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ASSESSMENT OF HEAVY METAL CONTAMINATION OF SOILS AROUND AGARAK (RA) COPPER-MOLYBDENUM MINE COMPLEX

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The aim of the present study was assessing the heavy metal pollution of soils around Agarak copper-molybdenum mine complex and related environmental risks. The study was implemented in 2013. The level of soil contamination by heavy metals was assessed by Contamination factors, Degree of contamination, Pollution load index and Geoaccumulation index. The study revealed that almost in all studied sites Cu, Mo, Pb and Cd were the main polluting heavy metals and this was conditioned by Agarak copper-molybdenum mine complex activity.

***Keywords*:** Agarak copper-molybdenum mine complex, environmental risks, heavy metals, soil, Pollution load index, Geoaccumulation index.

Introduction. Heavy metals enter and continuously circulate in the environment as a result of human activities such as mining, smelting, electroplating, energy and fuel production, power transmission, intensive agriculture, sludge dumping, and melting operations [1–5]. Among the mentioned activities the mining industry is considered as one of the most dangerous anthropogenic activities in the world. The harmful impact of mining activities in the environment has been repeatedly emphasized by many researchers [6–11]. Mining operation, grinding, concentrating ores and disposal of tailings are the overt sources of contamination in the environment [12]. The growth of mining industry results in severe pollution of soils by heavy metals, which threatens ecosystems, surface and ground waters, food safety and human health [13–17]. The development of mining industry is gaining paces now in Armenia, therefore, the pollution of soils by heavy metals is the alarming problem not only in the world, but also in our country [18, 19]. The extent and degree of heavy metal contamination around the mines vary depending on the capacity of mining activities and geochemical characteristics of the area.

Hazardous elements in the tails of mining and metallurgical operations are often dispersed, included in particulate material or in aqueous solution by wind and/or water after their disposal [20]. Due to transport process, pollution could emerge as a result of primary contamination, formed by residues placed close to the contamination sources, secondary contamination, produced as a result of trace element dispersion out of its production areas, through water and wind, whereas

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tertiary contamination involves trace element mobilization [21]. The contamination of soils by heavy metals is causing a strong concern due to the potential effects on human health and the possible long-term sustainability of food production in contaminated areas, since it is well known that contaminants circulating in the environment can pass through food chains [22, 23].

Therefore, the investigation of accumulation and migration of heavy metals in soils is currently a very important and relevant issue. The main objective of this study is the assessment of soil pollution level of territories adjacent to open mine, processing plant and tailing dumps of Agarak copper-molybdenum mine complex by heavy metals. The total concentration of heavy metals in the soil is a useful parameter indicating contamination intensity. In addition the degree of pollution in soil was also assessed using Contamination factors (*Cf*), Degree of contamination (*CD*), Pollution load index (*PLI*) and Geoaccumulation index (*I-geo*). By this study it was possible to obtain baseline data regarding the accumulation of toxic metals in soils and these findings will help in designing the pollution abatement strategy to control the spread of pollutants in the environment surrounded by mining activity.

Materials and Methods.

1. *Study Area*. Agarak copper-molybdenum mine complex is situated in the South-East of Armenia (Syunik marz). The soils of 5 riskiest sites of this region were studied:

- surroundings of the open mine (samples №№ 1–5);
- the sites adjacent to processing plant of Agarak copper-molybdenum mine complex (samples №№ 6–7);
- surroundings of Darazam active tailing dump (sample № 8);
- recultivated tailing dam of "ravine-2" (sample № 9);
- recultivated tailing dam of "ravine-3" (sample № 10).

The main soil type in study sites is the mountain cambisol with its 2 subtypes: the typical mountain cambisol (samples №№ 1–5) and the carbonate mountain cambisol (samples №№ 6–10 and the control sample).

In Armenia this soil type is distributed 500–1700 *m* a.s.l. and on arid southern slopes it reaches up to 2400 *m* [24].

It was revealed that the typical mountain cambisol occupied intermediate position between the carbonate and decalcified subtypes by its geographical location, morphological and physicochemical characteristics. This type of soil, compared to carbonate type, occupied higher position a.s.l. (1100–1300 *m*), the gradient was 0–35 degrees, the microrelief was smooth. The soil of this subtype was not fertile and useful for agricultural purposes, except the samples №№ 4 and 5. The carbonate mountain cambisol subtype of soil was distributed 700–1000 *m* a.s.l., on the gradients of 0–30 degrees, the microrelief was mainly smooth. This soil was mainly very rocky, carbonates were distributed from top to bottom. This subtype of soil was not fertile and useful for agricultural purposes, except № 7 and control samples.

2. *Sample Collection*. For studying purpose 10 sampling sites were selected in 2013. The control section was done in the site, which was 2 *km* away from the processing plant in the direction of the Agarak Town (at a distance of 550 *km* from motor road). The coordinates of sampling sites were recorded by GPS.

The sampling of soils was carried out in a traditional way, well-known in soil science. All labware and sampling apparatus were pre-soaked in 5% nitric acid

solution followed by distilled water for a day prior to sampling for removing trace concentrations of metals.

The samples of soils were taken from a depth of 0–20 *cm* at 5 *m* intervals on a grid measuring 20×20 *m* and with the center point of the grid at the sample location. The sections were done manually. All samples were collected into polyethylene sampling bottles and transported to the laboratory. After homogenization and removal of unwanted content (stones, plant material, etc.), the samples were air-dried at room temperature, sieved to pass a 1 *mm* mesh and stored in an all-glass jar for analysis of their properties.

3. *Pretreatment and Heavy Metal Analysis of Soil Samples*. Before analysis the samples need required digestion. Soil was grounded in a mortar and pestle to pass a 0.42 *mm* nylon mesh. Total concentration of heavy metals was determined using Aqua Regia (HCl–HNO_3, 3:1) extraction method (3 *g* of soil sample were digested for 2 *h* at 180°*C*). Heavy metals were determined by atomic absorption spectrometry method (AAS) using Atomic-Absorption Spectrometer PG990 (PG Instruments LTD).

4. *Assessment of Metal Contamination*. The level of soil contamination by heavy metals was assessed by contamination indices. *Cf*, *CD*, *PLI* and *I-geo* were used.

Cf and *CD* were calculated as suggested by Håkanson [25] through following formulas:

$$Cf^i = Cs^i / Cb^i, \tag{1}$$

$$CD = \sum Cf^i, \tag{2}$$

where Cs^i is the measured concentration of the examined metal *i* in the soil sample and Cb^i is the background value of heavy metal *i* in the uncontaminated soil (control). Hakanson suggested four classes of Cf to evaluate the metal contamination levels as shown in Tab. 1 [25]. Four categories of *CD* as suggested were used to evaluate metal contamination levels (Tab. 1). If the *CD* value exceeds 20, then it is necessary to take immediate counter measures to reduce heavy metal contamination in the soil.

Furthermore, each site was evaluated for the extent of metal pollution by employing the method based on the *PLI* developed by Thomilson [26], as follows:

$$PLI = \sqrt[n]{(Cf^1 \cdot Cf^2 \cdot Cf^3 \cdot \ldots \cdot Cf^n)}, \tag{3}$$

where *n* is the number of metals studied and *Cf* is the contamination factor calculated as described in (1). The *PLI* provides simple, but comparative means for assessing a site quality. The rank of values of *PLI* and its implication is shown in Tab. 1 [26].

I-geo was used to calculate metal contamination level in the soils. The *I-geo* was originally defined by Müller in 1969 [27], in order to determine and define metal contamination in sediments, by comparing current concentrations with pre-industrial levels. The index is calculated as [27]:

$$I\text{-}geo = \log_2\left(\frac{Cs^i}{1.5\ Cb^i}\right), \tag{4}$$

where Cs^i is the concentration of element *i* in the samples, Cb^i is the background value of the element *i*, and the factor 1.5 is used to take into account the possible lithological variability. The rank of *I-geo* values and implication are shown in Tab. 1.

Table 1

Different types of model and the categories for describing soil contamination

Model	Class	Description	Sources
Geoaccumulation index	$I\text{-}geo \leq 0$	uncontaminated	[27]
	$0 < I\text{-}geo \leq 1$	uncontaminated to moderately contaminated	
	$1 < I\text{-}geo \leq 2$	moderately contaminated	
	$2 < I\text{-}geo \leq 3$	moderately to strongly contaminated	
	$3 < I\text{-}geo \leq 4$	strongly contaminated	
	$4 < I\text{-}geo \leq 5$	strongly to very strongly contaminated	
	$5 < I\text{-}geo$	very strongly contaminated	
Contamination factor	$Cf \leq 1$	low	[25]
	$1 < Cf \leq 3$	moderate	
	$3 < Cf \leq 6$	considerable	
	$6 < Cf$	very high	
Degree of contamination	$CD \leq 11$	low	[25]
	$11 < CD \leq 22$	moderate	
	$22 < CD \leq 33$	considerable	
	$33 < CD$	very high	
Pollution level index	$PLI < 1$	perfection	[25]
	$PLI = 1$	base line level of pollution	
	$PLI > 1$	deterioration of site quality	

5. *Statistical Analysis*. Analysis of variance was used to compare the mean metal concentrations among the sites. Further evaluation was done via Duncan's multiple range tests. Statistical analysis was performed using SPSS software, version 15.

Table 2

The mean concentrations (mg/kg) of some heavy metals in the studied samples of soil

№	V	Cr	Mn	Co	Ni	Cu	Zn	As	Mo	Pb	Cd
1	12.5±3.4	1.4±0.4	54.3±14.9	2.1±0.4	1.3±0.4	53.3±18.0	4.9±1.8	0.6±0.2	5.2±1.7	1.1±0.3	0.03±0.01
2	16.3±4.6	0.6±0.2	61.5±21.1	1.6±0.3	2.3±0.7	145.5±35.8	10.4±3.7	1.5±0.4	13.2±3.8	2.3±0.7	0.06±0.02
3	11.9±2.9	0.7±0.2	32.3±9.1	1.3±0.3	0.7±0.2	123.5±36.1	7.1±2.1	1.0±0.3	8.5±3.0	3.9±0.9	0.04±0.001
4	14.1±5.1	3.8±1.1	99.9±24.5	2.1±0.5	4.1±1.1	23.4±6.0	11.7±2.0	0.8±0.3	1.2±0.4	1.5±0.4	0.05±0.002
5	7.5±1.8	2.7±0.9	70.0±19.2	1.3±0.2	2.2±0.6	25.3±5.4	7.7±2.2	0.4±0.1	1.5±0.4	1.7±0.4	0.03±0.01
6	11.2±3.1	2.0±0.7	184.5±34.9	1.6±0.4	1.7±0.4	56.2±16.9	19.2±5.4	1.6±0.5	4.2±1.5	6.4±1.9	0.22±0.07
7	8.9±2.2	1.3±0.4	113.0±35.1	1.4±0.4	1.0±0.3	81.2±24.4	10.8±3.8	1.0±0.4	6.2±2.0	3.5±1.4	0.04±0.01
8	9.5±3.0	6.8±2.1	50.9±11.2	1.6±0.5	4.1±1.3	35.4±13.1	6.6±1.9	0.8±0.2	1.2±0.3	1.2±0.4	0.04±0.01
9	9.2±2.8	6.0±1.6	49.0±12.4	1.4±0.3	3.7±0.9	39.7±11.7	6.5±2.3	0.9±0.2	0.9±0.2	1.4±0.4	0.05±0.02
10	8.2±3.1	2.8±0.5	39.2±9.5	1.2±0.3	1.7±0.5	17.4± 4.9	4.4±1.4	0.7±0.3	0.6±0.2	0.6±0.2	0.01±0.004
Ctrl	7.5±1.4	2.6±0.7	56.4±13.1	1.3±0.4	2.0±0.7	9.5±3.6	8.0±1.7	0.6±0.2	0.5±0.2	0.8±0.3	0.02±0.007

Results and Discussion. The concentrations of V, Cr, Mn, Co, Ni, Cu, Zn, As, Mo, Pb and Cd in the soils adjacent to open mine near Agarak copper-molybdenum

mine complex were determined, and the degree of heavy metal pollution in the soils was assessed. The ranges of mean concentration (*mg*/*kg*) of heavy metals in 5 studied areas are the following: V (8.2–16.3); Cr (0.6–6.8); Mn (39.2–184.5); Co (1.2–2.1); Ni (0.7–4.1); Cu (17.4–145.5); Zn (4.4–19.2); As (0.4–1.6); Mo (0.6–13.2); Pb (0.6–6.4) and Cd (0.01–0.22) (Tab. 2). Since the contents of metals in soils are specific and depend on the compound of rocks producing soil and the conditions of soil formation, for determination of pollution level, the obtained results were compared with the control sample which was considered as a background.

1. *Contamination Evaluation Based on Geoaccumulation Index. I-geo* was used to calculate metal contamination level in the soils (Tab. 3).

Table 3

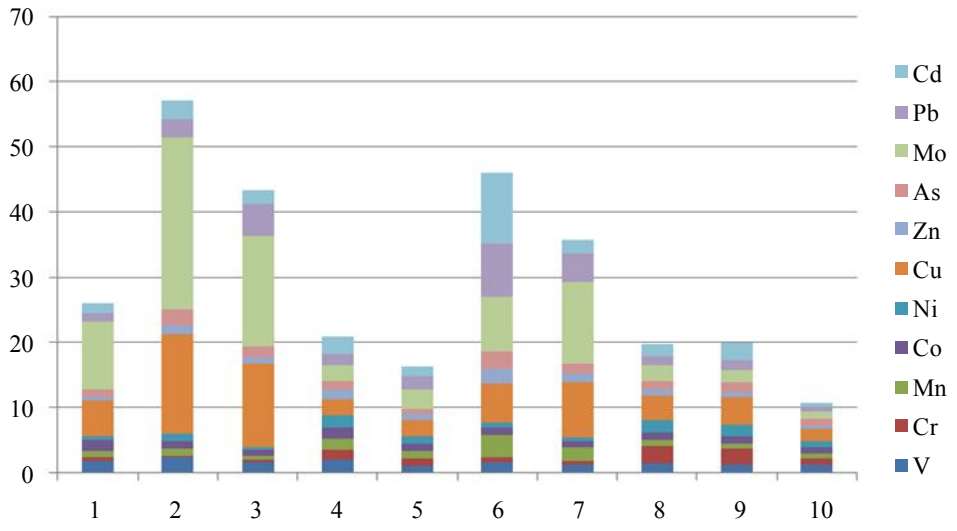
The degree of heavy metal pollution of soil samples according to the Geoaccumulation index

№	V	Cr	Mn	Co	Ni	Cu	Zn	As	Mo	Pb	Cd
1	0.15	–1.48	–0.64	0.11	–1.21	1.90	–1.29	–0.58	2.79	–0.13	0.00
2	0.53	–2.70	–0.46	–0.29	–0.38	3.35	–0.21	0.74	4.14	0.94	1.00
3	0.08	–2.48	–1.39	–0.58	–2.10	3.12	–0.76	0.15	3.50	1.70	0.42
4	0.33	–0.04	0.24	0.11	0.45	0.72	–0.04	–0.17	0.68	0.32	0.74
5	–0.58	–0.53	–0.27	–0.58	–0.45	0.83	–0.64	–1.17	1.00	0.50	0.00
6	–0.01	–0.96	1.12	–0.29	–0.82	1.98	0.68	0.83	2.49	2.42	2.87
7	–0.34	–1.58	0.42	–0.48	–1.58	2.51	–0.15	0.15	3.05	1.54	0.42
8	–0.24	0.80	–0.73	–0.29	0.45	1.31	–0.86	–0.17	0.68	0.00	0.42
9	–0.29	0.62	–0.79	–0.48	0.30	1.48	–0.88	0.00	0.26	0.22	0.74
10	–0.46	–0.48	–1.11	–0.70	–0.82	0.29	–1.45	–0.36	–0.32	–1.00	–1.58

The *I-geo* values for V show that 60% of the samples fall in the uncontaminated class (≤ 0) and 40% in the uncontaminated–moderately contaminated class (0–1). *I-geo* values for Cr show that 80% of the samples fall in the uncontaminated class (≤0) and 20% in the uncontaminated–moderately contaminated class (0–1), for Mn show that 70% of the samples fall in the uncontaminated class (≤0), 20% in the uncontaminated–moderately contaminated class (0–1) and 10% are moderately contaminated (1–2), for Co show that 80% of the samples fall in the uncontaminated class (≤0) and 20% in the uncontaminated–moderately contaminated class (0–1), for Ni show that 70% of the samples fall in the uncontaminated class (≤0) and 30% in the uncontaminated–moderately contaminated class (0–1), for Cu show that 30% in the uncontaminated–moderately contaminated class (0–1), 40% are moderately contaminated (1–2), 10% are moderately to strongly contaminated (2–3) and 20% are strongly contaminated (3–4), for Zn show that 90% of the samples fall in the uncontaminated class (≤ 0) and 10% in the uncontaminated–moderately contaminated class (0–1), for As show that 60% of the samples fall in the uncontaminated class (≤ 0) and 40% in the uncontaminated–moderately contaminated class (0–1), for Mo show that 10% of the samples fall in the uncontaminated class (≤ 0), 40% in the uncontaminated–moderately contaminated class (0–1), 20% are moderately to strongly contaminated (2–3), 20% are strongly contaminated (3–4) and 10% are strongly to very strongly contaminated (4–5), for Pb show that 30% of the samples fall in the uncontaminated

class (≤ 0), 40% in the uncontaminated–moderately contaminated class (0–1), 20% are moderately contaminated (1–2) and 10% are moderately to strongly contaminated (2–3), for Cd show that 30% of the samples fall in the uncontaminated class (≤0), 60% in the uncontaminated–moderately contaminated class (0–1) and 10% are moderately to strongly contaminated (2–3). The average *I-geo* for the observed metals were in decreasing order of Mo (1.83) > Cu (1.75) > Pb (0.65) > Cd (0.50) > > As (–0.06) > V (–0.08) > Co (–0.35) > Mn (–0.36) > Zn (–0.56) > Ni (–0.62) > > Cr (–0.88). No *I-geo* value was greater than 5 (i.e. very strongly contaminated), and only one value for Mo (4.14) at sample № 2 was in strongly to very strongly contaminated class.

2. *Contamination Evaluation Based on Contamination Factors.* In Figure are illustrated the calculated *Cf* for measured heavy metals in soil samples. During the studies it was found that almost in all soil samples Cu, Mo, Pb and Cd are the main polluting heavy metals. Particularly, in case of Cu the maximum value of *Cf* was 15.32, in case of Mo was 26.4, for Pb was 8.0 and for Cd was 11.0. All these values correspond to the very high level of contamination. Such pollution of soils is directly conditioned by high content of above-mentioned heavy metals in ore processed on Agarak copper-molybdenum mine complex. The average *Cf* for the observed metals were in the decreasing order of Mo (9.36) > Cu (6.82) > Pb (3.19) > Cd (3.11) > >As (1.59) > V (1.5) >Mn (1.41) > Co (1.230) >Zn (1.18) >Ni (1.17) > Cr (1.08).



Values of *Cf* of different metals in surrounding territories of Agarak copper-molybdenum mine complex.

3. *Contamination Evaluation Based on Degree of Contamination and Pollution Load Index.* The values of *CD* are presented in Tab. 4. By *CD* value reduction the 10 investigated soil samples made the following series: 2 > 6 > 3 > 7> 1 > 4 > 9 > 8 > 5 > 10. By *CD* value the 10% of soil samples fall in the low level of contamination (*CD* ≤ 11), 40% of soil samples fall in the moderate level of contamination (11< *CD* ≤ 22), 10% in the considerable level of

contamination ($22 < CD \leq 33$), 40% of soil samples fall in the very high level of contamination ($33 < CD$). Pollution severity and its variation along the sites were determined with the use of *PLI*. This index is a quick tool in order to compare the pollution status of different places [28]. The Pollution load index is presented in Tab. 4. The values of Pollution load index were found to be mostly high ($PLI > 1$) in all the studied stations (with the exception of №10 soil sample). The highest value of the Pollution load index was observed in № 6 ($PLI = 2.87$) and № 2 samples ($PLI = 2.28$), and the lowest value was observed in № 10 sample ($PLI = 0.91$). The high value of *PLI* in № 6 soil sample is connected with the operation of ore grinding mill of Agarak copper-molybdenum mine complex in the immediate vicinity of this sampling point and the high value of *PLI* in soil sample № 2 collected from the South-West side of the open mine is conditioned by the peculiarities of relief and the wind direction in the surroundings of the open mine.

Table 4

CD and PLI in the open mine and tailing dump of surrounding territories of Agarak copper-molybdenum mine complex

Sample number	*CD*	*PLI*
1	25.93	1.40
2	57.27	2.28
3	43.21	1.66
4	20.81	1.85
5	16.30	1.33
6	46.00	2.87
7	35.61	1.92
8	19.85	1.63
9	19.87	1.62
10	10.64	0.91

Conclusion. The assessment of metal contamination (V, Cr, Mn, Co, Ni, Cu, Zn, As, Mo, Pb and Cd) in soils from selected sites in surroundings of Agarak copper-molybdenum mine complex (open mine, processing plant, one active and two recultivated tailing dumps) was made in comparison with control site. The distribution pattern of trace metals in the soil profile according to *I-geo*, *Cf* and *CD* values shows that the soil is much polluted. Almost in all studied sites Cu, Mo, Pb and Cd were the main polluting heavy metals. The high values of *CD* in soil samples № 2 and 3 (57.27 and 43.21 respectively) collected from the surroundings of open mine is conditioned by peculiarities of relief, in particular by comparatively low location above sea level and by direction of winds.

The high value of *CD* in № 6 soil sample is connected with the operation of ore grinding mill in the vicinity of this point. From three tailing dumps the maximum value of Degree of contamination was observed in Darazam active tailing dump, which is the operating tailing dump therefore the contamination level is comparatively high there.

And also the *PLI* values for the 9 sites were >1, which indicates deterioration of site quality.

It is necessary to state that the pollution problem becomes pressing as some parts of these highly polluted region are inhabited by population, and agriculture is highly developed there, therefore heavy metals can enter human body through soil-plant-human or soil-plant-animal-human chains and cause various diseases.

Since the induced pollution can pose serious threats to public health, further investigations on soil and vegetation pollution are recommended. Finally, calculating the contamination factor based on distance from the pollution source and wind direction can provide more reasonable results.

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