Postprint of: Journal of Environmental Management (280): 111848 (2021)

Rehabilitation of waste rock piles: Impact of acid drainage on potential toxicity by trace elements in plants and soil

Paula Madejón[1*], David Caro-Moreno[2], Carmen M. Navarro-Fernández[1], Sabina Rossini-Oliva[3], Teodoro Marañón[1]

[1] IRNAS, CSIC, Avenida Reina Mercedes 10, 41012 Seville, Spain

[2] Environmental and Water Agency. Regional Government of Andalusia, (Agencia de Medio Ambiente y Agua de Andalucía, AMAyA). Johan G. Gutenberg 1, 41092 Seville, Spain

[3] Department of Plant Biology and Ecology, University of Seville, Avda. Reina Mercedes s/n, 41012 Sevilla, Spain

*pmadejon@irnase.csic.es

ABSTRACT

The restoration of mining areas, in particular if they are located near towns or villages, is essential to reduce their potential risks for human health and to minimize their visual impacts. In this study, we assess the rehabilitation of a waste rock pile adjacent to the town of Tharsis (SW Spain). We measured vegetation cover and its diversity, and chemical composition of plants and soil, twelve years after remediation by lime amendments, added topsoil and planted vegetation. In general, the applied measures were successful covering with woody vegetation the upper part of the waste rock pile, and providing a greening visual landscape for the town nearby. The most abundant species were the gum rockrose (*Cistus ladanifer*) and the legume shrub *Retama sphaerocarpa*, this latter species most probably introduced in the seedbank of the added topsoil. Also in the soil seedbank, probably arrived the invasive *Acacia saligna*, of fast growth. In contrast, the lower part of the slopes was almost devoid of vegetation. We interpret that partial failure in the rehabilitation process as due to the acid mine drainage, which caused downslope a decrease of soil pH and increased availability of trace elements, thus impeding growth and establishment of plants. In addition, some plants, like *C. ladanifer*, growing at the base of the rock pile, had concentrations of Cd above the maximum tolerable level for animals, therefore representing a toxicity risk. Finally, we propose here an alternative technique to restore waste rock piles, by sorting and selectively handling the extractive wastes, thus reducing infiltration rates, seepages and the negative effect of the acid mine drainage. Those modified waste rock piles will be rehabilitated by the addition of topsoil and planted vegetation, as successfully worked out in the upper slopes of the study site.

Keywords: Alcolea Reservoir, Ecological restoration, Heavy metals, Mine closure, Soil seedbank, Tharsis mine

1. Introduction

The most common environmental impacts of historical mining activities are deforestation, deformation of the soil surface, biodiversity degradation, and contamination of soil, air, water and groundwater (Doley and Audet, 2013). Although current mineral extraction activities are more efficient and they produce less environmental impacts than the techniques used time before (Moore and Luoma, 1990); the legacy of centuries of unsuitable or lack of any environmental program for mine closure have caused that vast areas are contaminated world-wide, even near villages or small towns (UNEP, 2001).

One of the main environmental consequences of this inefficient process is the water contamination caused by the acid mine drainage (AMD; Akcil and Koldas, 2006). The main concern of the AMD is its potential high toxicity due to the extreme acidity of water discharges and their richness in toxic trace elements. In abandoned mine sites, the AMD is produced by the oxidative dissolution of sulphide minerals, when those sulphide-rich materials present in waste rock piles, pits or tailings are exposed to atmospheric conditions (Cánovas et al., 2019). This acid drainage is able to persist throughout the life of an active mine, and long after it has been abandoned (Jain et al., 2016).

Remediation of historical mining areas has turned into an important issue in the most developed countries during the last decades (IPBES, 2018; ICMM, 2019). Due to the low potential profit of these types of degraded zones, the rehabilitation process should be sustainable and low cost. Additionally, due to the huge number of abandoned mines and the high cost of the rehabilitation process, it is necessary to prioritize which areas are going to be treated first. One of the prioritizing criteria followed is the nearness to inhabited areas, to reduce the potential risks for human health and to minimize the visual impacts (Dentoni and Massacci, 2007; Mavrommatis and Menegaki, 2017).

Vegetation restoration in abandoned mine sites is arduous and time-consuming, due to the harsh environmental conditions, especially when slopes are high and steep (Zhang et al., 2014). In fact, this is a common scenario in old waste dumps, where slopes are usually placed at their angle of repose and soil erosion is a major long-term concern (Sengupta, 1993). Thus, these abandoned facilities normally are completely barren. When they are colonized by local vegetation (i.e., “passive restoration”), the highest plant cover and species richness normally occurs at the bottom of slopes. This topographical pattern of vegetation can be explained by the transport and accumulation down slope of seeds, fine soil particles, nutrients and water (Leavitt et al., 2000; Maltez-Mouro et al. 2005).

Post-mining restoration often requires human intervention (that is “active restoration”) to achieve its goals at short-term, by using physical, chemical and biological techniques (Cooke and Johnson, 2002; Festin et al., 2019). Among them, contouring and reshaping the rock piles will prevent erosion and runoff. The addition of lime amendments will increase the soil pH and reduce metal availability; while applying fertilizers and organic residues will improve nutrition and plant growth (Madejón et al. 2006; Kumpiene et al., 2019; Palansooriya et al., 2020). On the other hand, the application of topsoil is recommended when the physico-chemical quality of the substrate is not suitable to grow plants (Bradshaw, 1989). This added soil provides physical structure, mineral nutrients and organic matter, besides beneficial soil organisms and a bank of seeds and buds which facilitate vegetation recovery (Bulot et al., 2017; Ribeiro et al. 2018; Festin et al., 2019). Finally, the use of plants and their associated microorganisms, a technique known as phytoremediation, is the most cost-effective and environmentally sustainable method to reduce the toxic effects of soil contaminants and to promote a faster development of vegetation cover on mine wastes (Tordoff et al., 2000; Mendez and Maier, 2008; Madejón et al. 2018).

From the economic perspective, some examples of rehabilitation works done in the Iberian Pyrite Belt can be examined. The cost of revegetating acidic soils contaminated by metallic mine activities ranged from 10 to 20 €/m^2. These costs normally included modifying slopes, adding amendments, regulating runoff (earth moving operations are the major cost during reclamation), and planting or seeding (D. Caro, pers. comm.). In addition, when combined with other techniques, revegetation of acidic soils can reduce the acid drainage by up to 50% (ITGE, 1989).

We should have in mind that the rehabilitation of mining areas is a long-term process in which the dynamics of potential toxic elements is uncertain; therefore, a program of periodic monitoring and adaptive management is needed (Cooke and Johnson, 2002; Wu et al., 2013; Karan et al., 2016). In particular, plant tissues are the first entrance of contaminants from rocks and soil to food chain (Madejón et al., 2018). That soil-plant transfer is especially relevant when abandoned mining areas are used for food and crop production. Actually, local people often use plants growing around mining areas and they are exposed to health risks (Rossini-Oliva et al., 2020). Therefore, to monitor the concentration of potentially toxic elements in plants and soil, and to compare them with known toxicity levels should be highly recommended in the remediated areas.

In this study we assess the rehabilitation of an old AMD-generating waste rock pile with steep, long slopes, and mostly barren of vegetation, which had a strong visual impact for people living in the adjacent town of Tharsis (SW Spain). The specific questions were: 1) how was the revegetation success after 12 years, in terms of composition, abundance and nutritional status of woody plant species? 2) How much was the concentration of trace elements in the different plant species? Which is the accumulation pattern and the soil-plant transfer of each trace element and for each plant species? and 3) was there any toxicity risk for herbivores, due to high levels of potentially toxic elements in wild plants?. Finally, based on these evidences, we discuss the environmental and health impact of the acid mine drainage and its consequences at basin scale. We also propose an alternative technique for rehabilitation of waste rock piles, minimizing acid drainage.

2. Methods

2.1. Study area

The study area is located in a mine waste rock pile named "Cabezo de los Gatos" adjacent to the town of Tharsis, SW Spain (Fig. 1A). It was originated during the mining operations in the nearby open pit "Filón Norte". This spoil tip was made of heterogeneous, mixed sized rocks, mainly black shales and acidic/basic igneous rocks, extracted during the second half of the twentieth century (IGME, 1984; López-Pamo et al., 2008). The “Filón Norte” belongs to the Tharsis Mining District, within the Iberian Pyrite Belt (IPB), one of the largest massive sulphide deposits in the world, extending across SW Spain and Portugal. This mine has been exploited since about 4,500 BC, mainly for copper, sulphur, gold, silver and cobalt; it was finally abandoned in the 1990s (Tornos et al., 1998; Sánchez-España et al., 2005; Moreno-González et al., 2018). Currently, Tharsis town has about 1730 inhabitants and GDP per capita over 17000 € (64% of the Spain average).

Hydrologically, Tharsis mine is located within the Odiel river drainage basin, where a great number of abandoned mining areas and the associated acid drainages

(AMD) have caused a drastic metallic contamination of the river waters (Macías *et al*., 2017). Despite this scenario of concern about metal pollution, two large dams are planned in the Odiel river basin; one is the Alcolea Reservoir, which will have a capacity of 274 hm^3 and a final budget of almost €164 million (Olías et al., 2011).

The "Cabezo de los Gatos" waste rock pile has a significant extension (almost 40 ha), a large size (maximum height of 35 m) and steep slopes that stood at the rock's natural angle of repose (34°-36°). It was constructed by push-dumping method, on a flat topography, in an area with subsoils of low permeability (IGME, 1984; REDIAM, 2008). The huge volume of waste rocks (≈ 20,000,000 m^3) devoid of vegetation, had strong visual impact for the inhabitants of the nearby town, Tharsis (Fig. 1A).

In this waste dump, some rocks are geochemically reactive (IGME, 1984); they can oxidize and generate acid drainage, producing soil and water contamination (Fernández-Caliani et al., 2009). In fact, salt efflorescence is abundant in drainage channels, resulting from shallow sub-surface flow beneath the waste rock pile. Another environmental problem is the transfer of pollutants from these mine wastes to nearby agricultural soils, by acid drainage and by atmospheric deposition of wind-blown dust; see studies by Madejón et al., (2011) and Fernández-Caliani et al., (2019). However, the most immediate and serious impacts of all acid drainages are on natural waterways. The acid drainages from this waste rock pile are stored in the adjacent reservoir "Embalse Grande" (Fig 1A.); from where they flow to the Odiel River.

Before the reclamation started, this huge volume of waste rocks was practically devoid of vegetation. The scarce vegetation was composed by a limited number of plant species, like gum rockrose (*Cistus ladanifer* L.), false yellowhead (*Dittrichia viscosa* (L.) Greuter), wild olive (*Olea europaea* var. *sylvestris* L., hereafter named *O. europaea* for simplicity) and different rupicolous species (EGMASA, 2004). Soils of this area had coarse textures and high rock contents, which limited the water holding capacity and the effective rooting volume for plants.

The climate in the area is typically Mediterranean, which mild cold winters and long periods of drought in warm summers. The mean annual rainfall is about 630 mm, mostly concentrated from October to May (Valente et al., 2013).

2.2. Rehabilitation history

The "Cabezo de los Gatos" waste rock pile was partially restored by the Andalusian Regional Government, under the FAJA project (2003–2006), with a total budget of 1,787,850 €. The main steps of this rehabilitation process were: waste rock pile reshaping, soil reclamation, revegetation and surface runoff management (EGMASA, 2004). Those restoration works only were done in the area near the town of Tharsis (≈ 10 ha), to reduce the visual impact.

To reshape the area (in November 2005) overburden materials were profiled to a traditional slope for reclaimed waste rock stockpiles: bench slope 3H:1V (18,43º), lift height 10 m and bench width 3 m. This design would provide stability to the slope, allowing the use of machinery in the restoration process. After reshaping, sugar beet lime was added at a rate of 40 t ha^{-1} with a lime fertilizer spreader (in March 2006). Following the amendment addition, a topsoil layer was applied using a front loader with a maximum thickness of 20 cm. Sugar beet lime was not mixed with soil after spreading, due to the large size of soil particles.

Once soil reclamation was completed, the revegetation was carried out (in November 2006), to provide surface cover and erosion control for the new-built slopes. It was made in two steps: first, planting 1-year-old saplings of the trees: stone pine (*Pinus pinea* L.) and wild olive (*Olea europaea*), and the shrub gum rockrose (*Cistus ladanifer*), with a plantation density of about 1600 plants/ha. Saplings were protected from herbivores with shelters and they were watered during the first summer after plantation. Secondly, a mixture of native grasses and shrubs was hydroseeded onto the waste rock pile slopes. The list of seeded shrubs included *Cistus albidus* L., *C. ladanifer* and *Thymus mastichina* (L.) L. (EGMASA 2004; see Table S1). The hydroseeding application rate was 30 kg ha^{-1}.

Finally, the surface runoff management included permanent channels and culverts to convey runoff without causing erosion. Small stone barriers were installed across ditches or areas of concentrated flow to reduce runoff velocity. Filter sediment from runoff were also established to avoid erosion by water action, before growing vegetative cover offered protection.

2.3. Plant and soil sampling

The sampling of vegetation and soil followed a stratified random design to cover the main environmental gradient in the waste rock pile (Fig. 1B). According to the geomorphology, we selected three topographical zones: “Flat Top” (FT), located in the upper flat plain of the rehabilitated area; “Upper Slope” (US) on the upper part of the slopes; and “Lower Slope” (LS) on the lower part of the slope, near the bottom. We distributed ten sampling plots across the studied area, two at the top (FT1 and FT2), three at the upper slope zone (US1, US2 and US3) and three at the lower slope (LS1, LS2, LS3). In addition, we sampled one non-remediated site nearby the top (NR1) and other in the bottom, near the discharge channel (NR2) (Fig. 1B).

At each plot we measured the woody vegetation cover by the interception in a transect line of 20 m. To estimate diversity, we also recorded the extra species present within a belt of one meter wide at each side of the transect. For chemical analysis, we sampled shoots (leaves and twigs) of at least three plants for each dominant species within the community around the transect. To calculate the functional trait - leaf mass per area (LMA; kg m^{-2})-, a subsample of those leaves were scanned and analyzed with Image-Pro Plus 6 (Media Cybernetic, Rockville, MD, US) to measure area, and then dried (at 70ºC at least 48h) to measure dry weight (see methods in de la Riva et al., 2016). In the case of *Retama sphaerocarpa* (L.) Boiss., we measured photosynthetic stems (cladodes) and for *Acacia saligna* (Labill.) Wendl. the phyllodes or modified petioles, to calculate LMA. Leaf N concentration was another functional trait considered, potentially related with photosynthetic capacity and growth rate (Wright et al., 2004); see analytical methods below.

Soil was sampled at each plot, at two depths: 0-10 cm and 10-20 cm, using an Eijkelkamp bucket auger. We took three soil samples along the 20 m transect to make a composite soil sample per plot. The upper depth (0-10 cm) represented the topsoil layer added during the reclamation, while the lower depth (10-20 cm, sometimes reaching the rock before 20 cm) was a mixture of the added soil, the amendments and weathered mine wastes. Sampled soils were stored in plastic bags and transported to the laboratory for analysis. Sampling of plants and soil was carried out in November 2018 and June 2019.

2.4. Chemical analyses of plant and soil

Plant material was washed for 15 s with 0.1M HCl, followed by a 10-s washing with distilled water, and then oven-dried at 70 °C. Dried plant material was ground by passing them through a 500-μm stainless steel sieve. Total C and N in plants and soil were measured using an elemental analyser Leco TruSpec CN. Trace elements in plants were extracted by wet oxidation with concentrated HNO_3 in a Digiprep Ms Block digester, (SPS Science) equipped with a temperature-time programmable controller and polypropylene digestion tube. The extractants were measured by inductively coupled plasma optical emission spectrometry (ICP-OES) with a Varian ICP 720-ES (simultaneous ICP-OES with axially viewed plasma). The quality of the analyses was assessed using the reference INCT-OBTL-5 (tobacco leaves) and the obtained recovery rates were between 95 to 105%.

Soil samples were dried at 40 ºC and then sieved to <2 mm for analysis. Soil pH was measured in a 1M KCl extract (1:2.5, m/v) (Hesse, 1971) using a pH meter (CRISON micro pH 2002). Electrical conductivity (EC) of soil was measured in the extract 1:5 soil/water. Available-P was determined by extraction with sodium bicarbonate at pH 8.5 (Olsen et al., 1954). Available K, Ca, Na and Mg were extracted with 1M ammonium acetate and determined by atomic absorption spectrophotometry.

The pseudototal concentration of trace elements and S in soils was determined by ICP-OES after digesting the samples (<60 μm fraction) with a mixture of concentrated HNO_3 and HCl (`aqua regia') in a Digiprep MS block digester (SPS Science). While the available concentration of trace elements (except of As) was determined in 0.01 M $CaCl_2$ (1:10 m/v) extracts after shaking for three hours (Houba et al., 2000). In the special case of As, its availability was measured after extraction with 0.5 M $NaHCO_3$ and determined by ICP-OES, using a hydride-vapor generator system (Varian VGA 77P). The quality of soil analyses was assessed using the reference sample ERM-CC141 (loam soil) and we obtained recoveries between 95-101%.

2.5. Data analysis

Differences among the three topographical zones in soil and plant chemical composition were tested by one-way ANOVA and posthoc Tukey test (p<0.05). Before the ANOVA analysis, data normality (by Kolmogrov-Smirnov test) and homogeneity of the variance (by Levene test) were tested. When data were non-normal, even after a logarithmic transformation, Kruskal-Wallis test and Mann-Whitney tests were used for mean comparison. The non-remediated sample NR1 was used as reference point in tables and figures, but not for statistical analysis (only one sample).

To explore variation in soils and plants according to their chemical composition, the principal component analysis (PCA) was performed in both cases. To avoid closure effects in chemical compositional data, dataset was logarithmically transformed. We also included for the PCA both non-remediated samples NR1 and NR2. Bivariate relationships between pH values and available trace element concentrations in soils were explored using Spearman correlational analysis. All statistical analyses were performed using the program SPSS 25.0 for Windows.

Bioaccumulation factor (BF) for each trace element and plant species was calculated as the ratio between element concentration in plant leaves and its (pseudototal) concentration in soil (Buscaroli, 2017).

To evaluate the toxicity risk, the concentration of trace elements in plant leaves was compared with the maximum tolerable levels for animals (MTL), according to the National Research Council - USA (NRC, 2005). The level of soil contamination was evaluated comparing the obtained pseudototal concentration of trace elements with their geochemical baselines for soils of South-Portuguese Zone (Galán et al. 2008), and the UNEP (2013) for Cd. In addition, three soil pollution indexes were calculated (Appendix A).

3. Results

3.1. Vegetation diversity and species abundance

A total of 15 woody plant species were recorded in the study area (Table S1). The dominant shrub species were *C. ladanifer* (mean cover of 21%) and *Retama sphaerocarpa* (mean of 17%); both were present in all the remediated plots. While *C. ladanifer* was planted and seeded during the rehabilitation process, *R. sphaerocarpa* colonized the sites spontaneously. The planted saplings of *Pinus pinea* and *Olea europaea* were found in almost all the remediated plots, although with low cover (about 2-3%). It was remarkable the abundance of the Australian wattle, *Acacia saligna*, in some plots (mean cover 18%); this exotic species was not planted during the rehabilitation.

Across the topographical gradient, the woody vegetation successfully covered the remediated flat top (65%) and upper slopes (83%), while in the lower slopes the vegetation was sparse and bare soil made up about 80% (Fig. 2 and Table S1). In the non-rehabilitated plots, *C. ladanifer* was the most abundant woody species, although the barren soil dominated the landscape (Fig. 2).

3.2. Plant functional traits and nutritional status

Among the functional traits, the leaf mass per area (LMA) differed between plant species, from the thinner and lighter leaves of *C. albidus* to the denser needles of *P. pinea*, and the extreme case of the photosynthetic stems (cladodes) of *R. sphaerocarpa*. Leaf N concentration also differed between plant species; as expected, N-fixing legumes had the higher values, while pine needles had the lowest (Table S2). Both leaf traits were negatively correlated across species (r= -0.81, p<0.0001), excluding the leafless stems of *R. sphaerocarpa* (Fig. S 1).

The concentration of macronutrients in leaves also differed between species (Table S2), with exception of C and Na. The variability (measured by the coefficient of variation, CV) of the foliar concentration of elements ranged from 138 % for Na down to 5.7 % for C. We mentioned above the highest N concentration in the legume shrub *R. sphaerocarpa* and in the tree *A. saligna*. This invasive tree also had the highest accumulation of Ca, Mg and S in its leaves (phyllodes). On the contrary, *P. pinea* for N and Ca, and *O. europaea* for S and Mg had the lowest values (Table S2).

The variations of leaf C, K, P and the ratio C/N were relatively independent of the topographical zone. However, some plant species did show differences between zones for certain nutrients. Thus, leaves of *P. pinea* accumulated more Ca, Mg, Na and S in the lower slope (LS) plots. Other plant species showing higher leaf concentration of nutrients in the lower slope were *C. ladanifer* for Mg and Na, *O. europaea* for Na, and *R. sphaerocarpa* for Na and S. The only exception was *O. europaea* having the lower Ca concentration in the plots located downslope (data not shown).

3.3. Trace element concentration in plants

Trace elements showed a different accumulation pattern depending on the plant species and the topographical zone where they were growing (Table S3). In general, trace element levels in plants were within the range of normal contents. The greater concentrations were found in plants at the lower slope, showing in some cases significant differences. Among all studied plants, *C. ladanifer* tended to accumulate the highest concentrations of Cd, Co, Cr, Pb and Zn; some punctual values were above normal levels for plants, as for Cd (1.91 mg kg^{-1}), Co (9.88 mg kg^{-1}) and Cr (2.11 mg kg^{-1}). The trend of *C. ladanifer* leaves to accumulate more trace elements downslope was significant for Al, Cd and Co (Table S3).

The concentration of Cd, potentially toxic, surpassed the maximum tolerable level for animals in leaves of *C. ladanifer* from all the zones, and in stems of *R. sphaerocarpa* from the lower slope (Fig. 3). With regard to Pb, another potentially toxic element, the highest concentrations were for *C. ladanifer*, *O. europaea* and *P. pinea* at the lower slope plots, but always within the normal range for plants (Fig. 3 and Table S3); in all cases they were below the MTL value of 10 mg kg^{-1} (NRC, 2005).

The results of the principal component analysis (PCA) with the concentration of 16 chemical elements (8 macronutrients and 8 trace elements) in 47 samples of photosynthetic organs of 7 plants species showed graphically the clear difference among species and elements (Fig. S2). The first variation trend (PC1 with 26 % variance) was associated to the highest concentration of Zn, Co, Cr and Cd in *C. ladanifer* leaves. Meanwhile, the second trend (PC2 with 19.3 % variance) was associated with the highest values of Mg, S, Ca and Cu in *A. saligna* phyllodes.

3.4. Soil properties and concentrations of trace elements

Topsoil samples (0-10 cm depth) taken in the flat top (FT) zone of the rock pile are assumed to represent the properties of the added topsoil during the remediation process. They were characterized by almost neutral pH (mean around 6), low conductivity, some concentration of carbonates, and relatively high soil fertility, in particular of Ca, Mg and K (Table S4). In contrast, topsoil samples from the lower slope (LS), where the vegetation was sparse, had a lower pH (mean of 3.8), higher conductivity (x14 times), and significantly lower availability of K and Na (Table S4).

Soil samples from the deeper depth (10-20 cm) are assumed to correspond to a mixture of the added soil, the amendments (sugar lime) and the original rock pile. At the flat top zone and compared with the superficial soil, the pH was also almost neutral (mean 6.1), but deep soils had higher conductivity (x 4.5), lower carbonates (6 times less) and higher availability of Ca (x 3.0) and P (x 3.1). Comparing the deeper soil at the two topographical extremes (flat top versus lower slope), in downslope samples the pH was lower (mean 4.8), the conductivity much higher (x 10), also were higher the availability of P (x 4.2) and Ca (x 2.2), but lower of K (4.6 less) and Na (2.2 less) (Table S4).At the adjacent non-rehabilitated zone, used as reference for the original waste rock pile conditions, the soil was strongly acid (mean pH 3.6), with lower availability of Ca and Mg, but higher of P and total C, compared with remediated soils (Table S4).

The pseudototal concentration of trace elements in soils increased downslope, in general (Fig. 4); being statistically different between topographical zones for Al, Pb and S at both soil depths, for Zn at topsoil, and for As and Cr at deeper soil (Table S5). The reference soil samples from the non-remediated zone had the highest values of trace elements (except for Co), in particular in deeper depth (10-20 cm) (Fig. 4).

Among the remediated soils, the lowest pseudototal concentrations of trace elements were found in topsoil samples from the flat top zone. For example, the minimum values for As (10.4 mg kg^{-1}), Cu (9.10 mg kg^{-1}) and Pb (8.65 mg kg^{-1}) would correspond to the added topsoil from a donor site. In contrast, the maximum values of trace elements were in deeper soil samples from the lower slope; for example, they reached up to 132 mg kg^{-1} for As, 162 mg kg^{-1} for Cu, 846 mg kg^{-1} for Pb and 225 mg kg^{-1} for Zn.

Comparing the obtained values of trace elements with the geochemical baselines in the study area (South-Portuguese Zone (SPZ), SW Spain; Galán et al., 2008), we can observe that some elements, in particular As, Cu, Pb and Zn, had anomalous high concentrations in the deeper soils (10-20 cm) of the lower slope zone (Fig. 4). Those soils had a contamination factor (CF) "very strong" for Pb and "strong" for As (Appendix A). For the potentially toxic Cd (not included in the study by Galán et al.) all the soil samples were below the threshold of 1 mg kg^{-1} (UNEP, 2013).

The main variation trend (PCA1) of soil chemical properties (16 variables and 19 samples) accounted for 52% of the variability and corresponded to the contamination gradient; the highest positive scores were for soil samples from the non-remediated zones, associated to higher pseudototal concentrations of Cu, Cd, Ni, Zn, Al, As and S (Fig. S3). At the opposite extreme of the gradient (with highest negative scores), the soil samples from the flat top were ordered; they originated from the transferred soil during remediation, therefore with lower concentration of metals.

The concentration of trace elements available for plants is only a fraction of the total concentration in the soil and it is very dependent of the pH values. The estimated availability (by $CaCl_2$ extraction) of most elements in soils of the lower slope were significantly higher than in the flat top zone (Table S5 and Fig. S4). There was a significant negative correlation of soil pH with the availability of Al, Ni, S and Zn (Table S5). The "mobility ranking" (percentage of availability compared with pseudototal) was Cd>S>Zn>Co>Ni>Cu>As>Al>Cr>Pb (Table S5).

3.5. Bioconcentration factors

The bioconcentration factor (BF) depends on each trace element availability in the soil and on the capacity of each plant species to uptake and accumulate that element in their tissues; therefore, the BF values varied among species, chemical element and sampled soil (Table S6). In general, most of trace elements - Al, Cr, Cu, Ni and Pb – had BF values below 1, indicating that these elements were not accumulated in the shoots of these plants, compared to the soil. The exception was Cd, which presented high BF values for the six plant species; in particular *C. ladanifer* had the highest BF values for Cd (maximum of 61.4 times the concentration in soil). BF values for Zn also were higher than 1 for all plant species, although they were mostly between 1-2 units.

4. Discussion

4.1 Assessment of the revegetation success and nutritional levels

Vegetation has an important role in soil conservation and rehabilitation (Tambunan et al., 2017). The use and integration of a plant cover in rehabilitated soils is the best measure to avoid land degradation, because it provides protection against erosion by wind and water, and avoid dispersion of contaminants (Pérez de Mora et al., 2011). The revegetation of the waste rock pile "Cabezo de los Gatos" (Tharsis, Spain) was partly successful, covering most of the flat top and upper slopes, and providing a greening visual landscape for the inhabitants of the town nearby (Fig. 5). However, the revegetation failed at the lower slopes (about 80% area devoid of plants), due to the adverse environmental conditions originated by the acid drainage (discussed below).

Among the 15 woody species recorded in the study site, the gum rockrose (*C. ladanifer*) was the most frequent and abundant (Table S1). During the rehabilitation process, it was planted as sapling and hydroseeded; in addition, it was naturally abundant in the adjacent non-remediated areas from where it could disperse, and even it could be introduced in the seedbank of the added topsoil. This semi-deciduous shrub is widely distributed in SW Spain where it dominates successional vegetation over acidic soils. *C. ladanifer* is well-known for its tolerance to heavy metals and its potential for phytostabilization of mining areas (e.g., Rossini-Oliva et al., 2016; Santos et al. 2016).

The second most abundant plant species was *Retama sphaerocarpa*, a leguminous shrub, leafless, with photosynthetic stems (cladodes), which is abundant in dry environments of the Iberian Peninsula and northern Africa (Haase et al., 2000). It was not planted in the rehabilitation of the study area and, according to the project (EGMASA, 2004), it was not included in the hydroseeding mixture; neither it was present naturally in the surroundings; therefore, most probably it came as part of the soil seedbank with the added topsoil from an unknown donor site. As most legume shrubs, *R. sphaerocarpa* has hard seed coats forming persistent, long-term soil seedbanks (Pugnaire et al., 2006; Fabião et al., 2014). This woody species has a potential role for the restoration of degraded areas due to its ability to improve the soil by symbiosing with N-fixing bacteria, and its tolerance to drought and trace elements contamination (Caravaca et al., 2003; Moreno-Jiménez et al., 2011). Once established as a shrub layer protecting and improving the soil, they can be used as nurse plants for planting slow-growing trees, like holm oaks (*Quercus ilex*), in metal-polluted areas (Domínguez et al. 2015).

The Australian wattle, *Acacia saligna*, is a small tree with biological traits like fast-growth, symbiosis with N-fixing bacteria, wide niche amplitude, and long-lived hard seeds, which have favoured its success invading ecosystems world-wide (Castro-Díez et al., 2011). In particular, the capacity to build and maintain large seedbanks in the soil (e.g., about 46,000 seeds m^{-2} in a site of South Africa) makes difficult its eradication, once it has invaded and established in a site (Richardson and Kluge, 2008). This tree species has been included in the "list of invasive alien species of concern" by the

European Union (Brundu et al., 2018; EU, 2019). Obviously, it was not planted during the rehabilitation works and most probably it was accidentally introduced with the added topsoil.

The addition of topsoil (and lime amendments) was a successful technique for the revegetation of mine wastes in Tharsis, as found in other studies of mine rehabilitation (Ribeiro et al. 2018; Festin et al., 2019). An additional advantage was the unintended seeding of the legume shrub *R. sphaerocarpa*, which grew successfully, dominating the plant community and improving soil fertility. In contrast, transfer of topsoil can have negative impacts introducing non-desired invasive species (Bulot et al., 2016). That was the case of the Australian wattle, *A. saligna*, in this rehabilitated mine waste.

Planted saplings of trees -*P. pinea* and *O. europaea*- had a relatively high survival rate, being present in almost all the plots; although their relative cover was low due to their reduced growth. In general, all plant species had chemical composition of leaves indicating adequate nutritional status (Table S2; Marschner, 1995); however, *P. pinea* had low concentration of Ca in their needles (mean 0.25%), compared with the normal values for plants: 1-2% (Marschner, 1995; Markert, 1996). The reduced growth of tree saplings in the remediated mine waste can be explained for a combination of dry conditions, high irradiance, soil acidity and high concentration of metals in the subsoil, reached by deeper roots when penetrating below the layer of added topsoil (Domínguez et al. 2010).

Functional traits can explain the potential growth of different plant species, resulting from a trade-off between "fast" and "slow" strategies (Reich, 2014). The alignment of plant species along the "leaf economics spectrum" helps to explain the species individual response to resource gradients (Reich, 2014; de la Riva et al., 2018). Among the woody species growing in the remediated mine waste, *A. saligna* had a fast-growing strategy (Castro-Díez et al. 2011), being locally abundant and forming small woodland thickets. At the other extreme of the spectrum, *P. pinea* and *C. ladanifer* had slower growth and also lower nutrient requirements (de la Riva et al., 2018). An implication for restoration planning is the possibility of spread soil with low organic matter level (<2%) during landscaping operations, using such frugal species. This type of soil has a lower economic cost and less impact on the environment (avoiding removal of natural topsoil).

4.2. Trace elements in plants and toxicity risk

Plants, in general, represent one of the main pathways for the entrance of toxic elements from soils to humans (Love and Babu, 2006). There are several mechanisms leading to the plant absorption and accumulation of potentially toxic elements, including the availability of those elements in the soil, and the processes in the rhizosphere that influence these availability levels and the root absorption rates. One of the main objectives of this study was to assess the transfer of potentially toxic elements from soil to plant, and the level of toxicity risk. This is a key aspect to evaluate the long-term success of the rehabilitation of mine-affected lands (Cooke and Johnson, 2002; Karan et al., 2016; Madejón et al. 2018).

The concentrations of trace elements in the plant leaves collected in the remediated zones of Tharsis were far below the maximum tolerable levels for animals, according to the National Research Council of USA (NRC, 2005). However, some authors (Chaney, 1989) are more cautious and consider that Cd concentrations above 0.5 mg kg^{-1} in plant tissues can be toxic for animals (instead of the 10 mg kg^{-1} level proposed by NRC, 2005). In the study site, leaves of the gum rockrose (*C. ladanifer*) reached the highest levels of Cd (up to 1.9 mg kg^{-1} at the lower slope), therefore presenting a potential risk for herbivores (according to Chaney, 1989). Rossini-Oliva et al. (2016) found even higher Cd accumulation in *C. ladanifer* leaves (mean of 6.8 mg kg^{-1}) from the near Rio Tinto mine tailings, confirming the accumulator strategy for some trace elements of this shrub.

The bioconcentration factor (BF) can be used as a tool to assess the accumulation of toxic elements in plants, and to quantify the potential risk associated to their consumption (Boim et al., 2016). However, this is not a constant value for a particular soil and will depend on the plant species considered (Swartjes et al., 2013). In the Tharsis case study, the BFs for Cd in a particular soil -topsoil of upper slopes- varied from very low values (0.005) for the wild olive, which can be considered excluder of Cd, up to very high values (54.9) for the gum rockrose, which is a Cd accumulator. In general, BF values for most trace elements (excepting Cd and Zn) were below one, indicating a general low risk because of their relatively low accumulation in plant tissues (Table S6). Similar low BF values were found for fruits and vegetables cultivated in the nearby town of Tharsis

(Madejón et al., 2011). The exception of Cd (and Zn in a lesser way) is due to their higher mobility in soils and, that, in consequence, they are easily taken up by plants, mainly when the soil availability is high because of the acid pH (Kabata-Pendias, 2011).

The interpretation of BF values should take in account that at very low concentrations in soils the accumulation in plants (also low) can be slightly higher than in soil, therefore BF>1. Those BF values above unity in non-contaminated sites correspond to low absolute values of the trace element, in both soil and leaves, and therefore they are not relevant in terms of toxicity risk (Madejón et al. 2017).

Among the planted woody species during the mine waste rehabilitation, *O. europaea* and *P. pinea* showed very low accumulation of trace elements in their leaves and therefore low toxicity risk for herbivores. From this perspective, they are good candidates for reforestation of mine wastes and contaminated soils (Madejón et al. 2018). However, they had slow growth and should be complemented with shrubs and herbs to protect the soil with a vegetation cover, as soon as possible. On the other hand, the gum rockrose (*Cistus ladanifer*) was successful growing fast and dominating the vegetation; it has been recommended for mine rehabilitation in dry Mediterranean environments (Rossini-Oliva et al., 2016; Santos et al. 2016). Both, the stone pine (*P. pinea*) and gum rockrose (*C. ladanifer*) are widely used for land reclamation after metallic mining in SW Spain; in fact, they are recommended for revegetation of extremely acid mine soils by the Andalusian Regional Government (Saiz-Díaz and Ceacero-Ruiz, 2008). Nevertheless, the leaves of *C. ladanifer* accumulated high concentrations of trace elements (mainly Cd, Zn, Co and Al) in the study site, which could have potential toxic effects on browsing animals, like goats; therefore, browsing in the remediated area of Tharsis should be banned.

Despite not being planted, the legume shrub *R. sphaerocarpa* (probably introduced with the added topsoil seedbank) colonized successfully most of the remediated area and its cladodes had low concentration of trace elements; therefore, their toxicity risk for herbivores was very low. These results confirm the suitability of this species for the remediation of contaminated sites (Moreno-Jiménez et al., 2011).

4.3. Residual contamination in soils and mine acid drainage

Twelve years after the rehabilitation started, soil conditions at the flat top of the waste pile remained favourable for vegetation establishment and growth; that is, they had neutral pH, low conductivity and relatively high fertility (available Ca, K, and Mg). In contrast, the adjacent non-remediated zones had acidic and low fertility soils, and there the vegetation was sparse. This rehabilitation success was partly due to the positive effects of lime amendments, increasing pH and reducing the availability of trace elements; in particular, the applied sugar beet lime is rich in $CaCO_3$ (70-80 %; Clemente et al., 2015), and its effectiveness can last more than 12 years (Madejón et al., 2018). The second factor contributing to this rehabilitation success was the addition of *ex-situ* topsoil; this technique provides physical structure and nutrients, besides the seedbank discussed above (Bulot et al., 2017; Ribeiro et al. 2018; Festin et al., 2019).

Despite this apparent success with the vegetation recovery and greening of the waste rock pile, thus reducing the former visual impact, it should be had in mind that the residual contamination by metals still represents an environmental problem. The concentrations of trace elements were higher in the deeper soils (in contact with the waste rocks), and in particular at the lower slope of the waste rock pile. We have already mentioned that the concentrations of As, Cu, Pb and Zn were higher than the geochemical baselines (Galán et al., 2008). However, the trace elements in the remediated site were below the generic reference levels (GRL) for Andalusia; that is, below those concentrations in soil with a risk higher than the acceptable maximum for human health or for ecosystems (CMAYOT, 2015). The exceptions was As with values up to 132 mg kg^{-1}, well above the GRL (36 mg kg^{-1}). Some authors have criticised these official GRL values as too high, for example 25 mg kg^{-1} for Cd, 595 mg kg^{-1} for Cu, and 10000 mg kg^{-1} for Zn, thus underrepresenting the reality of soil contamination in the region (Aguilar et al., 1999). Nevertheless, even considering the more conservative guide values by Aguilar et al. (1999), the concentrations of trace elements in the soils of the remediated waste area were below intervention levels.

Plant uptake of trace elements is mainly related with their available fraction in the soil (Aljerf and Choukaife, 2008); in this case, they were extracted with $CaCl_2$ 0.01M to estimate that availability. In comparison, (pseudo)total trace element content in soil is a poor indicator of the potential uptake by plants (Ciadamidaro et. al., 2017). In addition, the available fraction reflects the risk of the potentially toxic trace elements to the ecosystem (Adriano et al., 2004). In this study, the available concentration of trace elements was much higher at the lower slope of the rock pile (Fig. S4), affected by acid drainage. This pattern was related to the acid pH in the most contaminated soils downslope. It is well-known that most cationic elements are negatively and significantly correlated to pH (Kabata-Pendias, 2011). Although available values were low in general, especially for As, they can surpass the maximum permitted values for water soluble concentrations; that is (in mg kg^{-1}): 0.04 for As, 0.02 for Cd, 0.7 for Cu, and 0.5 for Zn (Aguilar et al., 2004). This study confirms that trace elements availability decreases when soil pH is above 4 in acidic soils contaminated by metallic mine in the Iberian Pyrite Belt. This finding was also reported by Saiz-Díaz and Ceacero-Ruiz (2008), supporting that soil pH have to be raised, at least, above this limit (Cánovas et al., 2019), when they are limed in order to reduce soil acidity and trace elements mobility.

4.4. Model of the hydrogeological dynamics in the waste rock pile

From top to bottom of slopes, there is a complex environmental gradient including changes in microclimate, water availability, and transport and accumulation of soil nutrients, generally resulting in higher plant biomass at the bottom (Maltez-Mouro et al., 2005). We found an inverted gradient of vegetation abundance and diversity, contrary to the expected response to the top-bottom soil fertility gradient (Fig. 5). As we explained above, the acid mine drainage (AMD) caused a decrease of soil pH and increase availability of trace elements impeding growth and establishment of plants.

The waste rock pile "Cabezo de los Gatos" contains sulphine minerals that are oxidized in the presence of oxygen (air), water and microorganisms leading to acid drainage (MEND, 2008). Although each pile is unique, in terms of configuration, internal structure and material characteristics (Broda *et al*., 2014), here we present a simple model (adapted from previous studies) describing the sources and pathways of the acid rock drainage (Fig. 6).

According to this model, the supply of oxygen into the rock pile can occur by diffusion, by advection driven by pressure gradients (barometric pumping or wind-driven advection), and by thermal convection caused by sulphide oxidation (Vriens et al., 2018).

Regarding water flow, rain can move through the cover of the reclaimed waste rock pile, either laterally as runoff or infiltrates at the soil surface. Water stored in the soil pores can be removed from the soil by evaporation and plant transpiration, or can move through the active soil zone and can infiltrate into the underlying waste rock (net infiltration) (Swanson and O´Kane (1999).

Additionally, the internal structure has a major impact on water and oxygen movement inside a pile (Broda *et al*., 2014). Because of the traditional method of building a waste rock pile, that is by push-dumping or end dumping, its internal structure is characterized by inter-layered beds of coarse-grained and fine-grained materials, inclined at the angle of repose (INAP, 2014). In unsaturated conditions, the fine-grained layers are the main pathways for water flow in the desiccated pile, whereas the coarse-grained layers act as a vent for the flow of water vapour (Azam et al., 2007). Consequently, and given the existence of a low-permeable layer beneath the pile, the acid seepage may exit along slope faces or at the toe of the rock pile (INAP, 2014). Hence, at the bottom of the slopes, the vegetation suffers the acid runoff and the residual acid seepage emerging near the toe of the restored waste rock pile (EPA, 1971).

Therefore, it is necessary to keep in mind these complex internal-flow systems to successfully restore waste rock piles, and to prevent the generation of acid rock drainage.

4.5. Implications for management of mine waste and consequences at basin scale

The "Cabezo de los Gatos" rock pile is located within the basin feeding the future Alcolea water reservoir (Fig. 7). In this hydrographic unit, around 1,000 ha are occupied by mine waste dumps (Grande, 2016), therefore generating huge amount of acid drainages. In consequence, the water of the Alcolea reservoir, intended mainly for crop irrigation, will be acidic and rich in metals (Olias et al. 2011; Macías et al, 2017). On the other hand, the European Water Framework Directive (EU, 2000) compel to achieve a good ecological and chemical quality of the Odiel river waters by 2027. For all these reasons, it is necessary to put into practice a basin-scale remediation program, low-cost and with no consumption of materials, to prevent acid mine drainage from the many waste rock dumps like "Cabezo de los Gatos".

A common technique for preventing acid drainage in waste storage facilities is the application of a dry cover system, which minimize both water and oxygen fluxes into the reactive waste rocks (Ayres et al., 2002). However, this technique has several drawbacks:

first, the cover does not stop entirely infiltration, secondly the high cost and durability of materials, and lastly the long-term maintenance (MEND, 2004). For example, the average cost of capping a waste rock pile with an impermeable and low-flux dry cover is about 55 €/m^2 in the Iberian Pyrite Belt (D. Caro, pers. comm.).

We propose here a different approach to restore waste rock piles, by sorting and selectively handling the extractive wastes. This is a simple technique consisting in to manage the excavation residues and select them by their geochemical properties; thus, extractive wastes are segregated into two main categories: the potentially acid generating (PAG) and the non-acid generating (NAG) materials (Garbarino et al., 2018), according to the EU (2009) limit values. In this case, instead of selecting excavation residues from an active open pit mine, the managing is made with the deposited waste materials in a rock pile. In order to provide the information required to this selection of waste materials, previously to the reclamation it should be carried out a careful geological mapping of the waste rock pile. Then, the non-acid generating waste should be placed around the periphery of the rock pile, as a protective cover that prevents water contamination (Martin et al., 2017).

The external zones will be made of non-reactive waste rock, built in benches (maximum height of 10 m and 18.43º slope angle) with semi-horizontal compacted layers; this construction method will reduce convective air flow (Fig. 8). When non-reactive wastes are not enough, benches can be also constructed with both non-reactive (at the periphery of the pile) and acid-generating wastes, this latter will be isolated by the non-reactive wastes. In addition, an inclined fine-grained material layer can be placed over the coarser waste rock, thus creating a capillary barrier effect, in order to divert water toward the edge of the pile (Broda et al, 2014; Martin et al., 2017; Dimech et al., 2019).

A basal drain collection system (a deep trench) can be also placed along the basal perimeter of the existing pile, to contain all water seeping from its base. These seepages can be treated in a dispersed alkaline substrate (DAS) plant; that is, a passive remediation system for low flow rates which successfully treats metal-rich acid drainages (Macías et al., 2017).

At the last stage, the modified waste rock will be rehabilitated with added topsoil, and planted vegetation as successfully worked out in the upper slopes of “Cabezo de los Gatos”. We expect that the proposed restoration technique, that involves higher evapotranspiration losses, lower infiltration rates and therefore less generation of seepages, will result in higher plant cover and species richness along slopes. One interesting example of this technology is the successful reclamation of the giant waste rock piles in Bingham Canyon Mine, Utah (USA) (RTKC, 2020).

5. Conclusion

The rehabilitation of abandoned mines is compulsory to restore ecosystems services, mainly to mitigate contamination of air, soil and water, to reduce potential risks for human health and to minimize visual impacts. We assessed the revegetation of a waste rock pile in Tharsis (SW Spain); it was partly successful on the upper slopes, protecting the soil and providing a greening visual landscape for the town nearby. The applied techniques included reshaping the rock pile, adding lime amendments and topsoil, and planting native vegetation. The addition of topsoil had the advantage of introducing valuable legume shrubs by the seedbank, while the disadvantage was the introduction of non-desired invasive trees with potential environmental impact.

In contrast, the revegetation of lower slopes failed because of the adverse environmental conditions originated by the acid mine drainage. The flow of air and water inside the rock pile and the reaction with sulphide minerals produced acid runoff at the bottom of slopes. In consequence, soil pH decreased and availability of trace elements increased impeding growth and establishment of plants. The few tolerant shrubs, like the gum rockrose (*C. ladanifer*), accumulated trace elements in their leaves, in particular the potentially toxic cadmium reached concentration levels that can be toxic for animals. The rehabilitation of waste rock piles is a long-term process and must include the monitoring of potentially toxic elements in soil and plants.

To cope with the problem of acid mine drainage we propose an alternative approach to restore waste rock piles. We suggest to select extractive wastes and to place the non-acid generating ones around the periphery of the rock pile, as a protective cover. That modified waste rock will be rehabilitated with added topsoil, and planted native vegetation.

Credit authorship contribution statement

Paula Madejón: Conceptualization, Methodology, Formal analysis, Investigation, Writing - Original Draft, Writing - Review & Editing, Project administration, Funding acquisition. **David Caro-Moreno:** Conceptualization, Investigation, Writing - Review & Editing. **Carmen M. Navarro-Fernández:** Investigation, Writing - Review & Editing. **Sabina Rossini-Oliva:** Investigation, Writing - Review & Editing. **Teodoro Marañón:** Conceptualization, Methodology, Investigation, Writing - Review & Editing, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

We thank the Spanish Ministry of Science and Innovation for the grant CGL2017-82254-R – INTARSU and European FEDER funds. Also, we thank the support provided by the General Directorate for Environmental Quality and Climate Change of the Andalusian Regional Ministry of Agriculture, Livestock, Fisheries and Sustainable Development. The chemical analyses of soil and plants were carried out by the Analysis Service in the IRNAS, CSIC.

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A)

B)

Fig. 1. A) Location map of the remediated study area "Cabezo de los Gatos", the open pit "Filón Norte" and the town of Tharsis; B) Location of the sampling plots within the waste rock pile.

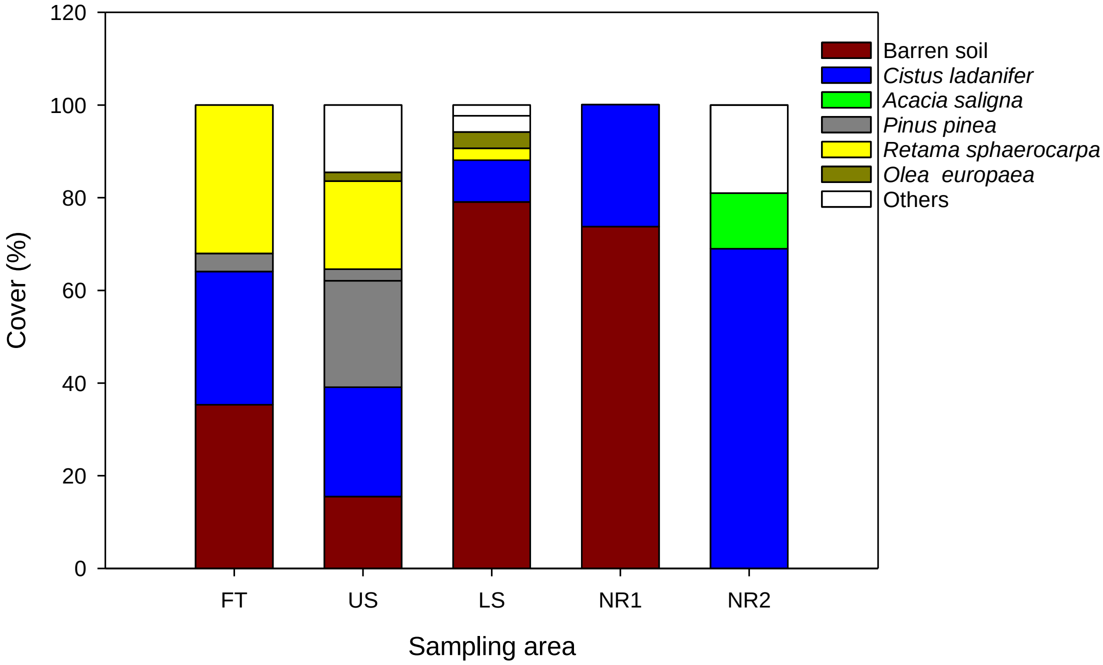

Fig. 2. Percentage of abundance of the main woody species and the uncovered soil, across the topographical gradient of the remediated plots in "Flat Top" (FT), "Upper Slope" (US) and "Lower Slope" (LS), and in the non-rehabilitated plots (NR1 and NR2).

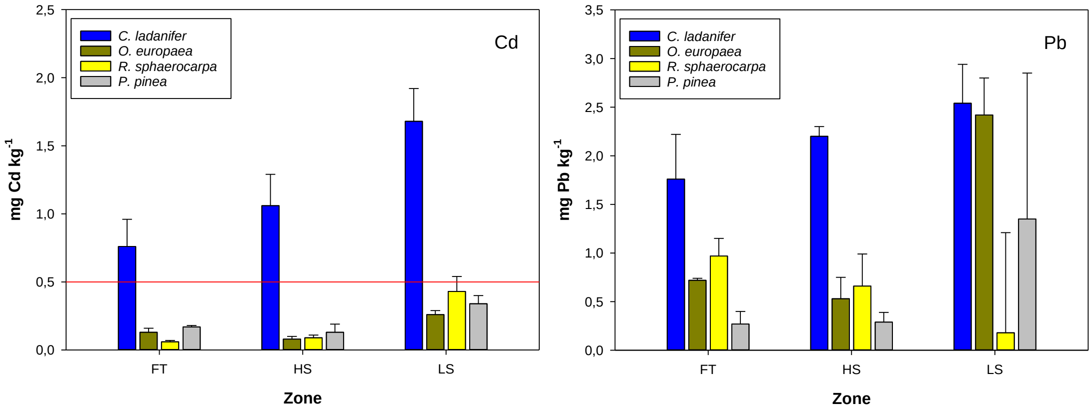

Fig. 3. Cd and Pb concentrations of the most abundant plants at the different studied zones (mean values ± standard deviation). Red line indicates maximum tolerable level for animals (MTL) according to Chaney (1989) for Cd.

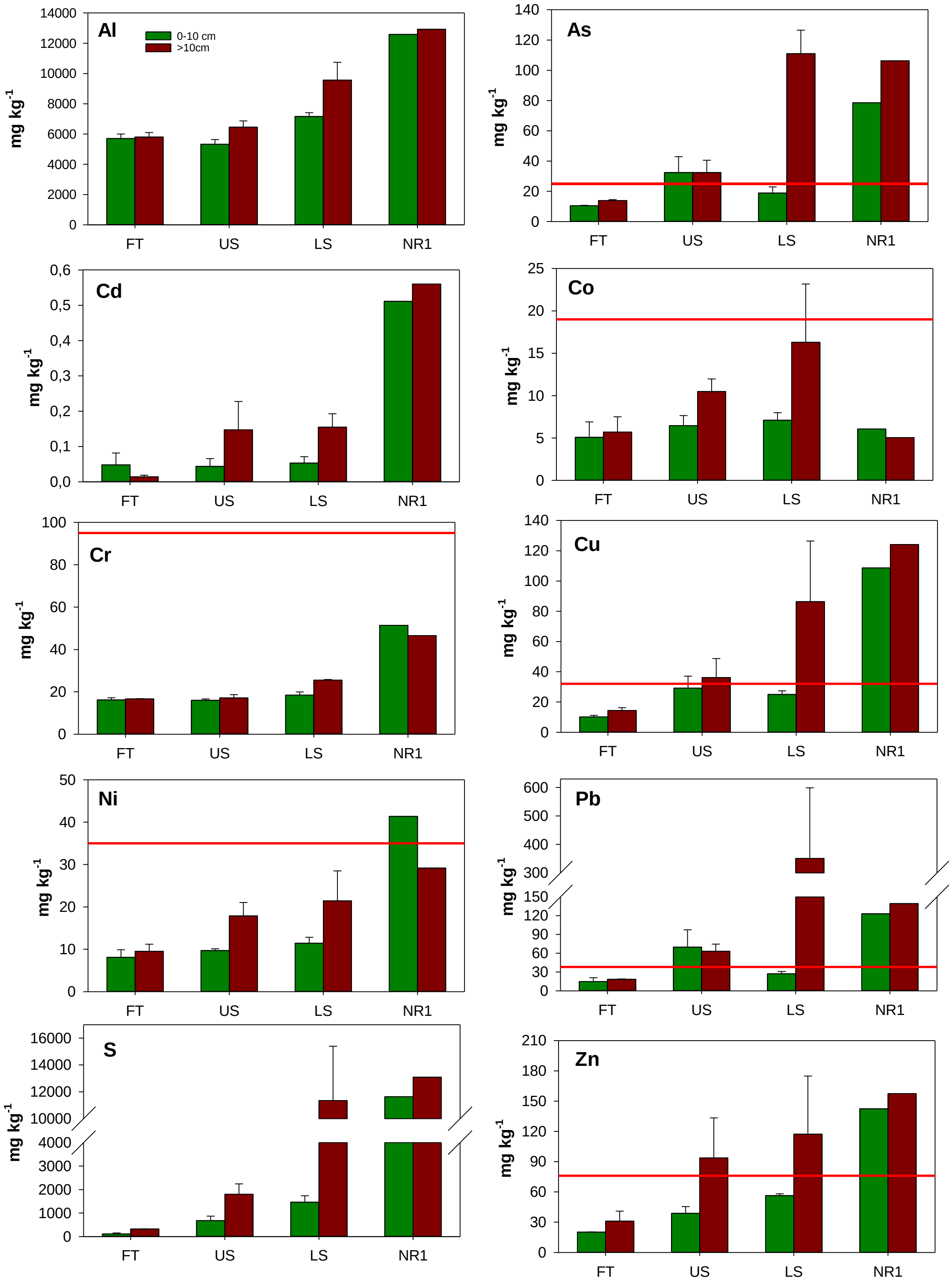

Fig. 4. Pseudototal concentration of S and trace elements in soils from three remediated zones (flat top-FT, upper slope-US and lower slope-LS) at two depths (0-10 cm and >10 cm), and in a non-remediated zone (NR1) used as reference. Mean values ± standard error. Red lines indicate normal values of regional soils (according to Galán et al. 2008).

Fig. 5. General view of the waste rock pile from the town of Tharsis; in 2005, when the rehabilitation started (left top), and in 2017, once covered by vegetation (left bottom). The contrast between revegetated plots at the upper slope (right top) and the barren soils at the lower slope (right bottom), affected by the acid mine drainage. Photos by EGMASA (2005 picture) and T. Marañón

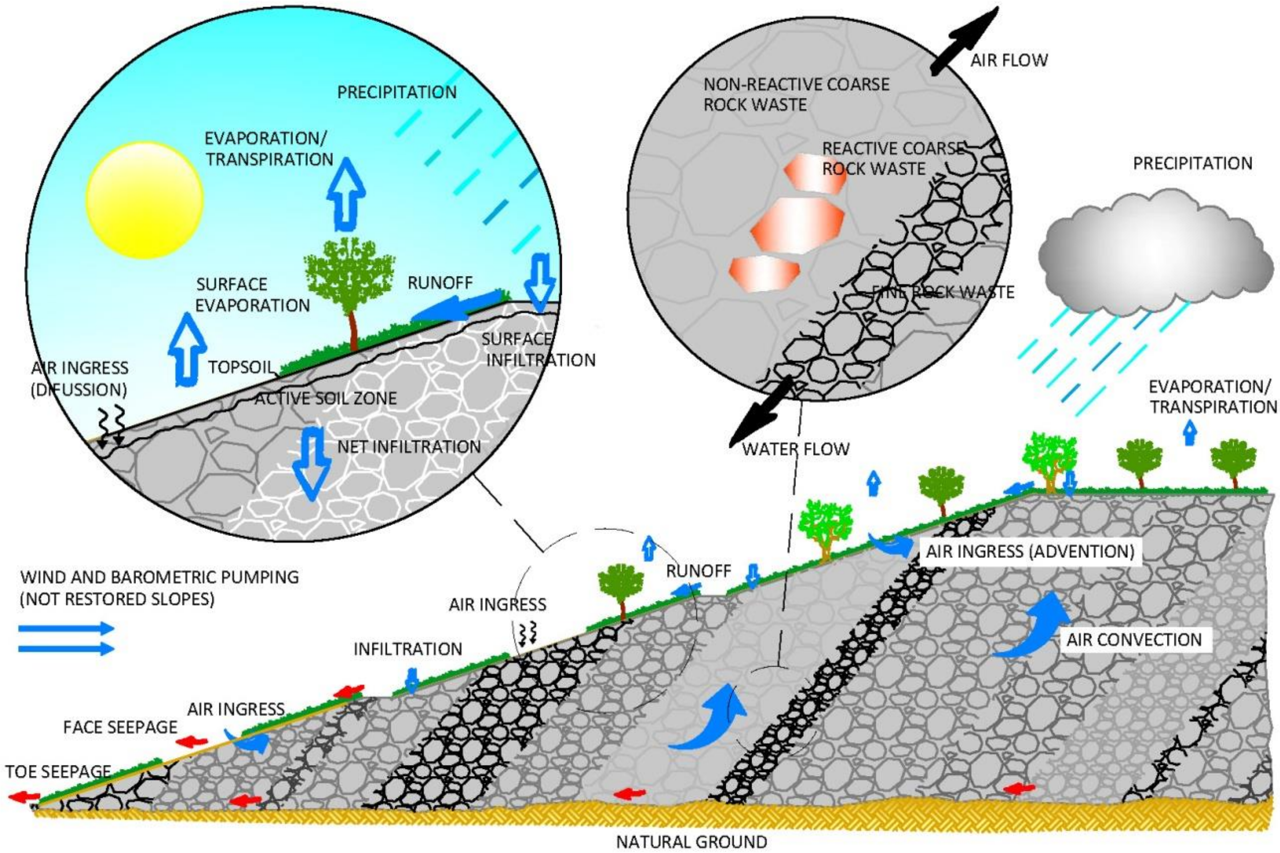

Fig. 6. Model of the hydrogeological dynamics in the waste rock pile "Cabezo de los Gatos", Tharsis, SW Spain. Adapted from Swanson and O´Kane (1999), Aubertin et al. (2005), Azam et al. (2007), INAP (2014) and Vriens et al. (2018).

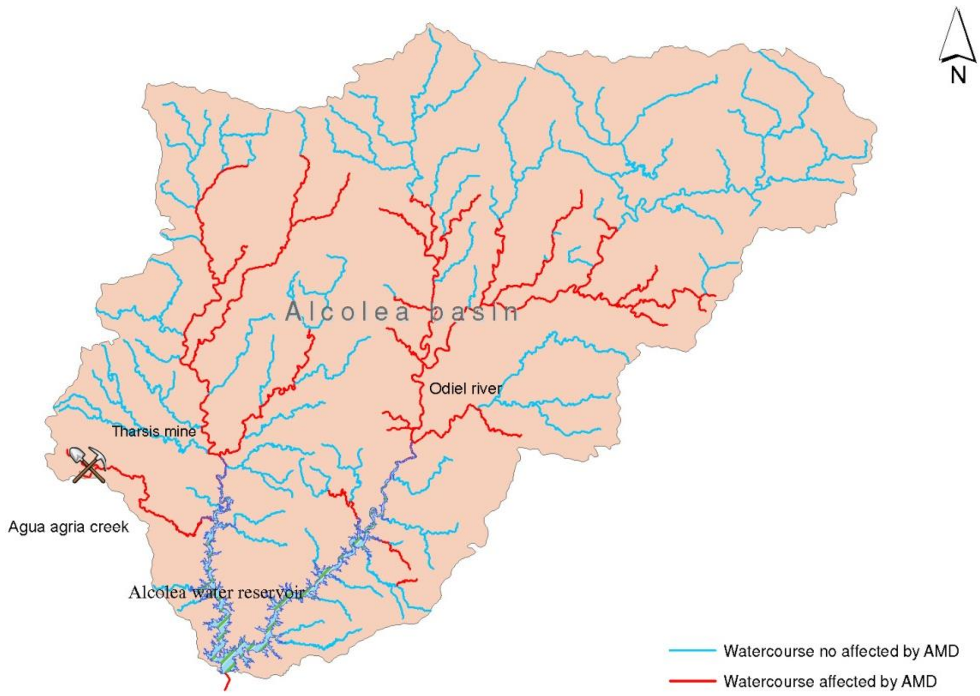

Fig. 7. Drainage basin of the Alcolea water reservoir, indicating which watercourses are affected by acid mine drainage (AMD), and the location of the studied waste rock pile at Tharsis mine.

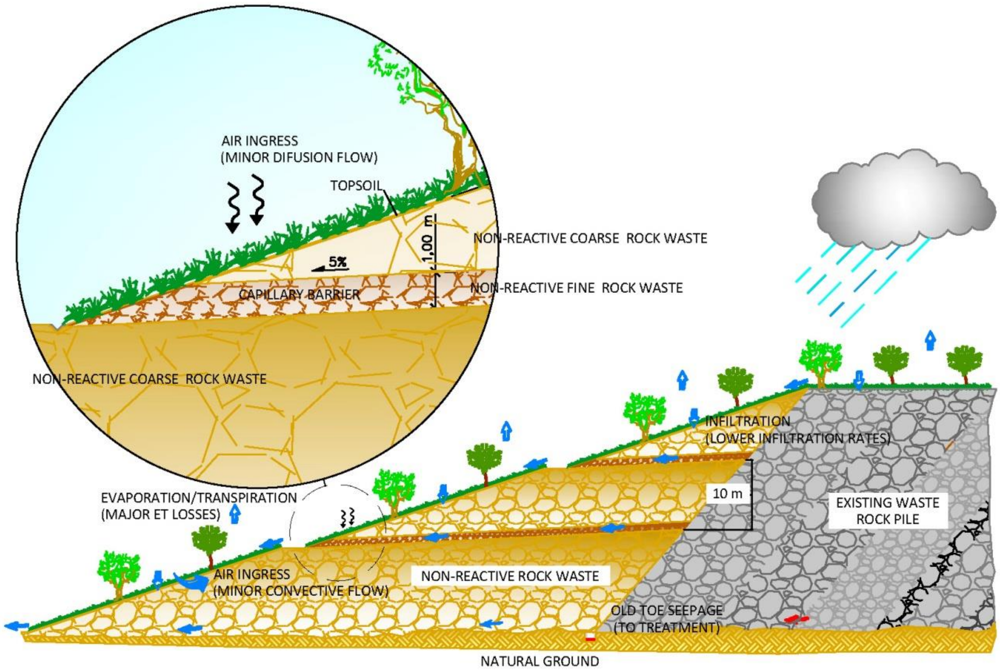

Fig. 8. Proposed configuration of the modified waste rock pile, by selection of the waste materials. Adapted from Broda et al. (2014), Martin et al. (2017), Dimech et al. (2019) and RTKC (2020).

Appendix A

Soil pollution indexes

We calculated three soil pollution indexes: contamination factor (CF), geochemical index (Igeo) and mobility index (MI).

- **Contamination Factor** (CF) was calculated for each element as the ratio between its concentration in the study area and its background value, following the equation CF = M_{soil} / $M_{background}$, where M is the soil element concentration (Rossini-Oliva et al., 2016). As background values, we used those reported by Galán et al. (2008) for the South-Portuguese zone, with exception of Cd for which we used the UNEP (2013) value: 1 mg kg^{-1}. According to the resulting CF, the contamination level of each element for a particular site is ranked from 1 to 6: 0=none, 1=none to medium, 2=moderate, 3=moderate to strong, 4=strongly polluted, 5=strong to very strong, and 6=very strong (Pandey et al. 2016).

- **Index of geoaccumulation** (Igeo) was calculated for each element in base of the concentration of the element *n* in the contaminated soil (C_n), compared to its background value (B_n), following the equation $I_{geo} = \log_2 [C_n/1.5\ B_n]$, where the factor 1.5 is used because of possible variations of the background data due to lithological variations (Cánovas et al., 2019). We used the background values reported by Galán et al. (2008) for the South-Portuguese zone, with exception of Cd for which we used the UNEP (2013) value: 1 mg kg^{-1}. This index was originally stated by Müller (1969) in order to determine metal contamination in bottom sediments by comparing current concentrations with pre-industrial levels. However, it can also be applied to the assessment of soil contamination. According to the I_{geo} values the contamination level of a particular element and site are: <0 Uncontaminated soil, 0-1 Uncontaminated to moderately contaminated, 1-2 Moderately contaminated, 2-3 Moderately to strongly contaminated, 3-4 Strongly contaminated, Strongly to extremely strong contaminated, >5 Extremely contaminated.

- **Mobility Index** (MI) of each element in a soil was calculated as the proportion (%) of the available fraction (0.01 M $CaCl_2$ extracted) respect to the total in the soil (extracted by aqua regia). This index assumes that the reagent $CaCl_2$ extracts the "mobile" or "bioavailable" form of the element in the soil (Houba et al. 2000).

Soil pollution indexes for 8 trace elements in the topsoil (0-10cm) and deep soil (10-20cm) of four zones from the waste rock pile: FT flat top, US upper slope, LS lower slope, and NR1 non-rehabilitated.

	As	Cd	Co	Cr	Cu	Ni	Pb	Zn
	Contamination factor (CF)							
Topsoil								
FT	0.43	0.05	0.27	0.17	0.32	0.23	0.39	0.27
US	**1.31**	0.04	0.34	0.17	0.91	0.28	**1.84**	0.51
LS	0.77	0.05	0.37	0.19	0.78	0.33	0.72	0.74
NR1	**3.18**	0.51	0.32	0.54	**3.40**	**1.18**	**3.25**	**1.88**
Deep soil								
FT	0.56	0.01	0.30	0.18	0.45	0.27	0.49	0.41
US	**1.31**	0.15	0.55	0.18	**1.13**	0.51	**1.67**	**1.24**
LS	**4.49**	0.15	0.86	0.27	**2.70**	0.61	**9.26**	**1.55**
NR1	**4.40**	0.56	0.27	0.49	**3.88**	0.83	**3.67**	**2.08**
	Geochemical index (I_{geo})							
Topsoil								
FT	-1.81	-5.45	-2.58	-3.14	-2.25	-2.73	-2.08	-2.49
US	-0.33	-5.57	-2.19	-3.16	-0.82	-2.44	0.05	-1.59
LS	-1.05	-5.05	-2.03	-2.96	-0.96	-2.22	-1.09	-1.01
NR1	**1.08**	-1.55	-2.23	-1.47	**1.18**	-0.34	**1.11**	0.32
Deep soil								
FT	-1.4	-6.77	-2.39	-3.09	-1.75	-2.48	-1.62	-1.95
US	-0.28	-4.33	-1.47	-3.07	-0.60	-1.61	0.11	-0.56
LS	**1.55**	-3.37	-1.28	-2.48	0.49	-1.52	**1.84**	-0.39
NR1	**1.52**	-1.42	-2.49	-1.61	**1.37**	-0.85	**1.29**	0.47
	Mobility index (MI)							
Topsoil								
FT	0.80	6.12	0.19	0.24	0.44	0.84	0.0000	3.73
US	0.49	45.51	2.53	0.30	0.53	1.85	0.0000	2.60
LS	0.64	116.5	20.68	0.07	8.71	13.88	0.24	23.71
NR1	0.57	7.86	3.99	0.01	0.69	1.70	0.002	3.16
Deep soil								
FT	0.81	33.33	0.99	0.24	0.29	0.73	0.000	0.12
US	1.02	29.53	0.17	0.22	0.19	0.18	0.000	0.35
LS	0.55	15.19	5.36	0.40	4.25	6.06	0.04	7.07
NR1	0.22	3.29	1.13	0.001	0.99	1.14	0.007	1.93

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Supplementary Material

Table S1. Vegetation diversity and species abundance in the study plots, including the percentage of uncovered soil. Some trees and shrubs were planted as saplings (SAPL), while other shrubs and herbs were propagated by hydroseeding (HYDR), according to EGMASA (2004).

Plant species	Source	***Flat Top***		***Upper slope***			***Lower Slope***			***Non-remediated***	
		FT1	FT2	US1	US2	US3	LS1	LS2	LS3	NR1	NR2
Acacia saligna		0	0	0	36.1	17.1	0.1	0	0	0	0
Cistus albidus	HYDR	0.5	0.5	14.5	11.4	9.5	0.1	0	2.5	0	0
Cistus crispus		0	0	0	0	0	0	0.1	0	0	0
Cistus ladanifer	SAPL/HYDR	48.2	9.5	20.0	34.5	25.2	12.0	5.3	11.0	26.3	12.0
Cistus monspeliensis		0,5	0	2.4	0	5.7	0.1	0	0.1	0	0
Cistus salvifolius		0	0	1.2	0	0	0	0	0	0	0
Dittrichia viscosa	HYDR	0	0	0	0	0	0	0	0	0	0.1
Erica australis		0	0	0	0	0	0	0	0	0	7
Halimium halimifolium		0	0	0	0	0	0	0	0.1	0	0
Olea europaea	SAPL	0.5	0.5	0.5	0	1.9	3	0.1	5.5	0.1	0
Pinus pinea	SAPL	3.7	4.0	0.5	3.5	0.5	2.5	1	5.5	0	0
Retama sphaerocarpa		12.5	51.5	55.7	5.5	3.3	3.5	0.1	4.5	0	0
Rumex scutatus	HYDR	0	0	0	0	0	0	0.1	0	0.1	12.0
Thymus mastichina	HYDR	0	0	0.5	0	0	0	0	0	0	0
Ulex sp.		0	0	0	0	0	0	0	0	0	0.1
Barren Soil		**35.6**	**35.0**	**6.3**	**9.0**	**37.1**	**79.0**	**93.8**	**71.1**	**73.8**	**69.0**
Species richness		6	5	8	5	7	7	6	7	3	5

Table S2. Carbon and nutrient concentrations of the studied plants at the different studied areas (mean values ±standard deviation). Results of the factorial ANOVA (*F*-parameter) or *Kruskal-Wallis (*H* parameter) are indicated. Per each element and species, significant differences among zones are marked with different letters.

Plant	Nº	C (%)	N (%)	Ca (%)	K (%)	Mg (%)	Na (%)	P (%)	S (%)	C/N
Acacia saligna	6	47.5±3.03	2.34±0.28cd	2.21±0.50f	0.60±0.15ab	0.87±0.29d	0.03±0.04	0.11±0.02a	1.50±0.40d	20.2
Cistus albidus	5	48.3±2..27	2.07±0.27bc	0.90±0.08f	0.94±0.13c	0.25±0.04ab	0.02±0.01	0.24±0.04d	0.29±0.04b	23.9
Cistus ladanifer	9	50.7±3.68	1.34±0.35a	0.70±0.10bc	0.56±0.10a	0.34±0.06bc	0.02±0.01	0.17±0.02c	0.34±0.10bc	22.3
Olea europea	9	48.4±1.91	1.63±0.42ab	1.48±0.40e	0.78±0.10bc	0.21±0.06a	0.04±0.04	0.15±0.04bc	0.18±0.05a	33.9
Retama sphaerocarpa	8	48.9±2.44	2.68±0.42d	0.53±0.11b	0.48±0.18a	0.49±0.13c	0.03±0.02	0.11±0.02ab	0.45±0.13c	30.5
Pinus pinea	8	47.8±2.83	1.14±0.17a	0.25±0.08[a]	0.48±0.05a	0.29±0.09ab	0.02±0.02	0.13±0.01abc	0.37±0.17bc	39.1
F		*1.331*	***6.629***	***76.801***	***14.969***	***28.815***	*0.487*	***15.636***	***31.561****	
P		*0.271*	***<0.001***	***<0.001***	***<0.001***	***<0.001***	*0.784*	***<0.001***	***<0.001***	

Table S3. Trace element concentration (mg kg-1) of the studied plants at the different studied areas plots (mean values ±standard deviation). Per each element and species, significant differences among zones are marked with different letters.

Plant species	Area	Al	As	Cd	Co	Cr	Cu	Ni	Pb	Zn
Acacia saligna	US (n=5)	45.2±9.70	<d.l.	0.15±0.13	0.70±0.20	0.33±0.05	16.35±4.55	1.33±0.44	0.66±0.21	75.4±18.21
	LS (n=1)	76.1	0.01	0.35	1.10	0.59	16.76	1.37	1.50	59.0
Cistus albidus	TF (n=1)	72.6	<d.l.	0.32	1.74	0.42	8.90	2.50	1.75	47.6
	US (n=4)	83.4±35.7	<d.l.	0.58±0.17	2.17±0.72	0.39±0.09	13.2±1.33	3.01±0.73	1.61±0.74	68.3±0.79
Cistus ladanifer	TF (n=2)	**109±3.12a**	<d.l.	**0.76±0.20a**	**3.53±1.26a**	0.65±0.08	7.01±0.74	3.88±0.51	1.76±0.46	113±61.8
	US (n=3)	**128±30.9ab**	<d.l.	**1.06±0.23ab**	**5.26±2.19ab**	0.62±0.11	8.35±1.33	4.40±1.13	2.20±0.10	99.9±15.4
	LS (n=3)	**210±45.0b**	0.09±0.11	**1.68±0.24b**	**8.62±1.39b**	1.41±0.73	9.03±1.09	7.62±0.43	2.54±0.40	150±19.5
	NR (n=1)	104	0.05	0.84	7.81	0.51	5.67	8.61	1.01	67.4
Olea europaea	TF (n=2)	21.8±4.89	<d.l.	**0.13±0.03a**	0.18±0.01	0.24±0.03	**9.54±0.47b**	3.30±0.84	**0.72±0.02a**	33.3±7.35
	US (n=3)	25.9±4.01	<d.l.	**0.08±0.02a**	0.22±0.08	0.27±0.02	**8.65±0.59ab**	1.97±0.31	**0.53±0.22a**	31.8±5.07
	LS (n=3)	66.6±31.4	0.04±0.03	**0.26±0.03b**	0.19±0.22	0.24±0.10	**6.66±1.18a**	6.22±4.49	**2.42±0.38b**	20.3±4.31
	NR (n=1)	31.5	0.01	0.31	0.05	0.25	3.48	1.56	0.58	12.1
Retama sphaerocarpa	TF (n=2)	42.0±2.82	<d.l.	**0.06±0.01a**	0.70±0.41	0.32±0.04	13.5±3.81	**3.13±1.28a**	0.97±0.18	46.2±2.10
	US (n=3)	36.9±2.78	<d.l.	**0.09±0.02a**	0.84±0.27	0.33±0.06	12.1±5.59	**2.57±0.33a**	0.66±0.36	62.9±13.5
	LS (n=3)	69.0±53.8	0.02±0.01	**0.43±0.11b**	1.41±1.12	0.23±0.20	19.2±3.66	**14.73±2.69b**	0.18±1.03	98.3±28.9
Pinus pinea	TF (n=2)	**159±22.0a**	<d.l.	**0.17±0.01a**	**0.37±0.06a**	0.16±0.01	5.36±1.96	4.87±1.08	**0.27±0.13a**	37.3±0.14
	US (n=3)	**172±93.1a**	<d.l.	**0.13±0.06a**	**0.59±0.29a**	0.24±0.04	3.99±0.28	4.53±2.52	**0.29±0.10a**	35.8±3.54
	LS (n=3)	**394±75.9b**	0.04±0.05	**0.34±0.06b**	**1.42±0.21b**	0.50±0.20	3.91±1.32	5.11±2.68	**1.35±1.50b**	52.0±16.9
Normal levels [a] (mg kg^{-1})		-	0.01-1	0.1-1	0.01-0.3	0.1-0.5[c]	3-20	0.1-5	2-5	15-150
Phytotoxic levels [a] (mg kg^{-1})		-	3-10	5-700	20	5-30 [c]	25-40	50-10	30-300[c]	500-1500
MTL (Maximum Tolerable Levels For Animals) [b] (mg kg^{-1})		-	30	0.5[a]	50	50	40	100	10	500

Abbreviation of dl means detection limit

[a] Chaney (1989)

[b] NRC (2005)

[c] Kabata-Pendias (2011)

Table S4. Mean values ± SE of the main soil properties of the studied soils, comparing between topographical zones: Flat top (FT), Upper slope (US) and Lower slope (LS). Results of the factorial ANOVA (*F*-parameter) or *Kruskal-Wallis (*H* parameter) are indicated.

Zone	**Depth (cm)**	**pH**	**Cond. (µs/cm)**	**Carbonates (%)**	**P-$_{available}$ (mg kg^{-1})**	**TN (%)**	**TC (%)**	**Ca-$_{available}$ (mg kg^{-1})**	**Mg-$_{available}$ (mg kg^{-1})**	**K-$_{available}$ (mg kg^{-1})**	**Na-$_{available}$ (mg kg^{-1})**
Topsoil											
FT	0-10	5.95±0.72	33±7.4	2.40±2.10	1.70±0.30	0.09±0.01	0.58±0.12	923±7.40	395±133	155.9±23.5	47.6±23.5
US	0-10	4.60±0.70	151±92.2	0.63±0.13	1.53±1.03	0.08±0.01	0.48±0.07	890±170	227±21.9	104.5±8.11	67.1±8.1
LS	0-10	3.78±0.01	473±190	0.50±0.03	2.97±0.88	0.06±0.01	0.48±0.08	678±68	380±139	33.7±0.70	22.7±0.70
NR1	0-10	*3.57*	*215*	*0.6*	*18.7*	*0.18*	*2.88*	*140*	*106*	*26.7*	*23.6*
F		3.22*	2.55*	0.032	1.24	4.55*	0.32	1.21	2.00*	**31.33**	**6.27**
P		0.20	0.17	0.984	0.37	0.10	0.74	0.37	0.36	**0.00**	**0.04**
Deep soil											
FT	>10	6.14±0.18	149±4.00	0.40±0.10	5.35±3.95	0.15±0.08	0.47±0.02	2751±958	277±60.3	75.1±1.03	39.9±1.03
US	>10	6.15±0.08	556±75.0	3.07±2.37	19.5±26.8	0.08±0.01	0.79±0.32	4101±1386	257±47.6	82.4±19.2	57.8±19.2
LS	>10	4.77±1.30	1506±655	0.77±0.09	22.5±16.0	0.05±0.01	0.77±0.06	6170±3565	512±305	16.3±3.48	18.0±3.48
NR1	>10	*3.50*	*128*	*0.50*	*24.8*	*0.22*	*3.88*	*83.0*	*41.0*	*22.0*	*139*
F		0.56*	2.77*	3.51	0.43	2.51	0.88	0.35	0.25	**8.23**	**2.51**
P		0.76	0.16	0.17	0.67	0.18	0.47	0.72	0.79	**0.03**	0.17

Table S5. Comparative analysis among the three zones of pseudototal and available concentrations of S and trace elements in soils, by factorial ANOVA (F- parameter). Significance values (p-values) are indicated (p < 0.05); with exception marked by asterisk* where Kruskal-Wallis (H parameter) was applied. Fraction of the total concentration that is available (in %). Correlation between available concentration of each element and soil pH; *r* coefficient and significance (*p*) are indicated.

Element	***Pseudototal concentration***		***Available concentration***		***Fraction Available/ Total (%)***	***Correlation available with pH***	
Topsoil (0-10 cm)							
	F	*p*	*F*	*p*	*%*	*r*	*p*
Al	**11.90**	**0.01**	5.38	0.06	0.46	**-0.733**	**0.025**
As	3.45	0.11	1.21	0.37	0.62	-0.183	0.637
Cd	0.05	0.96	**8.83**	**0.02**	56.20	-0.583	0.099
Co	0.62	0.58	**7.38**	**0.03**	8.22	-0.567	0.112
Cr	1.52	0.30	**54.7**	**0.00**	0.18	0.433	0.244
Cu	2.76	0.16	**9.60**	**0.02**	3.25	-0.600	0.088
Ni	1.80	0.26	**4.83**	**0.07**	5.62	**-0.683**	**0.042**
Pb	**4.30**	**0.08**	**-**	-	0.08	-0.606	0.084
S	**8.71**	**0.02**	2.14	0.21	21.40	**-0.683**	**0.042**
Zn	**14.12**	**0.01**	5.12	0.08	9.95	**-0.717**	**0.030**
Deeper soil (10-20 cm)							
Al	**5.66**	**0.05**	0.03	0.97	0.63	-0.485	0.185
As	**19.19**	**0.00**	2.18	0.21	0.73	-0.267	0.488
Cd	1.48	0.31	1.12*	0.57	22.70	-0.303	0.429
Co	1.15	0.39	4.52	0.08	2.19	-0.450	0.224
Cr	**25.14**	**0.00**	0.21	0.82	0.26	-	-
Cu	1.67	0.28	2.13	0.21	1.65	-0.533	0.139
Ni	1.18	0.38	3.48	0.11	2.37	-0.550	0.125
Pb	**5.26**	**0.06**	**-**	-	0.02	-0.621	0.074
S	**16.37**	**0.01**	**5.68**	**0.05**	20.40	-0.483	0.187
Zn	0.77	0.51	1.17	0.38	2.71	**-0.833**	**0.005**

Table S6. Bioconcentration factor (BF) for each species at different studied zones considering the two studied depths separately

Plant species	Area	Al		Cd		Co		Cr		Cu		Ni		Pb		Zn	
		0-10	>10	0-10	>10	0-10	>10	0-10	>10	0-10	>10	0-10	>10	0-10	>10	0-10	>10
Acacia	US (n=5)	0.01	00.01	**9.69**	**8.86**	0.09	0.06	0.02	0.02	0.72	0.57	0.12	0.08	0.2	0.01	**1.78**	**1.30**
saligna	LS (n=1)	0.01	0.01	**4.22**	**1.55**	0.12	0.06	0.03	0.02	0.63	0.10	0.10	0.04	0.5	0.01	**1.04**	0.26
Cistus	FT (n=1)	0.01	0.01	**22.1**	**16.9**	0.53	0.44	0.03	0.03	0.80	0.71	0.31	0.32	0.08	0.10	**2.35**	**2.24**
albidus	US (n=4)	0.02	0.01	**24.6**	**18.5**	0.31	0.20	0.03	0.02	0.53	0.48	0.32	0.18	0.04	0.03	**1.72**	**1.03**
	FT (n=2)	0.02	0.02	**26.9**	**61.4**	0.72	0.63	0.04	0.04	0.69	0.50	0.49	0.41	0.15	0.09	**5.60**	**3.56**
Cistus	US (n=3)	0.02	0.02	**54.9**	**45.0**	0.79	0.51	0.04	0.04	0.33	0.32	0.45	0.26	0.04	0.04	**2.66**	**1.64**
ladanifer	LS (n=3)	0.03	0.02	**45.7**	**12.9**	**1.22**	**1.03**	0.08	0.05	0.37	0.18	0.68	0.52	0.10	0.02	**2.66**	**2.33**
	NR1 (n=1)	0.01	0.01	**1.65**	**1.50**	**1.29**	**1.54**	0.01	0.01	0.05	0.05	0.21	0.30	0.01	0.01	0.47	0.43
	FT (n=2)	0.004	0.004	0.004	**9.78**	0.04	0.03	0.01	0.01	0.95	0.67	0.45	0.37	0.06	0.04	**1.64**	**1.13**
Olea	US (n=3)	0.005	0.004	0.005	**3.10**	0.04	0.02	0.02	0.02	0.33	0.30	0.20	0.12	0.01	0.01	0.84	0.50
europaea	LS (n=3)	0.010	0.007	0.010	**1.96**	0.03	0.05	0.01	0.01	0.27	0.12	0.56	0.26	0.09	0.02	0.36	0.36
	NR1 (n=1)	0.003	0.002	0.003	0.56	0.01	0.01	0.00	0.01	0.03	0.03	0.04	0.05	0.00	0.00	0.08	0.08
Retama	FT (n=2)	0.01	0.01	**2.68**	**4.64**	0.18	0.15	0.02	0.02	**1.32**	0.98	0.43	0.36	0.08	0.05	**2.28**	**1.67**
sphaerocarpa	US (n=3)	0.01	0.01	**3.75**	**2.78**	0.14	0.08	0.02	0.02	0.42	0.37	0.27	0.15	0.02	0.01	**1.68**	0.89
	LS (n=3)	0.01	0.01	**12.5**	**2.94**	0.22	0.12	0.01	0.01	0.80	0.35	**1.35**	**1.05**	0.02	0.00	**1.76**	**1.47**
	FT (n=2)	0.03	0.03	**7.42**	**13.1**	0.09	0.07	0.01	0.01	0.52	0.39	0.65	0.54	0.02	0.01	**1.84**	**1.33**
Pinus pinea	US (n=3)	0.03	0.03	**3.93**	**2.37**	0.09	0.06	0.02	0.01	0.16	0.15	0.47	0.26	0.01	0.005	0.98	0.59
	LS (n=3)	0.06	0.04	**8.72**	**2.62**	0.21	0.21	0.03	0.02	0.15	0.07	0.43	0.31	0.05	0.01	0.91	**1.00**

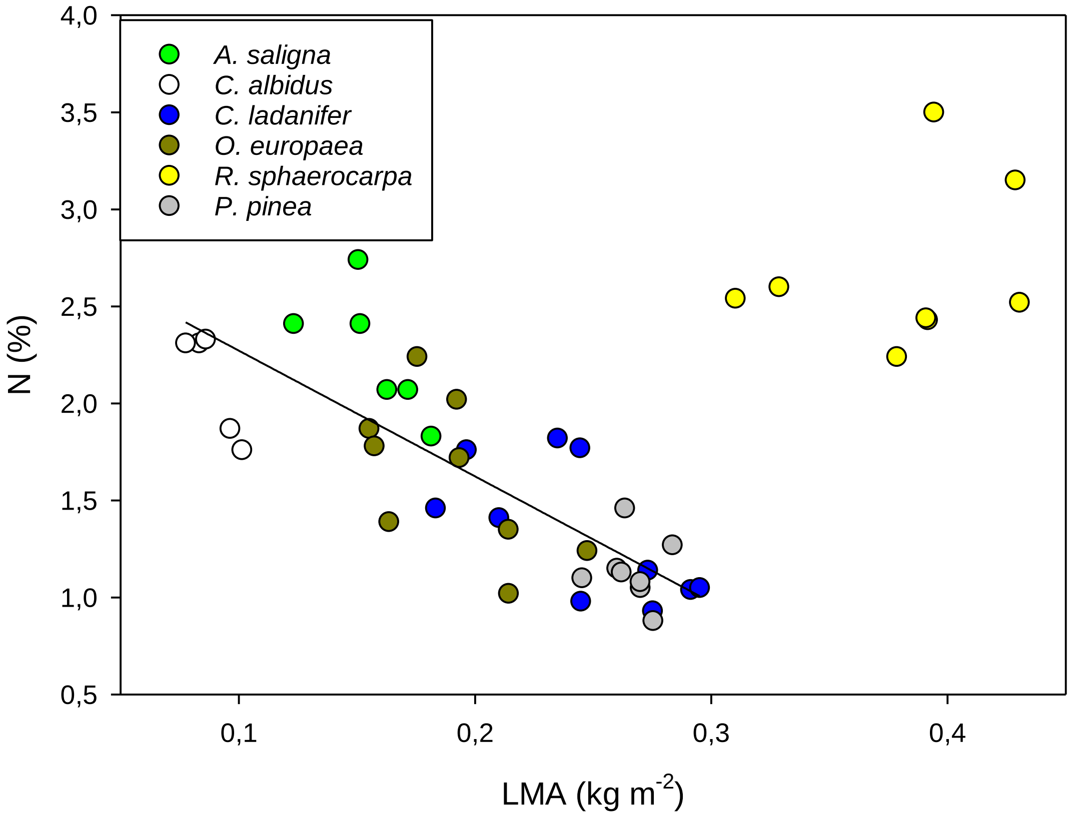

Fig. S1. Functional traits: leaf mass per area (LMA) and nitrogen concentration of the woody species colonizing the reclaimed waste stock piles of Tharsis

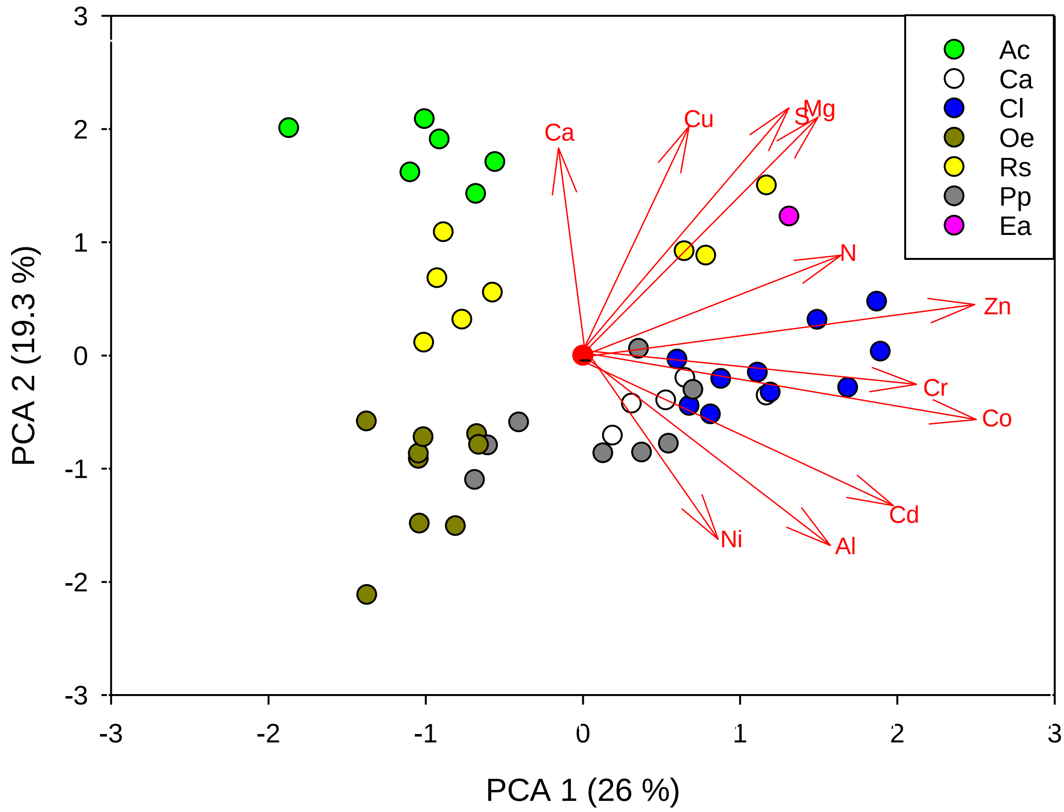

Figure S2. Results of the principal component analysis (PCA) of chemical composition of plants. Species names are: Ac: *Acacia saligna*, Ca: *Cistus albidus*, Cl: *Cistus ladanifer.* Oe: *Olea europaea*, Rs*: Retama sphaerocarpa*, Pp*: Pinus pinea*, Ea*: Erica andevalensis.*

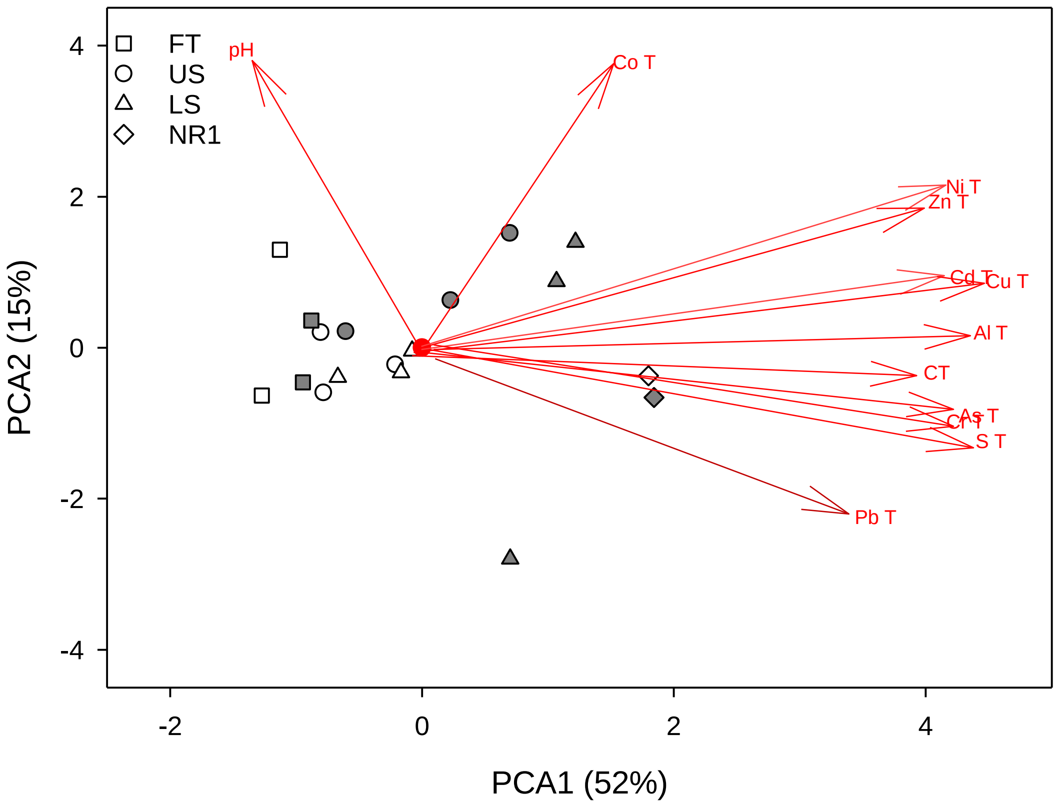

Figure S3. Analysis of the soil principal components using main chemical properties and pseudototal trace elements contents. Vectors represent the eigenvector coefficients (multiplied by five, for clarity) of the 16 elements. Symbols in white represent depth 0-10 cm, and grey symbols represent >10 cm depth.

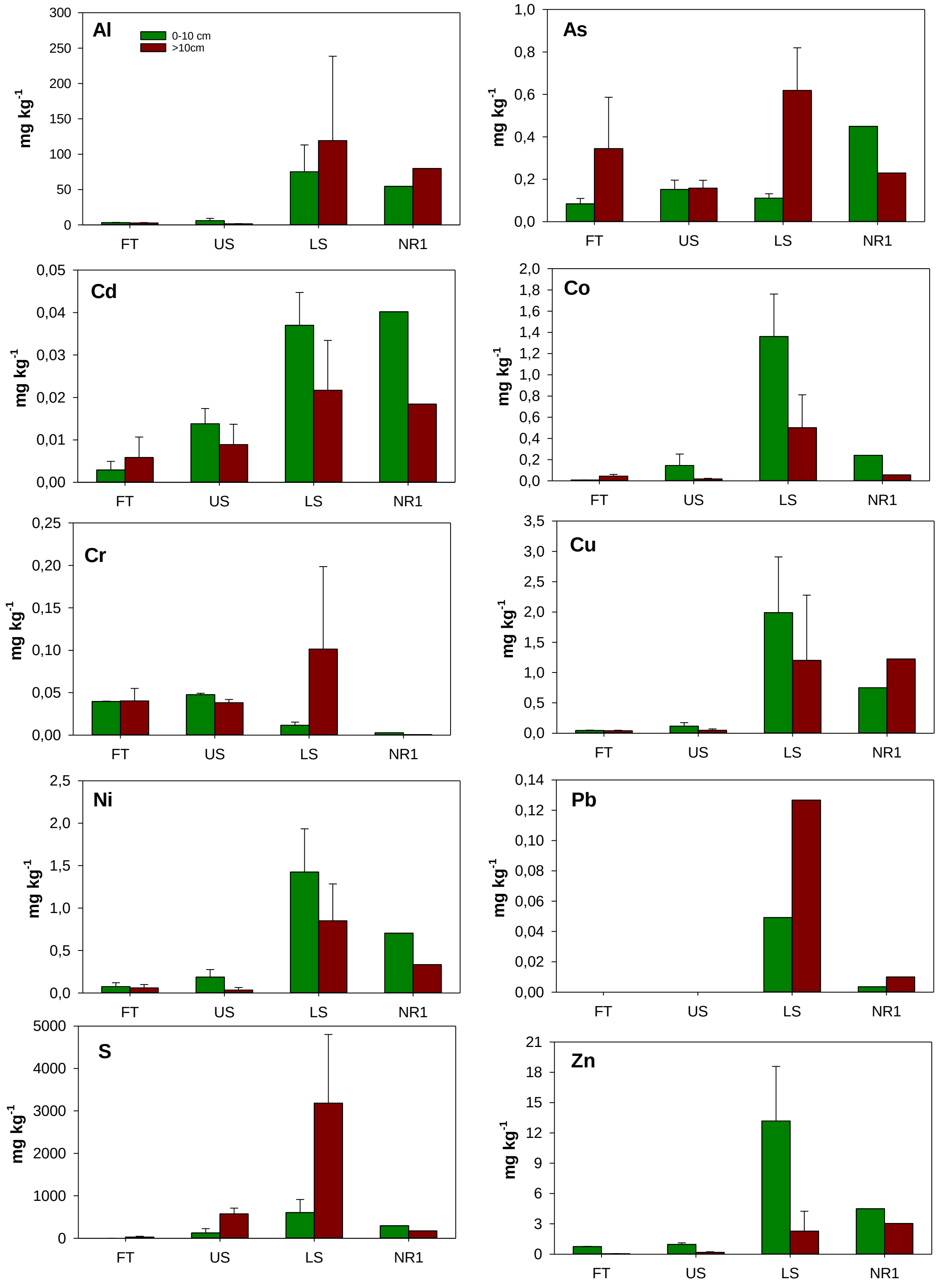

Figure S4. Available trace elements and S concentrations in the studied soils at two different depths (0-10 cm and <10 cm). Mean values ± standard error