

ISLAM ZUZAKU (ORCID: 0000-0003-2224-6872)¹

JOSTINA DHIMITRI (ORCID: 0000-0002-1367-8457)²

MAKFIRE SADIKU (ORCID: 0000-0002-1367-8457)¹

JETON HALILI (ORCID: 0000-0002-2015-1610)¹

BETIM ISMAJLI (ORCID: 0000-0001-8942-8940)³

SKENDER DEMAKU (ORCID: 0000-0001-5582-5323)¹

THE ECOLOGICAL RISK OF CONTAMINATION WITH TOXIC METALS IN SOILS AROUND THE TREPÇA COMPLEX, THE KOSOVO THERMAL POWER PLANTS, AND A NEW CO FERRONICKELI COMPLEX

Toxic waste, soil, and ash samples were collected in the landfill (solid environmental hot spots) near the Trepça complex, New Co Ferronickeli, and Kosovo thermal power plants. They were analyzed by the ICP-OES method to measure the concentration of some toxic metals. The pollutant with the highest mean concentration (in an acidic medium) was Fe (36 400.0), followed by Mn (8683.0), Cr (6575.0), As (4739.0), Pb (3364.0), Zn (2394.0), Ni (922.6), Cu (297.6), Co (46.6), and Cd (61.8) (all concentrations in mg/kg). Three pollution indices were used such as the geoaccumulation index (I_{geo}), contamination factor (CF_i), and pollution load index (PLI). The CF_i values determined for Fe, Mn, Cr, As, Pb, Zn, Ni, Cu, Co, and Cd indicated high contamination. In all soil samples, the PLI values showed the presence of soil pollution.

1. INTRODUCTION

Because of the toxic waste left over from mining, chemical fertilizers, and pesticides, heavy metal pollution of agricultural soil is a severe ecological problem [1, 2]. Heavy metal concentrations in the soil rise because of repeated use of manure and chemical fertilizers [3]. While some heavy metals are essential for plant growth, others, when absorbed and stored by plants, can be hazardous to humans if consumed [4].

¹University of Pristina, "Hasan Prishtina", Faculty of Mathematical Natural Sciences, Department of Chemistry, Pristine, Kosovo, corresponding author S. Demaku, email address skender.demaku@hotmail.com

²University of Pristina, "Hasan Prishtina", Faculty of Education, Pristine, Kosovo.

³University of Tirana, Faculty of History and Philology, Department of Geography, Tirana, Albania.

In the region of Mitrovica, Obiliq, and Glllogoc in Kosovo, soil samples from three different land uses and traffic conditions were discovered to include heavy metals such as Pb, Zn, Fe, Ni, Cr, Mn, and As [5, 6]. The waste in Kelmend, Cikatovo, and Obilic landfills contain Pb, Zn, Fe, Mn, and As [7, 8]. Fe, Zn, Ni, Pb, Mn, Cr, and As concentrations in agricultural land in rural Mitrovica, Obilic, and Glllogoc areas were found to be higher than 1.0 mg/kg [5, 6]. There is still a serious problem with toxic metal poisoning of the biosphere, which started with the industrial revolution and urbanization [2–5].

With a cumulative effect and risk to the ecosystem, environmental pollution by toxic metals has become a substantial global problem in recent decades [6, 7]. Even while there exist harmful metals in the Earth's crust naturally, anthropogenic activities have increased their amounts to toxic levels in many ecosystems [6, 7]. Industrial activity is the principal source of hazardous metals in the soil surrounding factories [6–9]. Significant sources of soil pollution include mining and thermal power facilities [1–6]. Urban areas, mining areas, and locations close to thermal power plants are high-risk sites for soil contamination [3]. Toxic metals such as Cd, Zn, As, and Pb are released during the combustion of coal in thermal power plants [3–5].

Through the diffusion or leaching of dust, leftover tailings slag, and waste rock produced by mining and transportation activities, toxic metals infiltrate the soil [1–4]. A preventative method is required since remediating toxic metal-contaminated soil is expensive and complicated [5, 6].

Studies have been done to evaluate the site's soil quality [6–9], however, there is little information available on soil contamination and ecological assessments in Kosovo's industrial zones. According to research, Kosovo's urban and industrial soil has large concentrations of harmful metals, including those found in minerals, the old metallurgical sector, the Fe-Ni manufacturing plant, and Zn-Pb concentrate [8–10]. Instead of hazardous metals, industrial zones are typically investigated for other pollutants [9, 10]. Thermal power plants and mineral ore mining in the vicinity of Mitrovica, Obiliq, and Glllogoc have an impact on the levels of hazardous metals in the soil and represent ecological problems [8–10]. Sources of pollution from the atmosphere, fertilizers, pesticides, and other pollutants have been investigated to assess the level of toxic metal contamination in soils and the ecological concerns close to the Trepça, New Co Ferronickeli and Kosovo thermal power plants. This investigation aims to offer useful data for ecological risk reduction and long-term planning in industrial areas [4, 5].

2. MATERIALS AND METHODS

Study area. Three significant industrial sites in Kosovo: the Kosovo power plant, Trepça complex, and New Co Ferronickeli complex (Fig. 1) were the focus of this study. Large volumes of trash from the New Co Ferronickeli, which contain heavy metals and

other pollutants, are emitted into the atmosphere through a variety of technical procedures [4, 5]. These activities can produce dust that can spread over great areas and contain harmful materials in the air, water, and soil [8]. With its ore mining and flotation procedure, the Trepça complex, an inactive lead and zinc mine close to Mitrovica city, has a substantial negative environmental impact [8–10]. The community and ecosystem in the area are at risk from the mine's production of sterile material.

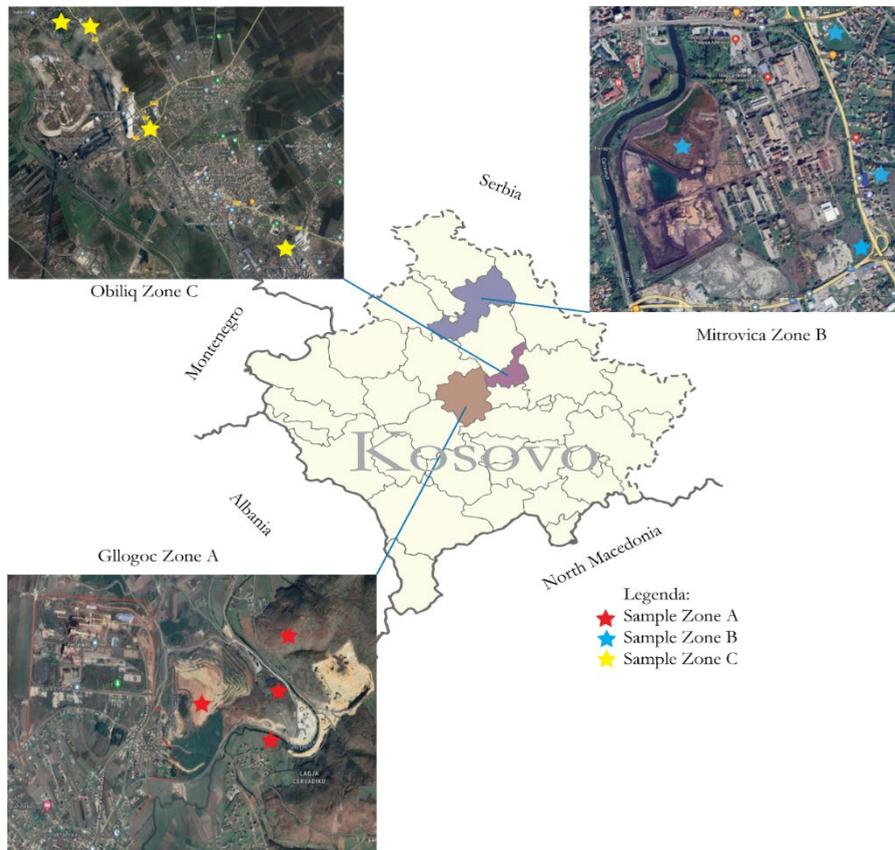


Fig. 1. The sampling site of scoria-sterile, toxic waste, ash, and soil at industrial zones of Obiliq, Mitrovica and Glogoc

The Kosovo Power Plant generates power from lignite coal in two basins in the Kosovo and Dukagjini areas in an estimated 10–12 billion tons [4–7]. About 6 million tons of coal, which has a calorific content of 1800–2000 kcal, is produced each year, and the process results in the release of ashes [11]. These ashes could potentially contaminate groundwater and surface waters with harmful metals such as Pb, Ni, Cd, Al, and Cr [12].

Environmental samples for chemical analyses were gathered and prepared using modern analytical techniques. In December 2022, field sampling was carried out in the study

regions. Samples of sterile material were collected from three sites in the New Co Ferro-nickeli (M1, M2), and soil samples were collected from a waste area close to Old Cikatovo Village (M3). Sterile samples were collected at the Kelmend (M4) landfill's toxic waste layer as well as in what is known as landfills but are known to the locals as IPM (Industrial Park of Mitrovica) (M5). Additionally, in the proximity of these zones, soil samples were collected in Shupkocv hamlet (M6). Toxic waste and soil samples were examined for the content of the following metals: Fe, Mn, Cr, As, Pb, Zn, Ni, Cu, Co and Cd.

Lignite combustion releases hazardous gases such as sulfur oxides (SO_x), nitrogen oxides (NO_x), carbon dioxide, hydrocarbons, ammonia, and hydrogen sulfide in the form of smoke and dust. Ash samples were gathered in two places. Additionally, ash dumps contain inorganic substances, the most significant of which are hazardous metals like Pb, Ni, Cd, Al, and Cr [2-5]. Two locations – TC Kosovo A (M7) and TC Kosovo B (M8) provided ash samples for analysis. 5 km from TC Kosovo, at the town of Plemetin (M9), soil samples were collected.

Sample digestion. 1 g samples of the soil, waste toxic, ash, and scoria-sterile were placed in Teflon containers. After the addition of 10 cm³ of aqua regia, the samples were heated in a microwave. Following digestion, the components were filtered (quantitative filter paper) and diluted with distilled water to create a 100 cm³ solution [13–15].

Instrumentation and statistical analyses. With the aid of inductively coupled plasma-optical emission spectrometry (Optima 2100 DV), the quantities of heavy metals were calculated (ICP-OES). For each group of analytical samples, two technique blanks and two spiked blanks were simultaneously processed. Statistics charts created using the Minitab 19 program were displayed separately for each component [16].

Pollution indices analyses. Müller's geoaccumulation index (I_{geo}) [17–19], established to measure contamination of sedimentary bottoms, was used to assess soil contamination. Soil pollution assessment can also be done by comparing the current toxic metal presence with preindustrial levels of concentration [8, 24]. I_{geo} has been widely used to comprehend the pollution levels of toxic metals in the soils [24], that is, for the calculation to assess the toxic metal pollution status of soil [17]. The I_{geo} was computed as follows:

$$I_{\text{geo}} = \log_2 \frac{C_n}{1.5B_n} \quad (1)$$

where C_n is the metal concentration in soil, mg/kg, B_n geochemical background concentration of the corresponding metal, mg/kg.

The contamination factor (CF_i) was established following the Tomlinson model [19] as the ratio of the maximum metal concentration in soil, waste, or ash to the reference value:

$$CF_i = \frac{C_n^i}{C_0^i} \quad (2)$$

where C_n^i is the metal concentration in soil (waste, ash), mg/kg, C_0^i – its pre-industrial concentration (the reference value), mg/kg.

According to international regulations, the following reference values for the analyzed metals in soil were used, mg/kg: Pb 100, Cr 100, Ni 100, Zn 300, Cu 100, Co 50, Fe 56.9, Mn 26, As 50, and Cd 5 [25, 27]. As the reference values vary from country to country, the CF_i values may be different, even if the metal concentrations are similar [8]. The CF_i is an important factor that is used to monitor metal contamination in the soil [22]. It has four categories according to the degree of contamination in the soil. $CF_i < 1$ indicates a low degree of contamination, $1 \leq CF_i < 3$ moderate pollution, $3 \leq CF_i < 6$ considerable pollution, and $CF_i \geq 6$ very high degree of contamination [24, 25].

The pollution load index (PLI) was calculated based on contamination factors calculated for each considered heavy metal. The PLI can assess the level of metal contamination and the actions that must be taken. The PLI was calculated according to:

$$PLI = (CF_1 CF_2 \times \dots \times CF_n)^{1/n} \quad (3)$$

The potential ecological risk index (ERI) put forward by Hakanson [17] was used to evaluate the soil ecological risk [17, 24]:

$$ERI = T_r CF_i \quad (4)$$

where T_r is the toxic response factor and CF_i the contamination factor. T_r values for metals are: Pb 5, Cr 2, Ni 5, Zn 1, Cu 5, Co 5, As 4, and Cd 7. The soil classification concerning ecological risk values involves five classes: $ERI < 40$ corresponds to low ecological risk, $40 < ERI < 80$ moderate ecological risk, $80 < ERI < 160$ appreciable ecological risk, $160 < ERI < 320$ high ecological risk, and $ERI > 320$ serious ecological risk [17, 24].

3. RESULTS AND DISCUSSION

3.1. HEAVY METAL CONTENTS IN THE STUDY AREA

The focus of this study was to evaluate the presence of Pb, Zn, Fe, Ni, Co, Cr, Mn, As, Cd and Cu in scoria-sterile material (M1, M2) and soil (M3) in sampling zone A (Gllgoc, New Co Ferronickeli), in the concentrate Pb-Zn toxic waste (M4, M5) and soil (M6) in sampling zone B (Mitrovica, Trepça), and in ash (M7, M8) and soil (M9) in

sampling zone C (Obiliq, TC Kosovo A and B, Power plant) (Table 1) as possible pollution sites.

Table 1

Heavy metal concentrations in zones A, B and C [mg/kg]

Sample	Pb	Zn	Fe	Ni	Co	Cr	Mn	As	Cd	Cu
Sampling zone A – Glogoc, New Co Ferronickeli										
M1	47.5	422.2	36400	922.6	46.6	6575	2541	21.1	39.9	18.3
M2	49.9	410.6	36340	1257	30.8	1792	326.2	65.3	31.4	29
M3	43.7	344.5	21280	766.4	39.5	635.5	762.9	13.8	15.1	19.1
Sampling zone B – Mitrovica, Trepça										
M4	3364	2394	4043	36.6	8.8	25.8	8683	4739	61.8	297.6
M5	90.3	428.3	913.6	7.4	0.2	16.2	59.6	13.4	2.1	21
M6	693	1847	2241	88.8	19	125.2	1657	41.7	27.3	76.1
Sampling zone C, Obiliq, TC (power plant)										
M7	65.5	235.7	28370	349.4	10.1	2839	1302	278.5	20.2	82.9
M8	50.5	238.4	25940	318.8	9.7	4262	1272	74.1	22.1	36.9
M9	96.26	460.14	23685	346.14	16.71	971	1054	58.14	8.29	51.57

It is important to note that the metal concentrations can change over time and location and also depend on the agency. The official documents for the soil quality guidelines are: *Soil quality – urban technical note No. 3* (established by EPA) [24] and *Dutch target and intervention value* [27]. These databases contain soil quality guideline values for a variety of contaminants, including heavy metals, as well as information on how the values were derived (mg/kg, As – 22, Cd – 0.4, Cr – 100, Cu – 1500, Pb – 400, Hg – 0.2, and Ni – 100 mg/kg).

The EU countries may have different soil quality guidelines for heavy metals than the EPA and the US [26]. However, in general, for essential elements such as Zn, Fe, Co, and Mn, guideline values are not usually set, as these elements are important for plant and animal growth and are beneficial (effects in small amounts). However, excess amounts of these elements can be harmful, so it is important to consider other factors such as pH, organic matter, and the presence of other contaminants when assessing soil quality. It is suggested to check the guidelines of each specific country, as they may have different rules and values.

The concentration of some heavy metals (Table 1), such as Fe (36 400 mg/kg) and Ni (922.6 mg/kg) was higher in study zone A than in sites B and C. Long-term use of minerals (scoria-sterile), machine tools, paints, pigments, and industrial equipment in study zone C may have caused the highest concentration of Fe and Ni in the soil samples in this area [9, 10].

The Trepça complex's ongoing toxic waste disposal (sampling zone B) which enriches the ore Pb-Zn, is to blame for the high concentration of heavy metals in toxic waste and soil. Our findings suggest that mining operations have a significant impact

on the presence of metal contamination in study zone B (M4). A high metal concentration was observed for Mn (8683 mg/kg), As (4739 mg/kg), Fe (2241 mg/kg), Pb (3364 mg/kg), and Zn (2394 mg/kg).

Study zone C is located very close to settlements, whereas ash waste contains natural magnetic minerals, which can raise heavy metal concentrations [4–7]. At study zone C, the contents of Fe (28 370 mg/kg) (M7), Cr (4262 mg/kg) (M8), and Mn (1302 mg/kg) (M7) were higher than those of other elements, which are suspected to be from the wasteland ash, agricultural fertilizers [9–11], as well as traffic waste. This is known because of the proximity of zone C to the power plant TC Kosovo factory.

3.2. CORRELATION ANALYSIS

A strong correlation between the Ni with Pb, and Cr with Pb (Table 2) indicates that their base arises from toxic waste and ash landfill side [19–23]. These toxic metals in the soil are a very common occurrence in the vicinity of landfill toxic waste and power plants (coal-fired), and lignite combustion and its unburned residuals are responsible for this situation [18–23]. Correlations between As with Pb and Cd with Ni (in zone C), shown for the level of significance $p < 0.05$, are negative. The results for Ni and Pb have a strong positive correlation, which means that high x -variable scores go with high y -variable scores (and vice versa). Whereas, As, Cr, and Cd have a negative moderate correlation.

Table 2

Heavy metal correlation analyses in zones A, B, and C

Sampling zone A – Gllgoc				
↓ x	Pb	Ni	Cr	As → y
Ni	0.944			
Cr	0.309	-0.022		
As	0.867	0.983	-0.206	
Cd	0.74	0.476	0.869	0.306
Sampling zone B – Mitrovica				
Ni	0.012			
Cr	-0.267	0.96		
As	0.986	-0.156	-0.425	
Cd	0.966	0.269	0.01	0.91
Sampling zone C – Obiliq, TC (power plant)				
Ni	0.684			
Cr	-0.993	-0.766		
As	-0.258	0.528	0.142	
Cd	-0.98	-0.526	0.95	0.445

3.3. CONTAMINATION FACTOR (CF_i) AND POLLUTION LOAD INDEX (PLI)

The soil was classified as having a high contamination degree in three tested regions, with middle contamination degree due to Pb, Ni, As, and especially high contamination degree by Cd and As (M4) (Table 3, according to criteria given in Table 6). As a result, the contamination factor may provide a thorough assessment of the overall enrichment and contamination impact of various pollutant groups in the soil and toxic waste. Significant correlations indicate that they are thought to be industrial, originating from industrial activities such as the production of Fe-Ni and Pb-Zn ores. The three analyzed areas are about 20 km of airlines the one from other, which is thought to contribute to the accumulation of heavy metals in soil and toxic waste in these areas [3–8].

Table 3

Contamination factors (CF_i) and pollution load indices (PLI) of heavy metals in toxic waste, ash and soil in zones A, B and C

Sample	CF_i					PLI
	Pb	Ni	Cr	As	Cd	
Sampling zone A – Glogovc – New Co Ferronickeli						
M1 (scoria)	0.48	9.23	65.75	0.42	7.98	1.18
M2 (scoria)	0.50	12.57	17.92	1.31	6.28	1.13
M3 (soil)	0.44	7.66	6.36	0.28	3.02	1.10
Sampling zone B – Mitrovica						
M4 (waste)	33.64	0.37	0.26	94.78	12.36	1.20
M5 (waste)	0.90	0.07	0.16	0.27	0.42	1.00
M6 (soil)	6.93	0.89	1.25	0.83	5.46	1.09
Sampling zone C – Obiliq TC (power plant)						
M7 (ash)	0.66	3.49	28.39	5.57	4.04	1.14
M8 (ash)	0.51	3.19	42.62	1.48	4.42	1.16
M9 (soil)	0.96	3.46	9.71	1.16	1.66	1.10
Baseline reference value, mg/kg	100	100	100	50	5	$PLI > 1$ (contaminated)

The PLI was derived from the CF_i values to calculate the toxic metal pollution [18–23]. In all toxic waste, ash, and soil samples, the analyzed $PLI > 1$ (Table 3) indicates the presence of sample pollution.

3.4. ECOLOGICAL RISK ASSESSMENT (ERI)

The ERI index stands for the potential ecological risk factor of toxic metals [21, 24, 26]. The estimated ERI values for chosen metals (Pb, Ni, Cr, As, and Cd) are presented in Table 4. From the obtained results and criteria (Table 6), all toxic waste, ash, and soil samples showed a high ecological risk [17, 18, 26]. The maximum ERI value (379.12) was observed for As and the lowest one (0.32) was noted for Cr. This study revealed

that Cd in all zones posed the highest ecological threat, whereas, in zone A (M1), Cr exhibited the highest ecological risk (*ERI* amounted to 131.50). As (M4) and Pb (M4) in zone B posed the highest ecological threat (the *ERI* values reached 379.12 and 168.20, respectively). The study also revealed that Cr in zone C (M8) posed the highest ecological threat (*ERI* amounted to 85.24).

Table 4

Ecological risk assessment (*ERI*) of heavy metals in scoria, waste, soil and ash samples

Sample	Pb	Ni	Cr	As	Cd
Sampling zone A – Gillogovc – New Co Ferronickeli					
M1 (scoria)	2.40	46.15	131.50	1.68	55.86
M2 (scoria)	2.50	62.85	35.84	5.24	43.96
M3 (soil)	2.20	38.30	12.72	1.12	21.14
Sampling zone B – Mitrovica					
M4 (waste)	168.20	1.85	0.52	379.12	86.52
M5 (waste)	4.50	0.35	0.32	1.08	2.94
M6 (soil)	34.65	4.45	2.50	3.32	38.22
Sampling zone C – Obilic, TC (power plant)					
M7 (ash)	3.30	17.45	56.78	22.28	28.28
M8 (ash)	2.55	15.95	85.24	5.92	30.94
M9 (soil)	4.80	17.30	19.42	4.64	11.62

3.5. GEOACCUMULATION INDEX (I_{Geo}) OF TOXIC METALS IN SOIL

The calculated values of the geoaccumulation indices (I_{Geo}) for toxic metals in soils at the industrial zones A, B, and C are presented in Table 5. I_{Geo} is highly variable, implying that the soil surrounding the sampling area was from strongly to moderately contaminated with the metals tested. The average values obtained for I_{Geo} (zone A) were as follows: Pb 9.61, Ni 16.00, Cr 17.61, As – 11.31, and Cd – 10.21. It can be seen that the maximum value has been reached by Cr (17.61). For values of $I_{\text{Geo}} > 5$ (Table 6) [22, 23] soil is extremely contaminated. Based on the average values of all toxic metals (zone A) it can be concluded that the soil belongs to the class extremely contaminated (Table 5). For zone B, the average values obtained for I_{Geo} were as follows: Pb 1.20, Ni –1.76, Cr –1.43, As 5.60, and Cd 5.66. It can be seen that the maximum value has been reached for Cd (5.42). Based on the average values for all toxic metals in zone B, it can be concluded that the soil belongs to the partially contaminated class. As for zone C, for the same elements, their values were Pb .08, Ni 1.17, Cr 4.16, As – 2.05, and Cd – 4.81 (strongly to extremely contaminated). Also, this zone is practically contaminated.

The distribution of items that were compared throughout the three research topics is shown in Fig. 2. The letters A, B, and C are included in brackets to denote the research area. The metals are divided into three categories by the first main component [24] (at different zones).

Table 5

Geoaccumulation index (I_{geo}) of heavy metals in soils

Sampling zone A – Glogovc, New Co Ferronickeli		
Metal	I_{geo} value	I_{geo} class
Pb	9.61	extremely contaminated
Ni	16.00	extremely contaminated
Cr	17.61	extremely contaminated
As	11.31	extremely contaminated
Cd	10.23	extremely contaminated
Sampling zone B – Mitrovica		
Metal	I_{geo} value	I_{geo} class
Pb	1.20	uncontaminated to moderately contaminated
Ni	-1.76	practically uncontaminated
Cr	-1.43	practically uncontaminated
As	5.60	extremely contaminated
Cd	5.66	extremely contaminated
Sampling zone C – Obiliq, TC (power plant)		
Metal	I_{geo} value	I_{geo} class
Pb	-3.08	practically uncontaminated
Ni	1.17	uncontaminated to moderately contaminated
Cr	4.16	strongly contaminated
As	2.05	moderately to strongly contaminated
Cd	4.81	strongly to extremely contaminated

Table 6

Pollution/ecological indices, classification, and interpretation [17–26]

Index	Classification	Interpretation
I_{geo}	$I_{geo} < 0$	uncontaminated
	$I_{geo} < 2$	between uncontaminated and moderately contaminated
	$2 < I_{geo} < 3$	between moderately and strongly contaminated
	$3 < I_{geo} < 4$	strongly contaminated
	$4 > I_{geo} < 5$	between strongly and extremely contaminated
	$I_{geo} \geq 5$	extremely contaminated
CF_i	$CF_i < 1$	low contamination
	$1 \leq CF_i < 3$	moderate contamination
	$3 \leq CF_i < 6$	considerable contamination
	$CF_i \geq 6$	very high contamination
PLI	$PLI = 0$	perfection
	$PLI < 1$	baseline level
	$PLI > 1$	contaminated
ERI	$ERI < 40$	low ecological risk
	$40 \leq ERI < 80$	moderate ecological risk
	$80 \leq ERI < 160$	considerable ecological risk
	$160 \leq ERI < 320$	high ecological risk
	$ERI \geq 320$	very high ecological risk

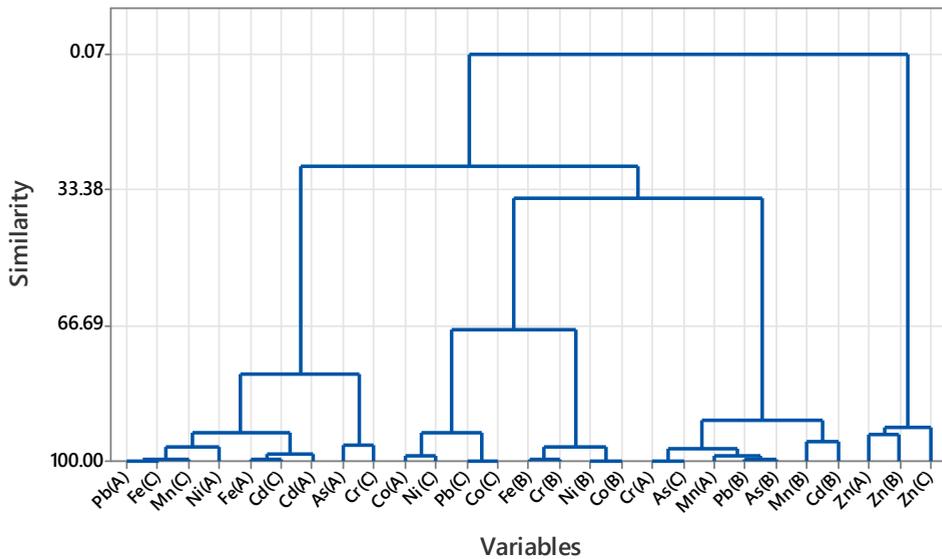


Fig. 2. Dendrogram analysis of the elements present in the locations zone (zone A, B and C)

The group consists of three component groups for metals: Cr, As, Mn, Cu, Pb, Cd, and Co, and one element, Zn. The first basic component group for metals is Pb, followed by Fe, Ni, Mn, Cu, Cd, As, and Cr. The second basic component group which comprises the metals is made up of the following elements: a) Pb, Fe, Ni, Mn, Cu, Cd, As, and Cr, b) Cr, As, Mn, Cu, Pb, Cd, and Co, and c) Zn. This supports the regression results, confirming their pollution source. Generally, the above results show the metal pollution distribution within the different sampling positions. The study confirms that the use of toxic waste, scoria-sterile material, ash and soil, can serve to monitor heavy metals [25–28].

4. CONCLUSIONS

A vital part of the biosphere is the soil, and any deterioration in its quality can have negative effects on human life. Heavy metals in the soil pose a serious hazard to human health, especially when they enter the food chain. Fe, Ni, Cr, Mn, Cu, Zn, and Pb contamination levels were significantly higher than those recommended by the Dutch Target and Intervention Values 2000 and Environmental Protection Agency.

Mining activities that can release substantial amounts of heavy metals into the environment have a significant impact on the occurrence of metal contamination in the study zones. The soil around the test site was contaminated with heavy metals, according to the highly variable geoaccumulation index, a measure of soil contamination. In zone A, heavy metal concentrations were 2–3 times higher than those in zones B and C. Based on the average values of the geoaccumulation index for all toxic metals (zone B)

it can be concluded that the soil belongs to the partially contaminated class. Also, zone C is practically contaminated.

The obtained results call for mandatory and long-term monitoring of toxic metals in the soil, as well as comprehensive research about adverse environmental and health factors. The national government and the local residents in general need to acknowledge the grave risks of toxic metal contamination and prioritize the work on sustainable solutions to mitigate the current risks.

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