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Study of metals leaching from nonferrous mine tailings in organic and inorganic extractants

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Abstract: Metals (Fe, Cr, Cu, Zn, Pb, Mn, Ba, Al and Sr) leaching from tailings resulted from non-ferrous ores processing was assessed using different organic (ethylenediaminetetraacetic acid - EDTA, citric acid - $C_6H_8O_7$, oxalic acid - $C_2H_2O_4$, acetic acid – $C_2H_4O_2$, succinic acid - $C_4H_6O_4$) and inorganic (nitric acid – HNO_3) extractants. The variation of metals leaching with the sampling depth (0-5 cm, 5-10 cm, 10-20 cm and 20-40 cm) was also studied. Generally, the highest leaching was obtained for Fe, Cu, Zn, Mn, Al in $C_2H_2O_4$ followed by Pb, Ba in EDTA and Sr, Mn in HNO_3. The enrichment factor (*EF*), contamination factor (*CF*) and geoaccumulation index (I_{geo}) were calculated to reveal the pollution pattern. The *EF* revealed minor enrichment with Cr, very severe to extremely severe enrichment with Zn and Pb, moderate to moderately severe enrichment for Fe and Sr, and severe enrichment for Cu and Mn. The *CF* indicated low contamination with Al and Cr, moderate contamination with Fe and Sr, considerable contamination with Cu and Mn, very high contamination with Zn, Pb and considerable to very high contamination with Ba. I_{geo} showed no pollution with Cr, Fe, Sr, Al, moderate Mn and Cu pollution, moderate to heavy Ba pollution and heavy to extreme Pb and Zn pollution.

Key-Words: metals, tailings, batch leaching, organic acids, pollution indices

1 Introduction

The large volume of wastes generated following exploitation of mineral resources determined an increase of metal contamination in soil and water, and raised important environmental concerns due to the mobility, toxicity, bio-accumulation and persistence of metals [1-3]. Generally, potentially toxic metals in tailings are found to be associated to various primary or secondary minerals, and can be mobilized in different environmental conditions [2, 4].

Leaching in different extractants allows to estimate the metals mobility in different conditions through the solubilization of various metal pools [4]. Single extractions are fast, easy to use and pose low risk for sample losses during phase separations and washing steps. Their main disadvantage is lack of selectivity [5].

Water is the universal solving agent that dissolves the most mobile metal fraction [2]. Leaching test with water can solubilize the metals weakly bound or the inorganic soluble salts [6].

Metal dissolution by low-molecular-weight organic acids such as oxalic, acetic, succinic, appears to be typical for mobile metal fraction assessment [7]. Citric-, oxalic- and succinic-acids favor mobilization of metals from tailings in mild acidic condition, by replacing adsorbed metals and by forming stable metal-organic complexes [1, 8].

EDTA is a non-selective multi-dentate chelating agent that forms stable complexes with metals, and estimates the potential phytoavailable metal fraction [9, 10]. EDTA is able to displace the exchangeable, carbonate, and reducible fractions of metals [7]. EDTA was used in numerous studies to assess the phytoavailable metal fractions [11].

Acetic acid is a weak acid that dissolve the easily mobile and mobilizable metals, usually bound by ionic exchange with slight change in pH of tailings. [10].

Various contamination indices as the contamination factor (*CF*), the enrichment factor (*EF*) and the geoaccumulation index (I_{geo}) are used to assess the intensity and presence of anthropogenic pollution. For the calculation of these indices, a metal concentration is normalized with respect to a reference element [3, 12].

The main objectives of the present study were to:

(i) determine the concentration and profile distribution of Fe, Cu, Zn, Pb, Mn, Ba, Al and Sr in tailings, *(ii)* investigate the metals leaching in various extractants (Fig. 1), and *(iii)* evaluate the pollution pattern using the enrichment factor (*EF*), contamination factor (*CF*), and geoaccumulation index (I_{geo}).

The paper is structured as follows: section 2 describes the extraction procedure for total metal content from the tailings and the leaching procedure used to determine the concentration and profile distribution of Fe, Cr, Cu, Zn, Pb, Mn, Ba, Al and Sr in the investigated tailings using various extractants. Section 3 presents in details the leaching efficiency of metals (%) from tailings sampled at different depths from the studied area, the pollution element patterns using different indices and their profile distribution. Finally, the paper ends with our conclusions.

2 Materials and methods

2.1 Study area

Romania has important nonferrous ore deposits, grouped mainly in the North and North Vest of the country [13]. The history of precious and base metals mining gets back to ancient time and led to important amounts of wastes, usually piled up in the nearby river bed. In Rodna mining area, Au and Ag were mined since the 12th century, and starting from the 19th century Pb and Zn associated with Cu, Sb, Bi, Ca, Au, Ag were exploited. The ores were treated by flotation and the resulting tailings stored in several impoundments. Situated in the Somes Mare river basin, Anies impoundment was built in 1982 and stores 1.2 million m^3 of tailings. From 2005, all mining activities in the area ceased and presently, the impoundment is completely revegetated and stabilized.

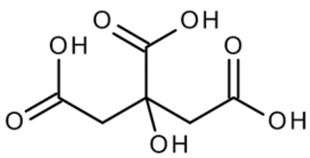
2.2 Sample collection

The samples were collected at 0-5, 5-10, 10-20 and 20-40 cm depths using a stainless steel hand auger. The tailings were dried in an oven at 60 ºC for 8 hours and grounded to pass the 2 mm sieve.

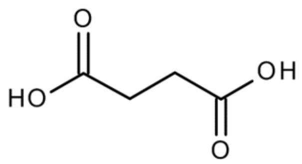
2.3 Extraction procedure for total metal content

All used chemicals were of analytical grade and purchased from Merck, Germany.

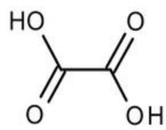
Total metal content was determined following a three steps procedure in Teflon vessels according to Froger et al. [14] *(i)* addition of 5 ml of 30% HF and 1.5 ml of 67% $HClO_4$ to 100 mg tailings, heating at 180°C for 3 h and evaporation to dryness, *(ii)* addition of 1.5 ml of 65% HNO_3 and 4 ml of 37% HCl to the residue of step one, heating at 150°C for 3h and evaporation to dryness, *(iii)* addition of 3 mL of 65% HNO_3 to the residue of step 2, heating at 150°C for 3h and evaporation to dryness. The residue of step 3 was dissolved in 1 mL of 65% HNO_3 and transferred in 50 mL volumetric flasks.



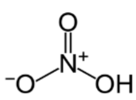
Citric acid



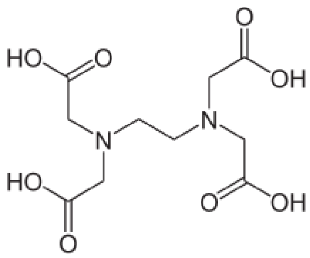
Succinic acid



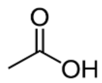
Oxalic acid



Nitric acid



EDTA



Acetic acid

Fig. 1. Chemical structure of used organic acid

2.4 Leaching procedure

Leaching experiments were conducted by using 0.1 N ethylenediaminetetraacetic acid (EDTA), citric acid ($C_6H_8O_7$), oxalic acid ($C_2H_2O_4$), acetic acid ($C_2H_4O_2$), succinic acid ($C_4H_6O_4$) and nitric acid (HNO_3) and H_2O.

To 1 g of tailings 25 mL of extractant was added and then shaken at 15 rpm, at room temperature, for 4 hours. The extracts were centrifuged for 10 minutes at 3500 rpm and then filtrated through 0.45 µm cellulose acetate membrane filters. The experiments were performed in duplicate.

The extracts were analyzed using an inductively coupled plasma optical emission spectrometer (OPTIMA 5300 DV, Perkin Elmer, Norwalk, USA).

2.6. Anions and pH determination

The anions (SO_4^{2-}, Cl^-, NO_2^-, NO_3^-, PO_4^{3-}) concentrations were measured using a 761 Compact ion chromatograph, (Methrom, Switzerland) in 1:10 tailings to water extract. The pH of the tailings was measured in the 1:10 extract using a Seven Excellence multiparameter (Mettler Toledo, Switzerland).

3 Results and discussions

3.1 Total metal content

Table 1 shows the total metal and anion content in the investigated tailings.

The pH values were circumneutral, ranging between 7.60 - 7.87.

The highest Cr, Cu, Al, Sr and the lowest Pb, Zn, Fe contents were found at 20-40 cm depth. Samples collected from 10-20 cm depth had the lowest Cr, Ba, Cu, Mn, Al and the highest Zn contents, while those from 0-5 cm depth had high Mn and Pb. The highest Ba amount was determined at 5-10 cm depth. The Fe contents were comparable up to 20 cm depth, while at 20-40 cm depth a slight decrease was noticed.

Generally, the Cr, Mn and Al contents decreased with the depth up to 10-20 cm and rapidly increase at higher depths. Cu, Ba and Sr contents increased up to 5-10 cm, then decreased at 10-20 cm and increased again at 20-40 cm depth. The Zn content decreases with the increase of depth, while the Pb content decreases up to 5-10 cm and remains constant after.

The SO_4^{2-} content was high up to 10 cm depth and very high at 10-20 cm, but decreased sharply under the detection limit after 20 cm depth. The PO_4^{3-}, NO_2^- and F^- contents were low and almost constant, while the highest peak of NO_3^- was observed at 5-10 cm.

3.2 Soluble metal content

The leaching efficiency of each extractant was calculated as the ratio between the metal content in the extract and the total metal content.

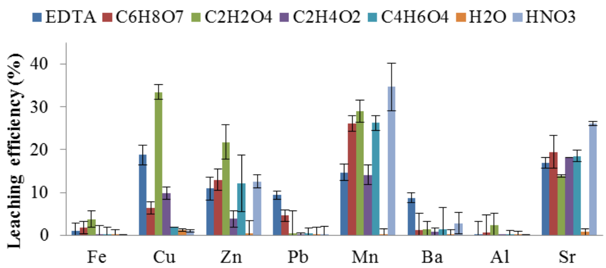


Fig. 2. Leaching efficiency of metals (%) from tailings sampled at 0-5 cm depth

The results showed that $C_2H_2O_4$ was the most

Table 1. The total content of metals and anions in the Anies samples (mg/kg)

Parameter	0-5 cm	5-10 cm	10-20 cm	20-40 cm
Fe	56120	49260	54242	36671
Cr	39.8	21.3	9.5	43
Cu	95.4	107.3	50.1	122
Zn	2063	1871	2584	1403
Pb	734	483	598	418
Mn	3405	2204	1852	2305
Ba	3545	10139	2463	2897
Al	31940	21434	10835	35621
Sr	247	560	361	756
F^-	<0.25	<0.25	2.85	<0.25
Cl^-	26.5	27	18.5	30
NO_2^-	<0.25	<0.25	3.28	<0.25
NO_3^-	13	35.3	9.1	19
PO_4^{3-}	<0.25	<0.25	<0.25	<0.25
SO_4^{2-}	1700	1900	10000	<0.25

effective reagent for Fe (3.8%), Cu (33.4%), Zn (21.8%) and Al (2.3%) (Fig. 2), while EDTA for Pb (9.4%) and Ba (8.7%). HNO_3 gave the highest leaching efficiency for Mn (34.6%), and Sr (26.1%).

In case of tailings collected at 5-10 cm depth (Fig. 3), the highest leaching efficiency for Fe (8.7%), Cu (29.8%), Zn (20.8%) and Al (1.9%), were achieved with $C_2H_2O_4$, for Pb (17.8%), and Ba (17%) in EDTA, while for Mn (29.5%) and Sr (19.8%) in HNO_3.

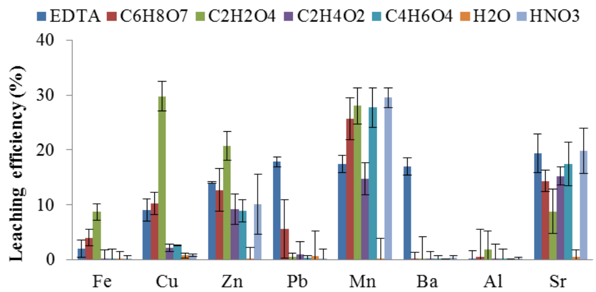


Fig. 3. Leaching efficiency of metals (%) from the tailings sampled at 5-10 cm depth

For tailings at 10 - 20 cm depth, the leaching efficiency of Fe (3.9%), Cu (34.5%), Zn (16.6%), and Al (2%) were higher by using $C_2H_2O_4$ as extraction reagent, with the exception of Pb (32.8%), and Ba (0.5%) elements that appeared to be leached in a higher percentage by EDTA extraction reagent (Fig. 4). Moreover, the HNO_3 was more efficient for Mn (40%), and Sr (30.3%) leaching from the tailings collected at 10-20 cm depth.

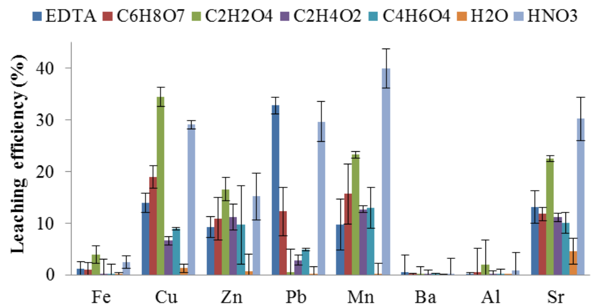


Fig. 4. Leaching efficiency of metals (%) from the tailings sampled at 10-20 cm depth

For tailings at 20-40 cm depth, as shown in Fig. 5, $C_2H_2O_4$ gave the highest leaching efficiency for Fe (12.4%), Cu (29.5%), Zn (33.3%) and Al (3.1%), while EDTA gave the highest leaching efficiency for Pb (36.8%) and Ba (1.5%). The highest percentage of Sr (21.4%) and Mn (35.3%) leaching was obtained using HNO_3.

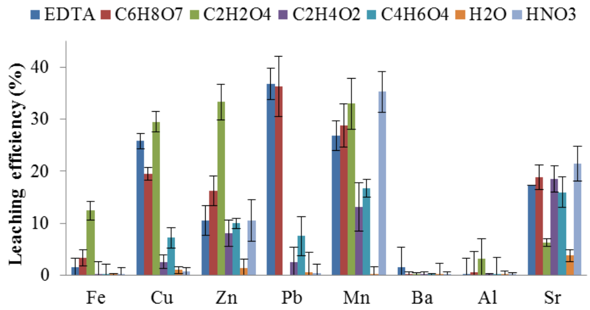


Fig. 5. Leaching efficiency of metals (%) from the tailings sampled down to 20-40 cm depth

3.3. Profile distribution of metals mobility in tailings

The profile distribution of Fe, Cr, Cu, Zn, Pb, Mn, Ba, Al and Sr mobility in tailings at different depths is presented in Figure 6.

The maximum value for Ba leached in EDTA (17.0%) was obtained in the tailings collected from 5-10 cm depth. The maximum values for Cu (34.5%) leached in $C_2H_2O_4$, Sr (30.3%) and Mn (40.0%) leached in HNO_3 were obtained in the tailings collected from 10-20 cm depth. The highest leaching percentage for Zn (33.3%), Fe (12.4%) and Al (3.1%) leached in $C_2H_2O_4$, and Pb (36.8%) leached in EDTA was obtained from the tailings sampled at 20-40 cm depth.

Generally, the highest leaching efficiency for Fe, Cu, Zn, Mn and Al was obtained in $C_2H_2O_4$, for Pb and Ba in EDTA and for Sr and Mn in HNO_3.

3.4 Contamination degree assessment

In order to evaluate the degree of contamination of the considered elements, the enrichment factor (*EF*), contamination factor (*CF*) and geoaccumulation index (I_{geo}) were calculated using the total metal content (Table 1).

The background values used in current study in the indices count were: Fe 35000, Cr 35, Cu 25, Zn 71, Pb 20, Mn 600, Ba 550, Al 80400 and Sr 350 mg/kg [15, 16].

3.4.1 Enrichment Factor (*EF*)

The Enrichment Factor (*EF*) was used to determine the impact of the human activities [17]. *EF* was calculated according to Eq. 1:

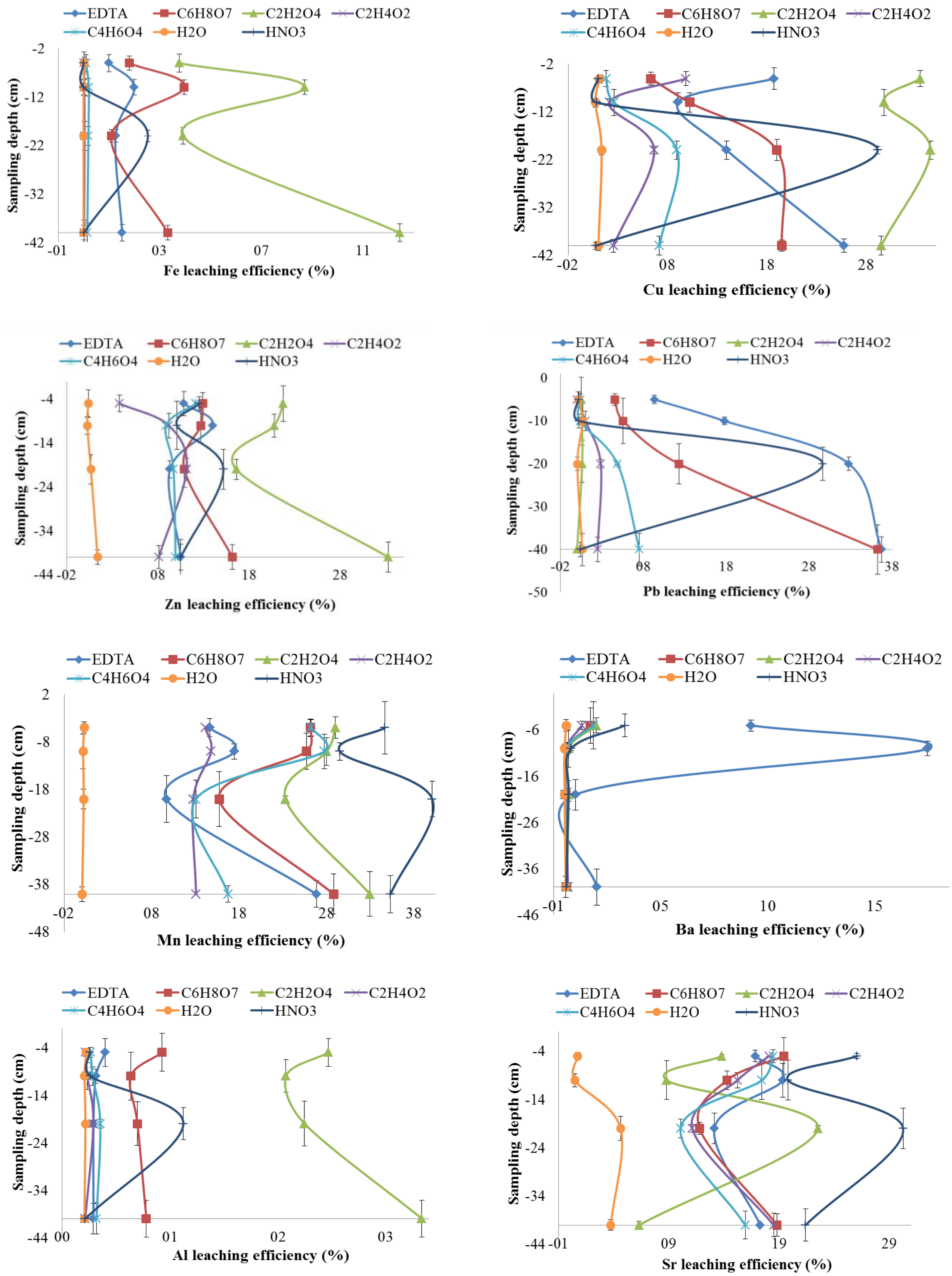


Figure 6. Profile distribution of Fe, Cr, Cu, Zn, Pb, Mn, Ba, Al and Sr in tailings at different depths

$$EF=\frac{M_S/Al_s}{M_b/Al_b} \quad (1)$$

where, M_s is the metal content from the tailings, Al_s is the Al content from the tailings, M_b is the metal background value and Al_b is the Al background value in the upper continental crust according to Taylor and McLennan [15] and McLennan [16].

The $EF < 1$ indicates no enrichment, $1 < EF \leq 3$ minor enrichment; $3 < EF \leq 5$ moderate enrichment; $5 < EF \leq 10$ moderately severe enrichment; $10 < EF \leq 25$ severe enrichment, $25 < EF \leq 50$ very severe enrichment, $EF > 50$ extremely severe enrichment. [12, 13].

The *EF* values (Table 2) revealed a very to extremely severe enrichment with Zn, and Pb, moderate to moderately severe enrichment with Fe, and Sr, severe enrichment with Cu and Mn and minor enrichment with Cr.

Table 2. The enrichment factor (*EF*) results

	0-5 cm	5-10 cm	10-20 cm	20-40cm
Fe	4.0	5.3	11.5	2.4
Cr	2.9	2.3	2.0	2.8
Cu	9.6	16.1	14.9	11.0
Zn	73.1	98.8	270.1	44.6
Pb	92.4	90.6	221.9	47.2
Mn	14.3	13.8	22.9	8.7
Ba	16.2	69.1	33.2	11.9
Sr	1.8	6.0	7.7	4.9

Generally, increasing enrichment trend with depth was found up to 20 cm depth for Fe, Zn, Pb and Sr, followed by an important decrease of the enrichment at 20-40 cm depth. The enrichment of Cr, Cu, Mn, was comparable at all depths. Comparable enrichment degrees were found for Cu, Pb and Zn in tailings located in the Aries river basin, Western Romania [13]. In another study, Muzerengi, (2017) found significant contamination with As ($3.5<EF>11$) and Cd ($4.8<EF>9.5$) for soil near tailings from a gold mine from Limpopo Province of South Africa [18]. In Mongolia the *EF* indicated very severe enrichment with Cu and moderately severe enrichment with Ni in mining tailings [19].

3.4.2 Contamination factor (*CF*)

The Contamination factor (*CF*) index can be calculated using the following equation:

$$CF=\frac{C_m}{C_b} \quad (2)$$

where C_m is the concentration of the element in the tailing and C_b is the background concentration of the element [12].

According to *CF* values, tailings can be classified as having low ($CF < 1$); moderate ($1 < CF \leq 3$); considerable ($3 < CF \leq 6$); and very high contamination ($CF > 6$) with metals [12, 13].

The tailings presented low contamination with Al and Cr, moderate contamination with, Fe and Sr, considerable contamination with Cu and Mn, and very high contamination with Zn, Pb and considerable to very high contamination with Ba (Table 3).

Table 3. The contamination factor (*CF*) results

	0-5cm	5-10cm	10-20cm	20-40cm
Fe	1.6	1.4	1.5	1.0
Cr	1.1	0.6	0.3	1.2
Cu	3.8	4.3	2.0	4.9
Zn	29.1	26.4	36.4	19.8
Pb	36.7	24.2	29.9	20.9
Mn	5.7	3.7	3.1	3.8
Ba	6.5	18.4	4.5	5.3
Al	0.4	0.3	0.1	0.4
Sr	0.7	1.6	1.0	2.2

The *CF* for Fe, Cu, Zn, Pb, Mn, Al, Cr and Sr was comparable at all depths. The *CF* for Ba increases at 5-10 cm and decreases comparable at 10-20 cm depth.

Rashed (2010) found that the tailings from gold mine Allaqi Wadi Aswan, Egypt are characterized as very strong contaminated with As and Hg ($CF>6$), considerable contaminated with Pb ($CF>4$), moderate contaminated with Cd, Co, Cr, Ni and Zn ($CF>1$) [20].

3.4.3 Geo-accumulation index (I_{geo})

The I_{geo} was calculated using Eq. 3.

$$I_{geo}=\log_2 \frac{C_m}{1.5 \times C_b} \quad (3)$$

The I_{geo} categorizes the tailings into practically unpolluted ($I_{geo} < 0$), unpolluted to moderately polluted ($0 < I_{geo} \leq 1$), moderately polluted ($1 < I_{geo} \leq 2$), moderately to heavily polluted ($2 < I_{geo} \leq 3$), heavily polluted ($3 < I_{geo} \leq 4$), heavily to extremely polluted ($4 < I_{geo} \leq 5$) and extremely polluted ($I_{geo} > 5$) [12, 13, 21].

Based on the I_{geo} results (Table 4), the tailings were considered practically unpolluted with Cr, Fe, Sr and Al, moderately polluted with Mn and Cu, moderately to heavily polluted with Ba and heavily to extremely polluted with Zn and Pb.

Table 4. The geoaccumulation index (I_{geo}) results

	0-5 cm	**5-10 cm**	**10-20 cm**	**20-40cm**
Fe	0.03	-0.1	0.1	-0.5
Cr	-0.4	-1.3	-2.5	-0.3
Cu	1.4	1.5	0.4	1.7
Zn	4.3	4.1	4.6	3.7
Pb	4.6	4.0	4.3	3.8
Mn	1.9	1.3	1.0	1.4
Ba	2.1	3.6	1.6	1.8
Al	-1.9	-2.5	-3.5	-1.8
Sr	-1.1	0.1	-0.5	0.5

The I_{geo} results regarding the Fe, Mn, Zn, Pb, Al and Sr concentration were similar at all depths. I_{geo} for Cu decreases at 10-20 cm depth and increases at 20-40 cm depth. Comparable I_{geo} values were found for Cr, Pb and Cu in tailings located in the Aries river basin [13].

4 Conclusion

The metals content in mine tailings was assessed by using diverse extraction procedures. $C_2H_2O_4$ gave the best leaching efficiency for Fe, Cu, Zn, Mn and Al, while EDTA for Pb and Ba, and HNO_3 for Sr and Mn. Tailings were found to be non-contaminated with Cr and Al, moderately contaminated with Fe, and Sr, considerably contaminated with Cu, Mn, and highly contaminated with Zn, Pb. The high percent of mobile metal amounts found in tailings can pose a serious threat to the ecosystems. The obtained results can be a good reference for further studies for mining tailings remediation.

Acknowledgments

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