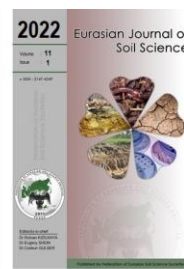




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A review on nanobioremediation approaches for restoration of contaminated soil

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Abstract

Nanotechnological approaches are emerging as one of the most contemporary restoration strategies that may be used to remove a variety of contaminants from the environment, including heavy metals, organic and inorganic pollutants. The application of nanoparticles (NPs) is entrenched with biological processes to boost up the removal of toxic compounds from contaminated soils. Many efforts have been taken to increase the effectiveness of phytoremediation such as the addition of chemical additives, application of rhizobacteria, and genetic engineering, etc. In this context, the integration of nanotechnology with bioremediation has introduced new dimensions to the reclamation methods. Thus, advanced remediation methods that combine nanotechnology with phytoremediation and bioremediation, where nano-scale process regulation aids in the absorption and breakdown of pollutants. NPs absorb/adsorb a variety of contaminants and also catalyze reactions by lowering the energy required for their breakdown due to unique surface properties. As a result, these nanobioremediation procedures decrease the accumulation of contaminants while simultaneously limiting their dispersal from one medium to another. Therefore, the present review is dealing with all the possibilities of the application of NPs for restoration of contaminated soils.

Keywords: Phytoremediation potential, phytoremediation strategy, NPs, contaminated soils, plants, microorganisms.

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Introduction

Soil is the essence of agriculture and it is enriched with vital macro and micronutrients that promote healthy growth of the crops that ultimately impart health benefits to humans (Joshi et al., 2020). Several anthropogenic activities contaminate the soil with a load of synthetic organic compounds, heavy metals, agrochemicals, and an excess of nutrients as well (Minkina et al., 2019; Ghazaryan et al., 2020). Similarly,

industrialization/urbanization is adding solid wastes, chemicals and solvents, and other persistent organic and inorganic materials to different environmental matrices (Midhat et al., 2019).

Advancement in nanotechnology and nanoscience provide new directions to research and development in almost every field of science. It is an expanding research field that involves structures, devices, and systems with unique properties owing to the arrangement of their atoms at the nanoscale (1–100 nm) (Bayda et al., 2019; Rajput et al., 2020b; 2021b). In recent decades, nanotechnology has been used in a range of contexts, notably medicine, textiles, pharmaceuticals, electronics, optics, cosmetics, sports, and many others. The application of NPs in agriculture was accepted at the beginning of the twenty-first century (Fraceto et al., 2016), and more than 232 products are available for various agricultural uses (Rajput et al., 2021a). Also, it has not remained static in the field of environmental restoration (Guerra et al., 2018; Singh et al., 2020).

Recently, nanobioremediation (NBR) is declared as a technology for cleaning up environmental contamination by accelerating natural biodegradation processes using NPs. NBR is defined as a process that uses NPs with microorganisms, or plants to eradicate hazardous contaminants from the soils (Cecchin et al., 2017). Following that, distinct NBR procedures are defined based on the type of organism used for contaminants remediation (i.e., nanophytoremediation, and microbial nanoremediation (Burachevskaya et al., 2020; Rajput et al., 2020a,c; Singh et al., 2020; Kumari et al., 2021). The intensification in the expenses of chemical as well as physical processes, microbes- and plant-mediated NBR technologies are receiving more attention.

Coming to the benefits of NBR, there is a multitude of reasons why nanotechnology is integrated with bioremediation. For example, NPs have a large surface area per unit mass, which means that a greater number of particles can come into contact with the environment, boosting the remediation process (Fernández-Luqueño et al., 2018; Kaur et al., 2018). Thus, NBR efforts to minimize pollutant concentrations to risk-based thresholds while also decreasing secondary environmental impacts. Furthermore, this method of reclamation also combines the advantages of nanotechnology and bioremediation to create a remediation process that is more efficient, faster, and environmentally benign than the individual methods (Patil et al., 2016; Kumar et al., 2021).

However, every advance of the process of remediation has particular explicit merits as well as demerits that need to be taken into consideration for each location. In a nutshell, after the extensive literature survey, it can be concluded that integration of bioremediation with nanotechnology appears to be a feasible alternative to conventional remediation technologies either in sequence or in parallel to them. However, there are still more studies and development measures necessary to bring these types of sustainable technology to the market for full implementation.

Recent advances in bioremediation of polluted soil

Chemical and physical remediation, incineration, and bioremediation are some of the NBR technologies that are currently in use. With recent advances, NBR provides an environmentally friendly and economically viable option for removing contaminants (Patra Shahi et al., 2021). The fundamental principle behind the NBR is depicted as the degradation of organic wastes employing nano-catalysts as a medium that allows them to enter deep within contaminants, thereby executes the whole process safely without modulating the environment (Rizwan et al., 2014; Cecchin et al., 2017; Chauhan et al., 2020). The overview of NBR is presented in figure 1.

As bioremediation relies on live species to clean up contaminated environments, thereby a good relationship between NPs and living organisms is critical for the efficacy of this phenomenal technique (Sangwan and Dukare, 2018; Paterlini et al., 2021). In this context, it is documented that the physical and chemical interactions between NPs, biota, and contaminants are influenced by numerous factors viz., NPs' size and shape, surface coating, and chemical nature. Plus, the nature of contaminants, the type of organism used, the media, pH, and temperature are also recorded to impact the process considerably (Ibrahim et al., 2016; Tan et al., 2018).

These events grow complicated due to the large number of potential parameters that have a direct or indirect influence on such interactions. For example, temperature and pH of media are reported as important factors for the optimal development of biological organisms (Patra and Baek, 2014). Now pinpointing the different actions, such as dissolution, absorption, and biotransformation may occur when NPs and biota interact (Kranjc and Drobne, 2019; Vázquez-Núñez et al., 2020). On the other hand, interactions of NPs and biota can be toxic or stimulating which results in a biocidal or bio-stimulant effect,

thus the performance of organisms involved in the NBR process could be impacted (Juárez-Maldonado et al., 2019).

Some of the most important NPs used in NBR are nano-iron, nanosized dendrimers, carbon nanotubes, single enzyme NPs, engineered NPs, etc. (Kaur et al., 2018; Patra Shahi et al., 2021). In the NBR technique, the contaminants are first broken down by NPs to a level that is conducive to biodegradation, and then the contaminants are biodegraded. The main advantages of bioremediation over conventional strategies are high competency, reduced generation of chemical and biological wastes, selectivity, no additional nutrient requirements, bio-sorbent regeneration, the probability of metal recovery, etc. (Juwarkar et al., 2010; Rizwan et al., 2014; Chauhan et al., 2020).

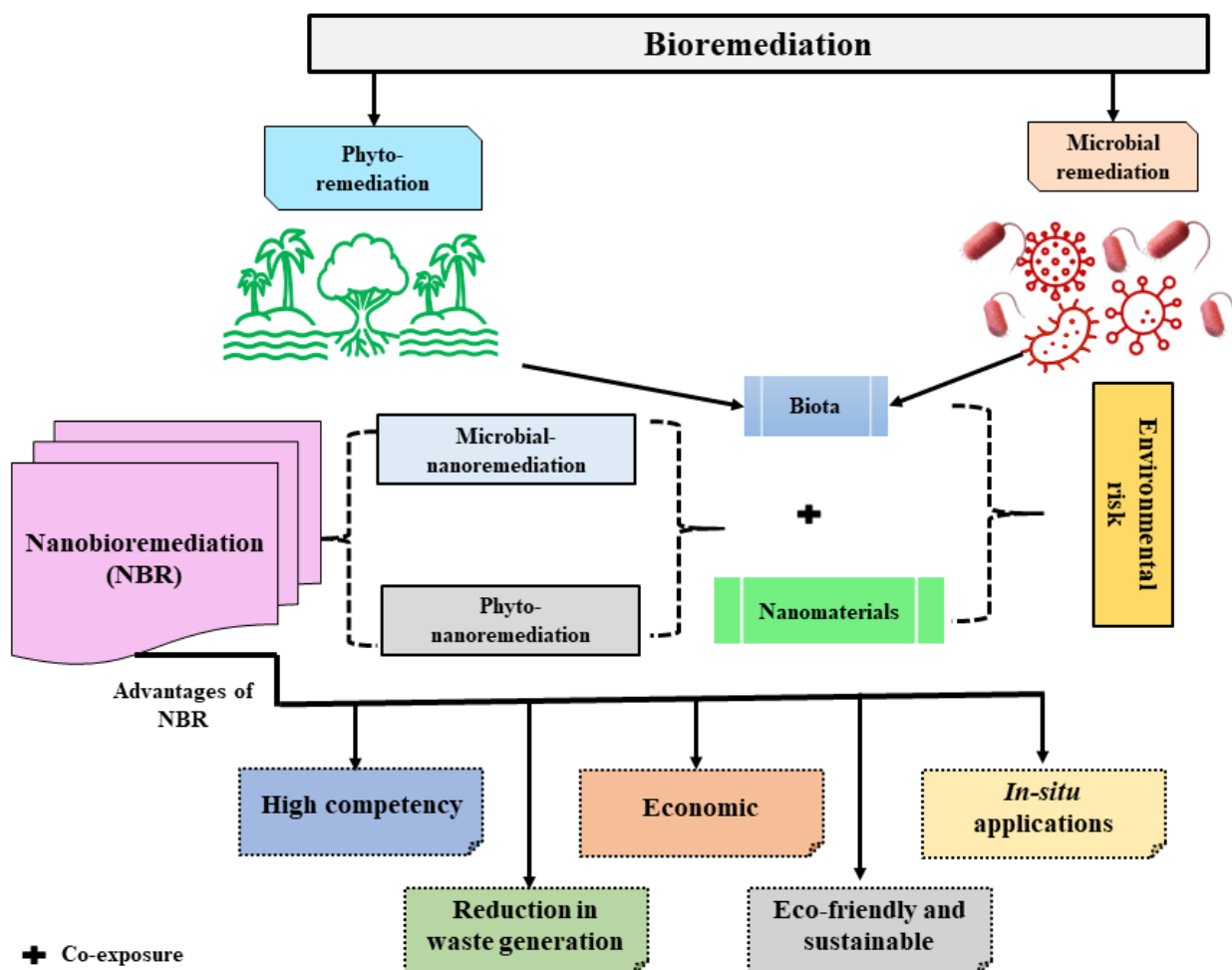


Figure 1. The pictorial representation of nanobioremediation and its types along with environmental risks

On top of it, most currently available conventional remediation procedures are based on the classic *ex-situ* strategy, which entails excavating contaminated material and then treating them with conventional means. Plus, some of these processes are energy-intensive, which makes them expensive, and they may also leave concentrated hazardous waste residues that require additional treatment and disposal (Wuana and Okiyeimen, 2011; Chauhan et al., 2020). On the flip side, *in-situ* remediation methods benefit greatly from the peculiar characteristics of NPs (Kumari et al., 2021; Rajput et al., 2021b). Thus, *in-situ* NBR can annihilate the need for draining out of groundwater and transportation of contaminated soils to treatment and disposal sites.

Nanophytoremediation of polluted soils

Nano-encapsulated enzymes also have greater potential in treating some complex organic pollutants, for example, persistent pesticides (organochlorines) and long-chain hydrocarbons are hard to degrade by microbial or plant remediation process (Chauhan et al., 2020). Few successful field applications of NPs have been done in past for the bioremediation of soils.

Heavy metals

Heavy metal pollution of arable soils is an increasing problem, as it poses a serious threat to food safety, public health, and the food chain and ecosystem. For *in-situ* treatment of polluted soils, phytoremediation is

documented as a favored and cost-effective method by researchers (Liang et al., 2017). Phytoremediation of soils polluted with cadmium, chromium, lead, nickel, and zinc was improved with the introduction of NPs, according to several studies (Wani et al., 2017; Ekta and Modi, 2018; Kanwar et al., 2020). It is well-established that exposure to heavy metal pollutants has major health risks to the well-being of humans (Rajput et al., 2020d; Zamani et al., 2020).

Phytoextraction is the most familiar method adopted to eliminate heavy metals from polluted soil (Ali et al., 2013). The application of NPs to enhance the phytoextraction efficiency has been a successful strategy towards nanophytoremediation (Ebbs and Kochian, 1998; Ghazaryan et al., 2018; Ghazaryan et al., 2019). Iron NPs are used as a strong reductant for those pollutants that require a reduction process for degradation (Sun et al., 2006), whereas, zerovalent iron (nZVI) has great potential in phytoremediation of a range of pollutants as it is a highly reactive reducing agent. Plants treated with the lower concentrations (100- 500 mg/kg) of nZVI have exhibited the maximum accumulation (1175.40 g per pot with 100 mg/kg of nZVI). Whereas, a higher dose of nZVI NPs (500-1000 mg/kg) caused oxidative stress in *Lolium perenne* thereby reducing the uptake of Pb (Huang et al., 2018). Another study reports the similar characteristics of nZVI of concentration 100-500 mg/kg that improves Pb uptake up to 857.18 µg per pot (at 500 mg/kg) in the ragweed (*Kochia scoparia* also known as *Bassia scoparia*).

The TiO₂ NPs of 100, 200, and 300 mg/kg spiked in soil have shown Cd accumulation in *Glycine max* by 1.9, 2.1, and 2.6 folds in the shoots and 2.5, 2.6, and 3.3 folds in roots, respectively. However, 1534.7 mg/kg per pot of Cd was reported to be the maximum accumulation (Singh and Lee, 2016). The inoculation of *Acaulospora mellea* considerably enabled the immobilization of heavy metals. The acceptable concentration of nZVI was 50 mg/kg to 1000 mg/kg (Cheng et al., 2021).

The concentration of nZVI at 100, 500, and 1000 of mg/kg showed effective uptake of Cd in the *Boehmeria nivea* L. root, stem, and leaves by 16–50%, 29–52%, and 31–73%, respectively (Gong et al., 2017). Arbuscular mycorrhizal (AM) fungi, *A. mellea* along with nZVI have shown uptake of Cd, Pb, and Zn from the acidic soil by *Sorghum bicolor* L. A table has been appended below that exhibits the role of NPs in the phytoremediation of heavy metals (Table 1).

Organic pollutants

Organic pollutants are a major threat to agricultural soil, food chain, ecosystem, and human health. They are majorly released from industrial operations and agricultural applications (Alharbi et al., 2018). Phenols, polycyclic aromatic hydrocarbons (PAHs), organochlorine insecticides, and polychlorinated biphenyls (PCBs) are all examples of cyclic organic compounds that are documented as persistent organic pollutants (Sushkova et al., 2016, 2018). Many of them are lipophilic, thereby they tend to get bioaccumulated and biomagnified in adipose tissues of several organisms in the food chains of aquatic and terrestrial ecosystems (Penell et al., 2014).

Hence, phytoremediation is always considered as a cost-effective and sustainable approach to remediate these organic pollutants (Kang et al., 2018). Application of NPs in phytoremediation of organic pollutants like trichloroethylene, endosulfan, and trinitrotoluene have been reported in the past (Pillai and Kottekkottil, 2016). Fullerene NPs have been reported to enhance the uptake of trichloroethylene using *Populus deltoides*, 2 and 15 mg/L of fullerene NPs have enhanced the uptake by 26% and 82%. *Plantago major* with the appropriate adsorbent (activated charcoal) and solubilizing agent, SiO₂ as green synthesized NPs of Fe and Ag namely *Ficus*-FeNPs (F-FeO) (size 2.46 nm–11.49 nm), *Ipomoea*-Ag (Ip-Ag₀) (size 6.27 to 21.23 nm) and *Brassica*-AgNPs (Br-Ag₀) (size 6.05 to 15.02 nm) were able to remove 93.7%, 91.30%, and 92.92%, respectively of chlorfenapyr (Romeh and Saber, 2020). Studies have also reported that pollutants like chlorpyrifos, molinate and, atrazine could be removed and broken by nanosized zerovalent iron.

Plants that absorb contaminants in their tissues to breakdown and detoxify from the environment are used in nanophytoremediation. Plants that are favored for phytoremediation purposes should have characteristics such as:

- Fast grower with higher biomass producer
- The highly branched and well-developed root system
- Potential to tolerate and accumulate pollutants
- Higher sink potential that allows hyperaccumulation
- Easy harvesting of plant's sink organs
- Genetic manipulation should be easier, and
- It should be non-consumable by humans

Table 1. Role of engineered NPs in the phytoremediation of Pb and Cd from the soil

Pollution	Applied NPs	Mode of action of NPs	Plant name	Remarks	Reference
Pb	nZVI	Lower concentration of nZVI promoted plant growth	<i>Kochia scoparia</i>	100-500 mg/kg of nZVI enabled 857.18 µg per pot of Pb accumulation in the plant	Zand and Tabrizi, 2020
	Nanohydroxyapatite	Promoted plant growth through phosphate mobilization in the soil	<i>L. perenne</i>	With nano-hydroxyapatite the concentration of Pb in the root was reduced by 2.86- 21.1% and in the shoots 13.19-20.3% reduction of Pb was observed	Ding et al., 2017
	nZVI	Lower concentration of nZVI promoted plant growth	<i>L. perenne</i>	Accumulated maximum concentrations of Pb in the root and shoot of the plants	Huang et al., 2018
	Nanohydroxyapatite	Reduced phytotoxicity, and enhance plant growth	<i>L. perenne</i>	The 21.97% remediation efficiency was observed within 6 weeks	Jin et al., 2016
Cd	TiO ₂	Improved germination and photosynthetic capacity of the plant	<i>Glycine max</i>	Concentration (100 to 300 mg/kg) dependent increase in the uptake of Cd was observed (128.5 µg -507.6 µg of Cd per plant)	Singh and Lee, 2016
	nZVI	Promoted plant growth	<i>Boehmeria nivea</i>	Increase in the accumulation of Cd in the leaves by 31-73%, stems by 29-52%, and roots by 16-50% were recorded	Gong et al., 2017
Pb, Cd	nZVI	Promoted plant growth	<i>Sorghum bicolor</i>	Enhanced uptake of Pb and Cd of the concentration 50 mg/kg to 1000 mg/kg	Cheng et al., 2021
Pb, As	CNT with biochar	Reduced seed germination; however, toxicity was modulated by biochar	<i>Brassica rapa</i>	Pb was reduced by 1.2–3.8-folds and significantly reduced As accumulation in the soil	Awad et al., 2019
As, Cd, Pb, Zn	nZVI	Stabilized HMs	<i>Helianthus annuus</i> , <i>L. perenne</i>	Reduced up to 60% uptake of As, Cd, Pb, and Zn in roots and shoots compared to the control plants	Vítková et al., 2018
Cd, Pb	Nano-silica	Improved the availability of Pb and Cd to the plants, and also promoted the growth of the plant	<i>Secale montanum</i>	Accumulation of Pb in the roots was achieved up to 533.6 mg/kg DW and Cd up to 208.6 mg/ kg DW.	Moameri and Khalaki, 2019

Microbial nanoremediation

Microbes-mediated nanoremediation, a novel and efficient approach, involved the cellular enzymes secreted by microorganisms that successfully degraded and cleaned up the broad variety of organic pollutants in the contaminated ecosystem (Sangwan and Dukare, 2018; Torimiro et al., 2021). Numerous environmental conditions limit and influence the efficiency with which pollutants are degraded by microbes in contaminated soils. Within a microbial association, the biological response to environmental pollutants is differed, and the presence of co-contaminants may bring out changeable reactions to the bioremediation process (Sangwan and Dukare, 2018; Rajput et al., 2021c; Shende et al., 2021). Despite this, NBR offers a proficient and lucrative approach for contaminated soil and waste or groundwater treatment.

Microbes-mediated nanoremediation of xenobiotics is a fundamental environment-friendly approach to eradicate persistent toxic compounds gathered in the surroundings. The capacity of microbes to metabolize, transform, as well as degrade, xenobiotic compounds has been documented as a competent approach to remove dangerous and toxic wastes (Agarry and Solomon, 2008). Microorganisms are preferably appropriate to remove pollutants due to the enzyme system present that allocates them to utilize ecologically noxious pollutants as their energy and food source. The progressions in bioremediation science have been accredited to the individual as well as interdisciplinary contribution afforded by scientific areas of analytical chemistry, microbiology, biochemistry, molecular biology, environmental engineering, and very recently, nanobiotechnology (Hu et al., 2014; Sangwan and Dukare, 2018; Singh et al., 2020). The process of bioremediation includes mineralization and detoxification, in which the transformation of waste into inorganic compounds, like water, methane, and carbon dioxide has been carried out (Liu et al., 2018a;

Vázquez-Núñez et al., 2020; Paterlini et al., 2021). Microbes can alter almost all organic materials, with catalytic mechanisms and wider diversity (Paul et al., 2005). They can function still in anaerobic plus extreme environmental conditions, which constructs them a smart candidate for the process of bioremediation.

Additionally, microorganisms play a significant function in biogeochemical cycles as well as the ecosystems' sustainability. The conversion of xenobiotic contaminants by microbes may occur either in an anoxygenic or oxygenic environment. Nevertheless, molecular oxygen contributes to aliphatic as well as aromatic xenobiotic compounds (Cao et al., 2009; Sinha et al., 2009). Amid the different microorganisms, bacteria have been established as the most competent and prevailing in the natural bioremediation processes. In both the conditions, i.e., aerobic as well as anaerobic, bacteria have developed an approach for acquiring energy from nearly every compound by electron acceptors like ferric ions, sulfate, nitrate, etc. Several genera of bacteria, e.g., *Alcaligenes*, *Acinetobacter*, *Bacillus*, *Escherichia*, *Gordonia*, *Moraxella*, *Micrococcus*, *Pseudomonas*, *Pandoraea*, *Rhodococcus*, *Streptomyces*, and *Sphingobium* either independently or in amalgamation are implicated in oxygenic breakdown. In contrast, bacterial genera concerned with the anaerobic degradation of xenobiotics include *Azoarcus*, *Clostridium*, *Desulfotomaculum*, *Desulfovibrio*, *Geobacter*, *Methanococcus*, *Methanosaeta*, *Pelotomaculum*, *Syntrophobacter*, *Syntrophus*, and *Thauera* (Jindrova et al., 2002; Van Hamme Jonathan et al., 2003; Kulkarni and Chaudhari, 2007; Weelink et al., 2010; Sangwan and Dukare, 2018)

The remediation of extremely persistent and xenobiotic water and soil contaminants, such as hydrocarbons, heavy metals, dye in textile (acid dyes, cationic dyes, azo dyes), pharmaceutical constituents (antibiotics and antiseptics), and other such contaminants are critical for wastewater and soil treatment and its future application. These contaminants increase pollution and pessimistically affect the environment (Koul et al., 2021; Sushkova et al., 2016).

Since NPs have a larger surface area and are smaller, they can act as catalysts or adsorb contaminants above a larger surface area. Numerous reports documented the catalytic properties of various NPs together with the biological components have been assessed to reduce harmful pollutants (Zhao et al., 1998; Kharissova et al., 2013). Many microorganisms have been utilized to hone NPs exploitation for the NBR process as several researchers reported encouraging outputs in the application of microbe-mediated NPs in the process of remediation.

An extensive recognition of microbes for this scientific approach was recognized owing to their exceptional chemical, physical, biological, as well as optical properties like super-hydrophobic and filtering nature, sensitive affinity membranes, modifiable functionality, as well as a higher surface-to-volume ratio (Sarwar et al., 2017; Wang et al., 2015; Sangwan and Dukare, 2018).

A detailed description of microbes-mediated nanoremediation has been given in the forthcoming sections.

Hydrocarbons

Many researchers have been reported that the microbes-mediated nanoremediation of persistent organic pollutants; i.e., hydrocarbon. It was reported the electrostatic interaction of magnetic NPs functionalized by *Rhodococcus erythropolis* harnessing system that substantively bio-desulfurize hydrocarbon component dibenzothiophene (DBT) by 56%. Thus, validating the advantage of magnetic NPs functionalized by *R. erythropolis* above the solitary exploitation of every component for bioremediation (Ansari et al., 2009). The efficient synergistic effect of the nZVI with *Sphingomonas* sp. as an effectual twosome towards the de-bromination and gradual polybrominated diphenyl ethers (PBDEs) degradation in aqueous solution (Kim et al., 2012). Alternatively, the feasibility of the combined employ of bimetallic (Pd/nFe) NPs and *Sphingomonas wittichii* for the NBR 2,3,7,8-tetrachlorodibenzo-p-dioxin hydrocarbon was also recognized (Bokare et al., 2012). The active dechlorination facilitated by integrated hybrid (nano-bioredox) resulted to form dibenzo-p-dioxin.

A study has revealed the applications of *Sphingomonas* sp. as a bio-functionalized tool for carboxymethyl cellulose (CMC) stabilized bimetallic (Pd/Fe) NPs (Singh et al., 2013). The nano-composite was found to be triumphant for the deprivation of gamma-hexachlorocyclohexane (γ -HCH), generally identified as lindane and the main component in cosmetics (Singh et al., 2013). The study was performed to remove Aroclor 1248- a congener of PCBs, where the noteworthy de-chlorination, as well as conversion of the contaminant, was observed by the treatment of bimetallic (Pd/Fe) NPs under anoxic surrounding resulted in the formation of biphenyls (Le et al., 2015). Progressive bioremediation of the resulting biphenyls further catalytically decreased the persistent Aroclor 1248 from $33.8 \times 10^{-5} \mu\text{g/g}$ to $9.5 \times 10^{-5} \mu\text{g/g}$ with *Burkholderia*

xenovorans (Le et al., 2015). The silica NPs biofunctionalized with lipid bilayers of *Pseudomonas aeruginosa* was investigated to clean up PAH (benzo[a]pyrene) (Wang et al., 2015). The 1,2-dimyristoylsn-glycero-3-phosphocholine, lipid molecule playing a dynamic role, to improve the sequestration or adsorption of the PAHs, when conjugated by silica NPs. The biofunctionalized graphene oxide NPs with laccase enzyme developed by *Trametes versicolor* were studied for their potential as well as combine enhance for the biodegradation of PAH (anthracene) (Patil et al., 2016). The amalgamation of laccase enzyme from fungi as conjugant was reported to have the enhanced ability of degradation than their single application and also extended their stability. The polymer (polyallylamine hydrochloride)-layered magnetic NPs functionalized by *Alcanivorax borkumensis* established an opportunity for vigorous hydrocarbon degradation (Konnova et al., 2016). Exceptional features like forming the neutral lipid inclusions in biofilms of *A. borkumensis*, the biosurfactant micelle ascertain the opportunity of hydrocarbon decomposition.

Bacillus licheniformis-mediated nanoremediation process was evaluated bio-functionalization of $Zn_5OH_8Cl_2$ modified Fe_2O_3 NPs with *B. licheniformis* to break crude oil into naturally degradable compounds. Additionally, demonstrate some prospects on the promising improvement of microbial bio-surfactants for efficient NBR of widespread oil pollution (El-Sheshtawy and Ahmed, 2017). The synergistic effect concerning iron oxide NPs and *Alkaligenes faecalis* improved the crude oil biodegradation in the contaminated environment (Oyewole et al., 2019). The authors observed that assessing variable deliberations of *A. faecalis* with iron oxide NPs, at 200 mg efficiently cleans up crude oil pollution.

Heavy metals

Microbes-mediated nanoremediation of heavy metals corroborates the potential of microorganisms in cleaning up the environment. NPs' effectiveness in bioremediation was accomplished during the *in-situ* fabrication of palladium (Pd) NPs from Pd (II) ions intervened by *Clostridium pasteurianum* acquired from sandy aquifer matter. The biosynthesized Pd NPs evidenced positive remediation in the alteration of hexavalent chromium; i.e., Cr (VI) into insoluble Cr (III) and, therefore, leading to the production of hydrogen gas (Chidambaram et al., 2010). In this study, the removal rate of Cr (VI) was considerably improved, reaching 7.2 g, indicating the importance of nano-catalysts over traditional *in-situ* bio-simulation techniques. A comparable strategy accomplished was channeled towards reduction of Cr (VI) by sodium alginate, polyvinyl alcohol (PVA), as well as a matrix of carbon nanotubes (CNTS) immobilized upon *Pseudomonas aeruginosa* cells (Pang et al., 2011). The biogenic Cr (VI) reduction to soluble Cr (III) was shown in wastewater by the immobilized bacterial cells (Nancharaiyah et al., 2010).

In the NBR process, algae also have revealed their significance. Iron NPs fabrication by *Chlorococcum sp.* demonstrated a noticeable elimination of Cr (VI) to Cr (III) about 92% of 4 mg/L (Subramaniyam et al., 2015). Iron NPs synthesized from algae was mediated with the biomolecules from algal cell illustrated more excellent stability, high reactivity, and proficient toxic pollutants reduction in the environment. On the other hand, the biogenic role of *Lysinibacillus sphaericus* in the production of magnetic oxide NPs intended to remove Cr (VI) contamination from the surroundings (Kumar et al., 2019). The authors reported the employ of exopolysaccharides (EPS) matrix of biofilm derived from *L. sphaericus* as a superior reducing, capping, and stabilizing agent, acquiring several binding sites for different metal ions. Magnetic oxide NPs functionalized with EPS illustrated the improved potential to adsorb Cr (VI). In another study, it was reported the integration of *Chlorella vulgaris* in ultrafine bi-metallic i.e., TiO_2/Ag chitosan nanofiber mats, as a functionalized agent, elucidated the significance of algae in the photo-removal strategy of Cr (VI) under UV light irradiation (Wang et al., 2017a). The discharge of organic substances such as chlorophylls, carboxylic acids, etc., through *C. vulgaris*, was documented to have an improved photocatalytic reduction of Cr (VI) on the TiO_2/Ag chitosan nanofiber mats, confirming the synergistic way of hybrid NPs by algae and TiO_2/Ag .

The fabrication of lead sulfide i.e., PbS NPs from *Rhodospiridium diobovatum* demonstrating the prospect of a straightforward breaking down of Pb(II) ions into less toxic and helpful forms by fungi (Seshadri et al., 2011). The triumphant elimination of Cd in Cd-polluted water illustrated the competence of *Pseudomonas aeruginosa* improved Cd bioreduction which in turn hasten the cadmium sulfide (CdS) NPs biosynthesis (Raj et al., 2016). Likewise, the removal and bioremediation of Cd from Cd-polluted soils also evaluated (Liu et al., 2018b). The authors demonstrated that the co-treatment of *Bacillus subtilis* and nano-hydroxyapatite (NHAP) efficiently eliminated the Cd contamination, encouraging the propagation of microbial community of rhizosphere along with the diversity of bacteria in the remediated soil (Liu et al., 2018b).

The evaluation of somewhat variable biofunctionalized approach including polyvinylpyrrolidone (PVP)-coated iron oxide NPs intermingled with *Halomonas sp.* isolated from the oil-contaminated soil, has been reported (Alabresm et al., 2018). Selenium NPs were found efficient in NBR of mercury polluted soil; those

NPs were formed by the occurrence of *Citrobacter freundii* (Wang et al., 2017c). The alteration of elemental form of (Hg⁰) to the insoluble form mercuric selenide (HgSe) with biogenic selenium NPs evaluated under aerobic as well as anaerobic conditions accounted for a bioremediation value 39.1-48.6% and 45.8-57.1%, respectively. The nickel compound was removed in the effluent by introducing *Microbacterium* sp. resulting in the production and recovery of nickel oxide NPs (Sathyavathi et al., 2014). In another study, the potential of *Hypocrea lixii* was discovered to reduce noxious metals, specifically nickel, in contaminants and devising the nickel oxide NPs biosynthesis from the waste for further applications (Salvadori et al., 2015).

Recently, it was demonstrated that the silver (Ag) NPs synthesized through greener way assisted by *Bacillus cereus* was supported with alumina, found efficient in NBR of pharmaceutical effluents restraining heavy metals, mostly chromium (Cr) and lead (Pb) (Kumari and Tripathi, 2020). The bacterial cell-mediated nano-adsorbent method certified to remove about 98.13% (Cr) and 98.76% (Pb) that were discharged from pharmaceutical industries as waste effluents. The possibility of nanobioremediation of cadmium (Cd) and lead (Pb) in the soil by the mutual exploitation of *Escherichia coli* along with metal NPs towards the elimination of these heavy metals (Zhu et al., 2020).

Pharmaceutical ingredients

The recurrent emancipation of pharmaceutical ingredients (antiseptics and antibiotics) in wastewater is considered a serious concern. These mainly originate from domestic and industrial effluents, which have polluted not only the environment but also enhanced the appearance of antibiotic-resistant microbes in wastewater (Adesoji et al., 2020). Nevertheless, the prospect of eliminating these pharmaceutical ingredients by the NBR strategy was evaluated as per the many research studies. The biosynthesis of both Au and Ag NPs using *Turbinaria conoides*, an alga which was found useful as an antimicrofouling agent (Vijayan et al., 2014). Hydrogen peroxide, a common pharmaceutical ingredient, yet a pollutant of the environment, was proficiently removed from waste effluents from industries by the electrocatalytic reduction of the compound aided with Pd NPs synthesized *Sargassum bovinum* (Momeni and Nabipour, 2015).

Micro-accumulation of triclosan, which has been found to be linked with cancer, has frequently been used as an antibacterial and antiseptic agent. Nevertheless, the significance of fungi (*Trametes versicolor*) as an essential biofunctionalized agent for bimetallic (Pd/Fe) NPs to remove triclosan in liquid effluents was established (Bokare et al., 2010). In this work, *T. versicolor* was observed to secrete laccase enzyme that was found to play a vital role in the two-step redox strategy, which involved the anaerobic dechlorination as well as sequential oxidation of 2-phenoxy phenol. Similarly, (Adikesavan and Nilanjana, 2016) described the magnesium oxide (MgO) NPs biofunctionalization by yeast (*Candida* sp.). The myco-nano approach was found to have hastened the process of Cefdinir degradation and treatment in an aqueous environment. A group of bacteria conquered by *Bacillus* and *Pseudomonas* spp. accountable for the biosynthesis of manganese oxide (MnO) NPs was found to efficiently eradicate 1,2,4-triazole from wastewater (Wu et al., 2017). This study established the prospective of biogenic manganese oxide NPs to remove a variety of recalcitrant pollutants from bio-treated chemical industrial wastewater.

The efficiency of Pt and Pd NPs biosynthesized from *Desulfovibrio vulgaris* to remove effluents containing pharmaceutical compounds was reported. The numerous chemical compounds contribute greatly to the pharmaceutical industry. Likewise, 1,2,4-triazole used in different clinical applications because of a large number of compounds of the ring system. Besides, 1,2,4-triazole is also applied in the production of pesticides that often contributes to groundwater pollution during leaching (Martins et al., 2017). Similarly, picric acid (2,4,6-trinitrophenol (TNP)), is a valuable constituent in the production of antiseptic, posing hazard to the environment as a pollutant in an aqueous solution. The study established the progressive application of *Pseudomonas aeruginosa* mediated Fe₃O₄ NPs as a portion of multiwalled carbon nanotubes (MWCNT) to produce nanocomposite moderately employed for NBR of picric acid (Yousefi et al., 2020).

Dyes in textile

Dyes have been widely recognized as an essential component in a multitude of sectors, including cosmetics and textiles. Nevertheless, it is disposed-off mainly as liquid waste matter into the surroundings, which is poisonous to living beings (Asaduzzaman et al., 2016). A study ascertained the coalesce effect of biofunctionalized Ag NPs by *Chromobacterium violaceum* as a biosorption strategy to remediate washing water employed to process cotton fabrics (Durán et al., 2010; Duran et al., 2017). This process demonstrated the successful removal of organic compounds as well as dyes used in the production of fabrics. This treatment further illustrated its effectiveness for eliminating used Ag NPs and the revival of bacteria, posing lesser harm to the environment. The application of Ag NPs synthesized from *Bacillus pumilis* have been used

to remediate the Congo red dye from wastewater, which was applied on cotton fabrics (Modi et al., 2015). The goal was to develop and implement an efficient method for removing Congo red dye because it is less resistant to light and washing. The highest revival of Ag NPs leached in the effluents to evade harm to the environment.

In another study, it was observed that Ag NPs competently decolorize the organic dyes during the catalytic activity and confirmed that NPs might be employed as catalysts in industries to degrade organic dyes with higher competence (Sharma et al., 2015). It has been reported that Ag and Au NPs demonstrated good catalytic activity in the removal of organic dyes. These NPs reduced the time requisite for eliminating dye while also competently improved the rate of reaction (Suwith and Philip, 2014). The Au NPs could also be employed as adsorbents for organic dyes. As Au NPs, comprising surface proteins produced from fungus *Cladosporium oxysporum* AJP03, efficiently enhanced the rhodamine-B organic dye adsorption (Bhargava et al., 2016). The roles of different NPs and nanocomposites such as TiO₂ NPs, FeNPs, magnetic NPs, bimetallic NPs, nanotubes, nanoclays, and nanosponges in the NBR of soil are also revealed (Koul and Taak, 2018). The authors accentuated that the synthesis of NPs by green methods might be an efficient approach for treating water and soil pollution. The efficient catalysis of Congo red dye by Ag NPs synthesized from green alga *Caulerpa serrulate* was reported (Aboelfetoh et al., 2017).

Even though methyl orange dye is infrequently employed in textile because of its susceptibility to acids, they still find expediency as a dye for wool fabrics, a type of contaminant in wastewater. Mechanism of NBR evaluated the consortium of *Cellulosimicrobium* sp., *Micrococcus lylae*, and *Micrococcus aloeverae* to produce TiO₂ NPs (Fulekar et al., 2018). The active degradation of methyl orange dye was achieved in a reactor by the influence of UV light. These rhizospheric root-associated microorganisms demonstrated the opportunity and efficiency of normal sources for the biosynthesis of NPs and around ~99 % of methyl orange dye photocatalytic degradation, a signal for the significance of photocatalytic process for a safe environmental and passable nanobioremediation system. A comparable discovery was recognized for algae *Hypnea musciformis* [wulfen] J.V Lamouroux-mediated synthesis of Ag NPs and their dynamic efficacy in humiliating methyl orange dye solution under visible light (Ganapathy Selvam and Sivakumar, 2015). An effort on the Azo dyes bio-reduction, which are imperative synthetic colorants are generally used in textile, paper manufacturing, printing, etc. was conceded out by Pd NPs fabricated from *Klebsiella oxytoca* (Wang et al., 2018). The synthetic organic colorants were effectively bio-reduced with recovery from the effluent liquids. The biosynthesized polysulfone nanofibrous web and *Chlamydomonas reinhardtii* were originated a synergistic effect that removes reactive dyes from wastewater (San Keskin et al., 2015).

The Ag NPs synthesized from microalgae *Caulerpa racemosa* and *Chlorella pyrenoidosa* were reported for the photo-catalytic degradation of methylene blue and the treatment of liquid effluent containing hazardous dye produced significant results i.e., dropping the level of the contaminants under controlled experimental conditions (Aziz et al., 2015; Edison et al., 2016). In recent work, the descriptive information on various approaches for the NPs synthesis using microbial cells; their applications in agriculture, bioremediation, diagnostics, and medicine; and their prospects are provided (Koul et al., 2021).

Other toxic chemicals

Besides these major groups of pollutants, there are found some other toxic chemicals in the environment. The biogenic synthesis of manganese oxide NPs by *Pseudomonas putida* documented the bacteria potential for sufficient removal of organic micropollutants (Furgal et al., 2015). Bisphenol A (BPA), generally known as an essential chemical substance exploited in the industries for developing resins and plastics, requisite for storage of food and beverages, has become an aggravation to the ecology. The elimination of bisphenol A by a route focused on applying MnO NPs biosynthesized from algae (*Desmodesmus* sp.) (Wang et al., 2017b). Commercially produced nitro compounds for solvents or chemical intermediates create a relatively extensive volume in effluents from industries (Torimiro et al., 2021).

The application of *Chlorella vulgaris* on nitrate removal from liquid effluents, in which algae played a dual role in biogenic production of Pd NPs and its immobilization on nanofibre mats prepared by an electrospun method that improves the catalytic activity of the complex to remove nitrate from liquid effluents was demonstrated (Eroglu et al., 2013). NBR mechanism evaluated in *Sargassum tenerrimum* and *Tubinaria conoides* for the biological production of Au NPs applied to reduce the nitro compounds in wastewater (Ramakrishna et al., 2016).

Environmental concerns and future perspective

Environmental contamination is a serious issue that humanity is currently struggling with (Litvinov et al., 2017; Sushkova et al., 2017; Rajput et al., 2017b,2018). Numbers of techniques are being used and some others are under trial for the remediation of contaminants of the environment (Song et al., 2019; Baig et al., 2021; Kumar et al., 2021; Kumari et al., 2021; Paterlini et al., 2021). There are several examples which are come under the category of contaminants, such as pesticides, herbicides, sewage and organic compounds, toxic gases, fertilizers, trace metals etc. (Vaseashta et al., 2007; Khan and Pathak, 2020). Therefore, to deal with these challenges, the engagement of NPs in the expansion of emerging green remediation technologies has been the subject of recent investigations (Tratnyek and Johnson, 2006). NBR is a unique technology employed in transforming the adverse effects of pollutants into safer molecules through NPs.

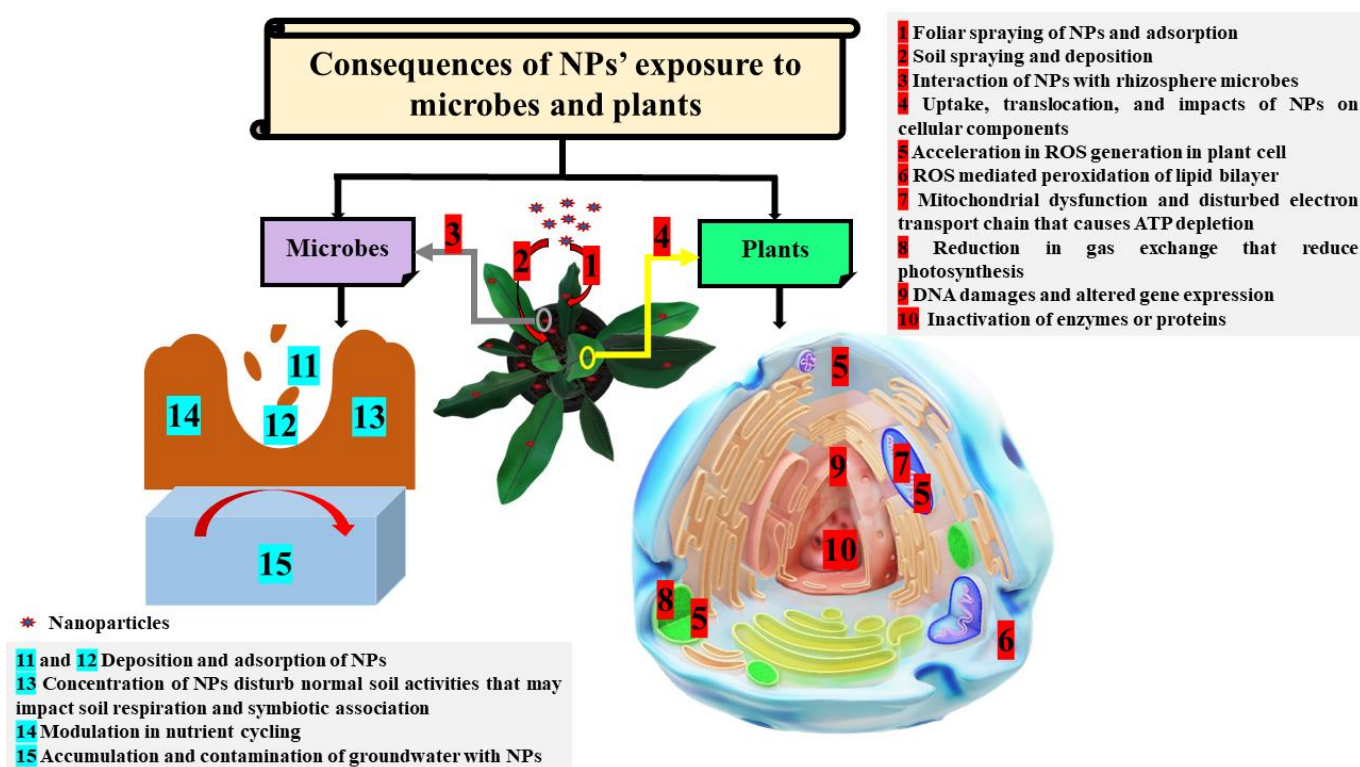


Figure 2. The exposure of NPs and associated impacts on microbes and plants

However, nanotechnology has gained very much importance in recent years because of its extraordinary properties. It has been accounted to play a major function in tackling diverse efficient and inventive resolutions to several ecological confront (Yan et al., 2013; Reddy et al., 2014). But pioneering thoughts for progress are similar a twice impacts. Every unique approach has been connected to pros and cons. It depends on researchers how they tackle and apply the new approach. In the turf of NBR, the negative aspects related to NPs are very significant and crucial which cannot be disregarded (Jiang et al., 2018; Rajput et al., 2021b).

Besides their positive effects, some negative aspects of NPs are also being seen in the environment. It is documented that NPs do not supply any profit in the situation of bioaugmentation, since they stop the microbial inhabitants in contaminated surroundings (Nzila et al., 2016; Amoatey and Baawain, 2019). The appliance of NPs for ecological action intentionally injects NPs into the soil or water body. This has finally involved rising anxiety from all stakeholders. The compensation of NPs such as their minute size, elevated activity, and immense capacity, could develop into a possible deadly feature by inducing unfavorable cellular toxic and damaging properties, abnormal in small-sized counterparts (Figure 2).

In stiff water and seawater, NPs tend to aggregate and are greatly influenced by the type of natural material or other natural colloids present in freshwater. The situation of dispersal will change the ecotoxicity, but several abiotic factors that influence this, such as pH, salinity, and the attendance of organic matters stay to be methodically investigated (Handy et al., 2008). It was demonstrated that the hindering effect of the nZVI in soils happens when the NPs begin to be putting on the facade of the soil particles, accumulating in such a method that they attract additional constituent parts in suspension, jamming the way of fluids (Reddy et al., 2014). Since the strainer result occurs as the concluding phase of deposition, which ends up providing a

“clogging” of the soil hole, not allowing for the channel of the element. It has been confirmed that carbon nanotubes (CNT) abridged the biodegradation pace by hindering bacterial expansion and microbial action (Zhang et al., 2015).

Nanosorbent have a significant impact in explaining ecological subjects like the filtration of water that established immense interests because of their unique physicochemical properties. However, the use of nanosorbent material in water bodies can also have certain negative consequences (Yaqoob et al., 2020). Major drawbacks are the probable negativity of the remaining NPs in the water and their large size which causes that few probable functioning is not used (Zhu et al., 2019). The exercise of silver NPs in many products direct them to their discharge to the water body and befall a source of suspended silver and thus produce negative impacts on marine organisms (Navarro et al., 2008).

Several microbes are present in the water body; therefore, it is a very natural process for NPs to encounter microbes after they are released into the water body. When nZVI is in straight connection with bacterial cells, it results in oxidative stress and membrane demolition (Figure 2). The current study represents the thrashing of intracellular components and the disturbance of communication between the outside and inside environment of the bacteria (Lv et al., 2017).

Carbon nanotubes change the oxidation nature of enzymes in water molecules, which causes adverse impacts on microbes (Chen et al., 2016). Graphene oxide enhances the active oxygen application, but it does not harm cells. However, a higher concentration of silver NPs are used, the enzymatic action was retarded, but the genes for resistance were augmented (Li et al., 2019; Kolesnikov et al., 2021). The application of silver NPs and zinc oxide NPs on the activity of bacterial is reported to depend on the dimension of particles, and the microorganism concentration (Mboyi et al., 2017). Treatment of zinc oxide NPs to anaerobic fermentation, zinc ions are engrossed in the mud, however, bacterial quantity, cell activity, enzymatic activity, and zinc ion concentration significantly decreased (Figure 2).

Generally, the negative impacts of NPs on microbial activity largely engage membrane devastation and oxidative stress (Rajput et al., 2017a; Chen et al., 2019). Conclusively, microorganisms and planktons are highly vulnerable to the toxicity of NPs. Furthermore, these water-loving organisms are pretentious by the adverse effects of NPs, and occasionally it is quite hard to recover due to those NPs not simply root of cell injury, but also harm genes and influence reproduction.

Challenges associated with nanobioremediation

- Nanophytoremediation studies are yet to be adopted widely and need to explore rigorously.
- Most studies using nanophytoremediation approaches are microcosm therefore *in-situ* and realistic studies in future research could bring a new direction in this scope
- Time series and long-term research using NPs are also necessary, that can enable us to observe the actual effects of NPs in phytoremediation progression and also their effect on soil characteristics, microbiome, and nutrients
- NPs may get aggregate, dissolved, undergo dissociation in different soil pH, or it can also undergo photodegradation. These processes certainly affect their mobility. Application of doping, composite, or polymeric structure for nanophytoremediation must be explored in this regard.
- Assessment of effects and safety of NPs application in agriculture or polluted soil should be mandatory. Sustainable nanophytoremediation largely relies on climatological conditions hence our exploration should also include the identification of a naturally stable NPs

Conclusion

The advent in nanobiotechnology as a research field brings up possibilities for developing nanoremediation methods for the restoration of contaminated soils. Several investigations' experimental findings revealed the potential of nanobioremediation for the removal of various inorganic and organic pollutants from terrestrial ecosystems. Also, these techniques could be applied to decontaminating air, or water, in cost-effective ways; however, significant environmental concern regarding the application of NPs should be in the regulatory framework, and eco-friendly. Thus, the understanding of NPs interaction with plants, microbes, pollutants, and human health is of utmost importance as these effects might be negative or positive. Thus, nanobioremediation will undoubtedly be a promising tool for achieving environmental sustainability once these research gaps regarding its environmental concerns will have been revealed.

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