REVIEW

Remediation of mercury-contaminated soils and sediments using biochar: a critical review

Qian Yang[1,3] · Yongjie Wang[2] · Huan Zhong[1,4]

Abstract

The transformation of mercury (Hg) into the more toxic and bioaccumulative form methylmercury (MeHg) in soils and sediments can lead to the biomagnification of MeHg through the food chain, which poses ecological and health risks. In the last decade, biochar application, an in situ remediation technique, has been shown to be effective in mitigating the risks from Hg in soils and sediments. However, uncertainties associated with biochar use and its underlying mechanisms remain. Here, we summarize recent studies on the effects and advantages of biochar amendment related to Hg biogeochemistry and its bioavailability in soils and sediments and systematically analyze the progress made in understanding the underlying mechanisms responsible for reductions in Hg bioaccumulation. The existing literature indicates (1) that biochar application decreases the mobility of inorganic Hg in soils and sediments and (2) that biochar can reduce the bioavailability of MeHg and its accumulation in crops but has a complex effect on net MeHg production. In this review, two main mechanisms, a direct mechanism (e.g., Hg-biochar binding) and an indirect mechanism (e.g., biochar-impacted sulfur cycling and thus Hg-soil binding), that explain the reduction in Hg bioavailability by biochar amendment based on the interactions among biochar, soil and Hg under redox conditions are highlighted. Furthermore, the existing problems with the use of biochar to treat Hg-contaminated soils and sediments, such as the appropriate dose and the long-term effectiveness of biochar, are discussed. Further research involving laboratory tests and field applications is necessary to obtain a mechanistic understanding of the role of biochar in reducing Hg bioavailability in diverse soil types under varying redox conditions and to develop completely green and sustainable biochar-based functional materials for mitigating Hg-related health risks.

Keywords Biochar · Methylmercury · Soils · Sediments · Bioavailability

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1 Introduction

Mercury (Hg) is a highly toxic pollutant worldwide (Chen et al. 2018; Nascimento and Edmar 2003) that is released into the environment mainly through natural processes (such as forest fires, volcanic and geothermal activities, and re-emission in soils and seas) and human activities (such as metal mining and refining, fossil fuel combustion, garbage incineration and other industrial activities) (Collado et al. 2015; Li et al. 2009; Pirrone et al. 2010; Beckers and Rinklebe 2017; O'Connor et al. 2019). Inorganic Hg (IHg) species, such as Hg^{2+} and Hg^0 (Hu et al. 2013), neutral Hg sulfides (Drott et al. 2007) and low molecular mass Hg thiols (Schaefer and Morel 2009; Schaefer et al. 2011; Lyu et al. 2019), can be converted into methylmercury (MeHg) primarily through a group of anaerobic microorganisms in sediments and soils (Ullrich et al. 2001; Beckers and Rinklebe 2017). Since Minamata Bay disease was first reported in the

1950s, the world has been concerned about the accumulation of Hg (especially MeHg) in food and the dietary exposure to MeHg. Research has shown that MeHg can accumulate in fish, shellfish and other aquatic products (García-Hernández et al. 2018) and has a high concentration in rice (Cui et al. 2017; Feng et al. 2008; Zhang et al. 2010), thus threatening food safety.

Mercury-contaminated soils and sediments have been recognized as 'hotspots' of MeHg production and the main sources of MeHg in crops (e.g., rice) (Feng et al. 2008; Frohne et al. 2012; Meng et al. 2014; Wang et al. 2018b) and aquatic organisms (e.g., deposit feeders) (Lawrence et al. 1999; Ullrich et al. 2001). The adverse effects of Hg-contaminated soils and sediments on human health have been reported in the literature (Feng et al. 2008; Zhang et al. 2010; Bank 2020; Natasha et al. 2020; Jiang et al. 2021). The consumption of Hg-contaminated food such as rice, vegetables, and meat is one pathway for human exposure to Hg, although fish consumption is considered to be the primary pathway for human MeHg exposure (Mergler et al. 2007). Therefore, a number of studies have been devoted to the development of technologies for the remediation of Hg-contaminated soils and sediments (Wang et al. 2012). Compared with ex situ remediation technologies (e.g., thermal destruction and foreign soil replacement), in situ strategies (e.g., Hg immobilization and phytoremediation) have the following advantages (Pavel and Gavrilescu 2008): (1) generally low cost (due to the avoidance of infrastructure construction and long-distance transportation, among other considerations), (2) environmental friendliness, (3) easy operation and maintenance, (4) simple equipment operation and (5) a small degree of damage to the soil structure. Biochar is recognized as a functional material for in situ remediation of Hg-contaminated sites due to its advantageous properties (e.g., easy operation, little environmental degradation and high adsorption efficiency) and has also received considerable attention for the remediation of contamination by other metals (Chen et al. 2019; He et al. 2019).

Biochar is a solid and stable high-carbon material that is produced through thermal decomposition of organic material (biomass such as wood, manure or leaves) in the absence of oxygen or under oxygen-limited conditions at relatively low temperatures (< 700 °C) (Liu et al. 2015; Xiao et al. 2018). Studies have indicated that biochar has a relatively high porosity and surface area and presents a large number of functional groups and adsorption sites on the surface (Ahmad et al. 2014; Xiao et al. 2018; Tang et al. 2019). The properties of biochar are significantly influenced by many parameters, including pyrolysis temperature, residence time, and biomass feedstocks (Liu et al. 2015; Rajapaksha et al. 2016; Li et al. 2017a; Liu et al. 2016). Owing to the above-mentioned advantages of biochar, its adsorption of Hg is believed to be the major mechanism for reducing the mobility and bioavailability of Hg (Cao

et al. 2011; Inyang et al. 2016; Li et al. 2017a). For example, studies have demonstrated that biochar can reduce the IHg concentration in soil leachate (O'Connor et al. 2018) and the bioavailability of IHg in sediments (Gomez-Eyles et al. 2013; Bundschuh et al. 2015). In addition, biochar can remove MeHg from solution (Gomez-Eyles et al. 2013; Liu et al. 2017a; Wang et al. 2019b), and biochar amendment of Hg-contaminated soil can also reduce the bioavailability of MeHg to Indian mustard (*Brassica juncea*) (Shu et al. 2016a) and rice plants (Shu et al. 2016a; Wang et al. 2019b). These pioneering studies have provided initial evidence of a reduction in Hg risk upon using biochar and suggest that biochar has great potential for the in situ remediation of Hg-contaminated agricultural soils and environmental sediments.

However, it is worth noting that studies suggest that the effect of biochar on the mobility and bioavailability of Hg (IHg and MeHg) is complex, considering the varying characteristics of biochars derived from different materials and differences in environmental conditions and application rates. For example, biochar has been reported to increase the MeHg content in soil (Shu et al. 2016b), and biochar addition can alter the physical and chemical properties of soil/sediment (Beesley et al. 2011; Xiao et al. 2018) and thus affect the biogeochemical processes of nutrient elements in soils or sediments (Beckers et al. 2019). These changes could indirectly influence Hg mobility and bioavailability. However, the underlying mechanisms are far from clear. These knowledge gaps hinder the comprehensive understanding of biochar's effect on Hg in environmental media, as well as the application of biochar for environmental remediation.

Here, we summarize recent investigations on the influences of biochar amendments on Hg mobility and bioavailability in soils/sediments and discuss the potential mechanisms responsible for these influences. Furthermore, potential problems arising from biochar amendments to soils/sediments are briefly discussed. Finally, future development directions for exploring biochar-impacted Hg bioavailability are proposed. This review highlights the possible mechanisms of the interactions among biochar, soil, and Hg under redox conditions and suggests future avenues for developing effective in situ Hg remediation strategies and mitigating the risk of MeHg production and exposure. These strategies will expand the practical application of biochar to the remediation of other heavy metals. We hope this review will provide a reference for researchers, teachers, students, and soil remediation practitioners for the in situ remediation of Hg-contaminated soils and sediments.

2 Effects of biochar addition on the mobility/bioavailability of Hg in soils and sediments

A large number of studies on biochar applications for removing Hg (IHg and MeHg) indicate that biochar has a strong sorption affinity for IHg and MeHg in water/solution (Kong et al. 2011; Dong et al. 2013; Gomez-Eyles et al. 2013; Boutsika et al. 2014; Feng et al. 2020; Lyu et al. 2019). However, the effects of biochar on the mobility and bioavailability of Hg in soils/sediments compared to those in water/solution could be complex. The relationships between the physicochemical properties of biochar (e.g., porosity, surface area, pH, surface charge, functional groups, and mineral contents) (Yuan et al. 2017) and Hg sorption in water have been well summarized in a published review paper (Li et al. 2017b). Here, we focus on the physical and chemical characteristics of biochar that could affect the mobility and bioavailability of Hg (IHg and MeHg) and influence Hg biogeochemical processes in soils/sediments, especially under redox conditions.

2.1 Biochar reduces the mobility/bioavailability of IHg

Although biochar cannot decrease the total Hg concentration in soils/sediments, it can effectively reduce the mobility and bioavailability of IHg and consequently decrease Hg accumulation and toxicity to animals and plants. In the last decade, biochar has been proven to be effective in mitigating the risks of Hg in soils and sediments due to the sorption of IHg and MeHg by biochar (Table 1). For example, using Hg isotope tracer methodology, Bussan et al. (2016) found that biochar (an amendment with 5% pinewood-derived biochar) significantly reduced the IHg methylation rate in wetland sediment by 88% without having much impact on the demethylation rate. This result suggests that biochar may decrease the bioavailability of IHg for methylating microorganisms. Bundschuh et al. (2015) found in a field experiment that Hg bioaccumulation in *Hyalella azteca* decreased when the sediments were mixed with two different biochars but that the efficiency of the biochar depended on the initial particle size and contact time.

Recent studies suggest that biochar could decrease the bioaccumulation of total Hg (THg) in rice grain. For example, a pot experiment showed that dissolved THg in soil pore water decreased by 34–44% throughout the rice-growing

Table 1 Biochar utilization for the remediation of Hg-contaminated soils and sediments

Feedstocks (pyrolysis temperature, °C)	Matrix	Added doses	Effects	Mechanisms	References
Commercially available biochar	Sediment	10% w/w	↓ Bioavailability	Adsorption	Bundschuh et al. (2015)
Pinewood (~830)	Sediment	5% (dry weight)	↓ 88% in the methylation rate	Complexation, electrostatic interactions, ion exchange	Bussan et al. (2016)
Switchgrass (300 and 600)	Water and sediment	1:20:160 (biochar, sediment, water)	↓ Reduction in aqueous THg and MeHg	Adsorption	Liu et al. (2017a)
Switchgrass, poultry manure and oak (300, 600 and ~700)	Sediment	5% w/w	↓ 8.0–80.0%	Adsorption, formation of Hg-sulfide minerals and precipitation	Liu et al. (2018a)
Rice husks and a mixture of rice husks and elemental sulfur (550)	Soil	1–5% w/w	↓ 94.9–99.3%	Formation of low-solubility HgS (cinnabar)	O'Connor et al. (2018)
Rice shells (480–660)	Soil	24–72 t/ha	↓ 31–62% Hg in bran, 25–43% Hg in hull	Combination of sulfide with Hg to form Hg sulfides	Xing et al. (2019)
Sewage sludge (600)	Soil	5% w/w	↓ 73.4% MeHg, 81.9% THg in rice grain	Adsorption	Zhang et al. (2019)
Bamboo (600)	Soil	0.5–5% w/w	↓ 49–73%	Formation of Hg complexes on the biochar surface	Wang et al. (2019b)
Rice shells (480–660)	Soil	24–72 t/ha	↓ 36–32% THg 47–53% MeHg	Immobilization through binding to thiols (e.g., cysteine) in biochar	Xing et al. (2020)

season, and consequently, the polished rice THg content decreased by 58–70% with 24–72 t/ha rice shell biochar amendment (Xing et al. 2019). Another pot experiment indicated that THg in rice grain decreased by 81.9% with 5% w/w sewage sludge biochar amendment despite a promotion of Hg methylation in this acidic soil (Zhang et al. 2019). In addition, the Hg levels in soil leachate decreased by more than 94% with rice husk-derived biochar amendment (1–5% w/w), similar to the results of activated carbon amendment (THg reduction by 99.9% with 3% w/w amendment) (O'Connor et al. 2018). These results show that biochar could be a potential green environmental sorbent for the in situ remediation of Hg-contaminated soils/sediments.

2.2 Biochar affects net MeHg production and MeHg mobility/bioavailability

Given the sorption of IHg to biochar, biochar amendments can decrease the microbial methylation of IHg, and reduce net MeHg production. For example, Bussan et al. (2016) used Hg stable isotope tracers (^{202}Hg) to explore the effect of biochar on Hg methylation potential in sediments. The results showed that biochar addition reduced the Hg methylation rate by 88% compared with that in the control group (without biochar addition). Gilmour et al. (2018) used biochar to perform in situ mercury remediation in the Penobscot River salt marsh. The results showed that biochar could reduce the MeHg content in porewater. Wang et al. (2019b) reported that bamboo-derived biochar decreased net MeHg production by ~ 70% at a 5% addition rate in paddy soils. However, Shu et al. (2016a, b) found that rice straw-derived biochar could significantly increase the concentration of MeHg in paddy soils. In addition, a long-term microcosm incubation study showed that two peaks in MeHg occurred during incubation, although the aqueous and soil solid concentrations of MeHg were generally lower in the amended systems than in the controls (Liu et al. 2018a).

With the development of more in-depth research, biochar studies have shifted from initial research work in the laboratory to practical applications. Recently, additional studies have confirmed that biochar can reduce the bioavailability of MeHg and its accumulation in crops. For example, Shu et al. (2016b) reported that straw biochar amendment could substantially reduce MeHg levels in rice plants (reduced by, rice grain 49–92%, straw 28–83%, root 29–61%), although biochar enhanced net MeHg production. One possible explanation for this phenomenon may be the decreased phytoavailability of soil MeHg (defined as the "MeHg immobilization effect"), as reflected by decreased extraction rates of MeHg by $(NH_4)_2S_2O_3$. Alternatively, an increase in rice yield could partly contribute to reducing the rice MeHg concentration (defined as the "biological dilution effect"). To verify the effect of biochar on bioaccumulation

and bioavailability, Zhang et al. (2018) studied the effects of the co-application of biochar and sodium nitrate on MeHg bioavailability and found that the content of MeHg in rice grain was significantly reduced following the co-application of biochar and sodium nitrate. Moreover, Wang et al. (2019b) reported that biochar amendments (0.5% w/w) further reduced MeHg accumulation (by 82–87%) in rice grains grown in selenium-amended paddy soil. The results provide new insights into the combined effects of biochar and other materials on reducing the bioavailability of MeHg in Hg-contaminated soils.

3 The interaction mechanisms of Hg with biochar and soils/sediments

Relative to aqueous solutions, the various substances in the soil/sediment environment biogeochemical cycle are extremely complex; therefore, the influence of biochar on the transformation and bioavailability of Hg is complex. According to reports, two main mechanisms have been proposed, as shown in Fig. 1. One mechanism is the sorption of Hg to biochar, which directly reduces the mobility and bioavailability of Hg. The other is that biochar indirectly affects the mobility and bioavailability of Hg associated with biochar–soil interactions under different conditions.
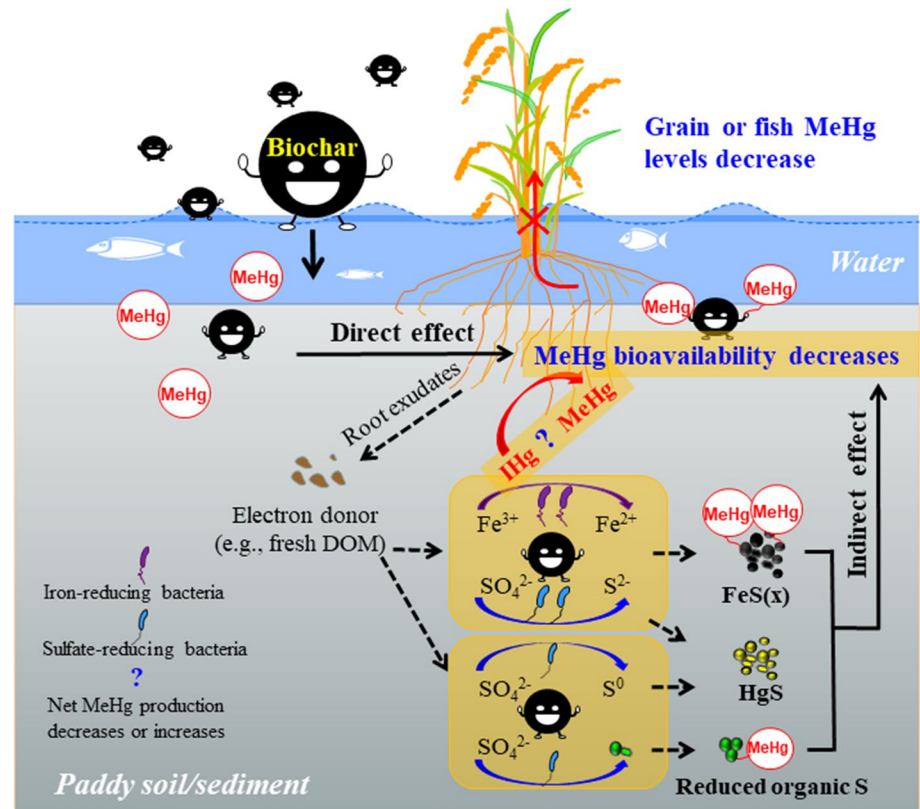
3.1 Direct interactions

The direct interactions between Hg and biochar are governed by the structure and surface chemistry of the biochar. The porosity and surface area are critical components of the structure of biochar, and the surface chemistry of biochar is dominated by surface functional groups and element contents (Ahmad et al. 2014; Liu et al. 2015; Tan et al. 2016; Xiao et al. 2018). The various mechanisms proposed for the interactions of biochar with Hg in aqueous solutions, including complexation, precipitation, ion exchange, electrostatic interaction (chemisorption), and physical sorption, have been well summarized in published review papers (Li et al. 2017a; Deng et al. 2020). The major mechanisms of the direct interactions between Hg and biochar in soil solution based on these published papers are summarized in Fig. 2. Here, we focus on the mechanisms of complexation and precipitation that affect the biogeochemical processes of Hg in soils/sediments because these mechanisms may be critical for reducing Hg bioavailability and for remediation applications.
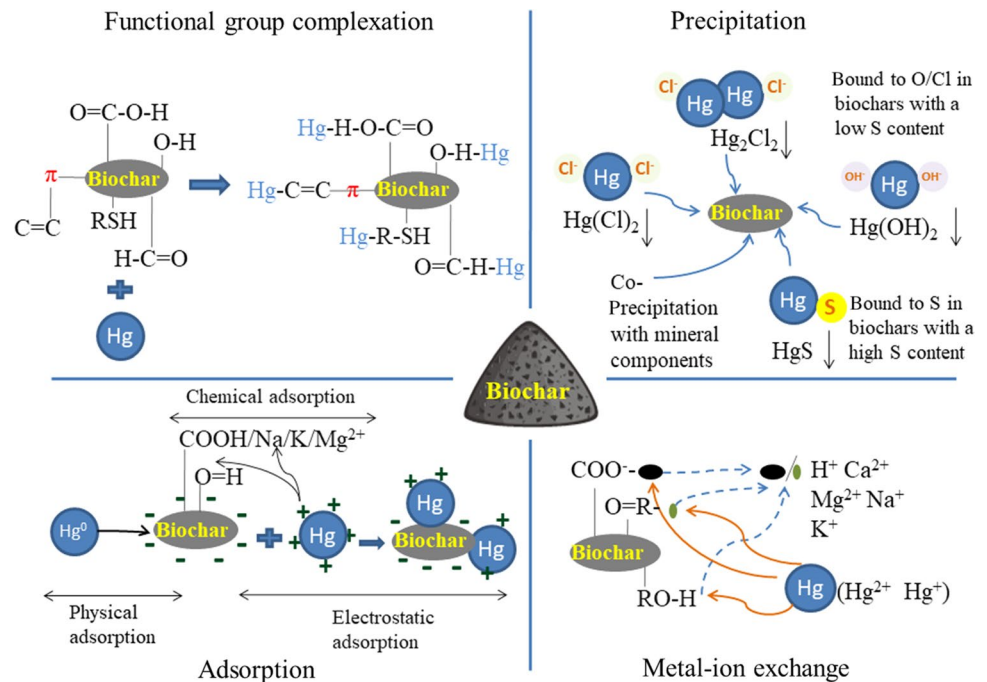
3.1.1 Complexation

The different types of functional groups on the surface of biochars are critical for the complexation of Hg with

Fig. 1 Proposed mechanisms of the interactions between Hg and biochar in soils and sediments

Fig. 2 Possible mechanisms of the direct interactions between biochar and Hg (modified from Inyang et al. 2016; Li et al. 2017a)

biochar, as shown in Fig. 2. For example, the sorption of Hg(II) by bagasse-derived biochar was mainly attributed to the complexation of Hg(II) with phenolic hydroxyl (COH) and carboxylic (COOH) groups and the formation of $(-O)_2Hg^{II}$ and $(-COO)_2Hg^{II}$ (Xu et al. 2016), while the interactions of Hg(II) with C=C and C=O to form Hg–π bonds could be mainly responsible for Hg(II) sorption by hickory chips or wood-derived biochar (Xu et al. 2016; Park et al. 2019). Dong et al. (2013) suggested that Hg was irreversibly sorbed via complexation with phenolic hydroxyl

and carboxylic groups in low-temperature biochars (Brazilian pepper biochar, 400 and 500 ℃) and by graphite-like structures in high-temperature biochar (600 ℃). In addition, sulfur-containing functional groups in biochars have been identified by advanced techniques, such as X-ray absorption near edge structure (XANES) (Cheah et al. 2014; Liu et al. 2016; Wang et al. 2019b) and X-ray emission spectroscopy (XES) (Holden et al. 2018), and are regarded as a key factor controlling the complexation of Hg with biochar. For example, Hg extended X-ray absorption fine structure (EXAFS) revealed that Hg(II) is bound to S in biochar with a high S content and to O and Cl in biochar with a low S content, indicating that binding of Hg(II) to reduced S is usually favored over binding to other functional groups, e.g., hydroxyl and carboxyl groups (Liu et al. 2016). Moreover, sulfurized biochars produced with reduced sulfur-containing chemical reagents (e.g., calcium polysulfide, dimercapto compounds, 3-mercaptopropyltrimethoxysilane (3-MPTS) and sodium sulfide) exhibited enhanced Hg(II) removal efficiency due to the binding of Hg(II) to reduced S, such as that in polysulfur-like structures (Liu et al. 2018a), thiophenic groups (Park et al. 2019), thiols (Huang et al. 2019) and sulfides (Tan et al. 2016; Feng et al. 2020), in biochars prepared via different modification methods.

Although a previous study suggested that IHg and MeHg sorption to biochars involves different sorption mechanisms (Gomez-Eyles et al. 2013), the mechanisms of MeHg binding to biochars could be similar to the mechanisms of IHg binding to biochars. Several studies have investigated the mechanisms of complexation between biochar and MeHg, and the interaction mechanism remains unclear. Studies based on microcosm anoxic incubation experiments showed that rice straw- or bamboo-derived biochar can significantly decrease the MeHg concentration in overlying water during soil incubation (Shu et al. 2016a, b; Wang et al. 2019b). Interestingly, the fraction of extractable MeHg (% of total) obtained with $(NH_4)_2S_2O_3$ presented a decreasing trend with increasing biochar dose in soils with or without selenium addition (Wang et al. 2019b) and showed a negative relation with MeHg log K_d values (Fig. 3). These results suggest that the dissolved MeHg is partitioned into the biochar, which is most likely a result of adsorption and complexation of MeHg by organosulfur groups in the biochar (Wang et al. 2019b). Furthermore, Huang et al. (2019) found that the active sites (–SH) on modified biochar surfaces play an important role in Hg(II) and MeHg scavenging from aqueous solution by surface complexation with –SH.

3.1.2 Precipitation

Another important direct interaction between Hg and biochar is precipitation, whereby IHg and MeHg are immobilized in soils/sediments. One study proposed that Hg(II)
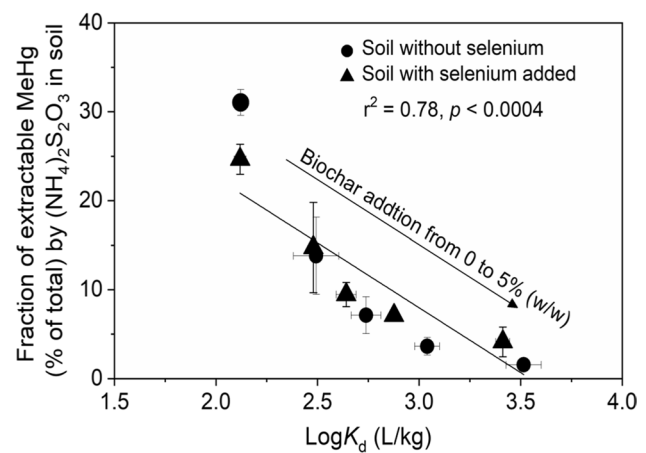
Fig. 3 Relationship between the fraction of extractable MeHg (% of total) obtained with $(NH_4)_2S_2O_3$ and the MeHg logK_d values in the microcosm anoxic incubation experiments. Data are shown as the mean ± standard deviation ($n = 3$) (adopted from our published paper, Wang et al. 2019b)

could be reduced via Hg_2Cl_2 or $Hg(OH)_2$ precipitation on the biochar surface (Kong et al. 2011). Furthermore, Tan et al. (2016) reported that sodium sulfide impregnation of corn straw biochar was an efficient way to enhance Hg(II) removal from aqueous solution due to the formation of HgS precipitates. A long-term (1030 days) anaerobic microcosm experiment indicated that biochar amendment could stabilize the unstable fraction of Hg (e.g., dissolvable Hg, HgO, colloidal Hg, nano Hg, etc.) in sediment as less soluble Hg-sulfide phases on the surface or within biochar particles (Liu et al. 2018a).

3.2 Indirect interactions

The physical, chemical, and biological properties of soils can be altered following biochar amendment (Joseph et al. 2015; Lian and Xing 2017; Bandara et al. 2019), and such changes can impact the biogeochemical cycling of Hg and other elements related to Hg cycling.

Biochar-driven alterations in biogeochemical redox processes that impact the mobility and bioavailability of Hg in soils/sediments have barely been investigated, apart from a few studies. The published literature focuses on biochar-induced biogeochemical processes involving the redox elements S and Fe that are associated with the mobility and bioavailability of Hg. Positive effects of biochar amendment on sulfate-reducing bacteria (SRB) activities have been reported (Eastona et al. 2015; Sande 2016). Thus, biochar-induced changes in S cycling may indirectly affect Hg mobility/bioavailability. For example, Shu et al. (2016b) found that rice straw-derived biochar contained high levels of sulfate, which elevated sulfate concentrations in soil and could subsequently enhance microbial

production of MeHg under anoxic conditions. Liu et al. (2018a) found that changes in MeHg concentration in the aqueous phase could be attributed to the activity of fermenters, iron-reducing bacteria (FeRB), and SRB in the early stages of microcosm incubation and to the activity of methanogens in later stages. Community shifts induced by biochar amendment may be correlated with changes in the concentrations of carbon sources and organic acids as electron donors and electron acceptors (NO_3^-, Fe, and SO_4^{2-}) (Liu et al. 2018b). In addition, Xing et al. (2020) detected a significant amount of thiol compounds (e.g., cysteine) in the biochar-treated paddy soil compared to the control. These thiol compounds might complex with MeHg to form MeHg–thiol complexes, thereby immobilizing MeHg in the soil. Similarly, Wang et al. (2020a) found that nanoactivated carbon as a soil amendment significantly reduced Hg uptake by rice plants and induced a change in Hg binding from organic matter to nano-HgS in the soil. XANES of S and Hg and transmission electron microscopy linked with energy-dispersive X-ray (TEM–EDX) spectroscopy revealed that Hg speciation transformation might be coupled to the reduction of sulfoxide to reduced sulfur species (S^0) by nanoactivated carbon. On the other hand, the precipitation of FeS under anoxic conditions could be induced by biochar due to the redox properties of biochar (Klüpfel et al. 2014; Joseph et al. 2015; Prévoteau et al. 2016; Yuan et al. 2017). Positive effects of biochar amendment on the activities of sulfate-reducing bacteria (SRB) may enhance the production of FeS, as reported by a recent study (Wang et al. 2020c). The formed FeS can react with Hg(II) and

MeHg to precipitate as metacinnabar (β-HgS(s)) and consequently immobilize Hg (Jeong et al. 2010; Jonsson et al. 2016). In summary, we propose that biochar-induced biogeochemical processes involving the redox elements S and Fe and resulting in the formation of inorganic and organic S species could play a key role in impacting the biogeochemical redox processes of Hg (Fig. 1).

4 Advantages of biochar amendments for the remediation of Hg-contaminated soils and sediments

Over the past 10 years (from 2011 to 2020), the number of references related to the use of biochar to remediate Hg-contaminated soils and sediments has increased by 99% (as shown in Fig. 4), and the number of citations for articles has increased from 3 to 1103. Among these articles, more studies have been published on biochar used to treat mercury-contaminated soils than mercury-contaminated sediments. Table 2 shows the published in situ remediation technologies (stabilization/solidification and immobilization) used to reduce MeHg accumulation in rice grain in pot experiment and field studies. The results show that biochar can decrease THg and MeHg in rice grain by 30–82 and 45–88% (% total content of THg and MeHg in rice grain), respectively. All the results indicate that biochar is a promising material for the in situ remediation of Hg-contaminated soils and sediments due to its advantageous properties.

Fig. 4 Number of articles and citations on mercury (Hg) pollution in soils and sediments under biochar amendment in the last 10 years (2011–2020). The data were collected by searching the Web of Science Core Collection (http://www.isiknowledge.com) using the terms "mercury", "biochar", "soil" and "sediment" on July 7, 2020
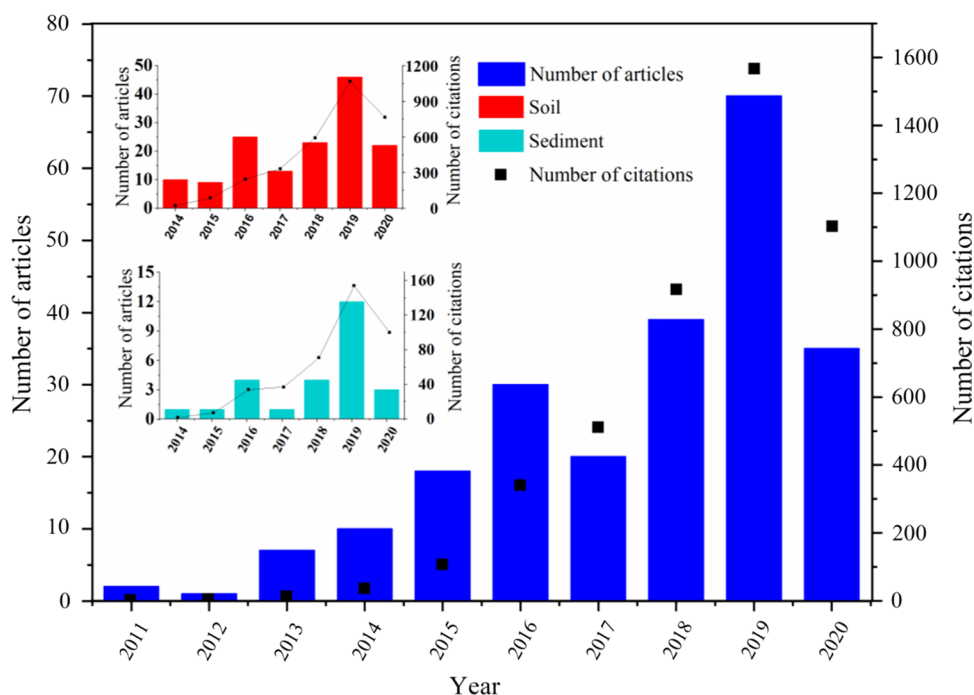
Table 2 Published in situ remediation technologies (stabilization/solidification and immobilization) for reducing MeHg accumulation in rice grain in pot experiments

Soil	Agent/material addition	Decrease in rice grain (% of total)	References
Hg-amended soil[a]	5.0 mg/kg Se as selenite	55% MeHg	Wang et al. (2014)
Hg-amended soil[a]	3.0 mg/kg Se [c]	46–49% MeHg	Wang et al. (2016)
Hg-contaminated soil[b]	1.0% w/w rice straw-derived biochar	~50% MeHg	Shu et al. (2016b)
Hg-amended soil[a]	100 mg/kg elemental S	~60% THg	Li et al. (2017b)
Hg-amended soil[a]	100 mg/kg S as thiosulfate	~31% THg	Li et al. (2018)
Hg-amended soil[a]	3.0 mg/kg Se[d] and 0.5% w/w bamboo biochar	88% MeHg	Wang et al. (2019b)
Hg-contaminated soil[b]	24–72 t/ha (1.2%, w/w)[e] rice shell biochar and 72 t/ha (3.6%, w/w)[e] wheat straw biochar	58–70 and 38% THg	Xing et al. (2019)
Hg-contaminated soil[b]	5% w/w sewage sludge biochar	73% MeHg and 82% THg	Zhang et al. (2019)
Hg-contaminated soil[b]	1–3% w/w nanoactivated carbon	47–63% THg	Wang et al. (2020a)
Hg-contaminated soil[b]	4–72 t/ha (1.2–3.6%, w/w)[e] rice shell biochar	47–53% MeHg and 32–36% THg	Xing et al. (2020)

[a]Soils were spiked with inorganic Hg solution to simulate Hg pollution from wastewater discharge

[b]Soils were collected from a Hg mining area

[c]Se was added using sodium selenite or selenate

[d]Aged (3 years) Se-spiked soil was prepared by mixing Se(IV)- and Se(VI)-spiked soil

[e]The added dose, presented in %, was calculated from a depth of 15 cm and a bulk density of 1.3 g/cm^3

5 Existing problems with the use of biochar to treat Hg-contaminated soils and sediments

Although biochar has potential advantages, it has not yet been applied on a large scale for the in situ remediation of Hg-contaminated soils/sediments, which may be related to the following problems.

5.1 Dosage and price

Table 2 lists the amendment dose and the reduction rate of MeHg in rice grain for the application of different biochars to Hg-contaminated soils/sediments. From the results, the reduction rate of Hg was higher when biochar was applied at a higher rate. However, most of the studies on soil/sediment remediation with biochar are based on pot experiments, and the amendment dose is high (1–5%, w/w). High cost may be a problem in practical applications with a high application rate of biochar.

The mean price of biochar was US$ 2650/t (ranging from US$ 90/t in the Philippines to US$ 8850/t in the US) (Zhang et al. 2017). According to a published report, the current break-even price for biochar is 246 US$/t, which is approximately 1/6th the price of commercially available activated carbon (1500 US$/t) (Maroušek et al. 2017). However, the economic costs of biochar cover feedstock collection, hauling, storage, and processing. The operating expenses include production, maintenance, labor costs, and transport and distribution costs (Galinato et al. 2011; Kung et al. 2013; Dickinson et al. 2015; Ahmed et al. 2016). Although the price

of the raw materials (e.g., livestock manure, crop straw, and wood) is low, transportation contributes the most to the total cost (Zhang et al. 2017). Furthermore, maintenance, labor, fuel, and staff expenses may also restrict the application of biochar.

5.2 Long-term effectiveness

The long-term effectiveness of biochar amendments is an important theoretical basis for discussing the environmental risks of biochar. In field experiments, many factors during biochar aging can affect biochar stability, and the adsorption capacity of biochar for Hg is still unclear. The effects of the raw materials, pyrolysis conditions, and soil properties on biochar stability should be taken into account. For example, Cui et al. (2012) and Jones et al. (2012) found that during the biochar aging process, the surface of the biochar is oxidized and many oxygen-containing functional groups (such as COH and COOH groups) are formed again, thus resulting in more negative charges and a higher ion exchange efficiency on the biochar surface. Furthermore, biochar has a highly aromatic structure and a high degree of chemical and biological stability after pyrolysis, and thus, it is very resistant to degradation. However, biochar persistence in the environment does not mean that it remains unchanged in a soil or sediment environment (Schmidt and Noack 2000). When biochar enters soil or sediment, with changes in environmental conditions (temperature, wind, moisture, rainfall, and soil type) (Zama et al. 2018) and the passage of time, the composition and surface chemical properties of the biochar change greatly, which may affect the passivation stability

of biochar for Hg. For example, Bundschuh et al. (2015) found that although biochar can reduce the bioavailability and bioaccumulation of total mercury in sediments, the extent of the reduction decreased over time, resulting in the re-release of Hg into soils, sediments or water body and a subsequent increase in Hg bioavailability and bioaccumulation. In addition, changes in environmental conditions (e.g., flooded or unflooded) and/or the microorganism community may promote the biochar aging process and thus decrease the adsorption capacity (Wang et al. 2018a). For example, micropores can become blocked during aging, thereby decreasing the surface area of the biochar.

6 Outlook of using biochar for the remediation of Hg-contaminated soils and sediments

Biochar is a promising platform for the synthesis of many other functional materials due to its easily tuned surface functionality and porosity (Liu et al. 2015; Rajapaksha et al. 2016). Therefore, the development of biochar-based functional materials and modified biochars is important (Liu et al. 2015; Ahmed et al. 2016). For example, Feng et al. (2020) conducted an experiment to improve the surface characteristics of biochar and found incorporated Fe, S, and Cl species in Fe-modified biochar, which makes the modified biochar a prospective material for Hg(II) removal (Gong et al. 2019). Lyu et al. (2019) reported that thiol-modified biochar can achieve higher removal rates of Hg(II) and MeHg (320.1 and 104.9 mg/g) than unmodified biochar. In addition to using engineered biochar alone, the multiple applications of biochar and other chemical agents (e.g., sulfur or selenate/selenite) may have great potential to improve the effectiveness of remediation. For example, Wang et al. (2019b) suggested that multiple applications of selenium and biochar could be a novel remediation strategy to mitigate MeHg accumulation in rice. Moreover, Hg bioavailability, rather than the total concentration, should be the focus of risk management regarding Hg-contaminated soils and sediments because biochar amendment cannot decrease the total concentration of Hg in soils/sediments. Thus, the development of methods for Hg bioavailability risk assessment under biochar amendment is required for quantitative risk assessment and to meet remediation objectives.

To achieve a more effective mitigation of the risks from Hg in soils and sediments, more efforts should be made to investigate the long-term effects of biochar amendments on reducing Hg bioavailability apart from the above-mentioned technological applications. The major influencing factors that should be considered include the application rate, cost reductions, large-scale commercial availability and sustainability, and possible ecological environmental risks.

Most importantly, minimizing the transfer of MeHg from soils/sediments to the food chain is a primary goal of the remediation of Hg-contaminated sites. Thus, the long-term benefit of combining biochar-based mitigation strategies with several other methods, including the minimization of Hg inputs, adoption of appropriate water management and changes in land use to grow low-accumulation crops, should be investigated.

7 Conclusions

Biochar amendment could be a practical and effective solution to mitigate the risk of Hg transfer from the environment to the biosphere. This review provides an overview of the current state of the development of biochar for the in situ remediation of Hg-contaminated soils and sediments and highlights the proposed mechanisms involved in the interactions between biochar, Hg and soils/sediments. Although the positive effects of biochar on the in situ remediation of Hg-contaminated soils and sediments have been identified, some of the remaining challenges and goals associated with biochar application remain to be investigated, including smart biochar design, multiple applications of biochar with other materials, long-term effectiveness measurements, low-dose applications, and multitier risk assessments. Implementing these strategies will further improve our ability to mitigate the ecological risks from Hg in the environment and to expand the practical application of biochar to the remediation of other heavy metals.

Compliance with ethical standards

Conflict of interest No conflicts of interest exist in the submission of this manuscript, and the manuscript is approved by all authors for publication.

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