

Critical Analysis of Remediation Methods of Metal Contaminated Lands

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Abstract Methods of land remediation continue to evolve. A number of reviews have been recently published on different aspects of land reclamation with a focus on bioremediation. In fact, bioremediation of metal contaminated sites using plants and microorganisms is currently the most recommended strategy to remove toxic metals from ecosystems. Recently, the use of nanoparticles for remediation of metal contaminated sites has been reported as a promising tool to restore lands. This because of their exceptional adsorption and mechanical properties combined with unique electrical property, high chemical stability and their application to a large surface area. The most efficient strategy in many cases is to combine different remediation methods for a sustainable effect. This review is a critical analysis of key findings on physical, chemical and biological restoration techniques used to reclaim metal contaminated lands.

Keywords: *Metal contamination, Metal toxicity, Anthropogenic activities, Physical, chemical, and biological remediation, Phytoremediation, Land reclamation, Soil restoration, Soil liming*

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1. Introduction

Soil is the Earth's delicate outermost layer where all life is found. It is among the most valuable resources to human [1,2] and a significant component of the earth's biosphere [3]. A number of species that form a dynamic and complex ecosystem and plays significant roles in terrestrial ecosystems are found in soil [1,2]. Soil composition includes minerals particles (such as sand, silt and clay), organic materials (soil organic matter [SOM] or humus resulting from the living organisms, gases, liquids and living organisms including bacteria, fungi, algae and insects [1,2,3]).

Most metal(loid)s in the periodic table can be naturally found in soil at varying concentrations. Soils can naturally have high concentrations of metals as a result of weathering of parental material with high amounts of metal minerals (lithogenic source) [4,5,6,7]. Anthropogenic activities such as mining, smelting, atmospheric deposition, fossil fuel combustion, wastewater irrigation, corrosion, sewage sludge and agriculture practices have also resulted in metal contamination of soils [4,6,7]. This can be localized or widespread. Point-source pollution results from a predominant single source that is discernible and confined. An example is the release of poisonous chemicals and toxic gases directly into the air via a chimney such as a smelters [8]. Specific metal(loid)s can be found in soil at industrial sites which is dependent on the type of industry, the use of raw material and product

output [5]. Non-point source pollution comes from various diffuse sources; it is more dispersed in nature. The non-point sources of pollution in air can originate from the release of chemicals such as oxides of nitrogen (N), unburned hydrocarbons from gasoline from vehicles and the burning of fossil fuels containing high sulfur (S) content which produces and releases sulfur dioxide (SO₂) into the atmosphere [8].

Volatile metals found in the atmosphere can adhere to particulates resulting in their widespread distribution in the atmosphere from their release site. The smaller and lighter metal particles are more persistent in the atmosphere [9,10]. Pollutants can enter the soil from the air via acid rain and also from the vehicle exhaust. Nonpoint source pollutions usually take a longer period to have a substantial impact on the environment but over time, they will likely have significant negative effects [8]. The determination of the nonpoint source pollution is challenging since these sources are widespread, coming from varying locations making it impossible to determine the exact source of the pollution. Soils act as a sink for all chemicals generated from various natural and anthropogenic activities mentioned above. The retention time of many substances in soil ecosystem is longer as contaminants accumulate quickly in soils but deplete at a slow rate [4,7]. The release of metals cause soil pollution because they accumulate in the soil over time because they are persistent, cannot be biodegraded and are stable in the environment [1,4,7,11]. Significant progress has been reported in the understanding of mechanisms that govern metal accumulation and detoxification in

accumulating plants [12]. Methods of land remediation continue to evolve. A number of reviews have been recently published on different aspects of ecosystems remediation. Rehman et al. [13] reviewed remediation of heavy metal contaminated soils by using *Solanum nigrum*. Hayat et al. [14] described current studies on microbial biotechnology as an emerging industrial wastewater treatment process for arsenic mitigation. Sruthi et al. [15] on the other hand summarized current literature on heavy metal detoxification mechanisms in halophytes. Camargo et al. [16] compiled and discussed studies on removal of Toxic Metals from Sewage Sludge through Chemical, Physical, and Biological Treatments. This review summarizes key studies on physical, chemical and biological restoration strategies of terrestrials sites contaminated with metals.

2. Remediation of Soils Contaminated with Metals

Land is static, the pollution in soil remain in the soil unless that area is remediated. Increasing soil metal contamination has important health and economic implications. Metals can be taken up by plants or leached into groundwater leading to food chain contamination. Several cleanup methods have been investigated and regulatory steps have been implemented and executed to restrict the release of metal pollutants into soils [17]. They are divided into two groups: 1) those that remove contaminants and 2) those that transform metals into harmless forms [4,11,18]. These cleanup technologies can be applied on or off-site (*in situ* or *ex situ*), using three types of remediation treatments: 1) physical, 2) chemical and 3) biological techniques [4,10,11,18,19]. Some contaminated sites may require a combination of procedures for an efficient remediation. Therefore, physical, chemical and biological technologies may be used in conjunction with one another to reduce the contamination to a safe and acceptable level.

2.1. Physical Remediation

Physical remediation includes soil replacement, thermal desorption and electrokinetic migration [4,18,20]. Soil replacement consists in using clean soil to replace contaminated soil to dilute the concentration of pollutant [18]. Soil replacement effectively decreases the effect of metals on environment but it is expensive and suitable for severely polluted small areas [18]. Thermal desorption consists in heating soil in a chamber where organic contaminants and certain metals can be vaporized [18,20]. Hence, thermal desorption depends on metals volatility and temperatures (e.g. mercury [Hg] and arsenic [As]) [18,20]. It is divided into low temperature (90-320°C) and high temperature desorption (320-560°C) based on temperature in the operating chamber [18,20]. This technique has several advantages. It is simple and less expensive to process, remediated soil is reused and is environmentally friendly [18,20]. Its disadvantages include, high cost of device and long desorption time. It is not equally effective for all soil types, and it cannot be used for all metals [18,20].

Electrokinetic process consists in passing a low intensity electric current between a cathode and an anode imbedded in the contaminated soil [4,21]. Anions move towards anode (positive electrode) and cations towards cathode (negative electrode) [21]. Buffer solutions are used to maintain the pH at the electrodes as it is essential in the optimization of the process efficiency [4,21]. Metals are removed by electroplating or precipitation/co-precipitation at the electrodes, using ion exchange resins or they can be recovered by pumping the waste to the surface [4,21]. Metals can be removed from soil since they are bound to it as oxides, hydroxides, and carbonates [4,21]. In Europe, this technology is used for copper (Cu), zinc (Zn), lead (Pb), As, cadmium (Cd), chromium (Cr) and nickel (Ni) [21]. This method has several advantages: 1) can be used *in situ* or with excavated soil, 2) effective with clay soils of low permeability, 3) accelerated rate of contaminant transport and extraction can be obtained [4,21]. However, heterogeneities, large metal objects, rocks, amounts of oxides, foundations, rubble, other obstacles, moisture content, temperature and other contaminants can interfere with the process [4,21]. Electrode duration and spacing is site-specific and may need to be optimized [21].

2.2. Chemical Remediation

Chemical remediation technique includes all the methods involving reagents or external compounds [4,21]. This includes chemical leaching and fixation, vitrify technique and chemical immobilization [4,18,21]. Chemical leaching is a volume reduction and waste minimization treatment done on excavated (*ex situ*) soil or on-site (*in situ*) [20,22]. It involves washing contaminated soil with fresh water and solvents which have the ability to solubilize metals [18,20,22]. Leaching solution and chelators are added to extract metals from soil [4,18,20]. Chemical fixation technique involves addition of reagents to metal contaminated soils. These agents are then used with metals to form insoluble and nontoxic forms. This process decreases the migration of metals into the environment [18].

Vitrify technique involves heating soils to extremely high temperatures between 1,400 °C and 2,000 °C [18]. The mobility of metals is reduced by high temperature treatment which results in the formation of vitreous materials, usually a solid oxide [18,22]. Increased temperature melts contaminated soil, buried wastes or sludge's rendering the material nonhazardous [18,22]. This technique can be applied to water, debris and various soil types. *In situ* vitrifications are preferred due to low energy requirement and cost. *Ex situ* process includes excavation, pretreatment, mixing, feeding, melting and vitrification, gas collection and treatment and forming or casting of the melted products [18,22].

Chemical immobilization is an *in-situ* technique where inexpensive materials (e.g. lime, fertilizers, fly ash, iron/manganese [Fe/Mn] oxides and cement) are added to contaminated soil. They can remove and/or stabilize metals in soils, resulting in a substantial reduction of costs [4,22,23,24,25]. This relies on a fundamental understanding of natural geochemical processes governing the speciation, migration, and

bioavailability of a given metal in the environment [4,26]. *In situ* chemical immobilization technique provides long term remediation solution by decreasing the concentration of contaminants by sorption or precipitation [4,22,25,26]. Mobility and bioavailability play a huge role in solubility of metals. Increased sorption and decreased solubility reduce pollutant transport and redistribution into the environment [7,25,26]. Chemical immobilization treatments serve also as reactive barrier which prevents metal seepage from recovery pits and other processing areas on active and inactive sites [26]. The redistribution of metals by means of solute transport mechanism can adversely affect ecosystem, water resources and human populations [22,26].

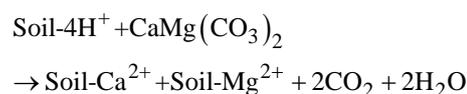
Chemical immobilization of metals uses phosphate and alkaline based materials to adsorb, chelate or complex metals in soils [22,26]. Metal contaminated soils treated with phosphates (apatite) reduce metal solubility by forming metal phosphate precipitates and minerals [22,26]. Addition of phosphate material and presence of sufficient level of soil phosphorous (P) have proven to be extremely effective as chemical immobilization of Pb and Zn [22,26]. In addition to reducing metal solubility, it decrease bioavailability of metals to plants and animals [22,26].

Chemical immobilization using alkaline amendments (lime) reduces metal solubility in soils by increasing pH and metal sorption to soil particles [23,24,25,27]. Increased soil pH and carbonate buffering lead to the formation of metal-carbonate precipitates, complexes, and secondary minerals [23,24,26,27]. Soils become acidic because basic cations are replaced by hydrogen (H^+) ion on the soil colloidal complexes [23,24,26,27]. As pH decreases, the availability of P, potassium (K), calcium (Ca) and magnesium (Mg) is reduced, while the availability of Zn, Mn, Cu and Fe increases [23,24,26,27]. Various studies have used lime (Ca containing inorganic material) as chemical amendment for remediation of soils contaminated with metals [7,23,25,27].

In addition to increasing pH, lime is used to increase mineralization of soils with Ca and in some cases, Mg [23,24,26,27]. It further, reduces metal solubility, improves soil structure and stabilizes soil nutrients, which helps reduce soil erosion resulting in an increase in SOM [23,24,27]. Lime can be applied as calcium hydroxide ($Ca(OH)_2$), calcium oxide (CaO) or calcium carbonate ($CaCO_3$) to effectively neutralize soil acidity [26,28]. Addition of lime neutralizes acidity, and increases microbial activity in soils. In the Greater Sudbury Region (GSR) in Northern Ontario (Canada), dolomite lime ($CaMg(CO_3)_2$) was applied as it primarily neutralizes acidity but also provides Ca and Mg for plant uptake [23,24]. The role of both Ca and Mg together is crucial to metal toxicity as they create a competitive exclusion of metal ions [23,24,27,29].

The increase in pH when lime is added to soil depends on the cation exchange capacity (CEC) [28,30]. Soils with low CEC will show a more marked pH increase compared to soils with high CEC [28,30]. Also, low CEC is associated with rapid leaching of added basic cations and a quick return of original acidity unless additional liming is applied [28,30]. Ions in liming

materials combine with hydrogen ions in the soil to produce water (H_2O) and carbon dioxide (CO_2) reducing soil acidity as described in following equation:



The reaction neutralizes H^+ and releases Ca^{2+} and Mg^{2+} , resulting in an increase in soil pH and CEC [28,30]. Ca^{2+} and Mg^{2+} ions replace H^+ on the existing pH-dependent CEC whose magnitude changes under the influence of increased alkalinity [28,30]. After lime application, higher exchangeable Ca than Mg is observed due to higher selectivity of the colloidal negative sites for Ca^{2+} than for Mg^{2+} [28,30]. As long as the soil supplies acidity in the form of H^+ , the above reaction will continue.

The primary factor limiting plant growth in GSR was low pH combined with elevated Cu, Ni and aluminum (Al) concentrations [23,24]. Experiments on germination and early growth of grasses in GSR revealed the inhibitory effects of Cu and Ni on root growth [23]. Synergistic effect was observed where these elements enhanced each other's phytotoxic effect, resulting in total toxicity that is more than the sum of the individual toxicity [23]. On the contrary, interaction between Ni and Al was antagonistic, as Al was protecting the plants from increased Ni concentrations [23]. In Sudbury, liming application to metal contaminated soils created positive feedback loops throughout the system and has an immediate detoxifying effect [23,24,29,31]. Application of limestone to toxic and barren soils triggered immediate colonization of native plant species through germination of existing seed and incoming wind disseminated seeds [23,24]. It also depressed the adverse effects on the early root and mycorrhizal development on planted seedlings [23,24]. Therefore, liming application on metal contaminated soils in GSR became a trigger factor. Recently, the use of nanoparticles for remediation of metal contaminated sites is promising [32]. This because of their exceptional adsorption and mechanical properties combined with unique electrical property, high chemical stability and their application to a large surface area [32].

Chemical immobilization has its advantages and disadvantages. Advantages include: 1) low cost, 2) simplicity and rapidity, 3) broad spectrum of pollutants can be targeted and 4) high public acceptability [18,20,22]. The main disadvantages are: 1) difficulty in removing permanently the contaminants (temporary solutions), 2) change in soil physiochemical properties can activate pollutants, 3) only the surface layer is reclaimed and 4) permanent monitoring is required [18,20,22]. Therefore, chemical immobilization is often performed along with biological remediation. Recent studies are being conducted to develop technologies that are cost-effective and highly efficient to remediate metal contaminated sites [33]. The use of plants and microorganisms for the remediation of these contaminated sites, is considered to be a green solution [10].

2.3. Biological Remediation

Biological technique is a sustainable remediation

technology to rectify and re-establish soil natural conditions [4,34]. This technology consists of *in-situ* remediation using plants (phytoremediation) and microorganisms (bioremediation) to clean up contaminated soils and water bodies [4,18,35,36]. In many developed and developing nations, phytoremediation has been accepted widely for its potential to clean up polluted sites [4,18,34,37,38,39]. Phytostabilization, phytovolatilization and phytoextraction are the three main types of phytoremediation [18,34,35].

Phytostabilization consists in immobilization of metals in soil through absorption and accumulation by roots, or adsorption onto roots or precipitation within the root zone. This reduces metal bioavailability and migration into ground water or the food chain [18,34,35]. However, metals that are absorbed by plants get converted into volatile forms. Subsequently, they are released into the atmosphere by the process called phytovolatilization. This has been used for removal of volatile metals like Hg and selenium (Se) from polluted soils [18,35]. This technology is only suitable for volatile contaminants and is limited because it does not remove the metal but rather transfers them from one medium (soil or water) to another (air) from which they can re-enter soil and water [18,34,35].

Phytoextraction refers to the uptake of metals from soil or water by plant roots and their translocation and accumulation into any harvestable plant parts [18,34,35]. Some accumulator species possess exceptionally high metal accumulating capacity and are known as hyperaccumulators. They can survive and even thrive in heavily contaminated soils [18,35,40]. Recently, removal of metal through phytoremediation, especially by hyperaccumulators has received wide attention due to its efficacy and cost efficiency [18,32,35]. The main criteria used for hyperaccumulation classification varies according to metals, ranging from 100 mg of metal/kg dry mass of plant for Cd to 1000 mg/kg for Cu, cobalt (Co), Cr and Pb [35,40]. According to the US department of energy, to be considered as hyperaccumulators plants should have the following characteristics: 1) have high accumulating efficiency under low contaminants concentration; 2) accumulate high concentrations of contaminants, 3) accumulate different types of metals, 4) grow fast and with large biomass and 5) have pest and disease resistance ability [18,35,40]. Most of the commonly known metal hyperaccumulators belong to the *Brassicaceae* or *Fabaceae* families [34,40]. However, more than 400 plants species have been reported to be hyperaccumulators, and a considerable number of species show the capacity to accumulate two or more elements [34,40]. It should be pointed out that phytoremediation greatly depends on plant biomass yield, a key factor in determining the time required to complete land restoration [39].

Phytoremediation technology has certain advantages and disadvantages as well. Advantages include: 1) low-cost, 2) low-energy, 3) being environmentally friendly, 4) far less disruptive to the soil environment, 5) avoids excavation and is socially acceptable and 6) plants are easy to implement and maintain [35,40]. The disadvantages include: 1) time consuming due to slow growth, 2) affected by changes in climatic conditions, 3) proper disposal of plant biomass after remediation, 4) contaminants may enter soil again due to litter

formation and 5) root exudates may enhance the solubility of pollutants and consequently increase the distribution of metals into soil environment [35,40].

The main drawback of phytoremediation is the storage and accumulation of pollutants in plant materials. The remediation process slows down and often becomes inadequate when the contaminated site has multiple pollutants. The appropriate solution to these problems is to combine microbe-plant symbiosis within the plant rhizosphere or to introduce microbes as endophytes to allow degradation of pollutants within the plant tissues [35,41]. Studies have shown that microbial populations in rhizosphere is much higher than in vegetation-less soil, as plants provide essential nutrients for microorganisms [35,41,42]. Microorganisms in turn are involved in various processes which benefit plant health, growth and development [7,35,41]. Microorganisms affect metal mobility and availability by releasing chelating compounds, acidification, and changing redox potential [43,44]. The incorporation of beneficial microorganisms to seeds or into soils are seen to be beneficial to plant growth and nutrition via mechanisms including N₂ fixation, phytohormones and siderophore production, as well as the transformation of nutrient elements [45,46,47]. Thus, phytoremediation efficiency is significantly increased when plants and microorganisms are combined [10,44,48]. This approach accelerates removal of pollutants and supports high plant biomass production for bioenergy.

In the last decade, attention has been drawn to biological remediation which involves the use of microorganisms to clean up contaminated soil and water. Microorganisms cannot degrade and destroy metals, but affect the migration and transformation by changing their physical and chemical characterization [18,35,41]. Bioremediation is divided into two categories: 1) biosorption and 2) bioaccumulation. Biosorption is a passive adsorption mechanism that is fast and reversible where, metals are retained by means of physicochemical interaction (ion exchange, adsorption, complexation, precipitation and crystallization) between metals and the functional groups (carboxyl, phosphate, sulfate, phenyl, amide and hydroxyl groups involved in metal binding) present on the cell surface [35,41,49]. Both living and dead biomass can occur for biosorption because it is independent of cell metabolism. On the other hand, bioaccumulation is an active process of metal removal by living biomass which uses both intra- and extra-cellular processes [35,41,49].

Microbes mobilize metals by leaching, chelating, methylation and redox transformation [18,35,41]. Leaching is a simple and effective technology for extracting valuable metals and has potential for remediation of mining sites, treatment of mineral industrial water products, detoxification of sewage sludge and for remediation of soils and sediments contaminated with metals [18,35]. Microorganisms use metals as terminal electron acceptors or reduce them through detoxification mechanism. They then remove them from contaminated environment [35,41]. In addition, they also remove metals through mechanisms that they employ to derive energy from metals redox reaction, to deal with toxic metals through enzymatic and non-enzymatic processes [35,41]. The redox reaction takes place in soil between metal and microorganism where, microorganisms act as oxidizing agents for metal and

cause them to lose electrons. In anaerobic conditions, microbes oxidize organic contaminants by reducing electron acceptor, while in aerobic condition, oxygen acts as electron acceptor [35]. For example, microorganisms reduce the state of metals and change their solubility. Dixit et al. [35] reported that *Geobacter* species reduce uranium (U) from a soluble state (U^{6+}) to insoluble state (U^{4+}).

Several studies have shown that many microorganisms have a natural capacity to biosorb metals and use different defense systems (exclusion, compartmentalization, complex formation and synthesis of binding proteins and peptides) to reduce stress developed by toxic metals [35,41,49]. In the presence of metals, some microorganisms produce cysteine-rich metal binding proteins and peptides, such as glutathiones (GSH), phytochelatins (PCs) and metallothioneins (MTs), which can bind and sequester metal ions into biologically inactive forms [35,41,49]. In *E.coli*, expression of different metal binding proteins and peptides regulates the accumulation of Cd [35,49]. Dixit et al. [35] reported that co-expression of GSH along with PC resulted in two fold increase in Cd accumulation. Coelho et al. [49] on the other hand, demonstrated the ability of certain fungi (e.g., *Aspergillus* and *Penicillium*) and some yeasts (e.g., *Saccharomyces cerevisiae*) to remove metals from contaminated sites.

Studies have shown that metal toxicity to plants can be reduced by the use of plant growth promoting microorganisms which exert some beneficial effects on plant development [35,41]. The use of endophytes and rhizospheric microorganisms associated with degradation of pollutants in soil is promising for remediation of metal contaminated sites. *Khuyvera ascorbate*, metal resistant bacterium, has been reported to promote canola (*Brassica campestris*) growth in the presence of high concentrations of Ni [41]. Mycorrhizal fungi reduce metal toxicity to their plant host by binding metals to their cell wall or surrounding polysaccharides [18,41]. Mycorrhizae reduce the concentrations of metals therefore, reducing their hazardous effect to plants [18,41]. Yao et al. [18] reported that Cd uptake from hyphal compartment was higher in mycorrhizal than non-mycorrhizal plants and a large proportion of the increased Cd content was sequestered in roots. It was concluded that AM fungi can transport Cd from soil to plants through extraradical hyphae, but that transfer is restricted due to immobilization of metals.

The application of bioremediation on a large scale can be challenging. Bioremediation can be affected by different factors such as temperature, oxygen, moisture and pH [18,35,41,48]. There are some limitations to the application of this technology. Some microorganisms can only degrade special contaminants, and native bacterial populations might be affected if new microbes are added to the ecosystem to cleanup contamination. This might incur secondary pollutions [18,35,41,49]. Also, industrial scale application would not be of interest if the microorganism is difficult to cultivate [35,49].

3. Conclusion

Currently, there is a wide range of techniques that are

available to restore metal contaminated sites. Each technique has its own peculiarities. Land remediation using plants and bacteria is more attractive because it is the most environmentally friendly. In many situations, reclamation of metal contaminated sites require a combination of different approaches such as soil liming and land revegetation or physical removal of metals and bioremediation.

4. Summary

Anthropogenic activities such as mining, smelting, atmospheric deposition, fossil fuel combustion, wastewater irrigation, corrosion, sewage sludge, and agriculture practices have contributed significantly to soil metal contamination. Several cleanup methods have been investigated and regulatory steps have been implemented to restrict the release of metal pollutants into soils. These detoxification methods can remove metals or transform them into harmless forms. The main three types of remediation treatments discussed in this review include physical, chemical, and biological techniques. Physical remediation consists in soil replacement, thermal desorption, or electrokinetic migration. Chemical remediation technique includes all the methods involving reagents or external compounds. Recently, the use of nanoparticles for remediation of metal contaminated sites has been reported as a promising tool to restore lands. This because of their exceptional adsorption and mechanical properties combined with unique electrical property, high chemical stability and their application to a large surface area. On the other hand, biological technique is a sustainable remediation technology used to rectify and re-establish soil natural conditions. This technology consists of *in-situ* remediation using plants (phytoremediation) and microorganisms (bioremediation) to clean up contaminated soils. It is currently the most recommended strategy to remove toxic metals from ecosystems because it is usually more environmentally friendly. The most efficient strategy to reclaim metal contaminated land is to combine different remediation methods.

List of Abbreviations

SOM: Soil Organic Matter
 N: Nitrogen
 S: Sulfur
 SO₂: Sulfur Dioxide
 Hg: Mercury
 As: Arsenic
 Cu: Copper
 Zn: Zinc
 Pb: Lead
 Cd: Cadmium
 Cr: Chromium
 Ni: Nickel
 Fe: Iron
 Mn: Manganese
 P: Phosphorous
 H⁺: Hydrogen ion

K: Potassium
 Ca: Calcium
 Mg: Magnesium
 Ca (OH)₂: Calcium Hydroxide
 CaO: Calcium Oxide
 CaCoO₃: Calcium Carbonate
 GSR: Greater Sudbury Region
 CaMg (CO₃)₂: Dolomite Lime
 CEC: Cation Exchange Capacity
 H₂O: Water
 CO₂: Carbon Dioxide
 Al: Aluminum
 Se: Selenium
 Co: Cobalt
 U: Uranium
 GSH: Glutathione
 PC: Phytochelatin
 MT: Metallothionein

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Conflict of Interest

Authors declare no conflicts of interest

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