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Risk assessment of potentially toxic elements in soil surrounding the Golesh ferronickel mine, Kosovo

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Abstract

Purpose. The objective of this study was to assess the risk of potentially toxic elements in soil samples surrounding ferronickel mines in the Golesh massif, Republic of Kosovo.

Methods. In total, 14 potentially toxic elements (Al, As, Cd, Co, Cr, Cu, Fe, Li, Mg, Mn, Ni, Pb, V and Zn) were investigated. Basic statistics, Pearson correlation, Principal Component Analysis (PCA), and Pollution indices (CF, PLI, I_{geo}, and EF) were used to explain better the data on metal concentrations in the soil samples.

Findings. Five groups of elements were identified by PCA, based on their geogenic or anthropogenic origin. The contamination factor for nickel ranged from 6.9 to 166, with a mean value of 65.17. Cobalt and magnesium also had high mean values of contamination factor: 10.38 and 9.76, respectively. The PLI_{site} for 14 locations were highly polluted with metals ($PLI > 4$), and the PLI_{zone} of the whole territory investigated was 3.5. The mean value of I_{geo} for nickel was 5.44, for cobalt (2.79) and for magnesium (2.7). The mean value of enrichment factor (EF) for nickel, cobalt and magnesium was 233.7, 35.26 and 19.16, respectively.

Originality. Soil samples were collected from 30 different locations in accordance with the soil sampling protocol. The samples were sent for further analysis at the ACME, Ltd. laboratory in Vancouver, Canada. The soil samples were digested with aqua regia, and the content of 14 chemical elements was determined using inductively coupled plasma-mass spectrometry (ICP-MS).

Practical implications. Based on statistical analysis and pollution indices, we concluded that most soil samples were highly polluted with Ni, Co, and Mg, resulting from the ferronickel and magnesite mines located in the region under investigation.

Keywords: *heavy metal, soil, ICP-MS, ferronickel mine, Golesh, Kosovo*

1. Introduction

The mining, smelting, and processing of metal ores have been identified as significant sources of potentially harmful elements (PHEs) within the environment [1]. Soil pollution is a type of land degradation that occurs when a range of natural or anthropogenic constituents exceed maximum permissible levels in natural soil environments [2]. One of the most pressing problems, as we now know, is that soil pollution by heavy metals is widespread in every country worldwide and has become a serious environmental issue [3]-[5]. Geochemical background assessment is an appropriate approach for determining whether concentrations of potentially toxic elements (PTEs) in soil are geogenic, resulting from natural factors and processes, or have been influenced by human activities, such as those from industrial plants, vehicle traffic, and agriculture [6]-[10]. Potentially toxic elements can occur naturally in soils in varying concentrations, depending on the geochemical composition of source rocks and soil formation processes, such as weathering, sedimentation, and volcanic eruptions [2].

The Republic of Kosovo is rich in natural mining resources, among which energy and colored metal resources represent the most significant potential for overall development. In this aspect, it is worth explicitly noting lignite, lead, zinc, silver, and gold, as well as silicate mines of nickel and cobalt, iron-nickel, bauxite, manganese, and a considerable number of non-metallic minerals, including industrial and construction geological materials [11], [12]. The development of Kosovo's economy, which is primarily based on its natural resources, is closely tied to the growth of the mining sector. A significant role in the exploitation of these assets is played by the underground extraction of valuable minerals, including those of magnesium, iron, nickel, and cobalt in the Golesh mines [11]-[13].

Nickel and Cobalt occurrences are found in the centre of Kosovo, located near Glavica, north of Magurë (Golesh), and in Çikatovë (Drenas). Explorations on Ni in Kosovo began in 1961 in the Golesh massif [14]-[17].

The Golesh ultramafic massif forms a roughly pyramidal body, whose base is outlined by the villages of Stankovc (west), Harilaq (northeast), and Magurë (southeast), extending approximately 7 km in length and 4.5 km in width. The massif represents a segment of the Balkan ophiolite belt, which is part of the Vardar Zone, formed during the closure of the Neotethyan Ocean [18], [19].

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The Golesh mine itself is located near the village of Magurë, within the municipality of Lipjan, on Golesh Mountain (1019 m), approximately 3 km west of Pristina International Airport [20]. Initially exploited as an open-pit operation, the magnesite ($MgCO_3$) deposit later transitioned to underground mining before its closure in 2002.

The Gllavica silicate nickel mine, located about 20 km southwest of Pristina, lies within the peridotite massif of Golesh on the eastern slope of Gllavica Hill. The deposit extends north-south over approximately 1050 m and covers around 32 hectares [21]. The Kodrina and Çikatova deposits are part of the exogenous type in the silicate-lateritic group of nickel ores, which were created before the beginning of the Tertiary. The average ore content of Kodrina is: 1.33% Ni, 0.065% Co, 49.35% SiO_2, 20.24% Fe_2O_3, and 13.1% MgO. The ore is part of a mineralization with an increasing content of nickel, whose bearers are nontronite, pimelite, and garnierite [17].

The Golesh massif is composed mainly of ultramafic rocks – predominantly harzburgite, with subordinate lherzolite, enstatite-dunite, and dunite – all strongly affected by serpentinization along their margins and major fracture zones. Locally, the massif is capped by relics of Paleogene lateritic weathering crusts, indicative of prolonged subaerial exposure [22].

The massif exhibits extensive serpentinization, resulting in the formation of secondary serpentine-group minerals, including lizardite, antigorite, and chrysotile, alongside iron oxides and hydroxides [23]. These minerals impart the greenish-gray color and friable texture typical of serpentinized ultramafic complexes within the Balkan ophiolite belt [24].

The Golesh vein magnesite deposits are located on the Golesh, which is predominantly composed of ultramafic rocks (the well-known Golesh ultramafic massif), 15 km southwest of Prishtina, Kosovo. This is the largest vein magnesite deposit in both Yugoslavia and the world, with total reserves of cryptocrystalline magnesite of approximately 5 million tons [25]. The veins typically range from 0.5 to 3 m in thickness, reaching up to 20 m at Magura, and extend 100-500 m in length (with a maximum of ~1200 m at Magura). The mineralization persists to depths exceeding 300 m at Magura. Such features are characteristic of hydrothermal vein systems developed in brittle ultramafic hosts [26].

The industry development in a country increases the well-being of the population, but it also causes environmental damage [27], [28]. Recently, numerous research studies have been conducted in Kosovo regarding the impact of industry and various industrial and municipal landfills on the pollution of water, air, soil and food with heavy metals [29]-[35].

The primary objective of this study was to evaluate the indicators of heavy metal contamination in soil samples collected from the area surrounding the ferronickel mines in the Golesh massif. This research work also serves as a basis for subsequent studies, with a particular emphasis on analyzing the deeper layers of the soil profile to gain a more comprehensive understanding of the vertical distribution and potential sources of contamination.

Fourteen chemical elements were analyzed by using inductively coupled plasma-mass spectrometry (ICP-MS). Statistical analysis was used to calculate Pearson correlation and Principal component analysis between different metals and locations. Four pollution indices: contamination factor (*CF*), pollution load index (*PLI*), geoaccumulation index (I_{geo}) and enrichment factor *EF*), were calculated to assess the level of soil pollution with heavy metals.

2. Methods

2.1. Study area

The mountain of Golesh (with a surface area of 22.2 km^2) belongs to the central mountain chain of the Carraleva Mountains in Kosovo, and its highest peak rises 1019 m above sea level (a.s.l). It is the highest mountain system in the central part of Kosovo [36], [37]. The Golesh mine is located near the village of Magura, within the municipality of Lipjan, on the Goleshi Mountain, approximately 3 km west of Pristina Airport. The Gllavica silicate nickel mine is located approximately 20 km southwest of Pristina, situated within the peridotite massif of Golesh on the eastern slope of Gllavica Hill. The deposit runs almost exactly north to south, stretching approximately 1050 meters in length and covering an area of around 32 hectares [15], [16], [38].

2.2. Sampling

In this study, we first prepared a network of maps for sampling in the Golesh region and then proceeded with the collection of soil samples at 30 different locations (Fig. 1). From each location, we collected samples from the soil surface to a depth of 50 cm [39].

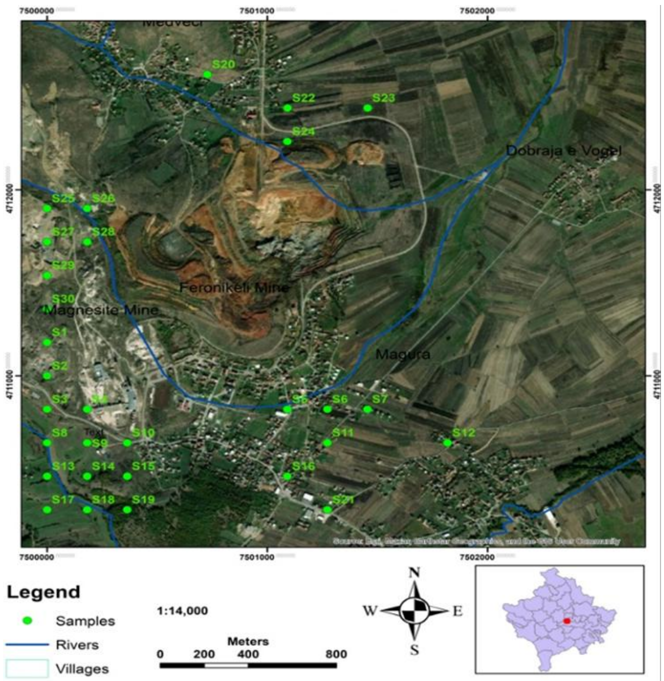

Figure 1. Location map of the study area and sampling points

The samples were stored in plastic bags, cleaned of non-soil elements (such as stones, sticks, and other debris), and dried at room temperature for 2 weeks. They were then crushed and sieved through a 63 μm sieve to obtain a fine powder [40]. Chemical analyses of soil samples were performed at the ACME, Ltd. laboratory in Vancouver, Canada. The soil samples were digested with aqua regia, and the content of 14 chemical elements (Al, As, Cd, Co, Cr, Cu, Fe, Li, Mg, Mn, Ni, Pb, V and Zn) was determined by using inductively coupled plasma-mass spectrometry (ICP-MS). The lower detection limits for elements Al, Fe, and Mg were 0.01%; for Cr, Mn, and V, they were one ppm; for As, Co, Li, Ni, Pb, and Zn, they were 0.2 ppm; and for Cu and Cd, they were 0.01 and 0.01 ppm, respectively.

2.3. Statistical analysis

The statistical analysis was conducted using PAST 4.11 software. This included calculating parameters such as minimum, maximum, mean, median, standard deviation, Pearson correlation and Principal component analysis. Pollution indices (*CF*, *PLI*, I_{geo} and *EF*) were calculated based on European soil samples [41].

2.4. Pollution indices

To assess the level of pollution, four pollution indices were calculated: contamination factor, pollution load index, geo-accumulation index and enrichment factor [41]-[43].

The contamination factor (CF) of each metal in the soil samples was calculated by the formula:

$$CF = \frac{c_m^i}{c_r^i}, \quad (1)$$

where:

CF – the contamination factor for a heavy metal;

c_m^i – the measured value of the heavy metal in the soil;

c_r^i – the parameter for calculation, with reference to the background values for heavy metals in European soil samples [41].

Contamination factor values are categorized by Fernandez [44] as follows:

– $CF < 1$ – no contamination;
– $1 \le CF \le 2$ – suspected contamination;
– $2 \le CF \le 3.5$ – slightly contaminated;
– $3.5 \le CF \le 8$ – moderate;
– $8 \le CF \le 27$ – severe;
– $CF > 27$ – extreme contamination factor [35].

For the total assessment of the contamination degree in soil, the *PLI* is calculated by the formula:

$$PLI = \sqrt[n]{CF_1 \cdot CF_2 \cdot CF_3 \cdot \ldots \cdot CF_n}, \quad (2)$$

where:

CF – the contamination factor of each metal, and n is the number of metals calculated.

When *PLI* is greater than 1, the contamination exists, and if *PLI* is less than 1, there is no metal contamination [42].

To calculate metal concentrations above background levels, the geo-accumulation index I_{geo} was used, as shown below:

$$I_{geo} = \frac{\log_2 c_n}{1.5 B_n}, \quad (3)$$

where:

C_n – is the concentration of the element;

B_n – is the average of European soil samples [41].

Müller [45] proposed the following classification:

– $I_{geo} > 5$ – extremely contaminated;
– $4 < I_{geo} < 5$ – strongly to extremely contaminated;
– $3 < I_{geo} < 4$ – strongly contaminated;
– $2 < I_{geo} < 3$ – moderately to strongly contaminated;
– $1 < I_{geo} < 2$ – moderately contaminated;
– $0 < I_{geo} < 1$ – uncontaminated to moderately contaminated;
– $I_{geo} \le 0$ uncontaminated.

Enrichment factor (*EF*) is a measure of the possible impact of anthropogenic activity on the concentration of heavy metals in soil. The Enrichment factor (*EF*) was calculated using the formula originally introduced by Adnan et al. [10], [46]:

$$EF = \frac{\left|\frac{C_n}{C_{ref}}\right| sample}{\left|\frac{B_n}{B_{ref}}\right| background}, \quad (4)$$

where:

C_n – the content of the target element;

C_{ref} – the content of the reference element (generally elements such as Mn, Al, Sr, Fe, and Ti). B_n is the content of the studied chemical element in the soils, and B_{ref} is the reference element content in the soils of Europe or the world [46].

According to Adnan et al. [10], [46]:

– $EF<2$ – no to minimal enrichment;
– $EF = 2\text{-}5$ – moderate enrichment;
– $EF = 5\text{-}20$ – significant enrichment;
– $EF = 20\text{-}40$ – very high enrichment;
– $EF > 40$ – extremely high enrichment.

3. Results and discussion

The determination of 14 potentially toxic elements (Al, As, Cd, Co, Cr, Cu, Fe, li, Mg, Mn, Ni, Pb, V and Zn) was carried out in 30 soil samples collected near the ferronickel and magnesite mines in Magura village (Golesh massif). To present and better understand the data on chemical element concentrations, we employed a statistical data processing method. In Table 1, we present the basic statistics of our data, along with the median values for European soils [41].

The median values for Al (16850 mg/kg), Li (10.35 mg/kg), and V (34 mg/kg) in our study were lower compared to the median values for European soils. The median values of our samples for As and Zn were very close to the European medians. In contrast, the median values of all other elements were many times higher than those of European soils. The high average concentration was for elements: 47200 mg/kg Fe, 44890 mg/kg Mg, 15353 mg/kg Al, 1179.9 mg/kg Mn, 1173 mg/kg Ni, 191.8 mg/kg Cr and 80.98 mg/kg Co. Nickel, cobalt, magnesium, chromium, manganese and iron had median values that were 65, 10.2, 5.56, 2.87, 2.23 and 1.9 times higher compared to European soils [41]. Numerous recently published research studies in Kosovo, using plants (moss and pollen) as bioindicators, confirm that the magnesite mine in Magura and the ferronickel mines affect air pollution with nickel, chromium, and cobalt [47]-[50].

The Pearson correlation coefficient (*r*) and its significance level (*p-values*) for 14 potentially toxic elements in 30 different locations are presented in Table 2. The absolute value between 0.50 and 0.70 exhibits a good correlation, and from 0.70 to 1.00, a strong correlation is observed [35],[42].

Aluminum exhibited five strong and positive correlations: Al-V (0.97), Al-Li (0.94), Al-As (0.69), Al-Zn (0.66), and Al-Cu (0.63). Arsenic showed five strong and positive correlations: As-Cu (0.83), As-Zn (0.83), As-Pb (0.78), As-V (0.74), and As-Li (0.72). Additionally, copper exhibited four strong and positive correlations: Cu-Zn (0.89), Cu-Pb (0.69), Cu-V (0.66), and Cu-Li (0.60). Cobalt had three correlations: Co-Fe (0.90), Co-Ni (0.89) and Co-Mn (0.56). Chromium and lithium had two strong and good positive correlations: Cr-Fe (0.58), Cr-Mn (0.56), Li-V (0.93), Li-Zn (0.67), and Fe-Ni (0.82), Mg-Ni (0.75), Pb-Zn (0.72), V-Zn (0.67) were with one strong and good positive correlations.

Table 1. Descriptive statistics for the content (in mg/kg) of the metals in soil samples

Element	N	Min	Max	Mean	Stand. dev	Median	P25	P75	Skewness	Kurtosis	EU Median [41]
Al	30	800	42400	15353	9466	16850	7350	20800	0.53	0.81	58000
As	30	0.6	18.9	8.07	4.09	8.9	4.4	10.55	0.19	0.36	7
Cd	30	0.07	1.03	0.25	0.18	0.22	0.138	0.295	3.04	12.62	0.15
Co	30	20.1	149	80.98	45.35	79.9	34.58	124.5	0.09	-1.50	7.8
Cr	30	45	496	191.80	108.7	172	106	257.75	0.92	0.73	60
Cu	30	9.8	38.5	23.28	7.67	23.2	16.95	27.825	0.31	-0.39	13
Fe	30	23800	74600	47200	14124.4	47750	32900	60000	0.15	-0.96	25000
Li	30	1	25	9.38	5.73	10.35	3.65	13.15	0.50	0.24	20
Mg	30	3500	100000	44890	39819.5	25600	12450	100000	0.57	-1.58	4600
Mn	30	390	2270	1179.9	461.5	1115	869	1427.5	0.67	0.08	500
Ni	30	124	2990	1173	842.2	1170	287	1805	0.38	-0.93	18
Pb	30	1.6	98.9	41.65	22.6	46.45	19.38	53.6	0.09	0.10	23
V	30	4	69	30.03	14.6	34	16.5	39	0.11	0.29	60
Zn	30	26.6	107	64.01	22.5	64.85	42.78	80.93	0.14	-0.82	52

Table 2. Pearson correlation coefficients between metal concentrations in soil samples (n = 30)

Element	Al	As	Cd	Co	Cr	Cu	Fe	Li	Mg	Mn	Ni	Pb	V	Zn
Al		0.00	0.45	0.04	0.98	0.00	0.41	0.00	0.00	0.77	0.00	0.03	0.00	0.00
As	0.69		0.06	0.00	0.41	0.00	0.02	0.00	0.00	0.17	0.00	0.00	0.00	0.00
Cd	0.14	0.34		0.64	0.11	0.03	0.74	0.70	0.39	0.38	0.33	0.01	0.27	0.01
Co	-0.38	-0.55	-0.09		0.00	0.08	0.00	0.00	0.01	0.00	0.00	0.03	0.03	0.06
Cr	0.01	-0.16	0.30	0.65		0.37	0.00	0.28	0.73	0.00	0.01	0.36	0.99	0.35
Cu	0.63	0.83	0.40	-0.33	0.17		0.30	0.00	0.00	0.02	0.00	0.00	0.00	0.00
Fe	-0.16	-0.44	-0.06	0.90	0.58	-0.20		0.05	0.02	0.02	0.00	0.01	0.31	0.25
Li	0.94	0.72	0.07	-0.57	-0.20	0.60	-0.36		0.00	0.68	0.00	0.03	0.00	0.00
Mg	-0.81	-0.82	-0.16	0.49	0.07	-0.74	0.42	-0.85		0.27	0.00	0.00	0.00	0.00
Mn	0.05	0.26	0.17	0.56	0.56	0.43	0.43	-0.08	-0.21		0.23	0.07	0.69	0.06
Ni	-0.64	-0.75	-0.19	0.89	0.45	-0.58	0.82	-0.77	0.75	0.23		0.00	0.00	0.00
Pb	0.41	0.78	0.46	-0.40	0.17	0.69	-0.46	0.40	-0.61	0.33	-0.59		0.01	0.00
V	0.97	0.74	0.21	-0.39	0.00	0.66	-0.19	0.93	-0.86	0.08	-0.64	0.47		0.00
Zn	0.66	0.83	0.45	-0.35	0.18	0.89	-0.22	0.67	-0.76	0.34	-0.60	0.72	0.67	
Significance $p < 0.01$														

Most elements with a strong positive correlation had the exact geogenic or anthropogenic origin [48], [49]. Also, some metals had strong and good negative correlations with each other, for example, magnesium had the following correlations: Mg-Al (-0.81), Mg-As (-0.82), Mg-Li (-0.85), Mg-V (-0.86), Mg-Zn (-0.76), Mg-Cu (-0.74) and Mg-Pb (-0.61).

Based on data from Table 2, a total of 40 associations with absolute values between 0.5 and 0.97 had a significance level of less than 1% ($p < 0.01$).

To identify the distribution of potentially toxic metals in soil samples and possible sources of contamination, we used principal component analysis (PCA). From the results presented in Figure 2, it can be seen that PC1 accounted for 53.28% of the total variance, while PC2 accounted for 22.09%. In Figure 2, we can see that, based on the origin and correlations between chemical elements, five different groups of elements can be distinguished. The first group, formed by magnesium (highly loaded in PC1), was the most concentrated element, since the Magure magnesite mines are located in this area. This metal did not have good correlations with other elements, and as a result, formed a separate group, which is mainly of geogenic origin [48]. The 2nd group is formed by iron, cobalt and nickel. Their vectors loaded highly in PC1 and PC2, indicating a shared geological origin, but are also influenced by anthropogenic factors. The vectors of Cr and Mn (3rd group) were similar and loaded highly in PC2, indicating that they have both geogenic and anthropogenic origins.

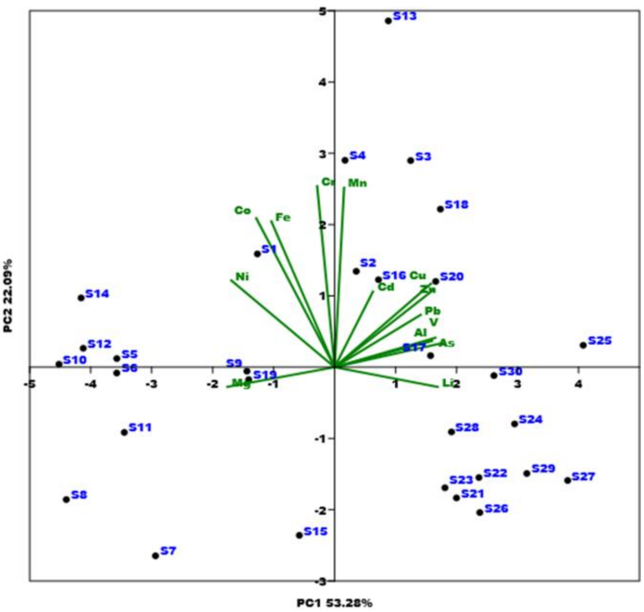

Figure 2. Principal component analysis (PCA) of 14 metals for 30 soil samples

Potentially toxic metals (Al, As, Cd, Cu, Pb, V, and Zn) form the fourth group and are present in large quantities due to anthropogenic pollution. Lithium forms the 5th group, which is loaded in PC1, and is influenced by anthropogenic factors, similar to the 4th group of elements.

Based on the data presented in Table 1, we calculated the contamination factor for chemical elements that had higher concentrations compared to the European median values. The contamination factor (*CF*), minimum, maximum and mean, for nine potentially toxic elements (Cd, Co, Cr, Cu, Fe, Mg, Mn, Ni and Pb), are presented in Table 3.

According to the categorization of the *CF* values provided by Fernandez [44], the zone investigated was highly contaminated with nickel, with CF values ranging from 6.89 to 166.11, and a mean value of 65.17. The zone was severely contaminated with cobalt, with a *CF* value ranging from 2.58 to 19.1 (CF_{mean} = 10.38), and magnesium, with a CF ranging from 0.76 to 21.74 (CF_{mean} = 2.36). The zone was slightly contaminated with chromium (CF_{mean} = 3.2) and manganese (CF_{mean} = 10.38), and other elements had the mean value of *CF* between 1 and 2 (suspected contamination).

The *PLI* site values for nine metals (Cd, Co, Cr, Cu, Fe, Mg, Mn, Ni, and Pb) throughout the sampled area are presented in Figure 3. According to Zhang [51] categorization, none of the soil sampling sites falls into the category of unpolluted sites. Three samples (26, 27, and 23) fall into the moderately to unpolluted category. Seven samples were moderately polluted, and 6 of them were moderately to highly polluted. Nine soil samples correspond to the highly polluted category, and five of them were very highly polluted. The most polluted sites were locations 1, 4, 13 and 19, which are close to the magnesite mine. The PLI zone for the sampled area was 3.5, corresponding to moderately to highly polluted category.

From the median value of our results, we have also calculated an important pollution indicator, the geoaccumulation index (I_{geo}), as presented in Figure 4. Based on Müller classification [45], the soil surrounding the Fero-nickel mines in Magure was highly contaminated with nickel (I_{geo} = 5.44), moderately to strongly contaminated with cobalt (I_{geo} = 2.79) and magnesium (I_{geo} = 2.7), moderately contaminated with Cr (I_{geo} = 1.09) and other elements fall into the uncontaminated category.

Table 3. Contamination factor (CF) for nine metals in soil samples (n = 30)

Element	Cd	Co	Cr	Cu	Fe	Mg	Mn	Ni	Pb
S1	1.87	15.90	5.62	1.60	2.12	5.70	2.60	133.33	2.22
S2	1.67	14.87	4.25	1.68	2.14	4.83	3.28	71.11	1.76
S3	1.47	18.21	5.07	2.17	2.72	3.70	3.68	77.78	2.33
S4	1.73	19.10	4.67	1.95	2.67	6.50	4.28	90.56	1.93
S5	0.87	16.15	2.78	1.24	2.55	20.35	2.36	131.11	0.52
S6	0.93	14.62	3.42	1.29	2.44	21.74	2.02	124.44	0.71
S7	0.67	8.47	1.62	0.75	1.16	21.74	1.45	71.67	2.21
S8	0.53	12.42	1.52	1.03	1.96	21.74	1.62	104.44	0.07
S9	2.27	11.42	2.65	1.61	2.03	21.74	2.42	82.78	1.49
S10	0.73	17.95	2.50	0.88	2.78	21.74	2.30	133.89	0.40
S11	0.87	12.44	3.32	1.31	1.91	21.74	1.85	98.89	0.59
S12	1.00	17.18	3.27	1.39	2.39	21.74	2.78	122.78	0.28
S13	2.87	18.72	8.27	2.95	2.48	6.17	4.54	90.00	3.15
S14	0.47	18.85	4.18	1.08	2.98	19.83	2.24	166.11	0.50
S15	1.47	5.33	2.38	1.12	1.27	8.63	1.17	38.56	0.89
S16	2.27	13.85	4.43	1.83	1.98	3.17	3.08	61.11	1.82
S17	1.60	9.96	2.95	1.81	1.76	3.20	2.68	44.28	2.03
S18	1.80	10.53	6.65	2.62	1.91	3.63	3.38	48.94	2.85
S19	2.53	8.46	5.65	1.37	1.61	ma	1.62	68.89	2.95
S20	6.87	8.40	3.98	2.20	2.02	10.87	1.78	52.83	2.31
S21	1.07	3.77	1.82	1.72	1.22	2.41	1.41	14.28	2.85
S22	2.47	3.24	2.08	1.98	1.26	5.43	1.13	16.50	2.33
S23	1.27	4.65	1.32	1.76	0.95	0.76	2.56	7.39	2.41
S24	1.27	3.54	1.33	2.96	1.47	1.35	2.22	9.94	2.17
S25	2.67	3.04	0.98	2.85	1.18	2.11	3.78	9.94	4.30
S26	1.47	2.58	0.88	2.02	1.12	1.09	1.91	6.89	1.66
S27	0.53	3.00	2.12	2.24	1.89	1.33	0.78	9.22	1.45
S28	1.73	5.09	2.13	2.13	1.50	3.15	2.08	25.06	2.03
S29	1.13	3.17	0.75	2.09	1.33	1.85	1.84	9.89	2.13
S30	1.07	6.58	3.32	2.09	1.86	2.80	1.95	32.39	2.01
Min	0.47	2.58	0.75	0.75	0.95	0.76	0.78	6.89	0.07
Max	6.87	19.10	8.27	2.96	2.98	21.74	4.54	166.11	4.3
Mean	1.64	10.38	3.20	1.79	1.89	9.76	2.36	65.17	1.81

The enrichment factor (*EF*) is a measure of the potential impact of anthropogenic activity on the concentration of heavy metals in soil [10], [43]. This factor was calculated as a ratio between the obtained median values for specific metals and the European median values of surface soil [41], [46]. In our study, aluminium was used as the reference element in European median soils with values of 58000 mg/kg [41]. The median value of aluminium in the soil of the whole study area was 16850 mg/kg. In Figure 5, we presented the *EF*, calculated from median values of the metal concentrations.

The whole zone had extremely high enrichment with nickel (*EF* = 233.7), very high enrichment with cobalt (*EF* = 35.26), seven elements (Cd, Cr, Cu, Fe, Mg, Mn and Pb) fall into the significant enrichment category, two of them (As and Zn) fall into the moderate enrichment category, and three others (Al, Li and V) fall into no and minimal enrichment category.

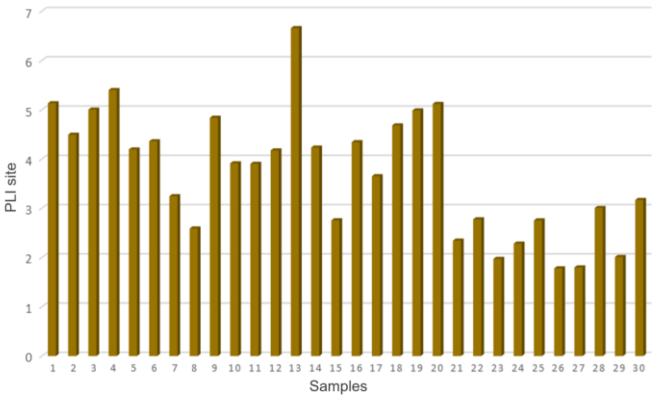

Figure 3. Pollution load index calculated for nine metals in soil samples (n = 30)

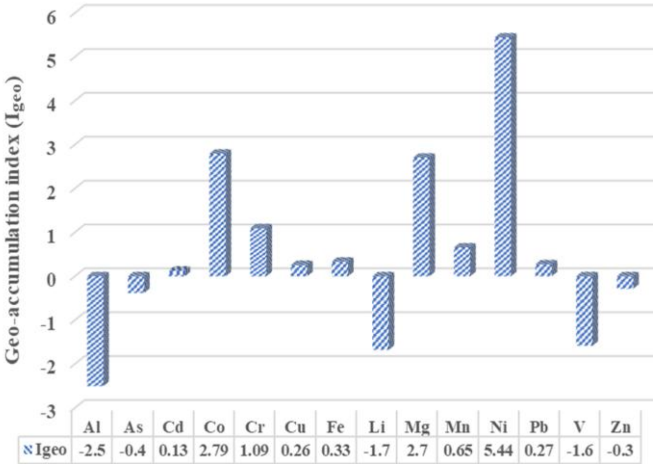

Figure 4. Geo-accumulation index (I_{geo}) for 14 metals in soil samples (n = 30)

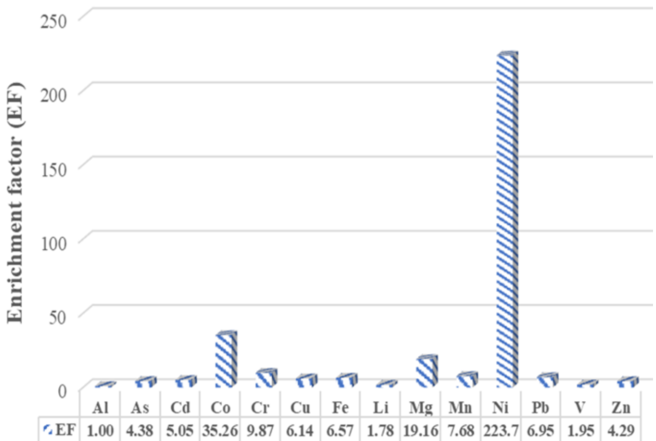

Figure 5. Enrichment factor of 14 metals in the whole study area (n = 30)

The very high concentration of nickel, cobalt, and other potentially toxic elements in the analyzed soils also affects the pollution of the surrounding soil, water, plants, and food [31], [35], [47]-[50]. This occurs because dust particles are dispersed by winds over various distances, up to 20-30 km.

4. Conclusions

In this work, we investigated 14 chemical elements (Al, As, Cd, Co, Cr, Cu, Fe, Li, Mg, Mn, Ni, Pb, V, and Zn) in soil samples surrounding the Golesh Ferronickel mine in Magure using inductively coupled plasma-mass spectrometry (ICP-MS).

The median values of Al, Li, and V were lower compared to European median values; however, the median values of 11 other elements were significantly higher than comparable values. Using the Pearson correlation coefficient, we identified 40 associations with absolute values ranging from 0.5 to 0.97, with a significance level of less than 1% ($p < 0.01$). Strong positive correlations were observed between the following elements: Co-Fe, Co-Ni, Fe-Ni, Cu-Zn, As-Cu, and As-Zn. Principal component analysis identified five groups of elements based on their geogenic and anthropogenic origin: 1st group was formed by magnesium, 2nd group was formed by Fe, Co and Ni, 3rd group – by Cr and Mn, 4th group – by Al, As, Cd, Cu, Pb, V and Zn, and the last group was formed by lithium.

Four different pollution indices (*CF*, *PLI*, I_{geo} and *EF*) were used as tools for the comprehensive evaluation of the degree of contamination of soil samples with metals. All pollution indices verified that the soils of the whole area investigated was highly contaminated with nickel ($CF = 65.17$, $I_{geo} = 5.44$ and $EF = 223.7$), strongly contaminated with cobalt ($CF = 10.38$, $I_{geo} = 2.79$ and $EF = 35.26$) and magnesium ($CF = 9.76$, $I_{geo} = 2.7$ and $EF = 19.16$). Other elements also contributed to soil contamination. The pollution load index for 14 locations had $PLI_{site} > 4$, and the whole area investigated had $PLI_{zone} = 3.5$, resulting from ferronickel and magnesite open mines in Magure.

Author contributions

Conceptualization: TS, EL; Data curation: EL, MP; Formal analysis: EL, MP; Funding acquisition: EL; Investigation: EL, MP; Methodology: TS, MP; Project administration: TS, EL; Resources: EL; Software: EL, MP; Supervision: TS, MP; Validation: EL, MP; Visualization: EL, MP; Writing – original draft: EL; Writing – review & editing: EL, TS, MP. All authors have read and agreed to the published version of the manuscript.

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Conflicts of interest

The authors declare no conflict of interest.

Data availability statement

The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

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Оцінка ризику потенційно токсичних елементів у ґрунті навколо феронікелевого рудника Голеш, Косово

Е. Лечай, Т. Серафімовскі, М. Пачарізі

Мета. Метою цього дослідження було оцінити ризик потенційно токсичних елементів у зразках ґрунту навколо феронікелевих рудників у масиві Голеш, Республіка Косово.

Методика. Всього було досліджено 14 потенційно токсичних елементів (Al, As, Cd, Co, Cr, Cu, Fe, Li, Mg, Mn, Ni, Pb, V і Zn). Базові статистичні показники, кореляція Пірсона, аналіз головних компонент (PCA) та індекси забруднення (*CF*, *PLI*, I_{geo} та *EF*) були використані для кращого пояснення даних щодо концентрації металів у зразках ґрунту.

Результати. За допомогою аналізу головних компонент (PCA) було визначено п'ять груп елементів, виходячи з їхнього геогенного або антропогенного походження. Коефіцієнт забруднення нікелем коливався від 6.9 до 166, із середнім значенням 65.17. Кобальт і магній також мали високі середні значення коефіцієнта забруднення: 10.38 і 9.76 відповідно. Індекс навантаження забрудненням (PLI_{site}) для 14 локацій вказував на високий рівень забруднення металами ($PLI > 4$), а значення PLI_{zone} для всієї досліджуваної території становило 3.5. Середнє значення I_{geo} для нікелю становило 5.44, для кобальту – 2.79, і для магнію – 2.7. Середнє значення індексу збагачення (*EF*) для нікелю, кобальту та магнію становило відповідно 233.7, 35.26 і 19.16.

Наукова новизна. Зразки ґрунту були зібрані з 30 різних локацій відповідно до протоколу відбору проб ґрунту. Зразки були відправлені для подальшого аналізу в лабораторію ACME Ltd. у Ванкувері, Канада. Зразки були піддані розкладанню за допомогою царської води, а вміст 14 хімічних елементів був визначений за допомогою мас-спектрометрії з індуктивно зв'язаною плазмою (ICP-MS).

Практична значимість. На основі статистичного аналізу та індексів забруднення, ми дійшли висновку, що більшість зразків ґрунту були сильно забруднені Ni, Co та Mg, що є наслідком діяльності феронікелевих та магнезитових рудників, розташованих у досліджуваному регіоні.

Ключові слова: *важкі метали, ґрунт, мас-спектроскопія з індуктивно-зв'язаною плазмою, феронікелевий рудник, Голеш, Косово*

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