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Application Mechanism and Efficiency Optimization of Carbon Nanomaterials in Heavy Metal Contaminated Soil Remediation

Ying Zhang

College of Chemical Engineering and Energy Technology, Dongguan University of Technology, Dongguan, China
PenicillinnNoah@outlook.com

Abstract. Heavy metal (Cd, Pb, As, Cr, Hg, Ni, Cu, Zn) soil pollution threatens food security, ecology, and human health. Carbon nanomaterials (CNMs) for in-situ remediation feature high surface area, adjustable functional groups, π-electron systems, and redox activity, simultaneously triggering adsorption, ion exchange, precipitation, and valence transformation in soils. Based on experimental evidence, this review summarizes key mechanisms by "metal-material" pairs: Pb/Cd undergo alkalization-complexation and carbonate/phosphate precipitation; As involves coordination with Fe/Al/Mn oxides and Mn-mediated As(III)→As(V) oxidation; Cr uses carbon-loaded iron for Cr(VI) reduction and Cr(III) precipitation; Hg forms stable Hg-S(-Se) coordination through sulfur/selenium anchoring; Cu/Ni/Zn coordinate with carboxyl/phenolic groups and undergo carbonate/hydroxide precipitation at higher pH. Key trade-offs include graphene oxide potentially enhancing Cd bioavailability in rice systems and phosphorus-rich biochar promoting As migration without iron binding. Multi-metal competition and rhizosphere processes affect long-term stability. A performance optimization framework emphasizes "material-process-index" synergy: material-level heteroatom doping, defect control, Fe/Mn/Al and S/Se functionalization, magnetic composites; process-level dosage optimization, moisture management, precise pH/EH control, and application sequencing.

Keywords: carbon nanomaterials, biochar, graphene oxide, carbon nanotubes, heavy metals

1. Introduction

Heavy-metal contamination of soils has emerged as a persistent barrier to safe food production, ecosystem health, and sustainable land use. Unlike organic pollutants that can be mineralized, metal ions such as lead (Pb), cadmium (Cd), arsenic (As), chromium (Cr), mercury (Hg), nickel (Ni), and copper (Cu) do not degrade; they transform, complex, or redistribute among solid, liquid, and biotic phases [1]. Mining and smelting, fossil-fuel combustion, wastewater irrigation, e-waste dismantling, and excessive inputs of fertilizers and pesticides all contribute to elevated concentrations that frequently exceed guideline values [2]. The resulting risks are multifaceted: phytotoxicity that depresses crop yields, trophic transfer that compromises human health through dietary intake, and

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the erosion of soil functions—from enzymatic activity to nutrient cycling—that underpin agricultural resilience [3]. Compounding the challenge is the heterogeneity of soils: texture, mineralogy, organic matter, pH, cation exchange capacity, and redox regime jointly determine metal speciation and bioavailability. Any technology that aims to mitigate risk must therefore perform across diverse geochemical contexts while preserving the ecological integrity of the soil matrix [4].

Conventional remediation technologies—excavation and landfilling, soil washing and flushing, solidification/stabilization with mineral binders, electrokinetic treatments, and phytoremediation—are well established but face practical constraints. Ex situ options are capital- and carbon-intensive and produce secondary wastes that require further management. In situ chemical immobilization is cost-effective but may sacrifice long-term stability if the sorbent dissolves, ages, or becomes passivated. Electrokinetic methods improve mass transfer yet demand continuous energy inputs and may struggle in highly buffered soils [5]. Phytoremediation can be land- and time-intensive, and its efficacy depends on plant species, rhizosphere chemistry, and climate. Against this backdrop, carbon nanomaterials (CNMs) have gained traction because they synergize multiple capture and transformation pathways in a single platform.

Materials based on graphene sheets, carbon nanotubes, fullerenes, nano-diamonds, carbon dots, nano-engineered biochars have features appealing to soil applications: very high specific surface area, hierarchical porosity, which is facilitative diffusion, rich defect chemistry and surface functionalities (–COOH, –OH, $–NH_2$, –SH), π-electron system, and redox, which can mediate changes in valency Cr(VI) to Cr(III) [6]. All these properties allow inner- and outer-sphere complexation, ion exchange, surface precipitation, and electron-transfer reactions to act in concert. Strategic engineering approaches have further enhanced the effectiveness of advanced nanomaterials for heavy-metal remediation applications [7]. What is more, CNMs may be agglomerated with iron or manganese oxides to nucleate and co-precipitate, agglomerated with magnets to be magnetized post-treatment, or agglomerated with polymers and ligands to bind to a particular metal based on hard-soft acid-base principles.

The review is devoted to application mechanisms and performance optimization of CNMs in remediation of heavy-metal-contaminated soils with the emphasis on how the material chemistry and process conditions influence the immobilization efficacy and risk reduction jointly. The scope spans pristine CNMs (e.g., graphene oxide, reduced graphene oxide, single- and multi-walled nanotubes, fullerenes), nano-structured biochars and activated carbons, and hybrid composites that integrate CNMs with minerals or polymers. Aqueous-only systems are referenced only as mechanistic analogues when they illuminate soil processes; the primary context remains real soils in batch, column, pot, and field settings. The review addresses three guiding questions [8]. First, by what dominant mechanisms—adsorption, complexation, redox transformation, surface precipitation, and biologically mediated pathways—do CNMs modulate metal speciation and bioavailability in heterogeneous soils? Second, which material attributes—heteroatom doping, defect density, pore architecture, functional group density, and composite formulation—most strongly control performance for specific metals and soil chemistries, and how can they be rationally tuned? Third, how should operating variables—dosage, contact time, moisture regime, amendment sequence, co-additives, and aging—be optimized to balance removal or immobilization with environmental safety, scalability, and cost? In addressing these questions, the review also considers potential trade-offs and unintended consequences, including the possibility of mobilizing metalloids such as arsenic under certain redox conditions, perturbations to soil microbial communities, and the need to ensure that CNMs themselves do not introduce new hazards.

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2. Target-Metal Mechanisms and Material Matchin

2.1. Lead (Pb)

Carbon nanomaterials (CNMs) in Pb-contaminated soils immobilize the risk, in part, through conversion of labile Pb^{2+} to sparingly soluble or strongly complexed forms and at the same time reduces plant availability. Biochar, especially at moderate-high pyrolysis temperature or post-modified, increases soil pH and alkalinity and causes carbonate equilibria to shift to cerussite/hydrocerussite, creating negative surface charge which contributes to outer-sphere attraction before inner-sphere complexation to carboxyl/phenolic O groups anchors Pb at edge sites. In soils containing phosphate (e.g. biochar is intrinsically P-rich such as biosolids biochars, or has been deliberately loaded with Pb), Pb can also convert to chloropyromorphite-like phases in situ, a process repeatedly observed to play a role in long-term immobilization in urban and agricultural soils [9]. This can be reinforced by composing CNMs with Fe/Mn oxides, which enhance the nucleation of co-precipitates and introduction of Lewis-acid metal centers to selectively form inner-sphere bonds; co-stabilization of co-occurring metals can also be realized with the same composites. New microbial approaches such as immobilizing phosphate-solubilizing bacteria (PSB) onto biochar surfaces further hasten the generation of pyromorphite and Pb fixation through preserving phosphate activity in the rhizosphere coupled with presenting high-surface area carbon scaffolds to be colonized [10]. This integrated approach exemplifies innovative strategies that combine nanomaterials with plant growth-promoting bacteria for enhanced contaminated soil remediation [11]. Taken together, these routes push Pb out of exchangeable/carbonate pools into oxide/residual fractions and reduce $CaCl_2$/DTPA-extractable Pb, the most policy-relevant measures of bioavailability [12].

2.2. Cadmium (Cd)

For Cd, the same alkalization–complexation–precipitation triad dominates, but the details are more pH-sensitive and crop-specific. Biochar's liming effect and carbonate alkalinity decrease Cd^{2+} activity and favor $CdCO_3$/$Cd(OH)_2$ precipitation; concurrently, abundant $–COO^-$/–OH groups provide inner-sphere complexation and ion-exchange sites that move Cd from exchangeable to oxide-/organic-bound pools. Meta-analyses across rice systems now indicate modified biochars (Fe/Mn minerals, phosphate, or heteroatom doping) reliably reduce DTPA-Cd in soils and Cd accumulation in shoots/grains, often with agronomic co-benefits [13]. Fe-modified biochars add co-precipitation with Fe phases and, upon aging, can keep Cd sequestered despite soil cycling; process-wise, soil pH and redox potential (Eh) remain master controls of efficacy [14]. Carbon nanotubes and graphene derivatives contribute via high-affinity oxygenated defects and cation–π interactions; magnetic/aminated GO composites show strong Cd binding in water systems and are increasingly reported in soil pots where they are best immobilized within larger matrices to avoid nanoparticle dispersion [15].

2.3. Arsenic (As(III/V))

Because As occurs as anions ($H_2AsO_4^-$/$HAsO_4^{2-}$) or neutral H_3AsO_3 in soils rather than as cations, the most effective CNM strategies invert relative to Cd/Pb. Bare biochars that raise pH or add labile phosphate can actually increase arsenate desorption by enhancing electrostatic repulsion and competitive anion exchange, potentially mobilizing As in paddy and upland soils. Reports now show

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that some biochars—and graphene oxide nanoparticles at modest doses—mobilize As even while immobilizing cationic metals, a caution that persists across textures and Eh regimes . The remedy is to pair carbon frameworks with Fe/Al (and often Mn) phases that provide strong, specific inner-sphere complexation sites for arsenate/arsenite; Fe-modified biochars, Fe_3O_4-coated hydrochars, and Fe–Mn co-modified carbons consistently pull As into less mobile fractions and lower porewater concentrations by ligand exchange and surface precipitation reactions, with Mn adding oxidative capacity that converts As(III) to As(V) before capture [16].

2.4. Chromium (Cr(VI/III))

Chromium uniquely invites exploitation of CNM redox capacity. In oxidizing soils and porewaters, the hazardous and mobile oxyanion Cr(VI) can be chemically reduced to Cr(III) by electron-donating moieties on biochar (quinone-like structures), by π-conjugated graphene surfaces, or—most effectively—by zero-valent iron supported on carbon. Biochar-nZVI (BC-nZVI) composites accelerate Cr(VI) reduction while the carbon scaffold mitigates Fe agglomeration and passivation; the resulting Cr(III) is then immobilized via precipitation ($Cr(OH)_3$) and complexation to carboxyl/phenolic O or to freshly formed Fe/Mn (hydr)oxides on the composite [17]. Field-proximal studies and controlled pot experiments now indicate that BC-nZVI not only depresses $CaCl_2$-extractable Cr but can also avoid large disruptions to microbial function when dosed judiciously.

2.5. Mercury (Hg^{2+})

Hg^{2+} is a soft Lewis acid; the most powerful CNM lever therefore is sulfur (and, to a lesser extent, selenium) chemistry integrated into a carbon matrix. Sulfur-modified biochars and S/Zn- or FeS–Se-functionalized carbons bind Hg through highly stable Hg–S or Hg–S–Se inner-sphere complexes that resist desorption and can approach the thermodynamic stability of metacinnabar-like domains, while the carbon backbone disperses reactive S sites and minimizes particle aggregation. Across soils and sediments, S-rich biochars repeatedly depress dissolved Hg and often suppress net methylmercury (MeHg) production or plant MeHg uptake by altering sulfur cycling and reducing microbial methylation niches [18]; From a design standpoint, pairing S with Fe (as FeS or magnetite@S-carbon) enhances both selectivity and retrievability, and adding Se can further passivate Hg through HgSe-like bonding while inhibiting re-activation under fluctuating redox. Risk-wise, careful control of dose and monitoring of methylation indicators (hgcAB gene abundance, MeHg:Hg ratios) are essential to confirm net benefit [19].

2.6. Copper/Nickel/Zinc (Cu^{2+}/Ni^{2+}/Zn^{2+})

Cu, Ni, and Zn are prototypical divalent cations whose immobilization on CNMs depends on the density and pKa of oxygen- and nitrogen-bearing groups, the availability of basic ash components (Mg/Ca oxides), and pH-controlled precipitation equilibria. Oxidized carbons—biochars aged in soil or deliberately oxidized, GO/rGO hybrids, and carboxyl-enriched CNTs—display enhanced inner-sphere complexation to carboxylate and phenolate sites; in biochars, a well-documented relationship exists between carboxyl-group abundance and stabilization capacity for Cu and Zn, with Cu typically exhibiting the strongest bonding and resistance to desorption because of its preference for chelate and mixed-ligand complexes . In soils, immobilization is therefore robust when amendments both supply these surface ligands and push pH toward neutrality/alkalinity, where carbonate/hydroxide precipitation of Zn^{2+} and Ni^{2+} complements complexation. Graphene-based

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sorbents and CNTs offer high-capacity uptake in water via cation–π and donor–acceptor bonding [20], but soil applications again benefit from hybridization—e.g., magnetic GO for recoverability or amine/thiol grafting to elevate selectivity—while biochar remains the scalable platform demonstrating consistent decreases in extractable Zn and Cu across soil types. Competition matters: in mixed systems or at higher ionic strength, adsorption sites fill preferentially with Pb/Cu, sometimes depressing Ni/Zn uptake; pH- and time-dependent models capture these trade-offs and can guide dose and co-amendment choices.

2.7. Mixed-metal system

Real contaminated soils rarely present a single-metal challenge, and the mechanistic behavior of CNMs under multi-metal stress reflects both competitive sorption and deliberate material selectivity. The most successful strategies are deliberately asymmetric: Fe/Al-rich carbon composites to capture anionic species (As) through inner-sphere complexation and surface precipitation, paired with carboxyl-/phenolic-rich carbon domains and basic oxides to immobilize cationic Pb/Cd/Zn/Ni via complexation, ion exchange, and carbonate/hydroxide precipitation. Mn addition can provide catalytic oxidation (e.g., As(III)→As(V)) and extra high-affinity sites, while sulfur/selenium grafting targets soft-acid Hg^{2+} without sacrificing cationic-metal capacity [21]. In practice, Fe–Mn-modified biochars have simultaneously depressed extractable Pb, Cd, and As and shifted all three into more recalcitrant fractions, whereas phosphate-rich carbons that excel for Pb can inadvertently mobilize As unless phosphate is co-confined within Fe domains. Batch/column tests and pot/field trials also confirm that competition reduces apparent capacities—especially for weaker binders like Ni/Zn—so tuning ligand density and pore architecture to preserve kinetic access is vital. In the presence of plants, rhizosphere processes (acidification, ligand exudation) may cause mobile forms of otherwise stable forms; CNM designs buffering pH, and redox-active minerals can suppress this effect. Finally, magnetization of graphene/biochar matrices not only facilitates retrieval and recycling but also improves dispersion and contact, an advantage in heterogeneous field soils.

3. Practical Optimization Framework for CNM-Based Remediation

3.1. Material tuning principles

The optimization of carbon nanomaterials (CNMs) to heavy-metal remediation is initiated by ensuring that surface chemistry is aligned to target-metal speciation in realistic soil conditions. For cationic species such as Pb^{2+}, Cd^{2+}, Cu^{2+}, Ni^{2+}, and Zn^{2+}, dense populations of oxygenated ligands—carboxylate, phenolate, and lactone—promote inner-sphere complexation, while basic ash components and mineral heterodomains supply alkalinity that drives carbonate and hydroxide precipitation. Graphene oxide and oxidized carbon nanotubes are valuable scaffolds because defect-rich basal planes and edge sites can be tuned by controlled oxidation or reduction to balance ligand density with electrical conductivity; the latter supports electron-transfer reactions important for redox-active metals like Cr [22]. For anionic species such as arsenate and arsenite, the strategy inverts: carbon frameworks must be hybridized with Fe/Al/Mn oxides or oxyhydroxides to create strong ligand-exchange sites that outcompete phosphate and silicate in soil porewaters. Sulfur or selenium anchoring on carbon matrices is the most selective approach for soft-acid mercury, forming highly stable inner-sphere Hg–S(–Se) complexes that resist desorption even as redox fluctuates.

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3.2. Process variables

Even an ideal material can underperform if the process window is poorly chosen. Dosage should be scaled to the soil's buffer capacity, contaminant loading, and competing ion inventory; in agricultural settings, mass fractions on the order of one to a few percent are typical for biochars, while highly active composites can achieve similar risk reductions at lower loadings when well dispersed. Mixing method governs contact efficiency: thorough incorporation into the root zone via tillage or blending yields more reproducible outcomes than surface topdressing; for hotspots or engineered fills, slurry injection can place CNMs precisely in high-concentration layers. Moisture controls diffusion and redox; maintaining field-capacity moisture during the initial curing period accelerates sorption, co-precipitation [23], and redox transformations. pH is a master variable and should be managed explicitly: liming or alkaline biochars may be introduced first to stabilize pH before adding Fe-rich composites meant for arsenic, thereby avoiding conditions that would mobilize oxyanions. Redox management is equally strategic; for chromium, slightly reducing microenvironments favor Cr(VI) reduction followed by Cr(III) precipitation, whereas for arsenic, mild oxidation combined with Fe phases promotes conversion of arsenite to the more easily sequestered arsenate.

3.3. Lab-scale recyclability

Recyclability diverges by platform. Biochar-based amendments are typically intended for in situ immobilization rather than retrieval; their "recyclability" is expressed as functional persistence across seasons and their capacity to retain metals through aging, leaching, and root turnover. In contrast, magnetic graphene/biochar composites and granular carbon carriers loaded with nanoscale actives can be physically recovered from slurries, columns, or thin reactive barriers. Regeneration protocols should be tailored to bonding mechanisms: mineral acids or saline eluents desorb outer-sphere-bound cations; chelators strip inner-sphere complexes on oxygenated sites; reducing agents convert Cr(VI) captured on carbon to soluble Cr(III) species during off-site regeneration, or, alternatively, fix Cr(III) in place while regenerating iron surfaces [24]; thiol-rich regenerants scavenge mercury but must be managed to prevent secondary pollution.

3.4. Evaluation metrics and reporting

Transparent, comparable reporting is essential to turn mechanistic insight into deployable guidance. Soil chemistry should be documented at baseline and after treatment: pH, electrical conductivity, cation-exchange capacity, texture, organic carbon, and redox potential. The evaluation of metal speciation and bioavailability should be complemented by such tools as quick extractions using neutral salts (e.g. $CaCl_2$) to recover the most labile pool; complexing extractants (e.g. DTPA or EDTA) to estimate plant-available fractions; and sequential extractions schemes to monitor the changes between exchangeable, carbonate-bound, oxide-bound, organic and residual pools. Carefully, the formation of target phases should be confirmed by direct spectroscopic evidence, X-ray absorption techniques, X-ray photoelectron spectroscopy [25], or by micro-imaging; this could be Pb-phosphates, Cr(III) hydroxides, or Hg-S complexes. Plant endpoints anchor risk reduction: tissue concentrations, translocation factors, and yield effects under agronomically realistic fertilization and irrigation regimes. Leaching risk should be probed with column tests or field lysimeters under realistic hydraulic gradients. For composites, mass balance on co-constituents (iron, manganese, phosphate, sulfur) clarifies which co-precipitates form and whether any nutrient

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runoff is plausible. Reporting should always include amendment characterization (surface area, pore distribution, functional-group density, ash mineralogy, point of zero charge), dose normalized to soil mass, mixing protocol, and contact time. Finally, uncertainty should be propagated: replicates across soil types, seasonality, and plant varieties will reveal the operational envelope of each material-process pairing.

4. Safety Notes, Evidence Snapshot, and Research Outlook

4.1. Environmental safety notes

Safety hinges on preventing the solution from becoming a new source of risk. Powdered nanoparticles disperse readily; form factors that limit respirable dust and subsurface transport—granules, pellets, or CNMs embedded in larger carbon matrices—reduce exposure to workers and non-target organisms. Biochars can carry residual polycyclic aromatic hydrocarbons and soluble salts depending on feedstock and pyrolysis conditions; thermal regimes and post-treatments should be selected to minimize these impurities, and pre-washing may be warranted for sensitive crops. For arsenic-bearing sites, pH elevation and phosphate release can mobilize oxyanions; coupling phosphate with iron domains and monitoring porewater anions guards against unintended consequences. Mercury amendments that rely on sulfide chemistry must be dosed carefully to avoid creating anaerobic niches that favor microbial methylation; tracking methylmercury alongside total mercury provides a clearer risk picture. Magnetized or metal-oxide-loaded carbons introduce secondary metals.

4.2. Evidence

Across lab, pot, and an increasing number of field demonstrations, well-matched CNM systems consistently reduce labile pools of divalent metals and depress plant uptake, with the strongest and most durable responses observed where material chemistry aligns with soil speciation and where pH/redox are actively managed. Lead stabilization is especially robust when phosphate is present within Fe-bearing carbon frameworks, often translating into marked declines in extractable Pb and tissue concentrations. Cadmium responses are more pH-sensitive but are enhanced significantly by Fe/Mn-modified or heteroatom-doped carbons which are a combination of alkalinity, complexation, and co-precipitation. In the case of arsenic, iron-bearing composites are more likely to be found than bare carbons, especially in soils with elevated phosphates; co-immobilization of cationic metals can be reduced without compromising reductions in porewater arsenic. Chromium An example of such redox-transformation is chromium in which carbon-supported zero-valent iron and conductive carbons are used to reduce Cr(VI) and immobilize Cr(III). Sulfurized carbons controlled with mercury are promising and are likely to remain in wet- dry cycles when the sulfur is firmly attached to the matrix. Mixed-metal soils demand asymmetric designs.

4.3. Practicality (light-touch)

Practical implementation is pegged on supply, management, and integrating with the available land management. Biochar presents good economics and scale due to the fact that biomass that is available in the region can be utilized to make biochar, and the characteristics of the biochar can be customized by using feedstock choice and process management. Additional steps like composite fabrication include iron loading, sulfurization and magnetization, which are expensive to undertake but may be justified when the contaminants are difficult to remove or recovery/regeneration is

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required. Logistically, pelletized or granulated forms are easier to transport and apply using common farm equipment and lead to less dust. When incorporated into the top soil during normal cultivation operation decreases the threshold to operations on cropland; when it comes to industrial land or mine tailings, slurry injection or overlay reactive covers can be better. Agronomic inputs, such as liming, fertilization, irrigation, are coordinated to maximize co-benefits such as pH buffering, micronutrient management and water-retention benefits.

4.4. Gaps and future work

A number of scientific and engineering issues require a joint effort prior to CNMs being worldwide accepted soil management tools. The long-term stability under true pedoclimatic cycles is constrained only in part: multiyear studies that trace the speciation along wet-dry and freeze- thaw- and crop-rotations are required to ensure that the immobilized metals are not re-entering the labile pools. Co-contaminant interference is not fully mapped; factorial designs need to measure the extent to which arsenic, phosphate, silicate, and dissolved organic ligands reorganize the performance of cationic metals, and vice versa. Effects at the microbial scale- positive and negative- should be studied in a mechanistic fashion using metagenomics and metabolomics to differentiate between transient community altersations and long-standing functional benefit or loss. Safety-by-design manufacturing practices must be advanced to upstream to include putting limits on the amount of leachable impurities, form factor management to reduce the escapes of nanoparticles, and recycling of spent composites.

5. Conclusion

Carbon nanomaterials provide a rich, versatile platform to reduce heavy-metal risks in soils, combining high surface area, functional tunability, and (when suitably composited) selective mineral chemistry, including adsorption/complexation, redox transformation, and surface precipitation and maintaining rhizosphere stability. The data on the elements of Pb, Cd, As, Cr, Hg and Cu/Ni/Zn reveals that the performance depends not on any one magic sorbent, but on rational combinations of material design (heteroatomic doping, hierarchical porosity, ligand grafting, Fe/Mn/apatite or S/Se domains, magnetization) with process windows optimizing pH, redox, moisture and co-amendments to promote speciation towards low-mobility pools without incurring the collateral ecological cost. Practical deployment should prioritize safe-by-design form factors (pellets, granules, embedded.

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