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STUDY REGARDING POLLUTED SOILS WITH HEAVY METALS FROM MARAMURES MINING BASIN, ROMANIA, IN VIEW FOR THE REMEDIATION OF AFFECTED AREAS

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Abstract

Heavy metal contamination of soils is associated to mining activities and has impact on plants, micro-organisms and life support functions such as immobilisation, mineralisation, nitrification. The increasingly frequent contamination of soils with heavy metals represents a serious problem for Maramures county in Romania. The phytoremediation method can recover the infertile soil that has been polluted with excessive concentrations of heavy metals, it is a friendly environment method of remediation, can be applied on large areas, it is an in situ method and conserves topsoil. The objective of this paper consists in creating an objective and concrete image, also detailed and actual about the pollution of soils with heavy metals from different areas of Maramures county, in Romania with the purpose of highlighting the necessity of remediation and protection of the environment due to the magnitude of soil pollution. The paper has a fundamental approach based on data from specific literature and technical documentations.

***Key words**: heavy metals, Maramures, phytoremediation, Romania.*

INTRODUCTION

The industry and the technological processes for extraction and for processing the underground minerals are key factors in producing the materials needed for the development of the society and of the economy.
The heavy metals, through their chemical and physical properties, are non-degradable elements in nature and can easily migrate in all environmental factors trough repeatedly transformations.
Starting with the decline of the Romanian mining industry, from the year 1990, the effects on the environment began to appear by polluted soils such as unmonitored and abandoned mining sites and drainage of the acid mines (Doroțan et al., 2015).
Through The EU Accession Treaty from the 1th of January 2007, Romania made a commitment to close the activity of all mining waste dumps (tailings management facilities), because of the non-compliancy with the environmental Community requests.
The closing of mining perimeters and of mine-sterile heaps, of preparation plants and of tailing ponds continues from the perspective of implementing national regulatory procedures to ensure the compliance with the environment protection requirements. The implementation of measures for making waste dumps safe, also for the ecological reconstruction of the affected areas require solutions, investments and works to ensure their stability and safety.
The technical projects which represent the base of the closing and rehabilitation works solution are submitted to currently specific legal regulations on fields such as health, water, safety in the mining industry, construction, environment etc.
The aim of this paper is to represent the effects of heavy metal pollution of mining activities on the affected terrains in order to restore them to their use by applying the technology of phytoremediation, an innovative technology largely accepted by public, eco-friendly, self-sustained, with a large coverage area and without disturbance of the ecosystem.

MATERIALS AND METHODS

The pollution of soils with heavy metals from Baia Mare mining basin, Maramures County
The mining exploitations from Maramureș county in Romania are found in the vicinity of the regions: Ilba, Nistru, Băița, Baia Sprie, Șuior, Cavnic, Băiuț, Poiana Botizei, Țibleș, Baia Borșa and Vișeu de Sus, (Figure 1).

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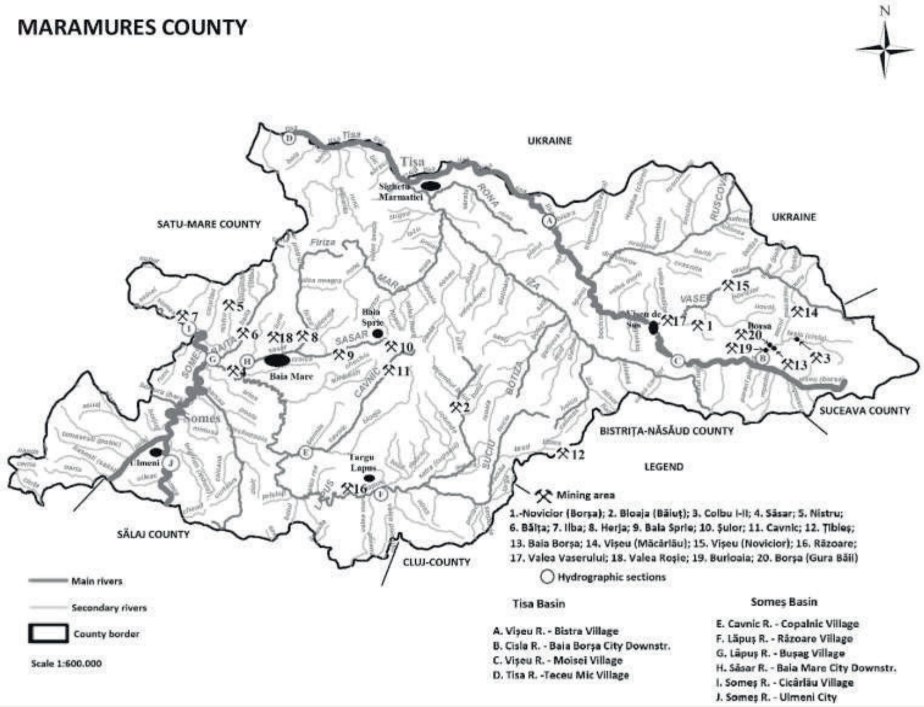

Figure 1. Mining perimeters in Maramureș county (Romania) (Smical et al., 2015)

The propagation and migration of heavy metals in soils could affect the groundwater and also trough contamination by migration and settling on lands.

After some field investigation, was found that depreciated mining concentrates, minerals and flotation sterile were dumped at The Ore Preparation Plants from Baia Sprie, Cavnic, Băiuț, Răzoare, Borșa and Săsar (Figure 2).

Figure 2. Dumps of mining depreciated concentrate and sterile in The Ore Preparation Plant in Băiuț, Maramureș county, Romania (photo: Ioana Petrean)

Because of the surface water entrainment, soil infiltration and because of the oxidation of pyrite from the pyrite dumps it is produced the acidification and heavy metals contamination of soil, underground waters and surface waters (Figures 3 and 4).

Figure 3. Leaks from the sterile dump situated at the exit from Strâmbu-Băiuț village to Lăpuș commune and their path flow on soil and in Lăpuș river, Maramureș county, Romania (photo: Ioana Petrean)

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Figure 4. Mine gallery with the flow of mine water from Băiuț-Vărătec metalliferous mining area and the path of the water from the mine in Băiuț valley, Maramureș county, Romania (photo: Ioana Petrean)

To the present day, the technical projects, technical assistance and the implementation of these works did not solve the environmental problems from the mining field (Figure 5).

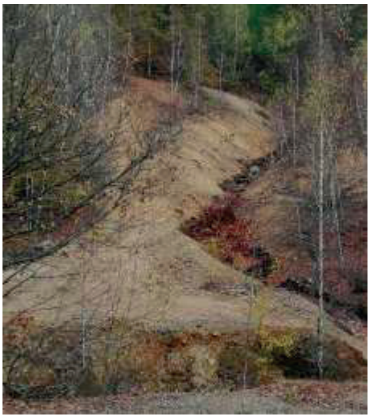

Figure 5. Destroyed coastal fences and withered saplings on rehabilitated waste mining dump in Nistru mining perimeter, Maramureș county, Romania (photo: Ioana Petrean)

In urban soils of Baia Mare were determined the following values for heavy metal concentrations: 0.3-16.6 mg/kg Cd, 23-404 mg/kg Cu, 151-3261 mg/kg Pb and 180-2695 mg/kg Zn, and in the East part of the area, close to the copper smelter, the concentrations were: 40375 mg/kg Pb, 6122 mg/kg Zn and 5823 mg/kg Cu (Mihali et al., 2013).
Also, close to Baia Mare, in Băiuț-Vărătec metalliferous mining area, the following concentrations were determined: Cu from 17.23 to 2184.1 ppm; Pb from 18.98 to 6362 ppm; Zn from 101.12 to 2834.7 ppm; Cd from 0.12 to 20.9 ppm; Ni from 1.52 to 311.4 ppm and As from 0.004 to 266.7 ppm (Chira et al., 2014) (Figure 6).

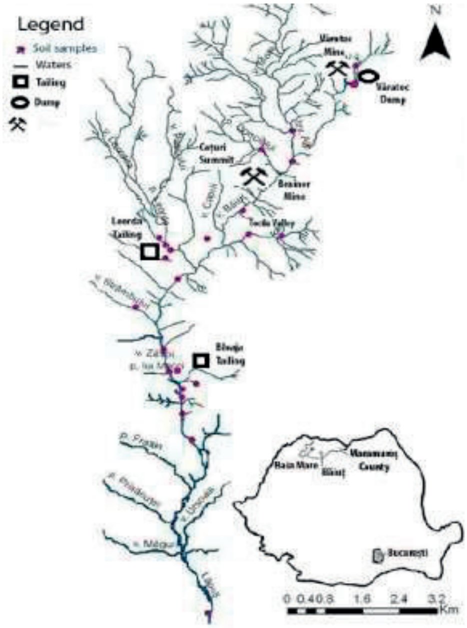

Figure 6. Mining site Băiuț, Maramureș county (Damian et al., 2014)

In agricultural and forest areas from Maramureș county, Romania, the following values for heavy metal concentration were found in soils: 5.6-48 mg/kg Cu, 31-243 mg/kg Zn, 17-639 mg/kg Pb, 0.1-2.0 mg/kg Cd, 45-1265 mg/kg Co, 0.1-31 mg/kg Cr, 1.6-86 mg/kg Ni and 0.2-100 mg/kg Mn (Manea et al., 2018).

The soil remediation

Soil depollution methods can be broadly divided into three categories: physical, chemical and biological (Figure 7). The depollution methods of heavy metals from contaminated soils can be used in combination to remediate contaminated sites. Innovative and cheap technologies are needed to be able to decontaminate soils (Gomes, 2019).
Physical, chemical, biological and combined remediation methods are researched and adopted to address the problems of contaminated soil and contaminated sediments, taking into account the environmental impact assessment and environmental criteria in setting remediation targets.

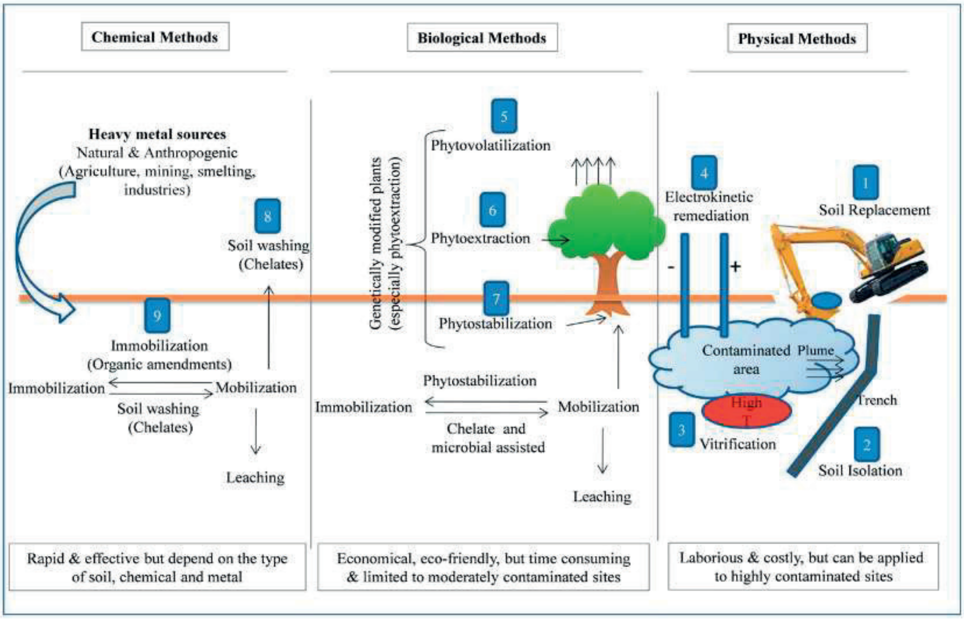

Figure 7. Comparison of different soil depollution methods (Khalid et al., 2017)

Physical remediation methods include: (1) soil replacement, (2) soil insulation, (3) vitrification, and (4) electrokinetics; biological methods include: (5) phytoevaporation, (6) phytoextracttion, and (7) phytostabilization; chemical methods generally include (8) soil washing and (9) immobilization (Khalid et al., 2017).
Physical remediation of soils refers to the partial replacement of contaminated soil with uncontaminated soil and the treatment by heat desorption of the soil. The thermal desorption method involves heating the contaminated soil so that the pollutant volatilizes in the soil. These volatile metals are collected using vacuum pressure and thus removed from the ground. This method is laborious, expensive and has limited applicability, is possible only for small portions of soil (Sidhu, 2016).
Chemical leakage refers to the washing of contaminants from soils with water, reagents, fluids and gases that help the pollutant to drain from the soil, and the recovery of metals extracted in leachate is done by using various chelating agents, surfactants, etc. In the method of chemical fixation, some reagents are added that form insoluble bonds with heavy metals and decrease their mobility in soils. Electrokinetic remediation involves the application of high voltage to the ground to remove the metal. The vitrification process involves heating the soil to very high temperatures (1400-2000°C), so that the pollutant volatilizes or decomposes, but it is an expensive, laborious and complicated process with limited application (Gong et al., 2018). The method performs well in soil with low permeability (Chao et al., 2014).
Biological methods refer to phytoremediation, microbial remediation and animal remediation for the removal of heavy metals from the soil.
Phytoremediation, in situ treatment, consists in growing plants that have hyperaccumulative properties (mainly in the root zone) on soil contaminated with heavy metals. The use of plants and their microcellular absorption system is a new technology, the success of the method consisting in finding suitable plants with affinity for the accumulation and tolerance of heavy metals from over 400 known species (Chao et al., 2014).
Phytoremediation (Figure 8) is advisable for sites contaminated with hydrophobic pollutants such as: benzene, toluene, ethyl benzene, xylenes, chlorinated solvents, PAHs, nitrates, ammonium, phosphate and heavy metals (Suthersan et al., 2017).

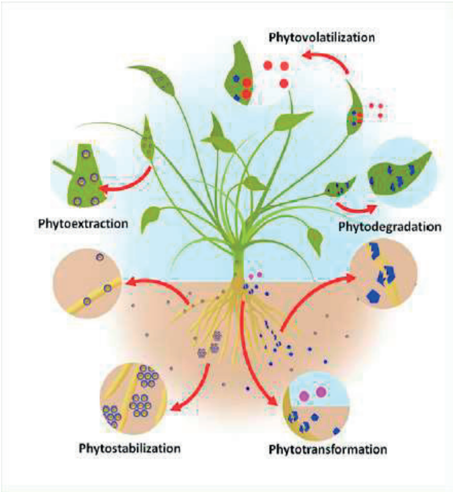

Figure 8. Schematic representation of phytoremediation approaches (Parmar et al., 2015)

Phytovolatization is an approach that involves the absorption and transpiration of metals into their volatile forms and their release or modified forms into the atmosphere through stomata (Tangahu et al., 2011).
Phytoextraction involves the transfer of metals from the soil to parts of the plant, and remediation using microorganisms refers to their ability to change the physical and chemical properties of pollutants, affecting the mobility and transformation of heavy metals in soils.
Plants act as systems for pumping and treating with solar energy, and contaminants solubilized in water are taken up by their roots and transported and translocated through various plant tissues, where they can be metabolized, sequestered or volatilized.
Restoration of polluted soil areas using phytoextraction consists in the in situ cultivation of suitable plant species, harvesting their biomass loaded with heavy metals and treating it (by composting, compaction, drying, thermal decomposition) to reduce its volume and mass, which will later be eliminated as hazardous waste or can be used for the re-extraction of trace elements (Suman et al., 2018).
In phytostabilization or phytoimmobilization, plants are chosen for their tolerance at the sites conditions and the contaminants are sequestered in the lignin of the cell wall of the root tissue (Bansode, 2015).
The mobility of heavy metals is reduced by phytostabilization, through reduced soil erosion and wind dust and low solubility of contaminants. Heavy metals are concentrated due to erosion through sorting and deposition of different sizes of soil fractions (Suthersan et al., 2017).
In phytostabilization, long-term monitoring of mobilization, bioavailability, heavy metal toxicity and ecological impact is necessary.
Microbial remediation refers to the use of microorganisms to achieve the absorption, precipitation, oxidation and reduction of heavy metals in the soil.

The quantification of phytoremediation

The bioconcentration factor (BCF), defined as the ratio between the total concentration of the element in the harvested plant tissue (C_{plant}) and its concentration in the soil in which the plant grew (C_{soil}), is calculated as follows (Favas et al., 2014):

$$BCF = \frac{C_{plant}}{C_{soil}}$$

The translocation factor (TF), defined as the ratio between the total concentration of the elements in the aerial parts of the plant (C_{shoot}) and the concentration in the root (C_{root}), is calculated as follows (Favas et al., 2014):

$$TF = \frac{C_{shoot}}{C_{root}}$$

The metal removal efficiency (ER) is calculated as follows (Gayatri et al., 2019):

$$ER = \left[C_{shoot} + \frac{C_{roots}}{C_{soil}} \right] * 100$$

A hyperaccumulating plant must have BCF>1 or TF>1, and the total concentration of Cu, Co, Cr or Pb>1000 mg/kg, or Fe, Mn or Zn>10000 mg/kg in the aerial parts (Wahsha et al., 2012).

Phytoremediation used plants

In hyperaccumulating plants, the content limits of metal elements in dry biomass are 100 mg/kg for Cd and Se, 1000 mg/kg for Co, Cu, Ni and Pb and 10000 mg/kg for Zn and Mn. These values are up to 100-1000x than in non-hyperaccumulating plants under the same conditions (Suman et al., 2018).
Grasses are the most frequently evaluated plants in phytoremediation because compared to trees and shrubs, herbaceous plants, especially grasses, have characteristics of fast growth, large amount of biomass, strong

resistance, efficiency in soil stabilization and ability to remediate different soil types, are adapted low soil nutrient content, stress environment and to shallow soils (Laghlimi et al., 2015).

Most of hyperaccumulator plant species belong to the plant families: *Brassicaceae* (25%) (Figure 9), *Asteraceae*, *Caryophyllaceae*, *Tiliaceae*, *Cyperaceae*, *Cunouniaceae*, *Fabaceae*, *Scrophulariaceae*, *Myrtaceae*, *Proteaceae*, *Flacourtiaceae*, *Lamiaceae*, *Poaceae*, *Violaceae*, *Euphorbiaceae*, *Rubiaceae, Cruciferae*, including genus *Brassica*, *Alyssums* and pennycress (*Thlaspi*), marigold (*Calendula officinalis*), Mexican marigold (*Tagetes erecta*) (Hassan et al., 2019; P. Ahmad, 2016; Parmar et al., 2015; Chao et al., 2014).

Figure 9. The most important hyperaccumulators from the fam. *Brassicaceae*. (1)*Arabidopsis halleri* (2) *Arabidopsis thaliana*, (3) *Brassica juncea*, (4) *Thalspi caerulescens*, (5) *Thalspi praecox* (Anjum et al., 2012)

Alpine penny grass (*Thlaspi caerulescens*), has been shown to accumulate Zn up to 2000 mg/kg and even 4000 mg/kg. The Indian mustard plant (*Brassica juncea*), has been found to accumulate a significant amount of lead. Indian hemp (*Apocynum* sp.) and common ragweed have also been observed as significant lead accumulators. *Aeollanthus subcaulis* var. *lineris*, a species of the *Lamiaceae* mint family, and bay grass (*Paspalum notatum*) are other hyperaccumulative plants known to accumulate Cu, respectively Cs. Hyperaccumulative plants can access contamination from shallow soils only up to 61 cm deep. If the contamination is between 1.80 m and 3 m, poplar trees can be used for phytoextraction and accumulation of heavy metals by sequestration (Suthersan et al., 2017).

The thale cress plant (*Arabidopsis thaliana*) has a tolerance to Cd without visible signs of toxicity of 1 μm in the soil substrate, but concentrations higher than 5 μm lead to visible morphological changes. The roots of the plant can contain up to 89% Cd under experimental conditions and only a very small part is transported in shoots. Similar results were obtained for *Arabidopsis halleri* plants, with hyperaccumulative root for Cd, but grown in soil have only 20% Cd in the root, the rest of Cd ions are found in the aerial parts (Anjum et al., 2012).

Gayatri et al. (2019) studied the potential of Indian mustard (*Brassica juncea*) for the removal of heavy metals in urban red soil fertilized with manure from cattle, in powder form and found that the metal absorbed in the largest amount by the plant was Pb (151.4 ppm), due to the already available form, followed by Zn (55 ppm) < Cu (15.4 ppm) < Cr (9.6 ppm) < Ni (3.1 ppm) and the percentage of recovery was higher at Zn (51.8%) < Cu (41.6%) < Pb (20.8%) <Cr (11.5%) < Ni (6.1%).

In the study by Ghazaryan et al. (2019) were investigated native species of wild plants that grow in soils contaminated with Cu, the content of Cu (d.s.) determined in the root varied between 55 mg/kg in perforate St John's-wort plant (*Hypericum perforatum*) and 775 mg/kg (*Thymus kotschyanus*). In plant shoots it ranged from 33 mg/kg in Oriental Germander plant (*Teucrium orientale*) to 243 mg/kg in timothy grass (*Phleum pratense*). *Thymus kotschyanus*, *Phleum pratense* and common yarrow (*Achillea millefolium*) had the highest phytostabilization potential.

Cheng et al., (2016) investigated the feasibility of phytoremediation using the silver grass (*Miscanthus floridulus*) on soil contaminated with high concentrations of lead (up to 6000 mg/kg). After one year, the root content reached 806.7 mg/kg, and the plant immobilized 1.13 kg/ha Pb from soil annually (Gong et al., 2018).

In a temperate climate, the best species for vegetating degraded sites belong to the genus: fescue (*Festuca*), ryegrass (*Lolium*), wheatgrass (*Agropyron*), meadow-grass (*Poa*), medick

(*Medicago*) and vetches (*Vicia*), while the trees that assure a good phytostabilization of the underlayers are: poplar (*Populus*), acacia (*Robinia*), willow (*Salix*), alder (*Alnus*), birch (*Betula)* și maple (*Acer*). Other plants such as: rye (*Secale cereale*), oat (*Avena sativa)*, barley (*Hordeum sativum*), common wheat (*Triticum aestivum*), orchard grass (*Dactylis glomerata*), red fescue (*Festuca rubra)*, Kentucky bluegrass (*Poa pratensis*), annual ryegrass (*Festuca perennis*), perennial ryegrass (*Lolium perenne*), tall meadow oat (*Arrhenatherum elatius*), Timothy grass (*Phleum pratense*) have a major role in nursery culture (Gajić et al., 2018).

In the list of hyperaccumulating plants for Cu are the Indian mustard (*Brassica juncea*), water hyacinth (*Eichornia crassipes*), sunflower (*Helianthus annus*), lentil (*Lemna* sp.), pistachio or marsh lettuce (*Pistia stratiotes*) and *Larrea tridentata* which have a bioconcentration factor of 1000x. The hyperaccumulative species for Zn (and other metals) are: field grass (*Agrostis castellana*), accumulator for Zn, Al, Mn, Pb and hyperaccumulator for As; Indian mustard (*Brassica juncea*), hyperaccumulator for Zn, Cu, Ni, Pb and accumulator for Cd, Cr, U; rapeseed (*Brassica napus*), proposed for phytoextraction of metals Zn, Hg, Cr, Pb, Ag, Se; sunflower (*Helianthus annus*), proposed for phytoextraction of heavy metals, willow (*Salix viminalis*), accumulator for Zn, Ag, Cr, Hg, Se; sage (accumulator for Zn and hyperaccumulator for Cr, Ni, Pb; penny cress (*Thalspi caerulescens*), hyperaccumulator (concentration factor 10000x) for Zn, Cd, Cr, Co, Mo, Ni, Pb. The hyperaccumulators for Cd are a relatively small number of species, including *Thalspi caerulescens*, *Arabidopsis halleri*, *Aramanthus retroflexus* (Oros V., 2011).

Other genres of trees and shrubs that can grow on mining deposits are: wattles tree (*Acacia)*, maples tree (*Acer*), *Azadirachta*, *Albizia*, false indigo shrub (*Amorpha*), *Cassia*, *Dalbergia*, *Eucalyptus*, ash tree (*Fraxinus*), *Grevillea*, leadtrees (*Leucaena*), chinaberry tree (*Melia azedarach)*, mulberries tree (*Morus)*, plane trees (*Platanus*), Indian Beech Tree (*Pongamia pinnata*), Indian gooseberry (*Phyllanthus emblica*), rose (*Rosa*), *Rubus*, tamarisk (*Tamarix*), teak (*Tectona grandis*) (Gajić et al., 2018).

To obtain a stable persistent cover, it is important to use a mixed crop and combinations of grasses, shrubs and trees in phytoremediation of mining soil, as they are types of plants with different roles (Laghlimi et al., 2015).

Cultivation of *L. perenne* with *Alyssum murale* can help the former to accumulate Cu up to 10 mg/kg. Mn mobilization by *Alyssum* hyperaccumulating species can significantly increase Mn levels in *L. perenne* (Anjum et al., 2018).

Among the fast-growing woody plants, in addition to the genus Populus, there are also willow, pine, aspen, birch, beech, eucalyptus.

It is encouraged the selection of native plant species which do not require much maintenance and in time will form self-sustaining communities (Gajić et al., 2018).

The experimental data obtained by Malschi et al. (2013) after tests for phytoextraction, bioaccumulation and bioremediation on samples collected from waste dumps and tailings ponds from mining exploitations in Romania (Rodna, Bistrița-Năsăud, Fundu-Moldovei, Suceava, Aurul in Baia Mare and Târnăveni Chemical Installation platform, Mureș county), indicate that the species of perennial ryegrass (*Lolium perenne* L.), and common water lentils (*Lemna minor* L.), are useful as bioaccumulators and bioindicators for heavy metals and metalloids; *Lolium perene* is a strong bioaccumulator for Mn, Zn, Pb, As, Ba, Cu, Cd and moderate for V, Cr, Co, Ni.

Bacteria-associated plants can improve phytoremediation by altering solubility, bioavailability, and transport of heavy metals and nutrients by altering soil pH, releasing chelates (sideropores, organic acids, biosurfactants, glycoproteins), methylation, P solubilisation, or redox exchange (Ansari et al., 2018).

RESULTS AND DISCUSSIONS

Methods and discussions over the cost and the choice of remediation techniques

Combined remediation involves the application of two or more physical, chemical and/or biological remediation technologies. Thus, the limitations of using a single technology are completed, with various advantages in order to improve the efficiency of remediation (such as: chemically assisted phytoextraction, electrokinetic remediation coupled with complexing agents, electrokinetic remediation combined with phytoextraction, heat treatment facilitated by citric acid, soil washing coupled with chemical stabilization, chemical stabilization and phytoremediation (Gong et al., 2018).

Achieving the expected effect of the depollution technology should be based on phytoremediation, supplemented by physical and chemical microbial methods, to increase the bioavailability of heavy metals (Yang et al., 2019).

Despite their high efficiency, most of the depollution methods are expensive, environmentally destructive (do not allow natural recovery) and time consuming (Gomes, 2019).

The choice and applicability of a particular technology depends on the following factors: cost, long-term efficiency and performance, commercial availability, its general acceptance, applicability in cases of mixed soil pollutants (organic and inorganic compounds), reduction of toxicity, reduced mobility, reduced volume (Wuana and Okieimen, 2011).

Phytoremediation is cheaper (60-80%) than the physico-chemical process, because it does not require expensive equipment or exceptionally meticulous recruits (Jan et al., 2016).

The US Environmental Protection Agency (2004) reported that the total value for phytoremediation of the soil varied from 25 USD/t to 100 USD/t, compared to 300-500 USD/t for vitrification and 75-210 USD/t for soil washing. The Federal Remediation Technologies Roundtable (FRTR, in 2007) reported a cost range between 50 USD and 117 USD/m^2 for electrokinetic remediation and 33-32 USD/m^2 for soil washing. Martin and Ruby (2004) estimated a cost of 40-65 USD/m^3 for in situ chemical stabilization. Chang and Yen (2006) estimated a cost of 834 USD/ m^3 for a large-scale thermal desorption process (750°C for 3 hours) for the treatment of mercury-contaminated soil. The cost of landfill for a contaminated site and chemical recycling of contaminants varies between 100 and 500 USD/t, and the cost for electrokinetic monitoring is about 20-200 USD/t, while the costs involved in phytoextraction are 5-40 USD/t (Parmar et al., 2015).

For a large area of contaminated soil or sediment, in situ remediation is more suitable because it causes less disruption to the ecosystem, is simpler as a method and with lower costs than ex situ remediation (Song et al., 2017).

Methods of replacing contaminated soil, removing soil and isolating the soil will cost a large amount of labor and material resources, so they can be applied only on small areas of soil (Chao et al., 2014; Gomes, 2019).

Soil washing is another strategy to depollute soil contaminated with heavy metals, but it has been reported that it is not suitable for plant growth and development due to the impediment of biological and chemical activities. Chemical methods are not preferable due to changes in soil texture and structure, costs and generation of large amounts of sludge (Hasan et al., 2019).

Phytoremediation rejuvenates the vegetal soil layer, does not leave solid wastes and can successfully replace incineration, thermal vaporisation, solvent washing and soil washing, which are procedures that disturb the physico-chemical and biological qualities of the soil and form non-biodegradable waste (Jan et al., 2016).

Phytoremediation depends on climatic and meteorological conditions (Gong et al., 2018).

To reduce the contamination of soils polluted with heavy metals, the planting and harvesting of hyperaccumulating plants must be repeated. Depending on the target metal and the selected plant, the duration of the process can vary from 1 to 20 years (Parmar et al., 2015).

CONCLUSIONS

In Maramureș mining area, with the closure of mining activities near the mines of Cavnic, Borșa, Baia Sprie, as well as around the municipality of Baia Mare, numerous of polluted dumps of sterile remained.

The special problems regarding the quality of the environment in the county are determined by a historical pollution, resulting from the activities of extraction and processing of polymetallic and gold-silver ores deposits, which have affected for a long time with specific pollutants (gases, dusts and heavy metals) the environment factors.

In order to support development and urbanization, the areas used in the past in industry in Baia Mare mining basin must be introduced in use, so it is necessary the implementation of measures for the remediation of the affected areas.

In situ remediation offers a number of potential technical, economic and environmental benefits. In some cases, on-site remediation is the only means of eliminating pollutants when considering the extent of the contaminated area and cost-effectiveness.

The method presented in this article to restore the areas affected by heavy metal pollution is by implementing phytoremediation technology for soils contaminated with heavy metals.

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