2	Langmuir		
Rice straw (100 °C, 6h)	Aluminum	Water	4.3	10.7	Langmuir	Complexation of aluminum with carboxyl and hydroxyl groups, surface sorption, and co-precipitation of aluminum with silicate particles (as $KAlSi_3O_8$)	[129]
Rice straw (400 °C, 6h)	Aluminum	Water	4.3	3.5	Langmuir		
Rice straw (700 °C, 6h)	Aluminum	Water	4.3	9.2	Langmuir		
Spartina alterniflora (400 °C, 2h)	Copper	Water	6.0	48.5	Langmuir	Surface complexation, Cπ–metal interaction, metal (hydr)oxide precipitation	[130]
Miscanthus (500 °C, 1h)	Copper	Water	6.0	15.4	Freundlich	Surface complexation	[131]

3.1.2. Adsorptive removal of phosphorus and nitrogen compounds

If biochars are used as soil amendments, it is necessary to investigate the bioavailability of nutrients, especially N and P [132]. They are plant essential nutrients and potential water pollutants. Knowledge of N and P retention and release mechanisms is needed when biochars are applied as amendments. Compared with N, P is relatively less available in soil. Chintala et al. studied the P sorption and the availability from switchgrass, corn stover and ponderosa pine wood residue derived biochar [133]. The corn stover derived biochar showed the most remarkable P sorption (79% of initial P concentration), followed by switchgrass derived biochar

412 (76%). The P sorption capacity of ponderosa pine wood residue derived biochar was
413 ca. 31%. The biochar amendments to acidic soil (4%) increased the equilibrium
414 concentration of P solution and increased the availability of sorbed P. Cornstover and
415 switchgrass derived biochars (alkaline biochars) enhanced P sorption and reduced
416 available sorbed P in calcareous soil. Similar investigation was conducted by Xu and
417 co-workers [134]. They studied the mechanisms underlying the P sorption changes.
418 Via inorganic P fractionation, it was found that biochar amendment greatly enhanced
419 the Ca-bounded P and slightly increased the Al-retained P. And biochar amendment
420 reduced the Fe-bounded P. The results revealed that the enhancement of P sorption
421 with biochar amendment was mainly caused by Ca-induced P sorption or precipitation
422 and less attributed by Al and Fe oxides. The effects of biochar on clay soil aggregate
423 stability and P sorption were investigated [135]. One sandy and two clayey soils were
424 amended with biochar. After three weeks of incubation, the biochar addition did not
425 increase the P sorption in incubated soils. But for clayey soils, biochar addition
426 enhanced the aggregate stability and induced changes in soil properties beneficial to
427 erosion control and thus reduced the loss of particulate P from soil. These effects of
428 biochar amendment were mainly dependent on the acidity of soil, which is the
429 important factor for soil amendment and soil productivity. And the proposed
430 mechanisms also include alteration of bulk density, soil water retention, plant-
431 available water and nutrient use efficiency [136]. These findings were further
432 supported by Schneider and Haderlein [137], Manolikaki et al. [138], Dari et al. [139],
433 and Wei et al. [140]. Generally, biochar can be regard as the mediator in soil

434 remediation. Biochar amendment is found to have altered P availability by changing
435 the P sorption and desorption capacities of the soils, and these biochar effects are
436 mainly dependent on soil acidity. Moreover, due to the different soil properties and
437 biochar characteristics, biochar as a source or sink for P in soil amendment depends
438 on the circumstances. Advantages from biochar amendment are likely to emerge when
439 a higher or lower concentration of soil nutrient is identified.

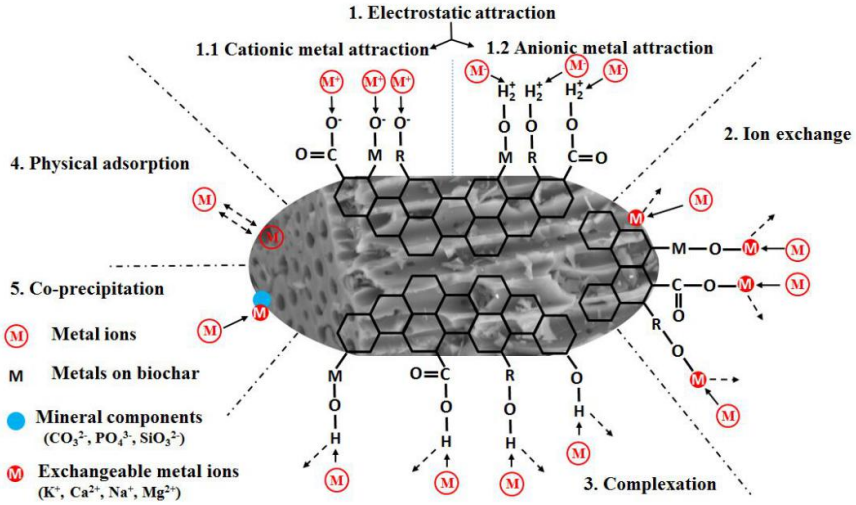
440 Literatures described the ability of biochar in retaining ammonium-N (NH_4-N)
441 and nitrate-nitrogen (NO_3-N), which decreased N losses through soil leaching [136,
442 141, 142]. Sorrenti and co-workers [136] performed a series of experiments to study
443 the potential of hardwood biochar as a source or sink for ammonium-N (NH_4-N),
444 nitrate-nitrogen (NO_3-N), potassium (K), phosphorus (P), and magnesium (Mg) in
445 solution. The results indicated that the NH_4-N, P, K and Mg concentrations in solution
446 were increased and positively correlated with the rates of biochar. However, it was
447 ineffective in the removal of NH_4-N, P, K and Mg from enriched solutions, while 52%
448 of initial NH_4-N was removed at the rate of 40 g L^{-1} biochar. In a sandy-loam soil
449 commercial nectarine orchard (Italian Po Valley), biochar amendment of 5, 15 and 30
450 t ha^{-1} were efficient in reducing the leached NH_4-N in the topsoil (0.25 m) during 13
451 months. However, independent of the rate, biochar amendment did not affect soil pH,
452 soil moisture, and the availability of soil N. And they concluded that in non-limiting
453 conditions the benefits of biochar amendment are hidden or negligible in commercial
454 nectarine orchard. Zheng and co-workers [143] studied the N loss, retention and
455 bioavailability in biochar amended soils fertilized with NO_3-N and NH_4-N via pot and

leaching tests. Fan et al. reported that bamboo biochar could effectively adsorb ammonium ion in dilute aqueous solutions. Precipitation and surface complexation were the main adsorption mechanisms [144]. Their results revealed that the mitigation of N leaching in biochar amendment soil was attributed to the enhancement of the soil water holding capacity and the sorption of NH_4^+.

The discussion above indicated the capacity of biochar in retaining ammonia. And the biochar adsorbed ammonia is bioavailable, which was proved by Taghizadeh-Toosi et al. [145]. They utilized nitrogen isotope to present that biochar adsorbed ammonia-N was stable in ambient air, but readily bioavailable when it was added in soil. Another observation by Spokas et al. added credence to the utilization of biochar as a carrier for nitrogen [146].

3.1.3. The mechanisms of biochar for adsorptive removal of inorganic contaminants

For better understanding of the removal behaviors of inorganic contaminants by lignocellulose biomass derived biochars, the study of the involved mechanisms of the sorption behavior is very necessary. The sorption behaviors of biochars for various contaminants are different and the sorption mechanisms also depend on the various properties of biochars. The proposed mechanisms for the sorption of inorganic contaminants onto biochars, especially metals, N and P, are summarized in Fig. 4.



475

476 **Fig. 4. The proposed mechanisms of biochar for remediation of inorganic**
477 **contaminants**

478 The surface functional groups, especially O-containing hydroxyl, carboxyl and
479 phenolic functional groups, have strong sorption capacity with inorganic contaminants,
480 e.g. surface complexation, ion-exchange, and electrostatic attraction. As discussed
481 earlier (Table 1), Hg, Pb, Cr, Cd, Cu, and Al ions are adsorbed via interactions with
482 surface functional groups. And these effects can be confirmed by the changes of the
483 surface groups before and after the sorptions [147]. The porous structure and surface
484 area of biochar also affect the sorption of inorganic contaminants. However, these
485 properties of biochar seem to have less influence on metals sorption than O-
486 containing surface functional groups [117, 148, 149]. Moreover, the mineral
487 components of biochars also play crucial roles in the sorption process [122, 150].
488 CO_3^{2-} and PO_4^{3-} originated from the feedstock serve as additional sorption sites,
489 contributing to high sorption capacity for metals. As for NH_4^+, the sorption process
490 was controlled by the cation exchange capacity of biochar and NH_4^+ sorption was

491 correlated positively with cation exchange capacity but negatively with surface area of
492 biochar [151]. Compared with the sorption of NH_4^+ by biochars, the NO_3^- and
493 phosphate sorption capacity are relatively lower. The NO_3^- and phosphate sorption
494 mechanisms mainly include partition and anion exchange, and the sorption capacity of
495 biochar is affected by soil water-holding capacity and its surface area. [133].

496 **3.2. Biochar for adsorptive removal of organic contaminants**

497 Biochar has been applied to the remediation of organic contaminants [152-154].
498 Typical organic contaminants found in waters and soils include dyes [155-158],
499 pesticides [159, 160], antibiotics [161-165], herbicides [166-168], environmental
500 endocrine (e.g. polycyclic aromatic hydrocarbons [169], polychlorinated biphenyls
501 [170], phthalic acid esters [171], etc.) and others [172-175]. Large proportion of
502 research focused on the remediation of organic contaminants in aqueous solution,
503 while a few studies were applied to soils (Table 2).

504 **3.2.1. Adsorptive removal of organic contaminants**

505 The textile-dyeing industry produces large amounts of wastewater in the process
506 of dyeing and finishing. Sun et al. prepared anaerobic digestion residue, eucalyptus,
507 and palm bark biochars for the sorption of methylene blue (MB) [176]. The results
508 were fitted by the Langmuir isotherm model, with a maximum monolayer sorption
509 capacity of 9.5 mg g^{-1} for anaerobic digestion residue biochar. Innovative
510 technologies incorporating engineered nanomaterials into biochar would improve the
511 functions of biochar for wastewater treatment. Hybrid multi-walled carbon nanotube-

512 coated biochars have been used for the treatment of MB. The results revealed that
513 electrostatic attraction was the main sorption mechanism for MB and diffusion
514 controlled its rate [177].

515 Jin and co-workers studied the properties of wheat straw, rice straw and swine
516 manure biochar amended soils and the removal of isoproturon, atrazine and
517 imidacloprid. The increased sorption capacity was due to the increase in surface area
518 and organic carbon content as well as the decrease in hydrophobicity. However, the
519 sorption capacity of the biochar amended soils would exceed or be below of predicted
520 values without assuming a cross-effect between biochar and soil [178]. Pignatello and
521 Xiao investigated the interactions of triazine herbicides with biochar. The results
522 revealed that the difference in polar effects was caused by π–π electron donor–
523 acceptor interactions on the polyaromatic surface. And they found that mesoporosity
524 was critical, that the sorption rate was affected by the size and charge of solute
525 molecular, that steric bulk suppressed equilibrium adsorption, and that π–π electron
526 donor–acceptor forces played an important role in triazine polar interactions with
527 biochar [179]. The effects of biochar addition on the sorption and desorption of
528 herbicides and herbicide metabolites (e.g., isoproturon, bentazone, pyraclostrobin,
529 aminocyclopyrachlor, etc.) in soils have been evaluated by Cabrera et al. [180],
530 Dechene et al. [181], and Eibisch et al. [182].

531 Antibiotics are a type of antimicrobial drug used for the treatment and prevention
532 of bacterial infections. Sulfamethazine is a highly frequently used veterinary drug and
533 its concentration is up to 900 mg kg^{-1} in manure. Pignatello and his team studied the

534 sorption of sulfamethazine in biochar-amended soil and the speciation of the ionizable
535 sulfamethazine on biochar [183]. The results revealed that only highly surfaceous,
536 carbonaceous biochars could be helpful for the stabilization of soil contaminants such
537 as sulfamethazine and aging may weaken the effectiveness of native biochar in
538 adsorbing such contaminants within soils and/or sediments. Our team combined
539 biochar with emerging nanotechnology to synthesise hybrid carbonaceous
540 nanocomposites and studied its sorption capacity for sulfamethazine in aqueous
541 solution. The primary sorption mechanisms for sulfamethazine include partition,
542 hydrogen bonding, and π-π interaction. We also studied the influences of harsh aging
543 on the sorption of sulfamethazine in the presence of soil and/or biological and
544 chemical aging [103]. Jia et al. studied the influences of pH and metals on the
545 oxytetracycline sorption by maize straw biochar [184]. Surface complexation via π–π
546 interaction and metal bridging were the dominant sorption mechanisms and cation
547 exchange also played a role in the sorption. The sorption of antibiotics like
548 tetracycline [185], levofloxacin [186], fluoroquinolone [187], sulfamethoxazole [188],
549 and sulfamethazine [189] have been studied.

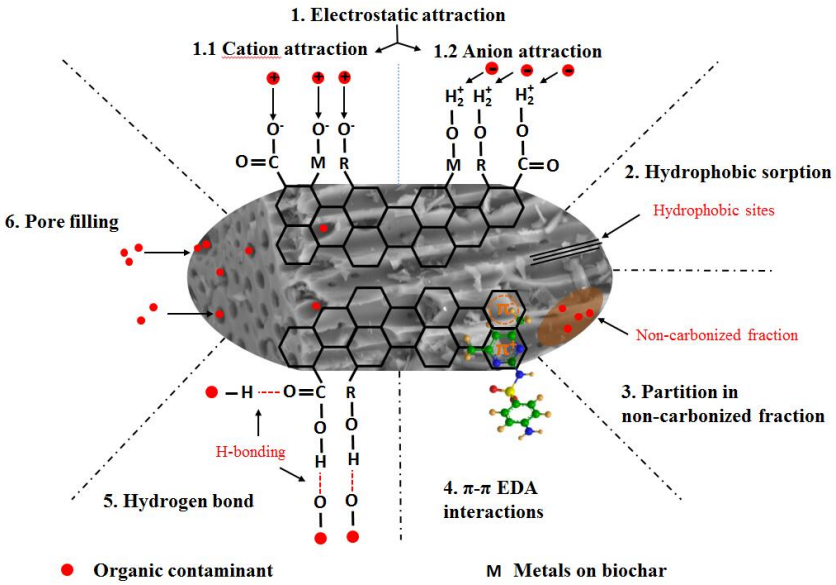
550 Hale et al. studied the sorption of pyrene, the best polycyclic aromatic
551 hydrocarbon for the prediction of total and freely dissolved concentration of other
552 polycyclic aromatic hydrocarbons, to activated carbon and biochar [169]. Moreover,
553 they investigated the effects of biological, physical, and chemical aging as well as soil
554 addition on the sorption of pyrene. The sorption of pyrene onto biochar was less
555 affected by soil. Biochar and activated carbon maintained a high sorption capacity

both in the presence of soil and during the aging process. Sun and co-workers [190] investigated the influence of deashing on the structure of biochar and its sorption behavior of phenanthrene. As for polychlorinated biphenyls, the in-situ remediation using biochar and activated carbon has been carried out [191]. They found that the concentrations of polychlorinated biphenyls in the root tissue of Cucurbita pepo were reduced by 72%, 64%, and 74%, with the addition of 2.8% Burt's biochar, Blue Leaf biochar, and activated carbon, respectively. Beless et al. compared the efficacy of biochar, activated carbon, carbon nanotubes, graphene and grapheme oxide for the sorption of eleven polychlorinated biphenyl congeners in aqueous solution [170]. Sorbent-water distribution coefficients (Ks) calculation revealed that activated carbon was superior to grapheme oxide, grapheme, carbon nanotubes and biochar for the sorption of polychlorinated biphenyls by an average of 1.1, 1.1, 1.3, and 2.5 orders of magnitude, respectively. A pot test was carried out to study the effects of straw biochar and bamboo biochar on the bioavailability of di-(2-ethylhexyl) phthalate (DEHP) in two soils. The results revealed that biochar amendment reduced the concentration of DEHP in shoots grown in the low organic carbon content soil, but there was no significant effect in soil with high organic carbon content.

3.2.2. The mechanisms of biochar for adsorptive removal of organic contaminants

The adsorption mechanisms of biochars for organic contaminants include pore-filling, electrostatic interaction, hydrogen bonds, hydrophobic effect, and π–π electron–donor–acceptor interactions (Fig. 5). The various mechanisms depend on the

578 various properties of biochars, the types of organic contaminants, and the
579 physicochemical properties of amended medium.



580

581 **Fig. 5. The proposed mechanisms of biochar for remediation of organic**
582 **contaminants**

583 The surface of biochar is heterogeneous including non-carbonized and
584 carbonized fractions, and the sorption mechanisms of non-carbonized and carbonized
585 fractions of biochar are different. The sorption of organic contaminants is caused by
586 partition into the non-carbonized phase and adsorption onto the carbonized fraction
587 [103]. Zhang et al. studied the mechanism of sulfamethazine sorption onto biochar-
588 based carbonaceous material [103]. The primary mechanisms were proposed
589 including partition caused by Van der Waals forces and adsorption caused by
590 hydrogen bonding and π–π interactions. The porosity of biochar makes it an excellent
591 sorbent via pore-filling. The sorption of organic compounds onto biochars is related to
592 the surface properties and the sorption capacity is directly proportional to the surface

area of micropore [192]. This adsorption mechanism was also reported by Teixido et al. [161] and Gao et al. [193] who studied the sorption of sulfamethazine and phthalic acid esters. The results of various experiments revealed that electrostatic attraction was an important mechanism for the sorption of organic contaminants with other contributed sorption mechanism [177, 194]. Moreover, several other mechanisms including hydrophobic effect, π–π interactions and hydrogen bonds are the involved sorption mechanisms [185].

Table 2. Biochars for organic contaminants treatment in soil and water

Biochar type	Contaminants	Matrix	pH	Q_{max} (mg g^{-1})	Isotherm	Dominated sorption mechanisms	References
Anaerobic digestion residue (400 °C, 0.5 h)	Methylene blue dye	Water	7.0	9.5	Langmuir	Monolayer adsorption	[176]
CNT modified bagasse (600 °C, 1 h)	Methylene blue dye	Water	7.0	6.2	Langmuir	Electrostatic attraction	[177]
Peanut straw (350 °C, 4 h)	Methyl violet	Water	9.19–9.41	104.4	Langmuir	Electrostatic attraction; carboxylate and phenolic hydroxyl groups; surface precipitation	[194]
Rice, wheat straw (600 °C, 1 h)	Imidacloprid, isoproturon, and atrazine	Soil	8.6	-	-	Organic carbon content and surface area as well as the decreased hydrophobicity	[178]
Pig manure (700 °C, 2 h)	Carbaryl	Water	6.5	-	-	Hydrophobic effect, pore-filling and π–π interactions	[195]
Hardwood (400 °C, 2 h)	Triazine herbicides	Water	7.4	-	Freundlich	π–π electron donor–acceptor interactions	[179]
Beech wood	Polar herbicides and	Soil	5.4	-	-	Higher organic matter	[181]

(550 °C)	herbicide metabolites					content	
Corn digestate, miscanthus, andwoodchips (750 °C, 45 min)	Isoproturon	Soil	-	-	-	Surface sorption as well as by diffusion and subsequent occlusion in micropores	[182]
Wood chips (750 °C, 45 min)	Aminocyclopyrachlor, bentazone and pyraclostrobin	Soil	6.0	-	-	High surface areas and low dissolved organic carbon contents	[180]
Hardwood (600 °C,)	Sulfamethazine	Soil	7.4	-	-	Surface area and organic carbon content	[161]
CNT modified rice straw (600 °C, 2 h)	Sulfamethazine	Water	5.0	79	Freundlich	Partition, hydrogen bonding and π-π interactions	[103]
Maize straw (300°C, 1.5 h)	Oxytetracycline	Water	5.5	7	Freundlich	π–π interactions and metal bridging	[184]
Methanol modified biochar (500°C)	Tetracycline	Water	2	-	-	π–π interactions and hydrogen binding	[185]
Wood chip (600°C)	Levofloxacin	Water	6.5	7.72	Langmuir	Hydrophobic interaction	[186]
Corn stover (600°C, 20 min)	Pyrene	Water	-	-	-	Nano-porosity	[169]
Softwood (450°C, 2 h)	Polychlorinated biphenyls	Soil	7.7	-	-	Sorption	[191]
Poplar leaves (300°C, 2 h)	Dibutyl phthalate	Young leachate	5.0	26	Freundlich	Porosity and organic carbon contents	[193]
Poplar leaves (300°C, 2 h)	Dibutyl phthalate	Old leachate	7.5	24	Freundlich		
Straw biochar (500°C, 3 h)	Di-(2-ethylhexyl) phthalate	Soil	5.8-6.0	-	-	Organic carbon content	[196]

3.3. Biochar for reactive removal of organic contaminants

Recent recognition of biochar as versatile media for organic contaminants

603 degradation. Biochar, as other pyrogenic carbonaceous matters, is able to promote
604 electron transfer, mediate some certain reactions, and generate reactive oxygen
605 species (ROS). Biochar for mediated or catalyzed removal of organic contaminants
606 and the underlying mechanisms still need further research. The aim of this section is
607 to summarize the existing research findings and suggest the research needs.

608 **3.3.1. Reactive removal of organic contaminants**

609 Previous studies regarded biochar as a versatile and cost-effective adsorbent. The
610 latest studies prove that biochar can promote the long-range electron conduction
611 between molecules and molecules/microbes, and facilitate local redox reactions and
612 hydrolysis reactions [107]. Recent research has proved that biochar can activate H_2O_2
613 to produce ·OH, which is frequently used for the treatment of environmental
614 contaminants. Biochar contains persistent free radicals (PFRs), typically ~10^{18}
615 unpaired spins/gram. And PFRs are the main contributor to the generation of ·OH.
616 Single-electron transfer from PFRs to H_2O_2 has been proposed as the H_2O_2 activation
617 mechanism by biochar [197]. Yang et al. studied the degradation of *p*-Nitrophenol
618 mediated by biochars [198]. The results revealed that ·OH could attack *p*-Nitrophenol,
619 and *p*-Nitrophenol contact with PFRs could be an important contribution to the
620 degradation. Huang et al. reported the important role of biomass types and its
621 compositions on the formation of PFRs in biochar. It was found that the amounts of
622 PFRs in biochar decreased sharply with the decrease of the initial phenolic
623 compounds and metals existing in biomass, and the effect of metals contents on PFRs
624 formation was much greater than that of phenolic compounds contents [199]. Similar

results were reported by Yang et al. [200]. Hydroxyl radical generated by the combination of biochar and H_2O_2 is able to degrade organic contaminants. The reported examples include *p*-nitrophenol [200], diethyl phthalate [201], sulfamethazine [202], 1, 3-dichloropropene [203], 2-chlorobiphenyl [197], polychlorinated biphenyls [197], and alachlor [204].

Moreover, biochar can transfer an electron to peroxymonosulfate ion ($HOOSO_3^-$), forming sulfate radical ($SO_4^{\cdot-}$), which is also an efficient oxidant for the degradation of organic contaminants. Compared with ·OH (1.9-2.7 V), $SO_4^{\cdot-}$ has a higher redox potential (2.5-3.1 V) and stability [205]. Biochar is a low-cost, efficient, and environmentally friendly activator, when it is used to activate persulfate to degrade organic contaminants. Fang et al. found that biochar could activate persulfate to produce $SO_4^{\cdot-}$, and the catalytic ability of biochar for the degradation of polychlorinated biphenyls was evaluated [206]. The type and concentration of PFRs determined the activation of persulfate by biochar and the results indicated that superoxide radical anions account for almost one third of $SO_4^{\cdot-}$ generation. Another removal mechanism was also reported. Persulfate activated by rice straw biochar for aniline degradation was studied [207]. The results revealed that the predominant reactive species responsible for aniline degradation might be holes instead of ·OH and $SO_4^{\cdot-}$. Biochar combined with Fe_3O_4 [208], $MgFe_2O_4$ [209], and nanoscale zero-valent iron [210] were applied to activate persulfate for the removal of organic contaminants. Recently, the photocatalytic potential of biochar, including pyrochar and hydrochar, has been reported. Fang et al. studied the photogeneration of reactive oxygen species

generating from biochar suspension [211]. It was found that diethyl phthalate was degraded and mineralized in biochar suspension under UV and simulated solar lights. Biochar matrix contributed for 63.6%-74.6% of ·OH and 10%-44.7% of $^1O_2$ formation, and dissolved organic matter derived from biochar accounted for 3.7%-12.5% of ·OH and 46.7%-86.3% of $^1O_2$ formation. Zhang and his group investigated the photochemistry of hydrochar [212]. Compared with pyrochar derived from the same feedstock, hydrochar was able to produce much more H_2O_2 and ·OH under daylight irradiation, which increased the degradation rate of sulfadimidine 6-fold more than that found without light. A series of characterization tests indicated that the higher ROS generation of hydrochar under daylight irradiation contributed from the abundant photoactive oxygenated functional groups. Compared with the traditional metal catalysts, activated carbon, and carbon nanomaterials, biochar is sustainable and versatile. It is of great significance to fully explore the reactivity of biochar. In addition, biochar based composite has been fabricated as catalyst in photo-Fenton reaction, and 93% of 40 mg L^{-1} tetracycline removal was obtained in 2 h in near neutral pH [213].

Another interesting phenomenon has been reported. Biochar can act as electron shuttles between contaminants and soil microorganisms to improve the microbial degradation [214]. Changes in sorption of atrazine [215], pentachlorophenol [216], thiacloprid [217], and petroleum hydrocarbons [218] and microbial transformation in biochar amended soils have been reported. Biochars produced at different pyrolysis temperatures have different effects on biochar-mediated microbial remediation.

669 Amendments with biochar produced at 300 °C promoted the biodegradation of
670 thiacloprid by increasing the microbe abundance and nitrile hydratase activity.
671 However, biochar produced at 500 and 700 °C inhibited the biodegradation by
672 decreasing the thiacloprid availability and changing the activity of nitrile hydratase
673 [217]. Kong et al. found that biochar amendment could accelerate the biodegradation
674 of polycyclic aromatic hydrocarbon (PAHs), which could be attributed to the biochar
675 benefit making the amended soil a better habitat for soil microorganisms [219]. The
676 combined use of biochar and compost for reducing PAHs has systematically
677 investigated. The sorption coefficients for organic contaminants including
678 phenanthrene, fluorene, carbazole, dibenzofuran, dibenzothiophene, and pyrene in
679 soils increased tenfold with 10% compost addition and a hundredfold with the adding
680 of 5% biochar. The increase of PAH degradation rate was probably due to the
681 introduction of exogenous microorganism [220]. An investigation studied the
682 combined effects of root exudates and biochar on the degradation of PAHs and the
683 microbial community structures in amended soil. The results indicated that a
684 synergetic effect of biochar and oxalic acid has been occurred on the shifts in
685 microbial community structures and on the degradation of PAHs, especially for high-
686 ring PAHs [221]. Soils and amended biochars are complex systems, and investigating
687 the key roles of biochar on microbial activity and the multiple affecting factors are
688 essential.

689 **3.3.2. The mechanisms of biochar for reactive removal of organic contaminants**

690 Biochar contains resonance-stabilized radicals (semiquinones, phenoxyls, and

cyclopentadienyls), and also referred as PFRs, which can be formed via the thermal decomposition of catechols, phenols, hydroquinones, etc., in the presence of metal oxides [222]. The types and concentrations of PFRs can be detected via electron paramagnetic resonance (EPR). And the spectral splitting factor of EPR (*g*-factors) can be used to identify the types of EPR. Normally, the *g*-factors of persistent carbon-centered free radicals are less than 2.0030, while *g*-factors of persistent oxygen-centered free radicals are more than 2.0040. *g*-factors of persistent carbon-centered free radicals connected with an adjacent oxygen atom are usually in the range of 2.0030-2.0040 [223]. PFRs are formed via the chemisorption of substituted aromatic molecular adsorbates on the metal cation center. For example, at temperatures from 150 to 400 °C, the formed PFRs on Fe_2O_3/SiO_2 surface were phenoxyl-type radicals with *g*-factors of 2.0024-2.0040 and second semiquinone-type radicals with *g*-factors of 2.0050-2.0065 [224]. Therefore, *g*-factors can be used to determine whether PFRs are oxygen-centered or carbon-centered free radicals. The formation of PFRs occurred when fresh biochars are cooled and exposed to air. C-centered dangling bonds will combine with dioxygen, non-dissociatively or dissociatively, producing both valence-saturated and radical products [107]. Many radicals can persist for days or longer due to extensive π-delocalization or their inaccessibility in the matrix.

The core issue of biochar for reactive removal of organic contaminants is PFRs. Thus, the manipulation of PFRs for the desired purpose is meaningful. The effects of metals (Cu^{2+}, Fe^{2+}, Ni^{2+}, and Zn^{2+}) and phenolic compounds loaded on biomass on the formation of PFRs in biochar was reported [206]. The results indicated that metals

713 and phenolic compounds could increase the concentrations of PFRs and change the
714 types of PFRs. Normally, the concentration of PFRs in biochars is ~10^{18} to 10^{19}
715 spins/g [197, 225]. The feedsocks and pyrolysis temperature also affect the formation
716 of PFRs. Increase of the pyrolysis temperature could enhance the PFRs intensity and
717 decrease the oxygen-centered/carbon-centered free radicals ratio. These results
718 revealed that manipulating pyrolysis temperature, the amount of metals and phenolic
719 compounds might be an efficient way to regulate PFRs and provided evidence to
720 elucidate the PFRs formation mechanism.

721 The mechanisms of biochar for reactive removal of organic contaminants are
722 shown in Fig. 6. There are two main mechanisms contributing to the reactivity of
723 biochar. PFRs can degrade organic contaminants directly. The direct contact with
724 PFRs on biochar both in water and soil is a common process in organic contaminants
725 degradation [198]. Biochar also exhibited excellent reactivity or catalytic activity to
726 activate oxidants, including H_2O_2, persulfate, O_3 and O_2 via the direct electron transfer
727 to generate ROS for reactive removal of organic contaminants [226]. Yang et al.
728 reported that the induced ROS resulted in about 20% of *p*-nitrophenol degradation,
729 and about 80% of *p*-nitrophenol directly reacted with PFRs [198]. Due to lack of the
730 support of publications, it is difficult to review the reactivity and selectivity of PFRs
731 in biochar. Moreover, previous investigations have demenstrated that biochars also
732 have catalytic redcution ability [227].

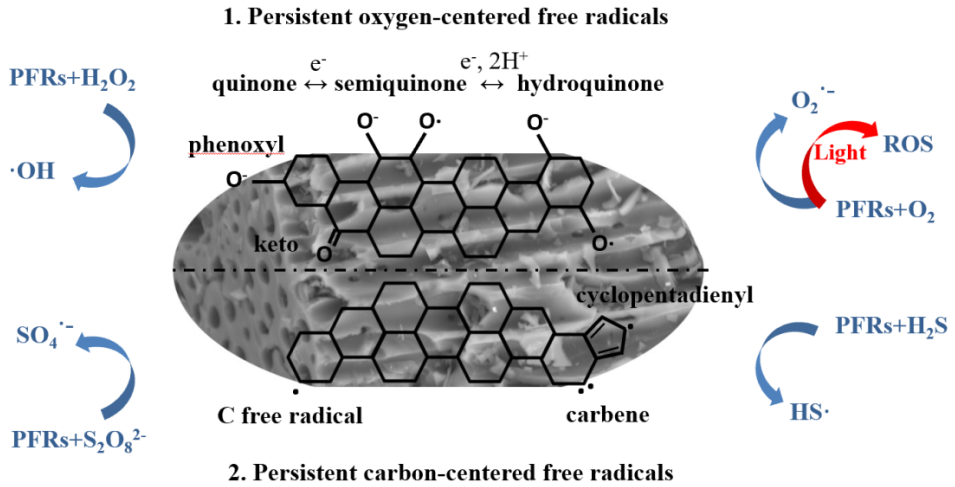
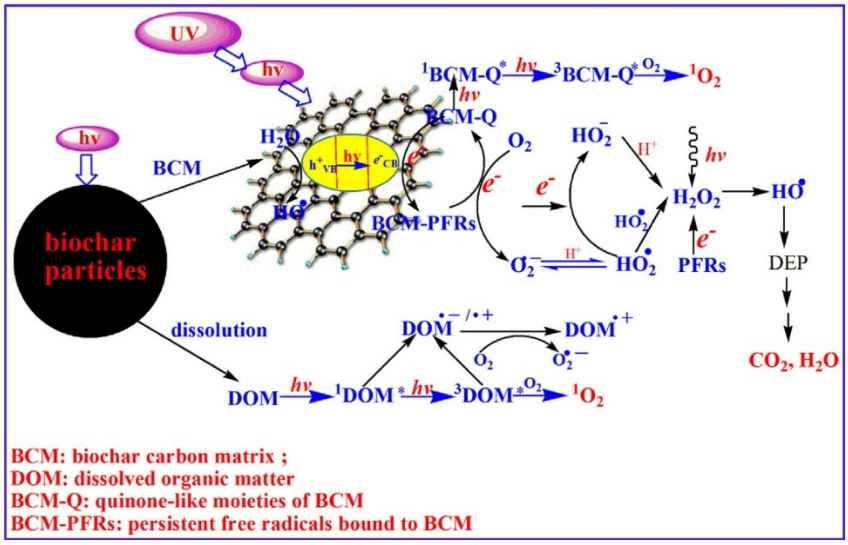


Fig. 6. The proposed mechanisms of biochar for reactive removal of organic contaminants

Recently, another interesting phenomenon is the enhanced reactivity of biochar under daylight irradiation. Zhang and his colleagues reported that comparing with pyrochar derived from the same feedstock, hydrochar could generate much more ·OH and H_2O_2 under daylight [212]. The results indicated that the higher reactive oxygen species generation ability of hydrochar was attributed to its photoactive surface oxygenated functional groups. Almost the same period, Fang et al. found that biochar suspension could generate reactive oxygen species and degrade diethyl phthalate under UV and simulated solar lights [211]. ·OH and $^1O_2$ were found to be the dominant ROS, which worked for the degradation. PFRs and quinone-like structure of biochar carbon matrix were the main factors affecting the formation of hydroxyl radicals and singlet oxygen under light. Detailed ROS generation pathways are shown in Fig. 7. The dissolved organic matter of biochar contributed to the formation of ·OH and $^1O_2$ by photogenic electron transfer. Quinone-like parts of biochar could form

749 excited triplet states under UV and also induce $^1O_2$ formation. Moreover, UV
750 promoted the formation of biochar PFRs, which could form $O_2^{\cdot-}$, further yielding
751 H_2O_2. H_2O_2 would decompose to form ·OH via photo-Fenton reaction and biochar
752 PFRs activation.



753

754 **Fig. 7. The proposed mechanisms for ROS formation from biochar suspension**
755 **under light [211]. Adapted and reprinted from ref. [211]. Copyright 2017**
756 **Elsevier.**

757 Biochar amendment also changes soil pH, water retention, dissolved organic
758 matter, and nitrogen and phosphorus levels, which further influences the community
759 structures, microbial activity, and soil enzyme activities. Ultimately, it affects
760 microbial degradation of soil contaminants [217]. The possible mechanisms of
761 biochar-mediated microbial degradation include two aspects. On the one hand,
762 biochar contains PFRs which assist biochar as an electron shuttle to enhance the
763 electron transfer between soil microorganisms and contaminants, thus accelerating the

764 degradation of organic contaminants and the removal of heavy metals. On the other
765 hand, biochars provide a suitable living microenvironment (adequate nutritions and
766 free from predators) and prevent the harsh environmental changes including soil pH
767 and soil physical properties (water holding capacity and aggregation) to ensure the
768 microbial growth [228].

769 4. Negative aspects of biochar application

770 Any strategy for extensively adopting of biochar amendment is constrained by
771 the lack of large field scale data on soil quality, crop response and environmental
772 impact. Influences of biochar application as a strategy for environmental management
773 are often inconclusive or even contradictory. Biochar application is extensively
774 regarded as a potential strategy for carbon sequestration and enhancement of crop
775 yield, mitigation of climate change, and improvement of soil quality. In 2014,
776 Mukherjee and Lal [50] reviewed the negative aspects of biochar amendment on crop
777 yield, soil quality, and associated financial risk. Besides, almost there is no review to
778 compile the negative aspects of biochar amendment. In this section, we focus on the
779 possible negative aspects of biochar application in environmental management.

780 4.1. Contamination originated from biochar

781 Biochar as amendment has been widely recognized, but relatively little attention
782 has been focused on the contamination originated from biochar, especially PAHs and
783 heavy metals. The concentrations of coproduced PAHs have been reported in the
784 order of 1 to 100 mg kg^{-1} for the sum of the 16 USEPA defined PAHs.[229] Moreover,

785 biochar produced from sewage sludge and tannery residue generally contained heavy
786 metals at high levels (e.g., Cu, Cr, Zn, etc.) [230]. Thus, high levels of biochar
787 amendment must take these into consideration and need further assessment.

788 The feedstock type and pyrolysis temperature affect the composition and
789 concentration of PAHs which are formed in the pyrolysis process [231]. The
790 properties of biochars determine their strong sorption of PAHs and high sorption
791 coefficients. Suitable concepts and methods for assessing the solvent extractable
792 PAHs in biochar have been reported [229, 231-234]. Keiluweit et al.[231] quantified
793 eleven unsubstituted three- to five-ring PAHs and alkylated forms of anthracene and
794 phenanthrene in wood and grass biochars produced in a temperature range of 100 to
795 700 °C. And they found that the concentrations of solvent extractable PAHs at 400
796 and 500 °C are greatly higher than those obtained at lower and higher temperatures.
797 The maximum extractable PAH for grass was 22 μg g^{-1} at 500 °C, which was greatly
798 higher than that for wood (5.9 μg g^{-1}). Meanwhile, it has been reported that biochars
799 obtained at 700 °C or greater could generate heavily condensed PAHs [235]. Further
800 research was conducted by Mayer and co-workers[232]. They assessed cyclodextrin
801 extractions, tenax extractions, sorptive bioaccessibility extractions, contaminant traps,
802 and equilibrium sampling for the determination of PAHs. But the results revealed that
803 none of the methods was suitable for the direct measurement of the readily desorbing
804 fractions of PAHs in the tested biochars. The high sorption capacity of biochar may
805 limit the leaching of PAHs from soils, but it depends on the biochar types and the soil
806 microbial catabolism. Previous studies reported that the conventional pollutants of

PAHs, heavy metals, furans, and dioxins remained at relatively safe concentration ranges due to the adsorption capacity of biochar. However, the latest study revealed that application of biochar to soils might result in human cancer risk because of the exposure of PAHs [236]. Field-scale and greenhouse-studies evaluated 35 commercial and laboratory biochars, and the bioavailable and total PAHs in biochars were in the range of below detection limits to 2792 μg kg^{-1} and 638 to 12347 μg kg^{-1}, respectively. The health risk assessment was carried out with the benzo[a]pyrene toxic equivalency quotient and the incremental lifetime cancer risk (ILCR) to investigate the exposure risk for human via ingestion of PAH-contaminated vegetables. The value of ILCR (above 10^{-6}) indicated a risk to human health. Thus, biochar for field-scale application should be pretreated to remove PAHs.

PFRs exist significantly in pyrogenic carbonaceous materials, including biochars. As discussed in the section of biochar for reactive removal of organic contaminants, PFRs can be used to degrade organic contaminants. However, PFRs are considered to be a new class of pollutants [237]. PFRs are highly stable and persistent. The toxicological effects of PFRs arise from the molecular byproducts and more importantly from reactive oxygen species, which are generated from the catalytic cycling of PFRs. Xing and his team detected the PFRs in biochars and determined their ability to inhibit the germination and growth of rice, wheat and corn seedlings [238]. The strong ·OH induced by PFRs significantly inhibited the germination and growth of tested plants. But inhibition of germination and the damage of plasma membrane were not obvious for biochar with low PFRs. Meanwhile, the opposite

voices are also often reported. Using the model organism *Caenorhabditis elegans*, Lieke et al. put forwards a new perspective that the risks of biochar PFRs triggered neurotoxicity in *Caenorhabditis elegans* are overlooked [239].

4.2. Negative alteration to soil properties

The improvement of soil physicochemical properties as reviewed above via biochar amendment includes the increase of soil surface area, water holding capacity, cation exchange capacity, availability of plant nutrients, and the reduction in soil acidity. Meanwhile, negative alteration to soil properties was also observed.

Soil pH is critical to soil properties. A meta-analysis has been conducted and the results revealed that the greatest positive influences of biochar probably occur in acidic and neutral soils, indicating that liming effect of biochar may be the dominated mechanism [240]. However, biochar amendment may also have undesirable soil properties alteration. The feedstock type and pyrolysis temperature affect the composition of biochar and also affect the pH values of biochars. As a rule, biochars derived from plant feedstock tend to have lower pH than biochars pyrolysed from manures or animal biomass [27, 241, 242]. Alkaline biochars may limit the availability of specific soil nutrient and have negative influence on soil cation exchange capacity. Lee et al. [243] found that pH would influence cation exchange capacity, ranging from -10 to 30 cmol kg^{-1} with the pH increasing from 5.0 to 8.5. The biochar cation exchange capacity was related to the increase of O:C ratio, which revealed the content of oxygen-containing functional groups [244, 245]. High-

850 temperature biochars, which have large amounts of condensed aromatic rings and
851 lower content of open chain or rings available for oxidation, may not increase the
852 cation exchange capacity of amended soils in a few months [246].

853 As for soil physical property changes (water holding capacity, surface area,
854 aggregation) with biochar application, negative results were also reported [247, 248].
855 For example, Herath et al. [249] studied the influence of biochar on the physical
856 properties of an andisol and an alfisol. The results revealed that up to 11.3 Mg ha^{-1} of
857 maize stover derived biochar (350-550 °C) did not increase the available water
858 capacity in amended soils, even after incubation for nearly 10 months. And it was
859 caused by the clogging of micropores by mineral/ash. In general, biochar improves
860 soil porosity. However, there are data proving the opposite. In a degraded Crosby silt
861 loam soil (18 m^2 g^{-1}), biochar (214 m^2 g^{-1}) amendment with an application rate of 7.5
862 Mg ha^{-1} did not significantly increase the surface area over four months [247]. This is
863 probably attributed to the clogging of pores caused by the stimulation of microbial
864 activity. Moreover, aged biochars usually have higher surface area for various
865 interaction types with soils [50]. The effect of biochar on soil aggregation is disputed
866 [250]. An increase in soil aggregate sizes as a result of an increase in soil organic
867 carbon when biochar is applied to soil has been observed [251]. And the reverse trend
868 has also been reported [252, 253]. Fungo et al. [254] studied the aggregate size
869 distribution in a biochar-amended tropical Ultisol. They found that biochar alone did
870 not affect a mean weight diameter but applied with either T. diversifolia or urea
871 increased mean weight diameter by 34 ± 5.2 μm (8%) and 55 ± 5.4 μm (13%),

872 respectively. The results indicate that biochar is stored predominantly as free
873 particulate organic carbon in the clay and silt fraction and promotes the movement of
874 native soil organic carbon from larger-size aggregates to smaller-sized fraction in a
875 two-year test.

876 **4.3. Negative alterations to soil biota**

877 Biochar amendment has been proved to have various effects on the alterations of
878 soil physicochemical properties including positive and negative effects as detailed
879 above. Soil biota is inevitably affected and an unambiguous identification of the
880 interactions between biochar and soil biota should be sufficiently investigated.

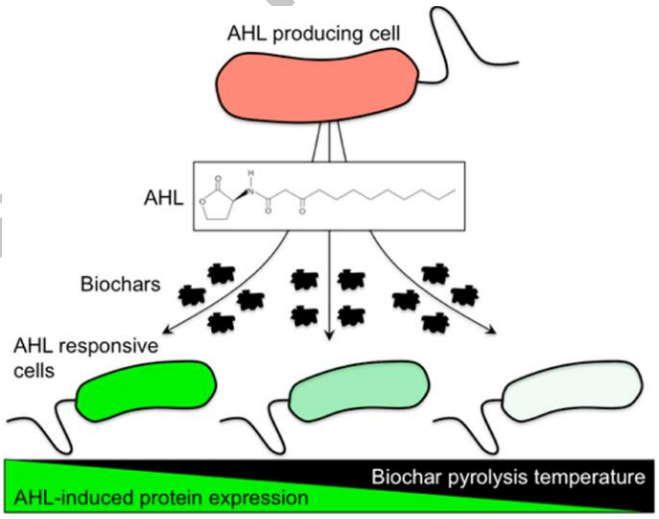
881 Biochar is devoid of biota when it is produced. However, inoculation with
882 microbes would happen and it may be inadvertently added to ecosystem, which would
883 change the native soil biota [21]. Biochar is not always beneficial to the abundance of
884 soil biota and unfavorable changes of increase in pathogenic microbes and decrease in
885 beneficial soil-biota may also occur [21]. The effect of biochar amendment on the
886 transport of *Escherichia coli* through a sandy soil has been investigated [255]. Poultry
887 litter derived biochar (350 °C) enhanced the transport of two of the *Escherichia coli*
888 isolates, whereas no obvious differences in the transport were found between 350 °C
889 and 700 °C biochar amendments for one *Escherichia coli* isolate. Biochar (700 °C)
890 with a 2% (w/w) rate of application had no influence on its transport behavior. But
891 with a rate of 10%, a significant decrease (five orders of magnitude) in the quantity of
892 *Escherichia coli* transporting through the soil has been found. Mycorrhizal fungi, an

893 important beneficial soil biota for plant health in soils, can be decreased by the
894 addition of biochar across several different soils [256]. Hol et al. reported that the
895 transient negative effects of biochar on plant growth were observed [257]. Flowering
896 was delayed in the biochar amended plots. The change of pH in biochar amended soil
897 and toxic effects caused by salt, heavy metal, and PAHs content in biochar were the
898 main reason. Another interesting observation was reported by Mickan et al. [258] who
899 investigated the interactions between biochar and mycorrhizal fungi in a water-
900 stressed agricultural soil. The results revealed that biochar had little influence on
901 arbuscular mycorrhizal fungi or plant growth under well-watered conditions. The
902 biochar induced enhancement in mycorrhizal colonization was effective under water-
903 stressed conditions.

904 Negative responses of earthworm were also observed. The negative alterations
905 include weight loss and decreased survival [259, 260]. These effects were associated
906 to desiccation caused by the water retention of biochar or an increase of soil pH or
907 toxic effects of ammonia. Tammeorg et al. [260] found that earthworms would avoid
908 biochar after half a month incubation which was probably caused by the reduction in
909 soil water potential. And after 4.5 months, biochar almost had no effects on
910 earthworms.

911 Another interesting investigation is the effect of biochar on microbial
912 communication. Bacteria communicate with each other via the biochemicals for
913 intraspecies communication. And plants synthesize flavonoids which regulate
914 microbial behaviors, e.g., the establishment of root nodules, plant hormones, and

915 microbes synthesize nodulation signals, which affect the nutrient uptake and
916 development of plant. Masiello et al. [261] studied the adsorption of an acyl-
917 homoserine lactone intercellular signaling molecule (N-3-oxo-dodecanoyl-L-
918 homoserine lactone) used by gram-negative soil microbes for the regulation of gene
919 expression (Fig. 8). The results suggested that biochars disrupted the communication
920 within a growing multicellular system. Compared with low-temperature biochar
921 (300 °C), high-temperature biochar (700 °C) inhibited cellular communication 10
922 times higher than an equivalent mass of low temperature biochar. The negative
923 impacts of biochar amendment on soil microbial community are still poorly known.
924 Sufficient studies on this aspect are necessary to make further insights.



925

926 **Fig. 8. The effects of biochar production conditions on microbial communication**
927 **[261]. Copyright 2013 American Chemical Society.**

928 **4.4. Negative impacts of biochar on GHG emissions**

929 As reviewed in section 2, biochar has been widely accepted as a material to
930 enhance soil carbon sequestration. However, negative impacts of biochar on GHG

emissions were reported. One study found that there is no effect of biochar on soil respiration across Chinese agricultural soils. The addition of biochar did not change soil CO_2 efflux and the carbon use efficiency by soil microbes [262]. Biochar addition was even found to increase CO_2 emissions and the enhancement was attributed to abiotic release of inorganic carbon, the decomposition of labile components of biochars, and the decomposition of organic matters or humus by biochar [50, 263]. Biochar amendment can also enhance the GHG emissions. A field study with wheat straw biochar amended soil over two consecutive rice growing cycles found that the emission of CH_4 enhanced by 49% and 31% at the application rates of 40 Mg ha^{-1} and 10 Mg ha^{-1}, respectively [39]. An initial enhancement of N_2O emission was observed by Singh et al. [264]. And it was caused by the labile N of biochars and microbial activity. Similar results were reported by Sánchez-García et al. [265] These data were mostly obtained from laboratory or greenhouse. Extra precaution needs to be carried out before interpreting these results in large scale field applications.

4.5. Negative impacts of biochar migration

Recently, scientists have begun to realize that the migration of biochar may also have negative impacts on ecological environment and human health [266, 267]. Biochar amendment is expected to last hundreds of years in soils. However, the calculation of the residence time does not account for the carbon loss due to soil erosion by wind and/or water in previous literatures. And the erosion potential of biochar and downstream impacts need to be studied. As discussed above, biochar particles effectively adsorb inorganic and organic contaminants and pathogens from

soil and water. The preferential erosion of biochar particles by wind may produce bioavailable contaminants in the airborne dust. Hence the migration of contaminants loaded biochar from amended soils may be a potential health hazard. Moreover, biochar can absorb short wave solar radiation and change the properties and distribution of clouds, influencing climate, air quality and biogeochemical cycles [267]. Ravi and co-workers carried out wind tunnel experiments to study the particulate matter emission of a sand and two agricultural soils amended with biochar. The obtained results revealed that the mechanisms of the increased particulate emissions were the accelerated emission of biochar particles, and the generation and emission of biochar particles resulting from abrasion of large biochar particles by sand grains. Therefore, the geomorphological processes and soil properties should be considered during the biochar applications [266]. Biochar as a potential contaminant carrier, the migration and transformation process in water should also be paid sufficient attention. The erosion and soil osmosis are the main migration approaches.

5. Conclusions and outlook

This review highlights the requirement for collaboration among interdisciplinary researchers exploring in different fields of study: environmental management including biochar for mitigating GHG emissions and biochar for contaminant management, and negative impacts of biochar application, which are often ignored, but are significant aspects for biochar application. Although environmental applications of biochar were reviewed several years ago (mainly on technical and economic aspects of biochar production, climate mitigation, contaminant

975 management), the rapid renovation of scientific knowledge on biochar and the
976 accumulation of new data encourage us to summarize the issues from both positive
977 and negative aspects simultaneously. In addition to the general consideration outlined
978 above, current knowledge gaps and research needs have been identified in this review.
979 These research priorities are listed as below:

980 Pyrolysis and carbonization of lignocellulosic biomass are originally used to
981 replace fossil resources for the production of biofuels and chemical products. Biochar,
982 as a by-product of pyrolysis for production of biofuels, is attracting much more
983 attention. However, the carbonization technology is still in the relatively extensive
984 stage. In terms of equipment, the commercialized process of production is backward.
985 It is a highly necessity that technical innovation and equipment should be developed
986 for the high-quality biochar. The biomass feedstock and the process chosen for the
987 biochar production including detailed operating conditions determine the properties of
988 biochar and the appropriate applications. The appropriate biochar should be applied to
989 the appropriate water or soil. Data about the biochars produced from various
990 feedstocks (e.g., lignocellulosic biomass, animal residue, sewage sludge, etc.) and
991 preparation parameters (e.g., peak temperature, pressure, moisture contents, etc.) for
992 various applications (e.g., mitigating GHG emissions, contaminant management,
993 increasing productivity, etc.) have been reported independently. Database related to
994 feedstock, preparation parameters, biochar properties, and potential function can be
995 built for rational utilization of biochar via further experiments and computer
996 simulation. Meanwhile, the production process of biochar should avoid secondary

997 pollution (e.g., heavy metals, PAHs, dioxin, etc.).

998 Previous findings suggest that biochar induced degradation of contaiminants is
999 more prevalent than currently recognized. More attention should be paid to the
1000 catalytic activity of biochars. It is significant to concern the potential for biochars to
1001 promote the degration of adsorbed contaminants. This feature makes biochar can
1002 solve the pollution fundamentally as opposed to just enrichment and sequestration.
1003 The regulation of PFRs in the process of biochar production is one hot topic and
1004 whether the generation of ROS is advantageous or disadvantageous in the natural
1005 environment has attracted the attention of the scientific community recently. The
1006 adsorptive removal of contaiminants has been widely investigated, but the reactive
1007 removal is still a broad space for research. Photogeneration of ROS from biochar, the
1008 catalytic performance of the biochar dissolved organic matter, the regulated formation
1009 of biochar PFRs, and the use of ROS may be the following research focuses.

1010 Biochar undergoes weathering by abiotic and biotic ageing and the deposition of
1011 organic matters and minerals, which tests its stability and will impact the activity and
1012 reactivity. Years of observation have been made. However, very little information is
1013 now available about biochar amendment at the field scale over decades or even
1014 hundred years. Reliable data on carbon sequestration, GHG emissions, contaminant
1015 management, and agronomic potential are necessary. These effects are real and the
1016 quantification is needed. Another important issue is the environmental risk assessment.
1017 Urgent attention should be paid on knowledge gaps, including biochar effects on long-
1018 term carbon sequestration, contaminants release, negative soil chemicophysical

properties alteration, and negative soil biota influence as well as microbial communication, enzymes, and plant pathogens. Although biochar has achieved great success, there is a long way to go.

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Highlights

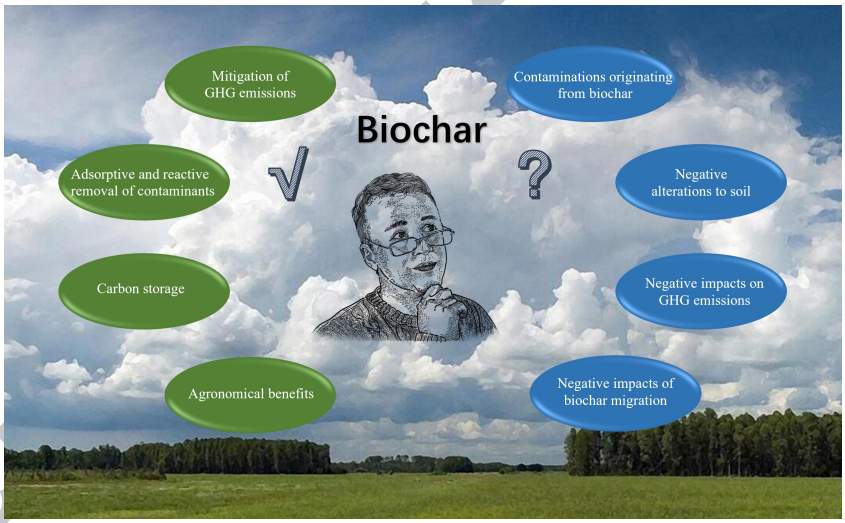
► Biochar for mitigating GHG emissions is reviewed.

► Activity and reactivity are significant for the removal of contaminants.

► The involved removal behaviors and mechanisms are reviewed.

► Potential negative aspects of biochar applications are also discussed.

► Possible improvements and outlooks of biochar applications are proposed.



Graphical Abstract