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HEAVY METALS SOIL POLLUTION STATE IN RELATION TO POTENTIAL FUTURE MINING ACTIVITIES IN THE ROŞIA MONTANĂ AREA

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Abstract: The aim of this paper is to establish the abundance of heavy metals in the soils affected by the past Rosia Montana gold and silver ore mining, and in currently unaffected soils that will be impacted by the proposed Rosia Montana project that foresees the expansion of the ore exploitation and a new processing facility. The soil cover of the Rosia Montana area consists of five soil types: Eutricambosols, Districambosols, Regosols, Lithosols, and Aluviosols. The first two types are prevalent; they cover 73.83% of the total researched surface (1,646 ha). In the soils from the areas where mining activities have been carried out, the total content of the heavy metals (Cd, Co, Cr, Cu, Mn, Ni, Pb, Zn) vary from the region's pedogeochemical background level up to the alert threshold for heavy metals pollution set down in the Order of the Ministry of Waters, Forests, and Environment Protection no. 756/1997. The analysis of soils from and surrounding the existing ore processing facilities shows that the heavy metals contents in few cases is above the intervention threshold, for copper, lead and zinc. The soils generally have low heavy metals contents and the values are at the region's pedogeochemical background level. The barren rocks, generally, have low heavy metals contents, close to the clark values. Taking all this into account, as well as the technology that the Canadian company intends to apply, there is a low probability that a significant heavy metals pollution of the soils left un-stripped would occur due to the proposed project.

Keywords: mining activities, auriferous ore, heavy metals, abundance index, pedogeochemical background

1. INTRODUCTION

Due to significant role played by heavy metals in the environment biogeochemical cycles, they have been amply studied in recent years in our country, in areas heavily polluted (Damian et al., 2008 a & b), including in urban areas (Lăcătuşu et al., 2008 a & b). Have been less studied these chemical elements in soils located in territories with underground or open pits mining. Area of Rosia Montana has offered a complex example on this line. There, the ancient underground mines, but later the open pits, including the ore processing industry could influence abundance of heavy metals in the area of impact of such activities.

Since ancient times Rosia Montana has been well known for its gold bearing ore. Underground gold extraction was practiced from Roman times, but major open pit extraction began in 1972 on one part of the ore body. This continued until 2006 when the State closed down the operation due to significant long-term financial losses and to the operation not meeting new environmental legal standards. In 1997 a Canadian company in joint venture with the state-owned mining company (80-20%) established an operating company, the Rosia Montana Gold Corporation, in order to re-evaluate the Rosia Montana ore deposits under an exploration licence. By 2002 they had delineated a world class gold and silver resource and had developed a plan for exploitation based on extracting ore from four large open pits – two centred on existing pits requiring expansion and from two new but smaller ones. The Company intends to industrially process the extracted ore in a plant situated near the pits and to deposit the waste material close by. The waste rock would be deposited in the Rosia Montana and Corna Valleys and the process tailings behind a rock dam in Corna Valley. These valleys are adjacent to each other and share the ore deposit in their upper parts.

In order to assess the impact that the possible exploiting works will produce on soils in the area, research regarding soil nature and their productivity level was carried out as part of the proposed development's Environmental Impact Assessment in 2003. The present soil pollution level was assessed in the areas where ore extraction and processing works were carried out in the past, and also in the area intended to be used for ore extraction and processing in the project footprint proposed by the Rosia Montana Gold Corporation. This paper presents these last aspects below.

The majority of countries with mining activities, especially open pit mining, that have environment protection requirements that apply to the affected zones, have published material with themes in line with this paper (Charles et al., 2005; Getaneh & Alemayehu, 2006; Ogoloa et al., 2002; Razo et al., 2004; Shumlyanski et al., 2005; Suciu et al., 2008). Similarly, many US EPA projects were developed (US Environmental Protection Agency, 1985; 1995) on these themes. This paper presents some new elements in the analytical interpretation of data to establish the nature and the intensity of heavy metals pollution.

2. MATERIAL AND METHODS

The total investigated area is about 1,646.32 ha, with the area occupied by soils being 1,521.38 ha and remaining 124.94 ha representing rock exposure and lakes. Districambosols and Eutricambosols, which represent 73.83% of the total surface, are the predominant soil types. In a decreasing order, these are followed by: Lithosols, Regosols, Andosols, and Colluvic Aluviosols (Tab. 1). Figure 1 presents the space distribution of these soils.

The land is mostly used for agriculture and forestry, with the remainder being unproductive (rock, waste dumps, detritus), or covered by habitation, roads, rivers or man-made lakes. The soil favourability for different crops oscillates between 44 and 55 points (on a 100 points scale) for pastures and grasslands and between 12 and 17 points for apple, plum, and potatoes.

In the investigated zone, which mainly comprises the area where the existing and proposed future gold-silver ore extraction and processing is to take place or lie adjacent to, 40 soil profiles were analysed, at variable distances between 100 m and 300 m depending on the relief conditions. The first step in the soil type's identification was morphologic characterization of the soil profiles. 153 soil samples were collected (according to the number of significant genetic horizons of each soil profile), for the chemical and physical analysis with the purpose to identify and characterize the soil types (National Research and Development Institute for Soil Science, Agrochemistry, and Environment Protection, 1987).

Table 1. Soil types distribution in the Rosia Montana area

	Soil type*	Area	
		ha	%
1	Cambisols		
	- Eutric Cambisols	429	26.09
	- Dystric Cambisols	786	47.74
2	Andosols	4	0.27
3	Leptosols	182	11.08
4	Regosols	100	6.12
5	Fluvisols	1	0.12
6	Rocks in association with leptosols	16	1.00
	Soils total	1,521	92.42
7	Pits	122	7.43
8	Lakes	2	0.15
	General total	1,646	100.00

* According to WRB (Bridges et al., 1998)

The 74 soil samples were collected to determine the heavy metals content from the upper horizon (0-10 cm). Sampling points were determined using a relatively uniform sampling network of approximately 100 m distance between points in the areas in which the past ore extraction and processing have been carried out. Soil samples were mineralised with a HNO_3 and $HClO_4$ solution and after that the precipitate was soluble with the hydrochloric solution. In this solution the total content of heavy metals (Cd, Co, Cr, Cu, Fe, Mn, Pb, Zn) was determined by means of atomic absorption spectrometry in air acetylene flames.

The contamination and pollution level was assessed according to a method developed by Lăcătuşu and Ghelase (1992, 1994). Analytical data were statistically calculated, with the grouping center parameters: arithmetic mean, geometric mean, median, module ($\bar{x}$, xg, Me, Mo) - and the spreading values: minimum value, maximum value, standard deviation (xmin, xmax,σ) being determined.

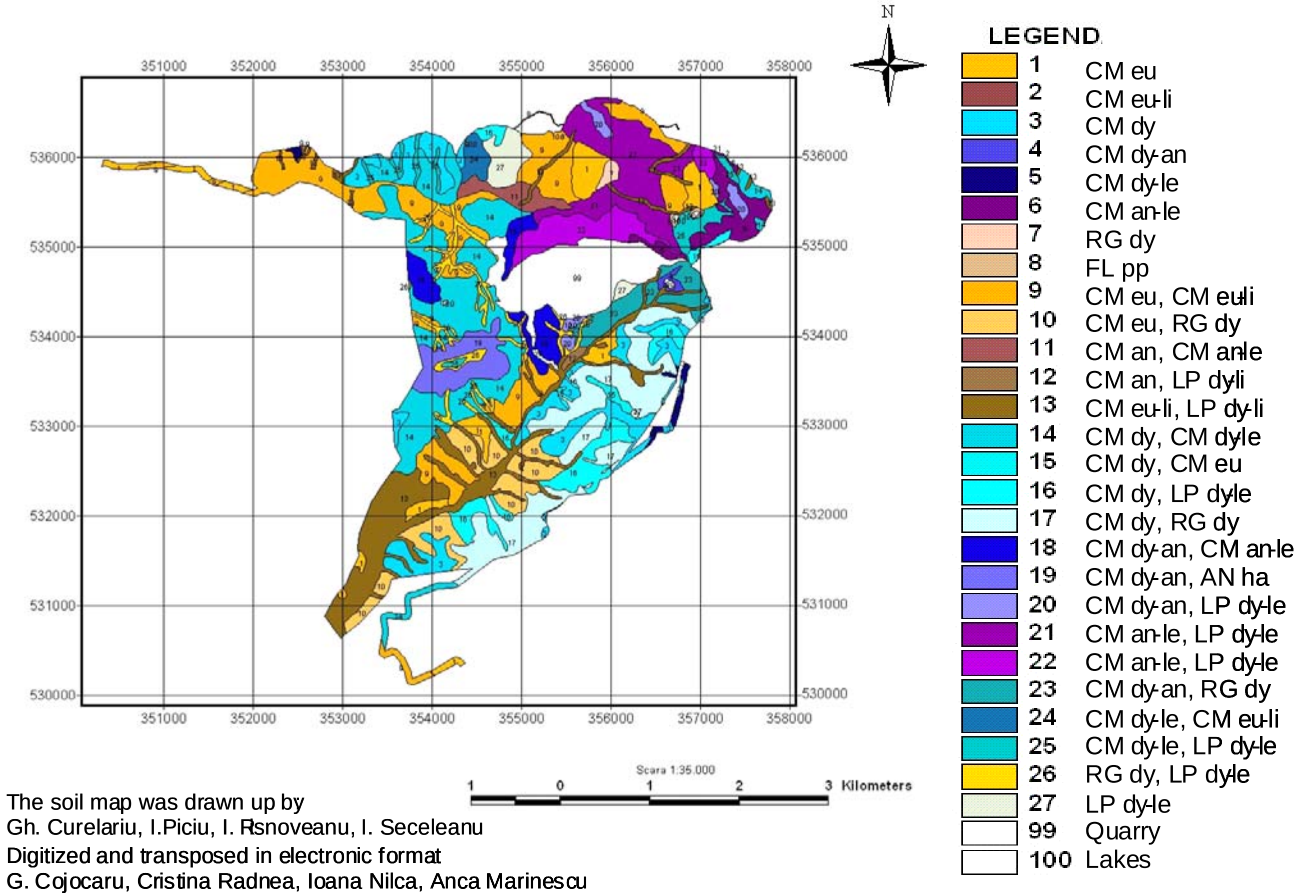

Figure 1. Soil map of the Rosia Montana area. The legend's abbreviations signification:
CM – Cambisol; RG – Regosol; FL – Fluvisol; LP – Leptosol
eu – eutric; li – lithic; dy – dystric; an – andic, le – leptic; ha - haplic

3. RESULTS AND DISCUSSIONS

3.1. The existing heavy metals contamination/pollution level of the soils from the areas already affected by historical mining

The statistical data regarding the soil's upper horizon (A), heavy metals content highlights a variation of values, from those specific to the region's pedogeochemical background, to values exceeding this background several times. However, both the individual values and the statistically calculated ones (Tab. 2) reveal that high values are few and isolated, and related to a certain old technological processes.

Table 2. Statistical parameters of the heavy metals distribution in the soils within the Rosia Montana area with mining activities as compared to the normal values (NV), alert threshold (AT), and intervention threshold (IT) values for land sensitive use (MAPPM Order no.756/1997)

Statistical parameter	Location	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn
		mg·kg^{-1}							
x_{min}	Pit and mine entry (n = 18)	0.7	10	25	40	910	39	74	170
x_{max}		2.1	14	43	142	2008	83	159	253
$\bar{x}$		1.4	12	32	84	1204	60	118	218
σ		0.6	2	10	40	711	18	37	27
x_g		1.2	10	29	75	1093	55	109	205
Me		1.1	9	27	69	954	56	97	187
x_{min}	Dumps (n = 24)	0.6	14	10	30	730	54	32	110
x_{max}		1.1	21	42	124	3512	89	80	274
$\bar{x}$		0.9	18	28	89	1419	71	52	193
σ		0.3	3	11	32	1501	14	21	61
x_g		0.7	15	20	74	1279	63	50	183
Me		0.6	12	18	69	1256	62	48	177
x_{min}	Processing works (n = 16)	4.0	12	20	210	510	54	140	625
x_{max}		12.0	56	41	1790	1159	102	327	1710
$\bar{x}$		7.0	28	32	429	830	82	221	1103
σ		3.0	11	8	832	381	20	101	504
x_g		6.0	27	27	315	712	76	202	1086
Me		5.0	21	28	307	716	77	196	1019
x_{min}	Ponds (n = 16)	0.5	13	35	32	629	62	60	84
x_{max}		1.2	29	52	59	1430	133	85	326
$\bar{x}$		0.8	19	43	41	992	104	73	204
σ		0.3	6	9	12	351	29	16	101
x_g		0.7	18	36	37	874	92	69	193
Me		0.6	17	38	38	851	91	70	198
NV		1.0	15	30	20	900	20	20	100
AT		3.0	30	100	100	1500	75	50	300
IT		5.0	50	300	200	2500	150	100	600

x_{min} - Minimum value; x_{max} - Maximum value; $\bar{x}$ - Average value; σ - Standard deviation;
x_g - Geometric mean; Median (Me)

The concentrations of all the analyzed heavy metals in samples collected both from the open pit area the mine entrance, waste dumps and waste material settling ponds were compared with the values set out in national and international guidelines. Table 2 shows the values of the heavy metals content in soils in different localities on the old mine activity areas. Here were detected the highest values for Cd, Cu, Pb and Zn with average values being found at the intervention threshold for land sensitivity use. In the samples collected from the old ore and processing facilities areas, the values for Cd were 1.4 times higher than the intervention threshold (IT). In addition, the values for Cu, Pb and Zn were 2.3, 2.2 and 1.8 times higher than the IT.

The percentage distribution of soils by loading/pollution classes (Tab. 3) reveals a localised surface polluted with heavy metals in the area around the processing works. This represents 20.9% (Cd), 19.7% (Co), 26.1% (Cu), 16.3% (Ni), 29.0 (Pb), and 25.4 (Zn). The analyzed samples, most of them from the ore processing works area, contain heavy metals at polluting level, from slight to strong. Table 4 shows that the analytical values for normal content at the alert threshold and intervention threshold present the same pattern. Thus, the interval up to the alert threshold contains most of the samples, between 18% (Ni) and 72% (Pb). The range between the alert and intervention threshold contains between 10% (Cu) and 82% (Ni), and the range over the intervention threshold contains between 3% (Cd) and 16% (Cu) of the analyzed samples.

Table 3. Percentage distribution by loading/pollution classes of the heavy metals contents from the soils (A horizon) of the Rosia Montana area, where auriferous ore extraction and processing activities were conducted

		Cd	Co	Cr	Cu	Ni	Pb	Zn
		%						
Loading	slight	14.0	10.0	44.2	17.0	24.0	8.8	8.7
	moderate	19.6	26.8	20.7	20.6	20.8	22.1	30.0
	strong	20.8	34.5	19.9	27.3	21.2	20.6	22.5
	very strong	24.7	9.0	15.2	9.0	17.7	19.5	13.4
Pollution	slight	10.2	12.4		7.4	16.3	16.3	16.1
	moderate	7.1	7.3		12.3		12.7	9.3
	strong	3.6			6.4			

The general pedogeochemical abundance index (GPAI) represents the ratio between an average chemical element's value in the analyzed samples and the general chemical element average content in a soil (Lăcătuşu & Ghelase, 1994). This index in the investigated soils worldwide is within the values over 1 (Tab. 5). These ratios show how many times the chemical element concentration in the analyzed samples is higher than the average content in the world's soil. By chemical elements, high values are noticed especially for Cd, Cu, Pb, and Zn. The highest values have been registered in the soils from the processing works.

The local pedogeochemical abundance index (LPAI), represents the ratio between a chemical element's average content in the soils from local area which has significantly higher heavy metals natural concentrations and our researched area (Lăcătuşu & Ghelase, 1994). From this index it can be noted that most of the values are below 1. The lower the index value is, the higher is the difference between the two considered parameters. Very low values of the LPAI are highlighted for Zn, Cd, Cu, and Pb in the soils from the ore processing works area (Tab. 5). The other values below 1 ranged in the 0.22-0.96 interval. The values over 1 show that in the soils from the open pit and mine entry area, element abundance such as Co, Cr, Pb, in the or Cd in the soils around the settling pond is lower than in the control area – the Southern half of the Apuseni Mountains. The average values of the heavy metals from the soils of this area ($mg\cdot kg^{-1}$) are 0.93 (Cd), 18 (Co), 32 (Cr), 41 (Cu), 800 (Mn), 34 (Ni), 48 (Pb), and 89 (Zn).

Table 4. Percentage distribution (%) of soil samples (n = 74) from the Rosia Montana area by heavy metals abundance classes, depending on the alert (AT) and intervention threshold values (IT), according to the Order no.756/1997 of the Ministry of waters, forests and environment protection

Chemical element	Value classes			
	normal	up to the alert threshold (AT) limit	between the alert (AT) and intervention threshold (IT) limits	over the intervention threshold (IT) limit
Cd	20	67	10	3
Co	17	43	35	5
Cr	49	51		
Cu	3	58	23	16
Mn	62	21	14	3
Ni		18	82	
Pb		72	28	
Zn	7	70	12	11

Table 5. Heavy metals general (numerator) and local (denominator) pedogeochemical abundance indices values from the soils of the Rosia Montana area where auriferous ore extraction and processing mining activities have been carried on

Localization	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn
Quarries and mine entries	$\frac{4.7}{0.66}$	$\frac{2.4}{1.50}$	$\frac{1,1}{1,00}$	$\frac{4,2}{0,49}$	$\frac{2,4}{0,66}$	$\frac{3.0}{0.57}$	$\frac{7.9}{2.45}$	$\frac{4.4}{0.41}$
Waste dumps	$\frac{3.0}{0.64}$	$\frac{3.5}{1.00}$	$\frac{0.90}{1.14}$	$\frac{4.4}{2.20}$	$\frac{2.8}{0.56}$	$\frac{3.6}{0.48}$	$\frac{3.5}{1.08}$	$\frac{3.9}{0.46}$
Processing works	$\frac{23.2}{0.13}$	$\frac{5.6}{0.64}$	$\frac{1.1}{1.00}$	$\frac{21.4}{0.10}$	$\frac{1.7}{0.96}$	$\frac{4.1}{0.58}$	$\frac{14.7}{0.22}$	$\frac{22.1}{0.08}$
Setting pond	$\frac{2.7}{1.16}$	$\frac{3.8}{0.94}$	$\frac{1.4}{0.74}$	$\frac{2.1}{1.00}$	$\frac{2.0}{0.81}$	$\frac{5.2}{0.33}$	$\frac{4.9}{0.65}$	$\frac{4.1}{0.43}$

Thus, the soils from the area where Rosia Montana ore has been extracted and processed have heavy metals content higher than the soil's general pedogeochemical background, with different values, from 1.1 up to 7.9 times and reaching over 23.3

times higher in the ore processing area. With few exceptions, in the same ore processing area, the heavy metals content of the soil in the area where ore extraction and processing activities are carried out are close to the pedogeochemical background of the Southern half of the Apuseni Mountains. It can be concluded that the mining activity carried on has not polluted the soil with heavy metals.

Only the soil from the ore processing facility and around this has heavy metals contents that exceed the intervention threshold. Excepting these small areas, it could be said that the Rosia Montana area ore extraction and processing is not a significant heavy metals soil pollution source.

3.2. The present heavy metals loading/pollution level of the soils from the area where ore extraction and processing activities are to be carried out

Analytical data regarding the heavy metal content in the soil (Tab. 6) reveals large value intervals, but the average contents are close to those usually determined in soils. They are within the normal content values, which define the alert threshold. Table 7 shows that there are only a few values of Cd, Mn, Ni and Pb situated between the alert threshold limit (AT) and the intervention threshold limit. Only 2% of all the Cd values are over the intervention threshold.

Table 6. Statistical parameters of the heavy metals contents ($mg \cdot kg^{-1}$) from the RMGC area where auriferous ore extraction and processing activities are to be carried on, as compared to the normal values (NV) and to the values of the alert (AT) and intervention (IT) thresholds for a soil sensitive use (Order no.756/1997of the Ministry of waters, forests and environment protection)

Statistical parameter	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn
n	153	153	153	153	153	153	153	153
x_{min}	0,5	11	11	8	80	13	12	26
x_{max}	10,1	67	79	39	2187	114	90	272
$\overline{x}$	1,2	30	30	18	645	49	36	88
σ	1,1	12	14	5	340	25	14	35
xg	1,1	28	27	17	331	43	33	82
Me	1,0	27	26	17	573	44	35	83
Mo	1,1	26	22	16	519	39	33	82
NV	*1,0*	*15*	*30*	*20*	*900*	*20*	*20*	*100*
AT	*3,0*	*30*	*100*	*100*	*1500*	*75*	*50*	*300*
IT	*5,0*	*50*	*300*	*200*	*2500*	*150*	*100*	*600*

The conclusions to be drawn from the above observations are better understood if data regarding the percentage repartition by soil heavy metals loading/pollution classes (Tab. 8) are examined. The notable fact is that most of the values belong to the loading domain and only three chemical elements (Cd, Co, and Ni) have 50% of the values included in the slightly polluted soils class.

Table 7. Percentage distribution (%) of the soil samples (n = 153) from the Rosia Montana area, by heavy metals abundance classes, depending on the alert (AT) and intervention threshold (IT) values, according to the Order no. 756/1997 of the Ministry of waters, forests and environment protection

Chemical element	Value classes			
	normal	up to the alert threshold (AT) limit	between the alert (AT) and intervention threshold (IT) limits	over the intervention threshold (IT) limit
Cd		97	1	2
Co		34	53	13
Cr	50	50		
Cu	64	36		
Mn	80	17	3	
Ni		83	17	
Pb		84	16	
Zn	52	48		

Table 8. Percentage distribution (%) by loading/pollution classes of the heavy metals (A horizon) from the Rosia Montana area

	Cd	Co	Cr	Cu	Ni	Pb	Zn
Loading slight			38,5			5,1	
moderate		5,1	56,4	46,1	7,7	51,3	25,6
strong	2,6	10,3	5,1	43,8	15,4	33,3	51,3
very strong	7,7				15,4	7,7	1,8
Pollution slight	76,9	66,7		2,6	53,8	2,6	10,3
moderate	12,8	17,9			7,7		

Besides, the maximum reached pollution level is the average one, with almost 18% of the values in the Co case. Cobalt is an exception to the above-described situation. Most of its values are between the limit of the alert threshold and that of the intervention threshold, and even above this. However, taking into account that this element also has a plant nutritional role (cobalt for instance is essential for nitrogen fixation in legumes and in root nodules of non-legumes) we consider that it cannot influence negatively the soil heavy metals balance.

Therefore, we can conclude that the natural soils located in the Rosia Montana area, where it is intended to exploit and process auriferous ore, are loaded with heavy metals without reaching, with some exceptions, low pollution levels.

3.3. Future soils heavy metals pollution

In the event of implementing the Rosia Montana project, the only soils heavy metals pollution source could be the mineral ore and country or barren rock, namely the gold-silver mineralization bearing rocks and their host rock. However, from the chemical analyses performed on a large number of samples and statistically computed (tables 9 and 10) it appears that both rock categories have normal heavy metals contents, around the clark. Part of the chemical elements in the mineralized and barren rocks have average contents below the clark value, in other words below the lithosphere average content value. Even if the other chemical elements have an average content higher than clark value, the overflow is small, the enriching factor being as a rule between 1 and 2 for the heavy metals in the barren rocks. In addition, the abundance of these elements is not very high either in the mineralization bearing rocks.

Table 9. Average heavy metals contents ($mg \cdot kg^{-1}$) from the Rosia Montana mineralization bearing rocks, as compared to the clark* values (according to analytical data supplied by the Roşia Montană Gold Corporation)

Localization	No. of samples (n)	Cd	Co	n	Hg	n	Pb	Zn
Carnic	394	0,51	7,05	229	0,17	394	41	129
Cetate	402	0,51	7,15	173	0,08	402	39	136
Jig	194	0,33	16,10	194	0,07	194	56	147
Orlea	118	0,40	11,30	118	0,03	118	42	132
TOTAL / average	***1108***	***0,47***	***9,13***	***714***	***0,10***	***1108***	***43***	***135***
Clark* from Fiedler and Rösler (1988)		***0,13***	***18***		***0,05***		***16***	***83***

Table 10. Statistical parameters of the total heavy metals contents ($mg \cdot kg^{-1}$) from the dead rocks of the Rosia Montana area (n = 64), as compared tot the clark values (computed for the analytical data taken from the RMGC Geochemical characterization report, 2005)

Chemical element	x_{min}	x_{max}	$\bar{x}$	σ	cv(%)	clark*	Enriching factor
Cd	0,03	1,20	0,27	0,38	142	0,13	2,08
Co	1,8	31	14	10	71	18	0,78
Cr	1	79	33	16	49	83	0,40
Cu	12	129	59	42	74	47	1,26
Mn	147	7380	1504	1430	95	100	1,50
Ni	2	95	25	26	104	58	0,43
Pb	8	133	29	20	67	16	1,81
Zn	6	424	116	89	77	83	1,40

* from Fiedler and Rösler (1988)

Mercury has an enriching factor of 2, and lead 2.7. Cadmium has different characteristics. The enriching factor value is 3.2 in the mineralization bearing rocks, and 2.1 in the barren rocks. The phenomenon is due to the higher mobility of this element.

Therefore, the heavy metals concentration in the raw material, which would be processed for gold and silver extraction, is low, and it does not constitute a soil pollution source. In any case, except for the open pit extraction and ore and rock transport, the other technological processes will take place in a closed circuit.

4. CONCLUSIONS

The soil cover from the Rosia Montana area consists of the following soil types and subtypes: a) Eutricambosols with the typical, lithic, and andic subtypes; b) Districambosols with the same subtypes; Regosols with the eutric and distric subtypes; d) Eutric and distric lithosols, and e) Colluvic Aluviosols. Districambosols and Eutricambosols are predominant, covering 73.83% of the completely researched area (1,646 ha).

The soils from the areas where gold-silver ore extraction and processing mining activities were carried out (the Cetate quarry, the surfaces in front of the mine entries, the barren rock dumps and the waste dumps from flotation) mostly contain heavy metals at the region's pedogeochemical background, up to the alert thresholds. The soils from the soil preparation works precincts and around it are the exception; the Cu, Pb, and Zn contents exceed in some samples the values of the intervention threshold for both sensitive and less sensitive use.

The soils from the area on which the proposed activities of open pit mining and ore processing, waste rock, stripped soil, and ore process waste depositing are to take place, are slightly polluted with heavy metals by geogenic origin. Heavy metals loading level of most of them is equivalent to the region's pedogeochemical background.

Heavy metals abundance in the mineralization bearing rocks and in the barren rocks is close to the clark values.

Taking into account the heavy metals abundance in the barren rocks and mineralization bearing rocks of the area, as well as the technologies which will be implemented in the construction and operation phases, there is a low probability of polluting the un-stripped soils to high levels that reach alert and/or intervention thresholds values.

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