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Review Article

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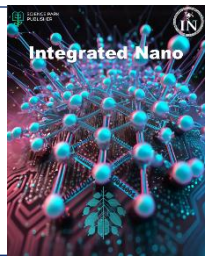

Modified biochar and its nanocomposites for efficient removal of aqueous Hg species from contaminated water

Kriti Shrivastava*

Centre for Renewable Energy & Storage, Suresh Gyan Vihar University, Jaipur, India

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Abstract

Mercury contamination in aqueous systems has emerged as a severe threat to the environment and human beings because of the toxicity of this heavy metal in different chemical forms and its persistent nature, leading to bioaccumulation followed by biomagnification. Efficient removal of all types of aqueous mercury species from the contaminated water can be done by different methods. Modified biochar and its nanocomposites are promising and sustainable adsorbents for this purpose, due to their low cost preparation and precursor abundance. This review presents the significant potential of modified biochar and its nanocomposites for effective recovery of aqueous mercury species to mitigate mercury pollution worldwide. This review comprehensively examines recent developments in the preparation and modification of biochar-based adsorbents for their specific application of aqueous mercury sequestration from the contaminated environment. Various methods of biochar preparation, modification, and biomass pretreatment techniques are discussed in correlation with the enhancement of physicochemical properties. Biochar-mercury binding mechanisms indicate complex interactions during the adsorption process, including electrostatic attraction, ligand exchange, redox transformations facilitated by metal nanoparticles, complexation with S-containing functional groups, charge shielding, cation bridging, etc. Biochar modification substantially improves mercury adsorption capacities by enhancing the selectivity, while preparation of nanocomposites demonstrates fast adsorption kinetics with high regeneration potential during several cycles. Major advantages like recyclability and low environmental impact make the modified biochar nanocomposites a potential candidate for scalable water treatment applications. However, it comes with the challenges of large-scale production processes, environmental safety issues, and comprehensive toxicity assessment. More scientific attention is required to optimize the regeneration strategies in order to facilitate a sustainable real-world mercury remediation technology.

Keywords: Mercury contamination; Biochar; Hg removal, Adsorption; Nanocomposites

1. Introduction

Heavy metal pollution is a global concern due to the chemical stability, extreme toxicity, and bioaccumulation capacity of the aqueous heavy metal species. Mercury contamination from different industrial sources such as mining, cement, fluorescent light bulbs, dry cell batteries, switches, metal plating, paints, fertilizers, fungicides, seed dressings, emissions from the coal power plants, etc [1]. Mercury (Hg) in different aqueous oxidation states is a severe threat due to its ability to bioaccumulate and biomagnify along the food chain. It not only disrupts the life cycle of aquatic flora and fauna but also leads to serious health issues in

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humans, such as neurological and developmental disorders. After its release into the environment, Hg can stabilize itself for about 3000 years through the process of global mercury cycle (**Figure 1**) [2]. Natural mercury pollution is caused by volcanoes and igneous rocks. Contaminated industrial effluents and small-scale gold mining are the major contributors of anthropogenic mercury pollution in the aquatic ecosystems. The United Nations (UN) initiative of Minamata Convention (2017) drew global attention towards the protection of human health and the environment from Hg and related species. Human activities to date have caused alarmingly high total atmospheric mercury concentrations of around 450% greater than the natural levels [3] In this convention, the proposed deadline of 2020 to curb mercury emissions by phasing out manufacture, import, and export of the listed Hg-products attracted significant scientific attention. It leads to several investigations related to the removal of mercury from the soil, air and water. Generally, Hg is transported through the atmosphere and subsequently deposited in land and water.

For wastewater discharge, the permissible limit of mercury contamination is 10 ppm, with a maximum guideline USEPA value of 2.0 ppm and a World Health Organization (WHO) value of 1.0 ppm for drinking water. Inorganic mercury remains in the natural waters in the form of Hg^{2+}, which is later converted into more toxic methylmercury (CH_3Hg^+) species by aquatic microorganisms such as iron-reducing, sulfate-reducing, and methanogenic bacteria through biological and non-biological methylation pathways [5]. Beyond the permissible limits, even the traces of mercury can adversely affect all the organisms associated with the relevant food chain, particularly the mental and nervous system, cardiovascular and immune system and reproductive systems, leading to physiological disorders and mortality [6].

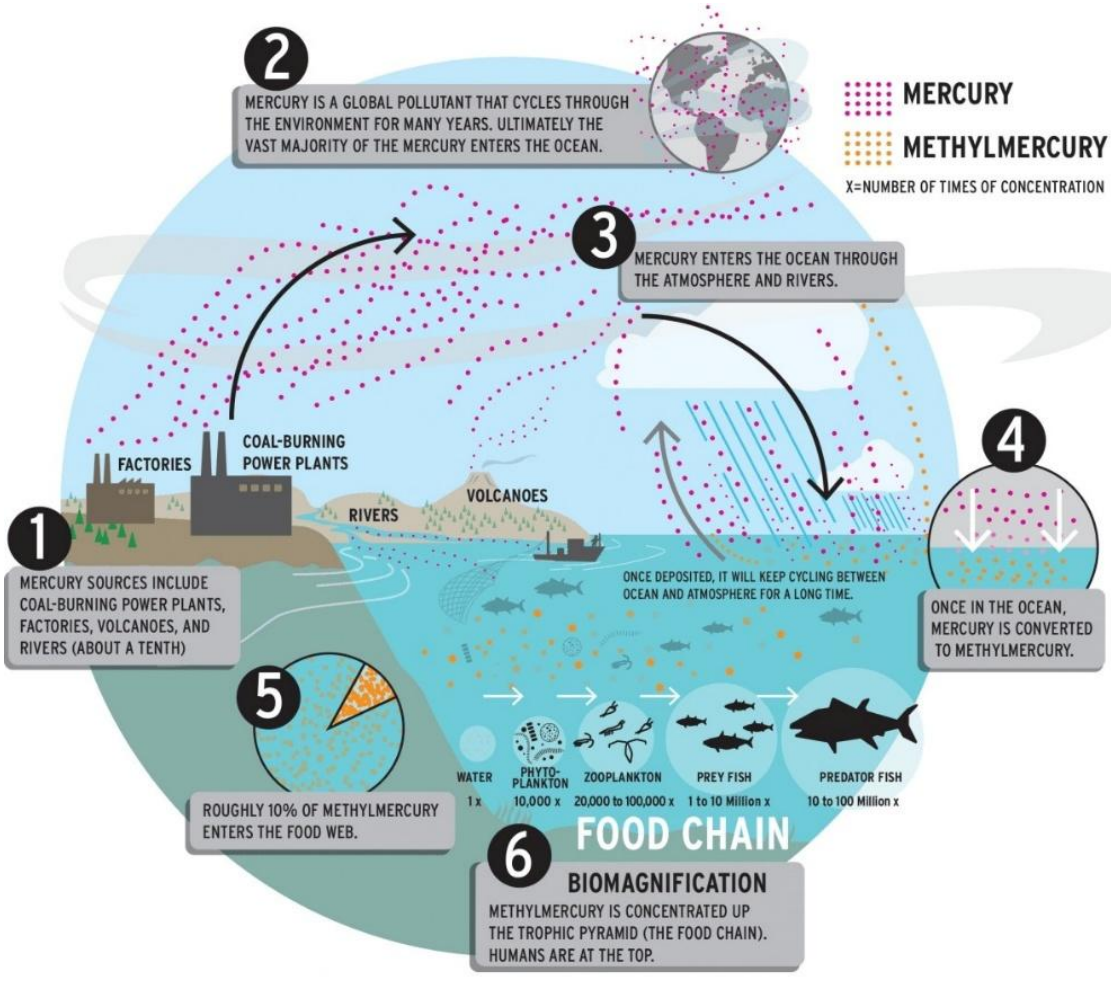

Figure 1. Global circulation of mercury in the environment and associated phenomenon [4].

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Conventionally, toxic mercury can be removed during wastewater remediation by the available technologies such as ion exchange, electrochemical treatment, coagulation, adsorption, chemical precipitation, membrane filtration, etc. However, their application for the large-scale aqueous mercury sequestration is limited in terms of the generation of secondary pollutants, high operational cost, and improper disposal of resultant toxic sludge. Among these technologies, adsorption stands as the most efficient and commercially employable method for the removal of mercury in trace amounts. Its feasibility is due to simple operation, low-cost, high removal efficiency, and ease of adsorbent regeneration [7].

In wastewater remediation, adsorption technology is commonly used for the removal of soluble organic and inorganic contaminants such as nutrients, hazardous metal ions, dyes, and pharmaceutically active compounds. An adsorbent is such a substance that is capable of simultaneously binding and holding molecules of gases, liquids, or dissolved solids on its surface by the process of physisorption and chemisorption. It is widely used in diverse applications of cleanup, purification, and separation, due to its high surface area and ability to selectively remove contaminants or impurities. The carbon-based adsorbents with typical advantages of having high surface area, porosity and tuneable surface chemistry are emerging as new promising candidate. Based on the carbon precursor, they can be classified into two types: Biochar and Activated Carbons [8]. When compared with the commercially available fossil fuel-based carbon materials, Biochar is considered as the most economical, sustainable, and low-cost, with the presence of natural heteroatom doping and surface functional groups. In addition to this, growing concern over the principles of circular economy and waste valorization has facilitated detailed research investigations into the development of biochar and biochar-based materials for different energy and environmental applications. Latest breakthroughs in the material science research and environmental engineering have further expedited research and development on biochar-based technologies due to their environmentally friendly nature and vast availability of precursor materials.

Biochar is a solid carbon-rich substance that is produced by the biomass pyrolysis under a limited oxygen environment. It is known for its potential to adsorb different soluble, colloidal, and suspended contaminants from the wastewater. Generally, pristine biochar displays limited adsorption performance towards aqueous Hg species at low concentrations as they contain insufficient surface functional groups and relatively low specific surface area. Therefore, it is generally pre-treated and activated through the processes of physical, chemical, and nanotechnological modifications. This helps in further enhancing reactivity and selectivity of biochar toward mercury. Such modified biochar can be converted into a tailor-made nanocomposite for effective mercury adsorption by the addition of metal or metal oxide nanoparticles, which incorporate additional pores with active sites, enhance surface multifunctionality for simultaneous immobilization of the contaminants, and increase overall stability.

This review article summarizes recent advances in modified biochar and its nanocomposites for the removal of aqueous mercury species, with an emphasis on their synthesis and modification techniques, mercury adsorption mechanisms, effectiveness in removing aqueous mercury species, regeneration capabilities, environmental safety, scalability, major challenges, and future directions.

2. Modified biochar and its nanocomposites for mercury removal

2.1. Mercury removal by pristine biochar

Pristine biochar is synthesized through thermochemical conversion methods like slow and fast pyrolysis, gasification, hydrothermal carbonization, microwave pyrolysis, torrefaction, etc. The process of pyrolysis is the most crucial, as the pyrolysis conditions generally control the biochar properties. Pyrolysis temperature greatly affects the physicochemical and surface characteristics of biochar. Higher pyrolysis temperatures increase the extent of carbonization, specific surface area, and aromaticity in the biochar, but it may reduce surface functional groups, which are important for Hg binding. Several studies have demonstrated pyrolysis temperature of 350–700 °C under low-oxygen environment as the most suitable pyrolysis condition for producing biochar with a diverse range of surface functional groups, such as -OH, -COOH, =CO, which play a critical role in pollutant adsorption. When different feedstocks were converted into biochar to investigate their potential for

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Hg adsorption from aqueous solution at environmentally relevant concentrations, the biochar generated at higher pyrolysis temperatures (600-700 °C) exhibited mercury removal efficiency exceeding 90%. In contrast, biochar produced at 300 °C generally showed removal efficiencies of 40-90%. Investigation of the biochar surface with Hg distribution maps indicated uneven distribution of Hg across the biochar particles because it binds to S-containing functional groups on the biochar surface having high sulphur content. In biochar with low sulphur content, Hg interacts with O and Cl functional groups [9]. Biochar prepared from corn straw [10], waste tea [11], municipal solid waste [12], and *Camellia oleifera* fruit shells [13] was found to show limited Hg removal capacity when compared with their counterparts with chemical modifications.

2.2. Mercury removal by modified biochar

Pristine biochar can be activated by several methods to improve its surface area, porosity, surface functional groups, and mechanical strength and can be converted into a tailor-made adsorbent for the effective removal of desired inorganic and organic contaminants. Among the physical methods of biochar activation, ball milling and sonication are two important methods to achieve nano-range particle size with enhanced surface area, porosity, and more reactive sites. Ball milling is a cost-effective top-down approach for nanoparticle generation [14], which can enhance surface reactivity and accessibility, thereby improving adsorption capacity and kinetics. Sonication helps in the exfoliation of biochar sheets, which increases micro and meso-porosity to enhance the fast and selective uptake of contaminant traces [15]. Chemical modification methods involve chemical activating agents such as acids, alkali, and salts, which helps to enrich the biochar surface with diverse surface functional groups. Functional groups or components can be attached to the biochar surface for highly selective adsorption of soluble impurities, which are otherwise difficult to remove by traditional methods or require high end technology-based methods. Acidic treatment with H_2SO_4, HNO_3, or HCl helps in the production of -COOH and -OH groups, facilitating metal ion removal by the process of complexation. In contrast, alkali treatments result in surface modifications in terms of change of surface charge, porous structure, and presence of binding sites.

For Hg adsorption, sulfurization using thiol groups has been found to be an effective activation technique due to the high affinity of Hg species for S-containing functional groups, which provide strong covalent binding sites and can achieve impressive Hg (II) sorption capacities of more than 300 mg/g [5]. Thiol-modified biochar with 3-mercaptopropyltrimethoxy silane (3-MPTS) has been reported to have high removal efficiency for aqueous Hg^{2+} (126.62 mg/g) and CH_3Hg^+ (60.76 mg/g) species. The introduction of -SH surface groups enhanced the selectivity for mercury sorption, especially methyl mercury species, through the ligand exchange as well as complexation processed between surface active sites and mercury. Compared to other surface functional groups, the important role of –SH was highlighted by the fact that increasing the 3-MPTS content during chemical activation enhanced Hg uptake capacity, even when partially hindered by the presence of natural organic matter (NOM), glucose, and humic acid in the natural waters [16]. Six biochar derived from six different feedstocks (rice, tobacco, corn, wheat, millet, and black bean straws) were activated by H_2S plasma modification and evaluated for mercury removal, which was enhanced from 26.4% to 95.5% after chemical modification. This can be attributed to the increase in the S-containing and carboxyl functional groups on the biochar surface during plasma treatment. Carboxyl and C–S are the main functional groups involved in mercury removal by the formation of HgS and HgO. However, this performance was obtained at low temperatures only, as active S is released from the C–S bonds at high temperature, to further oxidize Hg^0 in the gas phase [17].

In addition to sulfur-containing compounds, elemental sulphur can also be used for biochar activation towards Hg adsorption. Sulfurized wood biochar was directly impregnated with elemental sulfur to get Hg-uptake capacity of 107.5 mg g^{-1}, compared with 57.8 mg g^{-1} for the pristine biochar. Fast Hg adsorption was indicated by the intra-particle diffusion model, due to the boundary layer, which became slow when diffusing into the biochar pores. Mercury adsorption into sulfurized wood biochar was primarily governed by the Hg interaction with C-SO_x-C and thiophenic groups. This is in addition with the other interactions reported for the pristine wood biochar [18]. Biochar loses its adsorption potential during storage when it remains exposed to water and oxygen for longer

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durations, as the surface-active sites are occupied by water and oxygen. When such biochar was exposed to N_2-plasma treatment, decomposition of water molecules resulted in the formation of O-containing active radicals, which combined with the biochar surface to form oxygen-containing functional groups. Similarly, in the presence of surface-bound oxygen, biochar was easily sintered under plasma treatment, improving the mercury removal efficiency by oxidizing Hg^0 into HgO [19]. Post-pyrolysis exposure to sulfur stream was used to activate pine-needle biochar to demonstrate Hg (II) removal, which proceeded as an endothermic process depending upon irreversible C-Hg and S-Hg interactions, which did not allow the conversion of Hg(II) to elemental Hg during longer retention periods. Experimental analysis suggested increased Hg-adsorption with increasing solution pH and decreased Hg-adsorption with increasing content of dissolved organic matter [20]. Palm kernel biochar and coconut biochar (**Figure 2**) also displayed enhanced Hg uptake capacity upon sulfurization on the similar accounts, making them a potential candidate for treatment of Hg-polluted wastewater and suitable for large-scale natural water applications [21].

Recently, thiol-modified biochar has been reported for excellent but unstable mercury removal. When 3-mercaptopropyltrimethoxysilane was used as chemical thiolation reagent, the biochar surface exhibited greater numbers of surface sulphur, silicone, oxygen, and nitrogen elements with more negative charges on surface and surface defects, which results into faster initial reaction in the forward direction resulting into the mercury uptake due to thiol groups complexing with uncharged mercury species. Although some thiol groups were oxidized or precipitated, the resulting products like $HgSO_3$ and HgS were trapped by -OH groups and π bonds [5]. For Hg^0 removal, Br-modified biochar displayed better performance (up to 1786.85 μg/g), where the higher pyrolysis temperature and surface oxygen content increased the Hg^0 removal efficiency. In such cases, surface O* and Br* species cause oxidation of Hg^0, leading to its removal from the aqueous medium [22].

2.3. Mercury removal by biochar-based nanocomposites

When the biochar matrices are incorporated with the nanoparticles of metal oxides (such as iron, manganese, and zinc oxides), metals (such as zero-valent iron), layer double hydroxide (LDH), and sulfur-containing organic compounds (such as poly 2-aminothiophenol, 3-MPTS), it results into further enhancement of Hg-removal efficiency owing to the synergistic adsorption and redox reactions. To facilitate easy recovery and reuse of the adsorbent from aquatic systems, magnetic biochar nanocomposites can be prepared. Several biochar nanocomposites can be prepared by using synthetic routes such as hydrothermal, co-precipitation, and ball milling method.

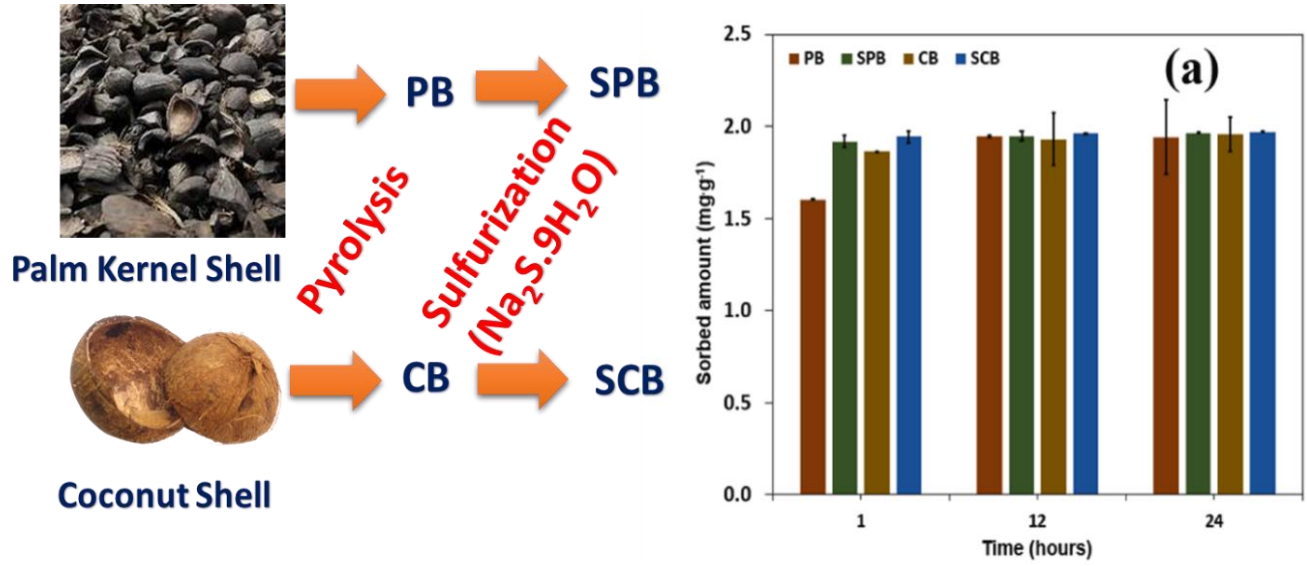

Figure 2. Mercury sorption capacity of pristine coconut shell biochar (CB), palm kernel shell biochar (PB), and their sulfur-functionalized derivatives SPB and SCB in a 1.0 mg/L Hg(II) solution at pH 6 ± 0.05 over 1, 12, and 24 h [21].

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$FeCl_3$- and $FeSO_4$-modified biochar prepared at higher pyrolytic temperature (900 °C) were reported to have higher Hg removal efficiency (>96%). EXAFS analysis revealed coordination of Hg with S-containing groups such as thiols and sulfide, which, in the case of other biochar, was reported to bind with O or Cl [23]. Ball-milled magnetic nanobiochar (BMBCs) derived from wheat straw exhibited a maximum removal capacity of 127.4 mg/g. The removal capacity decreased with increasing solution ionic strength but increased with increasing the solution temperature up to 45 °C. Adsorption of Hg (II) was mainly predominated by mechanisms of electrostatic attractions, surface complexation, and Hg–C π bond formation [14]. One-step co-pyrolysis was employed to prepare nano-adsorbent from waste rice straw and polyvinyl chloride using $CaCO_3$ as a template. It achieved over 90% Hg^0 adsorption performance at 120 °C. As compared to the earlier investigations, it highlighted synergistic effect of a high number of chlorine-containing active sites with well-developed hierarchically porous pores. In this Hg adsorption mechanism, chemisorption process occurred by simultaneous adsorption and oxidation. This finding emphasized the role of chemical species other than thiols in effective Hg-uptake. Micropore surface containing C–Cl bonds and reactive oxygen species served as active sites for the oxidation of adsorbed elemental Hg^0 into $HgCl_2$ and HgO, via the formation of HgCl as an intermediate [24]. Similarly, sulfurized magnetic biochar (SMBC) by single-step pyrolysis was investigated under acidic conditions, displaying thermodynamic spontaneity and an endothermic nature. This investigation helped in understanding the role of salinity in environmental remediation applications [25]. Zn-Al layered double hydroxides (Zn-Al LDHs) were another class of nano-materials utilized to form a stable support for grape stalk biochar combined with Mn-ferrite nanoparticles ($MnFe_2O_4$). The composite achieved 84.0% Hg-removal within 30 min; the process was governed by pseudo-second-order kinetics *via* intraparticle diffusion mechanism [26].

Biochar engineered with poly 2-aminothiophenol developed sulfur-containing functional groups on the surface, and as a result, maximum Hg-sorption capacity was reported for an adsorbent dosage of 0.75 mg/mL at the pH value of 9 [6]. When biomass was pyrolyzed at different pyrolysis temperatures, starting from 300 °C to 700 °C, and ball milled with 3-MPTS, it showed a preferential increase of Hg-uptake by 5.54 times as compared to the unmodified biochar prepared at low pyrolysis temperature [27]. Using the techniques of sol-gel followed by the co-precipitation method, Ce-Cu was used to modify iron-based biochar. The resulting 4%Ce-2%Cu-FeBC nanocomposite displayed maximum mercury removal performance. Mesoporous modified biochar with abundant metal active sites facilitates oxidative adsorption of elemental mercury. It exists on the biochar surface in the form of Hg^{2+}, Hg-OM, $HgCl_2$, and HgO. Modified biochar nano-adsorbent can be regenerated without any efficiency loss, as the chemisorption is the rate-controlling step of Hg adsorption [28]. With a novel approach, a biochar-microalgae complex was prepared with coconut shell activated carbon and Chlorella for remarkable Hg adsorption capacity of 46.8 μg/g of Hg at a concentration of 100 μg/L. The biochar-microalgae complex nano-adsorbent developed abundant oxygen-containing surface functional groups, e.g., -COOH, -OH and C-O-C, etc. and strongly adsorbed Hg through the processes of ion exchange and complexation between mercury and functional groups based on the single molecular layer theory of chemisorption [29].

Table 1 enlists some of the recently investigated biochar modification strategies for the adsorption of gaseous as well as aqueous mercury species. To further enhance the efficiency of biochar nanocomposites, the role of transition metals and their oxides cannot be ignored. These elements are known to display a catalytic effect in several applications and promote redox reactions by providing empty d-orbitals for electron transfer. Fe–Mn oxides were used to prepare modified biochar from waste straw biochar. It chemisorbed the Hg species by chemical complexation reaction with the formation of monodentate or multidentate inner-sphere complexes. Oxygen-containing functional groups and metal–π interactions favoured the rate kinetics. Fe/Mn atoms facilitated the electron transfer [39]. Magnetic pyrrhotite and magnetite-doped biochar were synthesized with a core-shell structure for the adsorption of Hg^{2+}, reaching a maximum capacity of 95.51 mg/g. It was found to lower down the Hg traces below 0.05 mg/L within 30 min. Core-shell structures with excellent conductivity ensured the minimal release of iron in aqueous media [40].

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Table 1. Mercury adsorption by modified biochar and biochar composites.

Biochar	Modification method	Adsorbed mercury species	Adsorption efficiency	References
Rice husk lignin	Macro-porous aerogel composite modified with MXene	Aqueous Hg (II)	98%	[30]
Brown seaweed "Sargassum muticum"	Pyrolysis	Aqueous Hg (II)	96.2%	[31]
Rice husk (*Oryza sativa* L.), mulberry twigs (*Morus alba* L.), and reeds (*Phragmites australis* (Cav.) Trin. ex Steud)	Co-pyrolysis with vermiculite	Aqueous Hg (II)	93.48%	[32]
Sugarcane bagasse-derived biochar	*In situ* oxidative polymerization	Aqueous Hg (II)	98.8%	[33]
Various Biochar	Sulphur functionalized	Gaseous Hg^0	90%	[34]
Rice husk biochar (RHB), Bamboo biochar (BB) and Tyre-modified biochar	Thermal Pyrolysis	-	72% - 85%	[35]
Cotton stalk biochar	H_2O_2 modification and NH_4Cl impregnation	Elemental mercury (Hg^0)	82.6 % and 116.2%	[36]
Iron-loaded biomass	One-step co-pyrolysis with in presence of brominated flame retarded plastic	Gaseous Hg^0	-	[37]
Industrial hemp stalks (*Cannabis sativa*) biochcar	Esterified with dimercaptosuccinic acid (DMSA)	Aqueous Hg (II)	99.8%	[38]

3. Mechanisms of mercury adsorption on modified biochar and nanocomposites

Mercury adsorption by activated/ modified biochar-based materials may undertake multiple pathways, including physisorption, electrostatic interactions, ion exchange, chemical reduction, complexation with functional groups, and chemical precipitation. The presence of surface functional groups such as =CO, -OH, -SH, and phenolic species provides the reactive centres to form stable Hg (II) complexes. Sulfurized biochar further facilitates covalent Hg–S bond formation, which is crucial for strong and selective Hg binding [41]. The high affinity between the thiol group and mercury (Log K_1 = 22.1 and 16.5 for Hg^{2+} and CH_3Hg^+ respectively) can be attributed to the significantly high mercury adsorption capacity of thiol-modified biochar [5]. The method of biochar pyrolysis and activation also affects the mechanism of mercury adsorption. Hydrothermal pretreatment at 180 °C could enhance the specific surface area of the precursor, and a more abundant porous structure can be constructed. It also promotes the ratio of chemisorbed oxygen and C=O bonds on the biochar surface, which can actually enhance the formation of HgO [42].

When metal/metal oxide nanoparticles are incorporated into biochar during nanocomposite formation, redox transformations increase due to the metallic electropositivity, such as the oxidation of elemental mercury Hg (0) to Hg (II), which enhances effective adsorption. DFT calculations revealed the presence of atomically dispersed surface metal nano-clusters, coordinated with O- and N-containing

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functional groups to provide favourable electronic environments for Hg adsorption. With the adsorption free energies up to −0.66 eV, the size of the metal clusters and their coordination sphere were found to affect the adsorption strength [20, 22, 40].

The physical adsorption mechanism predominates on the porous biochar surfaces in the case of organic mercury species, where specific surface area and distribution of pore size influence the overall sorption capacity and reaction kinetics. Mesopore volume can be increased using activating agents, such as $ZnCl_2$, during the process of pyrolysis. It increases Hg accessibility on the biochar surface in case of limited active metal sites. Environmental conditions such as the pH value of the aqueous medium, the presence of dissolved natural organic matter, and other competing ions may affect adsorption performance. Under acidic conditions, surface functional groups on biochar may undergo protonation, reducing sorption efficiency. On the other hand, the presence of co-contaminants can work in both directions, thus hindering or promoting Hg adsorption through synergistic effects [6, 25, 26].

Physical modification is favourable for the physisorption of elemental Hg^0 species. Chemical modification can also promote the uptake of Hg^0. In the presence of NH_4Cl impregnation, chemisorption of Hg^0 occurs due to the presence of C–Cl groups, which can transform elemental Hg^0 into $HgCl_2$ or other Hg–Cl complexes. The biochar modified by both the physical and chemical modifications display an excellent performance for Hg^0 removal [43]. To understand the reaction mechanism of Hg adsorption by biochar, corn straw biochar was fractionated into organic carbon (OC), inorganic carbon (IC), carboxyl-blocked carbon (BCC), and hydroxyl-blocked carbon (HBC). The reaction mechanisms for Hg (II) removal mainly include electrostatic adsorptive interactions, ion exchange, species reduction, precipitation, and complexation. Inorganic carbon displayed a slow rate but the highest capacity of Hg adsorption *via* the ion exchange mechanism. Organic carbon retains Hg species by the process of complexation with carboxyl and hydroxyl groups, which is the most dominant and majorly accountable for Hg adsorption. On the other hand, HBC and BCC adsorb Hg with a reduction–adsorption mechanism [44].

The performance of biochar-based nano-adsorbents in complex water matrices is generally affected by the presence of natural organic matter (NOM) and the ionic constitution of the aqueous media (**Figure 3**). Several S-containing sorption sites on the biochar surface bind Hg in the absence of natural organic matter. The negatively charged Hg(II)-chloro complexes were retained by the Ca^{2+} ion bridging. However, when natural organic matter is present, soluble Hg-NOM complexes are formed, which cannot enter biochar pores, suppressing the overall Hg removal efficiency. When ions like Cl^- and Ca^{2+} are also present, they aggregate Hg-NOM complexes to remove Hg(II) from the dissolved fractions. In the presence of Cl^-, weak O-containing functional groups of NOM were left behind, and in their place, small-sized Hg(II)-chloro complexes are formed, which could reach the additional intraparticle sorption sites. Evidence is available to claim that Cl^- ions can be outcompeted by S to immobilize Hg (II) species [45].

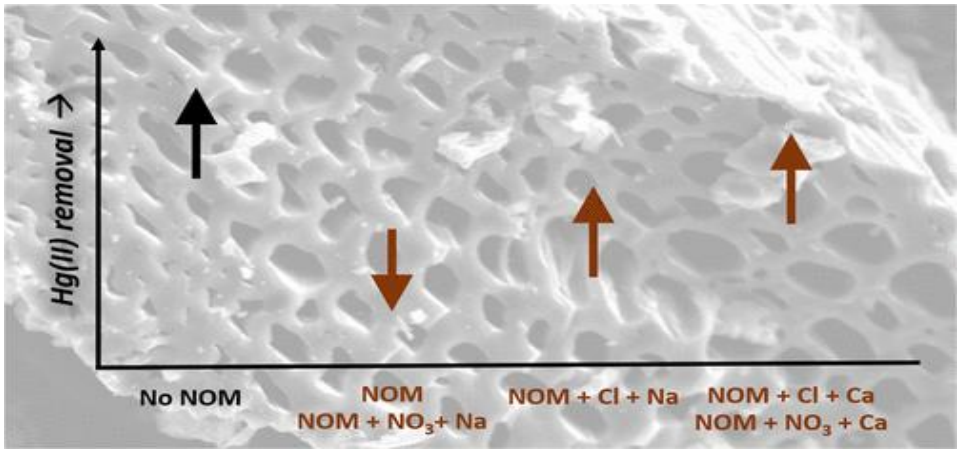

Figure 3. Effect of the presence of natural organic matter (NOM) and ionic constitution on biochar mercury sequestration [45].

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4. Current challenges and future perspectives

Extensive laboratory investigations have claimed that modified biochar can outperform pristine biochar for Hg removal. Biochar nanocomposites, when embedded with iron or copper oxides, display multi-fold enhancements in Hg and other heavy metal removal due to increased surface areas, O-containing groups, and redox activities induced by nanoparticles. Metal nanoparticle biochar composites also attained feasible kinetics, which is a crucial parameter for industrial-scale wastewater treatment. Multi-metal doped nanocomposites can sustain broader pH ranges and tolerate co-contaminants, further enhancing the wastewater remediation potential [8]. When compared with commercially activated carbon, modified biochar offers a competitive or superior mercury adsorption alternative with lower cost and greater environmental benefits, as they can be prepared from sustainable feedstocks *via* simple preparation methods. Regeneration potential is required for cost-effective yet large-scale applications. Among the various regeneration methods, including chemical washing, thermal desorption, and solvent extraction, recent studies have reported that iron oxide-biochar composites can retain high adsorption capacity (>90%) even after multiple regeneration cycles. Thiol-functionalized biochar is also capable of maintaining post-regeneration performance; however, gradual capacity loss is seen after multiple repeated cycles [46].

However, several challenges are also associated with the commercial utilization of these adsorbents, which are as follows:

- Limited regeneration due to the accumulation of Hg and other metals in biochar matrices.
- Issues of safe disposal of the spent adsorbent.
- Risk of the generation of secondary pollutants.
- Scaling up biochar production while maintaining the quality and performance consistency.
- Managing potential nanoparticle toxicity, their environmental fate, and future life cycle impacts.

Magnetic biochar nanocomposites may enable easier recovery of the adsorbent by decreasing the loss and contamination potential. It is necessary to develop green regeneration methods that not only preserve the surface functionality but also effectively remove the adsorbed mercury and maintain the active surface area [5, 21].

Application of modified biochar nanocomposites for Hg sequestration requires careful evaluation of the associated environmental risks and sustainability issues. On one hand, biochar offers advantages of being renewable, biodegradable, and having the potential for carbon sequestration. But on the other hand, the introduction of metal nanoparticles in biochar can enhance the efficiency and performance. However, it may have toxicity potential, nanoparticle release, and bioaccumulation in natural ecosystems. Various toxicological assessments have indicated limited acute toxicity of the nano-biochar towards humans and soil microflora, which needs to be further investigated. The presence of modified biochar can reduce metal bioavailability for plant uptake in soil systems. Life cycle assessments may suggest overall environmental benefits of the biochar usage for wastewater remediation and soil improvement but also emphasize optimization of its production and further application to minimize the negative impacts. Green synthetic routes for nano-biochar and their composites and the use of waste biomass further may improve sustainability profiles.

Modified biochar nanocomposites for mercury remediation still face obstacles before being widely used, despite the encouraging test results. Reproducibility and regulatory approval will be facilitated by standardizing characterization techniques and synthesis processes. The development of sophisticated adsorbents with improved selectivity, capacity, and regeneration capacities needs better knowledge of adsorption mechanisms at the molecular and atomic levels. Realtime applications can be determined by integrating functionalized biochar into the current water and wastewater treatment infrastructures and conducting cost-benefit assessments. Another interesting avenue is the creation of multifunctional biochar composites that may remove several pollutants at once.

5. Conclusion

There are several potential uses for biochar in water treatment research since it is an effective and promising adsorbent. Because adsorbent regeneration is reversible, adsorption is unique among wastewater treatment technologies in that it can provide high-quality treated effluent, operate with flexibility, and can be recycled. The

aqueous solution's pH has a big impact on how well heavy metals are removed *via* ion exchange. It has been discovered that carbon-based sorbents with functional groups like carbonyl, carboxyl, and hydroxy groups are efficient at eliminating heavy metals. Although unmodified biochar is still more economical, modified biochar, which is produced *via* chemical or thermal techniques, performs noticeably better in terms of adsorption efficiency [8]. Modified biochar and its nanocomposites represent a highly efficient, sustainable, and economically viable approach for the removal of aqueous Hg species from the contaminated water. Advances in synthesis techniques, such as chemical functionalization and composite preparation with metal or metal oxide nanoparticles, have significantly enhanced the adsorption capacities as well as the adsorption selectivity. Molecular-level insights into adsorption mechanisms have highlighted the significance of surface functional groups and atomically dispersed metal clusters. Although more toxicity research and life cycle analyses are necessary, environmental factors support these materials' ability to reduce ecological risks and minimize mercury contamination. These adsorbents' practical usefulness is increased by regeneration and reuse; however, optimization is necessary to avoid secondary contamination. All things considered, incorporating modified biochar-based nanocomposites into water treatment systems has a lot of potential to address the global problem of mercury pollution.

Authors' contribution

Kriti Shrivastava: Conceptualization, Original draft, Writing, Review and Editing.

Conflict of interest statement

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Author information

Corresponding authors: Kriti Shrivastava*
E-mail: kriti1885@gmail.com
ORCID iD: 0000-0001-9690-4124

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