



Potential of nano-phytoremediation of heavy metal contaminated soil: emphasizing the role of mycorrhizal fungi in the amelioration process

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Abstract

Heavy metal pollution has become a grave environmental problem drawing worldwide attention. Reclamation of polluted land using innovative and ecofriendly way is crucial to restore soil fertility. During the past two decades, various approaches of bio-/phytoremediation are gaining their acceptance to remediate contaminated soil. Recently, the use of reactive nanomaterials for phytoremediation or ‘nano-phytoremediation’ is becoming popular, claiming to improve the phyto-availability of heavy metals and reduce their toxicity through transformation or detoxification. Nanomaterials exhibit distinct properties concerning size, shape, reactivity, and ratio of surface area to volume rendering their potential for a range of applications including remediation of polluted soil environments with heavy metals, chlorinated organic solvents, organochlorine pesticides, polycyclic aromatic hydrocarbons, and polychlorinated biphenyls. Further, the combination of plants and associated microbes such as rhizospheric bacteria or arbuscular mycorrhizal fungi has the potential to significantly improve the nano-phytoremediation of heavy metal contaminated soil. This review focuses on recent developments on nano-phytoremediation, plant root associated microbes, and their interaction for developing an integrated and efficient nano-phytoremediation strategy for improved soil remediation in general and for soil contaminated with heavy metal. The application of nanomaterials combined with phytoremediation was discussed.

Keywords Nanoparticles · Plants · Arbuscular mycorrhizal fungi · Nanoremediation

Introduction

Soil contamination by heavy metals (HMs) and metalloids due to unremitting development of multifarious industrial technologies is a current global nuisance compromising safe agricultural production and groundwater quality. Major source of HMs is from metal mining and milling processes, industrial wastes from tanning, textile, electroplating, gas exhausts, energy, fuel production, and petrochemical (Chaney and Oliver 1996). Industrialization, air deposition,

manure/sludge, and extensive use of chemical fertilizers have resulted in HM deposition in agricultural soils (Mehr et al. 2021). Extensive use of pesticides in agriculture and horticulture containing heavy metals, for example, Cu Hg, Mn, Pb, Zn, etc., is another source of HM contamination of soils (Alloway 1995). Application of livestock manures, composites, municipal sewage, and sludge amassed in soil also contribute HM contamination such as Pb, Cr, As, Cd, Cu, Hg, Ni, Mo, Zn, Se, Sb, etc. (Basta et al. 2005; Alvarenga et al. 2015). Because HMs cannot decompose in soil, they disrupt the soil ecology by inhibiting the development of soil microorganisms and plants, accumulating in various environmental spheres. This indicates a strong chance that HM will bioaccumulate, spread up via the food chain to the ecological pyramid, and ultimately reach humans, creating a health risk (Reza and Singh 2010; Parvin et al. 2019). To address the issue of HM pollution, presently phytoremediation technology has been adopted as a potential tool.

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Phytoremediation strategy is based on the principle of exploiting the plants to decontaminate soil, sludge, sediment, and wastewater by using various methods. These are phytoextraction (i.e., deportation of heavy metal/toxic pollutant from soil/water), phytotransformation (i.e., degradation or chemical modification of organic contaminants), phytovolatilization (i.e., contaminants directly or after chemical modification are taken up by plants and then volatilized into the environment), rhizofiltration (i.e., filtering water through mass of roots, where contaminants remain adsorbed), phytostimulation (i.e., enhanced degradation of toxic substances by rhizospheric microbes), and phytostabilization (i.e., reduced bioavailability of the pollutants by immobilizing it within the soil). For example, many plants of Compositae family are highly tolerant to HM and can phytostabilize HM such as Cr, Pb, Ni, Fe, Mg, Cd, and Zn by accumulating in their roots and stems (Nikolić and Stevović 2015; Siddiqi and Husen 2016; Chand et al. 2016; Gautam and Agrawal 2017). In order to improve plant phytoremediation of heavy metals (HM), a number of researchers are interested in combining rhizospheric growth-promoting bacterium (PGPR) with symbiotic arbuscular mycorrhizal fungus (AMF).

While phytoremediation holds great potential for remediating soil contaminated with heavy metals, its implementation is time-consuming. To increase the effectiveness of phytoremediation in HM-contaminated soils, recent research suggests applying a combination of phytoremediation and nanomaterials (Zhu et al. 2019; Yu et al. 2021). Using nanomaterials in phytoremediation, also known as nanophytoremediation, is an environmentally friendly technique that use plants to absorb and accumulate toxic pollutants that have been destroyed or adsorbed through the use of nanomaterials. This new remediation method has been shown to be successful in the elimination of HM by absorbing them, changing the toxic valence state to a stable metallic state, and by catalyzing the process (Karn et al. 2009; Gil-Díaz et al. 2014). Because nanoparticle-aided phytoremediation techniques successfully and more efficiently remove pollutants from polluted soil or water in shorter time than traditional phytoremediation methods, nano-phytoremediation technology offers enormous promise for environmental remediation of HM, organic, and inorganic contaminants (Ma and Wang 2010; Pillai and Kottekottil 2016; Souri et al. 2017). Adsorption, heterogeneous catalysis, electro-nanoremediation, photodegradation, and nano-bioremediation involving microbes are some of the different technical processes that can be used to apply nanoparticles (NPs) to contaminated soils and remove or immobilize HMs (Mukhopadhyay et al. 2021; Ahmed et al. 2021). Enzyme-based bioremediation using NP can also be used in phytoremediation (Singh 2009; Yadav et al. 2015). However, it is to be noted that in nano-phytoremediation of

polluted soils, NPs employed should not possess any hazards to plants and rhizospheric microbes and should be capable of increasing the phyto-availability of pollutants for efficient phytoextraction. In this context, biogenic NPs produced by soil bacteria and plants are promising candidates as they stimulate the synthesis of plant growth hormones, resulting in enhanced biomass output and pollutant absorption from roots to above-ground tissues. Further, manufactured NPs have physicochemical, surface, and optical-electronic properties that are shown to address problems that cannot be tackled by standard approaches. New processes, tools, materials, and chemicals with excellent performance and low energy consumption can be created using this technique.

Though the technology has shown to have several advantages, there are limited field scale studies, and the technology requires a thorough research investigation in various aspects such as prices, toxicity, recycling issues, etc. (Alazaiza et al. 2021). Even though NPs have been widely studied in the field of HM remediation, majority of the available review works focused on nanotechnology as a solution for HM remediation from wastewater. A comprehensive and systematic review work on nanotechnology-assisted phytoremediation for HM remediation in soil environment is rather limited. From the background study, we discovered that a thorough assessment of the state-of-the-art body of knowledge spanning various facets of the technology, such as microorganisms associated to plant roots and their role in developing a cohesive and successful nano-phytoremediation technique, is desperately needed. In order to remediate soil contaminants and improve the phytoremediation performance of plants grown on HM-polluted soils, this review will concentrate on recent advancements in nano-phytoremediation, plant root associated microbes, and their role in developing an integrated and effective nano-phytoremediation strategy. This review aims to focus on recent developments on nano-phytoremediation, plant root associated microbes, and their role for formulating an integrated and efficient nano-phytoremediation-based strategy to achieve desirable soil decontamination level and to boost up the phytoremediation performance of plants grown on HM-polluted soils.

Properties and types of nanomaterial applied for environmental remediation and advantages of nanoparticle-assisted phytoremediation

Nanoparticles are molecular assemblages ranging between 1 and 100 nm having distinct physicochemical properties compared with bulk material (Tewari 2019). Nanomaterial can be broadly classified into two categories depending on their chemical nature; *organic* such as carbon NPs

(e.g., fullerenes), and *inorganic* (gold and silver, and semiconductor NPs, TiO₂, ZnO) (Yadav et al. 2017). Based on to origin, NPs can be grouped as *natural* such as volcanic dust; *incidental* generating from anthropogenic sources (coal combustion, diesel exhaust, welding fumes) and *engineered* NP such as Ag NP, Zn NP, etc. (Khan 2020). Nanomaterials have unique properties such as higher surface area, higher reactivity, variable shape, and morphologies, suitability for catalytic activity that made them advantageous for treating toxic chemicals in contaminated water and land (Khin et al. 2012).

The major benefits of employing nanoremediation for soil and groundwater remediation, particularly for substantial site cleaning, include lower costs and faster cleanup, as well as complete degradation of certain toxins without the need to transferring the soil (Karn et al. 2009). Through application of desirable material at nanoscale, a greater activity could be ensured due to its higher surface area because of which a larger fraction of the material comes into contact with the material (Ahmed et al. 2021). Due to their novel surface coating and small size, many engineering NPs are highly suitable for in situ remediation applications. Further, NPs have greater ability to diffuse and penetrate the subsurface and thus can infiltrate into a contamination zone where microparticles cannot penetrate and they have higher reactivity to redox-amiable contaminants (Alazaiza et al. 2021). Surface plasmon resonance, quantum effect properties shown by NPs help to detect the toxic chemicals and requires less activation energy to initiate the chemical reaction (Rizwan et al. 2014). Nanotechnology-mediated sustainable techniques, such as Raman scattering, surface plasmon resonance, fluorescence spectroscopy, and optical detection systems are applied in recent dates for efficient remediation of contaminated land and waterbodies (Ghasemzadeh et al. 2014). Nanoparticles can also facilitate immobilization of microbial cells responsible for degradation or reclamation of specific chemicals (Rizwan et al. 2014). Nanomaterials are reported to be more competent, economic, and eco-friendly alternatives to achieve both sustainable management and environmental remediation.

The NPs exploited for environmental cleanup possess specific size, solubility, surface area, surface charge, and surface chemical composition (Das et al. 2015). The two primary categories of NPs are inorganic and carbon-based (Yu et al. 2021). The most popular and thoroughly studied NPs for use in environmental applications include nano zero-valent iron, titanium dioxide (TiO₂ NPs), and carbon nanotubes (CNTs). A wide range of hazardous chemicals, including HMs, can be bioremediated using nanoscale materials such as nanoscale zeolites, metal oxides, Pd/Fe⁰ bimetallic NPs, zinc oxide, carbon nanotubes, chitosan and graphene oxide, Ag–iron oxide, TiO₂NPs, polysulfone–zero-valent iron, and poly (acrylic acid)-coated iron oxide

(Das et al. 2018). Organic nanomaterials used for metal removal, radionuclide remediation, and degradation of toxic chemicals are dendrimers, modified dendrimers, CNTs, calcium alginate, and multiwalled CNT (Das et al. 2018). Different types of organic and inorganic NMs/NPs used for heavy metal remediation are presented in Table 1.

Biogenic production of nanomaterials for remediation of heavy metal

Recent nano research has made tremendous progress in developing NPs with unique properties that are distinct from original source material. Customarily, NPs are generated exclusively by physical as well as chemical methods. However, to prevent the production of unwanted or harmful by-products, environmentally friendly synthesis is required for establishing a reliable, sustainable manufacturing processes. It is critical that materials used to clean up pollution must not become a new contaminant itself once put into application. Green synthesis method offers a safe, nontoxic, and ecologically friendly way to make metallic NPs because of its economic potential and viability. Appropriate natural resources are necessary to achieve this goal and biodegradable materials represent huge potential in this regard. Further, the need for biosynthesis of NPs evolved on account of higher expenses incurred in the aforesaid methods. Therefore, in search of cost effective and ecofriendly pathways of synthesis of NPs, microorganisms and afterward plants are exploited to develop NPs with different chemical configuration, sizes, and morphologies for diverse application. Metal compounds can be reduced by enzymes and phytochemicals secreted from microorganisms into their corresponding NPs due to their antioxidant and reducing properties. Biosynthesis of NPs has been evidenced in bacteria, yeasts, algae, fungi, and actinomycetes besides vascular plants (Sastry et al. 2003). Organic and inorganic NPs of biological origin is strongly needed considering their sustainability and inexpensive in nature (Rizwan et al. 2014). Below NPs synthesis from various biogenic sources is discussed.

Plant-based synthesis of nanoparticles

Presently, green synthesis of plant-based NPs is gaining importance due to single-step biosynthesis measure and reduced generation of toxic by-products. The plants are more advantageous source of NP for their availability, non-toxic nature, and a broad variability of metabolites that may serve as reducing agents. Plant leaf extracts include essential ingredients for the formation of NPs. The majority of the phytochemical components in plants that are involved in NP bio-reduction include carboxylic acids,



Table 1 Example of nanomaterials used for heavy metal remediation

	Nanoscale materials	Effects	References
1	Chitosan	Adsorption of Pb (II) and removal	Qi and Xu (2004)
2	CeO ₂ -CNTs	pH-dependent adsorption of As(V)	Peng et al. (2005)
3	FeS	Effectively immobilize Hg in a clay loam sediment	Xiong et al. (2009)
4	Bimetal iron (III)-titanium (IV) oxide (NHITO)	Removal of As(III) and As(V) from arsenic-rich ground water	Gupta and Ghosh (2009)
5	Manganese associated hydrous iron (III) oxide (MNHFO)	As(III) sorption occurred with catalytic oxidation of As(III) to As(V) on the surface of oxide	Gupta et al. (2010)
6	Modified magnetic chitosan chelating resin	Temperature dependent adsorption of Cu (II), Co(II), Ni(II)	Monier et al. (2010)
7	Chitosan derivatives (chitosan-GLA)	Pb (II) biosorption	Ngah and Fatinathan (2010)
8	Magnetite reduced graphene oxide (M-RGO)	Removal of arsenic (As) from wastewater	Chandra et al. (2010)
9	Alumina-coated multi-wall carbon nanotube (MWCNT)	Pb (II) adsorption and removal	Gupta et al. (2011)
10	Graphene nanosheets (GNSs)	Adsorption of Pb(II) from aqueous system	Huang et al. (2011)
11	Polypyrrole-reduced graphene oxide (PPy-RGO)	Selective adsorption of Hg (II)	Chandra and Kim (2011)
12	IONPs immobilized in PEG/nylon membrane	Pb (II) absorption, treatment of Pb (II) contaminated water	Tong et al. (2011)
13	IONPs embedded in orange peel pith	Cr (II) removal	López-Téllez et al. (2011)
14	Fe-Mn oxide nanoparticles	As (III) immobilization	An and Zhao (2012)
15	Polysulfone-zero valent iron	As(III) remediation	Fresnais et al. (2013)
16	Polyacrylic acid modified nZVI (PAA-nZVI)	Removal of arsenate	Laumann et al. (2013)
17	Magnetic chitosan/graphene oxide (MCGO)	Pb (II) removal, methylene blue	Fan et al. (2013)
18	Zeolites	Removal of heavy metals	Oliveira et al. (2004), Kocaoba et al. (2007), Guan et al. (2010) and Mallard et al. (2015)
19	Hydrous cerium oxide (HCO)	Removal of Cr (VI)	Albadarin et al. (2014)
20	Kaolinite supported Fe/Ni nanoparticles Ag-iron oxide/fly	Simultaneous catalytic remediation of lead and nitrate	Joshi et al. (2015)
21	Fe (II)-montmorillonite	Removal of Cr (II)	Vinuth et al. (2015)
22	Titanium phosphate	Removal of heavy metals	Pol et al. (2016)

amides, flavones, terpenoids, sugars, ketones, and aldehydes are mostly involved in NP bio-reduction (Roy et al. 2021). Additionally, plant-derived carbohydrates such as glucose and fructose take part in metallic NP formation (Panigrahi et al. 2004). Plants are more suitable for NP synthesis than microorganisms as incubation time for metal ion reduction is shorter when using plants metabolites (Yadav and Rai 2017). Plant tissue culture and downstream processing approaches are most commonly used to generate metallic and oxide NP on a large scale.

A number of plants have been reported for their role in the synthesis of NP. Several authors have reported rapid and potential synthesis of Ag, Au, and Cu NPs using extracts of plants such as *Aloe vera* (Chandran et al. 2006), *Medicago sativa* (Gardea-Torresdey et al. 2002), *Azadirachta indica* (Shankar et al. 2004), *Avena sativa* (Armendariz et al. 2004),

Emblica officinalis (Ankamwar et al. 2005), *Humulus lupulus* (Rai et al. 2006), *Spinacia oleracea* and *Lactuca sativa* (Kanchana et al. 2011), *Gnidia glauca* (Ghosh et al. 2012), *Capsicum annum* (Jha and Prasad 2011), *Tridax procumbens* (Gopalkrishnan et al. 2012), and *Sargassum muticum* (Mahdavi et al. 2013). However, effect of engineered NPs differs from plant to plant depending on their mode of application, size, and concentrations (Siddiqui et al. 2015). Stable silver NPs of different sizes can be prepared from berry extract of *Solanum xanthocarpum* [10 nm] (Amin et al. 2012), the roots extract of *Coleus forskohlii* [82.46 nm] (Baskaran and Ratha bai 2013), flower extract of *Hibiscus rosa-sinensis* [5–40 nm] (Surya et al. 2016), *Dioscorea alata* tuber extract [10–25 nm] (Pugazhendhi et al. 2016), the latex of *Thevetia peruviana* [10–30 nm] (Rupiasih et al. 2015), and the rind extract of *Citrullus lanatus* fruit [17.96 nm]



(Ndikau et al. 2017). Plant-based NP synthesis shows great promise, but qualitative research is needed to understand the molecular, physiological, and biochemical mechanisms behind nanoparticles.

Nanoparticles produced by bacteria

Bacteria have the potential to deplete metal ions, making them excellent candidates for the production of NP (Iravani 2014). Bacteria are well known for heavy metal remediation through mobilizing, immobilizing, reduction, and precipitation. The unique metal binding ability of the bacterial cell makes them worthwhile for nanobioremediation technologies. Bacteria act as promising ‘biofactory’ for the synthesis of NP such as silver, titanium, gold, platinum, titanium dioxide, magnetite, palladium, cadmium sulfide, etc. (Iravani and Varma 2020). Bacterial enzymes can control specific reactions that lead to the formation of several inorganic NPs ranging in size 100–200 nm (Iravani 2014). Although NPs synthesized by bacteria are relatively pure, its further purification is needed by filtration. Bacterial species reported in NP synthesis are shown in Table 2.

Genetic engineering can help to develop bacteria with desirable properties suitable for HM detoxification and metal remediation. Characteristics such as metal tolerance, protein overexpression that can chelate with metals, metallic bioaccumulation, and synthesis of NP could be incorporated to bacteria (Paliwal et al. 2012; Poirier et al. 2013). For example, in recombinant *E. coli* strain, co-expressing metallothionein and phytochelatin synthase enzyme can be incorporated for NP biosynthesis. *E. coli*

bacteria bearing the reductase genes of both *Shewanella* sp. ANA-3 and *Salmonella enterica* serovar Typhimurium were found responsible for the production of arsenic sulfide NP (Chellamuthu et al. 2019). Silver NPs could be generated within a short period by using *Klebsiella pneumoniae*, *E. coli*, and *Enterobacter cloacae* culture, where bacterial nitro reductase enzymes are responsible for reducing the silver ions (Shahverdi et al. 2007). Gold NPs were synthesized by applying *B. megatherium* D01, where NP was encased with thiol self-assembled monolayer (Wen et al. 2009). Bacterial species like *Delftia acidovorans* can synthesize pure gold NP (Johnston et al. 2013).

Nanoparticles produced by fungi

Fungi are good source for the green synthesis of NP because of their simple structure and cost-effectiveness in laboratory cultivation and in industrial production of NP. Fungal production of metal/metal-oxide NP is relatively faster and more eco-friendly since they include a variety of enzymes. Compared to bacteria, fungus can produce more NPs (Mohanpuria et al. 2008). Fungi-mediated synthesis of NP may be in vivo where it occurs intracellularly within living mycelia; or in vitro where NP is synthesized by using fungal extracts (Ramanathan et al. 2013; Zhao et al. 2018).

Filamentous fungi have the potential to synthesize a broad range of metal NPs such as gold (Vágó et al. 2016; Kitching et al. 2016), silver (Duran et al. 2005; Shaligram et al. 2009), iron oxide (Saif et al. 2016), and bimetallic NP (Castro-Longoria et al. 2011) (Table 3). *Aspergillus terreus* was found promising for the synthesis of gold (size 10–50 nm) and silver (8–20 nm) NP from mycelial-free

Table 2 Application of various bacteria for the synthesis of nanoparticles

	Nanoparticles	Bacterial species	References
1	Silver nanoparticles	<i>Pseudomonas stutzeri</i> <i>Corynebacterium</i> sp <i>Staphylococcus aureus</i> <i>E. coli</i> <i>Leuconostoc lactis</i> <i>E. Coli</i> <i>Bacillus cereus</i>	Klaus et al. (1999) Zhang et al. (2005) Nanda et al. (2009) Gurunathan et al. (2009) Saravanana et al. (2017) Kushwaha et al (2015) Sunkar and Nachiyar (2012)
2	Magnetic nanoparticles	<i>Magnetosirillum magneticum</i> <i>Sulphate reducing bacteria</i>	Mohanpuria et al. (2008) Gericke and Pinches (2006)
3	Palladium nanoparticles	<i>Desulfovibrio desulfuricans</i> NCIMB 8307	Nair and Pradeep (2002)
4	Cadmium sulfide nanoparticles	<i>Clostridium thermoaceticum</i> <i>Klebsiella aerogens</i> <i>Escherichia coli</i>	Mandal et al. (2006) Sweeney et al. (2004)
5	Gold nanoparticle	<i>Alkalothermophilic actinomycete</i> <i>Thermomonospora</i> sp <i>Pseudomonas aeruginosa</i> <i>Deinococcus radiodurans</i>	Sastry et al. (2003) Sastry et al. (2003) Husseiny et al. (2007) Li et al. (2016)
6	SnO ₂ nanoparticle	<i>Erwinia herbicola</i>	Srivastava and Mukhopadhyay (2014)
7	Copper nanoparticle	<i>Pseudomonas fluorescens</i>	Shantkriti and Rani (2014)



Table 3 Mycosynthesis of different nanoparticle

Types of nanoparticles synthesized (shape and size)	Fungal species	References
Fe (spherical, 10–24.6 nm)	<i>Aspergillus oryzae</i>	Tarafdar and Raliya (2013)
Ag (spherical, 5–50 nm)	<i>Calocybe indica</i>	Sujatha et al. (2013)
Au (Decahedral and Icosahedral, 20–40 nm)	<i>Colletotrichum sp.</i>	Shankar et al. (2003)
Si (Quasispherical, 5–15 nm)	<i>Fusarium oxysporum</i>	Bansal et al. (2005)
Ti (Spherical, 6–13 nm)	<i>Fusarium oxysporum</i>	Bansal et al. (2005)
Ag (Spherical, 5–50 nm)	<i>Pleurotus florida</i>	Sujatha et al. (2013)
Ag (Spherical, 5–50 nm)	<i>Rhizopus stolonifer</i>	Afreen et al. (2011)
Ag (Spherical or rodlike 5–40 nm)	<i>Trichoderma viride</i>	Fayaz et al. (2010)
Ag (Spherical 8–60 nm)	<i>Trichoderma sp.</i>	Devi et al. (2013)
Ag (13–18 nm)	<i>Trichoderma asperellum</i>	Mukherjee et al. (2008)

filtrate (Balakumaran et al. 2016). It has been determined that NADH and NADH-dependent reductase are responsible for *Aspergillus terreus*'s ability to produce NP through bio-reduction (Li et al. 2011). *Fusarium sp.* has good potential for different types of NPs synthesis (Zielonka and Klimek-Ochab 2017; Rai et al. 2021). Molnár et al. (2018) used a two-step approach to create gold NP from thermophilic filamentous fungal strains: first, Au³⁺ was reduced to Au⁰, and then the core of the NPs was stabilized by capping agents, a biopolymer with a molecular weight larger than 3 kDa. *Trichoderma reesei* is a well-studied fungus has been found to contain a wide variety of enzymes that may facilitate NP synthesis compared to others (Vahabi et al. 2011).

Mechanism of action of nanoparticles

Adsorption, transformation, and photocatalysis, as well as catalytic reduction are the primary detoxification or degradation processes followed by NP in the treatment of environmental contaminants.

Adsorption

Adsorption is an exothermic process mediated by surface and ionic interactions to bind the contaminants on the surface of an adsorbent. The smaller size and increased fraction of atom of NPs lead to magnified surface energy in its chemical potential (Gubin et al. 2005). Certain adsorption properties of the NPs govern the adsorption process (Borisev et al. 2020), such as definite surface area (large surface-to-volume ratio), available adsorption sites and porosity as well as precise interactions with other chemicals, making them one of the most suitable adsorbents (Santhosh et al. 2016; Sadegh et al. 2017). Several prokaryotes and eukaryotes can synthesize nanoscale adsorbents. Different biologically produced nonadsorbents are reported to trap heavy metals. When nanoadsorbent is applied to environment containing

HM, depending upon the metal concentration and accessible surface area on adsorbent, metals diffuse on the external surface of the nanoadsorbents owing to diffusion through physisorption or chemisorption (Parvin et al. 2019). The affinity of HMs for an adsorbent determines their extent of removal from water.

Several surface modifications on NP as a technique to improve their adsorption effectiveness have been researched to make the greatest use of NPs and maximize their application. Enhancement of adsorption capacity, efficiency, dispensability, and stability or inertness in challenging conditions are the primary goals of this modification. They are predominantly iron-, alumina-, and silica-based surface-modified NP (Manyangadze et al. 2020).

Metal oxides

Metal-oxide nanoparticles (MeONPs) are promising as HM adsorbent. Nanoadsorbents based on metals such as iron oxide, zinc oxide, cupric oxide, and titanium dioxide are well-known MeONPs for HM removal (Recillas et al. 2011). Iron-oxide NPs have the best adsorption capability among the cheapest adsorbents. Lunge et al. 2014 reported maximum capacity of Fe₃O₄ NPs for trivalent (188.69 mg⁻¹) and pentavalent (153.8 mg⁻¹) arsenic adsorption from tea waste. Pb (II) and organic dye adsorption from aqueous solution by applying CuO NP (CuONP) was also reported (Farghali et al. 2013; Mustafa et al. 2013). Green synthesis of CuONPs from mint leaves and orange peel extract was found to successfully remove Pb(II), Ni(II), and Cd(II) ions from the contaminated water through adsorption process (Mahmoud et al. 2021). A zero-valent silver nanoadsorbent synthesized from *Phyllanthus emblica* leaf extract was shown to adsorb Hg(II) ions with a maximum adsorption capacity of 312 mg^g⁻¹ (Siva et al., 2013). Proanthocyanidins-functionalized gold NPs have the potential as good adsorbent for HM ions such as Ni²⁺, Cu²⁺, Cd²⁺ and Pb²⁺ and organic dyes in an aqueous solution (Biao et al. 2018).

Because of their relatively low cost, high adsorption capacity, and ease of regeneration, MeONPs are a preferred nanoadsorbent material for removing HM (Parvin et al. 2019).

Another advantage of MeONPs is their possibility of impregnation on activated carbon or other porous materials aiding in concurrent removal of HM and organic contaminants (Hristovski et al. 2009). MeONPs are easy to regenerate and retain their efficacy even after several regenerations (Qu et al. 2013).

Bimetallic nanoparticles (BNPs)

Synthesis of BNP is a biocompatible approach that exploits bacteria, yeast, fungi, and plants for synthesis of NP, either intra- or extracellularly at room temperature. The nanoscale particles are then coated with discontinuous layer of noble metals to enhance the reactivity as compared to uncoated iron (Chang et al. 2011). Bimetallic Au/Ag NPs were produced by applying single-cell proteins of *Spirulina platensis* (Govindaraju et al. 2008). Plant biomass is used as non-toxic natural reductants to reduce metal ions for the synthesis of BNP (Sarkar et al. 2021). The flavonoids and phenolics secreted by plants are accountable for both the reduction and stabilization of BNPs. Phenolic hydroxyl and protein content in pomegranate fruit extricate/extract are assumed to play a significant part in reducing and settling the bimetallic Au–AgNPs (Kumari et al. 2015; Sarkar et al. 2021). Other BNP synthesized successfully through bio-reduction method are Ag/Se, Ti/Ni, Au/Pd, etc. (Das et al. 2018). Higher catalytic potential of BNPs make them superior for environmental contaminant remediation (Litter et al. 2014).

It has been reported that BNP has a larger adsorption capability for HM than single metal-oxide NPs (Zhang et al. 2010; Babaee et al. 2018). Parsons et al., (2009) observed that bimetal oxide has 20–50 times the arsenic adsorption capacity of Fe_3O_4 and Mn_3O_4 . Because these magnetic nanomaterials are paramagnetic, they can easily separate HM from solution (Zhang et al. 2010).

Modified nanoparticle

The outer layer of the MeONPs may be chemically modified either by organic and inorganic molecules such as surfactants, polymers, biomolecules or silica and metals, metal oxide, and metal sulfide to increase the stability as well as efficiency in application. Modified NPs show higher reactivity, surface energy, and magnetic properties, and therefore can be very easily oxidized in aqueous medium (Das et al. 2018). The mechanisms of adsorption by surface-modified NP comprise of surface site binding, magnetic selective adsorption, electrostatic interaction, and modified

ligand combination (Jeevanandam et al. 2016). Study by Singh et al., (2014) showed that citric acid-coated magnetite NP effectively removed Cd(II) ions from contaminated water. Green synthesized magnetic NPs using *Cinnamomum tamala* (CT) leaves and *Jatropha curcas* (JC) latex extract were found to adsorb Co(ii) and Cu(II) ion from wastewater effectively (Das et al. 2020).

Multifunctional nanocomposite

To reduce the time required for the removal of individual pollutants from industrial effluents, concurrent adsorption of multiple HM is required, and multifunctional NPs are now manufactured using low-cost technique to handle the issue. Alsabagh et al., (2015) reported the preparation of a multifunctional nanocomposite of chitosan, silver NP, copper NP, and carbon nanotubes that can remove Cu(II), Cd(II), and Pb(II) with a higher adsorption efficiency compared with chitosan and the bi-nanocomposites alone within a short time. Fato et al., (2019) showed that ultrafine mesoporous magnetite (Fe_3O_4) NPs can satisfactorily remove Pb^{2+} , Cd^{2+} , Cu^{2+} , and Ni^{2+} concurrently from contaminated river water. Similarly, Matos et al., (2017) evaluated the effects of CNTs in soil remediation, particularly to immobilize the HMs ions (Ni^{2+} , Zn^{2+} , Pb^{2+} , and Cu^{2+}).

Catalysis

Nanocatalysts may be used in many fields and are more effective than traditional catalysts. The adaptability and efficacy of nanocatalysts are due to their smaller size (usually 10–80 nm) yielding a tremendous surface area-to-volume ratio and unique features, which are not present in their macroscopic counterparts (Das et al. 2018). Photocatalysis is acceleration of photoreaction by nanocatalyst to photodegrade organic and inorganic contaminants. When exposed to light, nanocatalysts can produce powerful oxidative free radicals that can breakdown a variety of organic contaminants and reduce HM levels (Subramaniam et al. 2019). These semiconductors absorb photons and excite electrons in the presence of light, resulting in the transportation of electron pair holes across the surface, leading to the formation of reactive oxygen species (ROS) such as OH^- and O_2^- . In the presence of light, these holes and electrons undergo oxidation and reduction processes, destroying organic contaminants (Subramaniam et al. 2019). Recently, semiconductor materials such as silver, titanium, and zinc oxide NPs have drawn considerable attention for photocatalysis-based environmental contaminant removal. Photocatalytic degradation is gaining priority due to its low cost, high stability, and rapid oxidation process. Silver NPs synthesized from the stem, bark, and root of *Helicteres isora* extracts was investigated as reducing and capping agent in

the degradation of various organic dyes, including eosin methylene blue, methyl violet, and safranin (Bhakya et al. 2015).

Though not many works have explored photocatalyst in HM remediation, these appear to be promising tool for HM removal as it can transform metal ions into less harmful by-products. The reduction efficiency of titania nanoparticle (TiO_2) to hexavalent chromium has been reported by Loryuenyong et al., (2014).

Chemical modification

Another effective method for removal of organic and inorganic pollutants by NPs is chemical modification. It includes successive oxidation and reduction of the metal compounds, and thus altering metal speciation resulting in alleviation of metal toxicity. Zero-valent nanoparticles (ZV NPs) can effectively transform HM such as Cd, Cr, Ni, Zn, and Pb (Das et al. 2018). Reduction of Cr (VI) by ZV NPs where it serves as electron donor has been explored in various bench and field-scale studies (Singh et al. 2012). Singh et al., (2012) reported that ZV NPs help with the reduction of and removal of Cr(VI) from the water by acting as an electron donor, releasing ferrous iron during the reduction process and by releasing molecular and atomic active H_2 and solid minerals having Fe(II) from NZVI corrosion as byproducts that help with the reduction process. Metal ions and NPs in their oxidation states undergo redox cycling. Studies support that application of ZV NPs can successfully transform HM such as Cd, Cr, Ni, Zn, and Pb in the contaminated sites.

Nanoparticle alleviates heavy metal stress in plants

Nanoparticles are found to be highly effective in minimizing HM stress in plants and are indispensable tool for present day application in phytoremediation of heavy metals (Gunjan et al. 2014; Tripathi et al. 2015). Under abiotic and biotic stress, several physiological and biochemical processes in the plants such as CO_2 fixation, nitrogen metabolism, water acquisition, and antioxidant systems are found to be regulated by NPs (Wei and Wang 2013), thereby, mitigating the adverse effects of the stress (Lei et al. 2008; Wei and Wang 2013; Tripathi et al. 2015).

Because of their quantum dot properties, NPs have a strong affinity for metals like Cu and Pb, thus reducing phyto-availability by serving as a barrier (Worms et al. 2012). The commonly used TiO_2 NP has been discovered to reduce HM phytotoxicity and increase photosynthetic rate, relative water, and chlorophyll content, and mitigate the harmful effects of metals and metalloids in many

plants (Singh and Lee 2016; Faraji and Sepehri 2018). Following HM exposure, TiO_2 NP-treated spinach was shown to have lower free radical concentration (such as H_2O_2 , malonyldialdehyde, and superoxide) and improved antioxidant enzyme activity (Lei et al. 2008; Zheng et al. 2008). TiO_2 NP application has been shown to be a potential method for reducing Cd stress in several plants (Faraji and Sepehri 2018). Furthermore, nanoscale hydroxyapatite treatment was observed to alleviate Cd toxicity in mustard (Li and Huang 2014). In Cr-treated plants, another potential NP of Si has been shown to promote plant growth, nutrient absorption, photosynthesis, leaf nitrogen content, and antioxidant enzyme activities (Tripathi et al. 2015). When exogenously sprayed with Si NPs to a cucumber plant under oxidative stress, it was discovered to maintain an optimal K^+/Na^+ ratio and promoted the activation of numerous important enzymes involved in ROS detoxification (Kader and Lindberg 2010; Alsaedi et al. 2017; Siddiqui et al. 2017). ROS scavenging efficiency of CeO_2 NPs in plants under oxidative stress has been reported by a number of authors (Rico et al. 2013a,b; Gomez-Garay et al. 2014).

Nano-phytoremediation of heavy metals in contaminated soil

The use of nanomaterials for environmental cleaning is fast rising, as NPs can effectively eliminate or lessen the toxicity of contaminants such HM, organic, and inorganic pollutants. In nano-phytoremediation, factors regulating the efficient uptake of NPs by plants include size, type, chemical composition, and the route for NP penetration within plants (Schwab et al. 2016). Following uptake through the plant root, NPs are transported within the plants tissue either through apoplastic (transport outside the plasma membrane, through xylem vessel) or symplastic (along with the movement of water between cytoplasm and sieve plates) pathway (Sattelmacher 2001; Roberts and Oparka 2003).

Several authors have demonstrated that zero-valent iron NP (ZV FeNP) can be applied for remediation of metal/metalloid-contaminated soil systems (Alidokht et al. 2011; Gil-Díaz et al. 2014; Fajardo et al. 2015;). The possible mechanisms for removal of HM by ZV FeNP is through immobilization of HM by adsorption and/or surface precipitation and redox reduction (Martínez-Fernández et al. 2017). The particles of ZV FeNP have a core-shell structure made up to 98% Fe and wrapped by a shell composed of iron oxides and hydroxides such as $\text{FeO}/\text{Fe}_2\text{O}_3/\text{FeOOH}$, rendering the characteristics of both iron oxides (sorption) and elemental Fe^0 (reduction) (O'Carroll et al. 2013). Recent research shows that applying 10% ZV FeNP reduces As bioavailability by immobilizing it and enhance growth of *Hordeum vulgare L.* plant while limiting plant absorption of



As (Gil-Diaz et al. 2016; Zhang et al. 2018a, b). According to Huang et al., (2016), ZVNP may efficiently enhance Cd residual deposits, hence limiting Cd availability, which has several negative consequences on plants. Low levels of ZVNP inhibited the buildup of Cd in plant roots, stems, and leaves, promoting plant development (Su et al. 2016). Plants, when used in conjunction with ZVNP may decrease the acid soluble percentage of Pb, lowering Pb toxicity in sediments (Huang et al. 2018). Vitkova et al. (2018) found that applying ZVNP in As contaminated soil encourages sunflower roots to effectively phytostabilize the soil. Because high concentrations of ZVNP (1000 and 2000 mg/kg) induced significant oxidative stress in the plant, low quantities of ZVNP combined with plant remediation proved to be beneficial (Jiang et al. 2018).

TiO₂NP is another highly researched NP with photocatalytic activity, greater reactivity, chemical stability, and the potential to change the mobility of HM in sediments, soil, or water. These properties of TiO₂ made it a potential tool for removal of various toxic metal ions specially, Cr (VI) and As (III) (Yang et al. 1999; Kim et al. 2013). Higher concentration of TiO₂NP in soil has been reported to induce the accumulation of Cd in roots and buds of *Glycine max* (L.) Merr. (Singh and Lee 2016). Andersen et al., (2016) reported that average root length of the cucumber and onion plants was increased in the presence of TiO₂NP in Cd contaminated soil.

Nano-hydroxyapatite (NHAP) is efficient in HM immobilization due to its good HM sorption capacity, low water solubility, higher stability under reducing and oxidizing environment, easy availability, and affordability (Zhang et al. 2009). Several studies have demonstrated the potential of NHAP in chelating Pb and Cd in contaminated soil (Zhang et al. 2009; He et al. 2013). Spraying of 5 g/kg NHAP in Pb-contaminated soil increased the removal rate of Pb (Jin et al. 2016) and thus reduced its the phytotoxic effect in *Lolium perenne* L. (Ding et al. 2017). Similar result was also reported by Wei et al., (2016), where NHAP decreased HM availability to ryegrass in contaminated soil and improved plant biomass as it possesses substantial Pb fixing property in contaminated soil (Ding et al. 2017). *Helianthus annuus* L. and rapeseed are strong phytoextractors of Cd from soil (Sun et al. 2018). Supplementation with bentonite NP curtails the exchangeable fraction of Cd further and converts the majority of it to non-reactive form. The higher the amount of bentonite in the soil, the lower the Cd concentration in plants and the greater the remediation effect (Sun et al. 2015). Liang et al., (2017) discovered that 0.2% (w/w) NHAP substantially enhanced Pb accumulation in above ground parts of ryegrass as compared to control plants.

Manganese oxides (including hydroxides and oxyhydroxides) can also efficiently immobilize the metals

due to their large surface area, negative surface charge in soil (Dong et al. 2000; O'Relly et al. 2003). MnO NP can be safely applied for soil remediation as nanoscale biogenic MnO is a natural and ubiquitous soil component. MnO can also be prepared biogenically by using bacteria and fungi besides the classical synthesis methods (Essington 2004; Singh et al. 2010; Zhou et al. 2015). Application of MnO NP in contaminated soil resulted in significant reduction in metals/metalloid concentration and evidenced higher adsorption capacity for Pb, Cu, Zn, Cd, and As (Della Puppa et al. 2013; Micháľková et al. 2014). MnO has high oxidative characteristics and can undergo oxidation–reduction and cation exchange processes, making them ideal for As remediation since they increase immobility by converting As(III) to As(V) (Watanabe et al. 2013; Villalobos et al. 2014). But Mn oxides do not give satisfactory result for Cr amendment in soil as it is readily oxidized Cr(III) to more toxic and mobile Cr(VI) (Fandeur et al. 2009).

Fe oxide NP (goethite, hematite, maghemite, and magnetite) also play crucial role in remediation of metals and metalloids from the soil system due to their adsorbing properties (Shiple et al. 2011). Nanomaghemite (nFe₂O₃), formed from oxidation of magnetite, was reported as a powerful scavenger of Cr (VI), Pb(II), Cd(II), and arsenite (III) (Lin et al. 2012; Tuutijarvi et al. 2012; Jiang et al. 2013; Komárek et al. 2015). Factors controlling the sorption by nFe₂O₃ include presence of citrate complexes, organic acids, or other nutrients (Vitekova et al. 2015; Martínez-Fernández et al. 2014). Zhang et al., (2010) reported higher as immobilizing efficacy of nFe₂O₃ compared to that of iron sulfide or ZV FeNP. Another study documented faster adsorption rate and highest adsorption capacity of nFe₃O₄ for Pb from wastewater relative to other NP (Nassar 2010). Zhang and Zhang (2020) reported that the presence of Fe₂O₃ NPs may influence the synthesis of starch granules in response to HM stress (Pb and Zn for 48 days) in castor (*Ricinus communis*).

Factors affecting the performance of np with particular focus on role of rhizospheric microbes

Efficiency of nano-phytoremediation depends on different conditions which could be broadly categorized as soil properties, contaminants properties, NP properties, and plant properties, that ultimately affect the uptake and accumulation of HM within plant tissue. Figure 1 depicts the various factors related to NP, soil, plant, and contaminant that influence the efficacy of the nano-phytoremediation process.

Properties of soil environment such as pH and temperature are the primary factors influencing the



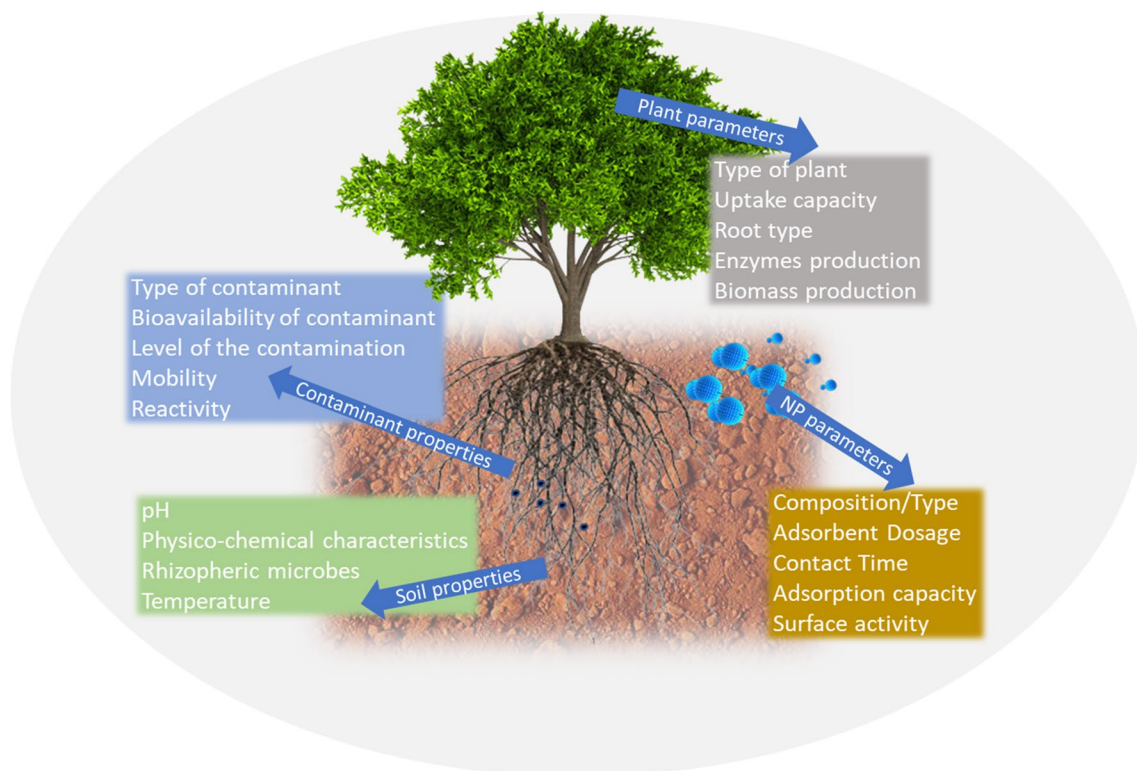


Fig. 1 Various factors related to nanoparticle, soil, plant, and contaminant that influence the efficacy of the nano-phytoremediation process

nano-phytoremediation process. Surface charge of NP is affected by the pH of the solution, which in turn influences its ability to adsorb HM and pH also has an effect on the bioavailability of HM ions and their speciation. A neutral pH generally favors higher removal of HM (Yu et al. 2021). Temperature is another key factor that may impact the equilibrium adsorption capability of NP and thus the removal efficiency. Metal immobilization is reported to increase as the temperature rises (Yu et al. 2021). The dosage of NP also affects the removal capacities of HM ions. It is crucial to identify the optimum application dosage for cost-effective and optimum removal efficiency. Significantly enhanced Cd uptake by *Trifolium repens* was reported by Zand et al., (2020) when TiO₂ NPs application dosage was increased from 0 to 500 mg kg⁻¹; however, a higher adsorbent dose, on the other hand, does not always imply a considerable increase in removal rate (Arshadi et al. 2014). The contact period between NP and HM also directly affects the removal efficiency during adsorption and redox process. The adsorption rate of HM ions onto NPs often peaks during initial phase and then slows until sorption equilibrium is attained (Gong et al. 2014). Cao et al. 2018 investigated Cd(II), Pb(II), and Zn(II) removal efficiencies from mine and agriculture contaminated soils using NZVI coupled with low-molecular weight organic acids. It was observed that the HM removal increased rapidly in the first 2 h and

gradually slowed until equilibrium was reached. The factors affecting the phytoremediation process were described in detail in previous publications (Srivastav et al. 2018; Yu et al. 2021). Hence, the current work focusses mainly on the role of rhizosphere microbes in the context of soil nano-phytoremediation.

A highly complex chemical, physical, and biological interactions exist between roots of plant and the ambient soil environment (Bais et al. 2006). Rhizosphere soil encompasses millions of endemic microbial species (Curl and Truelove 2012) that secrete various bioactive secondary metabolites such as siderophores, lipopeptides, and extracellular polysaccharides (Tyc et al. 2017). Plants are dependent on rhizospheric microbes in soil to collect nutrients that help mobilize the availability of essential nutrients (Burke et al. 2015).

Rhizosphere microorganisms influence the bio-accessibility of NP especially through the secretion of secondary metabolites, mediate NP lysis, and anti-aggregation, and may affect plant productivity (Dimkpa et al. 2012). Enhanced microbial siderophore synthesis was found to improve hematite (Fe₂O₃) NP dissolution, and liberated Fe were absorbed by plants (Barton et al. 2012). Siderophores may help dissolve metal NP and promote anti-hetero-aggregation of soil minerals and NP through metal chelation (Neubauer et al. 2000) (Table 4).



Table 4 Use of nanotechnology along with plant species for heavy metal remediation

Nanomaterial used	Target heavy metal	Species	Remarks	References
FeNPs	Cd	Wheat	Reduced Cd concentration and its phytotoxic effects; increased plant growth, rate of photosynthesis and activity of antioxidant enzymes	Hussain et al. (2019)
CMC stabilized nanoscale zero valent iron (nZVI)	Cr (VI)	Rape, Chinese cabbage	Reduced Cr run-off (82%) and bio-accessibility (58%)	Wang et al (2014)
Citrate-coated magnetite NPs	Cr (VI)	Wheat, oat, sorghum	Alleviation of metal toxicity	Lopez-Luna et al. (2016)
TiO ₂ NPs	Pb	Rice	Minimizes Pb accumulation in rice tissue and accumulate NPs in roots	Cai et al. (2017)
Salicylic acid nanoparticle (SANPs)	Arsenic	Woad (Isatis cappadocica)	705 ppm and 1188 ppm As accumulate in roots and shoots, respectively	Souri et al. (2017)

The greatest hurdle in the effective application of nanotechnology is homo-aggregation, which reduces surface area and bioavailability of NP (Ocoy et al. 2013). Rhizospheric microbes such as *Bacillus*, *Pseudomonas*, and *Streptomyces* produce lipopeptides, an amphiprotic molecule with surfactant properties, that may disperse NP and reduce aggregation formation through steric and depletion stabilization (Raaijmakers and Mazzola 2012; Kiran et al. 2011; Rangarajan et al. 2018). Use of *Bacillus* lipopeptides as capping and stabilizers can make Ag-NPs more stable (Rangarajan et al. 2018). It has also been shown that a lipopeptide produced from *Bacillus subtilis* that contains surfactin and fengycin is an excellent dispersant to form highly stable carbon NP (Khan et al. 2011). Another efficient NP stabilizer secreted by rhizospheric microbes is variety of multifunctional exopolysaccharides (EPS) (Khan et al. 2011; Lin et al. 2016). Rhizosphere of periwinkle (*Catharanthus roseus*) could generate stable Ag-NP that was found to aggregate only after 4 months (Sirajunnisa and Surendhiran 2014). Similarly, study of Xiao et al., (2017) showed that EPS of *Cordyceps sinensis* fungi restrained Se-NPs to form aggregate and upgraded their stability due to the intense interactions between Se-NPs and the –OH groups of the EPS to form new C–O···Se bonds. Other microbial exudates, for example, organic acids, thiosulfate, and cyanide may also steer metal solubilization (Wang et al. 2020).

The potential of rhizospheric microbes to enhance the biological activity of NPs through dissolution, dispersion, and anti-aggregation has been proved; however, it needs further research investigation. Considering the fact that various environmental factors such as pH, ionic strength, and organic matter have an impact on solubilization and homo-aggregation of NPs, practical outcome of the interaction of rhizospheric bacteria, and NPs in the agricultural field is quite unpredictable (Amde et al. 2017). However, positive

correlation between NPs and the colonization of the plant growth promoting bacteria on the root surface and enhanced production of secondary metabolites reported in number of works are very much promising (Palmqvist et al. 2015; Timmusk et al. 2018; Khan et al. 2018; Panichikkal et al. 2019).

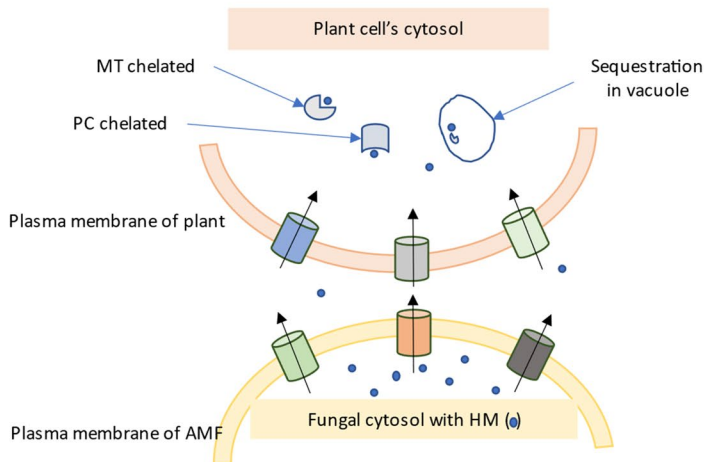
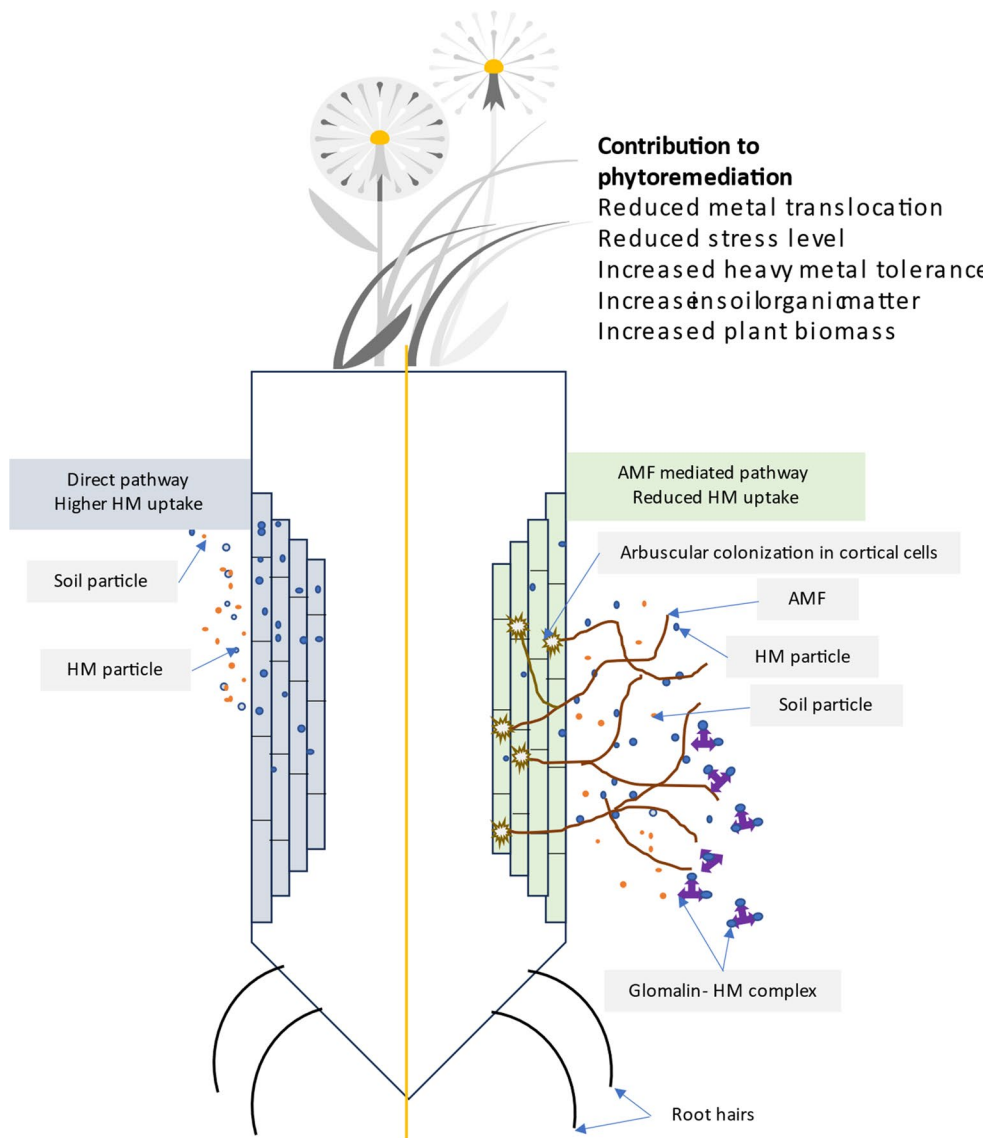
Role of Arbuscular mycorrhizal fungi in phytoremediation of heavy metal

Arbuscular mycorrhizal fungi (AMF) such as *Glomus intraradices* and *Glomus mosseae* are a widely recognized symbiotic group of organisms that play a significant role in enhancing ecosystem efficiency (Mitra et al. 2021). AMF can help in promoting plant growth and overall soil health ability as it enhances plant water and nutrient uptake, form a complex web of interactions with soil microorganisms, regulate pathogenic proliferation, influence soil structure and water retention through mycelial network, and alleviate abiotic stressors such as HM (Smith and Read 2008; Miransari 2011; Khan 2020).

Arbuscular mycorrhizal fungi mediated heavy metal uptake by plants

AMF plays an important ecological function in the phytostabilization of soils by sequestering toxic trace elements and thus aid the survival of mycorrhizal plants in polluted soils (Fig. 2). AMF cell wall proteins can sorb and sequester potentially toxic elements. The glycoprotein Glomalgin secreted by the AMF hyphae (Wright and Upadhyaya 1998; Gonzalez-Chavez et al. 2004; Driver et al. 2005) can decrease bioavailability of toxic metals through sequestration (Fig. 2). Glomalgin can also bind to soil and assists in stabilizing aggregates (Khan 2005).

Fig. 2 Schematic representation of contribution of AMF to plant heavy metal acquisition and distribution and tolerance. In direct pathway (upper left corner) metals are transported through high-affinity metal transporters located in root hairs. In AMF-mediated pathway (upper right corner), metal translocation occurs along the hyphae to fungal cell and then to the root at the symbiotic interface (lower panel). Thus, under toxic heavy metal condition mycorrhizal plants leads to increased removal of heavy metal from the soil (phytoextraction). Heavy metal tolerance in mycorrhizal plants is achieved by heavy metal binding to the fungal cell wall, chelation by glomalin, decreased uptake, and heavy metal compartmentalization in the vacuoles and chelation with phytochelatin (PC) and metallothioneins (MT) (lower panel of the diagram)



Mycorrhizae may be crucial for the survival of plants at HM-contaminated sites. Increased mycorrhizal colonization in the soil can be linked to a parallel rise in plant biomass and decrease in plant HM concentrations. Accordingly, depending on mycorrhizal inoculum, the root density of AMF affects heavy metal tolerance (Weissenhorn et al. 1995). The mycorrhizal association of *Trigonella foenum-graceum-Funneliformis mosseae* (*Glomus mosseae*) grown in as-contaminated soil showed an increased HM tolerance in the *Trigonella* compared to its non-mycorrhizal form (Trotta et al. 2006). The symbiotic association between mycorrhizal fungi and plants facilitates HM accumulation and uptake through the AMF. The symbiotic association of mycorrhizal fungi *Glomus mosseae*, *Glomus geosporum*, and *Glomus etunicatum* with *Plantago lanceolata* L. was reported to augment As accumulation (Wu et al. 2009; Orłowska et al. 2012). AMF can negate the negative effect of Cd on plant by restricting its translocation to the root of the plant (Janouskova et al. 2006). HM in soils added through chemical fertilizers can be significantly reduced by utilizing mycorrhiza (Atakan et al. 2018). Yang et al., (2016) reported significant enrichment of Pb in the roots of mycorrhizal plants to protect aerial portions of the plant. Similarly, Diaz et al. (1996) found that under varied levels of HM stress, mycorrhizal *Lygeum spartum* and *Anthyllis cytisoides* plants accumulated more Pb and Zn in their roots than non-mycorrhizal plants. The study found a greater root to shoot Pb ratio, showing that mycorrhizae inoculation boosted Pb uptake and accumulation in the root system compared to non-mycorrhizal plants.

Arbuscular mycorrhizal fungi induced heavy metal sequestration and tolerance in plants

Several studies documented the impact of AMF colonization on the expression of plant genes linked to HM tolerance and detoxification (Rivera-Becerril et al. 2005; Hildebrandt et al. 2007; Cicatelli et al. 2014; Shabani et al. 2016). Promising species for phytoremediation include *Gigaspora margarita*, *G. decipens*, *Scutellospora gilmorei*, *Acaulospora* sp., *Glomus* sp., *Acaulospora*, *Claroideoglossum*, *Diversispora*, and *Sclerocystis*. (Cabral et al. 2015). Synthesis of phytochelatins that plays an important role in intracellular detoxification of HM is enhanced by AMF (Garg and Aggarwal 2011; Garg and Chandel 2012). Synthesis of metallothioneins (MTs), another crucial protein family involved in the sequestration of toxic metals is triggered in presence of HM in number of species (Kumar et al. 2005; Folli-Pereira et al. 2012). MTs play a multifaceted role in regulating HM levels in plants and protecting cells from the metal-induced oxidative damage. Cicatelli et al. (2010) demonstrated activation of metallothionein MET genes in Cu and Zn contaminated soil following introduction of *F. mosseae* or *G. intraradices*

to *Populus alba*. Conversely, the mycorrhizal symbiosis could mitigate Cd stress in *Pisum sativum* by enhancing the expression of the PsMTA MET gene (Rivera-Becerril et al. 2002). Similarly, Shabani et al. (2016) observed a significant decrease in Ni transfer from the roots to the aboveground portions of *Festuca arundinacea* plants after *F. mosseae* inoculation as opposed to non-mycorrhizal plants. When compared to non-mycorrhizal plants, tall fescue plants colonized by *F. mosseae* exhibited higher amounts of ABC transporter and MT transcripts (Shabani et al. 2016). Studies reporting the contribution of AMF to phytoremediation and metal are listed in Table 5.

Application of Arbuscular mycorrhizal fungi in phytoremediation

One of the best ways to improve the effectiveness of phytoremediation is to manipulate microbial consortia, which entails changing how microorganisms interact with metals and optimizing the symbiotic relationship between them and plants. Furthermore, selection of specific microbes and their genetic modification for enhanced metal tolerance can improve phytoremediation efficiency. Some fungal species can reduce metals concentration and resist them, leading to the formation of gold, platinum, and silver NPs within their intracellular compartments, which is facilitated by hydrogenases present in the periplasmic and cytoplasmic regions.

Nonetheless, it is evident that AMF can regulate HM distribution by impeding its transportation from the root to the shoot (Christie et al. 2004; Wang et al. 2007; Wu et al. 2016). Previous study revealed ability of *Diversispora spurcum* and *Funneliformis mosseae* to retain HM in the belowground maize parts, thus decreasing Zn, Pb, and Cd in the shoot (Zhan et al. 2018). Thus, AMF can reduce HM stress of plants (Fig. 2). For example, *Calendula officinalis* developed slowly under HM stress (Hristozkova et al. 2016), which could be mitigated by AMF incorporation that reduced metal uptake and increased beneficial secondary metabolites. Mycorrhizal tomato plants exhibited decreased Zn absorption and concentration (Watts-Williams et al. 2013). Mycorrhizae can induce several biogeochemical changes in the rhizosphere including HM immobilization (Kangwankraiphaisan et al. 2013). AMF has been shown to mitigate metal stress of *Phragmites australis* and *Iris pseudacorus* through their conversion into metallic NPs (Manceau et al. 2008). Only limited studies report impacts of NP on AMF. Some NPs including Ag, FeO, and zero-valent iron have been reported to exhibit toxicity toward AMF by through oxidative damage (Pawlett et al. 2013; Cao et al. 2016; Feng et al. 2013). Wu et al. (2018) observed that the application of zero-valent iron (nZVI) NP at 0.5% (w/w) concentration along with AMF influenced the uptake and



Table 5 Contribution of arbuscular mycorrhizal fungi (AMF) to phytoremediation of heavy metals and increased HM resistance in plants grown in contaminated soil (pot experiment)

AMF species	Host plant	Heavy metal	Effects	References
<i>Glomus intraradices</i>	<i>Linum usitatissimum</i>	Ni	Enhanced tolerance to Ni toxicity, higher uptake of Ni	Amna et al. (2015)
<i>Glomus mosseae</i> , <i>Glomus fasciculatum</i> , <i>Gigaspora margarita</i>	<i>Panicum virgatum</i>	Pb, Cd	Accumulation of high concentrations of Pb and Cd in root; approximately twofold phytoextraction of metals from the Pb/Cd contaminated soil; increased tolerance to Pb and Cd	Arora et al. (2016)
<i>Funneliformis caledonium</i> , <i>Funneliformis mosseae</i>	<i>Helianthus annuus</i>	Cu, Pb, Cd, Zn, Ni, Cr	Enhanced P absorption and plant growth, decreased heavy metals in the shoots	Zhang et al. (2018a, b)
<i>Glomus</i> sp.	<i>Zea mays</i>	Hg	Higher Hg concentration in central cylinder of the AMF-colonized plants, increased overall biomass	Debeljak et al. (2018)
<i>Glomus intraradices</i> , <i>Glomus mosseae</i> , <i>Glomus fasciculatum</i>	<i>Acroptilon repens</i>	Cd	Higher biomass under stress Conditions, higher Cd accumulation in roots	Rasouli-Sadaghiani et al. (2019)
<i>Glomus etunicatum</i> , <i>Gigaspora margarita</i> , <i>Acaulospora tuberculata</i> , <i>Glomus Manihotis</i>	<i>Anthocephalus cadamba</i>	Pb	Reduced Pb concentration in roots, stems, and leaves; reduced phytotoxicity; increased seedling tolerance	Setyaningsih et al. (2017)
<i>Funneliformis mosseae</i>	<i>Lycopersicon esculentum</i>	Cd	Improved plant growth, The distribution of Cd from the root to the shoot was impeded.; increased Cd tolerance	Li et al. (2020)

translocation of metal(loid) elements in maize. Cheng et al., (2021) reported synergistic effects between nZVI and AMF inoculation in terms of Pb and Cd immobilization, in spite of the limited colonization of AMF by nZVI. AMF potentially may facilitate Fe compounds formation on root surface by utilizing nZVI or its derived oxide (Wu et al. 2018), resulting in the precipitation of metals (Hansel et al. 2002). Further, AMF can accelerate the degradation and deterioration of nZVI, resulting in a more porous and less compact surface structure, which can in turn increase the ability of nZVI to adsorb pollutants and a larger surface area available for interaction with the contaminants (Wu et al. 2018).

Nanophytoremediation-based technologies are being increasingly recognized as an effective measure to deal with various environmental contaminants. However, the scope of these studies is confined mostly to the controlled laboratory settings, necessitating additional research to establish their efficacy in practical context such as for soil and wastewater treatment plants. Further, it is to be noted that some literatures have also highlighted the potential ecological impact on the biological functions of AMF-plant symbiotic systems (Moll et al. 2016; Tian et al. 2019). It has been indicated that laboratory engineered NP may have

a direct impact on AMF infection, colonization intensity, and bush richness, directly impacting its structural stability and compactness within mycorrhizal systems (Xu et al. 2019). In a study of Ag and titanium dioxide NP treatment on sunflower roots (*Helianthus annuus*), the rate of AMF infection, intensity of colonization, and abundance of *Glomus intracellularis* correlated negatively with the concentration of NP applied. Cao et al. (2017) reported remarkable decrease in the variety AMF under AgNPs stress, while Yang et al. (2021) showed disruptive effect of ZnONPs on the composition and diversity of AMF. On the contrary, a number of studies reported positive impact of NPs on AMF colonization, diversity in soil and promotion of physiological function (Feng et al. 2013; Moll et al. 2016; Watts-Williams et al. 2013). According to Gatahi et al. (2016), incorporation of SiO₂NPs in a biological control agent could enhance the colonization of AMF in the rhizosphere of tomato. Joseph et al. (2015) suggested improved P solubility and nutritional conditions and decomposition of organic matter in the rhizosphere of wheat plants in the presence of magnetic FeNPs. Another important concern is the development of potential toxicity in aqueous systems due to the formation of intermediate complexes from NP application. Hence, it is



imperative to monitor the origin and fate of these compounds at various stages of the treatment process, and to implement appropriate removal measures if required.

Challenges and future prospect

Nanomaterials and nanotechnology have recently achieved significant advances in remediation of HM pollution, though they pose considerable environmental and health concerns. Most recent *in vitro* and *in vivo* investigations showed that NP treatment decreased HM absorption by plants and bioavailable HM levels in polluted soil (Ahmed et al. 2021). Despite their numerous potential applications, NPs still face significant issues in environmental cleanup that need specific research focus. Figure 3 shows the environmental, technological, research and economic challenges and concerns that need further investigation in nano phytoremediation.

There is a paucity of data regarding nano-phytoremediation studies as predominantly microcosm experiments have been executed so far, therefore, more effective prolonged investigations are required to infer the actual effects or efficacies of NPs. Further studies on large-scale manufacture and industrial use of NPs are required to assess their cost-effectiveness, including NP supply, NP incorporation techniques, and long-term stability in real-world applications. The intricate chemical and biological environment surrounding the plant rhizosphere in nano-phytoremediation should be further investigated, as it influences the plant's uptake of heavy metals in the presence of nanomaterials and the potential synergistic or antagonistic

effects between microbial activity and nanoparticles. Moreover, the function of NPs produced and released by rhizospheric bacteria in nano-phytoremediation is still unknown, which must be taken into account when applying nano-phytoremediation.

Extensive testing is required to determine the actual impacts of NPs on various spheres of environment including biological and environmental safety. NPs may also accumulate in plant and microbial systems, infiltrating the food chain and endangering human health. Different NPs will have varying impacts on soil biodiversity and soil characteristics. Monitoring and evaluation of the impact and safety of applying NP in contaminated sites are required. Currently not much information is available on what happens to the materials once they have been incorporated into the environment to degrade or remediate contaminants to prevent them from becoming a cause of environmental contamination itself. The environmental toxicity of leached NPs, NP leakage, and environmental effects must also be taken into account for a sustainable nano-based phytoremediation application. Developing biodegradable and biocompatible NPs should be the focus of future studies.

Choosing the optimal NPs to remove a pollutant from an environment needs consideration of the type of pollutant, accessibility to the location, quantity of material required, feasibility of recovering/recycling the remediation nanomaterial. Future research should address NP dosage optimization and safe targeted administration to realize their environmental benefits. Although certain materials are recyclable, it seems that their efficiency diminishes with time, limiting their usefulness within a short span. Therefore, regeneration and reusability of NP are of great

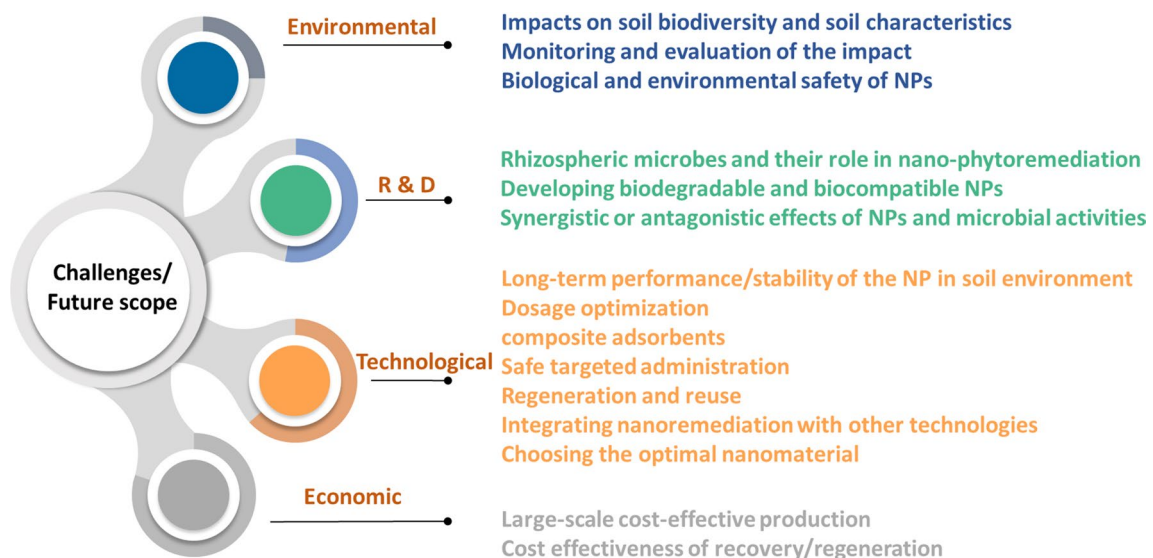


Fig. 3 The environmental, technological, research, and economic challenges and concerns that need further investigation in nano phytoremediation

importance to prevent these from becoming environmental contaminant itself.

Combining nanoremediation with other soil remediation technologies has been demonstrated to be a helpful soil remediation approach, as the synergetic effects may boost the sustainability of the applied process toward green soil remediation technology. Processing of the NPs could also be an attractive option before application to enhance the bioavailability or mobility of NP to plants. In this context, composite nano-based adsorbents are promising; however, physicochemical properties of the composite adsorbents such as hydrophilicity, porosity, thermal and mechanical stability, long-term performance, regeneration, and reuse are some of the key challenges that must be considered when developing new NPs for environmental remediation.

Conclusion

This study presents a comprehensive review on the prospects of use of NPs in HM remediation in soil. We have presented an overview of different NPs used in the context of environmental remediation, their functions, environmental issues, toxicity, and further, views on concerns and future prospects of NP-based HM cleanup were presented. We analyzed the benefits and challenges associated with the application each type of NP and particularly emphasized how arbuscular mycorrhizal fungi can influence the NP mediated soil heavy metal uptake. The review offers a theoretical foundation for the development of effective, efficient, and safe remediation technology for HM-contaminated soil, which is critical for furthering the theoretical and practical development of soil pollution control. To attain the optimum utilization potential of NPs for environmental applications, several obstacles must be addressed. Nonetheless, nanomaterial-based bioremediation technologies offer a plethora of opportunities for dealing with environmental degradation.

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Declarations

Conflict of interest The authors declare that they have no known competing interests.

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