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Review

The Role of Modified Biochar for the Remediation of Coal Mining-Impacted Contaminated Soil: A Review

Subhash Chandra [1,*], Isha Medha [2,3] and Ashwani Kumar Tiwari [4,*]

[1] Department of Civil Engineering, GITAM School of Technology, Gandhi Institute of Technology and Management University, Visakhapatnam 530045, India
[2] Department of Mining Engineering, Indian Institute of Technology Kharagpur, Kharagpur 721302, India
[3] Department of Civil Engineering, Vignan's Institute of Information Technology, Duvvada, Visakhapatnam 530049, India
[4] School of Environmental Sciences, Jawaharlal Nehru University, New Delhi 110067, India
* Correspondence: schandra3@gitam.edu (S.C.); ashwaniktiwari@mail.jnu.ac.in (A.K.T.)

Abstract: Land degradation and the release of contaminants such as heavy metals into the environment due to mining activities is a concerning issue worldwide. The bioaccumulation of heavy metals in the environmental matrix can severely damage flora and fauna and negatively impact human health. The poor physicochemical properties of mine spoil generated through mining operations make restoration of such contaminated and degraded lands challenging. In recent years, an exponential growth in the development and applications of biochar and its composites for the remediation of heavy metal-polluted environmental matrices such as soil and water has been observed. The literature review found that 95 review papers were published in the last five years reviewing the utility of biochar for heavy metals removal from the aqueous environment. However, no paper was published focusing on the application of biochar and its composites for the remediation of heavy metal-contaminated coal mine soil. The objective of the present review is to critically review the impact of mining activities on the environment and the role of biochar and its composites in the remediation of heavy metal-contaminated mine soil. This review presented a detailed discussion and sufficient data on the impact of mining practices in India on the environment. In addition, it critically discussed the methods of the production of biochar from various wastes and methods of modifying the pristine biochar to develop functionalized biochar composites. The detailed mechanism through which biochar and its composites remove and immobilize the heavy metals in the soil was discussed. The efficacy of biochar for the remediation of contaminated mine soil was also critically evaluated using various case studies and data from previously published articles. Thus, the major conclusion drawn from the review is that the application of various functionalized biochar composites could effectively manage and remediate heavy metal-contaminated mine soil.

Keywords: biochar; heavy metals; mine soil; pollution; remediation

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1. Introduction

The release of pollutants, particularly heavy metals, into the environment has become ubiquitous due to industrialization and urbanization. In the last decade, rapid growth in the worldwide population has created a massive demand for supply chain systems and energy requirements. To cater to the increased demands, particularly the energy requirement, which has almost doubled, most developing countries rely on coal-based thermal power plants to meet their energy demand [1]. Globally, coal production is around 7.90 billion tonnes (BTs) [2], of which China is the leading producer, followed by India and the USA. Recent energy data estimated that coal produces approximately 40% of the world's electricity. It has also been forecasted to be the most demanded fuel to fulfill energy needs over the next three decades [2]. India is the world's seventh-largest economy and the second most populated country [3]. The continuous increase in the population and

economy has increased India's energy demand per capita. In India, coal is the primary energy source to meet the continuously increasing energy demands. India has a total of 306.60 BTs of coal reserves [2]. During the financial year 2020–2021, India produced around 756.50 million tonnes of coal, which is forecasted to grow by one billion tonnes by 2022 [3]. During the last decade, coal production in India has increased by 32% to meet growing demands [4].

The production of coal and other minerals to fulfill the supply chain can only be done through mineral mining, such as coal mining, bauxite mining, and iron ore mining. Mining activities are often associated with changes in the landscape, deforestation, migration of humans and animals, loss of soil, acid mine drainage, and release of heavy metals into the environmental matrix [5]. India has 319.02 billion tonnes of coal reserves. It was reported that India produced around 778.19 million tonnes of coal in the year 2021–2022, which was 8.67% higher than last year's production figures ([6]). Coal production through mining involves the removal of the green cover and scavenging massive amounts of rock materials. The rock materials are dumped over a specified location of the active mining area [7]. The mine spoils in the form of dumps also pose a serious threat to the environment, as the mine spoil is loaded with heavy metals and mostly comprise a coarser fraction [8]. Coal mine spoils are rich in pyrite, often leading to acid mine drainage. Acid mine drainage is responsible for the leaching of heavy metals from the dump into the water resources due to the lowering of pH of the aqueous soil solution [9]. The release of heavy metals through the leaching process into the environment poses a serious threat to human beings, flora, and fauna. Therefore, it is essential to restore such contaminated sites with the most common approach of revegetation [10].

Various methods have been practiced and suggested for restoring such contaminated and degraded mine spoils lying over the overburden (OB) dumps, such as phytoremediation and application of mulch, lime, biosolids, and fly ash as soil conditioners ([11–13]). However, the application of the techniques mentioned above has certain limitations. For example, fly ash application in the soil is associated with the risk of heavy metals contamination [14]. Biosolids applications in the soil may cause contamination of soil with organic pollutants (pesticides and herbicides) and can increase the bioavailability of some heavy metals [15]. The lime and manure application is only suitable for acidic soils and can increase CO_2 emissions from the soil [16]. Other technologies are also available for soil remediation and pollutant removal, such as adsorption, biodegradation, and in situ chemical oxidation (ISCO), which can substantially retain and degrade the pollutants in the soil [17]. In this context, biochar has emerged as a wonderful material that can be used to improve the soil structure, immobilize heavy metals through adsorption and oxidation/reduction processes, and support vegetation growth [18–20]. Biochar is a porous carbon material derived from the thermochemical conversion of biomass waste under anoxic conditions in the temperature range of 400–700 °C for a duration varying between 1 and 3 h [21,22]. Biochar can be produced from a variety of raw materials, such as corn straw, rice straw, hardwood waste, food waste, and agricultural waste, by pyrolyzing them at the temperature mentioned above [23–26]. Recently, a ball-milling technique has been developed to produce very fine or nano-sized biochar particles for their application in the efficient removal of pollutants from the environmental matrix [23,27,28]. Biochar is known for its high porosity, functional groups, aromatic character, presence of inorganic elements, and high specific surface, making it a suitable material to be used as a soil ameliorant in contaminated soil [29,30]. For example, Turan [31] reported the application of olive pulp-derived biochar in combination with calcite in Ni-contaminated soil to reduce its mobility within the soil and in crop biomass based on a pot-culture study. In another study, Turan [32] reported the combined application of hazelnut husk-derived biochar and arbuscular mycorrhizal fungi that have substantially reduced the bioavailable and DTPA-extractable fraction Ni in the soil besides improving the soil enzymatic activity. In addition, the chemical characteristics of the biochar can be changed by doping its surface with other inorganic elements, acid or alkaline treatment, and the development of composites [33,34]. The modification methods

of biochar can be classified into five types: acid, alkali, steam, metals, and amination modifications [21]. The acid modification removes the impurities from pristine biochar and introduces acidic functional groups [21]. The alkali and steam modifications increase the surface area of pristine biochar and pore structure [21]. Metal impregnation is done to improve the adsorption capacity and catalytic performance of biochar [21]. Thus, the modified biochars are highly efficient in retaining nutrient loss and reducing the availability of heavy metals in the soil [35,36]. He et al. [37] reported the development of chitosan and alginite biochar from rice straw waste to be used as soil conditioner. Results indicated that the application of the chitosan and alginite-modified biochar substantially reduced the soil acidification and the availability of Cd^{2+} ions in the aqueous soil solution [37]. Li et al. [38] reported the development of MgO flake-modified rice husk biochar to immobilize heavy metals in the soil. Results of the study indicated that the MgO-modified biochar showed high adsorption capacity for the adsorption of Cd (II) (104.68 mg kg^{-1}), Cu (II) (173.22 mg kg^{-1}), Zn (II) (104.38 mg kg^{-1}), and Cr (VI) (47.07 mg kg^{-1}) in the aqueous solution and reduced the exchangeable fractions of these heavy metals by 9.60% in the soil [38]. Li et al. [39] reported the development of Mg/Al-modified biochar (MAB) through the one-pot pyrolysis method for immobilizing heavy metals and compared its efficiency with pristine biochar. Results indicated that compared with the pristine biochar, the application of the MAB in the soil increased the removal efficiency of As, Pb, and Cd from −62%, 17%, and 5% to 52%, 100%, and 66%, respectively [39]. Gholami and Rahimi [40] reported the synthesis of CH_4N_2S-modified potato peel biochar for application in the soil to remove heavy metals. Results of the study showed that the application of modified biochar at a rate of 8% (*w*/*w*) significantly reduced the soluble and exchangeable fractions of Cd, Zn, and Cu in the soil to 102.97, 94.40, and 76.18 mg kg^{-1}, respectively, compared with the application of pristine biochar [40]. In addition, the application of modified potato–biochar composite also improved the soil pH, cation exchange capacity, and organic carbon content in the soil [40]. Yang et al. [41] reported the development of pig carcass-derived phosphorous-rich biochar and plant-derived Fe-modified biochar and their application for immobilizing Cd and Pb in the soil. It was found that the application of P-rich biochar showed better immobilization of Pb^{2+} in the soil under redox conditions than Fe-modified biochar, whereas both P-rich and Fe-modified biochars showed lower immobilization of Cd^{2+} ions compared with pristine biochar under pre-maintained redox conditions (Eh ≤ -100 mV) [41]. Song et al. [42] reported the development and application of biochar-supported nano-zero valent iron (nZVI) composite in the multimetal-contaminated soil. Findings from the study demonstrated that the application of (nZVI)–biochar composite substantially immobilized the heavy metals (Cu, Zn, As, Cd, and Pb) and improved the pH, CEC, and enzymatic activities in the soil [42].

The motivation behind writing this review article is that during the last five years (2018–2022), a total of 481 research articles have been published, as shown in Figure 1 (sourced from Scopus, Google Scholar, and Web of Science). The articles were searched based on the keywords related to biochar production, biochar for soil amendment, and biochar for remediation of contaminated soil. Out of a total of 481 publications, only 95 review articles were published on the same topic. However, not a single article has been published highlighting the environmental impacts of mining practices in India and the role of modified biochar and biochar composites in the remediation of such heavy metal-contaminated mine soils. The main objective of the present review article is to critically evaluate the impact of the mining practices in India and abroad on the environment and how it can lead to the damage of sensitive ecosystems, apart from deteriorating the soil and water quality. In addition, this article also discussed the production methods of biochar and the physical and chemical methods to produce modified biochar and biochar composites. Finally, the role and efficiency of the modified biochar and biochar composites for the remediation of heavy metal-contaminated mine soil and the underlying mechanism were also evaluated. The detailed structure of the manuscript and the interrelation of various

topics discussed in this manuscript can be understood with the help of a diagram presented in Figure 2.

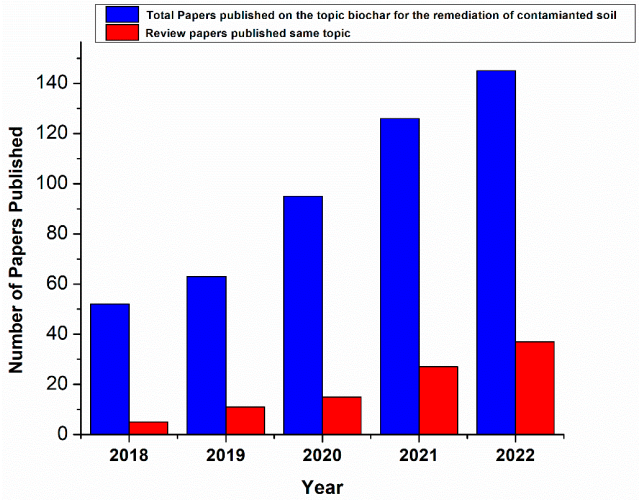


Figure 1. Total numbers of articles published in the peer-reviewed journals related to the keywords biochar, remediation, heavy metals, and contamination (articles were sourced from various research databases: Scopus, web of Science, and google scholar).

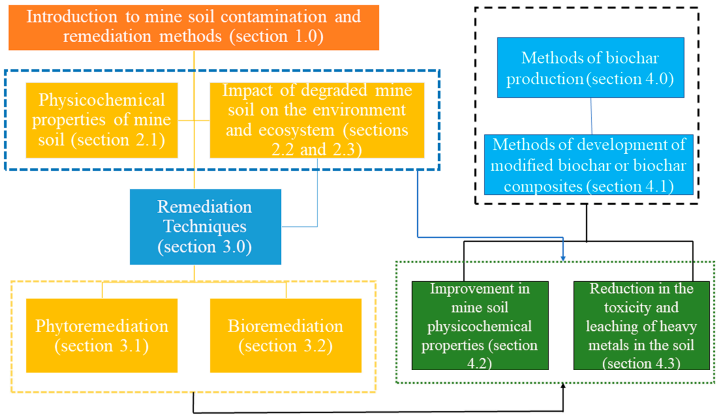


Figure 2. Overall structure chart of the review highlighting various topics covered and their interrelation.

2. Degraded and Contaminated Soils in Mining Areas

The mined-out degraded soils are artificial habitats developed through the deposition of overburdened materials during the mining process. These degraded soils experience a wide range of problems with establishing vegetation. The physicochemical properties of such soil inhibit vegetation growth due to the lack of nutrients and the presence of heavy metals [43]. The physicochemical properties, nutrient conditions, and contamination levels of coal mine-degraded soil are discussed in the subsequent sections.

2.1. Physicochemical Properties

Mining activities are associated with severe land and soil productivity damage. Mine soil is developed anthropogenically due to changes in the landscape after post-mining. The soil in the mined-out land is typically acidic, low in organic matter and nutrients, and has a high content of heavy metals [44]. The mine soil reportedly has poor structure, high bulk density, and a coarser texture. The high bulk density and poor structure of mine soil can be linked to heavy machinery during mining operations and the intrusion of rock fragments within the soil during mining that results in soil aggregation and low water-holding capacity [45]. The typical characteristics of mine soil are given in Table 1.

Table 1. Typical physicochemical characteristics of mine soil.

S. No.	Study Area	Texture Class	Bulk Density (Mg/m^3)	Moisture Content %	pH	Organic Matter (%)	Available N (mg kg^{-1})	Available P (mg kg^{-1})	Exchangeble K (mg kg^{-1})	CEC (cmol/100 g Soil)	References
1	Viswakarma opencast mines Jharia coal field	Sandy loam	1.16	6.56	6.53	1.67	82.07	8.76	26.98	14.88	[46]
2	Daliuta Mining area, Shanxi Province, Northwest China	Sandy loam	1.60	9.07	5.67	3.06	-	-	-	-	[47]
3	Mining city, Anhui Province, China	-	1.37	-	7.92	2.15	0.72	18.96	53.82	-	[48]
4	Pingshuo mine, China	-	1.62	-	8.70	1.78	20	6.25	52	-	[49]
5	Silesian Upland in Southern Poland	-	-		5.70	-	-	14.90	212.10	-	[50]
6	East-central Ningxia Province, Northwest China	-	-	7.58	8.04	3.20	3.24	5.80	158.70	-	[51]
7	Antaibao opencast coal mine, Pingshuo, Shanxi Province	Sandy loam	1.35	6.45	8.00	-	-	4.91	151.30	-	[52]
8	Cartagena-La Union mining district, SE Spain	-	-	-	7.40	1.64	-	-	-	-	[53]
9	Latiand Sambarata open coal mine sites, Berau Regency, East KalimantanProvince	-	1.65	-	4.50	0.87	-	2.40	103	-	[54]
10	Mahakam Sumber Jaya open-pit mines, East Kalimantan Province, Indonesia	Silty loam	1.83	19.09	3.27	1.42	-	5.02	296.40	14.69	[55]
11	Pingshuo Mine, China	Sandy loam	1.42	-	8.12	1.04	-	4.73	82.63	-	[56]
12	Bastacolla, Jharia Coalfield, Dhanbad, Jharkhand, India	Sandy loam	-	13.18	5.86	1.03	194.05	0.70	95.88	6.16	[57]

2.2. Impact on Soil Ecosystem

Ecology has two components, viz., organisms and environment, and combinedly, they are called the ecosystem. The ecosystem has two components: abiotic (includes all living organisms) and abiotic component (includes all non-living parts of the ecosystem) [12]. Ecosystem stability is another crucial component within the soil. A stable ecosystem maintains the species richness and nutrient pool dynamics within the soil structure. An ecosystem is considered stable if it maintains the species richness, structure, and functioning from year to year [12]. Mine soil is often considered to have poor ecosystem stability due to the loss of vegetation, nutrients, and frequent mining operations. Topsoil removal during mining operations disturbs the microbial activities and prevailing vegetation, which makes it hard to re-establish the vegetation cover [58]. The impacts of mining activities on the ecosystem can be visualized in Figure 3. The microorganisms and their microhabitats play an essential role in maintaining vegetation growth over degraded mine soil through nutrient recycling, humus formation, and mineralization [12]. Assessing microbial biomass gives information about the growth of the overall microbial community in the soil [45]. The mine soil respiration, cellulose decomposition, and soil enzymes are the most commonly measured parameters to estimate the growth of the microbial community in the soil. The soil enzymes are another critical parameter to evaluate the quality of the soil ecosystem within the soil matrix. The intercellular and extracellular microorganisms secrete the soil enzymes and perform essential biochemical functions to promote organic matter formation and nutrient recycling within the soil [45]. Mined-out land lacks active soil enzymatic activities within the soil matrix and, thus, has a meager nutrient recycling rate and poor soil ecosystem [59]. Previous studies have demonstrated that enzymatic activities in the soil could be used as indices for monitoring the degraded mine soil reclamation success [45,59].

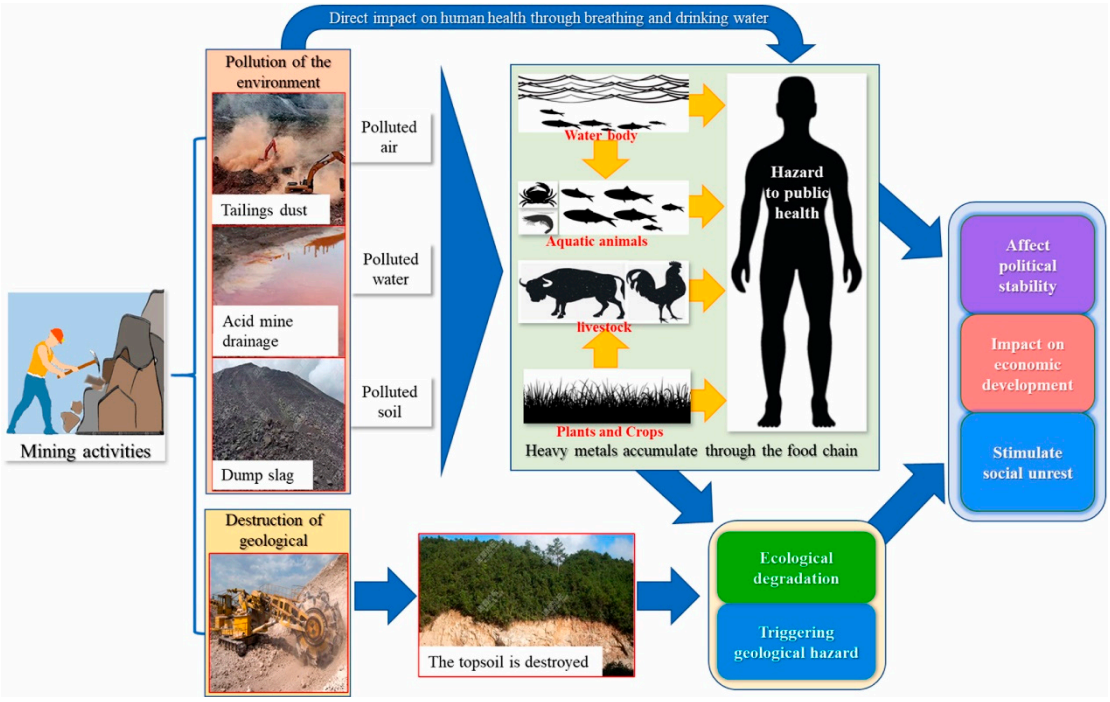


Figure 3. Impact of mining activities on the ecosystem and environment (reproduced with permission from [60] License no. 5406921447438).

2.3. Heavy Metals Pollution and Health Impacts

Metals such as lead (Pb), zinc (Zn), chromium (Cr), cadmium (Cd), mercury (Hg), copper (Cu), and nickel (Ni) having densities greater than 5 g/cm^3 are categorized as heavy metals. The presence of heavy metals in the soil in quantities greater than the permissible limit leads to soil pollution. The heavy metals in the soil may intrude into the

food chain and can profoundly impact the environment and human health. The toxicity and the chances of intrusion of heavy metals from the soil into the food chain can be measured using the geoaccumulation index (I_{geo}), which is used to evaluate the level of heavy metal contamination and their mobility in the soil [61]. The geoaccumulation index can be calculated using the following formula reported by [62].

$$I_{geo} = \log_2 (C_n/1.5B_n) \quad (1)$$

C_n is the heavy metal concentration measured in the mine soil and B_n is the background concentration of the respective heavy metal in the mining area. The calculated value of I_{geo} index can be evaluated using Table 2.

Table 2. Standards for the interpretation of geoaccumulation index values [62,63].

Class	I_{geo} Values	Pollution Level in the Soil
0	<0	Practically uncontaminated
1	0–1	Uncontaminated to moderately contaminated
2	1–2	Moderately contaminated
3	2–3	Moderately to heavily contaminated
4	3–4	Heavily contaminated
5	4–5	Heavily to extremely contaminated
6	>5	Extremely contaminated

The presence of heavy metals in the environmental matrix is associated with several health issues and ecological disturbances [64,65]. The impact of heavy metals on the environment and human health can be understood with the help of Figure 4 and Table 3, respectively.

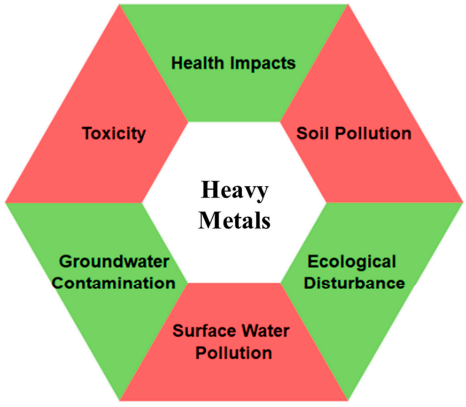


Figure 4. Impact of heavy metals on the environment and ecosystem.

Table 3. Effect of heavy metals on human health (Source: [66]).

S. No.	Heavy Metal	Target Organs	Health Impacts
1	Arsenic (As)	Pulmonary, Nervous System, Skin	Perforation of Nasal Septum, Respiratory Cancer, Peripheral Neuropathy, Dermatomes, Skin Cancer
2	Cadmium (Cd)	Renal, Skeletal, Pulmonary	Proteinuria, Glucosuria, Osteomalacia, Aminoaciduria, Emphysema
3	Chromium (Cr)	Pulmonary	Ulcer, Perforation of Nasal Septum, Respiratory Cancer
4	Manganese (Mn)	Nervous System	Central And Peripheral Neuropathies
5	Lead (Pb)	Nervous System, Hematopoietic System, Renal	Encephalopathy, Peripheral Neuropathy, Central Nervous Disorders, Anemia
6	Nickel (Ni)	Pulmonary, Skin	Cancer, Dermatitis
7	Tin (Sn)	Nervous System, Pulmonary	Central Nervous System Disorders, Visual Defects, EEG Changes, Pneumoconiosis

The soil in mining areas around the world is contaminated with heavy metals such as Cd, Cr, Cy, Ni, Zn, and Pb. The spatial distribution of the heavy metals varies from place to place and depends upon the soil's geology, rock formation, and physicochemical properties. The distribution of heavy metals in coal mining areas worldwide is presented in Table 4.

Table 4. Distributions of heavy metals in the soil in the mining areas around the world.

S. No.	Study Area	Heavy Metals (mg/kg)						Reference
		Cu	Pb	Zn	Cr	Ni	Cd	
1	Shizishan Mining Area	79	67	139	N.A.	33	0.71	[48]
2	All Opencast Coal Mining Pits, China	28.60	28.26	77.94	63.86	27.91	0.24	[66]
3	Lakhra Coalfield, Province of Sindh, Pakistan	n. a.	2.38	n. a.	n. a.	n. a.	3.46	[66]
4	Rohini Opencast Mining Area, North Karnpura, Jharkhand, India	n. a.	6.57	n. a.	15.29	n. a.	1.40	[67]
5	Raniganj Coal Mining, West Bengal, India	677	256.6	893.7	851.7	811.4	n. a.	[68]
6	Coal Mining Region, Tai'an City, Shandong Province, China	26.55	27.62	66.68	20.66	29.61	0.20	[69]
7	Jharia Coalfield, Jharkhand, India	11.36	11.43	19.90	23.40	11.38	0.80	[70]
8	Barapukaria Mining Area, Bangladesh	31.66	n. a.	101.96	82.37	56.54	n. a.	[71]
9	Wuhai Coal Mining Area, China	19.60	28	55.20	61.70	24.70	0.16	[72]

3. Remediation of Heavy Metal-Contaminated Soil

3.1. Phytoremediation

As per the World Health Organization (WHO) and the Environmental Protection Agency (EPA), heavy metals contamination in the soil and water resources is one of the most significant issues. There are various methods reported for the remediation of such contaminated sites. Among the reported methods, phytoremediation is an easy, eco-friendly, and sustainable approach to remediate heavy metal-contaminated sites [73]. The word phytoremediation was derived from the Greek word "Phyto", which means plants, and the Latin word "remedium", meaning to correct or remove [74]. Phytoremediation is an in situ remediation technique that utilizes particular types of plants called hyperaccumulators to extract heavy metals present in the soil and, thus, reduce their mobility [74]. The phytoremediation process can further be sub-divided into various processes, namely, phytoextraction, phytostabilization, phytodegradation, phytovolatilization, Rhizodegradation, and Rhizofiltration, which integratively contributes to the remediation process [74,75]. The details of the various types of phytoremediation processes and the mechanism of phytoremediation can be understood with the help of Figures 5 and 6, respectively.

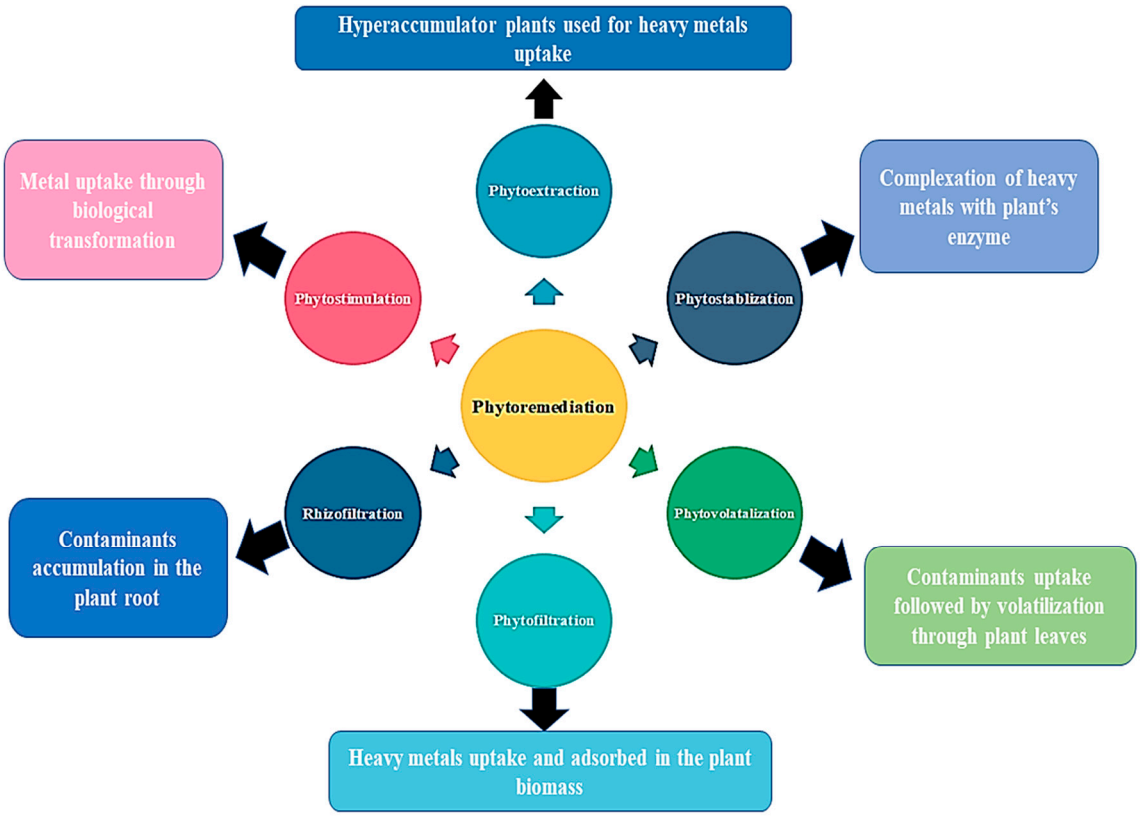


Figure 5. Processes of phytoremediation of heavy metals in the soil.

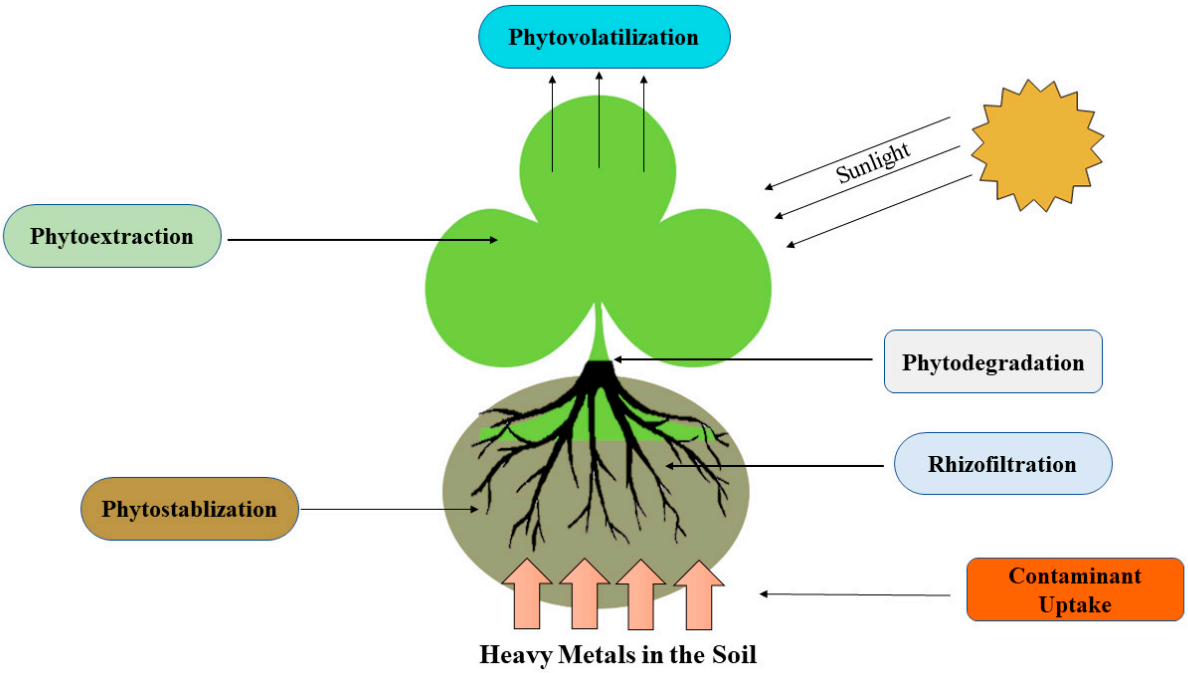


Figure 6. Mechanism of phytoremediation of heavy metals in the soil.

Various factors affect the effectiveness of the phytoremediation process. This includes the plant species, growth rate, selectivity of metals, plant immunity, and species' growing conditions [76]. In addition, geographical factors such as climatic conditions, temperature, and humidity also affect the phytoremediation potential of any plant species [77]. In addition, the plant species' immunity and root structure significantly affect the soil's

phytoremediation [77,78]. The details of different varieties of plant species used for the phytoremediation of heavy metal-contaminated soils are presented in Table 5.

Table 5. Plant species used for the phytoremediation of heavy metal-contaminated soil.

S. No.	Type of Contamination	Plant Species Used	Initial Concentration (mg kg^{-1})	BCF_{shoot}	Type of Study	Reference
1	Pb and Cr contaminated soil	*Parthenium hysterophorus* *Cannabis sativa*	Pb = 14.54 Cr = 4.48	Parthenium (Pb) = 1.01 (Cr) = 0.58 Cannabis (Pb) = 1.03 (Cr) = 0.54	Field study	[79]
2	Cu, Zn, and Cd contaminated soil	*Phyllostachys praecox*	Cu = 195 Zn = 2980 Cd = 14.5	Cu = 0.18 Zn = 0.33 Cd = 0.26	Field study	[80]
3	Cd contaminated soil	*Medicago sativa* *Bidens Pilosa*	Cd = 0.031	2.90	Pot-culture experiment (40 days)	[81]
4	Multi metals contaminated soil	*Rhazya stricta*	Cd = 50 Pb = 10 Cu = 10 Zn = 10	Cd = 1.48 Pb = 0.36 Cu = 0.52 Zn = 1.46	Pot-culture experiment (3 months)	[82]
5	Multi metals contaminated soil	*Switchgrass*	Zn = 68.1 Cr = 35.9 Pb = 16.9 Ni = 27	-	Pot-culture experiment	[83]
6	Multi metals contaminated soil	Wheat Crop	Cr = 70.2 Cu = 339 Zn = 202 Pb = 156 Ni = 113	Cr = 8.2 Cu = 2.2 Zn = 2.6 Pb = 2.8 Ni = 2	Pot-culture experiment (10 weeks)	[84]
7	Multi metals contaminated soil	*Robina pseudoacacia*	Cu = 3166.7 Cd = 3.66 Pb = 137.06	Cu = 0.044 Pb = 0.170 Cd = 1.358	Field study	[85]
8	Anshan Mining, Lioning Province, China	Soybean	Zn = 149.5 Pb = 42.1 Cd = 0.8	-	Field study (105 days)	[86]

3.2. Bioremediation

Bioremediation is a biological process that utilizes microbes (aerobic and anaerobic bacteria and fungi) to immobilize heavy metals in the soil. The main principle of the bioremediation process is to degrade and convert the pollutants to their less toxic form [87]. In the bioremediation process, microbes play an essential role. Two types of microbes participate in the remediation process: aerobic and anaerobic. The aerobic microbes consist of bacteria such as *Pseudomonas*, *Acinetobacter*, *Sphingomonas*, *Nocardia*, *Flavobacterium*, *Rhodococcus*, and *Mycobacterium*, which are capable of degrading and transforming the heavy metals in the soil [88]. The effectiveness of the bioremediation process is controlled by various factors such as the concentration and chemical nature of the pollutant, physiochemical characteristics of the environment (pH, temperature, oxygen, and nutrients), and the accessibility of the pollutant to the microbes [87]. The biotic factors that significantly affect the bioremediation process are the microbial cell enzymatic activities, mutation, gene transfer, microbial biomass production, and the microbes' population size [89]. The abiotic factors that affect the bioremediation process are soil temperature, pH, moisture, soil structure, water solubility, nutrient availability, redox potential, pollutants concentration, and toxicity level [89,90]. The bioremediation technique is frequently used for the remediation of heavy metal-contaminated soils. The mechanism through which microbes remediate the

heavy metal-contaminated soils is presented in Figure 7. Briefly, the heavy metals in the soil remain in their ionic form. Electrophilic heavy metals have the vacancy of electrons in their outer orbit. When such electrophilic heavy metals come into contact with microbes applied in the soil for bioremediation, the electron donor components present inside the microbes' cells, such as sugars or fatty acids, transfer the electrons to the electrophilic heavy metals. Thus, the heavy metals are reduced to non-toxic and stable forms by accepting the electrons. The details of the various types of microbes used for the remediation of heavy metal-contaminated soil are presented in Table 6.

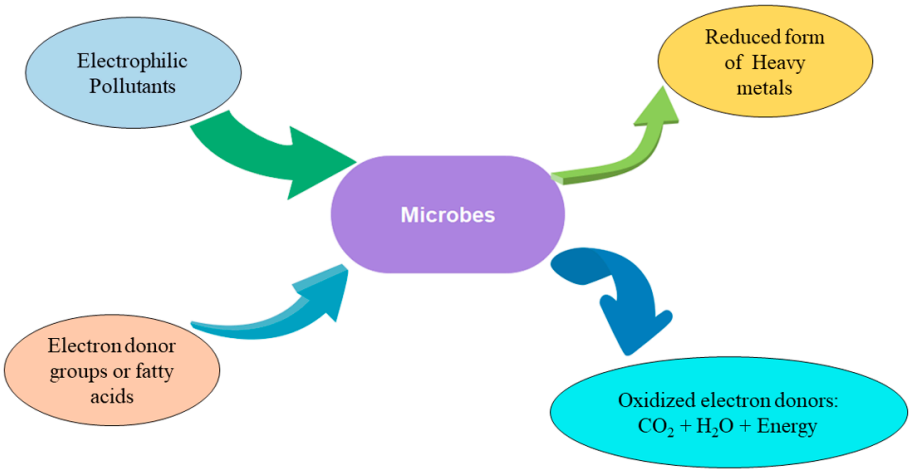


Figure 7. Mechanism of the bioremediation of heavy metals by the microbes present in the soil.

Table 6. Microbes used for the bioremediation of heavy metals in the soil.

S. No.	Type of Microbes Used	Site Description	Heavy Metals Remediated	Reference
1	*M. Circinelloides* *T. asperellum* *Mortierella* sp.	Metal-contaminated mine tailings, Anshan Mining Group, Lioning Province, China	Zn, Cu, Pb, and Cd	[86]
2	*Bacillus Idriensis strains* *B. subtilis BR151*	Neighborhood of Lead and Zinc smelters, Northern France	Cd	[91]
3	*Perenniporia subtephropora* *Cerrena aurantiopora* *Aspergillus niger MH541017* *Aspergillus fumigatus*	MSW Landfill, Jinjang Utara, Kuala Lumpur	Ni, Pb, and Zn	[92]
4	*Rhizobium* -legume	Heavy metal-contaminated soil	Cd, Cu, and Pb	[93]
5	*Agrocybe Aegerita*	Artificially contaminated soil	Ni and Cd	[94]
6	*Eisenia fetida* *Cylindrospermum stagnale*	Artificially contaminated soil	Cd	[95]
7	*Agrococcus* *Streptomyces* *Microbacterium*	Chromium-contaminated site in Henan Province	Cr (VI)	[96]
8	*Sporosarcina pasteurii*	Artificially contaminated soil	Pb	[97]
9	*Acaulospora mellea*	Vicinity of a Pb and Zn smelter located in Dongtang Town, Shaoguan City, Guangdong Province, China	Pb and Zn	[98]
10	*Phanerochaete chrysosporium*	Abandoned mining area in Hechi City, Guangxi Province, China	As, Cd, Cr, V, Sn, and Zn	[99]
11	*Bacillus thuringiensis* HM—311	Contaminated farmland soil in Jiangning District, Nanjing, Jiangsu Province	Pb and Cd	[100]
12	SRB 1 (*Clostridium, Desulfosporosinus, and Desulfovibrio genera*)	Huilong nonferrous metal smelter, Guangxi Province, China	As	[101]

4. Biochar and Its Composites for Soil Remediation

Biochar is a porous carbon material produced through the pyrolysis of various biomass wastes [102]. The biochar can be produced through various pyrolysis processes, such as slow pyrolysis, fast pyrolysis, gasification, and hydrothermal carbonization, as shown in Figure 8.

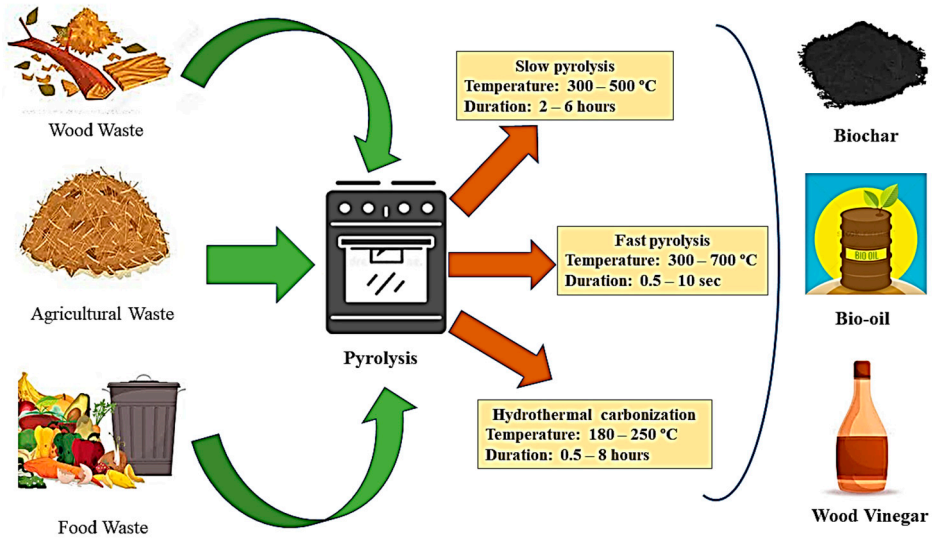


Figure 8. Various pyrolysis processes to produce biochar, bio-oil, and wood vinegar as the product through the pyrolysis of various biomass wastes.

Slow pyrolysis consists of prolonged biomass heating in the presence of insufficient oxygen over a temperature range of 300–800 °C [103,104]. Slow pyrolysis is primarily marked by a low heating rate (5–7 °C/min) and long solid and vapor residence time (usually longer than 1 h) [104]. The primary product of slow pyrolysis is biochar and a small quantity of bio-oil and syngas. Biochar yield through slow pyrolysis of biomass waste varies from 35–45%, and bio-oil and syngas gas yields vary from 25–35% and 20–30%, respectively [105–108]. Alternatively, the biochar can be produced by the fast pyrolysis process, which is characterized by high heating temperature, high heating rate, and short retention time. The biomass waste is typically heated at a temperature higher than 500 °C at a high heating rate ($\geq$300 °C min^{-1}) and heated at the final temperature for a few seconds (<2 s) [109]. The primary product of fast pyrolysis is reportedly the bio-oil having a yield of more than 60%, followed by syngas at 10–20%, and biochar having a yield of approximately 10% [108–110]. Another method is the gasification process, which is typically carried out at a very high temperature (>700 °C). During this process, heat transfer occurs from the bulk to the biomass particles leading to the evaporation of water followed by volatiles [108]. The primary product of the fast pyrolysis process is the syngas (>90%), followed by the generation of a small quantity of biochar (<5%) [111]. Hydrothermal carbonization (HTC) was developed to process wet biomass waste into carbon material called hydrochar. The advantage of HTC over other pyrolysis processes is that it can process wet biomass waste to hydrochar [112]. The HTC process is performed in the temperature range of 180–250° at an autogenic pressure (0.5–2 Mpa) for 8–10 h in a closed reactor or kiln [108,111]. The primary outcomes of the HTC process are the hydrochar having a yield of up to 80% and the production of dissolved carbon slurry having a yield of up to 20%. Unlike biochar, hydrochar is rich in nutrients and has a hydrophilic nature, as well as a low O/C ratio [108].

4.1. Various Synthesis Methods of Biochar Composites

Pristine biochar is a beneficial soil-ameliorating material owing to specific physico-chemical properties such as high porosity, functional groups, alkaline pH, and inorganic elements. However, the pristine biochar has certain limitations, such as hydrophobicity, low

specific surface area, and variations in acidic and basic functional groups that further limit its application areas. Various methods have recently been developed to modify biochar's physicochemical properties and develop metals or organics-doped biochar composites. The modifications in the biochar's physicochemical properties enable its application in various areas, such as for heavy metals remediation, water treatment, energy storage, and application as a slow-release fertilizer in the soil. The various methods for the modifications in the properties of biochar are graphically shown in Figure 9 and discussed in detail below.

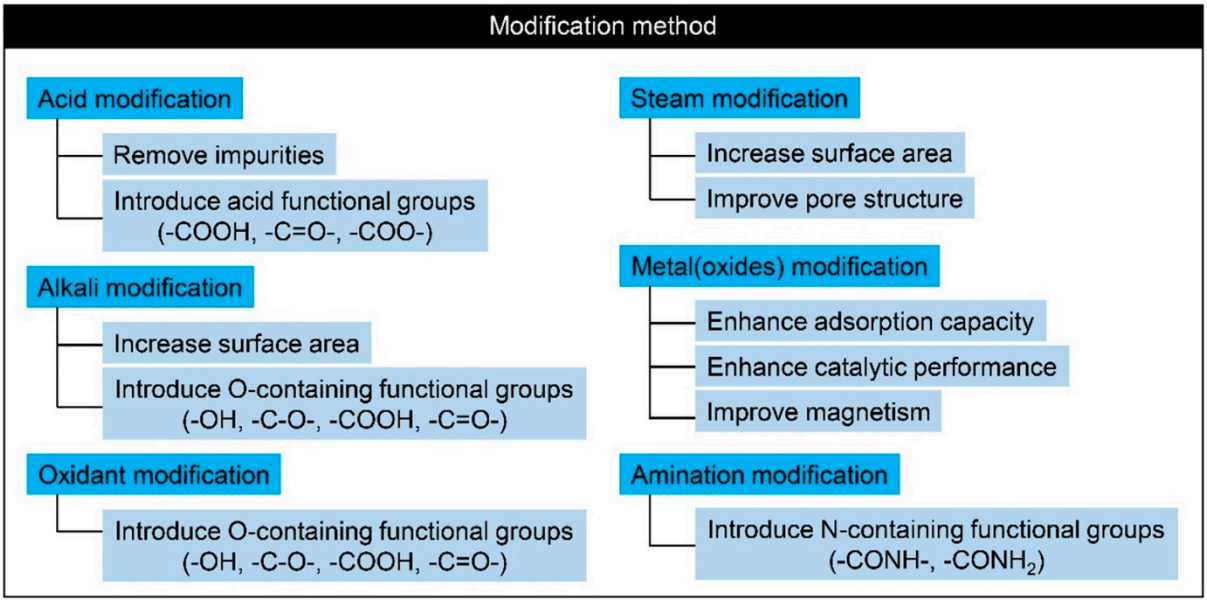


Figure 9. Various methods for the modifications in the properties of biochar (reproduced with permission from [34], License number: 5425161366000 obtained on 10 November 2022).

The changes in the properties of biochar can be done either by modifying the treatment methods (pre- and post-treatments) or by doping the biochar's structure with some foreign inorganic or organic elements and by surface modification methods. The surface modifications can be done by grafting the surface of biochar with carboxylic acid, amine, and amide functional groups to modify their surface chemical properties, such as an increase in hydrophilic character or hydrophobic micro and macro pores [113].

4.1.1. Modifications in the Treatment Methods

(a) Pre-treatment methods

The pre-treatment methods have been developed to impregnate the biomass waste with organic or inorganic elements through natural diffusion or bioconcentration processes by keeping the biomass waste in contact with the foreign element through solvent treatment. Various biomass wastes were reportedly pre-treated with $AlCl_3$, $ZnCl_2$, $MgCl_2$, and $KMnO_4$-based metal solvent for some time before pyrolyzing the biomass. Such pre-treatment of the biomass waste resulted in the impregnation of these nanoparticles within the biochar's structure and the formation of Al_2O_2, CaO, MgO, and ZnO nanocrystals [114,115]. Thus, the intrusion of foreign nanoparticles within the pristine biochar's structure significantly changes its physicochemical properties for its wide applications. Lyu et al. [116] reported the development of nano zerovalent iron–biochar composite (nZVI/BC) through various methods, such as hydrothermal carbonization, chemical precipitation, and ball milling, for the remediation of contaminated soil. Chen et al. [117] reported the development of an iron–biochar composite through the pre-treatment of barley grass with $FeCl_3$ followed by pyrolysis at 350 °C for the remediation of Cr-contaminated soil. Yu et al. [118] reported the development of MgO-modified biochar through the pre-treatment of corn straw biomass with $KMnO_4$ (0.079 mol/L) for 24 h, followed by pyrolysis at 600 °C for 0.5 h for its

application for the adsorptive removal of As from contaminated red soil. In addition, biomass waste can also be pre-treated with acid and alkaline solutions to change the functional groups and specific surface area. Acid pre-treatment of biomass waste before pyrolysis introduces acidic functional groups in the biochar along with a reduction in the mineral matter content.

In contrast, the alkaline pre-treatment increases the specific surface area of the biochar along with the enrichment of oxygen-containing functional groups [34]. Pap et al. [119] reported the synthesis of acid (50% H_2SO_4) fruit waste biomass-derived functionalized biochar to remove Pb and Cr heavy metals from the aqueous solution. Chen et al. [120] reported the application of alkali pre-treated biochar to remove Thorium (IV) from the aqueous solution, which was developed through the pre-treatment of duckweed biomass with 0.1 M NaOH followed by pyrolysis. Thus, it can be deduced that the modified or pre-treatment process can modify the properties of biochar as required to improve the removal of contaminants from the environmental matrix.

(b) Post-treatment methods

Post-treatment methods are different from pre-treatment methods, which leads to modifications in the biochar's physicochemical properties that also differs from the biochar produced through pre-treatment methods. Post-treatment refers to the process of modification of biochar properties after the pyrolysis of biomass waste. Post-treatment can be done using impregnation, evaporation, heating, and co-precipitation [34]. Dong et al. [121] reported the synthesis of magnetic biochar through the co-precipitation of biochar particles with $FeCl_3$ and $FeCl_2$ in an alkaline solution and its application for the remediation of contaminated soil. Imran-Shaukat et al. [122] reported the post-treatment of palm shell-derived biochar with 20% NaOH followed by acid treatment (HCl) to remove heavy metals from an aqueous solution. Samsuri et al. [123] reported the application of Fe (III)-coated biochar for the removal of As (III) and As (V) from the aqueous solution, which was developed by impregnation of biochar particles with Fe ions in a solution containing $FeCl_3$ salt for 24 h. Lyu et al. [124] reported the post-treatment of biochar with 0.063 moles of $FeSO_4$ aqueous solution to produce FeS-coated biochar particles to remove hexavalent chromium from the aqueous solution.

4.1.2. Co-Doped Biochar Composites

Co-doped mineral biochar is developed using chemical and mechanical methods such as co-precipitation, ball milling, and chemical doping, as shown in Figure 10. Co-doped or mineral biochar has a certain advantage over pristine or pre-treated biochar because the presence of mineral or co-doped elements significantly affects the performance of biochar through the change in its physicochemical properties. Co-doped biochars are applied in soil remediation, soil fertility improvement, heavy metals removal, and wastewater treatment [33]. For example, montmorillonite, a natural clay material, co-doped biochar significantly improved the metals and cationic retention within its interlayers through electrostatic attraction and cation exchange processes [125,126]. Li et al. [127] reported the development of Mg/Al-layered double hydroxide biochar by pre-treating the biomass waste with 75 mmol L^{-1} of $AlCl_3$ and by post-treating with 0.12 mol L^{-1} CH_4N_2O and 40 mmol L^{-1} $MgSO_4{\cdot}7H_2O$ followed by hydrothermal carbonization at 160 °C for 8 h. The application of Mg/Al-layered double hydroxide composite significantly increased Cd (II) and Cu (II) adsorptive removal from the aqueous solution. Wang et al. [19] reported the development of a clay–biochar composite derived through the co-pyrolysis of various clay minerals (Kaolin, Vermiculite, and Attapulgite) with rice husk biochar at 500 °C for 90 min. The study results showed that the application of mineral–biochar composite significantly improved the physicochemical properties of the contaminated soil and reduced the phytoavailability of the heavy metals in the soil [19]. Zhang et al. [128] reported the development of the EDTA and Chitosan bi-functionalized bamboo waste-derived magnetic biochar composite, which was produced through co-precipitation and chemical modification methods to remove Cd and Zn from the aqueous solution. Ma

et al. [129] reported the synthesis of microorganism-coated biochar composite known as *Bacillus* TZ5-coated biochar composite to immobilize Cd in the contaminated soil. Tu et al. [130] reported the development and application of a maize straw-derived NT-2-loaded biochar composite for Cd and Cu immobilization in the soil. Yin et al. [131] reported the application of rice straw-derived Fe–biochar composite synthesized through the co-precipitation method followed by the pyrolysis of biomass waste for the immobilization of Cd and As in the soil. Yu et al. [119] reported the development and application of corn straw-derived MnO-doped biochar composite to immobilize As in the soil and reduce their phytoavailability to Indian rice (*Oryza sativa* L.) cultivated in such contaminated soil. Apart from that, the biochar can also be simultaneously co-doped with metals and non-metallic elements, such as sulfur and iron co-doped functionalized biochar developed for the efficient stabilization of Cd and Pb in the contaminated soil [132].

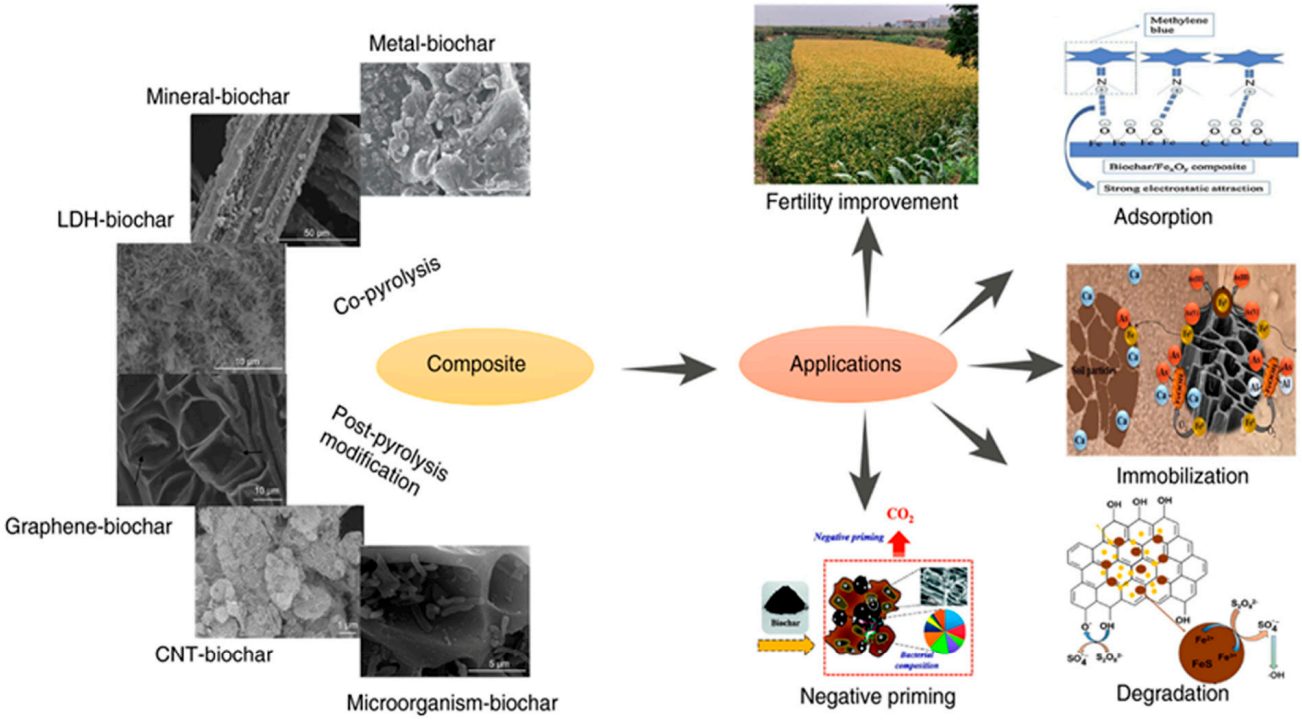


Figure 10. Co-doping of biochar with various metals and minerals to produce biochar composites for their applications in various areas (reproduced from [33] under open access Creative Common Attributions Non-commercial License).

4.2. Effect of Biochar and Its Composites on Soil Physicochemical Properties

The application of biochar into the soil is acknowledged to have benefits through the improvements in the soil's physical, chemical, and biological properties, as shown in Figure 11 [133]. Biochar is characterized as having a porous structure, functional groups, nutrients, and other inorganic elements, as shown in Figure 12. The porous structure of biochar helps to increase soil porosity and reduce its bulk density. Blanco-Canqui [134] reported that the application of biochar increased the soil porosity by 14 to 64% while reducing the soil bulk density by 3 to 31%. Further, the presence of functional groups and inorganic elements in the biochar structure helps it to retain nutrients within the soil and promote overall fertility [57]. However, pristine biochar has certain limitations regarding functionalized functional groups, inorganic elements, and high specific surface area, which are the key factors in retaining soil moisture and nutrients and promoting overall soil growth. In this context, the development of functionalized biochar and biochar composites has opened up a way to cater to such issues. Functionalized biochars are characterized by having a high specific surface area and the presence of functionalized metallic groups on their surface [135–138]. Lyu et al. [117] reported that applying iron–biochar composite

in the soil improved soil aggregation. This is because the iron oxide-coated surface of the biochar promoted the co-precipitation of dissolved organic carbon in the aqueous soil solution, and the organometallic interaction enhanced the cementing properties within the soil structure. Zhang et al. [139] reported that the application of Mg/Fe-layered double hydroxide–biochar composite in the soil enhanced the retention of NH^+ and NO_3^- ions through hydrogen bonding and anion exchange processes, respectively. Chen et al. [140] reported that the montmorillonite–biochar composite increased the retention of NH_4^+ and PO_4^{2-} ions through cation exchange and electrostatic attraction processes, which further regulated the controlled release of the nutrients in the soil. Chandra et al. [36] reported that the application of potassium–iron–biochar composite in the soil reduced the leaching of NH_4^+ and PO_4^{2-} ions and increased the plant availability of the nutrients in the soil. In another study, Shen et al. [141] reported that the application of rice husk-derived $MgCO_3$–biochar composite in the soil significantly increased the soil's water retention capacity and the retention of PO_4^{2-} ions through chemisorption. Thus, biochar composites have advantages over pristine biochar; however, both biochar and its composites can be used as an effective soil ameliorant.

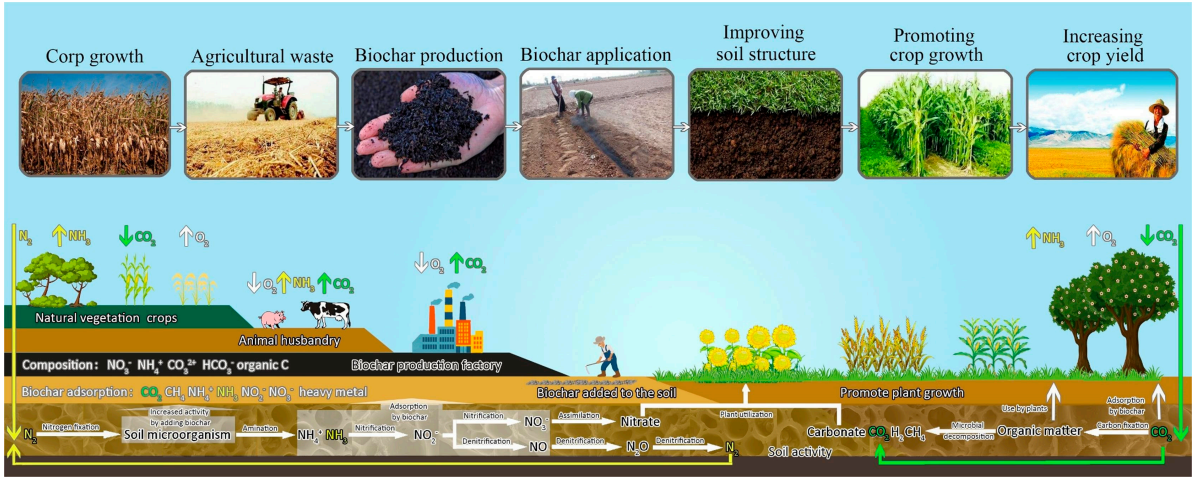


Figure 11. Effect of biochar application on soil physicochemical and biological properties, as well as on plant growth (reproduced with permission from [142] under License number: 5430160064883).

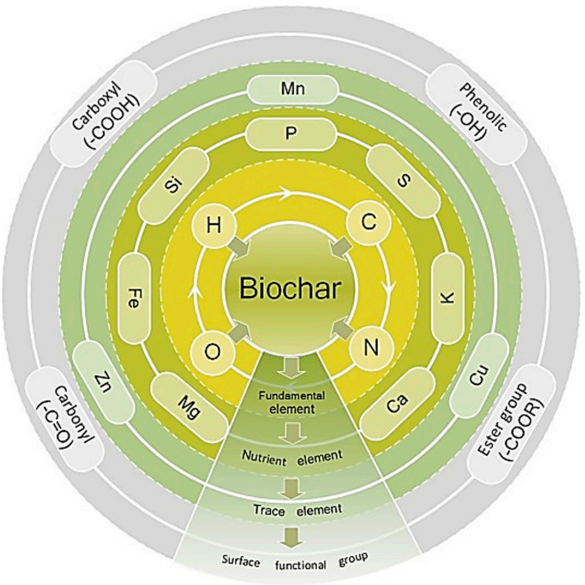


Figure 12. Chemical structure of biochar (reproduced with permission from Zhang et al. [142] under License number: 5430160064883).

4.3. Effect on Heavy Metals

The presence of heavy metals in soil and water resources is known to have a deleterious effect on the environment and human health. The various types of heavy metals found in contaminated mine soils are Pb, Cr, As, Zn, Cd, Cu, Hg, and Ni [57,143]. In the contaminated soil, the heavy metals are highly mobilized due to acidic pH and lack of organic matter and, thus, may cause the leaching of heavy metals into the water resources [144]. Various methods of immobilizing heavy metals in such contaminated soil include physical remediation, vitrify technology, chemical leaching, chemical fixation, electrokinetic methods, and phytoremediation [145]. Among the above-reported methods, the phytoremediation technique was well appreciated due to its high efficiency and low application cost [146]. However, with the phytoremediation technique, some limitations have been observed, such as the accumulation of heavy metals in the above-ground biomass, low growth rate of hyperaccumulators in the degraded and contaminated soil, and a prolonged process to achieve complete success [147]. The amendment of such contaminated soil with biochar as a soil ameliorant supports vegetation growth and assists in immobilizing heavy metals within the aqueous soil solution [148]. The various mechanisms through which biochar immobilizes or adsorbs the heavy metals in the aqueous soil solution are the electrostatic attraction, ion exchange, surface complexation, and precipitation processes [148,149]. The detailed mechanism of removing heavy metals in the aqueous solution using biochar can be understood with the help of the diagram presented in Figure 13. Although pristine biochar is a promising adsorbent for the removal of heavy metals, it also has certain limitations that restrict its application to certain types of heavy metals, such as Cr (VI), As, V, etc. This is due to having a negative charge on the biochar's surface [150,151]. In recent years, efforts have been made to develop various types of biochar composites to address the problems associated with pristine biochar. The modified biochar has the advantages of having functionalized organic groups, high specific surface area, improved pore structure, an abundance of active sites, and co-doped inorganic elements [152,153]. Liu et al. [154] reported the development of a magnetic macroporous biochar for the remediation and removal of Cd (II) and As (V) from the contaminated soil. Zhang et al. [155] reported the development of a corncob-derived multifunctional carboxymethyl cellulose sodium-encapsulated phosphorous-enriched biochar composite for the removal of Pb (II), Cd (II), and Ni (II) from the aqueous solution. Sajjadi et al. [156] reported the synthesis of a double-layered magnetized and silica-functionalized biochar composite. They observed that the application of biochar composite significantly improved the removal of Ni (II) and Pb (II) by 139 and 38%, respectively, compared with the pristine biochar. Chen et al. [118] reported the development and application of iron–biochar composite for the remediation of Cr-contaminated soil. The results indicated that the amendment of Cr-contaminated soil with Fe–biochar composite immobilized the Cr ions within the soil and substantially reduced their leaching into the groundwater. In addition, the recently developed biochar-supported microbial composite is an exciting development in biochar-based composites due to having a high potential for the remediation of heavy metals polluted soils. In this regard, Qu et al. [157] reported the development of bone char-supported biochemical composite carrying phosphate solubilizing bacteria, iron sulfide, and carboxymethyl cellulose on its surface for the remediation of Pb-contaminated soil. The major chemistry for the removal of heavy metals using biochar composites are surface complexation with the acidic or basic functional groups, co-precipitation with inorganic doped elements, oxidation or reduction by accepting electrons and protons, electrostatic attraction between oppositely charged heavy metals and biochar's surface, and π–π electron interaction between heavy metal ions and π-electrons [158]. The detailed mechanism of the chemistry behind the higher removal efficiency of heavy metals by biochar composites can be understood by referring to Figure 14. Further, the biochar was reported to have shown excellent heavy metals removal efficiency and remediation processes when combined with the phytoremediation technique [57,159]. Gong et al. [160] reported that the application of biochar varying from 100 to 5000 mg kg^{-1} in the soil improved the translocation of Cd from soil to biomass and, thus,

increased the efficiency of the phytoextraction capacity of the hyperaccumulators. Rathika et al. [161] studies the synergistic effect of biochar with EDTA (chelating agent) in the heavy metal-contaminated soil. It was found that the co-application of biochar with EDTA in combination with the growth a *Brassica juncea* in the Pb-contaminated soil significantly improved the phytoextraction of Pb from the soil. It was reported that the application of biochar enhanced the plant growth by providing nutrient support and reducing the leaching of heavy metals from the soil, which, in turn, increased the phytoextraction of heavy metals by the hyperaccumulators [162]. The details of the various types of biochar reported to assist in the phytoremediation of heavy metals from the soil is given in Table 7.

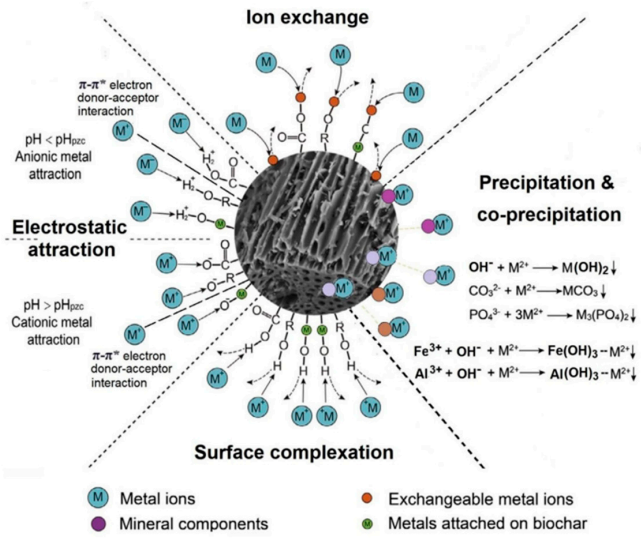


Figure 13. Mechanism of removal of heavy metals from soil using biochar (reproduced with permission from [148] under Creative Commons Attribution License).

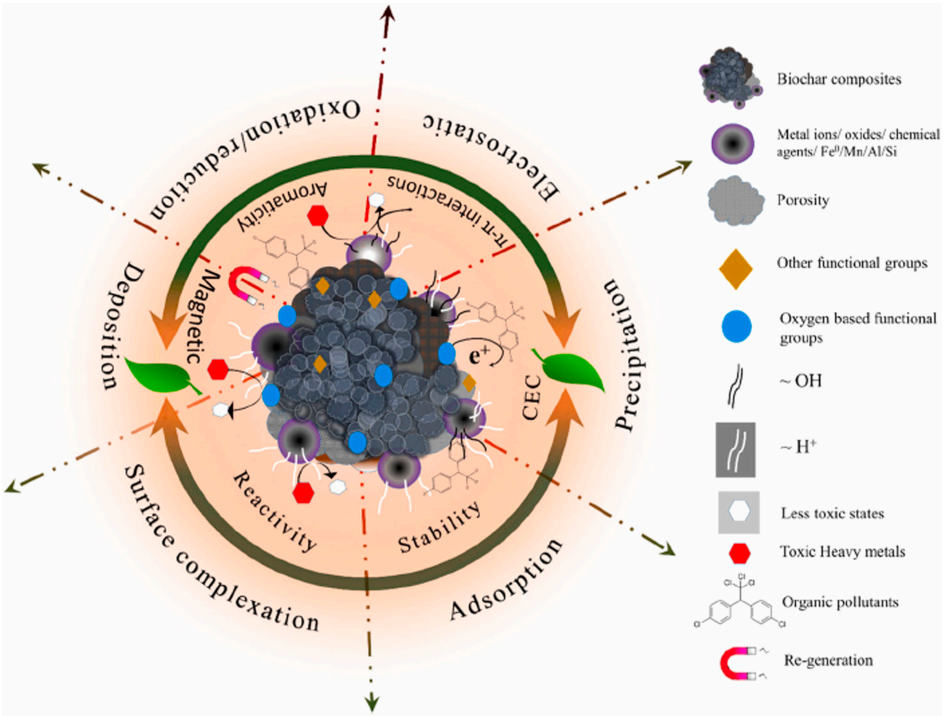


Figure 14. Mechanism of heavy metals adsorption or removal using various functionalized biochar composites (reproduced with permission from [158] under license number: 5431750879544).

Table 7. Biochar used as a soil amendment in combination with plant species for the phytoremediation of mine soil.

S. No.	Feedstock Used	Pyrolysis Conditions	Mine Soil Type	Heavy Metals Present	Plant Species Used	Inferences	References
1	Orchard pruning residues	Slow pyrolysis at 500 °C	La Mina Monaca Mine site, Spain	Cd, Cu, Pb, As, and Zn	Zea Mays	Biochar amendment significantly reduced the mobility of heavy metals in the aqueous soil medium	[163]
2	Rice straw	n. a.	Lanping Pb–Zn mines, Yunnan province, China	Cd and Pb	Maize	Biochar application decreased the availability of both total and plant-available contents of Pb and Cd in the soil	[164]
3	Sewage sludge	Slow pyrolysis at 500 °C	Contaminated soil, Sergipe experimental station, Northeast Brazil	Cu	Corn seeds	Biochar was applied at the rates of 30 t ha^{-1} and 60 t ha^{-1} reduced the exchangeable fraction of Cu by 96.2 and 57.5%, respectively	[165]
4	Hardwood waste	Slow pyrolysis at 500 °C for one hour	Degraded mined-out land	Cr, Cu, Zn, Pb, and Mn	*Oryza sativa*	Biochar applied at a rate of 3% (*w*/*w*) reduced the available contents of Cr, Cu, Zn, Pb, and Mn by 99.1, 71.7, 61.7, 36.4, and 47.9%, respectively	[166]
5	Chicken manure and Oat hull	Slow pyrolysis at 500 °C and 300 °C, respectively	Ventanas Cu smelter, National Copper Corporation of Chile (CODELCO), Puchuncaví, Valley of Central Chile	Cu	*Oenothera picensis*	Applications of biochars at the rates of 1 and 5% reduced the exchangeable and carbonate-bounded fractions of Cu in the contaminated soil	[167]
6	Eucalyptus wood	Slow pyrolysis at 400 and 600 °C for 150 min	Coal mine, Bastacolla area, Dhanbad, Jharkhand, India	Cr, Zn, Ni, and Co	*Accacia Auriculiformis*	Application of different temperatures biochar applied at the rates of 0.5, 1, 2, 3, and 5% (*w*/*w*) significantly reduced the concentrations of total and exchangeable heavy metals in the soil	[57]
7	Fishbone	Slow pyrolysis at 600 °C	Polluted soil located in an area near a gold mine in the governorate of Mahd ad-Dahab, Saudi Arabia	Pb, Cu, Zn, and Cd	Yecora Rojo	Fishbine biochar application at a rate of 30 g kg^{-1} reduced the mobility of Pb, Cu, Zn, and Cd by 43.0, 66.2, 55.6, and 33.8%, respectively	[168]
8	Rice straw Coconut shell Sludge	Slow pyrolysis at 500 °C	Mount Manao abandoned lead-zinc mine areas in Chenzhou city, Hunan Province, China	Cd, Cu, Pb, and Zn	-	Application of rice straw, coconut shell, and sludge-derived biochar at the rates of 0.5, 2.5, and 5% improved the soil's physicochemical properties and reduced the mobility of heavy metals	[169]

5. Conclusions and Future Prospects

The following conclusions can be drawn from the above analysis and discussions regarding the impact of mining on the ecosystem and environment, methods of remediation of heavy metal-contaminated soil, methods of production of biochar and its modification methods, and the role of the biochar and its composites for the remediation of such contaminated soils:

(i) The coal mining activities destroy the landscape, soil quality, and ecosystem in the impacted area to such an extent that restoring such degraded lands to their original state becomes difficult.
(ii) The degraded landscape triggers the release of heavy metals such as Cr, Cd, As, Mn, Co, Ni, and Zn from the contaminated mine soil into various surface and groundwater resources, which may have significant health impacts on humans and animals.
(iii) Phytoremediation is the most common and low-cost method to restore such degraded and contaminated mine soil by promoting the growth of metal-tolerant species in such soil. However, the poor physiochemical properties of the mine soil further limit the growth of hyperaccumulators in the mine soil.
(iv) The surface properties of pristine biochar can be modified by pre- and post-treatment methods. The acid- or alkali-treated biochar shows the change in the abundance and characteristics of functional groups and surface porosity. The biochar can also be combined with other minerals through thermal and chemical treatment methods to produce biochar composites having enhanced surface properties compared with the pristine biochar.
(v) The application of biochar to the contaminated and degraded soil improves the overall soil physicochemical properties and promotes the immobilization of heavy metals within the soil matrix, thus, promoting the phytoremediation of the soil. Precipitation, surface complexation, electrostatic attraction, and ion exchange are the dominant methods through which biochar adsorbs and immobilizes heavy metals in the soil matrix.
(vi) Remediating mine soils contaminated with heavy metals such as As, Cr, and Cd using pristine biochar is challenging. The application of modified biochar or biochar composites enhances the remediation process. The modified biochar's surface properties can be changed through the modification process to remove a particular class of heavy metals with anionic characteristics in the aqueous soil solution.

The modified biochar and biochar-based composites can have multiple applications other than the soil remediation process. The modified biochar can be used as an effective adsorbent to remove emerging pollutants of concern from wastewater, as their surface properties can be modified to promote the adsorption of such pollutants. In addition, modified biochar can be used as electrode material in electrochemical cells designed to electrochemically remove heavy metals, ions, and organic pollutants from wastewater.

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