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Agro-improving method of phytoextracting heavy metal contaminated soil

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Abstract

Phytoextraction of heavy metal contaminated soils is a promising remediation technology. Till now, more than several hundreds of hyperaccumulators or non-hyperaccumulators which can be used to clean polluted soils with heavy metals have been reported. However, phytoextraction is still not extensively applied. Thus, some measurements should be taken to improve phytoremediation. This paper introduced the basic mechanisms of phytoextration, its main restrictive factors, its relationship with agricultural technology and some agricultural improvement methods. We suggested that unavailable heavy metal activation, crop breeding, seed-coating and felicitous utilization of fertilizer and water, as well as the use of two-phase planting may be important and indispensable paths for phytoextraction to be widely applied at a commercial level in the future.

Keywords: Contaminated soil; Heavy metal; Improving method; Phytoextraction

1. Introduction

Remediation methods of contaminated soils with heavy metals can be roughly classified into physical or chemical, and phytoremediation [1]. Remediation mechanisms basically consist of two fundamental principles. The first is to completely remove contaminations from polluted sites and the second is to transform these pollutants to harmless forms by using one or more engineering technologies, which mainly include excavation, separation, extraction, electrokinesis, washing, oxidation, reduction, phytoextraction, phytovolatilization, or solidification, vitrification, among others [1–4]. In order to overcome some shortcomings of physical and chemical remediation methods such as the destruction of soil-structure, secondary pollution and huge costs, phytoextraction is introduced into remediation field. Actually, with some added advantages such as environmental beautification, easy acceptance by the public and potential application to a relatively large pollution area, phytoextraction of heavy metal contaminated soils is widely considered a promising remediation technology in the future [5–7].

Phytoextraction is a phytoremediation technology, and usually implies the use of hyperaccumulators to unusually accumulate metals from polluted soils, followed by the seasonal harvesting of plant biomass – in particular their above-ground parts – until the concentration of heavy metals in the soil decreases to an acceptable level [5,8]. Although some papers documented some non-hyperaccumulators which also have some potential in remediating heavy metal contaminated soils [9–11], ultimately hyperaccumulators are still the main or at least important plants to effective phytoextraction owing to their superior accumulation ability [1,9–11].

The term hyperaccumulator was first used by Brooks et al. [12] to name plants that can accumulate more than 1000 mg/kg Ni (dry weight) in their shoots [12]. Now, any plant that can exceptionally accumulate any kind of heavy metal can be termed a hyperaccumulator. Usually, the four main characteristics of hyperaccumulating plants can be summarized as follows: (1) accumulation capacity, i.e. the minimum concentration of As,

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Pb, Cu, Ni and Co in the shoots of a hyperaccumulator should be greater than 1000 mg/kg drymass, 10,000 mg/kg Zn and Mn, 100 mg/kg Cd and 1 mg/kg Au [13]; (2) translocation capacity, i.e. the concentration of heavy metals in the shoots of a plant should be higher than that in the roots [13]; (3) tolerance capacity, i.e. a hyperaccumulator should have a high tolerance to toxic contaminants, in particular, the shoot biomass of the plants tested under experimental conditions should not decrease significantly when growing in contaminated soils [14]; (4) enrichment factor (EF) index (concentration ratio in plant to soil), i.e. EF is often higher than 1; the EF value should be at least higher than 1 when the content of a heavy metal in soils reaches its critical concentration in a hyperaccumulator [14].

Even though hyperaccumulators have shown a strong removal potential and despite the effectiveness of more than 400 hyperaccumulators having been published [15,16], phytoextraction is still not extensively used to remediate heavy metal contaminated soils as a result of practical limitations. This paper discusses some of the science mechanisms and improvable countermeasures of using agricultural technologies to improve phytoextraction in order to provide a scientific base for a wide, commercial application of this technology.

2. Main restrictive factors of phytoextraction and its relationship with agriculture

2.1. Effects of heavy metal bioavailability and site conditions on phytoextraction

Usually, heavy metal bioavailability in contaminated soils is different even for the same element and is greatly affected by soil type and meteorological conditions. In addition, there is an indispensable relationship between heavy metal bioavailability and the source of soil contamination. Generally speaking, there are two main sources of soil contamination by heavy metals, namely, natural and anthropogenic. Natural sources mainly result from weathering of various mines (which are themselves anthropogenically created) rich in heavy metals. In the presence of environmental factors such as pH, oxygen, water and heat change, weathering due to a series of complicated reactions such as dissolution–precipitation, oxidation–reduction, and adsorption–desorption is greatly accelerated. Thus the concentration of heavy metals in soils around mines is often higher than its background or basal level. Sometimes, some ores are also important sources of heavy metals in soils even though they are buried under the soil's subsurface [17]. Many cases have documented that the most fatal effects on human beings are caused by anthropogenic contamination [1]. Pollution sources resulting from anthropogenic activities mainly include mining, smelting, ore-processing, irrigation with sewage-containing heavy metals, agricultural utilization of sewage sludge, application of pesticides and chemical fertilizers, release of automobile exhausts, and a pile-up of municipal wastes. Among the above-mentioned pollution sources, mining wastes and irrigation of wastewater containing high levels of heavy metals are very prominent [1,17].

Regardless of the form of heavy metal, they will continuously react with various components of soils after having entered into them. These reactions include dissolution-precipitation, adsorption–desorption, complexation–dissociation, and oxidation–reduction. Among many important factors, soil properties such as pH, electrode potential (Eh), content and composition of colloids, and conditions of climate, hydrology and biology are the main factors affecting heavy metal formation in soil. Though the bioavailability of heavy metals in soil is complex, there are some regularities of species [18] distribution in space and time, which can be usually classified into water-soluble, exchangeable, or bound to organic matter, bound to carbonates, bound to Fe–Mn oxides and residual forms bound in a mine crystal lattice [1,17]. As shown in Fig. 1, usually, and based on the uptake by plants, heavy metals in soil can be roughly sorted into three forms including available, exchangeable and unavailable fractions [1,18–20]. Available heavy metals include free ions and chelating ions that can be easily extracted by plants. Unavailable heavy metals including residual forms are very difficult to be absorbed by a plant. Between available and unavailable lies the exchangeable fraction such as bound to organic matter, carbonates or Fe–Mn oxides, which are not partly extracted by plant. Available and unavailable speciation of heavy metals in soils is often at an equilibrium. Once bioavailable heavy metals are reduced due to plant uptake, they would be supplied from unavailable heavy metals. When bioavailable heavy metals increase owing to input from external surroundings, some bioavailable and exchangeable fractions can change into unavailable heavy metals as a result of an external disturbance or a change in environmental conditions such as plant uptake, organic chelation or the fluctuation of temperature and moisture [20]. Thus, bioavailability of heavy metals is one limiting factor of phytoextraction.

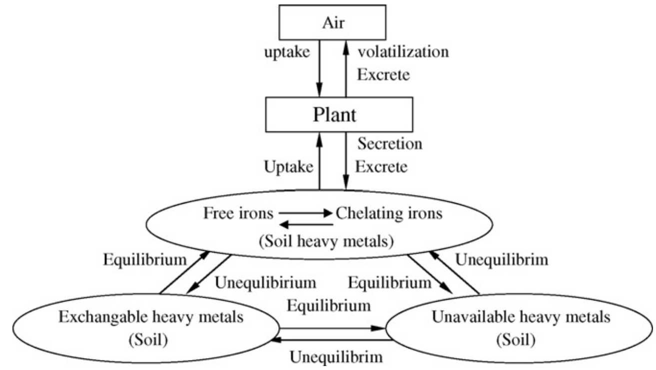

Fig. 1. Interactions among of a plant, heavy metals and soil.

Considering the main sources of soil contamination, heavy metals in industrial wastewater are basically dispersed on the surface of the soil and can be easily absorbed by plant root systems. Usually, this type of polluted soil contains low concentrations of heavy metals and is suitable to remediation by phytoextraction. As for soils contaminated by mining activities and their abandoned mineral materials, the contaminated soil layer is often deep and the concentration of heavy metals in soils is very high. Phytoextraction cannot be applied to sites in which the soil depth is so deep that the plant cannot reach contaminated soil. Furthermore, if the concentration of heavy metal in soil is too high, the plant will not survive, even though many plants have a very high tolerance to heavy metals [21]. This is because the endurance of a plant to heavy metals is not unlimited [22]. In addition, since phytoextraction is the application of plant to remove contaminants from polluted soils, contaminated sites whose environmental conditions are unsuitable for the survival of plants cannot be treated by phytoextraction either. This may be the most important restrictive factor, which cannot be overcome by phytoextraction itself. Considering the occurrence of heavy metals in soils, many of them are in an unavailable form [17]. Even if the heavy metal content in contaminated soils could be decreased to an available level after consecutively planting plant, the whole remediation process of changing unavailable into available forms would be too long and too slow [20]. It is thus necessary to take some measures to activate unavailable heavy metals to enhance the efficiency of phytoextraction.

2.2. Effects of the relationship between plant and heavy metal on phytoextraction

The ability of a plant to absorb and accumulate heavy metals is not only limited by its genotype, but also impacted by its rhizospheric microflora, by physical and chemical properties of the soil, and by the bioavailability of heavy metals in soil [23,24]. It is a universal phenomenon that a plant can absorb almost any heavy metal [25]. This is because a plant has no absolute selectivity during the absorption of nutrient substances in the soil, and there are some differences only in accumulation capacity. Some heavy metal concentrations in plants can be expressed as a percentage, some by traces in mg/kg and μg/kg, but some cannot be detected using current analytical methods [1]. The mechanisms of the active and passive uptake of heavy metals by a plant are still unclear [1].

Like animals, plants can continuously excrete redundant matter and metabolic products which are often vented as secretion and volatilization (Fig. 1). Generally speaking, there are two pathways of excretion [1,25]. The most important one is absorption by roots, and excretion of a substance or element through above-ground organs such as leaves or stems. For example, some plants extract Hg and Se from soil solution through their roots, and excrete them from their leaves. Another path is the uptake of heavy metals by leaves, and their elimination through roots. For instance, 1,2-ethylene dibromide is firstly absorbed by leaves of tobacco and radish, then excreted rapidly from roots [1]. Most heavy metals (in phytoextraction) follow the roots-to-leaves pathway, and not the other way. Usually, though heavy metals can be partially excreted by a plant, many of them integrate with some proteins or polypeptides [26–28] in a plant, so they are accumulated in some tissues and organs. Along with the growth of a plant, the contents of heavy metals accumulated in the plant gradually increase. Hyperaccumulation may take place in some special plants, the hyperaccumulators, which is one of the basic theories of phytoextraction.

The absorbing, excreting and accumulating processes of heavy metals by a plant are also dynamic. At the moment of plant growth, these processes may be at an equilibrium state, which could be broken with large changes in environmental conditions. When the concentration of bioavailable heavy metals in soils decreases owing to plant uptake, the process of transformation from unavailable into bioavailable forms will be accelerated (mostly require the proper conditions to be in place), and the accumulation of heavy metals in a plant will increase accordingly. Once the quantity of bioavailable heavy metals cannot meet the accumulation in a plant, the contents accumulated by a plant will not increase [1,22,25]. If bioavailable heavy metals from the external environment are greater than the critical concentration which a plant can accumulate maximally, the accumulation of heavy metals by the plant does not increase either. On the contrary, bioavailable heavy metals may be translated into unavailable speciation. Thus it can be seen that the accumulation of heavy metals by a plant is not unlimited. This process is also one of main restrictive factors for phytoextraction, so some strengthening measures should be taken to improve the efficiency of phytoextraction.

2.3. Relationship of agriculture with phytoextraction

Basically, the aim of agriculture insists in obtaining the highest seed or biomass like root, stem, leaf or inflorescence from crops. There are many crops in the world such as food crops corn, rice, wheat and vegetable crops cabbage, eggplant, and so on. Nearly every part of crops can be as agricultural product, for example, root of sweet potato, stem of potato, leaf of celery cabbage, inflorescence of daylily and seed of corn. In order to gain the largest quantity of agricultural product from crops, many measurements are applied such as watering, fertilizer, breeding and so on.

The technology of phytoremediation is mainly using plant to remove pollutants from heavy metal contaminated soils. Thus, plant or special plant like hyperaccumulator is the hard core of phytoextraction, which inevitably involve plant growth and propagation, especially for plant biomass. Agricultural production commits itself to how to improve crop growth and propagation so as to obtain the highest economical biomass (stem,leaf or inflorescence biomass) or seed yield. The hard cores of phytoextraction and agriculture are accordant (Table 1), i.e. plants are used by both of them. Thus, it is very important and feasible that agricultural technology is used to improve phytoextraction [1,14,18].

3. Improving measures of phytoextraction

3.1. Activating unavailable heavy metals in contaminated soils

After heavy metals enter into the soil, many of them are changed into insoluble precipitation owing to reactions with soil organic or inorganic matter, or are absorbed on the surface of soil particles. Thus it is difficult for the heavy metals to be absorbed by plants [20,29]. By taking some activating measures as discussed below, the efficiency of phytoextraction could be increased to some extent, resulting in the enhanced bioavailability of heavy metal concentrations in soil solutions.

Table 1
Relationship between agriculture and phytoextraction

Items	Agriculture	Phytoextraction
Research object	Crops like corn, rice, wheat, cabbage, eggplant, etc.	Many plants such as weed species, trees, sometimes, some crops and so on
Research aim	Obtain the highest seed yield or economical biomass (stem, leaf,etc.)	Using plant to remove heavy metals in contaminated soils
Using parts of plant	Different crops with different aim plant parts including seed, root, stem, leaf, inflorescence	Usually, stem and leaf is main organs to remove heavy metals
Common grounds	Plant growing including germination, growth and harvest Obtain the largest biomass (seed, root, stem, leaf or inflorescence) in limited time Need suitable conditions like nutrient, water, light,temperature to grow and enhance plant biomass	

Usually, the concentration of heavy metals in soil solution can be increased by decreasing the soil pH value, because the amount of H^+ will increase and the exchangeable capacity between heavy metal cations and H^+ adsorbed on the surface of soil particles will also increase after the pH decreases. Thus, a large quantity of heavy metal ions are desorbed from the surface of colloids and clay mineral particles, and then entered into soil solution. In the meantime, a decrease in pH can break the dissolution-precipitation equilibrium among heavy metal ions, and promote the release of heavy metals to soil solutions [29–31]. There are two common methods of reducing soil pH [20]. One is direct acidification, namely, concentrated sulfuric acid is diluted, and then sprayed onto the surface of soils; finally, the acid is adequately mixed with soils by some mechanical treatment such as farming tillage so that soil pH is decreased. The other is by adding a soil nutrient reagent which is made of organic fertilizers, chemical fertilizers or diluted concentrated sulfuric acid, which can be mixed with soils by scattering (by machine or manpower). The addition of soil nutrient reagents can both increase soil fertility and decrease soil pH. Certainly, the lowering of soil pH must not inhibit the growth of a plant, so acid-resistant hyperaccumulators are very useful for phytoextraction. Of course, the addition of acidification reagent has some potential environmental risks similar to those of soil chelating agents [32]. Safe operation procedures and protection measures must be taken to avoid secondary pollution, which is very important and challenging point in environmental science. In fact, after certain pH, the buffer system of the soil consisting of carbonate, aluminium etc. will deteriorate, so that a regeneration of the soil will become difficult. All over the world nations deal with the problem how to prevent acidification of the land. Therefore, it would not be wise to acidify the soil with too many times or too heavy. Moreover, a decrease in soil pH is not always useful for the activation of all heavy metals [25]. As for example, is an exception. In many cases, As content in soil solution increases with pH, because As often occurs as $AsO_4{}^{3-}$ or $AsO_3{}^{3-}$. When soil pH increases, the cations absorbed to the surface of soil particles will be reduced and the absorption of As will also decrease; subsequently As in soil solution can increase. The addition of alkaline materials such as quick lime can effectively increase soil pH [1].

The rise of Eh can also increase the concentration of heavy metals in soil solutions [1,20]. As for Cr, when Eh is enhanced, Cr^{3+} can be oxidized to Cr^{6+} which has strong water-solubility, so the Cr ion content in soil solution increases. Likewise, $AsO_4{}^{2-}$ can also be reduced to $AsO_3{}^{3-}$ and increase the solubility of As. But for slightly soluble sulfides which can firmly immobilize heavy metals, an increase in Eh will enhance the concentration of heavy metals in soil solutions, because the sulfides will be unstable and be oxidized under such conditions, so heavy metals can be released. Others like soils rich in Fe or Mn oxides, a decrease in Eh will make them dissolve only partially, so the heavy metal ions absorbed by or co-precipitated with them will be released. However, Eh adjustment is very complicated. For example, when Eh drops at the beginning, there is a mobilization. But when $SO_4{}^{2-}$ is reduced to S, the heavy metals are very tightly bound so that phytoextraction is very difficult. Similar when Eh is increased with the oxidation of Mn–Fe oxides. The method of regulating soil Eh can be conducted by farming techniques such as solar drying, intermittent irrigation of paddy fields, or crop rotation between paddy and dry fields [33]. Moreover an increase in soil organic material can also decrease soil Eh.

Chelating reagents can accelerate the release of heavy metals bound to soil solids [34], because they can break the equilibrium between the soil liquid and solid phases of heavy metals. Sequentially the intensity of soil adsorbed to heavy metal-chelating reagents is decreased, thus accelerating the transformation of equilibrium to desorption; most heavy metals are then released into the soil solution, whose concentration of heavy metals increases before a new equilibrium is attained [35]. However, the use of chelating agents has also some potential risks, i.e. mainly eluviation and secondary pollution of groundwater by heavy metals when no extraction by plants occurs. Moreover, residual chelating reagents may also result in a new source of pollution and activation of other metals by using some chemical inducers aimed heavy metals [32] (which would be removed or cleaned). Therefore, an environmental risk assessment should be made to assure environmental safety before some chelating reagents are used. Such chelating reagents would be ideal if they could be degraded naturally and toxic chemicals should be avoided as much as possible, e.g., the use of cyanides can induce the desorption of Au.

3.2. Application of crop breeding technology

In order to gain maximal resources from crops such as food, vegetables, oil, fodder, fiber, beverage and rubber, humans not only endlessly improve farm-culture technology, but have also optimized desired crop traits by advanced breeding methods. Most documented hyperaccumulators are wild plants [35] whose biological behaviors are relatively unknown by the public [36]. There are almost no ready-made modes of culture or perfectly rounded breeding technologies. Despite this, crop cultivars derive from wild plants, whose many disadvantageous properties can be reconstructed into available resources through continuously choosing and reuniting plant genes, i.e. by crop breeding technologies. By using system breeding technology (continuously choosing) some hyperaccumulator species with huge stems and leaf biomass can be selected [36], which is beneficial to the accumulation of heavy metals by plants [1]. In agricultural science, this point is relatively easy to achieve compared with practices to get a higher seed biomass [36]. Usually, the maturation period of wild plants is inconsistent and the growing period is too long [37]. By using crop breeding, a consistently ripe stage and short growing period of every hyperaccumulator plant can be obtained, then harvested biomass will increase according to season change in contaminated soils. In other words, when harvesting a hyperaccumulator, one can get accordant characteristics and higher individual plant biomass in a relatively shorter period. This implies a higher biomass within the same harvest period. Thus, phytoextraction efficiency can be increased [1,38]. Longer dormancy and easy grain drop of wild plant seeds [37] are disadvantageous and limit the application of hyperaccumulators in phytoextraction of contaminated soil and thus reduce phytoremediation efficiency. Because a longer dormancy of seeds will require much time to grow into a hyperaccumulator, and since easy grain drop makes it difficult to collect seed on a large scale, the subsequent efficiency of phytoextraction will decrease. By crop breeding, these shortcomings of hyperaccumulators can be easy reduced [36]. Phytoextraction is thus also directly improved. It may be feasible to use crop breeding to improve the characteristics of hyperaccumulators, and construct corresponding remediation technologies.

3.3. Application of seed-coating technology

Seeds of documented hyperaccumulators are usually small, and whose diameter or length is only several mm or even μm [36]. Such small seeds are not only inconvenient for sowing, but also difficult to transplant. Seed-coating is the use of a layer of materials commonly containing some fertilizers and pesticides to enclose them, which can improve germination, and prevent and cure seedling diseases and pests, and dispel mice [18]. Moreover, increased seed volume is convenient for mechanized sowing. Seed-coating is an indispensable technology for phytoextraction applied at a large, commercial scale. Furthermore, the joint remediation of hyperaccumultors with microbes which can largely absorb heavy metals is one of the important measures to efficiently improve phytoextraction. Abou-Shanab et al. [39] studied the effect of some bacterial strains on hyperaccumulator *Alyssum murale* accumulating Ni [39]. The results showed that the nine bacterial strains *Microbacterium oxydans* AY509223, *Rhizobium galegae* AY509213, *Microbacterium oxydans* AY509219, *Clavibacter xyli* AY509236, *Acidovorax avenue* AY512827, *Microbacterium arabinogalactanolyticum* AY509225, *M. oxydans* AY509222, *M. arabinogalactanolyticum* AY509226 and *M. oxydans* AY509221 significantly increased Ni extraction to some extent. Compared with uninoculated seeds, *M. oxydans* AY509223 significantly increased Ni uptake of *A. murale* grown in the low, medium, and high soils by 36.1%, 39.3%, and 27.7%, respectively. In order to examine the promotion of bacterial strains to rape (Brassica napus) extracting Cd, Sheng et al. [40] isolated a large of bacteria from heavy metal-polluted soil in Nanjing, China [40]. The results indicated that some Cd-resistance bacterial strains significantly increased water-soluble Cd concentration from Cd carbonate medium. Compared with non-inoculated control, Cd content extracted by this plant increased from 16% to 74%. Whiting et al. (2001) studied the possible mechanism of bacteria remediation heavy metal combined with plant by using Zn-hyperaccumulator *Thlaspi caerulescens* [41]. They speculated that bacteria could produce zinc-chelating metallophores, which might also be taken up by the plant. Generally speaking, microbial reagents can obviously enhance the infection rate of microbes close to plant roots. Practically speaking specific symbiotic microbes are applied to a treatment, and then combined with plants by scattering the mixture near the roots, using seed-coating technology.

3.4. Felicitous utilization of fertilizers and water

The above-ground biomass of a plant is one of the important factors impacting the efficiency of phytoextraction, which can be increased if the above-ground biomass of a hyperaccumulator is enhanced as much as possible [1]. Fertilization and irrigation are two important factors that promote the growth of a plant. Hamlin and Barker [38] determined the effects of nitrate fertilizers on growth and Zn accumulation in Indian mustard (*Brassica juncea* Czern.) [38]. The results showed that nitrate nutrient supply enhanced shoot biomass and stimulated Zn accumulation. When plants received 10% of the total N as NH^{4+} and 90% as NO^{3-}, the Zn phytoextraction potential of Indian mustard was maximized. Chou et al. [42] examined the effects of K fertilizer on plants extracting Cs^{137} [42]. The results revealed that rape (*B. campestris* L.) exhibited the highest production of above-ground parts and had the highest Cs^{137} transfer factor.

However, a plant may also suffer from over-watering and over-fertilization. Moreover, application of water and fertilizers may also result in increasing the diffusion of heavy metals in soils. Understanding the requirements of water and fertilizers by a plant, in view of the fact that seedling and flowering stages are the most sensitive periods, their appropriate application will affect the level of harvestable hyperaccumulator biomass [36,43].

3.5. Necessary measures to shorten the cycle of phytoextraction

Environmental factors such as sunlight, temperature, water, air, and heat can greatly affect plant growth, which are basic life factors in agricultural science [36]. According to the reaction of a plant to environmental conditions, the growth duration of a hyperaccumulator could be shortened as far as possible to decrease the cycle of phytoextraction, e.g. a greenhouse can accelerate the growth of a plant such as some outdoor plants that cannot survive well at low temperatures. Shade devices may promote the growth of a plant, mirroring faded light conditions. An increase in carbon dioxide concentration can enhance the plant's photosynthesis [44]. Transplanting can also shorten the cycle of phytoextraction, and hyperaccumulator seedlings that are kept under cover can be transplanted into contaminated soils once field conditions are fit for growth, shortening the remediation time from sowing to seedling [37]. Furthermore, taking advantage of the fact that for some hyperaccumulators the growth duration from seedling-transplantation to the flowering phase is short, and that the concentration of heavy metals accumulated in their shoots at the flowering phase is high, the efficiency of phytoextraction can be greatly improved using the method of two-phase planting, i.e. harvesting the hyperaccumulator at its flowering phase, then transplanting its seedlings again [14,45]. However, some of suggested measure costs may be very expensive. Economical evaluation should be considered to assure phytoextraction of heavy metal contaminated soils as cost efficient as possible.

4. Conclusions

In order to solve the main restrictive factors of phytoextraction, such as activation of unavailable heavy metals in soils, the finite accumulation of targeted metals by hyperaccumulators, and the long duration of remediation, some strengthening measures should be taken. No matter how strong the remediative ability of a hyperaccumulator, in the final analysis, the successful use of phytoextraction needs necessary regulation technology and useful strengthening measures to enhance its remediation efficiency. Thus it can be seen that phytoextraction is intimately tied-in with advanced agricultural technologies. Crop breeding can improve the biological characteristics of plants and enhance their remediation potential. Seed-coating technology can promote the germination of hyperaccumulator seeds in pollution sites. The use of crop cultural technologies should accelerate the growth, increase biomass, and shorten the remediation time of a hyperaccumulator. In short, the adoption of advanced agricultural technologies may be a short-cut to phytoextraction applied at a commercially large scale.

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