Application of biochar for the remediation of polluted sediments

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Abstract

Polluted sediments pose potential threats to environmental and human health and challenges to water management. Biochar is a carbon-rich material produced through pyrolysis of biomass waste, which performs well in soil amendment, climate improvement, and water treatment. Unlike soil and aqueous solution, sediments are both the sink and source of water pollutants. As for in-situ sediment remediation, biochar also shows unique advantages in removing or immobilizing inorganic and organic pollutants (OPs). This paper provides a comprehensive review of the current methods of in-situ biochar amendments specific to polluted sediments. Physicochemical biochar properties (pore structure, surface functional groups, pH and surface charge, mineral components) were influenced by pyrolysis conditions, feedstock types, and modification. Furthermore, the mechanisms and efficiency of remediation of pollutants (heavy metals and OPs) vary with the biochar properties. Biochar influences microbial compositions and benthic organisms in sediments. Depending on the location or flow rate of polluted sediments, potential utilization methods of biochar alone or coupled with other materials are discussed. Finally, future practical challenges of biochar as a sediment amendment are addressed. This review provides an overview and outlook for sediment remediation using biochar, which will be valuable for further scientific research and engineering applications.

Keywords: Engineering biochar; Heavy metal; Modification; Organic pollutant; Sediment remediation

1. Introduction

With the expansion of industry, high concentrations of heavy metals (HMs) and organic pollutants (OPs) are detected in sediments due to various inputs including those from sewage infiltration and atmospheric deposition [1]. Sediments are basic environmental components that provide nutrients for many organisms and serve as repositories of organic and inorganic pollutants from aquatic ecosystems, which may have negative influences on environment and ecology [2]. Nowadays, serious adverse impacts of polluted sediments have occurred in many countries due to mining or industrial processes, such as toxicity to aquatic flora and fauna, which has aggravated water pollution since the 1980s [3, 4]. Therefore, we must explore executable methods to achieve sediment remediation and maintain its ecological function.

Remediation strategies for polluted sediments include in-situ and ex-situ actions [5]. Conventional ex-situ sediment remediation mainly relies on relocating polluted sediments, and in-situ remediation involves natural recovery without human intervention or capping contaminants with special barrier materials to isolate pollutants [6, 7]. Nowadays, sediment remediation technology mainly involves dredging (ex-situ) and capping (in-situ) (Fig. 1). Mildly polluted sediments are relocated to maintain ditches and lakes after dredging, nevertheless, heavily polluted sediments may cause secondary pollution if they are reused prior to detoxification [8, 9]. Large investments required for large-scale, ex-situ clean-up projects and the detoxification of transported sediments in dredging work is also a crucial problem [10]. Hence, lower cost and more efficient in-situ sediment remediation technologies are increasing in popularity [11, 12].

Numerous laboratory experiments and full-scale field sediment treatments have shown that sequestration and immobilization of pollutants using a sorbent material has progressed to an innovative in-situ sediment remediation approach [13-16]. Various carbonaceous materials are available for in-situ sediment remediation, including activated carbon, biochar, and carbon nanotubes [11]. As a potential surrogate, biochar has shown superior qualities (relative abundance and comparative sorptive abilities) compared with other carbonaceous materials and provides new material for polluted sediment remediation [17]. For example, the cost of activated carbon is approximately 10 USD/m^2, whereas the cost of biochar is approximately 2 USD/m^2 [18]. Biochar is prepared through pyrolysis of biomass (agricultural waste, forestry waste, and sewage sludge) under insufficient or zero oxygen conditions. Biochar has been successfully applied as an amendment to reduce contaminant bioavailability, or as an active capping material, to reduce contaminant mobility in a laboratory and pilot-scale sediment remediation [19]. Ghosh et al. [20] proved that biochar has long-term effectiveness and physical stability in pilot-scale sites. Thus, biochar has a high potential in sediment remediation and ecological engineering via contaminant sequestration and immobilization [21].

The objective of this review was to methodically summarize and analyze the amendment of sediment using biochar. Sediments are generated from soil after natural weathering and transportation processes and accumulate on the substrate as a critical compartment of aquatic ecosystems [2]. Biochar is an outstanding remediation agent that has been widely used in wastewater decontamination to remove OPs, HMs, and other inorganic contaminants (NH_4^+, PO_4^{3-}, and NO_3^-), and as a soil amendment to immobilize or insulate HMs and OPs [22-25]. Moreover, many studies have focused on its performance in sediment remediation [8, 26]. The application of biochar for polluted soil and water remediation has been comprehensively reviewed [27, 28]. However, previous reviews have mainly focused on the application of carbonaceous materials (mainly activated carbon) in sediment remediation from technical and ecological perspectives, and there is a lack of detailed information regarding in-situ sediment remediation mechanisms and the potential risks posed by biochar applications [7, 12, 29-31]. To the best of our knowledge, this is the first comprehensive review that concentrates on the application mechanisms, methods, and risk of biochar for remediation of polluted sediments and presents an overview of the following: (I) the effects of physicochemical properties (pore structure, functional groups [FGs], pH, and surface charge) of biochar on sediment remediation; (II) analysis of the mechanism of biochar for sediment remediation of HMs and OPs; (III) the risks of biochar additions to biological systems; (IV) the engineering application approaches of biochar in sediments; and, (V)prospects and challenges for applying biochar in sediment remediation.

2. Effects of physicochemical properties of biochar on sediments remediation

In the utilization of biochar as an amendment for polluted sediments, the physicochemical properties of biochar significantly affect its ability for sorbing pollutants. The raw material, preparation conditions, and modifications are principal influencing factors, which is a breakthrough for improving the physicochemical properties of biochar to effectively remediate contaminated sediments [32]. Pore structure, pH, surface charge, FGs, and mineral contents are the main characteristics of biochar that influence the effectiveness of sediment remediation.

2.1 Pore structure

Pore structure, including surface area, porosity, and pore diameter, is an important physical property of biochar [32]. Pore surface area and porosity vary with the pyrolysis temperature used during biochar production. Therefore, it is of interest to summarize and determine the relationship between the pyrolysis temperature and the physical properties of biochar to identify the optimal preparation temperature for polluted sediment remediation. Liu et al. [39] selected switchgrass biochar (pyrolysis at 300 °C [switchgrass biochar-300] or 600 °C [switchgrass biochar-600]) to compare the effect of biochar on Hg stabilization in sediment, and reported that the surface areas of switchgrass biochar-300 and switchgrass biochar-600 were 2.6 and 230 m^2/g, respectively. Biochar obtained at 600 °C with a larger surface area and higher S content was conducive to stabilizing Hg through precipitation of Hg-sulfide minerals [39]. An assessment of the use of wheat straw biochar (pyrolysis at 400 °C [wheat straw biochar-400] or 700 °C [wheat straw biochar-700]) as an adsorbent, aiming to dissipate phenanthrene and pyrene in polluted sediments, was conducted by Chi and Liu [41]. The result showed that a high pyrolysis temperature improved the pore structure and promoted the aromaticity of biochar, resulting in wheat straw biochar-700 °C manifesting stronger immobilization of polycyclic aromatic hydrocarbon (PAHs) in sediments than wheat straw biochar-400 °C. Production of biochar at 600–700 °C usually results in a higher surface area (Table 1).

In addition to the pyrolysis temperature, the biochar feedstock is also an important factor. The raw biomass containing organic (cellulose, fats, and hemi-cellulous) and inorganic substance (N, P, S, K, and minerals) may affect the physical properties of the biochar. For example, Wang et al. [42] applied three kinds of biochar produced from crofton weed, macadamia, and wheat straw (BC-1, BC-2, and BC-3) to adsorb flubendiamide in sediments and showed that the surface areas of BC-1, BC-2, and BC-3 varied considerably, at 382.21, 0.55, and 24.73 m^2/g, respectively. According to sorption studies, the addition of BC-1 to sediments was the most effective material in reducing freely dissolved flubendiamide due to its microporous surface texture, high surface area, and a higher fraction of non-carbonized organic matter (OM) than other biochars [42]. Generally, biochars produced at higher temperatures with lower surface areas and pore volumes may arise from the extensive cross-linkages, cracking, or blockage of micropores when the feedstocks contain less volatile matter [54]. Therefore, biochars produced from various feedstock and pyrolysis temperatures have entirely different physical properties.

2.2 FGs

FGs, such as carboxylic group (–COOH), hydroxyl group (–OH), amino group ($–NH_4^+$), and aromatic compounds (–C=C–), in biochar determine the mechanism between biochar and pollutants in sediment [55]. Pyrolysis temperature and the raw biomass of biochar are the main factors related to FGs of biochar [56]. When the pyrolysis temperature is increased, the carbonization degree of biomass is higher, but the abundance of FGs decreases [32]. Using different types of biomass, the FGs in biochar produced at low temperatures (≤ 300 °C) were unchanged, whereas there were multiple different peaks in biochar produced at a relatively high pyrolysis temperature (≥ 400 °C) [32, 57].

FGs vary with the pyrolysis temperature using the same biomass, and the atomic ratios of H/C, O/C, and N/C reflect the chemical components of biochar. The O/C and H/C ratios of biochar significantly decrease as the pyrolysis temperature of biochar increases, indicating an increase in aromaticity [32]. It is beneficial to enhance contact between pollutants and biochar to enable a high immobilization ability of biochar-amended sediments [58]. Suliman et al. [32] observed decreased O/C and H/C ratios corresponding to carboxylic (–COOH) and hydroxyl (–OH) groups as the pyrolysis temperature increased from 350 °C to 600 °C for biochars from Douglas fir wood (DFW), Douglas fir bark (DFB), and hybrid poplar wood (HP), which was attributed to the increasing C content and decreasing O content. In general, the content of oxygen-FGs on the surface of biochar decrease as the pyrolysis temperature increases but the opposite result was proved by Hung et al. [55]. The same FGs were present in red algae-based biochar (RAB) produced at 300 °C or 500 °C; however, increased content of oxygen-FGs was reported for RAB-900 °C as the pyrolysis temperature increased from 700 to 900 °C [55]. Thus, the abundance of –COOH and –COH groups in RAB-900 °C achieve the highest 4-nonylphenol reactions with H_2O_2 to generate reactive radicals under alkaline conditions from marine sediments [55]. Hence, it is possible to obtain biochar with ideal FGs by appropriately balancing the feedstock and pyrolysis temperature.

2.3 pH and surface charge

The pH and surface charge of biochar also vary with different feedstocks and pyrolysis temperatures and are closely related to the efficiency of sediment remediation [39]. The alkaline matter in biochar caused by base cations (Ca, Mg, K, and Na) in raw biomass are transformed into oxides, hydroxides, and carbonates (e.g., ash) during pyrolysis [26, 59, 60]. The biochar feedstock influences all the properties related to the alkalinity. Previous studies had proved that the pH of sediment increased following the addition of biochar due to the alkaline materials and oxygen-FGs in biochar [26, 42, 61]. The pH of biochar is related to the pyrolysis temperature (Table 1), which significantly increases the pH of sediments [62]. With an increasing pyrolysis temperature, the enrichment of the ash content contributes to the increased pH of biochar [56]. HMs and OPs vary in their sensitivity to sediment pH, whereby in general, a higher sediment pH increases the stability of HMs but prevents the adsorption of OPs [4, 26, 34, 42].

The surface charge of biochar is strongly influenced by the FGs that are present. With an increasing pyrolysis temperature, the oxygenated FGs would be lost from the biochar surface, contributing to the negative charge of the biochar [63]. Among the sorption reactions between biochar and pollutants, electrostatic attraction is controlled by the surface charge of biochar. Dong et al. [64] measured the ability of water caltrop shell biochar (WCSB) surface with a negative charge to adsorb $S_2O_8^{2-}$ through electrostatic attraction and the redox process of Fe^{2+}/Ce^{3+} to Fe^{3+}/Ce^{4+} to catalyze $S_2O_8^{2-}$ to produce active oxidants capable of degrading phthalate esters (PAEs) on a Fe–Ce/WCSB surface. As for HM ions, biochar with a surface charge binds to pollutants with an opposite charge [8]. The pH and surface charge of biochar, which are related to the feedstock and pyrolysis temperature, have different roles in immobilizing pollutants in sediments.

The mineral composition of biochar is a small partition which can reduce the availability of pollutants by ion exchange or precipitation [56]. Feedstock and pyrolysis temperature control the contents of mineral components in biochar, and pyrolysis of biomass at a higher temperature could augment the mineral contents in biochar [32, 56]. In conclusion, the physicochemical properties of biochar mainly vary with feedstock and temperature via various mechanisms.

2.4 Modification

In comparison with modified biochar, pristine biochar has inappropriate physicochemical properties, making it difficult to achieve the optimal adsorption capacity and minimum standard concentration of target pollutants. Hence, modification technologies of biochar have attracted the interest of researchers. Common biochar modification methods for sediment remediation are chemical additive activation and biochar-supported nanocomposites [65]. Chemical additive (e.g. HCl, H_2SO_4, H_3PO_4, KOH, NaOH, and $MgCl_2$) activation can increase the specific surface area and ameliorate the undesirable pore properties and oxygen-FGs of biochar [66-69], which contributes to improving the performance of biochar in reducing contaminants in sediments. Among these chemical additives, acid treatment would augment the porosity and surface area of biochar and contribute to FGs on biochar surface [67]. For example, Liu et al. [40] found that activations using H_2O_2, $ZnCl_2$, HNO_3, H_3PO_4, and H_2SO_4 increased the specific surface area of rice husk biochar to varying degrees. Moreover, activation of biochar by HNO_3 significantly improved its oxygen-FGs compared to other activated biochar [40]. Furthermore, the decreased Cd content in overlying water indicated that the ability of biochar to adsorb HMs was increased by chemical activation [59].

The application of modified biochar is promising in sediment treatment. Currently, more researchers are directing attention toward biochar-based nanocomposites because of the high-efficiency in reducing environmental contaminants [70-72]. Biochar-based nano-materials have a higher carbon content, larger surface area, and are more porous than raw biochar [73-75]. For example, biochar-supported nano-chlorapatite (BC-nClAP) realized improved Pb immobilization in polluted sediments compared to pristine biochar and nano-chlorapatite, as shown by Huang et al. [43]. Biochar pores can also absorb excessive phosphorus released by chlorapatite and reduce the agglomeration of chlorapatite [43]. BC-nClAP performed high conglutination efficiency of Pb and increased the OM content in a sediment-nano-composite system [43]. Many biochar-based nano-composites have been used in recent years due to its unique physical and chemical properties for removing contaminants [76], such as graphene-coated biochar [77], MgO-biochar nanocomposites [78], and carbon nanotube-coated biochar [79, 80], but only a few are applied in sediments [40]. Metal-based biochars have been applied to activate oxidants for OPs degradation, among which iron-based catalysts have outstanding advantages, such as being environmentally friendly, non-toxic, and highly efficient [73, 81]. Dong et al. [15] found that Fe_3O_4-RHB accelerated $SO_4^{-}\bullet$ formation and exhibited high efficiency in PAE degradation, and that magnetic Fe_3O_4–RHB can facilitate the achievement of environmental sustainability. Besides iron-based biochar as catalysts, copper, cobalt, mixed metals, and heteroatom-based biochars have been widely employed to remove pollutants in Fenton-like systems [82]. Thus, the application of biochar-based nano-composites in sediment remediation has considerable prospects. Overall, biochar modification or activation provides opportunities to improve the physicochemical properties of biochar and verify the feasibility for practical applications in sediment remediation. To obtain desirable biochar with suitable properties for pollutant sequestration or immobilization in sediment, comprehensive understandings of the interaction mechanisms between biochar and pollutants is imperative.

3. Remediation of HM-polluted sediments by biochar

HMs can enter natural aquatic systems due to natural or anthropogenic causes, and industrial effluents are considered the main source [83, 84]. It has been reported that 90% of HMs eventually accumulate and settle into sediments, resulting in much higher HM concentrations in sediments than in the overlying water [85]. HMs cannot be biological/chemical degraded naturally, and are, therefore, infinitely persistent in sediments, ultimately affecting the quality of the water and sediments. Therefore, HMs in sediments is a focus of pollutant monitoring [2, 29]. Multiple biochars have been used to promote stabilization and reduce the bioavailability of HMs (Cu^{2+}, Pb^{2+}, Cd^{2+}, Zn^{2+}, and Hg^{2+}) in sediment [8, 86]. HMs bound onto biochar are dependent on the biochar characteristics, as discussed previously. In this section, studies on the mechanisms, distribution, and toxicity analysis of HMs stabilization in biochar-sediments systems are reviewed (Fig. 2).

3.1 Adsorption of HMs in sediments

Metal cations (Cu^{2+}, Pb^{2+}, Zn^{2+}, and Cd^{2+}) are common pollutants in soil, water, and sediments, present as divalent ions or compounds, which exhibit similar characteristics and behaviors in sediments [54]. The valences of metal anion species vary in different sediments [54]. Biochar can immobilize and diminish the bioavailability of metal cations and oxy-anion compounds due to several different mechanisms, such as sorption, cation exchange, surface complexation, precipitation, and electrostatic interactions for metal cations, reduction, and complexation of metal oxy-anion compounds (Fig. 3) [43, 59, 87, 88]. Biochar as an immobilization agent can enhance the stability of Cu^{2+} and Pb^{2+} in sediments by surface precipitation [89]. However, the distributions of metal fractions were not changed because only minimal soluble and carbonate metals in sediments could be captured by biochar [90].

Moreover, the mineral components of biochar perform as additional sorption sites for metals through electrostatic reaction and ion exchange [63, 91], surface complexation [92, 93], and induce the precipitation of metals by releasing soluble ions, such as PO_4^{3-}, CO_3^{2-}, and SO_4^{2-} [63, 94]. In addition, the pH of a biochar-sediment system has a significant effect on HM immobilization, and the addition of biochar may change the sediment pH [61]. In highly acidic conditions, competition between H^+ and Cu^{2+} for active FGs on the biochar intensifies, weakening Cu^{2+} adsorption [95]. Dong et al. [96] showed that ion exchange processes between HMs and hydrogen were related to oxygen-containing FGs onto biochar, which led to changes in the sediment pH. In addition, a higher pH enhanced the sorption and/or precipitation of HM ions with ligands (CO_3^{2-}, SO_4^{2-}, Cl^-, OH^-, etc.) [97]. Liu et al. [98] investigated the distribution and speciation of Hg in biochar-amended sediment that was allowed to react for 1030 days. Through synchrotron-related methods, it was reported that the stabilization of Hg resulted from Hg-sulfide minerals and precipitation on or within biochar particles, which could co-exist with S, Fe, Cu, and Zn after switchgrass biochar remediation [39]. In fact, metals could gradually stabilize in sediments because of natural attenuation [29]. Biochar accelerated this process and redistributed the metals fraction in sediments as long-term incubations due to the formation of stable compounds [99, 100]. Therefore, sediment particles with high OM content, which is an additional source for biochar production, could also be used as the biochar feedstock to remove contaminants. Some laboratory experiments had proved the feasibility of applying biochar derived from sediment to environmental remediation [96]. Dong et al. [96] prepared biochar derived from sediments to adsorb HM, and this biochar showed higher fixation capacities for Cu^{2+}, Cd^{2+}, and Pb^{2+} through complexation, ion exchange, and pore-filling. Hence, HMs could be effectively retained by biochar through adsorption.

3.2 Distribution of HMs in aqueous phase

The distribution of HMs includes two portions: the fraction in porewater and in the overlying water of the water-sediment phase [26]. Biochar can reduce metal concentrations in surface water, thereby lowering the pollution risk [63]. Organisms in sediments can somewhat reduce the amount of HMs in porewater and overlying water by feeding; however, this method cannot permanently or completely remove HMs [43]. The distribution of HMs in the aqueous phase will be influenced by different environmental conditions during the process of sediment remediation with biochar [101]. Sequestration and immobilization alter the ionic species and considerably lower the HM concentration. In the sediment-aqueous phase, for instance, the pyrolysis temperature of biochar is negatively correlated with the HM-removal rate in sediment porewater, as reported by Zhang et al. [101]. When biochar is prepared at temperatures > 600 ℃, the removal efficiency of HMs (Cd, Cu, Ni, Pb, and Zn) remained constant in sediment porewater [101]. The Cu concentrations in surface water and interstitial water after in-situ rice husk biochar remediation were investigated by Que et al. [26]. The Cu reduction rates in surface water and interstitial water with RHB amendment were increased by 8–60% and 11.1–48.1%, respectively. Furthermore, the stable Cu fraction in interstitial water increased due to the increased pH of sediments and bonding with FGs on the surface of biochar to capture the acid-extractable Cu [26]. Biochar-based composites also exhibit high adsorption performances. Wang et al. [102] prepared a composite with improved properties and well-dispersed attapulgite on biochar and showed that it significantly lowered the As and Cd concentrations by approximately 79–82% and 36–44% in the overlying water, respectively, and by approximately 68–82% and 38–48% in the sediment porewater, respectively. HM morphology is highly correlated with its content in surface water and porewater [101]. Besides the chemical properties, the adsorption of metals also depends on particle size, and powdered biochar has a larger external surface area and shorter diffusive path lengths of pollutants than granular biochar. For example, at equal dosages, biochar with a particle size < 380 μm did not affect HM (Cu, Pb, Ni, Zn, Cd, and Cr) removal in sediment porewater, however, the removal efficiency slightly decreased with the application of biochar with a particle size > 380 μm [101]. When sediment conditions change or human and natural disturbances occur, HMs may be released into the overlying water from sediments and cause re-pollution, and biochar could sever the transfer of HMs in sediment-aqueous systems [2]. Hence, research on the distribution of metals in the aqueous phase is of great significance for appraising the effect of in-situ sediment remediation by biochar.

3.3 Change of HMs species in solid sediments

The bioavailability and biotoxicity of HMs are affected by the addition of biochar in sediment and are closely related to their speciation fractions and binding states [103]. Therefore, the HM concentrations and species in sediment that requires amendment should be comprehensively considered. A risk assessment of HMs in lake sediment was conducted by Yang et al. [104], and intensive human activities inputs and speciation distribution analysis showed that four metals (Cd, Cu, Pb, and Zn) were dominated by non-residual fractions in highly urbanized areas. This study revealed that the pH and total organic carbon (TOC) concentration of sediment are closely related to the chemical fractions of HMs [104]. Consideration of the HM species in sediment is beneficial for assessing the bioavailability and toxicity of HMs following the biochar amendment. Huang et al. [43] investigated the speciation of Pb^{2+} in solid-phase sediments, consisting of the acid-soluble fraction (F1), reducible fraction (F2), oxidative fraction (F3), and residual fraction (F4). The potentially available fraction of metals (F1 + F2 + F3) usually decreased with the addition of biochar to sediments and different chemical forms of HMs pose varying ecological risks [104]. The acid-soluble and reducible fractions were regarded as the weakest bound fractions and caused direct toxicity to organisms. The basis of sediment remediation is to transform the active chemical forms of HMs (F1, F2 and F3) into the stable fraction (F4) due to the low bioavailability of F4 [2, 105, 106]. Wang et al. [102] applied biochar/attapulgite composites for As and Cd immobilization in sediment and showed that after 60 days of treatment, the F1 of As and Cd were reduced approximately 43% and 11%, respectively. At the same time, F2 increased to different degrees, and F4 increased in sediment [102]. Therefore, the use of a biochar/attapulgite composite as a sorbent could effectively enhance As and Cd immobilization in sediment. The chemical fractions of HMs are correlated with sediment properties (total organic carbon, pH, and clay content). When biochar is added to polluted sediments, HM stability enhances due to the change in sediments properties. Activated biochar and biochar-supported nanocomposites all lower the percentages of F1 and F2 of HMs in sediments [107, 108]. The bioaccumulation and acute toxicity of HMs in sediments decreased after biochar remediation, and accordingly, the stability was increased. Chemical fractions of HMs are prone to transform among each other if environmental conditions change. For example, the presence of humic acid (HA) in sediments was conducive for the conversion of Cd due to the formation of ternary biochar-HA-Cd surface complexes [60].

3.4 Influence on HMs toxicity in sediment

HMs in sediments may be released into the water when the concentration is higher than the maximum sorption capacity of sediments or if the surrounding environment changes. In turn, released HMs can be taken up and accumulated by organisms in food chains, causing human health or environmental risks. Hence, toxicity assessments of HMs in biochar-amended sediments is imperative for measuring remediation efficacy.

Toxicity bioassays can elucidate the adverse impacts of in-situ sediment remediation [109]. The influences of the OM content in sediments have been proved, i.e., to control the transfer of HMs and reduce their bioavailability and toxicity [110]. The toxicity characteristic leaching procedure (TCLP) method is used to assess the potential leaching toxicity of HMs in sediments under acidic conditions and could be used to evaluate the performance of biochar for amended sediments. Liu et al. [59] discovered that several activated biochars improved the effects on Cd immobilization in sediment, as evidenced by the decreased concentration of

TCLP-extractable Cd (by approximately 13–23%). Biochar as a remediation agent has a high solid OM content which increases the OM content in sediments. BC-nClAP with a higher OM content could decompose organic acids, resulting in more phosphate being released from BC-nClAP to immobilize Pb by precipitation and reduce TCLP-leachable Pb [43]. With increased dosages of biochar in sediments, the stability of HMs generally increased and HM concentrations in the TCLP leachate correspondingly decreased [89].

Biological tests have also been an important element of appraising the detoxification feasibility of biochar for sediments, and an increasing number of researchers are conducting toxicity tests. A phytotoxicity test of sediments polluted by organic compounds and HMs with the addition of biochar, activated carbon, and multiwalled carbon nanotubes were assessed by Jośko et al. [33]. It was reported that the reduction of phytotoxicity of sediment after the application of biochar and other carbonaceous materials (activated carbon and multiwalled carbon nanotubes) was 27.5% and 17.7–28.0% (reduction of root growth inhibition) and 70% and 30–40% (reduction of seed germination inhibition), respectively. In addition, the reduction of sediment toxicity was positively correlated to the biochar diameter [33]. Other factors (nutrient binding and water availability) must be considered in the analysis of phytotoxicity in biochar-amend sediments [33].

To summarize, biochar can alter the physicochemical properties of sediments and reduce the toxicity of HMs to aquatic organisms in sediments. Through toxicity analysis, researchers could estimate whether pure biochar and modified biochar have positive effects on sediments or if the side-effects of remediation are better than detoxification. In fact, the toxicological risk of sediments may result from not only metals but also other highly toxic OPs, such as pyrethroids or cypermethrin [51, 111]. Toxicity analyses to determine the effects of biochar in polluted sediment require further research.

4. Remediation of OP-polluted sediments by biochar

As a remediation agent, biochar exhibits a high efficiency for removing OPs, mainly by decreasing their bioavailability and mobility through adsorption and degradation processes (Table 2). For OPs at different initial concentrations in sediment, removal efficiencies of 90–99.7% have been reported using different types of biochar (Table 3). Pristine biochar, chemically modified biochar, and biochar-based nano-composites are used to remove OPs. The effects of biochar may be correlated with interactions between microorganisms, biochar, and pollutants in the system, in which adsorption–desorption of pollutants, microbial growth, and metabolism, biodegradation of pollutants, and chemical degradation processes occur simultaneously [117, 118]. The effects of biochar on the detoxication of OPs include increasing the stability of OPs in sediment, decreasing the concentration of OPs, bioaccumulation in benthic organism, formation of hypotoxicity product, and acute toxicity as evaluated by bioassays (Fig. 3). As for OP removal from sediments, the roles of biochar mainly involve adsorption, catalytic degradation, and bio-degradation, which are summarized in the following section.

4.1 Adsorption of OPs

A significant source of organic contaminants in surface water may be heavily polluted sediments, commonly polychlorinated biphenyls (PCBs), PAHs, pesticides, and herbicides. It has been reported that biochar has a superior capacity to absorb OPs, due its high surface areas, microporosity, special FGs (e.g., carbonyl, hydroxyl, and phenolic groups), and charge characteristics [28, 45]. Biochar in sediments may absorb OPs mainly via H-bonding, π-π interaction, electrostatic interaction, and pore filling (Fig. 4). The adsorption mechanisms depend on the characteristics of OPs, the physicochemical properties of biochar, and the sediment properties. For example, Wang et al. [42] assessed the bioavailability of flubendiamide in the presence of biochar. Through sorption studies, partitioning into the noncarbonized OM of biochar and π-π electron donor–receptor action were the dominant mechanisms between biochar and flubendiamide. Sun et al. [46] found that the hydrogen bonds of fluridone and norflurazon could interact with N-containing FGs of biochar. H-bonding between herbicides and FGs on biochar surface most likely plays an important role in their interactions [119]. The sorption of OPs occurs through hydrophobic partitioning, as reported by Xiao et al. [52].

Pristine biochar consists of carbonized and non-carbonized fractions and has a high affinity to organic compounds. The sorptive ability of biochar in sediment system is higher than that of sediments alone, although the aging effect is enhanced over time [120]. Biochar can increase the free sorption sites, so that movement of OPs was prevented by adsorption process within biochar, affecting their bioavailability in sediments. Typical OPs such as pesticides, antibiotics, PAHs, and dyes have been detected in sediments [121, 122]; however, further research is essential to evaluate the applicability of adsorption for remediation of various types of OPs.

4.2 Catalytic degradation of OPs

Besides sorption, reactive removal is also a method of OP removal because biochar, as an efficient electron transfer medium, can motivate electron conduction and facilitate redox reactions. In natural sediments, reducing agents are omnipresent and abundant, such as aqueous Fe(II) and reduced sulfur species (bisulfide, HS^-, and polysulfides) [72, 123]. For instance, Gong et al. [44] investigated the abiotic reductions of trifluralin and pendimethalin by sulfides in anoxic sediments using three types of black carbon. The result suggested that adding biochar as sufficient reductants could significantly accelerate the abiotic reduction of dinitroaniline herbicides. Moreover, Fe(II)-mediated (abiotic) reactions are conducive to the reductive transformation of halogenated OPs [124]. Adding biochar to sediments contributes to sulfide reduction, which results in high-toxicity OPs being degraded to hypotoxic products by effective catalytic action, thereby reducing the environmental risk [50]. Chen et al. [47] found that biochar amendment significantly enhanced bacterial iron-reducing process. The electron flow during iron cycling was considered as a "mediator" to generate electrons, and high Fe(II) levels in biochar-amended sediments may result in high reductive reactivity, ultimately facilitating the reductive debromination of 2,2,4,4-tetrabromodiphenyl ether, as shown in anaerobic mangrove sediments slurries [124].

Biochar with reactive free radicals contributes to OP removal, mainly the combination of biochar with hydrogen peroxide (H_2O_2) or peroxydisulfate (PDS). Fenton (activated H_2O_2) and sulfate radical-based methods (activated $S_2O_8^{2-}$) are presently the most popular methods to degrade OPs [82]. A biochar-based catalyst took part in H_2O_2-based chemical oxidation to generate •OH to efficiently degrade 2-chlorobiphenyl because biochar containing persistent free radicals could activate H_2O_2 [125]. Redox-based advanced oxidation processes (AOPs) involving PDS ($S_2O_8^{2-}$) and peroxymonosulfate (PMS) activations have attracted increasing interest due to the low cost, ease of operation, and versatility [126-128]. For example, a Fe_3O_4 and bamboo biochar (BB) composite was used as a catalytic material to activate $S_2O_8^{2-}$ for degrading PAHs in $SO_4^{-}\bullet$-based AOPs, in which the presence of BB promoted electron transfer and accelerated the formation of Fe^{2+} to generate $SO_4^{-}\bullet$ [73]. In addition, catalytic testing showed that the Fe_3O_4–BB composite achieved the highest activity toward PDS after 24 h treatment [129], which was in agreement with the findings reported by Dong et al. [81]. Fe_3O_4-RHB can be used as an activator for the PDS oxidation process to rapidly facilitate the removal of PAEs in marine sediments [81]. Biochar-based catalysts facilitate electron transfer, simultaneously expedite the formation of active radical ($SO_4^{-}\bullet$, •OH), and thus, accelerate OP degradation.

4.3 Biodegradation of OPs

Biodegradation refers to the living microorganisms on biochar degrading OPs in sediment. Biochar with a high surface area could supply habitat and nutrients for microbial communities (bacteria, algae, and fungi), which is beneficial for pollutant biodegradation by immobilized microorganisms [50, 130]. Some microorganisms are known to generate enzymes to degrade various OPs [36]. Microbial degradation is a natural attenuation process controlling the OP content in polluted sediments [131, 132]. Yang et al. [50] enhanced the efficiency of removing phenanthrene in sediments using a combination of biochar and nitrate, and showed that PAH-degraders (*Thiobacillus* and *Stenotrophomonas*) settling in the biochar surface pores could utilize phenanthrene as a carbon source. Then, the added nitrate was captured by biochar and served as an electron acceptor to stimulate phenanthrene degraders. Microbial community structures vary with different OP concentrations and biochar dosages [117]. There has been minimal research regarding the utilization of biochar for OP removal through bio-degradation in sediment; however, this approach is promising based on the capability of other carbonaceous materials [19, 117]. For example, Jin et al. [129] found that *Pseudomonas* sp. isolated from Jiaozhou Bay wetland sediment, with preeminent environmental adaptability, was immobilized in coal cinder and chitosan beads for benzopyrene degradation. Therefore, coal cinder with a larger specific surface area and pore size, and a greater amount of oxygen-FGs as a degrading bacteria carrier, is the optimal choice for benzopyrene degradation [133]. Through a similar means, biochar may also be used as a bacteria carrier material for OP biodegradation in further experiments.

5. Potential risks of biochar to biological systems in sediment

Using capping technology with biochar for in-situ sediment, remediation should consider not only the binding and isolation performance but also the potential risks posed to biological systems. Previous studies on the toxicological effects of carbonaceous materials have focused on analyses at the microbial community, fauna, and indigenous plant levels [7, 54], and the potentials risk of biochar to plants in sediment have not been investigated. The effects of biochar on microbial communities and enzyme activity could indirectly change the efficiency of sediment remediation. Thus, the potential risks of biochar were concluded at indigenous microbial composition and benthic organism levels in this section.

5.1 Effect of biochar on indigenous microbial composition

Whether or not biochar causes changes in the microbial community and enzyme activity in sediment ecosystems has been a focus of sediment remediation [45, 47]. Microbial dynamics are an indicator for the remediation process, which is related to the stability of the sediment structure, nutrient cycling and respiration, water content, disease resistance, and carbon storage capacity [19, 45]. Additionally, enzyme activity is used to appraise microbial activity due to its direct relationship with sediment functionality [36]. Researchers are increasingly concentrating on the effect of remediation techniques on microbial communities and enzyme activity in sediments [26, 40].

Biochar alters the structure and abundance of microbial communities, and the micronutrient content in sediments. Biochar may also increase the microbial community abundance of sediments on account of its porous structure, which provides habitat and nutrients [8, 134, 135]. For example, Chen et al. [114] showed that biochar derived from fresh biogas slurry residue exerted a positive effect on the microbial community by either promoting an increase in abundance or reducing the magnitude of loss via mediating reductions of As^{5+} and Fe^{3+}. By contrast, biochar was perceived to have a negative effect on the abundance of organisms in *Desulfosporosinus* and *Pedobacter* genera. Microbial community abundance often (directly or indirectly) reflects the physical–chemical properties of sediments [136]. Bacterial abundance and diversity in control sediments groups were higher than those in sediments treated by activated biochar [36, 137]. The reported results suggested that the application of biochar affects microbial community abundance and diversity in polluted sediments.

Moreover, enzyme activity in sediments is susceptible to biochar. Some studies have proved that enzyme activity in sediments was influenced by the biochar content [36]. A high biochar concentration in sediments caused decreases in invertase and alkaline phosphatase activity, and extractable Zn and Cd fractions declined, whereas the pH and OM content increased in biochar-amended sediments [36, 108]. Biochar addition promoted N and P recycling, thereby affecting enzyme activity [36]. However, phenol oxidase activity was positively correlated with the biochar content in sediments [45]. Meanwhile, biochar also poses a risk of an increase in the number of harmful microorganisms in sediments, competing with beneficial microorganisms for nutrients and habitats, which would render sediment remediation much less effective [138]. To facilitate biochar technology to achieve better remediation effects and reduce the risks of biochar application to sediments, further research regarding the mechanisms between various biochars and microbial communities and enzymes are needed.

5.2 Effect of biochar on benthic organisms

Various benthic organisms dwelling in sediment are exposed to biochar and pollutants, which have impacts on the water-sediment layer. Benthic organism behaviors such as digging, feeding, and excreting (recognized as bioturbation) can be an indicator for toxicity of biochar and pollutants, such as worm, tubificid. Bioturbation causes the movement of capping material (sediment, biochar particles) and the release of pollutants from sediment to water [139, 140]. The available reports have proved that carbonaceous materials (activated carbon, charcoal) did not have significantly negative effects on fish embryos and molluscs [7]. Thus, biochar may cause negative effects on aquatic organisms, due to feeding on biochar particles or pollutants, and habitat changes resulting from the presence of biochar. When tubifex worms were exposed to sediment containing biochar (produced at 700 °C), the bioaccumulated concentrations of As and Cd in tissue decreased and the abundance and biodiversity of microbial community increased [90]. To the best of our knowledge, toxicity experiments of biochar on aquatic organisms in sediment are currently inadequate and further studies are needed.

6. Engineering applications of biochar in polluted sediments

Polluted sediment treatment technologies mainly aim to reduce or immobilize pollutants, thereby reducing the risk posed by pollutants to organisms or humans. Nowadays, common sediments remediation strategies include dredging and capping. Dredging treatments can maximize the removal of pollutants from aquatic environments [10]. The benefit of dredging engineering for dislodging Cu, Cd, and Pb from lake sediments after six years was proved by Chen et al. [10]. Dredging reduces the labile fractions of metals, increasing the metal-binding capacity of available sediment solids and retarding the leaching of metals from solids [10]. Nevertheless, disadvantages of dredging technology cannot be ignored, including: (I) dredging cannot fully remove all contaminants; (II) new pollutants will migrate downward and again cause pollution owing to the short maintenance time after sediment relocation; (III) the cost of this technique is high; and, (IV) original benthic communities in sediments may be destroyed [5]. Considering the above disadvantages of dredging, the application of low-cost biochar in in-situ capping to immobilize contaminants is feasible and has become a current research hotspot. Capping technology for a mature sediment remediation method has already been successfully used by the US Environmental Protection Agency [141]. Forming a capping layer with biochar and other materials (calcite, zeolite, and apatite), or mixing biochar with sediments as a capping, are two commonly used methods [139]. There are three commonly used engineering methods for biochar application in polluted sediments (Fig. 5): (a) A capping layer combining biochar with other material (gravel, quartz sand, or zeolite) is used for low-flow water or closed lakes; (b) two permeable geotextile layers containing the capping material is suitable for high-flow water; and, (c) Biochar mixing sediments as a capping layer is appropriate for shallow water or small-scale water bodies.

6.1 Coupling biochar with other capping materials

The majority of pollutants in sediment come from sewage [104]. Establishing an isolation barrier between sediment and sewage would effectively reduce the risk of sediment contamination and prevent the release of pollutants from polluted sediment into the water [142, 143]. Biochar is very useful as a sorbent for active capping due to its potentiality to limit the interactive diffusion of pollutants between sediment and water [53]. Biochar as capping material must combine other materials that contribute to biochar deposition (Fig. 5a). Traditional materials such as sand and natural zeolite have been applied for capping with biochar, which is suitable for low-flow water or closed lakes. Capping treatment, an shaped technology, employs inert materials to effectively prevent the release of contaminants from sediments by binding and sequestration, which can create a protective barrier against contaminants [95]. By isolating sediments from water, the pollutant concentrations in the overlaying water are maintained at safe levels [6, 20]. Zhang et al. [95] incorporated RHB into the sediments, aided by covering with a quartz sand layer to prevent the biochar floating, to reduce the concentrations of Cu^{2+} and 4-chlorophenol. The results indicated that a certain thickness of RHB could limit the release of Cu^{2+} and 4-chlorophenol from sediments through adsorption, flocculation, or ingestion by microorganisms. Besides, it has been demonstrated that the biochar layer could adsorb more NH_4^+-N from water than soil layer and inhibit its endogenous release from sediment in capping systems [21]. To improve the stability of these clean materials and preventing the forming of a biochar-sediment mixture, two permeable geotextile layers could fasten the reactive core, as shown in Fig. 5b. With geotextile layers, these capping materials can resist high-flow water and extreme weather [54]. Thus, a dissolvable or biodegradable geotextile is needed when using biochar as a capping amendment.

6.2 Mixing biochar with polluted sediments

Instead of capping with clean amendment materials, mixing biochar with sediment is another method used to immobilize contaminants and reduce their bioavailability and accumulation in food chains (Fig. 5c) [30]. The capping treatment can potentially couple with in-situ bioremediation and provide feasibility for field long-term capping treatment technology [16, 53]. However, long-term capping treatment may weaken the sorption capacity of biochar due to the lack of adsorption sites [37, 144]. Mechanical mixing is only feasible in shallow water or small-scale sediments. For large scale sediment remediation, however, the direct mixing of the capping material is not feasible. Mixing biochar with polluted sediments to allow full contact could maximize isolation and fixation and reduce contaminant mobility, toxicity, and bioavailability in sediments [39, 120], and this method is commonly used in laboratory experiments for removing pollutants from sediments [34, 89]. Many laboratory experiments have been conducted with continuous shaking to intensively mix biochar and polluted sediment [96]. Sediment remediation methods using biochar can reduce costs and labor.

7. Conclusion and future perspectives

Biochar exhibits a high efficiency for treating polluted sediments, which is in line with the principles of environmentally, sustainable development. In capping technology, the low-cost of biochar provides a nonnegligible advantage as a capping material to absorb and degrade pollutants, and biochar could perform comprehensive functions in sediment remediation (Fig. 6). Despite biochar being considered a promising material for pollutants immobilization or sequestration in sediment remediation, some research gaps and uncertainties remain that require further investigation and development:

(1) Optimizing biochar properties and enhancing its ability to immobilize target pollutants in sediment remediation work. Under various sediment conditions, the major influencing process parameters of engineered biochar are the biomass feedstock and production conditions. Pre-treatment of biomass and post-modification of biochar could be designed to improve its properties, which must be environmentally friendly and low-cost. Moreover, the commercialized preparation of biochar is currently retarded and rough, and, therefore, the technical procedures and equipment should be improved for delicacy structure biochar. The physicochemical properties of biochar produced from various waste biomasses and pyrolysis conditions are entirely different in contaminant management, thus, a database of biochar feedstock, preparation conditions, physicochemical properties, and function could be built for further utilization.

(2) Improving the utilization patterns of biochar could save labor and costs. Lake, river, and marine sediment deposits are very different, which influences pollutant behavior. The expenditure of capping mainly contains three aspects: 1) Cost of biochar; 2) Efficiency of biochar, and; 3) Capping layer design and establish. Activated biochar amendment could reduce the thickness of traditional capping technology but may increase the immobility of pollutants. New and innovative technologies for capping or mixing should be invented to remediate polluted sediments. In addition, the optimal layer thickness of biochar in capping should effectively isolate pollutants from sediments to the overlying water for decades to centuries under extreme weather conditions. Long-term monitoring of different layers at polluted sites could provide a valuable database for sediment remediation.

(3) Microbial communities and enzyme activity are easily affected by biochar in sediments. Further studies are needed to ascertain whether other aquatic organisms are sensitive to biochar. Toxicity experiments are necessary to determine the practicality of different biochars and synthesis methods for large-scale applications. Effects of biochar-amendment on the eco-function and health of sediments and food chains are critical issues that require further attention. The complex physical and chemical parameters of actual sediment is a challenge that requires further field experiments.

(4) Increasing attention has been paid to the application of biochar for emerging contaminants in sediment, such as endocrine disrupters, medicines, and personal care products. The potential application of biochar for metals that are present in trace amounts but exhibit persistent accumulation in the natural storage system, and many other anthropogenic

contaminants should be also considered, such as Ni^{2+}, Sb^{6+}, Co^{2+}, and radionuclide ions such as Sr^{2+}, Cs^{+}. Therefore, the synergistic treatment of different pollutants should be investigated in natural sediments with various coexisting pollutants.

(5) Biochar as an amendment for sediments polluted by organic or inorganic contaminants generally has complex mechanisms that require further exploration. Variations in the properties of sediments caused by biochar, the types of biochar (pristine or modified), the time of treatment, and the environmental conditions (pH, temperature, etc.) may all influence the remediation efficiency. A deep understanding of these parameters is important for further applications of biochar.

(6) Capping technology has the potential for cost-effective and long-lasting sediment remediation. However, most capping experiments with biochar have been confined to laboratory- or pilot-scales. Amendments have been conducted in laboratories without consideration of field conditions, and, therefore, additional field data and pilot-scale experiments representing the reliable practicability of biochar in the environment are essential. For improvement, other clean materials coupling biochar in caps which conform to the principles of ease of availability, low-cost, and negligible toxicity should be selected.

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