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PII: S0304-3894(23)02419-6

DOI: https://doi.org/10.1016/j.jhazmat.2023.133135

Reference: HAZMAT133135

To appear in: Journal of Hazardous Materials

Received date: 11 September 2023 Revised date: 5 November 2023 Accepted date: 28 November 2023

Please cite this article as: Weicong Wang, Shuangqi Wu, Xueqing Sui and Shuiping Cheng, Phytoremediation of contaminated sediment combined with biochar: feasibility, challenges and perspectives, *Journal of Hazardous Materials*, (2023) doi:https://doi.org/10.1016/j.jhazmat.2023.133135

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Phytoremediation of contaminated sediment combined with biochar: feasibility, challenges and perspectives

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Abstract

The accumulation of contaminants in sediments is accelerated by human activities and poses a major threat to ecosystems and human health. In recent years, various remediation techniques have been developed for contaminated sediments. In this review, a bibliometric analysis of papers on sediment remediation indexed in the WOS database between 2009-2023 was conducted using VOSviewer. We describe the development of biochar and plants for sediment contaminant removal. However, the single processes of biochar remediation and phytoremediation can be impeded by (i) low efficiency, (ii) poor tolerance of plants towards pollutants, (iii) difficulty in biochar to degrade pollutants, and (iv) biochar aging causing secondary pollution. Fortunately, combination remediation, realized through the combination of biochar and plants, can overcome the shortcomings of their individual applications. Therefore, we suggest that the remediation of contaminants in sediments can be accomplished by combining biochar with macrophytes and considering multiple limiting factors.

achieving efficient and sustainable sediment remediation, including complex sediment environments, interaction mechanisms of biochar-macrophyte-microorganisms, emerging pollutants, and integrated life cycle assessments, which can provide references for combined biochar and plant remediation of sediments in the future.

Key words: sediment, biochar, macrophytes, phytoremediation, co-remediation.

1 Introduction

The environmental challenges confronting humanity have become more severe as industries and agriculture continue to expand. The development of agriculture and industry has caused more damage to natural waters, and high levels of heavy metals (HMs) and organic pollutants (OPs) have been detected in substrates along with the input of pollutants through water infiltration and atmospheric deposition [1, 2]. Sediment is a naturally occurring substance that exists in rivers, lakes, and oceans and consists of sand, silt, clay, organic compounds, and inorganic minerals [3]. Pollution in the water system eventually affects the sediment because of its deep interaction with the overlying water. Pollutants accumulated in the sediment can be re-released into the overlying water due to changes in environmental or biological conditions, which causes the sediment to become both a sink and a source of pollutants. Pollutants in sediments can also bioaccumulate through the adsorption and enrichment of macrophytes, which can further threaten human health through the food chain, increase the risk of cancer in humans, damage the antioxidant systems of cells, and cause organ damage [4-6]. Therefore, control and remediation of sediment pollution have become urgent priorities.

The remediation of pollution in sediments faces serious challenges owing to their cumulative, immobile, and complex nature. Considering the cumulative, immobile, and complex nature of sediment pollution, traditional physical and chemical remediation approaches often fail. These approaches may only offer temporary mitigation of pollutant toxicity, as the fate of most reaction

products remains uncertain. Moreover, changes in environmental conditions can result in additional pollution, further complicating the remediation process. As a result, phytoremediation has surged into prominence as an in-situ remediation technology that is both ecofriendly and cost-effective [7]. For instance, a vast array of macrophytes, including Hydrocharis dubia, Phragmites australis, Echinochloa stagnina, Nelumbo nucifera, Typha latifolia, and other similar specimens have shown notable potential for the efficient accumulation and sequestration of HMs such as Arsenic (As), cadmium (Cd), and plumbum (Pb) from sediments [8-10]. During this process, pollutants in the sediment are rendered immobile by flora via the root, stem, and leaf cellular structures. This structure acts as a barrier, preventing hazardous components from entering the water column and accompanying the food chain [11]. However, several primary challenges impede the successful implementation of phytoremediation in sediment remediation, including the complexity of sediments, elevated pollutant concentrations, and diminished quantities of organic matter [12, 13]. For example, environmental availability of HMs can be increased or decreased by humic substances in the sediment to affect phytoremediation processes [13]; anoxic sediments promote the formation of large iron plaques by plant roots, preventing the uptake and translocation of HMs [14]; plants exposed to high concentrations of HMs can cause growth limitation, root tissue disturbance, structural alterations of epidermis and endodermis, etc. [15]. Therefore, it is imperative to devise solutions that mitigate these hurdles and enhance the effectiveness of phytoremediation.

Carbon-based adsorbents have attracted academic attention because of their complex pore structures and unique adsorption properties. Owing to its strong adsorption capabilities and cost advantages, biochar has been recognized as possessing superior quality and high efficacy for removing polluting substances compared with other carbonaceous alternatives [16]. It not only facilitates the modification of sediment physicochemical properties but also enhances sediment microbial and enzymatic activity [17]. Previous studies have demonstrated that biochar application can increase plant biomass and promote root mycorrhizal colonization [18]. For instance, adding 1% corn stover

biochar has been shown to significantly boost root length and dry weight in ryegrass, as well as increase peroxidase, urease, and lipase activity in the soil [19]. Although there have been numerous studies on the use of biochar in combination with phytoremediation in soils, research on its combined application in sediments remains relatively limited.

Currently, most studies have focused only on the application of single techniques to sediments, such as plants, biochar, advanced oxidation, and microorganisms, owing to their complex environments. However, stable and efficient sediment remediation cannot be achieved using a single remediation technique. Currently, biochar and plants can fill their individual deficiencies through synergistic interactions, which have been studied in soils but not in sediments. The synergistic relationship between biochar and macrophytes in sediments deserves attention as a sustainable strategy for improving the sediment environment, reducing sediment contamination, and decreasing ecological risks. In this review, the number of published articles and keyword co-occurrences of "sediment remediation" in the past 15 years were clustered by using VOSviewer (version 1.6.19, Netherlands), and the developments and limitations of traditional phytoremediation techniques and biochar in sediment remediation were analyzed. The feasibility of co-remediation of biochar and plant with their interactions in soils was discussed, and the challenges that biochar synergistic phytoremediation in sediments may face were explored. We expect to provide insights into the development of biochar combined with the phytoremediation of sediments and to attract more studies on sediment remediation.

2 Literature collection and analysis

Using a cycle of every 5 years, search the Web of Science database for the last 15 years (2009–2023) using "sediment remediation" as the keyword (the number of papers is 6089). The studies on "sediment remediation" have been on the rise in the last 15 years (Fig. 1a). From 2009-2013, the United States has consistently been the country with the highest number of papers posted. Since 2014, China has overtaken the United States as the top country in terms of the number of publications on sediment remediation per year (Fig. 1a).



Figure 1 (a) Top 10 countries in terms of number of papers and number of publications per year from 2009 to 2023 (as of June 30, 2023) based on the Web of Science database for the keyword "sediment remediation." (b) Cooccurrence of keywords in "Sediment Remediation"; (c) Mapping on density visualization of key words.

Using the VOS viewer to analyze the above 6089 papers for keyword clustering, the minimum range of keyword occurrences in the results was determined to be 20 (Fig. 1b). There are six main clusters (the color indicates the type of cluster and the size of the circle indicates the frequency of keyword occurrence). However, the analysis of the visual graph of keyword density reveals (Fig. 1c) that the hot spots in these papers mainly focus on the following four areas: (A) remediation mechanisms and conditions, including mechanisms, toxicity, and pH; (B) Removal of HMs, including Cd, Pb, and Cu; and (C) pollutant removal materials, such as biochar and nano zero-valent iron (nZVI). (D) Bioremediation technologies, including biodegradation, bacteria, and anaerobic degradation.

Number	2009-2013		2014-2018		2019-2023	
	Key Words	0	Key Words	0	Key Words	0
1	sediment	461	sediment	584	sediment	810
2	remediation	311	remediation	490	remediation	676
3	soil	249	heavy metal	350	heavy metal	524
4	heavy metal	236	soil	315	soil	378
5	PAHs	154	PAHs	251	removal	359
6	bioremediation	129	water	203	PAHs	311
7	removal	126	removal	192	water	289
8	water	120	degradation	159	bioremediation	238
9	adsorption	115	bioremediation	152	cadmium	226
10	sorption	114	adsorption	140	degradation	215
11	biodegradation	100	biodegradation	133	adsorption	211
12	degradation	87	sorption	130	biodegradation	181
13	bioavailability	83	cadmium	123	pollution	179
14	speciation	77	contamination	112	biochar	163
15	phytoremediation	70	bioavailability	106	contamination	161
16	extraction	66	speciation	101	bioavailability	150
17	groundwater	62	contaminated soil	100	phytoremediation	130
18	transport	62	pollution	90	immobilization	122
19	metals	61	toxicity	87	toxicity	114
20	phenanthrene	60	phytoremediation	84	speciation	110

Table 1 The distribution of the top 20 most common keywords in the literature on sediment remediation from 2009 to 2023.

O is the Occurrence Number of the keyword.

According to the types of sediment pollution (Table 1), HMs have been the most frequent term over the years, followed by polycyclic aromatic hydrocarbons (PAHs), indicating that the focus of pollution removal from sediments in recent years has been on HMs and PAHs. Among the remediation techniques, bioremediation appears most frequently, followed by adsorption, sorption, degradation, and biodegradation, indicating that bioremediation and adsorption have become key technologies for current substrate remediation in recent years. Phytoremediation has been a popular research topic among bioremediation technologies since 2009. Regarding the adsorption technology, no adsorbent materials appeared in the top 20 buzzwords from to 2009-2018. Black carbon and activated carbon

(AC) were the dominant materials used for the adsorption of substrate pollutants during this period. However, the frequency of biochar was at 163 occurrences by 2019-2023, indicating that biochar has gradually become a major research focus for sediment adsorption materials.

With emphasis on sediment environments, various types of research have been steadily increasing. For example, AC was first applied in 2009 for the in-situ removal of polychlorinated biphenyls (PCBs) from contaminated substrates [20], and AC was first demonstrated in 2011 to reduce PCBs uptake by benthic organisms in an actual contaminated river [21]. In addition to the use of AC, an increasing number of studies have been conducted on the remediation of sediment pollution with other materials; for example, marine (harbor) sediments were restored for the first time using loess balls [22]. In-situ remediation of Hg and methyl-Hg in sediments via AC [23]. First, the adsorption of polycyclic aromatic hydrocarbons (PAHs) onto hardwood was evaluated [24]. However, single remediation techniques have limited effects on sediments. Consequently, scientists have begun experimenting with a combination of techniques that can improve the efficiency of restoration and overcome the shortcomings of individual techniques. In 2020, Che et al. first used an artificial oxygenation treatment combined with the introduction of exogenous microorganisms to purify urban river sediments and achieved a maximum removal efficiency of 22.3% of organic matter from sediments with no effect on the overlying water quality [25]. In addition to co-remediation techniques, sediment research has been extended to pollution removal in complex systems. In 2022, Sun et al. first used rice straw biochar to inhibit the release and transport of PAHs from the sediment of a rice-crab co-culture system, which reduced the enrichment of PAHs in crab and rice by 38.2-69.5% and 32.1-97.6%, respectively [26]. The development of environmentally friendly, economically viable, and long-term effective radioactive contamination removal methods is also a key focus of the current research. Vettese et al. used six contrasting treatment approaches to achieve removal efficiencies of 89 to >99% for U and 65-95% for Sr. Reduction and biomineralization mechanisms should be targeted during the remediation process to facilitate radionuclide-mineral incorporation [27]. In summary, during the

sediment remediation process, the above studies indicate that multi-technology associations, pollution characterization in complex environments, and persistent pollutants are key considerations for research.



Figure 2 The first innovations in the field of sediment remediation from 2009–2023 [20-32].

Based on the results of a cluster analysis of the literatures, we will discuss phytoremediation and biochar remediation of sediments separately in the following.

3 Phytoremediation of contaminated sediments: current research and limitations

Pollutants can be absorbed from the sediment and accumulated in the macrophyte, this process is called sediment phytoremediation [33]. Phytoremediation, a green and effective remediation technique, has been widely used in the in-situ remediation of contaminated sediment [34]. Compared with the

traditional physical, chemical, and microbial methods, phytoremediation is an effective method to prevent toxic pollutants from entering the food chain and to protect biodiversity, with the advantages of low cost, sustainability, no secondary pollution, and effective landscaping [35, 36].

3.1 Phytoremediation of sediment with heavy metals

Phytoremediation of pollutants is usually achieved through various physiological processes [37]. [38]. For example, HMs are immobilized and transformed mainly through absorption into plants by the roots [39, 40]. A large amount of evidence indicates that the adsorption of HMs from sediments by macrophytes occurs mainly through the root system, where HMs are taken up by plant roots through apoplastic transport (specific ion channels or carriers) and symplastic transport (interstitial cells) [41, 42]. Generally, there are two transfer pathways after HMs enter a plant: direct storage in plant root cells, and translocation to the aboveground parts of the plant (stems, leaves, etc.) (Fig. 3) [43, 44]. The accumulation of HMs in the roots has been observed in a variety of plants. Several studies have shown that HMs can enter plant root cells via Ca and nutrient uptake channels (N, P, and K) in plants [45]. After the entry of HMs into cells, they are mostly present in the cell wall and cell-soluble fraction of plant cells, but less so in organelles [46]. Following the adsorption of HMs into plant roots, some of them are transported to the aboveground parts. Several studies have shown that the accumulation of HMs in non-hyperaccumulating plants follows the order of stem < leaf < root [47]. This phenomenon can be explained by an increase in stem and leaf biomass with increasing plant growth, which results in the dilution of HMs concentrations in plant tissues [48]. Subsequently, an increase in exposure time causes large amounts of HMs to accumulate in the roots, causing severe physiological damage to plant roots and reducing their ability to transfer HMs to the aboveground parts of the plant [49]. Stems play a major role in the transport of HMs in plants, causing them to be transferred to leaves. Large amounts of HMs are stored in the vesicles of leaf epidermal cells by translocation and are then immobilized in the soluble fraction of cells by compartmentalization of the vesicles. When HMs are immobilized, some amino acids and organic acids in plant cells form metal complexes by complexation with HMs

ions through atoms such as O, N, and S, for long-term storage in vesicles [50].

Identifying and cultivating hyperaccumulators is another phytoremediation method. The hyperaccumulation ability of HMs in plants is generally described by the bioaccumulation factor (BAF) or translocation factor (TF) [51]. Larger BAF values indicate that the plant is more capable of enriching HMs, and larger TF values indicate that the plant is more capable of transporting HMs from the roots to the stems and leaves [52]. Generally, if the BAF and TF values of a plant for HMs are greater than 1, the plant is regarded as having a hyperaccumulation capacity for HMs [53]. El-Amier et al. demonstrated that *E. stagnina* can be used as a hyperaccumulator of Pb in the extraction and HMs accumulation capacity of the three emerging macrophytes (*E. stagnina*, *P. australis* and *T. domingensis*) tissues [54]. From the accumulation of HMs in the roots and shoots, the value of BAF > 1 for Cd, Mn, Pb, and Ni in all three macrophytes indicated that the three plants could save HMs effectively. The values of TF were < 1, except for *E. stagnina* for Pb [54]. This also demonstrates that HMs are preferentially fixed in the roots rather than being transferred to the shoots.



Figure 3 Mechanisms of phytoremediation of OPs and HMs.

3.2 Phytoremediation of sediment with organic pollutants

Phytoremediation of OPs focuses mainly on PAHs, polychlorinated biphenyls (PCBs), and

petroleum hydrocarbons (PHCs). Currently, plants used for the restoration of OPs include alfalfa (*Medicago sativa*), Chinese bushclover (*Lespedeza cuneate*), everlasting pea (*Lathyrus sylvestris*), eelgrass (*Zostera marina*), etc [55-58]. OPs are usually effectively degraded by plants through direct uptake, the release of root exudates, and the combined action of rhizospheric microorganisms (Fig. 3) [59, 60] The uptake of OPs by plant roots is much smaller than that of inorganic pollutants and HMs. Huesemann et al. showed that only 0.32% and 0.17% of the initial mass of total PAHs was present in the sediment after 60 weeks of treatment in eelgrass roots and shoots, respectively [55]. Similarly, only 0.29% of the total PCBs mass was detected in the roots but not in the shoots [55]. This indicated that plant uptake was not the main mechanism of OPs by plants. Previous studies have shown that root exudates and plant enzymes promote the removal of OPs from environmental media by a variety of plants [61]. For example, root secretions enhanced the degradation rates of PCB 44, 66, and 118. Plant secondary metabolites (PSMs) in root exudates (e.g., flavanone, flavone, isoflavone, 7-hydroxyflavanoe, and 6-hydroxyflavone) can also effectively increase the degradation efficiency of the seven PCB congeners and have also been used as growth substrates for microbial consortia [62].

The effects of plant remediation depend on the tolerance of the plant species to organic matter. For example, low concentrations of PAHs in the sediment (ranging from to 8-80 mg kg⁻¹) were effective in promoting the growth of *Potamogeton crispus*; however, the growth of plants was inhibited when the concentration of PAHs was increased to 180 mg kg⁻¹ [63]. Meanwhile, the presence of *P. crispus* increased the removal of phenanthrene and pyrene from the sediment by 18.3–34.1 and 14.1– 27.8%, respectively [63]. The increase in the removal efficiency was attributed to the presence of *P. crispus* in addition to its higher tolerance to PAHs and increased polyphenol oxidase activity. However, the effectiveness of phytoremediation is not related to tolerance. Numerous studies have shown that appropriate plant density is the key to improving the effectiveness of restoration. Nutrient availability for macrophytes depends on plant species and density. Faster plant growth and increased total root

surface area promote the dissipation of PAHs in sediment [64].

	C - 1:	T	D - 11	Descrite	Def
Macrophytes	Seament	Treatment	Pollutants	Kesuits	Kel
	types	duration			
Hydrilla	River	50 d	Cd	Cd accumulation in plants were $42.65 \pm$	[33]
verticillata	sediment			$0.52 \text{ and } 107.82 \pm 1.86 \text{ mg kg}^{-1} \text{ in soils}$	
				with 20 and 50 mg (kg DW) ⁻¹ of Cd,	
				respectively.	
Typha	River	-	HMs	The plants showed $BAF > 1$ and $TF > 1$	[65]
latifolia	sediment			for Zn.	
Limnobium	River	7 d	Cr	The accumulation of Cr in plant	[40]
laevigatum	sediment			increased from 20 ± 5 mg/kg to $2066 \pm$	
				216 mg/kg.	
Cannabis	River	2 months	Cr, Ni, Cu	The removal rates of Cr, Ni, and Cu on	[66]
sativa	sediment			a dry mass basis were 0.4%, 0.6%, and	
				0.9%, respectively.	
Acanthus	Mangrove	-	As, Pb, Cr, Cu, and	HMs content in plants (mg/kg). Leaf:	[67]
ilicifolius	sediment		Zn	Zn (107) > Pb (28. 7) > Cu (16.9) > As	
				(11.2) > Cr (4.99). Root: Zn (104.32) >	
				Pb (27.02) > Cu (15.29) > As (10.39) >	
				Cr (3.80).	
Zostera	Ocean	60 weeks	PAHs and PCBs	The concentrations of PAHs and PCBs	[55]
marina	Sediment			decreased by 73% and 60%,	
				respectively.	
Rhizophora	Mangrove	90 d	PAHs	Phytoremediation (60.76%) was better	[57]
mangle	sediment			than natural attenuation (49.57%)	
Potamogeton	River	54d	phenanthrene and	The removal rates of Phe and pyrene	[63]
crispus	sediment		pyrene	were increased by 18.3 - 34.1 and 14.1	
				- 27.8%, respectively.	
Vallisneria	River	54d	phenanthrene and	The dissipation ratios of Phe and pyrene	[64]
spiralis	sediment		pyrene	were up to 53.3-59.6% and 50.0-	
				53.6% in planted sediments	
Phragmites	Sediment	90 d	Pentachlorophenol	The removal rate was $90.35 \pm 0.03\%$.	[68]
communis			(PCP)		
T. orientalis	Sediment	90 d	РСР	The removal rate was $99.23 \pm 0.02\%$.	[68]
Scirpus	Sediment	90 d	PCP	The removal rate was $99.33 \pm 0.01\%$.	[68]
validus					

Table 2 Phytoremediation of sediment by different macrophytes

S. validus	Freshwater	18 months	PBDE	congener	The removal rate of was 92.84%.	[69]
	sediment		decabromodiphenyl			
			ether (BI	DE-209)		

3.3 Limitations and prospects of phytoremediation

Pollutants can be effectively absorbed and transformed by plants and discharged from the water through plant collection. However, some problems are currently associated with phytoremediation in sediment remediation.

Currently, most plants have a low TF for HMs, although they have certain HM removal effects. For practical environmental restoration, it is essential to ensure plant species richness and enhance the effects of phytoremediation. Therefore, several issues need to be addressed, such as enhancing the BAF and TF of common plants (non-hyperaccumulators) for pollutants, realizing the transformation of common plants into hyperaccumulators, and enabling common plants to accumulate higher amounts of HMs.

A long duration is a limitation of phytoremediation. Plant growth depends on the environment, climate, microorganisms, and other factors, and plant tolerance to pollutants must also be considered. Single phytoremediation cannot cope with severely contaminated environments, which can cause growth inhibition or plant death. Therefore, improving plant tolerance and adopting simple methods to reduce the impact of pollution on plant growth are the focus of current research.

The success of phytoremediation depends on the uptake, translocation, detoxification, and sequestration of the pollutants. Much of the current research focuses on identifying hyperaccumulators and suitable plant species for specific environmental conditions. However, there are still some problems that need to be addressed to achieve successful phytoremediation, including the role of different transporter proteins in vacuole segmentation, mechanisms of phytodetoxification, costs, benefits, and the long-term and environmental impacts of phytoremediation.

Therefore, the focus of phytoremediation should be on improving the efficiency. The search for suitable hyperaccumulators is a common solution for this problem. However, how to make conventional plants obtain better pollutant accumulation ability and higher TF is the focus of current and future research.

4 Remediation of sediment by biochar

4.1 Mechanisms of pollutant removal from sediments by biochar

During biochar adsorption, various physicochemical properties determine its performance and can have different effects (Fig. 4). The specific surface area (SSA) and pore size distribution (PSD) are key factors affecting the adsorption performance of biochar for sediment pollution. In general, an appropriate PSD offers mass transfer paths for pollutants, and a large SSA provides more active sites for adsorption [70]. A larger SSA results in a biochar with a stronger adsorption capacity and faster adsorption rate. Soares et al. confirmed the greater remediation capacity of biochar from larger SSAs by comparing the temporal variation of remediation in sediments by biochar generated from sugarcane pyrolysis at 350 °C (BC350), 550 °C (BC550), and 750 °C (BC750). Compared to BC350, the SSA of BC550 improved by 658% and BC750 improved by 1211%, respectively. The As content in the BC750 treated sediment was much lower than that in BC350 and BC550 after 180 d of remediation [71]. High temperatures can increase the SSA of the biochar. Under high-temperature conditions, biomass releases its original volatile components and tends to form graphene-like structures, leading to larger SSA [72].

However, a large SSA does not always result in high adsorption performance. The adsorption capacity and pore structure are also frequently connected. Bentley et al. found that the adsorption capacity of biochar for organic micropollutants (OMP) was positively and negatively correlated with microporous and non-microporous SSA, respectively [73]. Generally, micropores (pore diameter < 2 nm) on biochar provide the principal adsorption sites, mesopores (2 nm < pore diameter < 50 nm) provide both active sorption sites and intraparticle diffusion pathways, and macropores (pore diameter > 50 nm) mainly affect diffusion pathways [70].



Figure 4 Mechanism of biochar treatment of sediment pollutions.

In addition to the physical adsorption, the surface functional groups and chemical properties of biochar play crucial roles in the adsorption and degradation of sediment pollutants. Pollutants are attached to biochar through mechanisms such as ion exchange, co-precipitation, complexation reactions, and cation- π interactions [74, 75]. For example, the adsorption of phosphorus in sediments by peanut shell biochar (PSB) is mainly attributed to the ligand exchange of PO4³⁻ with -COOH and - H [76]. In addition, multiple mechanisms coexist for the adsorption of sediment pollutants by biochar owing to the complex elements and functional groups on its surface. By investigating algal biochar (ABCs) on SMX-contaminated marine sediments, Zhao et al. found that SMX sorption on the ABCs was dominantly through cation bridges, negative SMX⁻ could bridge with the ABCs via the abundant cations (e.g., Ca²⁺ and Mg²⁺) in seawater, thereby forming surface ternary complexes (SMX⁻-cations-biochars) [77]. Next to produce the effect are the negative charge-assisted H-bond, π - π electron donor-acceptor interactions, and pore filling. The–SO₂–NH– of SMX and the surface carboxyl or hydroxyl groups of ABCs can share protons to form (–)CAHB in seawater (pH 8.0). Also, the abundant graphite

C and graphite N in ABCs have been confirmed to form π - π electron donor-acceptor (EDA) interactions as donors with the acceptors of SMX heterocyclic N [77].

The functional groups of biochar can also play a direct role in the degradation of pollutants. For example, both hydroxyl (HO \cdot) and superoxide anion (O^{2- \cdot}) radicals have a strong ability to degrade pollutants [78]. Previous studies have shown that the presence of electron-rich groups such as hydroxyl (-OH) and carboxyl (-COOH) groups on the surface of biochar promotes the production of large amounts of HO \cdot [79]. 3.0 g/L of red algae biochar (RAB) can produce enough HO \cdot to effectively degrade 4-Nonylphenol (4-NP) with a conversion rate of 67% [80].

4.2 Biochar application for sediment remediation

Numerous experiments have demonstrated that biochar effectively adsorbs and immobilizes pollutants in situ during sediment remediation. Different raw materials such as straw, wood chips, corn cobs, sludge, etc. (Fig. 5) impart various physicochemical properties to biochar. However, unmodified biochar has limited removal of pollutants and needs to be modified or composited (Fig. 5). This section will focus on the removal of sediment pollutants by biochar composites, modified biochar, and biochar as a carrier for other substances.





Figure 5 Raw materials and classification of biochar [79, 81-86]

4.2.1 Modified biochar

Currently, the biochar removal capacity for environmental pollution during sediment remediation is still low. Biochar is usually modified to enhance its ability to remediate sediment pollutions [87, 88]. Currently, physicochemical modification methods include steam activation, gas modification, and chemical activation (KOH, H₂SO₄, KMnO₄, etc.) heteroatom doping, etc. [89-91]. Modified biochar typically has higher ratios of H/C, O/C, and (O + N)/C than native biochar. Additionally, it possesses

a larger specific surface area, greater surface charge, and more functional groups [92-95]. For example, nitrogen (N)-containing functional groups provide biochar with multifunctionalized environmental activities, including catalysis, nutrient supply, and the ability to adsorb HMs and OPs. The interaction of N-rich biochar with pollutants primarily depends on the type of N functional groups. During the synthesis of biochar, quaternary, pyridinic oxide, pyrrolic, and pyridinic types of N are mainly retained, and planar pyridinic-N and pyrrolic-N contents mainly determine the biochar's catalytic performance [81]. For the adsorption process, N-containing functional groups may facilitate the adsorption of OPs through π - π interactions and Lewis acid-base interactions. In other words, N doping can enhance both the physical and chemical properties of biochar to improve the removal performance of pollutants.

The spontaneous release of metal ions from sediments can modify biochar to some extent. Gao et al. found that the spontaneously released Ca^{2+} and Fe^{2+} from the substrate could be loaded into peanut shell biochar (PSB) in the form of Ca–O and Fe–O to obtain sediment-triggered biochar (P-PSB) and enhance the adsorption of phosphorus from the sediment [76]. The adsorption capacity of P-PSB is 2 times higher (10.72 mg/g) compared to PSB (adsorption capacity of 4.85 mg/g), and its high adsorption capacity is also due to the fact that P can form a Ca-O-P surface complex with Ca²⁺ on the surface of S-PSB, generating surface precipitation of Ca₅(PO₄)₃(OH) and CaHPO₄ [76].

4.2.2 Biochar composites

The adsorption performance of biochar is often limited by its underdeveloped porous structure and limited surface functional-group content. Researchers have proposed several approaches to overcome the disadvantages of biochar with limited adsorption performance, including modification with materials such as graphene, Fe₃O₄, and nZVI etc [96-98]. Compared with biochar, the biocharbased composite had a larger SSA and higher porosity. For example, the SSA of a composite composed of 40% MnMoO₄ nanoparticles (MM) and 60% microalgal biochar (MM40BC60) was larger than that of microalgal biochar, whereas maximum tetracycline removal was observed in the MM40BC60 composite [99]. However, SSA and porosity are not the only indicators of enhanced adsorption

performance, because the number of oxygen-containing functional groups, pH, and cation exchange capacity (CEC) are important factors that dominate pollutant adsorption [100, 101]. For example, iron-coated fine empty fruit bunch biochar (IC-EFBB) had a higher pH (9.910 \pm 0.01a) and functional group (OH:18 \pm 0.00a cmol₊kg⁻¹ and CO:20 \pm 0.00a cmol₊kg⁻¹) content compared to uncoated fine empty fruit bunch biochar (EFBB), and the adsorption capacity of IC-EFBB (142.86 mg g⁻¹) for Pb was higher than that of EFBB (103.09 mg g⁻¹) [102]. This indicates that the HM adsorption capacity of biochar can be increased by Fe modification.

Extensive research has been conducted to develop effective and simple methods for producing biochar composites. Clay-based mineral materials can increase the adsorption capacity of biochar of their high CEC, high SSA, and active hydroxyl groups [103]. Attapulgite/biochar composites can effectively reduce the content of HMs in the overlying water, pore water, and sediment solid phase compared with biochar [104]. The increased adsorption can be explained by the higher SSA and pore volume (PV) of the attapulgite/biochar composites, which provide more opportunities for the interaction of HMs with the composites. Similarly, more O-containing functional groups provide more adsorption sites on the composites, which can bind HMs (As and Cd) via surface complexation [104]. Biochar composites with excellent adsorption capacity can not only mix with sediments but also act as capping materials to avoid the release of sediment pollutants. For example, nano-Fe₂O₃@biocarbon composite (nFe₂O₃@BC) capping reduced the Cd release concentration from the overlying water by 1.6–11.0 times compared with biocarbon capping, while improving the disadvantage that biocarbons are not suitable for acidic and neutral water (pH 3, 5, and 7) [105]. Biocarbons plays a role in limit aggregation and enhance the reactivity of nano-Fe₂O₃ [105].

Similarly, biochar composites can be used as activators to improve the pollutant degradation efficiency. It can generate environmentally persistent free radicals (EPFRs) by effectively activating H₂O₂, peroxydisulfate (S₂O₈^{2–}), peroxymonosulfate (PMS, HSO₅[–]), percarbonate, etc. [106], such as oxygen-, carbon-, and oxygenated carbon-centered radicals, these radicals are powerful and promising

oxidants for the degradation of OPs [107, 108]. For example, the addition of Fe-Ce/water caltrop shell biochar (WCSB) composites to the persulfate (PS) system can accelerate the production of HO· and SO_4 , and with increasing the amount of Fe-Ce/WCSB composites from 0.4 to 1.7 g L⁻¹, the degradation rate of PAEs in the sediment increased sharply, with a maximum of 85% [79]. The oxygen vacancy concentration and oxygen mobility are the main reasons for catalytic activity. The improvement of Fe-Ce/WCSB degradation efficiency might be due to the excellent electron conversion between Ce⁴⁺/Ce³⁺ and Fe³⁺/Fe²⁺ redox pairs, which generated oxygen vacancies and triggered the reaction to produce SO₄, releasing iron or cerium ions and improving the catalyst efficiency [79]. With the increase of biochar composites, more redox centers will generate to activate PMS, thus enhancing the production of strong electron donors, specifically, SO₄⁻⁻ and ·OH [109, 110]. In addition, biochar is often used in such reactions to provide a special interphase surface where direct surface electron transfer to the pollutants and PS molecules also occurs [111]. According to the Haber-Weiss mechanism, efficient electron transfer can facilitate catalytic and redox reactions at the biochar-metal interface, leading to the degradation of pollutants [112, 113].

4.2.3 Biochar as a carrier

Although nanomaterials are highly efficient for sediment remediation, their application in reaction systems is severely affected by their tendency to polymerize [114]. Biochar can effectively prevent the agglomeration of nanocatalysts and improve their reactivity. Rice husk biochar, a support material for nanohydroxyapatite (Ca₁₀(PO₄)₆(OH)₆, nHAP), can effectively distribute nHAP to form a composite material (nHAP@BC) and enhance its adsorption capacity. The average pore size (2.88–29.2 nm) and SSA (92.30 m²/g) of nHAP@BC were higher than those of biochar (average pore size of 2.46 nm and SSA of 1.53 m²/g) [82]. The increase in pore size and SSA may be due to the formation of more pores by the interlayer-loaded nHAP. Subsequently, Zhao et al. introduced phosphate-solubilizing bacteria (PSB) into nHAP@BC to investigate their effects on Cd removal from sediments. Based on XRD and FT-IR analyses, three mechanisms of action were derived for the immobilization

of Cd. First, Cd is immobilized on the composite material by adsorption. Second, Ca^{2+} in nHAP forms Cd-HAP (Ca_{5-x}Cd_x(PO₄)₃(OH)) with Cd via ion exchange. Finally, PSB achieve Cd immobilization by converting nHAP into soluble phosphate, which reacts with Cd to form Cd-containing precipitates (Cd₂P₂O₇, Cd(H₂PO₄)₂) [82].

In environmental remediation, nZVI often exhibits degraded properties owing to defects such as precipitation, passivation, aggregation, and limited electron transport [115]. Therefore, it is necessary to have carriers with improved performance. Biochar is an effective support material for nZVI. Previous studies have also demonstrated that biochar can enhance the removal of pollutants by nZVI through electrostatic attraction, ion exchange, surface complexation with functional groups, and π - π electron donor-acceptor interactions [116]. For example, the dispersion of naked oat straw biochar to loaded with nZVI (BC-nZVI) was better than that of nZVI, proving that the composite could effectively reduce the aggregation and passivation of nZVI [117]. In addition, nZVI/BC showed excellent chemical stability and regenerative properties. Xu et al. found that the removal of Cr(VI) from nZVI/BC was still 56.85 mg/g (the first cycle was 63.29 mg/g) after the fifth cycle [118]. With increasing reaction time, the interaction mechanism of Cr(VI) with nZVI/BC was followed by adsorption (complexation and electrostatic interactions), reduction, and coprecipitation processes. The oxidation of Fe^0 to Fe^{2+}/Fe^{3+} (corresponding to the reduction of Cr(VI) to Cr(III)) and further coprecipitation of Fe⁰/Fe²⁺ with Cr(III) ions, followed by the disappearance of O=C-O and–OH groups after the reaction, also indicate the involvement of oxygen-containing functional groups in the removal of Cr(VI) [118].

4.3 Limitations and Prospects of Biochar Remediation

Although biochar has achieved better remediation results in sediment treatment and is an excellent sediment capping material, current research has some limitations.

Raw materials significantly influence the physicochemical properties of biochar. Although modification and compounding can improve the performance of biochar, the cost issues must be

considered. Impurities in biochar also cause environmental pollution in the sediment.

The sediment environment is more complex than the soil environment. The effectiveness of biochar remediation is influenced by various conditions such as the initial concentration of pollutants, ionic strength, natural organic matter in the sediment, contact time, and mixing method. In addition, the environmental conditions of sediments are influenced by hydrological conditions. Therefore, the remediation mechanisms of biochar in complex environments and the influences of various environmental factors require further investigation.

Biochar remediation is characterized by easy adsorption and difficult transformations. Biochar can easily adsorb different pollutants in sediment; however, its adsorption capacity is limited because of its complex sediment environment. Meanwhile, biochar is not easily transferred from the sediment after adsorption saturation, and the adsorbed pollutants remain in the sediment. With the aging and oxidation of the biochar, the adsorbed pollutants are released back into the sediment.

In the process of sediment remediation, current research has focused on the modification of biochar to improve its adsorption capacity and its ability to uptake and transform pollutants. Similarly, biochar combined with other remediation methods has begun to appear in sediment remediation. Exploiting the synergistic efficiency of multiple restoration methods is the focus of current and future research.

5 Biochar-phytoremediation technology

Due to the limitations of biochar remediation and phytoremediation, it is possible to combine these two techniques to address complex sediment environments. Biochar has been widely used for soil improvement because it can effectively enhance soil habitats and promote plant growth and development [119-121]. Plants can influence the adsorption and desorption processes of biochar through the release of root secretions, which can improve pollutant removal efficiency and increase the lifetime of biochar. However, the current research has combined the two techniques only in some settings and less in sediments.

5.1 Feasibility of biochar synergistic phytoremediation of sediments

Although relatively few studies have been conducted on the remediation of sediment pollution using biochar combined with phytoremediation, biochar has been shown to be effective in promoting the efficient removal of soil pollutants by terrestrial plants. Lebrun et al. confirmed that pinewood biochar can improve soil fertility and promote the growth of three willow species (*Salix alba, Salix viminalis and Salix purpurea*) by increasing pH and electrical conductivity (EC) and reducing lead mobility [122]. Kiran and Prasad found that a 5% Prosopis juliflora biochar (PJB) treatment reduced the accumulation of Pb in the root system of Ricinus communis by 59%, which increased plant height, leaf diameter, number of nodes, and number of leaves of Ricinus communis by 76%, 217%, 82%, and 69%, respectively [123]. It has also been shown that the biochar-Prosopis laevigata synergy can effectively reduce HMs, such as Cu, Pb, and Cd, in mine soils [124]. Fig. 6 shows the interaction mechanisms of biochar combined with phytoremediation. All of the above examples of successful biochar combined with phytoremediation in soils lay the foundation for the co-remediation of sediments.



Figure 6 Technologies for biochar synergistic phytoremediation of sediment pollutions.

5.1.1 Promote the uptake of pollutants by plants

Biochar can increase the uptake of HMs by plants through the adsorption of soil nutrients and the reduction in competition between HMs ions and nutrients [125]. Rees et al. showed in their study that the increase in hyperaccumulation was caused by the depletion of major ions in the soil due to biochar, and that the fixation of Ca by biochar application to the soil weakened the competitive uptake of metals in the Cd-Zn hyperaccumulator (*Noccaea caerulescens*) root system [126]. This phenomenon can be explained by the fact that a decrease in major cations in the soil solution can cause a more negative potential at the plasma membrane of root cells in the hyperaccumulator, which may lead to an increase in the activity of other cations close to the membrane [126, 127].

Nutrient uptake by plants promotes pollutant absorption. Numerous studies have shown that HMs can enter plants through the channels of essential elements and are subsequently absorbed by plants; for example, Cd can enter plants through the transport channels of K, Ca, and Mg [45]. Owing to its complex composition, biochar can release different nutrients (C, N, Ca, and Mg) into the sediment or soil. It accelerates the uptake of HMs by stimulating the uptake of nutrients and water by plants and changing the composition of root secretions during the inter-root activity of plants. For example, the addition of bamboo biochar to soil increases both S and Cd/Zn uptake in Salix, showing a positive correlation between S content and both Cd and Zn [128]. Gong et al. also found that tea waste-derived biochar (TB) containing certain amounts of Fe, Ca, and Mg might stimulate plant uptake of nutrients while promoting Cd uptake and transport in ramie [45].

5.1.2 Facilitates the transfer of pollutants in the plant

A higher TF of the plant indicates that it has a better ability to transfer pollutants from the roots to the stems and leaves. Previous studies have shown that biochar can effectively enhance the TF in plants. For example, the introduction of biochar increased the transfer factors of Cu, Mn, and Zn in *Jacaranda mimosifolia D*. Don and increased the total accumulated mass of Mn in the aboveground parts of *J. mimosifolia* [129]. The quantity of pollutants carried by a plant varies with the concentration

of biochar. Gong et al. explored the effects of biochar application to sediments on the behavior of HMs, biotoxicity, and microbial activity. As the concentration of TB increased from 100 mg/kg to 1000 mg/kg, the concentration of Cd in ramie stems increased from 12% to 20%, respectively [45]. The addition of TB changed the distribution of Cd in ramie cells and increased the proportion of Cd in the organelles of ramie leaves, stems, and roots [45].

5.1.3 Promotion of plant growth

Plant biomass in phytoremediation is the main factor determining the level of pollutants accumulated by plants. Plants tend to fail to develop normally in soils or sediments with high pollutant concentrations in the early stages, when phytoremediation rates are often constrained by their growth. Biochar can improve plant productivity by increasing nutrient retention, improving CEC, regulating pH, adsorbing pollutants, etc. [130]. For example, the addition of biochar resulted in a 10-20% increase in the length of the aboveground parts of *Vallisneria natans*, and the maximum increase in the fresh weight of the aboveground parts was 34.36%. The amount of biochar was positively correlated with chlorophyll content, aboveground height, and aboveground fresh weight in macrophytes based on the change in correlation coefficients [131]. In addition, biochar can increase the rate of nutrient and water uptake by plants, leading to stronger root absorption of Cd, Zn, and Cu [128]. Under the treatment, the water content of ramie leaves and stems increased by 4.29–6.69% and 4.01–8.25%, respectively [45].

Biochar can effectively immobilize HMs and adsorb OPs to reduce their phytotoxic effects. Mackenzie J. Denyes et al. observed that the addition of 2.8 wt% biochar to contaminated soil with PCB levels of 136 and 3.1 μ g/g resulted in a 77% and 58% decrease in PCB concentration in the root system of *Cucurbita pepo ssp. pepo*, respectively, and that the decrease increased with increasing biochar addition [132]. Biochar reduces the toxicity of HMs to plant roots by reducing their mobility, thereby promoting plant root growth. This may also lead to an increase in the uptake sites of HMs on the surface of plant roots, resulting in an enhanced phytoremediation capacity [133]. Previous studies have demonstrated that biochar can scavenge excessive ROS (including H₂O₂, ·OH, O₂⁻⁻, etc.)

produced by plants to alleviate the oxidative damage induced by HMs in plants [134, 135]. The accumulation of ROS is related to the concentration and type of HMs in the soil or sediment [136]. When biochar applies to soil or sediment, on the one hand, it reduces the accumulation of HMs in plants and increases their mineral supply; on the other hand, its rich pore structure and functional groups accelerate the oxidation of \cdot OH, O2⁻⁻ and other free radicals.

5.1.4 Influencing the adsorption-desorption properties of biochar

The adsorption-desorption process of biochar in association with phytoremediation is affected by root secretions released by the plant. Root exudates can promote the surface adsorption process of biochar by increasing its polarity and SSA as well as inhibiting hydrophobic partitioning, causing a decrease in biochar adsorption [137]. Similarly, root exudates can facilitate the bioavailability of biochar-sorbed OPs, thereby promoting their degradation [138]. The desorption of pollutants from biochar occurs through the action of root exudates, which leads to easier microbial degradation of contaminants in the environment [139]. The carboxylic acid groups in root exudates are most likely to promote the desorption of contaminants from biochar by disrupting the organic matter-mineral complex [140]. Biochar and root exudates can stimulate the biomass and activity of microorganisms in the soil as well as the abundance of genera and genes associated with OPs degradation, resulting in the diffusion of contaminants [141]. Song et al. found that hexachlorobenzene (HCB) in soil can be effectively immobilized and degraded by ryegrass (Lolium perenne) and biochar. Oxalic acid, a major component of ryegrass root exudates, promotes the desorption of HCB from biochar and increases the abundance of some genera (e.g., Azohydromonas, Pseudomonas, Fluviicola, and Sporocytophaga), further demonstrating that the biochar-plant tandem approach is an effective strategy for soil remediation [138].

5.2 Challenges and perspectives in sediment remediation by combining biochar with phytoremediation

Fig.7 shows future perspectives and challenges in sediment remediation using biochar combined

with phytoremediation. Challenges and opportunities for biochar synergistic phytoremediation in sediments mainly stem from the lack of biochar-plant interactions and synergistic mechanisms in real sediment environments, especially in the absence of sufficiently mature engineering practices to clarify the synergistic effects of biochar and phytoremediation in complex sediment environments (e.g., dissolved oxygen, organic matter, and salinity). The interactions between biochar and sediment involve various physical, chemical, and biological processes that can directly affect phytoremediation. For example, water in the sediment entering biochar pores directly causes the dissolution of soluble organic and mineral compounds on its inner and outer surfaces, which may lead to changes in organic carbon, conductivity, pH, and Eh in the sediment and may also depend on the type of biochar. Subsequently, environmental changes in sediments can affect macrophyte remediation processes. For example, lower Eh results in changes in the bioavailability of sediment HMs, which can affect phytoremediation. In summary, the impact on phytoremediation needs to be considered in the context of different environmental conditions and changes in sediment physicochemical properties caused by different types of biochar.

Synergistic biochar phytoremediation involves a complex mechanism of interactions between biochar, macrophytes, and microorganisms. Microscopic mechanistic studies and synergistic macroscopic experiments should be combined to clarify the interactions among biochar, macrophytes, and microorganisms. By exploring the correlation between the molecular structural changes in biochar, plant physiological indicators, root exudates, enzyme activities, microbial community structure, and the morphology, toxicity, and degradation intermediates of pollutants, which can fill the gaps in the mechanism of sediment pollutant removal, the mechanism of pollutant action in different degradation pathways can be clarified. In addition, varying methods of biochar addition can lead to differences in remediation efficiency. Biochar takes the form of a complete mix with the sediment, or is planted into the sediment in combination with plant roots, depending on the mechanism of interaction between the biochar and the plant. Therefore, it is necessary to clarify the interaction mechanisms for sediment remediation.



Figure 7. Future perspectives and challenges in sediment remediation by biochar combined with phytoremediation. With the development of industrial, technological, and medical industries and the frequent occurrence of disasters, several emerging environmental contaminants, including microplastics, rare earth elements, radioactive elements, and liquid crystal monomers (LCMs), have appeared in sediments, complicating their remediation. The changed sediment environment caused by coremediation may have many effects on the removal of emerging contaminants, such as more difficult removal from the sediment, changes in morphology, and increased toxicity. Therefore, it is important to understand the biochemical mechanisms of biochar combined with phytoremediation for emerging contaminants and assess their applicability to different types of emerging contaminants.

Currently, the remediation mechanism and resulting environmental behavior of biochar combined with phytoremediation cannot be accurately understood by relying only on short-term experimental results, thus failing to elucidate their application in real sediment environments. Therefore, long-term exploration is required to fully evaluate the effects of combined biochar and phytoremediation on contaminants, resistance genes, and microorganisms in sediments. A life cycle assessment is necessary

for the entire remediation system. It is not sufficient to consider only the secondary effects in the process of sediment remediation, but also the impacts of sediment reuse after remediation projects. Additionally, plant disposal poses several problems during long-term remediation. Macrophytes may die and release pollutants back into water if they are not harvested in a timely manner. Harvested macrophytes can also cause the re-release of contaminants owing to unsafe disposal methods.

6 Conclusions

This review summarizes the progress and limitations of phytoremediation and biochar remediation for sediment remediation, as well as the feasibility and challenges of the co-remediation of biochar with macrophytes.

Phytoremediation is an ecofriendly and sustainable environmental remediation technology. However, there are still some limitations to their application; for example, phytoremediation can only be used in environments with low pollutant concentrations and commercial value owing to its long remediation time. Phytoremediation efficiency can be improved through genetic modifications; however, its effectiveness is limited. Therefore, further studies on phytoremediation are required.

In recent years, biochar has achieved good results in various environmental remediation applications with the advantages of low cost, strong adsorption capacity, and green environmental protection. However, the impact of complex sediment environments on the adsorption performance and stability of biochar needs to be further explored because the mixing process of biochar and sediment greatly affects the remediation effect of biochar and easily causes the secondary release of pollutants from the sediment into the overlying water, which cannot be easily removed from the sediment, leading to secondary pollution. Therefore, enhancing the ability of biochar to absorb and transform pollutants and developing multiple techniques for combined remediation to enhance its service life are the focus of current research.

In the future, composite technologies will become a trend in environmental remediation. Biochar and macrophytes play synergistic roles in environmental remediation. Biochar can effectively improve

the sediment environment, promote macrophyte growth and development, and enhance pollutant enrichment and transfer. Macrophytes can remove enriched pollutants from sediment by harvesting, thus reducing sediment pollutant concentrations. Macrophytes indirectly extend the life of the biochar by creating habitats that promote microbial development and pollutant degradation. Therefore, it is important to investigate the interaction mechanisms between biochar and macrophytes and the effects of complex sediment environments on their joint restoration. Finally, considering the increasingly complex aquatic environment, key fields for future research are the elimination of composite and emerging pollutants through complex remediation measures and studies on intermediate transport and transformation processes. In summary, this study demonstrated the feasibility of combining macrophytes and biochar, revealing their challenges and perspectives. It also serves as a reference for improving the efficacy of in-situ sediment remediation and enhancing the efficiency of composite remediation measures.

Declaration of competing interest

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

Acknowledgements

This study was financially supported by the National Natural Science Foundation of China (52170168) and the Fundamental Research Funds for the Central Universities (22120220367).

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships

that could have appeared to influence the work reported in this paper.

Environmental Implication

Single remediation techniques cannot achieve stable and efficient sediment remediation. This review describes the feasibility of biochar combined with phytoremediation applied to sediment remediation from the perspective of the composite remediation method. Biochar and macrophytes show significant synergistic effects. Biochar can improve the efficiency of phytoremediation, while macrophytes can reduce the risk of secondary pollution caused by biochar. This paper provides references and insights for realizing sediment green remediation for the natural, efficient, and practicable development of environmental remediation technologies.

Highlights:

- Low TF and long remediation time are limiting factors for phytoremediation. •
- Complex sediment environments limit the efficiency of biochar remediation.
- Biochar and phytoremediation can exhibit significant synergistic effects.
- Challenges and perspectives of phytoremediation combined with biochar. •



Graphical abstract