REVIEW ON PHYTOEXTRACTION OF NICKEL IN THE CONTEXT OF PLANT-METAL INTERACTION

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ABSTRACT

Metal pollutants can be produced through industrial processes such as mining, refining, and electroplating.Due to continuous irrigation of sewage water in agricultural field, application of sewage sludge to the crops, indiscriminate disposal of electroplating industry effluent and some fertilizer usage lead to the Ni contamination in soil. Presently phytoremediation is a popular technology which deals with the rehabilitation of metal contaminated sites using plants.In phytoremediation, phytoextraction is the major mechanism which absorbs metals from the soil and accumulated in plant biomass. The major criteria for selecting plant species plant species for phytoextraction are hyperaccumulating, fast growing species with high biomass. Physico-chemical and biological processes of soil systems also significantlyaffect themetal bioavailability. The primary soil factors controlling bioavailability are soil pH, cation or anion exchange capacity, clay content, and soil organic matter.For chemically-assisted phytoextraction, the dynamics of metal chelates in the rhizosphere need to be examined, either to overcome the risks associated with low degradability of synthetic chelators, or to optimize the use of more biodegradable compounds. A complete understanding of plant metal tolerance will be essential to develop strategies to genetically enhance the metal accumulation ability of plants.

INTRODUCTION

The heavy metal toxicity causes serious threats to human and animal health due to their long-term persistence in the environment (Gisbert *et al.*, 2003). The increasing attention on threats caused by heavy metal toxicity necessitated the remediation of polluted soil by using plants and microbes to remove those hazardous metal ions (Winge *et al.*, 1985; Pence *et al.*, 2000). Plants have harvestable stem and leaves aboveground, which is suitable for subsequent post-processing, whereas the microbes possess a larger specific surface area that activate and remove heavy metals which cannot be harvested (Salt and Rauser, 1995; Kramer *et al.*, 1996). Therefore plants are more preferentially used for remediating the heavy metal contaminated soil. Phytoremediation is a cost-effective technology to remediate the contaminated soil using hyperaccumulating plants. In this technology, the living plants act as a solar-driven pump, which can extract and concentrate certain heavy metals from the environment (Raskin *et al.*, 1997). For phytoremediation of metal contaminated soils, it is essential to understand the interaction between metal-tolerant plant species and soil chemical properties controlling the bioavailability of metals. Numerous plant species have been identified for the purpose of phytoremediation. Hyperaccumulators plants are more appropriate as they are able to accumulate potentially phytotoxic elements to the concentrations of 50-500 times higher than the average plants (Lasat, 2002).

The term hyperaccumulator was first applied by Jaffre*t al.*

(1976) when they observed the accumulation of Ni in *Sebertia accuminata*. The present idea concerning the concentration of more than 1000 mg/kg of metal in plant tissues was introduced by Brooks *et al.* (1977), when they examined Ni concentrations in *Homalium* sp. and *Hybanthus* sp. For most elements the threshold uptake concentration is 1000 mg/kg (0.1%) dry mass, except for Zn (10,000 mg/kg), Cd (100 mg/kg). Around 400 plant species from at least 45 plant familieshyperaccumulate metals, most of which are Ni hyperaccumulators occurring in ultramaûc areas (Reeves, 2006). Some of the important Ni hyperaccumulators were given below (Table 1).

Plant metal interaction

Certain non-hyperaccumulating plants retain the heavy metals in their root cells and detoxify them by chelating in the cytoplasm or by storing them into vacuoles. Contrastingly, hyperaccumulators release metal chelating compounds (phytochelators/ phytosiderophores)to the rhizosphere, which increase the bioavailability of metals that are tightly bound to the soil fractions and help to translocate them into plant tissues (Eapen and D'Souza, 2005). Phytochelators are usually low molecular weight organic compounds such as malic, malonic, oxalic acids, acetic acid, succinic acid, sugars, amino acids and phenolics which can change the metal speciation and bioavailability (Cieslinski *et al.*, 1998; Ma *et al.*, 2001; Nascimento *et al.*, 2006). Certain metal chelating compounds (mugenic acid and avenic acids) are released during nutrient metal deficiency and thus increase the bioavailability of metals (Ma and Nomoto, 1996, Pellet *et al.*, 1995, Cakmak*et al.*,

1996) and help to carry them into plant tissues. Metal chelate complexes may also be transported across the plasma-membrane (Romheld, 1991). The rhizosphere provides a complex and dynamic micro-environment where microorganisms form unique communities that have potential for detoxification of hazardous waste compounds. Their interaction can improve metal bioavailability in rhizosphere through the secretion of proton, organic acids, phytochelatins (PCs), amino acids, and enzymes (Idris*et al.*, 2004; Yang *et al.*, 2005). Fungal symbiotic associations have the potential to enhance root absorption area and stimulate the acquisition of plant nutrients including metal ions (Khan *et al.*, 2000).

Mechanism involved in the phytoextraction of metals

Phytoextraction seems to be the most promising technique and has received increasing attention from researchers since it was proposed by Chaney (1983) as a technology for reclaiming metal polluted soils.Phytoextraction employs metal hyperaccumulating plant species to transport high quantities of metals from soil into the harvestable parts of roots and aboveground shoots (Kumar *et al.*, 1995; Chaney *et al.*, 1997). Ni hyperaccumulation was defined by Brooks *et al.* (1977) as the accumulation of at least 1000 mg kg^{-1} Ni in the dry biomass of plants grown on a natural substrate. Subsequently, hyperaccumulators of other heavy metals were identified. Metal concentrations achieved by naturally occurring hyperaccumulatingplant species can be more than 100 times those that occur in non-accumulator plants growing in the same substrates. The extraordinary ability of hyperaccumulator plants to tolerate and accumulate heavy metals has stimulated research into possible uses of phytoextraction (Chaney *et al.*, 1995).

Phytoextraction is the most feasible method and widely utilized to extract metals from soils. Hyperaccumulators absorb metals through roots and translocate them to shoots. Then, the metals can be harvested from the plant biomass by incineration techniques. Phytoextraction involves various steps such as solubilization of metal from the soil matrix, root uptake and transport to shoots, and finally translocation and sequestration of metals in the plant biomass (Fig.1). Salt *et al.* (2000) were unable to identify any high-affinity Ni-chelator compound in the rhizosphere of the Ni hyperaccumulator *Thlaspi g oesingense*. In contrast, they found that Ni-chelatorshistidine and citrate accumulated in the root exudates of the non-hyperaccumulator *Thlapsi arvense* exposed to Ni. Such findings led the authors to suggest that the release of these exudates by *T. arvense* may be a strategy to reduce Ni uptake and toxicity, but exudate releasing is not involved in the hyperaccumulation of Ni by *T.goesingense*. Persans *et al.* (1999) also established that Ni hyperaccumulation in *T. goesingense* is not determined by the overproduction of histidine in response to Ni. Since at non-toxic Ni concentrations, both plant species translocate Ni to shoots at equivalent rates (Kramer *et al.*, 1997), the existence of a more efficient translocation mechanism in *T. goesingense* does not seem to explain the capability of this species in accumulating Ni. Kramer *et al.* (2000) provided evidence that free histidine may be also involved in shuttling Ni across the cytoplasm into the vacuole in *T. goesingense*, which could be responsible for Ni tolerance and accumulation. Kramer *et al.* (1996) have already reported

a 36-fold increase in the concentration of free histidine in the xylem exudates of the Ni hyperaccumulator*Alyssum lesbiacum* after exposure to Ni, suggesting that histidine could be involved in the transport and storage of Ni in such species. Kerkeb&Krämer (2003) recently provided further evidence that histidine enhances the release of Ni from roots into the xylem, not only in *A. lesbiacum* but also in the non-hyperaccumulator *Brassica juncea*.

The main role in heavy metal accumulation is played by free amino acid such as histidine (His) and nicotinamine, which forms stable complexes with the bivalent cations. Free histidine (His) is considered as the most important ligand in the hyperaccumulationof Ni (Callahan *et al.*, 2006). Presence of high concentration of His in the roots of *Thlaspi* species which is Ni hyperaccumulators, suggest the same mode of operation of amino acid in other hyperaccumulators (Assuncao*et al.*, 2003). Because of the presence of carrier for the transport of Ni in plants, heavy metal get absorbed from the soil easily, crosses the cell wall and plasma membrane of the root and through xylem gets accumulated in the leaf vacuole (Fig. 2).

The root to shoot translocation in hyperaccumultor plants relies on enhanced xylem loading by constitutive overexpression of genes coding for transport system common to non-hyperaccumulators (Rascio and Navari-Izzo., 2011). Moreover, Heavy Metal Accumulation (HMAs) plays a vital role in metal homeostasis and tolerance (Axelsen and Palmgren, 1988). The MATE (Multidrug And Toxin Efflux) family of small organic molecule transporter seems to be another transport protein that is active in translocation of heavy metals in hyperaccumulation plants (Rascio and Navari-Izzo, 2011).

Metal transport from the cytosol to the vacuole is considered an important mechanism of both metal tolerance and accumulation in plants. For this reason, much work has been dedicated to investigating subcellular localization of metals in hyperaccumulators (Vázquez *et al.*, 1992; Kupper *et al.*, 1999; Hirschi *et al.*, 2000; Kramer *et al.*, 2000; Sarret *et al.*, 2002). Kramer *et al.* (2000) isolated vacuoles from Ni-tolerant *T. goesingense* and Ni-sensitive *T. arvense* aiming directly to address the role of vacuolar Ni storage in Ni tolerance. They found that *T. goesingense* accumulated two-fold more Ni in the vacuole than *T. arvense*. Since protoplast and apoplastNi contents were similar in both species, vacuolar compartmentalization in *T. goesingense* seems to play a major role in Ni-accumulation and tolerance.

Soil amendments for nickel (im)mobilization

A number of amendments are used either to mobilize or immobilize heavy metals in soils. The basic principle involved in the mobilization technique is to release the metals into soil solution, which is subsequently removed using higher plants. In contrast, in the case of the immobilization technique the metal concerned is removed from soil solution either through adsorption, complexation and precipitation reactions, thereby rendering the metals unavailable for plant uptake and leaching to groundwater.

Chelating and complexing agents

When a metal ion combines with an electron donor, the resulting substance is called a complex or coordination

Table 1: List of important Nickel hyperaccumulators

Hyperaccumulator plant	Family	References
Sebertia acuminate	Sapotaceae	Jaffre et al. (1976); Perrier (2004)
Allysum pindasilvae	Brassicaceae	Garcia-Leston et al. (2007)
Allysum bertolonii	Brassicaceae	Barzanti et al. (2011)
Allysum serpyllifolium	Brassicaceae	Becerra-Castro et al. (2009)
Phidiasia lindavii	Acanthaceae	Reeves et al. (1999)
Bornmuellera kiyakii	Brassicaceae	Reeves et al. (2009)
Thalapsi goeingense	Brassicaceae	Wenzel et al. (2003)
Berkheya codii	Asteraceae	Robinson et al. (1997); Moradi et al. (2010)

Table 2: Plants used for the Nickel remediation and their accumulating capacity

Plant	Family	Amount of Ni accumulated(mg kg^{-1})	References
Alyssum caricum	Brassicaceae	7576	Altinozlu et al., 2012
Alyssum peltariodes	Brassicaceae	4411	
Isatis pinnatiloba	Brassicaceae	275	
Aegopordon berardioides	Asteraceae	20.0	Ghaderian and Baker., 2007.
Cleome heratensis	Capparaceae	21.0	
A. serpyllifolium	Brassicaceae	38105	Freitas et al., 2004.
L. spartea (L.)	Scrophulariaceae	492.0	
A. serpyllifolium	Brassicaceae	670-31200	Lazaro et al., 2006
Cistus ladanifer	Cistaceae	3-50	
Plantago subulata subsp.Radicata	Plantaginaceae	46.4-267	
Silene armeria	Caryophyllaceae	2540	Lombini et al., 1998.
Cerastiumarvense		2685	
Minuartia laricifolia		2629	
Dianthus sylvestris		2501	
Biscutella laevigata		2399	
Alyssum bertolonii	Brassicaceae	2594	
Brassica juncea	Brassicaceae	18	Hsiao et al., 2007.
Cynanchum schlechtendalii	Asclepiadaceae	235	Reeves et al., 2007
Macroptilium gracile	Fabaceae/Papilionaceae	114	
Hyptis suaveolens	Lamiaceae	175	
Oxalis frutescens	Oxalidaceae	106	
Paspalum pectinatum	Poaceae	170	
Diodia teres	Rubiaceae	246	
Buchnera pusilla	Scrophulariaceae	185	
Russelia sarmentosa	Scrophulariaceae	130	
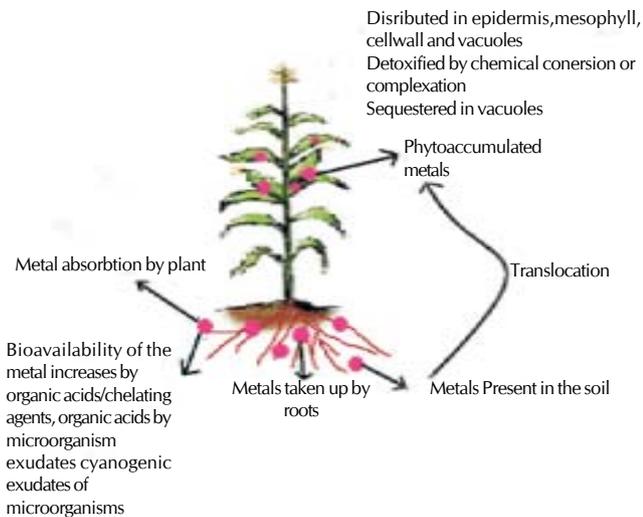
Figure 1: Mechanism of phytoextraction
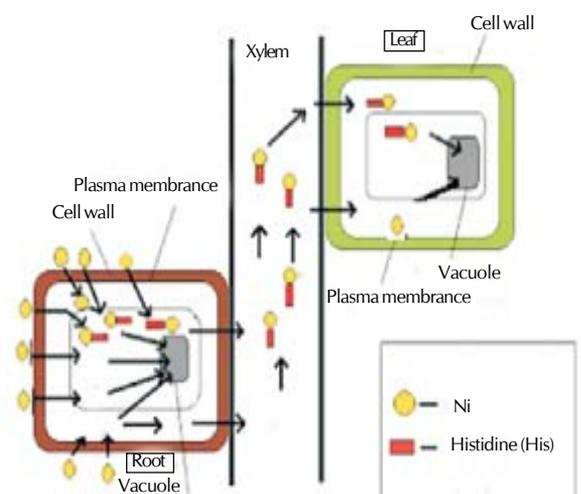
Figure 2: Schematic representation of translocation of Ni from root to leaf

compound. If the substance which combines with the metal contains two or more donor groups so that one or more rings are formed, the resulting structure is called a metal chelate, and the donor is called chelating agent.

The term 'chelate' is derived from the Greek word 'chela' which means 'claw' and it is so named because these species can coordinate at several or all positions around a central metal ion by literally wrapping themselves around the metal ion. Chelating agents which have high affinity for metal ions can be used to enhance the solubilization of metals in soils through the formation of soluble metal chelates. A number of synthetic chelating agents are available including EDTA, EDHA, DTPA and EHPG.

Liming materials

Although liming is primarily aimed at ameliorating soil acidity, it is increasingly being accepted as an important management tool in reducing the toxicity of heavy metals in soils (Bolan *et al.*, 2003a, 2003b). A range of liming materials is available, which vary in their ability to neutralize the acidity. These include calcite ($CaCO_3$), burnt lime (CaO), slaked lime ($Ca(OH)_2$), dolomite ($CaMg(CO_3)_2$) and slag ($CaSiO_3$). The acid neutralizing value of liming materials is expressed in terms of calcium carbonate equivalent (CCE), defined as the acid neutralizing capacity of a liming material expressed as a weight percentage of pure $CaCO_3$. Liming, as part of the normal cultural practices, has often been shown to reduce the concentration of Cd, Pb and other metals in edible parts of crops. Similarly, liming serpentine soils containing toxic levels of Ni has shown to alleviate the phytotoxic effects of Ni. In these cases, the effect of liming materials in decreasing metal uptake by plants has been attributed to both decreased mobility in soils (through adsorption/ precipitation) and to the competition between Ca^{2+} and metals ions on the root surface.

Organic composts

Although a number of studies have examined the role of biosolid as a source of metal contamination in soil, only limited work has been reported on the beneficial effect of organic amendments as a sink for the immobilization of metals in soils. Addition of organic amendments has often been shown to increase the CEC of soils, thereby resulting in increased metal adsorption (Bolan *et al.*, 2003c). The presence of phosphates, aluminium compounds and other inorganic minerals in some organic amendments, such as typical municipal sewage sludge, is also believed to be responsible for the retention of metals, thereby inducing the 'plateau effect' in metal uptake by crops and preventing the increased metal availability suggested in the 'time bomb' hypothesis.

Metals form both soluble and insoluble complexes with organic constituents in soils, a process which apparently depends on the nature of the organic matter (Bolan *et al.*, 2003d). As might be expected, the organic component of soil constituents has a high affinity for metal cations because of the presence of ligands or functional groups that can form chelates with metals. With increasing pH, the carboxyl, phenolic, alcoholic and carbonyl functional groups in soil organic matter dissociate, thereby increasing the affinity of ligand ions for metal cations. Sathya *et al.* (2016) found that the soil application of bioamendments resulted in the formation of complexes with fulvic and humic acid fractions of soil organic matter. While fulvic acid complex enhanced the bioavailability by mobilization of Ni ions, the humic acid complex resulted in immobilization of Ni and enhanced the adsorption of Ni ions on soil exchange sites. Arnesen and Singh (1999) found that the application of peat increased the amount of DTPA-extractable Ni. They suggested that the lowering of pH in the peat-amended to soil decreased the sorption of Ni in the soil and this effect became more significant after degradation of organic matter. Karaca (2004) reported that the amount of DTPA extractable Ni has been increased after the addition of mushroom compost due to the higher organic matter content. Tobacco dust application also increased the DTPA extractable Ni (Karaca, 2004). The concentration of exchangeable Ni was increased due to the application of poultry manure pressmud compost in an incubation study (Sathya and Mahimairaja, 2016). In a pot experiment, prosopis biochar was found to reduce the Ni concentration in marigold but pressmud compost and poultry manure were found to increase Ni content and uptake by marigold plants (Sathya *et al.*, 2016).

Accumulation of nickel in plants

Since Ni is ubiquitous in the environment, it is a normal constituent of plant tissues. Nickel content of field-grown crops and natural vegetation ranges from 0.05 to 5.0 ppm in the dry matter (Vanselow, 1966). In normal plants, the following ranges of values have been reported (Kabata-Pendias and Pendias, 1992): grasses, 0.10 to 1.7 ppm DW; clovers, 1.2 to 2.7 ppm; vegetables, 0.20 to 3.7 ppm; cereals, average of 0.50 ppm. Generally, the level of Ni in most plant species that may produce phytotoxicity ranges from 10 to 100 ppm (Adriano, 2001).

Nickel is usually absorbed in the ionic form Ni^{2+} from the soil or culture medium. Nickel is apparently more easily absorbed by plants when supplied in the ionic form than when chelated (Mishra and Kar, 1974). A possible explanation is that the charge on the chelated molecule inhibits its absorption by the roots, i.e., a molecule with no charge or a slight negative charge can be taken up, while a complexed molecule with strong negative charge cannot. High Ni content of the soil, particularly when the exchangeable form is also high, facilities the absorbtion of this element by plants when other conditions are also favourable (Adriano, 2001).

A nickel hyperaccumulator grown under natural conditions should contain 1000 mg kg^{-1} dry matter or more Ni in its aboveground tissues (Baker & Brooks, 1989; Reeves *et al.*, 2001, Reeves *et al.*, 2009). The nickel content of hyperaccumulators may range between 4200 and 20,400 mg kg^{-1} (Li *et al.*, 2003). Nickel-accumulating species comprise about three-quarters of all hyperaccumulator plants (Baker *et al.*, 2000; Reeves & Baker, 2000; Ghaderian *et al.*, 2007). The genus *Alyssum* L. (Brassicaceae) has the highest number of such members, with 48 taxa accumulating up to 3% Ni in their foliage (Brooks *et al.*, 1977; Reeves & Baker, 2000). The number of discovered Brassicaceae members, some of which are Ni hyperaccumulators, has increased day by day (Ozhatay *et al.*, 2009; Mutlu, 2010; Ozhatay *et al.*, 2011). Hyperaccumulators do not usually compete with normal flora (Baker *et al.*, 2000). Soleimani et al. (2009) reported that tall fescue grass accumulated more Ni than the Bermuda grass.

Not likewise other metal the Ni has accumulated more in shoots than the roots. Motesharezadeh and Savaghebi-Firoozabadi (2011) studied the effect of different inoculants on the efficiency of Ni phytoextraction by sunflower, amaranthus and alfalfa. They have obtained the highest accumulation with the *Bacillus safensis*inoculants in amaranthus. Bosiacki and Wojciechowska (2012) studied the phytoextraction capacity of different ornamental plants such as marigold, sunflower and amaranthus. They have assessed the Ni accumulation in different plant parts like inflorescences, leaves and stem. Among the three plants, marigold and amaranthus accumulated higher amount of Ni than the sunflower. Several researchers used wide variety of plant species to study the accumulation capacity and pattern for Ni in the selected plant species. The list of plants used so far in the remediation of Ni is given in the table 2.

Nickel toxicity in plants

Nickel is metabolically important and essential minor element for the development of the plants but increase in concentration results in toxicity (Fargasova, 2008). In general, when Ni concentrations in vegetative tissue of plants exceed 50 ppm DW, plants may suffer from excess Ni and manifest toxicity symptoms (Adriano, 2001). Different plants species have different resistivity against nickel. While some plants are introduced as Ni hyperaccumulators other are very sensitive and introduced as non-accumulators (Freeman et al., 2004). In the cytoplasm, high levels of free nickel generally avoid removal of the metal ions to the vacuoles and the formation of complexes with organic acids (Ernst et al., 1990). High concentration of nickel inevitably binds organic macromolecules and denatures them. Furthermore, nickel can replace iron, zinc and magnesium due to the chemical affinity with those elements, interfering with their metabolism (Woolhouse, 1983). Ni is transported to underground plant parts by the oxygen atoms either as metal complexes of organic acids or as hydrated cations (Salt et al., 2002). High Ni concentrations retard shoot and root growth, affect branching development, deform various plant parts, produce abnormal flower shape, decrease biomass production, induce leaf spotting, disturb mitotic root tips and produce Fe deficiency that leads to chlorosis and foliar necrosis. Additionally, excess Ni also affects nutrient absorption by roots, impairs plant metabolism, inhibits photosynthesis and transpiration, and causes ultrastructural modifications (Ahmad and Ashraf, 2011).

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