The Metal Removal Capability of Endemic Chilean Species

Subjects: Engineering, Environmental
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Among the species studied, *Cistanthe grandiflora* and *Puya berteroniana* can accumulate Cr and Zn but not translocate them, showing their potential to phytostabilize these elements, obtaining a bioconcentration factor for Zn close to 1.2 for both species, 1.5 in the case of *Cistanthe grandiflora* for Cr, and 1.7 for *Puya berteroniana* for the same metal. *Oxalis gigantea* only showed the ability to concentrate Zn in the roots, with a *BCF* close to 1.2, similar to the value obtained with the other species. In the case of Ni, its bioconcentration factor was under 1 in all species.

Keywords: mine tailings ; endemic plants ; metals

1. Introduction

Mining activities are carried out in the northern and central zones of Chile. For a long time, due to the lack of regulations, mine tailings were stored without major environmental consideration; therefore, at present, several hills of these residues without owners are located near population centers. The tailings are made up of fine-grain material and a variable content of water, which facilitate the occurrence of chemical reactions, the dissolution of toxic substances, and the generation of sulfuric acid, with the inherent risk of soil and groundwater contamination [1][2]. This holds special relevance for Chile, where there are 173 abandoned mine tailing impoundments, and nearly 40 of these have unknown owners [3]. Many of these impoundments are located between the Region of Tarapacá and the Region of Valparaíso in the vicinity of towns, which is further worsened by the fact that Chile has no standards for soil pollution [3].

One major point of concern for environmental and public health is contamination by mining activities [1], where the negative effects of heavy metal contamination have been observed in the surrounding water, soil, and air environments of the mine sites [4][5][6]. In the case of abandoned mine tailing impoundments, negative environmental impacts have been identified, including an increase in the concentration of metals in agricultural soils, sediments, animals, human beings, and plants [7][8][9][10][11][12][13][14][15]. Among the various heavy metals, chromium (Cr) is widely present in soils, with an average concentration of 60 mg kg^{-1} [16]. Chromium is a nonessential element for plants, and it produces toxic effects in most of them at 5 mg kg^{-1}, with a normal concentration in plants being less than 1 mg kg^{-1} [17]. In the case of zinc (Zn), it is an essential nutrient for plants, with a maximum value in soils for residential use of 200 mg kg^{-1} [18]. For crops, the required Zn concentration is between 15 and 20 mg kg^{-1} dry weight [19][20]. Finally, nickel (Ni) is an essential micronutrient for plants with a normal concentration between 0.05 and 10 mg kg^{-1} dry weight; at higher concentrations, it becomes toxic for plants [21]. In soils, the Dutch Environmental Standard indicates a background Ni concentration of 35 mg kg^{-1} as being acceptable [18]. The limits reported by the WHO for Zn, Cr, and Ni as permissible in plants are 0.60 mg kg^{-1}, 1.30 mg kg^{-1}, and 10 mg kg^{-1}, respectively [22].

Phytoremediation is an alternative bioremediation technique that employs plants to recover soil or a water source without adverse effects on the environment [23]. Among the known technologies developed in recent decades, such as physicochemical and thermal processes, phytoremediation has emerged as a cost-effective remediation technology [24][25]. The main mechanisms of phytoremediation include phytoextraction and phytostabilization, both of which were assessed in the present research; the former has exhibited high efficiency and comprises a reduction in the concentrations of pollutants in the soil through their uptake by harvestable parts of the plant, while phytostabilization restricts the pollutants to the area close to the roots, halting their movement [23][26][27][28]. Plants that accumulate high concentrations of pollutants are called hyperaccumulators. In the case of heavy metals, hyperaccumulator plants contain a concentration higher than 1000 mg g^{-1} of Ni, Cu, Pb, Cr, or Co; 10,000 mg g^{-1} in the case of Zn or Mn; or up to 100 mg g^{-1} of Cd and other rare metals [29]. Until today, approximately 721 hyperaccumulator species have been discovered, most of which hyperaccumulate Ni and Zn and belong to the *Brassicaceae* family [30]. In the case of Zn, some species, mainly located in metalliferous soils in Europe, can accumulate concentrations higher than 10,000 mg kg^{-1} dry weight, most of them belonging to the *Brassicaceae* and *Crassulaceae* families [30][31]. In the case of Cr, the maximum concentration values have also been found in species of the *Brassicaceae* family [32].

The phytoremediation studies of inorganic pollutants with native flora are limited; most of the latest research has been performed with flora from Eurasia and South America [24]. The work of Chandra et al. [33] obtained translocation factors higher than 20 and 3 with *Argemone Mexicana* and *Rumex dentatus*, respectively, for Zn. The same research showed a TF = 10.45 for Ni with *Tinospora cordifolia*. In the case of *Rumex dentatus* and *Saccharum munja*, a *BCF* >1 was obtained for Zn, Cu, and Ni. Jeleni et al. [34] assessed the use of *Aristida congesta* (native) to phytoremediate alkaline mine tailings in Namibia, obtaining promising results for Cr, Ni, and Zn, and classified it as a hyperaccumulator for Cr. Another promising species is *Solanum nigrum* L., a species native to Eurasia, which has been considered as an ideal phytoremediator for several metals due to its high tolerance and easy adaptation to different conditions [35][36]. In Chile, the endemic species *Adesmia atacamensis* has been suggested as an excluder of Zn (translocation factor TF ≤ 1), where the enrichment of tailings with additives might be associated with an improvement in its phytostabilization character for Zn [37]. In another study in Chile, conducted by Lam et al. [38], *Schinus molle* (native) and *Prosopis tamarugo* (endemic) showed some features of a Zn accumulation, while *Atriplex nummularia* (introduced) was the most promising Zn accumulator.

2. Current Insights

The mining industry in Chile is highly developed; however, for many years, it did not take responsibility for its environmental effects. In Chile, there is no legislation relative to pollutants in soils; for that reason, the Dutch Environmental Standard was used for comparison [39], and according to this, the intervention values for Zn and Ni are 720 and 100 mg kg^{-1} dry weight. They are not indicated for Cr [39], but Srivastava et al. [16] indicated an acceptable level in soils for human health and the environment of 64 mg kg^{-1}. The lack of regulations produced a high number of abandoned tailings in Northern and Central Chile. The problems generated by these environmental liabilities are deepening due to the limited resources of the municipalities that must take charge of these residues, which imposes the need to develop an economic, easy, and environmentally friendly technology to remediate the tailings. In 2019, the Chilean government detected 37 abandoned tailing impoundments with a potential risk for nearby towns, which constitutes less than 25% of the total abandoned impoundments and 5% of the total impoundments [3][40].

Moreover, tailings are not a nutritive medium, they have a poor water-holding capacity, and they are normally exposed to severe environmental conditions [41].

It is a well-known fact that using native or endemic species increases the chance that a phytoremediation process will be successfully implemented. This success lies in the establishment of a plant community, of species that adapt easily to local conditions, with the ability to remove the pollutants of interest [15][24]. Plant stress is increased when plants are exposed to a lack of nutrients and organic matter or large variations in temperature [15][41]. For the above, to carry out a phytoremediation process in tailings successfully, the use of native or endemic flora is considered essential. This research focused on the assessment of the ability of certain endemic species of the zone where mining activities are carried out to remove Cr, Zn, and Ni, considering that the most important pollutants in tailings are Cu, Cr, Ni, Zn, Pb, As, Cd, and Hg [42].

The selection of the species was based on their fast growth, good ornamental value, low price, and low water requirements. The latter is essential due to the existing water scarcity in the main mining areas and the drought experienced in the country.

2.1. Presence of Metals in the Original Tailings

As can be seen from **Table 1**, the initial concentration of Zn in the tailings exceeded the values indicated by the Dutch Environmental Standards. In the case of Ni, the initial level was very close to the intervention value, and, finally, levels of Cr in the original samples did not only exceed the indicated value for intervention but doubled it. Unfortunately, in Chile, there are several abandoned tailings close to rivers and population centers; nevertheless, it is not possible to label this pollution, because there is no legislation regarding soil pollution [3].

Table 1. Initial and final concentrations of elements in tailings ± standard deviation.

Element	Concentration mg kg^{-1} Dry Weight ± Standard Deviation			
	Initial Tailings	Final Tailings		
		Oxalis gigantea	*Cistanthe grandiflora*	*Puya berteroniana*
Zn	869.80 ± 31.54	832.17 ± 32.67	828.96 ± 25.67	803.88 ± 37.89
Ni	94.64 ± 2.57	87.98 ± 3.32	80.93 ± 2.89	83.27 ± 4.06
Cr	154.63 ± 5.41	142.79 ± 6.52	140.25 ± 5.98	141.83 ± 4.87

The initial concentrations of Ni and Zn were higher in the roots than in the aerial parts in the case of *Oxalis gigantea* in all samples; however, in the case of *Cistanthe grandiflora* and *Puya berteroniana,* the trend was reversed. Chromium was below the detection limit of the instrument in most samples. All concentrations in the plants were within normal values.

Control samples were maintained over the complete period of growth, and they were analyzed before and after this period, showing no significant differences in Cr, Ni, and Zn. The above indicates that the increase in the final concentrations of the target elements is only due to the effect of the elements contained in the tailings.

2.2. Efficiency of the Phytoremediation Process

There have been a few studies carried out with tailings in Chile under climatic conditions similar to those found in the central and northern zones. One of these studies was carried out by Lam et al. [43], who studied the phytoremediation of Cu, Fe, Mn, Pb, Zn, and Cd. From this research, carried out over one year of growth, they observed a reduced performance by three species (*Prosopis tamarugo*, *Schinus molle,* and *Atriplex nummularia*) in tailings without amendments, obtaining, in all cases, an improvement when a mixture with $CaCO_3$ + compost or $CaCO_3$ + compost + *mycorrhizal fungi* was used. None of these species exhibited the ability to stabilize the metals, but the last two showed high translocation factors for Zn, with a removal efficiency of 40 and 60%, respectively. The species used by Lam et al. [43] can reach several meters in height; the first two are trees, and the latter is a large shrub. In the case of the present research, all species can reach three meters as a maximum height; they showed a slight stabilizing character for Zn with removal efficiencies lower than 10%. None of them presented a translocation factor higher than 1, evidencing their poor performance regarding the phytoextraction of Zn and the rest of the target elements.

In the case of Ni, there is a lack of studies with endemic Chilean species. In other countries, there are native species with the ability to phytoremediate Ni, with a high capacity to bioaccumulate it. In the work of Wu et al. [15], several native Chinese plants were assessed for soil phytoremediation, and the authors obtained a TF > 1 with *Polygonum capitanum* and promising results for the phytoextraction of Ni with *Miscanthus floridulus*, Conyza canadensis, and *Rubus setchuenensis*. The concentration of Ni accumulated in the roots and aerial parts was similar to that obtained in this research. Jeleni et al. [33] used *Aristida congesta*, a native grass of Namibia, to phytoremediate tailings, obtaining a mean concentration in roots of 50 mg kg^{-1} and in aerial parts close to 25 mg kg^{-1}. The pH of the tailings used by Jeleni et al. [33] was 1.5 points more alkaline than the tailings in the present research. Comparing the concentrations, *Puya berteroniana* achieved a higher accumulation in the roots; however, the performance of all species was inferior in the case of the aerial parts. The removal efficiency for Ni with *Cistanthe grandiflora* was 15%, and the efficiencies for the other two species were very close to this value. However, only *Puya berteroniana* showed a *BCF* close to 1, and none of the species studied presented a TF higher than 1 for Ni.

For Cr, promising results were obtained using *Puya berteroniana* and *Cistanthe grandiflora* to stabilize this metal, but the removal efficiencies were close to 10% for seven months of growth. Considering the limited budget of the municipalities in charge of these liabilities, the extraction and incineration of the plants are a very unlikely alternative; thus, stabilization becomes an economic alternative to reduce the mobility of the metal and the possibility of leaching. Similar studies with Chilean native or endemic flora were not found; however, Manikandan et al. [44] screened several native species for the phytoremediation of Cr from soil contaminated by tannery effluent. The species *Acacia auriculiformis* was cataloged as a hyperaccumulator of Cr. In addition, other species, *Dalbergia sisso* and *Thespesia populnea*, concentrated large quantities of this element without falling into this category. Translocation and bioconcentration factors were not calculated; hence, it is not possible to compare these with the present research. Based on the concentrations in roots, a value of 200 mg kg^{-1} was exceeded with *Puya berteroniana* and *Cistanthe grandiflora*, but very low concentrations were achieved in the aerial parts < 20 and 75 mg kg^{-1}, respectively. The work of Manikandan et al. [44] indicated concentrations in the roots close to 10,000 mg kg^{-1} and 5000 mg kg^{-1} in the aerial parts with *Acacia auriculiformis* and 36,000 mg kg^{-1} in the roots and 1500 mg kg^{-1} in the aerial parts in the case of *Albizzia lebbeck*. However, only a few Cr hyperaccumulator species have been found up to now. Other species recognized for the phytoremediation of Cr are *Plunchea indica*, *Cynodon* *dactylon*, *Phragmites australis*, *Typha angustifolia*, *Pterocarpus indicus,* and *Jatropha curcas* [45]. No endemic Chilean species has been cataloged as a hyperaccumulator of Cr.

All the removal efficiencies were calculated using the mean value; however, in the case of Cr and Zn, some results were inconclusive. The analysis of the results and the discussion was mainly focused on the chemical analysis of the aerial parts and roots. Technically, a mass balance would solve this problem; however, in this case, due to the fine roots of the plants, the complete biomass could not be fully recovered from the tailings. Among the species studied, *Puya berteroniana* presented the best performance to stabilize Cr and Zn. Regarding the latter, future research will be focused on the use of nanoparticles and chemical chelators to improve the mobility of metallic ions and/or the ability of these species to concentrate the elements of interest; moreover, the use of different mixtures of amendments and compost will be explored, and the rate of accumulation of each metal and each species will be determined.

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