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NEW APPROACHES REGARDING REMEDIATION TECHNIQUES OF HEAVY METAL CONTAMINATED SOILS FROM MINING AREAS

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ABSTRACT. Currently, heavy metal contamination of the soil and water near mining areas is a critical environmental problem that concerned all humanity due to its impact on ecological environment and human health. In order to remediate the heavy metal contaminated soils from mining areas, some technologies, generally based on physical, chemical, thermal and biological approaches, have long been in use to clean-up heavy metal contaminated soils to an acceptable and safe level. However, effectiveness of these methods depends a lot of soil type and characteristics, level of pollution and mixed contaminants present in soil. Moreover, some conventional technologies pose a secondary risk on environment. Thus, efficient eco-friendly techniques, based on natural materials or natural constituents of the soil needs development and research. This paper will provide an overview of the recent exploration and research, attempts of the remediation effectiveness assessment and developments regarding decontamination technologies applicable for the removal of heavy metals from soils near mining areas, being focused on new approaches regarding remediation methods. Moreover, limitations, financial aspects and future remediation research needs are also summary discussed.

Key word: *soil, heavy metals, soil remediation methods, mining areas, remediation effectiveness.*

INTRODUCTION

Nowadays, heavy metal contamination of the soils near mining areas is a widespread environmental problem in both developing and developed countries. Heavy metals such as cadmium, copper, zinc, lead, mercury, arsenic, contained in the residues from mining and metallurgical operations are often dispersed by wind and/or water after their disposal (Navarro et al., 2008).

Unlike organic contaminants, heavy metals are somewhat unique by the fact that they are highly resistant to either biologically or chemically induced degradation. Therefore, total heavy metal contents of soil persist for a long time after being introduced into the soil causing severe environmental problems, making the land resource unavailable and causing risk on human health since soil is the main resource to grow a part of human food (Khalid et al., 2017). Thus, remediation of soil contaminated by heavy metals is necessary in order to reduce the associated risks, make the land resource available for agricultural production, enhance food security, and scale down land tenure problems (Wuana and Okieimen, 2011).

Many technologies are available nowadays to remediate heavy metal contaminated soil near mining areas. Among these, immobilization, soil washing, and phytoremediation are frequently listed among the best available technologies for cleaning up heavy metal contaminated soils and have been mostly demonstrated in developed countries (Wuana and Okieimen, 2011).

Inspite all of these, these technologies have limitations considering efficiency, cost involved and secondary impact on the environment and/or human health due to the release of additional contaminants to the environment.

Considering all these there is a need to develop efficient technologies based on using natural low-cost materials that didn’t pose secondary risk to the environment and/or human health.

Thus, this paper brief describes the long-term used technologies to remove heavy metals from contaminated soil near mining areas with their limitations and the recent research and exploration performed in order to develop new methods to remediate highly heavy metals polluted soils, as the ones from mining areas.

Overview on soil contamination with heavy metals

In mining areas, one of the source that lead to soil pollution is represented by the runoff formed (figure 1) when rainwater reach sulfide-bearing minerals contained by mining deposits which is highly acidic, contain a high level of dissolved metals, sulphate and iron (Varvara et al., 2013).

When runoff reaches the soil, some heavy metals are dissolved and enter into solution, while others remain adsorbed and/or precipitated and move with the soil particles causing an increase in pollution that pose a significant risk to the environment and human health (Navarro et al., 2008).

Fig. 1. *The runoff formed when rainwater reach sulfide-bearing minerals from sterile dump located on Larga de Sus mining perimeter (Zlatna, Romania)*

Heavy metals are listed as priority pollutants by the United States Environmental Protection Agency (UEPA). For the level of toxicity, lead, mercury, arsenic and cadmium are ranked first, second, third, and sixth, respectively, in the list of US Agency for Toxic Substances and Disease Registry (ATSDR) (Singh and Prasad, 2015). Among these, heavy metals are reported to cause several disorders in humans including cardiovascular diseases, cancer, cognitive impairment, chronic anaemia, and damage of kidneys, nervous system, brain, skin, and bones (Khalid et al., 2017).

Numerous studies have reported important quantities, which exceed the permissible limits, of heavy metals in soil in several regions around the world.

In Europe, the polluted agricultural lands likely encompass several million hectares (Lestan et al., 2008).High contamination of soil was found by Navarro et al. (2008) in soil near *Cabezo Rajao* abandoned mine (Spain): 231 mg/kg Pb, 0.80 mg/kg Cd, 335 mg/kg Zn, 24 mg/kg Cu, 16 mg/kg As, 1.1 mg/kg Fe and 669 mg/kg Mn. Extremely high metal and metalloid content was also found in Greece near a mining and metallurgy complex (*Lavrion Technology and Cultural Park*): 64195 mg/kg Pb, 7540 mg/kg As, 4100 mg/kg Cu, 55900 mg/kg Zn and 6500 mg/kg Mn (Moutsatsou et al., 2006).

The situation is the same in the USA (United States of America), around 600,000 ha area has been contaminated with heavy metals (Khalid et al., 2017). Heavy metals are prevalent at almost all sites targeted by major remediation programs. For instance, metals are present in 77% of the Superfund sites (National Priorities List), in 72% of the Department of Defense (DOD) sites and in 55% of the Department of Energy (DOE) sites. The USEPA estimates that over 50 million cubic meters of soil at current NPL sites are contaminated with metals (Dermont et al., 2008).

In China the situation is even worse; the degraded land associated with mining activities reached about 3.2 Mha by the end of 2004, and the figure is increasing at an alarming rate of 46,700 ha per year. The proportion of soils that exceeds environmental standard reaches 16.1% (Lestan et al., 2008).

Due to pollution from mining at country level (Romania) there are 24.432 ha, of which 23.640 ha are excessively affected (Băbuț et al., 2011).

Moreover, all these mining sites over the world aren’t polluted only with heavy metals, organic pollutants being present in this soil as well. Every site has unique features considering soil properties and it is not possible to transfer a remediation technology from one site to the other (Jőger et al., 2013).

Considering all these, it is imperative to deploy innovative and site-specific remediationtechnologies which could feasibly and efficiently remediate highly heavy metal contaminated soils.

AVAILABLE REMEDIATION TECHNOLOGIES OF HEAVY METAL CONTAMINATED SOILS FROM MINING AREAS

During the last two decades many remediation technologies has been investigated, developed and have long been in use in order to reduce the total and/or bioavailable fractions of heavy metals in soils near mining areas. The approaches include isolation, immobilization, toxicity reduction, physical separation, and extraction. These conventional technologies, presented in table 1, used to treat heavy metal contaminated soils are based on physical, chemical, and biological processes (Sluser et al., 2011).

Technologies based on *isolation* were generally designed to prevent the movement of heavy metals by restricting them within a specified area when other remediation methods are not economically or physically feasible (Zhu et al., 2012). An isolation and containment system can work adequately, is not expensive (50-150$/ton) but there is no guarantee as to the destruction of the encapsulated contaminant (Khan et al., 2004).

Table 1. *Technologies for remediation of heavy metal-contaminated soils (Wuana and Okieimen, 2011).*

Category	Remediation technologies
Isolation	(i) Capping (ii) subsurface barriers
Immobilization	(i) Solidification/stabilization (ii) vitrification (iii) chemical treatment.
Toxicity and/or mobility reduction	(i) Chemical treatment (ii) permeable treatment walls (iii) biological treatment bioaccumulation, phytoremediation (phytoextraction, phytostabilization, and rhizofiltration), bioleaching, biochemical processes.
Physical Separation	
Extraction	(i) Soil washing, pyrometallurgical extraction, in situ soil flushing, and electrokinetic treatment.

Immobilization refers to decrease in metal mobility, bioavailability and bio accessibility of heavy metals in soil by adding immobilizing agents to the contaminated soils (Khalid et al., 2017). Among amendments used to immobilize heavy metals are reported cement, clay, zeolites, phosphates, minerals, microbes and organic amendments (Sun et al., 2016). Other studies have reported the potential of low-cost industrial residues such as red mud, bark saw dust, chitosan from crab meat canning industry, rice hulls, leaves in immobilization of heavy metals in contaminated soils (Wuana and Okieimen, 2011). The reported cost is 110$/ton.

Biological treatments are most viable option to rectify the natural condition of the soil and are based on using microorganisms, tolerant and accumulating plants to remove, decrease toxicity and/or mobility of heavy metals from soils (Guemiza et al., 2017; Khalid et al., 2017).Cost involved is between 50$ and 90$/m^3. Bioleaching and phytoremediation are the most used biological techniques.

Bioleaching, or bacterial leaching, consists in the extraction by solubilization of the metallic elements from contaminated soil using bacteria. This method does not destroy (eliminate) the pollutants, but it favors their segregation from the contaminated environment, the microorganisms having the property to oxidize the metals, transforming them into a more soluble form. Sur et al. (2016) investigated during 16 weeks the extraction of heavy metals (Cu, Zn, Cd and Pb) from polluted soils near Baia Mare area by in situ bioleaching and aerated bioleaching, using *Thiobacillus ferrooxidans* type of microorganisms in 9K medium. Results (figure 2) indicated that the extraction efficiency of metals is much higher if aeration is introduced in the process (Cu: 17 - 27%; Zn: 14 -27%; Cd: 8 - 14%. Pb: 7 - 13%).

However, this techniques requires long periods to efficiently remove the contaminants from soils and it is only efficient for surface contamination and for the most mobile metals present into the soil. These metals present on the surface of the soil can also be extracted by electro kinetic processes which consist on the application of low intensity electric current between a cathode and an anode inserted into the contaminated soil, making the ions and the small charged particles to be transported to the anode or to the cathode according to their charges (Guemiza et al., 2017). Cost involved: 50-225 $/ton.

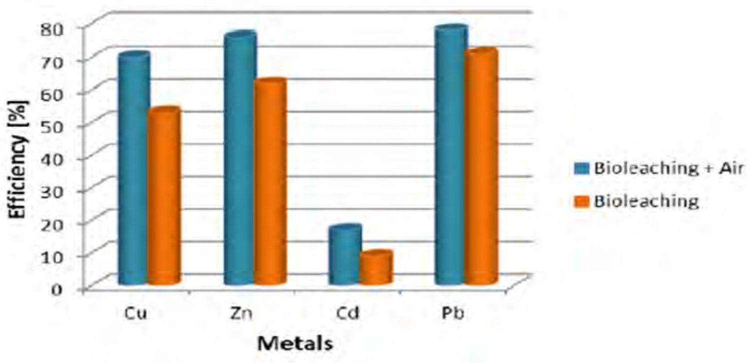

Fig. 2. *Efficiency of metals extraction when by in situ bioleaching and aerated bioleaching was applied (Sur et al., 2016)*

Chemical methods include, among others, soil washing and soil flushing. Soil washing is one of the few treatment alternatives for the elimination of heavy metals from highly contaminated soils such the ones from mining areas, which can be applied to pilot/full-scale field remediation (Dermont et al., 2008) being feasible also to remove toxic metals attached to the fine particles of soil. Also, cost varies between 75->150$/ton. Torres et al. (2012) demonstrated that Cd, Zn, and Cu could be washed with efficiencies up to 85.9%, 85.4%, and 81.5% respectively.

Over the past years, scientists have tried to optimize the extraction of heavy metals from contaminated soils by chemical leaching using different extractants. These chemical agents (synthetic and organic acids, bases, surfactants, alcohols, chelating agents and cyclodextrins) are used to transfer metal from contaminated soil to the aqueous solution (Guemiza et al., 2017).

All these soil washing extractants have been developed on a case-by-case basis depending on the contaminant type at a particular site. Removal efficiency of heavy metals depends on the type of the extractant used, contaminant type, presence of other contaminants and on the characteristics of the soil.

Despite the proven efficiency of acid extraction (more than 90%-figure 3) in full-scale applications for non-calcareous soils, strong acids attack and degrade the soil crystalline structure at extended contact times.

Thus, strong acids have been considerate inappropriate to remove heavy metals from soil (Wang at al., 2013).

For less damaging washes, organic acids and chelating agents are often suggested as alternatives to straight mineral acid use (Wuana and Okieimen, 2011).

The most used and studied chelating agents are: EDTA (ethylenediaminetetraacetic acid), NTA (nitrilotriacetic acid) and DTPA (diethylenetriaminepentaacetic acid) due to their effectiveness and low cost.

However it was reported that using EDTA for soil washing may destabilize the soil aggregate stability, and mobilize colloids and fine particles (Karthika et al., 2016).

Thus, in recent years biodegradable chelating agents such as EDDS ([S,S]ethylenediaminedisuccinic acid, which is a stereoisomer of EDTA), IDSA (iminodisuccinic acid), NTA (nitrilotriacetic acid), ASP (2-Amino-3-sulfhydrylpropanoic acid) and MGDA (methylglycinediacetic acid) have received increasing attention (Liu et al., 2015; Karthika et al., 2016).

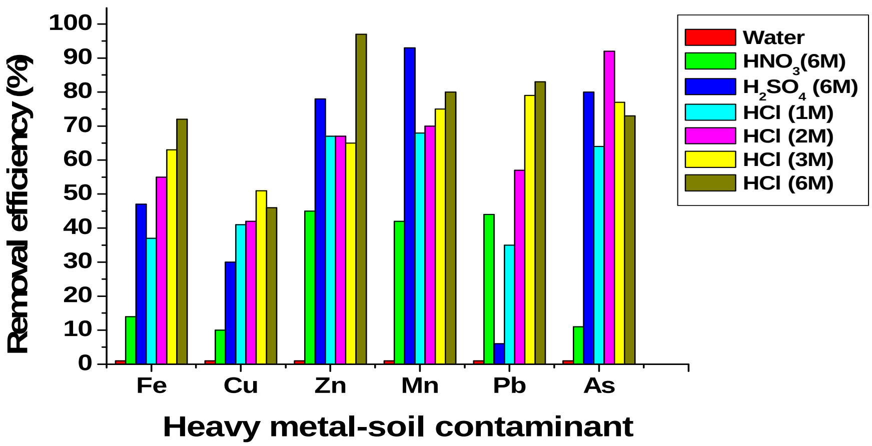

Fig. 3. *Removal efficiency of heavy metals from soil using acids as washing agents (according to Moutsatsou et al., 2006)*

Extraction of metals in the soil by washing with trisodium salt of EthyleneDiamine-Disuccinic acid (Na_3EDDS) was investigated by Eng. Maria Szanto (Prodan) under scientific coordination of Prof. Dr. Eng. Valer Micle. There were used: soil samples taken from the vicinity of S.C. Sometra Copşa Mică, an area highly polluted with heavy metals due to metallurgical activities.

Soil samples were subjected to washing with Na_3EDDS for 2, 4, 6, respectively 8 hours, the Na_3EDDS solution concentration was 0.4, 0.5 and 0.6 [%] (Szanto (Prodan), 2012).Results have shown that extraction of metals by washing with Na_3EDDS has a very high yield: 85.54 % for Cu; 98.91 % for Zn; 97.59 % for Pb; 100 % for Cd, when treating soil for 8 hours and using a concentration of 0.6 % Na_3EDDS (for Cu, Zn and Pb) and a concentration of 0.4 % Na_3EDDS (for Cd). Concentration of Pb in soil (figure 4) was below alert threshold , according to Order 756/97, when treating soil for 8 hours and using 0.5% Na_3EDSS.

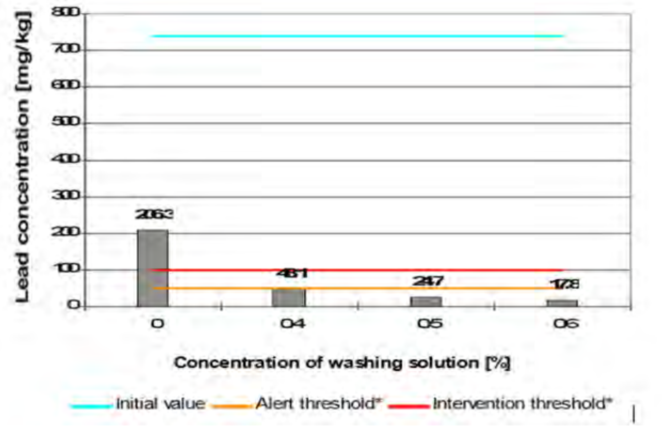

Fig. 4. *Lead concentration in the soil after Na3EDDS treatment for 8 hours (Szanto (Prodan) et al., 2012)*

Although high efficiencies were obtained in case of Cu, Zn, Pb, Cr, Ni and Cd when soil is washed with above chelating agents, it was reported that these are toxic and carcinogenic (Jiang et al., 2011).

Considering all above limitations of the previous long-term used technologies, nowadays there is a need to develop and investigate other inexpensive methods that may be efficient, eco-friendly and without posing risk to human health for remediating highly heavy metal contaminanted soil.

NEW APROACHES REGARDING REMEDIATION METHODS OF HEAVY METALS CONTAMINATED SOILS FROM MINING AREAS

The overall objective of any soil remediation approach is to create a final solution that is protective of human health and the environment (Wuana and Okieimen, 2011). None of the options available nowadays retain the healthy state of the soil being in the same time efficient and inexpensive. Thus, there is a need to increase research on this field. Some attempts, described below, were made until today on this line.

Soil washing. Currently, research is performed in the field of optimizing soil washing, increase efficiency on multi metal contaminated soils and reducing costs. Rinsing steps and repeated washing are conducted to improve the removal efficiency of heavy metals which can reduce the consumption of washing agents and the washing costs (Torres et al., 2012). Similarly, combined use of different extractants also improves heavy metals washing efficiency especially for multi-metal contaminated soils (Guo et al., 2016). For example, Wei et al. (2016) reported that phosphoric- oxalic acid-Na_2EDTA order based soil washing enhanced heavy metals removal efficiency by 41.9% for As and 89.6% for Cd.

On the other hand, investigations regarding efficiency of using other substances as reagents in removing heavy metals through soil washing is now conducted.

On this line, few studies have investigated chitosan and humic acids (that are natural constituents of the soil) to extract heavy metals from highly heavy metal polluted soil (Kulikowska et al, 2015a; Boechat et al., 2016; Jiang et al., 2011; Meng et al., 2017; Gusiatin et al., 2017). Nowadays cheaper sources of humic substances are under research. Kulikowska et al. (2015a) investigated the effectiveness of using humic substances extracted from sewage sludge compost for remediating soil polluted with heavy metals through soil washing.

The results indicated that under optimum conditions, a single washing removed 80.7% of Cu and 69.1% of Cd from polluted soil. The same research team revealed in another study that percent of metal removal from soils when humic substances from compost were used was 79.1–82.6%, 51.5–71.8%, 44.8–47.6%, 35.4–46.1%, 27.9–35.8% in case of Cd, Cu, Pb, Ni and Zn, respectively (Kulikowska et al., 2015b). Also, it was reported by Meng et al. (2017) that humic substances being environmentally benign, can improve soil physical, chemical, and biological properties leading to a healthy state of the soil.

Combined Remediation. Combined remediation has gained, over the last years, much attention of researchers from all over the world and involves two or more different types of physical, chemical, or biological remediation technologies. The combination of diverse technologies can not only overcome the problems caused by using any one technology alone, but also take advantages of all and enhance the remediation efficiency (Song et al., 2017).

Therefore, batch experiments (flow diagram of the experiments is illustrated in figure 5) were conducted in order to determine the effectiveness of *washing process combined with sieving* to remediate soil from an abandoned mine in China (Liao et al., 2016).

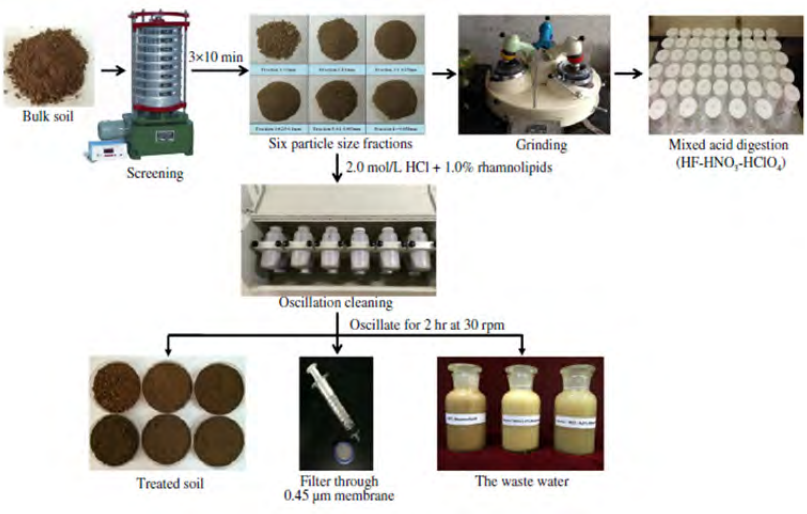

Fig. 5. *Flow diagram of the experiments conducted in order to determine the effectiveness of washing process combined with sieving (Liao et al., 2016)*

Results of the experiments (table 2) indicated that larger particle size did not necessarily result in greater removal efficiency for arsenic and heavy metals. The highest removal efficiencies by washing for Pb, Cd, Zn, and As were obtained in the fraction of >2 mm. However, the small particle size fractions can also achieve high heavy metal removal efficiencies. Compared with the original efficiency, the equivalent efficiencies for Pb, Cd, and Zn had been enhanced, whereas the equivalent efficiencies for As and Cu were lower (Liao et al., 2016).

Table 2. *Removal efficiency of the variously sized soil (Liao et al., 2016)*

Soil particle size	Pb	Cd	Zn	Cr	As	Cu
>2 mm	87.4%	87.3%	76.5%	10.4%	45.5%	64.9%
1–2 mm	74.7%	80.0%	66.7%	37.1%	26.5%	61.7%
0.25–1 mm	84.3%	80.6%	67.4%	25.0%	21.8%	65.5%
0.1–0.25 mm	84.3%	81.7%	60.9%	34.7%	23.5%	65.3%
0.053–0.1 mm	85.4%	78.1%	60.7%	25.9%	10.2%	62.4%
<0.053 mm	87.2%	79.5%	66.0%	25.1%	27.2%	66.6%

Chelate-Assisted (Induced) Phytoextraction. The use of biodegradable chelants (NTA, EDTA and EDSS) in improving the uptake of metals by plants and in limiting the leaching of metals from soil has become an attractive field of research.

When the chelating agent is applied to the soil, metal-chelant complexes are formed and taken up by the plant, mostly through a passive apoplastic pathway. Several previous studies showed significant increase in plant accumulation of Pb, Cd, Ni, Cu, and Zn from contaminated soil in the presence of synthetic chelates (Khalid et al., 2017). Also, it was reported that tartaric, acetic, malic, citric and oxalic acids, that are natural root exudates, can also be used for heavy metals phytoextration as an alternative to persistent synthetic chelates (Abbas et al., 2015).

Moreover, phytoremediation of heavy metals contaminated soils was ameliorated by adding exogenous humic substances, thus making contaminants more available to phytoextraction (Floris et al., 2017).

Chelate-assisted phytoextraction of heavy metals from soil has not gained considerable acceptance because of its high leaching risk, relatively low efficiency and high cost.

Microbial assisted phytoremediation. Recently, it was reported that inoculation of Burkholderia sp. (Z-90) enhanced heavy metals removal efficiency in soil by 31% for Pb, 32% for As, 44% for Zn, 37% for Cd, 52% for Mn and 24% for Cu (Yang et al., 2016; Ma et al., 2015) reported that Sedum plumbizincicola significantly enhanced Cd uptake (43%), whereas Bacillus sp. (E1S2) enhanced the Zn accumulation (18%) in Sedum plumbizincicola.

Modified plants. Another field of research refers to the possibility to create an *ideal plant species* for clean-up of heavy metals contaminated soil through the *introduction of foreign resistant genes*. Several researchers have proposed that establishing ideal crop hyper accumulator in the future can be an ideal choice due to its feasibility and applicability in the field of which current emphasis is scarce. By mean of genetic engineering, ability of a plant to accumulate, translocate and detoxify heavy metals can be significantly enhanced (Khalid et al., 2017).

Interestingly, in the last few years, the possibility of planting metal hyper accumulator crops over a low-grade ore body or mineralized soil, and then harvesting and incinerating the biomass to produce a commercial bio-ore has been proposed though this is usually reserved for use with precious metals. This process called *phytomining* offers the possibility of exploiting ore bodies that are otherwise uneconomic to mine, and its effect on the environment is minimal when compared with erosion caused by opencast mining (Wuana and Okieimen, 2011).

Ultrasonic combined with mechanical soil washing process. The effect of high-power ultrasound on the conventional mechanical soil washing process was investigated in a large lab-scale 28 kHz sonoreactor by Park and Son (2017) in order to remove heavy metals from soil. Results obtained indicated that removal efficiencies were enhanced with 70%, 140% and 55% in case of Cu, Pb and Zn, respectively.

CONCLUSIONS

Nowadays, remediating a highly metal-contaminated soil, containing slags and sulphur compound waste as a result of mining and metallurgical activities it is a challenge for researches from all over the world.

Available technologies have limitations considering efficiency in highly heavy metal contaminated soils, cost involved and environmental and health risk.

In this case, using combined technologies (described on this paper) based on natural low-cost materials, could be a feasible, inexpensive and efficient solution that could overcome the problems caused by using any one technology.

Inspite all of these, research and development actions are still needed for emerging technologies to achieve a healthy state of the soil and to bring them to the market place for full-scale implementation.

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