



Using plants to remediate or manage metal-polluted soils: an overview on the current state of phytotechnologies

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ABSTRACT. Soil contamination by metals threatens both the environment and human health and hence requires remedial actions. The conventional approach of removing polluted soils and replacing them with clean soils (excavation) is very costly for low-value sites and not feasible on a large scale. In this scenario, phytoremediation emerged as a promising cost-effective and environmentally-friendly technology to render metals less bioavailable (phytostabilization) or clean up metal-polluted soils (phytoextraction). Phytostabilization has demonstrable successes in mining sites and brownfields. On the other hand, phytoextraction still has few examples of successful applications. Either by using hyperaccumulating plants or high biomass plants induced to accumulate metals through chelator addition to the soil, major phytoextraction bottlenecks remain, mainly the extended time frame to remediation and lack of revenue from the land during the process. Due to these drawbacks, phytomanagement has been proposed to provide economic, environmental, and social benefits until the contaminated site returns to productive usage. Here, we review the evolution, promises, and limitations of these phytotechnologies. Despite the lack of commercial phytoextraction operations, there have been significant advances in understanding phytotechnologies' main constraints. Further investigation on new plant species, especially in the tropics, and soil amendments can potentially provide the basis to transform phytoextraction into an operational metal clean-up technology in the future. However, at the current state of the art, phytotechnology is moving the focus from remediation technologies to pollution attenuation and palliative cares.

Keywords: phytoremediation; phytoextraction; soil pollution; hyperaccumulators.

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Introduction

Industrialization and urbanization of developed and developing countries have enormously increased humankind's demand for metals. These continuous processes and hence mining activities to meet this growing demand led to the unfortunate side-effect of soil pollution. Agriculture can also pollute soils with metals through the excessive application of pesticides, fertilizers, manure, and sewage sludge with high heavy metal content. As a result, soil pollution has been identified as the third most important threat to soil functions in Europe and Eurasia, fourth in North Africa, fifth in Asia, seventh in the Northwest Pacific, eighth in North America, and ninth in sub-Saharan Africa and Latin America (Food and Agriculture Organization of the United Nations [FAO], 2015). Sixteen percent of all Chinese soils are categorized as polluted, while 3 million potentially polluted sites were identified in Europe, and more than 1,300 sites in the USA are included in the Superfund National Priority List (Eugenio, McLaughlin, & Pennock, 2018). Data on the number of potentially polluted sites in Brazil are inexistent. However, several studies have shown that agricultural soils and urban, industrial, and natural sites in the country have been seriously contaminated through human activities (Freitas, Nascimento, Souza, & Silva, 2013; Silva, Nascimento, Araújo, Silva, & Silva, 2016; Silva, Silva, Araújo, & Nascimento, 2017; Araújo, Biondi, Nascimento, Silva, & Alvarez, 2019). Thus, soil pollution poses a serious long-term threat to human health and the environment that requires affordable and sustainable solutions to reduce risk to an acceptable level.

The most common reasons for soil remedial action are: (i) unacceptable risk as suggested by a risk assessment; (ii) direct evidence of human or ecological harm; and (iii) regulatory threshold values for metal concentration in the soil are exceeded (Pierzynski, Sims, & Vance, 2005). Threshold values for metals in the

context of soil protection in Brazil are established on the national level by The National Environment Commission (CONAMA). According to this regulation (Resolução n. 420, 2009), metal concentrations in soil exceeding the Intervention Value (IV) indicate that soil remediation is necessary or mandatory. However, the cost of remediation versus addressing other significant social issues prevalent in developing countries such as Brazil makes the funds for site remediation scarce. In this competitive scenario, lower-cost strategies for soil remediation are much needed.

Since the conventional approach of removing polluted soils and replacing them with clean soils is very costly, not feasible at a large scale, and has low public acceptance, alternative methods that use plants to clean up contaminated sites, the so-called phytoremediation, have been developed. This group of environmentally-friendly technologies has been regarded as a promising tool to remediate metal-polluted soils (Nascimento, Amarasiriwardena, & Xing, 2006; Marques, Rangel, & Castro, 2009; Šuman, Uhlík, Viktorová, & Macek, 2018). However, examples of successful application of phytoremediation either in field trials or commercial operations are still scarce.

The main bottlenecks that hinder the further development of phytoremediation are the low efficiency of metal removed per unit of land and prohibitively long clean-up times (Evangelou, Conesa, Robinson, & Schulín, 2012; Freitas, Nascimento, & Silva, 2014; Silva et al., 2017). Given such limitations have not yet been overcome, phytomanagement was developed as an alternative approach. Phytomanagement is defined as the use of plants to control and mitigate risks arising from soil pollution while making profitable and sustainable use of the contaminated land by producing marketable biomass (Robinson, Bañuelos, Conesa, Evangelou, & Schulín, 2009; Bañuelos & Dhillon 2011; Evangelou et al., 2012). This review discusses the parallel evolution of phytoremediation and phytomanagement from their early beginnings to their current status. The main objectives are to address the main limitations that prevent these technologies from becoming widely applicable and to point towards future research and developments to overcome such drawbacks.

Phytoremediation: still a promising tool for remediating contaminated land?

Phytoremediation is an umbrella term that includes: i) phytoextraction (growing plants to concentrate metals in shoots for removal from the site); ii) phytostabilization (the use of plants to convert metals into less bioavailable or mobile forms, so they no longer pose a risk to the environment); and iii) phytovolatilization (a process in which plants uptake metals from soil and release them as volatile form into the atmosphere) (Chaney et al., 1997; Ernst, 2005; Nascimento et al., 2006). Phytovolatilization is intended for a limited number of sites as it applies to metals that exist in methylated, volatile forms, i.e., mercury (Hg), selenium (Se), and arsenic (As), and has shown the greatest promise with Se (Bañuelos & Dhillon, 2011; Schiavon & Pilon-Smits, 2017). The most employed and studied phytoremediation techniques are phytoextraction and phytostabilization (Figure 1).

Phytoextraction relies on the use of plants to uptake metals from soil and transfers them to aerial parts. It aims to reduce metal concentrations in contaminated soils to regulatory levels within a realistic time frame, say < 25 years; otherwise, phytoextraction cannot compete with the traditional, non-plant-based technologies (Nascimento et al., 2006; Robinson et al., 2009). Two approaches have been tested to reach this goal: a) natural phytoextraction, in which hyperaccumulating plants with exceptional natural metal accumulation ability are used to remove metals from the soil, and b) chemically assisted-phytoextraction, i.e., the utilization of high-biomass crop plants induced to accumulate metals through the application of chelators in the soil. Both approaches have pros-and-cons (Nascimento et al., 2006), but there is still a scarcity of examples of their potential at a large scale for the clean-up of metal-polluted soils; therefore, uncertainties over the longer-term effectiveness of phytoextraction persist (Dickinson, Baker, Doronila, Laidlaw, & Reeves, 2009).

Natural phytoextraction relies on a group of plants with exceptional ability to accumulate metals in harvestable parts, when growing in their natural habitat. Therefore, plants growing in hydroponics and spiked or chelator-treated soils are not included in the definition (Reeves et al., 2017). Metal hyperaccumulation is a rare phenomenon in nature whereby plants can accumulate metal or metalloid shoot concentrations hundreds to thousands of times higher than most other plant species. The threshold concentration that identifies the phenomenon of hyperaccumulation varies with the metal considered. Plants with more than 1,000 mg kg⁻¹ of nickel (Ni) in the leaf dry matter are considered hyperaccumulators of the element. For cobalt (Co), chromium (Cr), and copper (Cu), the minimum concentration is 300 mg kg⁻¹; zinc (Zn) and manganese (Mn) thresholds are 3,000 and 10,000 mg kg⁻¹, respectively (Ent, Baker, Reeves, Pollard, & Schat, 2013; Ent et al., 2015; Reeves et al., 2017).

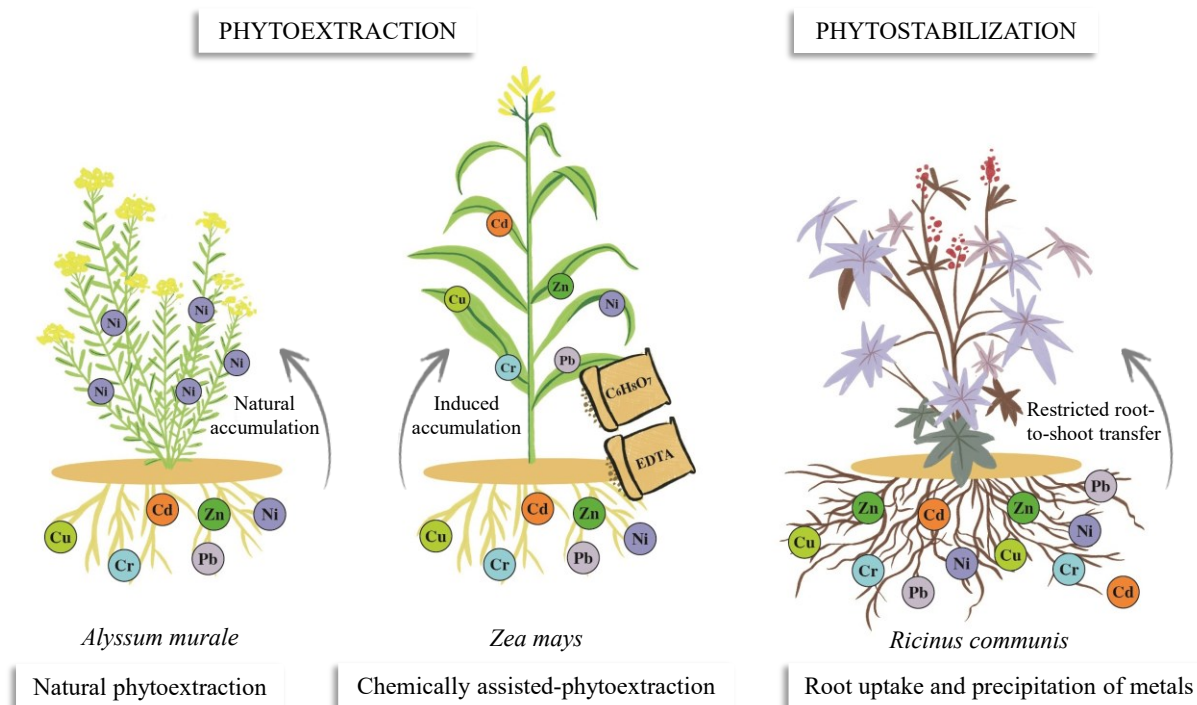


Figure 1. Overview of the phytoextraction (a) and phytostabilization (b) techniques.

Phytoextraction

Studies on Brazilian hyperaccumulators are still incipient. R. D. Reeves, A. J. M. Baker and R. R. Brooks carried out the first extensive plant survey and analyses in the ultramafic outcrops of Goiás State, Brazil, in the late '80s, in which dozens of Ni hyperaccumulators were identified (Brooks, Reeves, Baker, Rizzo, & Diaz Ferreira, 1990). Following a new plant collection in early 2005, Reeves et al. (2017) found notable new Ni hyperaccumulators, including *Justicia lanstykii* and *Lippia lupulina* (Figure 2). However, scientific reports on how the soil and plant management practices affect the ability of such species of extracting Ni (and other metals) are still lacking.

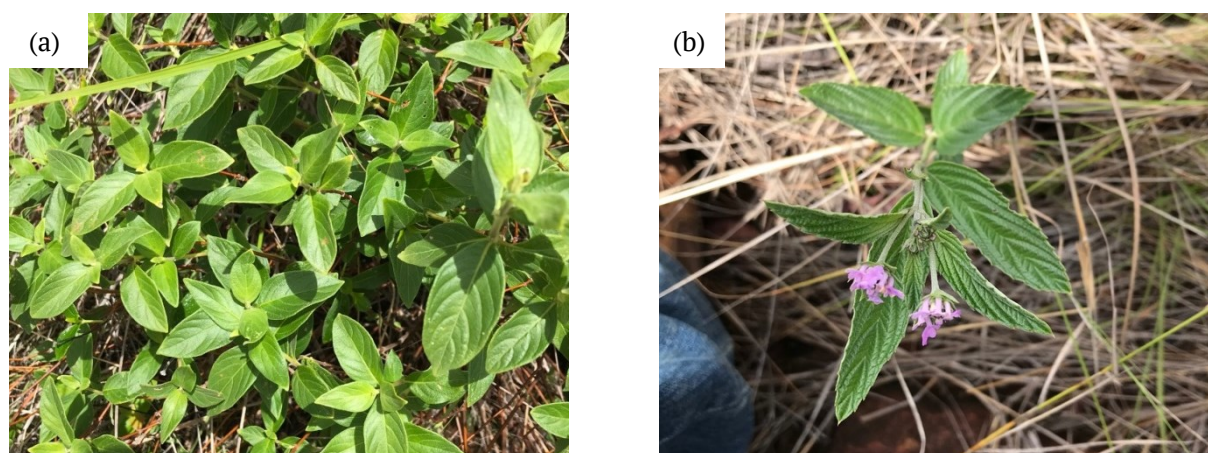


Figure 2. Nickel hyperaccumulators *Justicia lanstykii* (a) and *Lippia lupulina* (b) growing on an ultramafic outcrop in Niquelândia, Goiás State, Brazil. Photos: Clístenes Williams Araújo do Nascimento.

A few hyperaccumulators, notably *Alyssum murale* in Ni agromining from ultramafic soils (Bani, Echevarria, Sulçe, & Morel, 2015a; Bani et al., 2015b; Chaney & Baklanov 2017; Cerdeira-Pérez et al., 2019), have great potential for commercial operations. As the primary goal of agromining is not to remediate soils but rather a profitability, it is out of the scope of the present review. Outstanding reviews on this topic are available elsewhere (Ent et al., 2015; Nkrumah et al., 2016). It is clear from the literature to date that the use of hyperaccumulators to reduce the total concentration of metals to below threshold values has not been much convincing. Jacobs, Drouet, Sterckeman, and Noret (2017) showed the potential of using *Noccaea*

caerulescens, a Zn, Ni, and cadmium (Cd) hyperaccumulator for both Cd and Zn remediation of moderately contaminated soils once sufficient biomass yield would be reached; however, Cu and lead (Pb) possibly hampered plant development and could not be phytoextracted at a reasonable time frame.

The lack of success is generally attributed to most hyperaccumulators' low biomass and, therefore, the low net removal of metals from the contaminated land. The fact that most contaminated soils are multi-metal contaminated, while most hyperaccumulators are not tolerant to several metals, is also a limitation (Jacobs et al., 2017). For example, *N. caerulescens* can hyperaccumulate Ni, but it was severely affected by high Mn concentrations in the soil solution (Nascimento, Hesterberg, & Tappero, 2020a). Field studies on the performance of hyperaccumulators in reducing metal soil concentrations over successive croppings are scarce (Simmons et al., 2014; Tlustoš, Břendová, Száková, Najmanová, & Koubová, 2016) in comparison to pot experiments. Long-term field trials are crucial to assess how the decreasing metal availability with successive cropping and soil characteristics affect biomass yield and phytoextraction efficiency.

Chemically assisted-phytoextraction aims to overcome the slow-growing and low biomass yield of hyperaccumulators using high biomass crops induced to uptake metals from soils and transfer them to shoots through the application of chelators to the soil (Nascimento et al., 2006). The first results of Pb assisted-phytoextraction using EDTA were remarkable, with plants accumulating over 1 % of the metal in aerial parts (Blaylock et al., 1997; Huang, Chen, Berti, & Cunningham, 1997; Vassil, Kapulnik, Raskin, & Salt, 1998). It seemed that phytoextraction assisted by chelators was technically feasible and should be in commercial use within few years. However, synthetic chelators such as EDTA were soon shown to have a slow degradation rate and high persistence in the soil, which increased the metal leaching risk to unacceptably high levels (Chen, Li, & Shen, 2004; Freitas, Nascimento, Biondi, Silva, & Souza, 2009; Freitas & Nascimento 2016). Consequently, EDTA is no more considered to assist soil phytoextraction, and a search for environmentally-friendly chelators that could also induce the uptake of metals from contaminated soils started.

Low-molecular-weight organic acids (LMWOAs) can be alternatives to synthetic chelators for phytoextraction of metals (Nascimento et al., 2006; Arwidsson et al., 2010; Freitas et al., 2013). Unlike synthetic chelating agents, LMWOAs are quickly degraded in soil, significantly reducing the potential risk of groundwater contamination. However, only few works tested LMWOAs in field conditions. Freitas et al. (2013) showed that the use of citric acid at 40 mmol kg⁻¹ applied to an automobile battery waste polluted site effectively solubilized Pb from the soil and induced its uptake by maize. The time frame for reducing the soil Pb concentration below the regulatory level was shortened from 85 to 19 years when citric acid was used. On the other hand, Braud, Gaudin, Hazotte, Guern, and Lebeau (2019) found that applying 5 mmol kg⁻¹ of citric acid increased the buckwheat Pb extraction rate but not enough to efficiently remove Pb from a moderately contaminated soil. The calculated phytoremediation period would be 166 years, which makes the practice unfeasible.

It is important to point out that phytoextraction's estimated time depends on several factors, including plant species, the citric acid rate applied to the soil, metal considered, and total metal concentration in the soil being remediated. Phytoextraction is not viable for highly contaminated soils and has increased efficiency for mobile metals, for example, Cd compared to Pb (Silva et al., 2017). Using low citric acid rates (< 30 mmol kg⁻¹) also limits the phytoextraction process owing to insufficient metal mobilization from the soil (McGrath et al., 2006; Braud et al., 2019; Nascimento et al., 2020a), and this is the probable reason for unsuccessful results in some trials. Food grade citric acid could be used to make assisted-phytoextraction economically viable (Freitas & Nascimento 2016). A new approach of combining natural phytoextraction (hyperaccumulators) with the use of citric acid has also been proposed (Nascimento et al., 2020a; Nascimento, Hesterberg, Tappero, Nicholas, & Silva, 2020b), but field studies are needed to confirm the positive results obtained in controlled conditions.

Phytostabilization

Phytostabilization assisted by soil amendments such as phosphate fertilizers, lime, and organic matter has been shown to significantly decrease the bioavailability and leaching of metals, besides reducing wind blow and runoff of metal-contaminated soil particles, with beneficial effects to the environment and protection of potential receptors of the contaminant (Vangronsveld et al., 2009; Dickinson et al., 2009; Ali, Khan, & Sajad, 2013). Plants and the resultant root system can also provide litter through leaf fall and beneficial changes that include soil aggregation and metal binding (Pulford & Watson, 2003). Thus, phytostabilization primarily aims to reduce exposure to metals and prevent the risks associated with contaminated sites. In this scenario, the durability and efficiency of the stabilization process is crucial. Epelde, Burges, Mijangos, and Garbisu (2014)

demonstrated that chemical stabilization with sewage sludge but with no plants was only partly successful: it did improve soil quality and decreased Cd availability, but it did not reduce Pb and Zn bioavailable contents in the soil.

Several field trials showed the successful application of phytostabilization to metal contaminated sites, primarily mine tailings and brownfields (Johansson, Xydas, Messios, Stoltz, & Greger, 2005; French, Dickinson, & Putwain, 2006; Santini & Fey, 2013; Pardo, Martínez-Fernández, Clemente, Walker, & Bernal, 2013; Gil-Loaiza et al., 2016; Zgorelec, Bilandzija, Knez, Galic, & Zuzul, 2020). For instance, compost amendment allowed the establishment and sustained growth of a mix of native species on mine tailings over four years, demonstrating feasibility for phytostabilization (Gil-Loaiza et al., 2016). French et al. (2006) demonstrated that phytoextraction with *Salix*, *Populus*, and *Alnus* could reduce contamination hotspots of more mobile metals such as Cd and Zn within the crops' life cycle while phytostabilizing other less mobile metals (As, Pb, Cu, and Ni).

Phytostabilization is not a cleanup technology but rather a way of stabilizing metals in the soil to prevent further spreading and transfer into the food chain. Vangronsveld et al. (2009) highlighted that monitoring the contaminants must be part of the management when phytostabilization is used as a remediation tool. Such monitoring should include chemical (metal soil bioavailability and sequential extraction) and biological (metal plant uptake, microbial and ecotoxicological essays) tests.

In selecting plant species for phytostabilization, it should be observed that a high foliar concentration of metals is unwanted, unlike phytoextraction. However, metal removal can sometimes promote a valorization of the biomass. For instance, by producing Zn-enriched biomass that could be used as fodder (Fassler et al., 2010) or Se- and Boron-rich plants that could be used as organic fertilizers for soils deficient in these elements (Robinson, Green, Chancerel, Mills, & Clothier, 2007; Bañuelos, Arroyo, Dangi, & Zambrano, 2016). The use of bioenergy crops in phytostabilization is also an opportunity to enhance the biomass value (Melo, Guilherme, Nascimento, & Penha, 2012; Nascimento & Marques, 2018), making a strong case for phytoremediation by generating revenue in the land under remediation.

Phytomanagement: establishing a new paradigm?

Phytomanagement emerged as an attempt to solve a significant phytoextraction limitation, i.e., the lack of revenue from the land during the extended time frame required to cleanup (Evangelou et al., 2012; Burges, Alkorta, Epelde, & Garbisu, 2018) by focusing on the production of valuable plant biomass or integration with other ecosystem services. Phytomanagement includes synergies with biomass energy crops, biodiversity, watershed management, and protection from erosion, carbon sequestration, and soil quality (Regvar, Vogel-Mikuš, Kugonič, Turk, & Batič, 2006; Dickinson et al., 2009). Phytomanagement is not an actual remediation strategy but can provide a range of economic, environmental, and social benefits until the contaminated site returns to productive usage (Cundy et al., 2016; Burges et al., 2018).

To grow profitable crops to minimize the environmental risks in metal contaminated soils while guaranteeing a profit to economically support the remediation process itself or provide additional revenues for the landowner are the main objectives of phytomanagement. For instance, castor bean (*Ricinus communis*) had good development and restricted metal translocation to shoots in soils contaminated with Pb, As, Cd, and Zn surrounding an abandoned Pb smelting plant in Santo Amaro, Brazil (Silva et al., 2017). Given that castor bean oil is inedible, therefore with negligible risk to human or animal contamination, growing the crop in these marginal lands may be an alternative for an economic return resulting from biofuel production. *Jatropha curcas*, another oilseed crop, is also a viable alternative to revegetate sites contaminated with these metals in tropical regions (Marques & Nascimento, 2013; Nascimento & Marques, 2018). The cultivation of aromatic grasses (*Chrysopogon zizanioides*, *Cymbopogon citratus*, and *Cymbopogon winterianus*) has been proposed as an income source for the local population in sites affected by the Fundão tailing dam rupture in Minas Gerais State, Brazil (Zago, Dores, & Watts, 2019).

Due to the large biomass yield and extensive root system, several tree species have been recommended for phytomanagement approaches (Martinez-Oró, Parraga-Aguado, Querejeta, & Conesa, 2017; Wan, Lei, Chen, Tan, & Yang, 2017; Chalot et al., 2020). Growing fast-growing trees that yield a high quantity of biomass can bring significant economic benefits to the remediation of contaminated lands. However, the accumulation of metals in different parts of the tree (roots, stems – wood and bark tissues –, and leaves) must be considered to maximize the biomass valorization (Leclercq-Dransart et al., 2019). The clean, free-metal biomass can be

useful for plant-based industrial processes such as biomaterials and bioenergy, while metal-enriched biomass could be exploited in eco-catalysis processes (Ciadamidaro et al., 2019). Due to the higher metal concentration in bark tissues of poplar trees than the wood and the higher proportion of bark in branches compared with the wood, Chalot et al. (2020) recommended that only stem wood be harvested.

Agricultural areas affected by excessive soil metal concentrations are also suitable targets for phytomanagement. Meers et al. (2010) proposed using energy maize to reduce environmental risks and generate an alternative income for local farmers in the Campine region along the Dutch-Belgian border, which had roughly 700 km² contaminated with Cd, Zn, and Pb by atmospheric deposition from smelter activities. They found that the shoot metal concentration was too high for use as fodder but acceptable for feedstock for anaerobic digestion. Besides, it was calculated that energy maize cultivation could yield 30,000-42,000 kW of renewable energy per hectare, which would imply a reduction of up to 21 t ha⁻¹ year⁻¹ CO₂ compared to coal-powered power plants.

The cost-benefit during remediation of 700 ha of soil contaminated with Pb, As, and Cd by metal-enriched flooding water from the Beishan Pb-Zn mine, China, was assessed by Wan et al. (2017). The authors demonstrated that phytoremediation benefits could offset the project costs in less than seven years with the planting of cash crops (sugarcane and mulberry tree) intercropped with the As and Pb hyperaccumulators *Pteris vitata* and *Sedum alfredii*. One of the main reasons for the remediation project's success was the relatively low concentration of metals in the soil (on average, 36.0; 0.3; and 350.0 mg kg⁻¹ of As, Cd, and Pb, respectively) across a large area. In this particular situation, phytoremediation (or phytomanagement) seems to be a suitable and economically viable approach.

Conclusion

The extensive efforts on phytoremediation research in the last 30 years allowed for tremendous gains in understanding how plants can remediate or attenuate soil pollution. Phytostabilization has found a practical and commercial application to restrain the spread of metal contamination in industrial and urban sites. On the other hand, phytoextraction still lacks convincing field data and long-term operations to become an alternative for conventional remediation techniques. The main phytoextraction limitations are the low biomass yield of most hyperaccumulators and environmental risks or inefficiency of chelators used to induce accumulation in high biomass plants. Phytoextraction can only be successful if the time frame required for remediation is dramatically decreased from several decades to less than 20-25 years. In situations where this goal is not achievable, phytoextraction could be combined with sustainable and profitable site usage through biomass valorization. Such an approach, named phytomanagement, can overcome phytoextraction's main drawback (the extended remediation time) while gradually decreasing the soil metal concentration over time, attenuating the associated environmental risks. At the current state of phytotechnologies development, while phytostabilization and phytomanagement have a more promising future ahead, there is a need for phytoextraction experimentation at full-scale field operations to prove (or not) the efficiency of this cleanup technology.

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