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Ecological risk of heavy metal contamination in the municipality of Graçanica, Kosovo, from the tailings dump

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ABSTRACT

This study investigates the contamination and ecological risk of heavy metal concentrations in the residential area near the mining waste dump in Graçanica, a municipality in the Pristina District of Kosovo. Graçanica is located near the Kizhnica-Badovc-Hajvalia ore field and has historical significance in mining as part of the larger Trepça mining complex. The Kizhnica Flotation Plant processed lead and zinc ores from nearby mines, depositing its waste materials in tailings in Graçanica. Six soil samples were collected according to standards from areas near the waste dump and residential zones, and analyzed for arsenic (As), lead (Pb), and manganese (Mn) concentrations, as well as pH levels. Chemical analyses were conducted using inductively coupled plasma-atomic emission spectrometry (ICP-OES) equipment. The results showed that As, Pb, and Mn concentrations ranged from 7 to 1127 mg/kg, 69 to 1961 mg/kg, and 479 to 6440 mg/kg, respectively—exceeding the soil contamination limits set by Kosovo's Administrative Instruction (GRK), as well as permissible limits according to WHO and EU Directives. Moreover, soil pH ranged from 3.95 to 7.73, with acidic conditions potentially enhancing heavy metal mobility. The anthropogenic impact from this tailings dump may have penetrated deep into the natural environment of Graçanica. The increasing accumulation and mobility of heavy metals pose a risk of entering the food chain through water, plants, and animals, raising environmental and public health concerns.

Keywords: tailings dump, heavy metals contamination, ICP-OES, soil pollution, environmental risk.

INTRODUCTION

The mining industry in Kosovo is a key priority for state institutions, as it plays a crucial role in the country's economic development. The Republic of Kosovo has a diverse geological landscape rich in natural resources, particularly energy resources and non-ferrous metals, which have significant potential for economic growth. According to the Mining Strategy of the Republic of Kosovo (2012–2025) [ME, 2012], this sector is a major driver of national progress.

Kosovo has substantial deposits of lead, zinc, silver, gold, chrome, nickel, iron, cobalt, aluminum, copper, and manganese, along with rare metals such as antimony, niobium, lanthanum, cerium, scandium, zirconium, molybdenum, tungsten, indium, and rhenium [ME, 2012; Hyseni et al., 2010; Kelmendi, 2021; Hyseni et al., 2022]. In addition to conserving mineral resources sustainably, it is essential to protect ecosystems and human health. Effective resource management requires collaboration among the mining sector, government institutions, society, and environmental authorities. Furthermore, abandoned mining waste poses significant environmental risks. However, short- and long-term monitoring strategies can help mitigate its impact [Sheremeti et al., 2024; Ndërmarrja et al., 2010]. The environmental threat posed by mining waste dumps in Kosovo is a growing concern. More than 60 million tons of mining waste have accumulated across the country, spread across 11 tailings dumps near various mining sites and flotation concentrators [Hyseni et al., 2022]. The "Kizhnica-Badovc-Hajvalia" Pb-Zn mine, located in the eastern part of Kosovo, is one of the region's most significant sources of lead (Pb) and zinc (Zn) extraction [Mederskia et al., 2022]. This mine, along with its flotation concentrator, has historically been associated with the generation of large volumes of solid mining waste, particularly in the form of tailings, which are often stored in nearby dumps, such as the one in Graçanica (Fig. 1).

This tailings dump covers an area of 47.76 ha and contains approximately 11 Mt of Pb-Zn solid waste from the "Kizhnica-Badovc-Hajvalia" mine's Pb-Zn flotation plant [Sadriu, 2020] (Fig. 2 and 3). The nearest settlements to the Graçanica tailings dump are located just 30 meters to the west and about 150 meters to the southeast. (Fig. 4). These waste deposits pose a potential risk of spreading toxic metals and pollutants into residential and surrounding areas. The primary concerns are the contamination of soil, water, and air due to high concentrations of harmful metals such as lead, zinc, arsenic, manganese, and other associated elements.

While heavy metals are naturally present in varying concentrations in rocks and soils, their average content can be significantly exceeded due to geochemical anomalies or anthropogenic activities [Sheremeti et al., 2024]. Natural processes (e.g., geological weathering, atmospheric precipitation, chemical processes, wave erosion, wind) and human activities (e.g., mining, industrial wastes, urbanization) significantly contribute to the transport of heavy metals into soil and aquatic environments. [Kudjelka et al., 2002; Ali et al., 2022]. Once deposited in the soil, heavy metals can remain for extended periods, continuously contaminating the

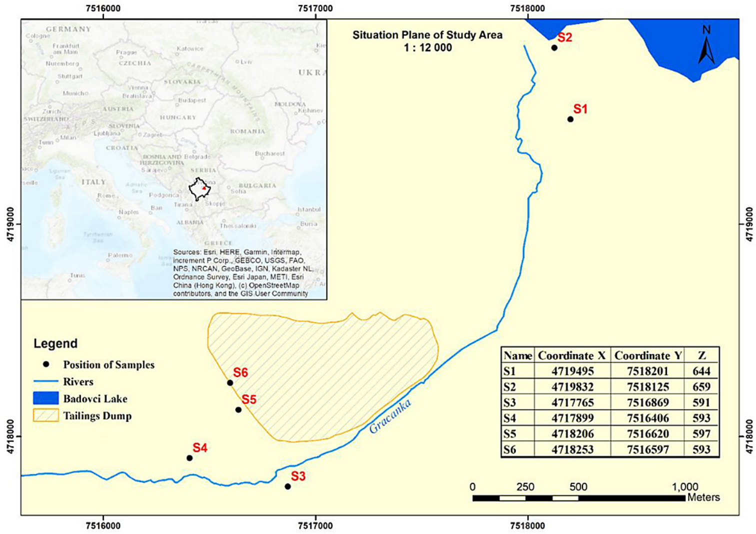

Figure 1. Location of the tailings dump in the Municipality of Graçanica, Kosovo

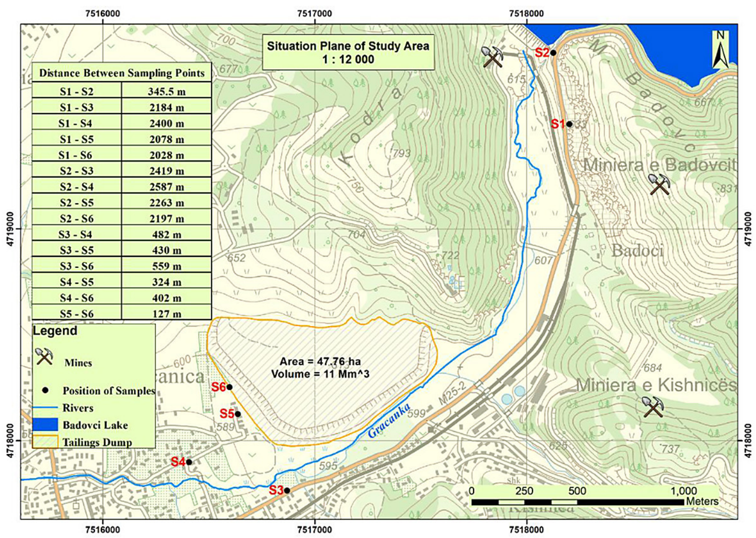

Figure 2. Location of the 47.76 ha tailings dump in Graçanica, with an industrial waste volume of nearly 11 Mt

Figure 3. Tailings dump, without effective containment and treatment measures, the risks of toxic substances leaching into the surrounding environment grow over time (Photo by Argjend Hajra, 2025)

Figure 4. The close proximity of the tailings dump to residential areas contributes to the contamination of water, soil, and air (Photo by A. Hajra, 2025)

environment and posing long-term risks to ecosystems and human health [Bhat et al., 2022]. Soil pH is fundamental to the understanding of soil systems, because it is an indicator of many reactions in the soils. Soil pH controls plant nutrient availability and microbial reaction in soils. In addition to the that, toxicity is also highly dependent on the pH and solubility of the metal, hence the form of metal binding [Anjali and Kumar, 2018]

This paper takes into account the anthropogenic accumulations of heavy metals that were be caused by mining activities at "Kizhnica-Badovc-Hajvalia" mine. It is crucial to investigate the rising concentrations of heavy metals such as lead (Pb), arsenic (As), and manganese (Mn) in both residential areas and near the mining tailings dumps in Gračanica, as well as to identify the correlation between these metals based on pH levels, and their mobility. Monitoring these metals is essential, as their elevated levels pose significant environmental and public health risks, particularly in areas where people reside. Regular assessments will help mitigate potential harm to both the ecosystem and the local population."

MATERIALS AND METHODS

Sample collection and handling

To assess the ecological risk of soil contamination with heavy metals from the tailings dump in Gracanica, a total of 6 samples were collected (Fig. 2) within the framework of the project

"Heavy metals pollution and ecological risk assessment of the Graçanica tailings dump" funded by the Ministry of Education, Science, Technology and Innovation of the Republic of Kosovo and Austrian Development Agency. Two samples, labeled S1 and S2, were collected approximately 1300 meters to the north / northeast of the tailings dump; three samples (S3, S4, and S5) were taken from arable land within settlements to the south and southwest of the tailings dump; and sample S6 was gathered at the edge of the tailings dump, to the southwest. Samples were taken using a soil probe at a depth of 30 cm, weighing about 3 kg per sample. Each sample was taken in the same manner to obtain a more accurate comparison. The sampling points were accurately determined by GPS (Fig. 1). The distance between the sampling locations is shown in (Fig. 2). After collecting samples, to avoid contamination, the samples were sent to Horn & Co. Analytics SH.P.K.-certified laboratory in Prishtina.

Chemical analysis

The prepared samples were analyzed for heavy metals (Pb, As, and Mn) using ICP-OES, in accordance with the DIN EN ISO 11885: 2009-09 [ISO 11885, 2019] standards for Pb, As, and Mn. pH determination followed the DIN EN ISO 10523:2012 standard [ISO 10583, 2012]. The instrument settings and operating conditions were adjusted and managed in accordance with the manufacturers' specifications, under the oversight of the Horn & Co. Analytics SH.P.K. laboratory.

Data analysis

This paper presents statistical evaluations of heavy metal and pH concentrations in soil samples, using both numerical and graphical representations. Changes in concentration in soil samples were analyzed in relation to pH. The Pearson correlation coefficient was calculated to assess the relationship between Pb, As, and Mn concentrations. To assess environmental pollution, the heavy metal concentration values in the soil samples were compared with the maximum permissible limits for contaminated soil set by regulatory bodies, including the Administrative Instruction (GRK - Government of the Republic of Kosovo) [GRK, 2018], EU Directives [EU-Directives, 1986], the World Health Organization (WHO) [Anjali and Kumar, 2018; 19], and WHO/FAO [WHO/FAO, 2001].

RESULTS AND DISCUSSIONS

pH value

Soil pH is often referred to as a "master variable" in soils due to its significant influence on numerous chemical processes. It plays a crucial role in determining the availability of plant nutrients by controlling the chemical forms in which nutrients exist and influencing the chemical reactions they undergo [Anjali and Kumar, 2018]. pH is also a critical factor influencing the mobility of heavy metals, as even small changes in pH can lead to significant variations in metal concentrations [Sheremeti et al., 2024; Krol et al., 2020]. The pH values for all soil samples are shown in Figure 5, with the corresponding statistical data summarized in Table 1.

The results clearly show the pH variation in the samples. In samples S1–S4 and S6, the pH values indicate a lower concentration of H+ ions, ranging from 6.56 to 7.73, which is significantly

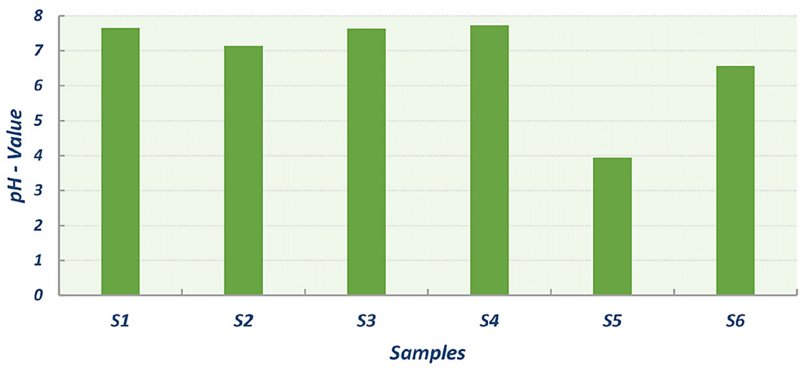

Figure 5. pH value of soil samples

Table 1. Statistical data on pH levels and heavy metal concentrations (mg/kg) in soil samples

Parameter	Ph	Arsenic (As)	Lead (Pb)	Manganese (Mn)
Minimum (mg/kg)	3.95	7.00	69.00	479.00
Maximum (mg/kg)	7.73	1127.00	1691.00	6440.00
Average (mg/kg)	6.78	295.53	632.00	2203.50
Median (mg/kg)	7.39	145.60	399.50	1078.00
Standard deviation (mg/kg)	1.45	425.33	663.46	2344.98
Coefficient of variation (-)	0.21	1.44	1.05	1.06

higher than the pH value of 3.95 in sample S5. This is because the soil's acidity has increased due to redox processes, which have caused the breakdown of minerals, making heavy metal ions become active [Kicińska et al., 2022; Bao et al., 2022]. The coefficient of variation has a low value of 0.2145, indicating that the data have a small dispersion, making them closer to the average value, except for sample S5.

Lead

Lead is classified as a chalcophile element due to its strong affinity for sulfur. The most abundant mineral containing lead is galena, a naturally occurring lead sulfide (PbS). Galena serves as the primary source of lead for production [Sheremeti et al., 2024]. Lead (Pb) is the second most toxic metal and constitutes only 0.002% of Earth's crust [Raj and Prasad, 2023] Although it is naturally present in small amounts, its primary source today comes from human activities, including industrial processes, especially mining, batteries, etc. These human-made sources have significantly contributed to lead pollution, which in turn negatively impacts both human health and the environment. As a result of this pollution, workers in mining industries and residents living near these areas face long-term respiratory diseases due to the constant inhalation of dust particles [Dey et al., 2023], as well as health risks from consuming food grown in lead-contaminated soil.

The variation in lead content across the soil samples reveals a substantial exceedance of the permissible Pb levels. According to the Administrative Instruction (Kosovo) [GRK, 2018], the limit is 200 mg/kg; EU Directives [EU-Directives, 1986] set a range of 50 to 300 mg/kg, while WHO/FAO guidelines [WHO/FAO, 2001] recommend a maximum permissible limit of 50 mg/kg. The highest lead concentration was found in sample S4, with a value of 1691.0 mg/kg (Figure 6). It is noteworthy that the Pb content in samples S1 and S2 is much lower, with a minimum value of 69 mg/kg and a maximum value of 105 mg/kg. These samples were collected approximately 1300 meters north/northeast of the tailings dump.

Sample S6, at the edge of the tailings dump, shows low lead values, 144 mg/kg, which is likely due to the mobility of lead as a result of the oxidative-reductive processes of its minerals [Sheremeti et al., 2024; Zehl, 2005]. A coefficient of variation of 1.0498 (Table 1) for lead indicates a relatively high level of variability in the data set. This variability could be attributed to factors such as differing contamination sources

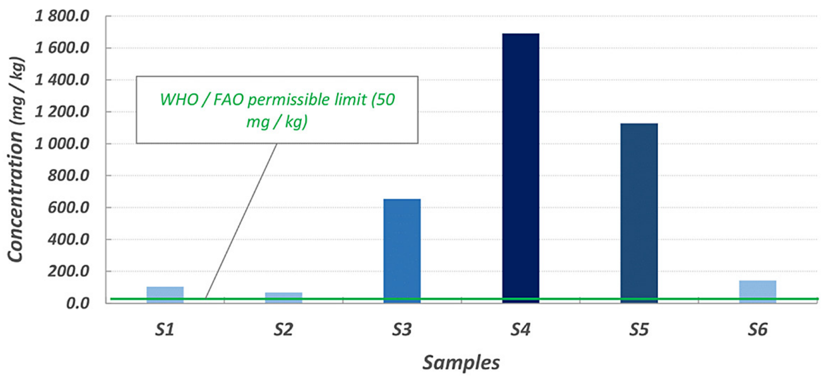

Figure 6. Lead concentration in soil samples

or environmental conditions that influence lead distribution and accumulation, as observed in samples S3, S4 and S5, taken from arable land within settlements.

Figure 7 shows a very weak negative correlation between pH and lead, with a correlation coefficient of -0.19. Higher concentrations of lead are observed in samples S3, S4, and S5, where the concentration of H^+ ions is low, indicating an alkaline environment. However, in sample S5, with a pH of 3.95, lead concentration remains high. Lead is available as a free mobile cation Pb_2^+ with pH value of 5–6 of somewhere 80–90% and at lower values, especially bioavailable in high acidic environment, [Sheremeti et al., 2024; Zehl, 2005].

Manganese

Manganese is the 12th most abundant element in the biosphere, with a concentration in the Earth's crust reaching up to 0.098 mass%. It is widely distributed across soil, sediments, water, and biological materials. It can also negatively impact the ecosystem by accumulating through the food chain [Anjali and Kumar, 2018]. Geochemically, it is a lithophilic element, generally concentrated in rock-forming minerals [Albarède, 2009]. The main sources of high anthropogenic manganese inputs into the environment include the metallurgical industry, battery factories, and industrial plants that emit dust containing manganese [Michalke and Fernsebner, 2014]. Excessive manganese in drinking water can pose potential hazards not only to crop production but also to human health through food consumption, groundwater contamination, and accumulation in food crops [Rahman and Zajm, 2015]. Elevated manganese levels can also have toxic effects on the nervous system, leading to symptoms such as speech impairment, loss of coordination, and, in severe cases, Parkinson's-like [Michalke and Fernsebner, 2014].

The manganese concentration in the soil samples ranges from 479.0 to 6440.0 mg/kg. All six samples, except for sample S5, exceeded the WHO/FAO permissible limit of 500 mg/kg (Fig. 8). A coefficient of variation of 1.0642 (Table 1) for manganese indicates a relatively high level of variability in the data set. Since the coefficient is greater than 1, this suggests that the manganese concentrations are widely dispersed around the mean value. This is due to the sampling being conducted in distant locations and near the tailings dump.

The manganese levels are extremely high in samples S3 and S4, which were taken from arable land within settlements near the tailings dump. In sample S5, due to high acidity, the manganese concentration is below the WHO/FAO permissible limit. According to [Zehl, 2005], increased soil acidity promotes the release of manganese, facilitating its transfer through flowing water (Fig. 9). A correlation coefficient of 0.49 between manganese and pH indicates a moderate positive correlation between the two variables. This suggests that as the pH of the soil increases, the concentration of manganese tends to rise as well. However, the relationship is not very strong, possibly because samples S1 and S2, which were taken farther from the tailings dump, may not be as directly influenced by the same factors affecting manganese concentration near the dump.

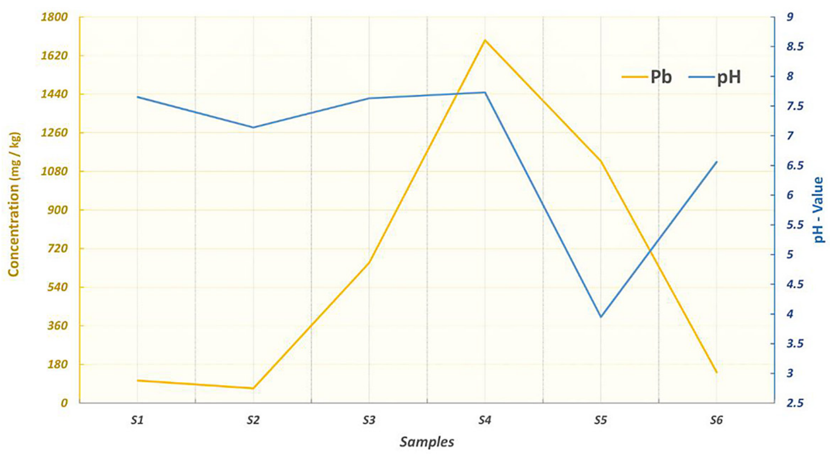

Figure 7. Variation in lead content as a function of pH in soil samples

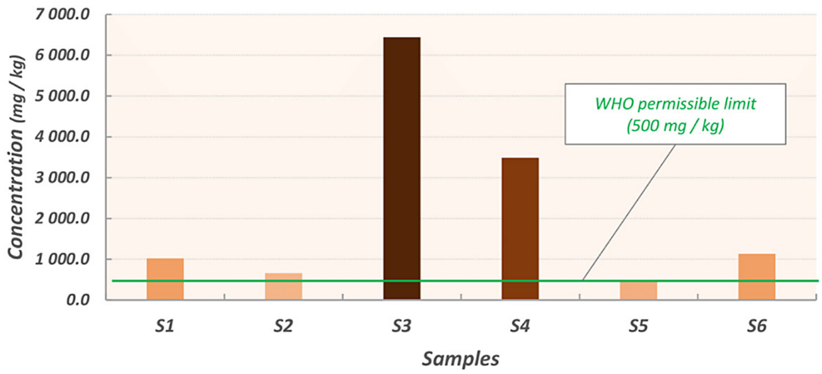

Figure 8. Manganese concentration in soil samples

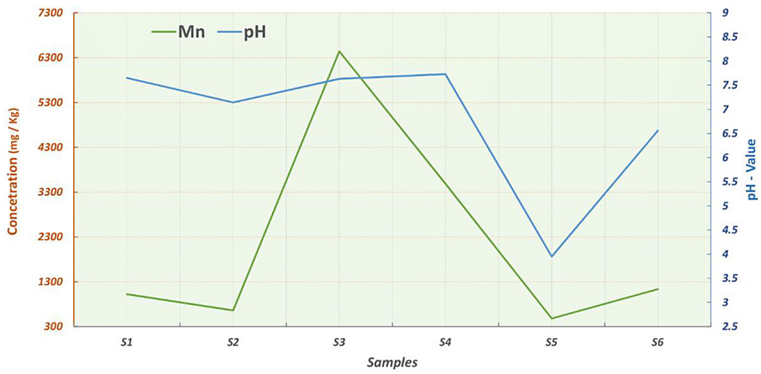

Figure 9. Variation in manganese content as a function of pH in soil samples

Arsenic

Arsenic is considered a heavy metal despite being a metalloid because of its high atomic weight and density, which are characteristics typically associated with heavy metals. Although it has properties of both metals and non-metals, its physical and chemical behaviors, such as toxicity and its tendency to accumulate in the environment, align more closely with those of heavy metals. Arsenic is a naturally occurring metalloid that is widely present in soil, water, food, and the environment. It enters the environment through various human activities, including mining, metal smelting, the burning of fossil fuels, and the use of agricultural pesticides [Fatoki and Badmus, 2022]. Arsenic is a potent neurotoxicant that shows negative effects on biological species [Kaur et al., 2024]. Figure 10 shows that arsenic is present in high concentrations in soil sample S5, located in arable land within settlements, 35 meters from the tailings dump, with a maximum value of 1127.0 mg/kg. In contrast, the minimum value is found in sample S2, located 1425 meters from the tailings dump, with a concentration of 7.0 mg/kg. In all samples, except for S1 and S2, there is a clear exceedance of the permissible arsenic content. According to the Administrative Instruction (Kosovo) [GRK, 2018], the limit is 30 mg/kg, while the WHO/FAO guideline [WHO, 1981] sets the limit at 20 mg/kg.

A coefficient of variation of 1.4392 for arsenic in soil samples (Table 1) indicates a high level of variability in arsenic concentrations, suggesting significant fluctuations in arsenic content. This variability could be influenced by factors such as the distance between sampling locations and the impact of soil pH (Fig. 11). In an acidic medium, arsenic tends to accumulate at higher concentrations because, in low pH conditions, arsenic becomes more soluble and mobile. Depending on the redox conditions and microbial activities, arsenic can occur in soils, surface water, and groundwater in two main forms: pentavalent arsenic (arsenate) and trivalent arsenic (arsenite) [Wiedemann, 2017]. When the soil or water is acidic, arsenic, often present as arsenate, can more easily dissolve and move through the environment. This increased solubility leads to greater bioavailability, making arsenic more readily absorbed by plants or organisms, thus accumulating in higher amounts in the environment [Matheß, 1994].

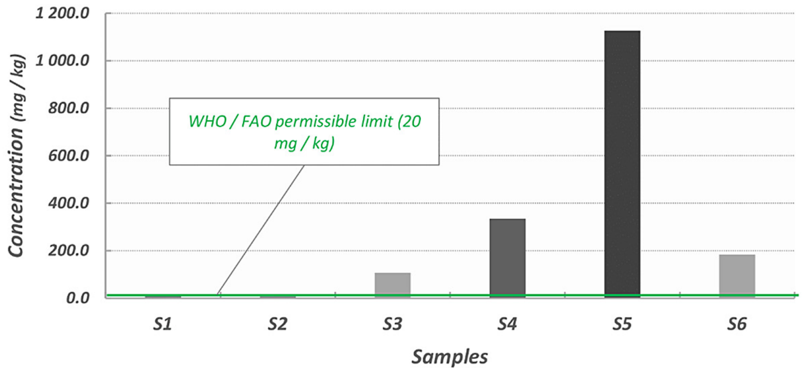

Figure 10. Arsenic concentration in soil samples

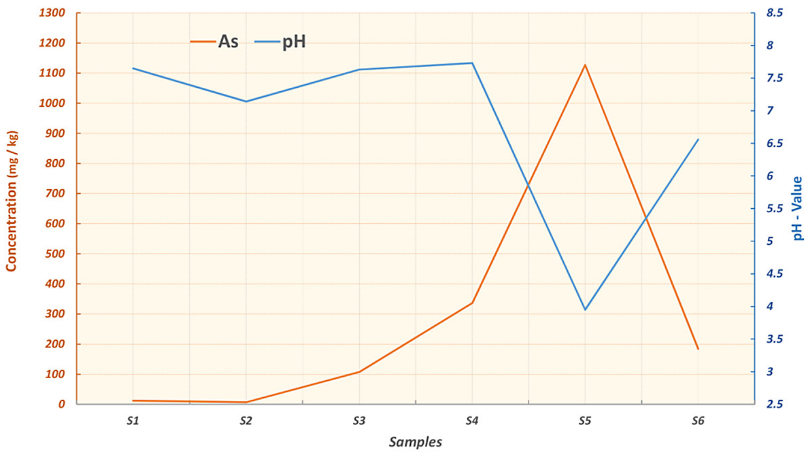

Figure 11. Variation in arsenic content as a function of pH in soil samples

Correlations matrix

The correlation matrix in Table 2 presents the correlation coefficients between arsenic, manganese, and lead.

The correlation coefficient between lead and arsenic in the matrix is 0.57921, indicating a moderate positive correlation. This suggests that as the concentration of arsenic increases, there is a corresponding moderate increase in the concentration of lead. This relationship is particularly evident in the samples near the tailings dump (S3, S4, S5, and S6), which exhibit higher levels of both lead and arsenic. In contrast, samples farther from the tailings dump (S1 and S2) show much lower concentrations of these elements. Additionally, soil pH appears to influence the concentrations of these metals. Arsenic concentrations tend to peak in acidic environments, while lead concentrations are highest in alkaline soils, as demonstrated in Figures 6 and 10. The correlation coefficient value of 0.33622 between lead and manganese in the matrix suggests a weak positive correlation, indicating a slight increase in the concentration of manganese as the concentration of lead increases. However, since the value is closer to 0 than to 1, the relationship between these two variables remains relatively weak. This weak correlation may be attributed to the differing sources of contamination in the samples. Specifically, samples S3, S4, S5, and S6 are primarily influenced by contamination from the tailings dump, while samples S1 and S2 are impacted by the ore field and geological processes. The concentration of manganese is lower in acidic media compared to other samples, indicating higher mobility [Block et al., 2016; Tian et al., 2021]. The correlation coefficient value of -0.25661 between arsenic and manganese in the matrix indicates a

Table 2. Correlation matrix for arsenic, manganese and lead

Parameter	As	Mn	Pb
As	1.00		
Mn	-0.26	1.00	
Pb	0.58	0.34	1.00

weak negative correlation. This suggests that as the concentration of arsenic increases, the concentration of manganese tends to decrease, particularly in acidic media, as observed in sample S5, and in alkaline media, as seen in sample S3 (Figures 8 and 10). Manganese shows high mobility in acidic media [Sheremeti et al., 2024]. The mobility of metals in acidic medium follows this order: Mn > Pb > As. In alkaline medium, the immobilization effect follows this order: Pb > Mn > As, resulting from redox processes, bonding with clay minerals, humic substances, soil solutions, and other factors [Zehl, 2005; Matheß, 1994; Kudjelka et al., 2002; Yang et al., 2021].

Spatial distribution

Based on the data presented in the map, the concentrations of heavy metals (lead – Pb, manganese – Mn, arsenic – As) as well as pH values in soils show significant spatial variations. These changes are mainly influenced by the presence of the tailing dump and by other natural factors.

The area near the waste tailing (S5 and S6) is the most polluted point, especially with high concentrations of As and Pb. Manganese (Mn) has higher concentrations in the south (S3, S4), while As and Pb are more concentrated near the mining waste (S5, S6). Low pH values in the polluted

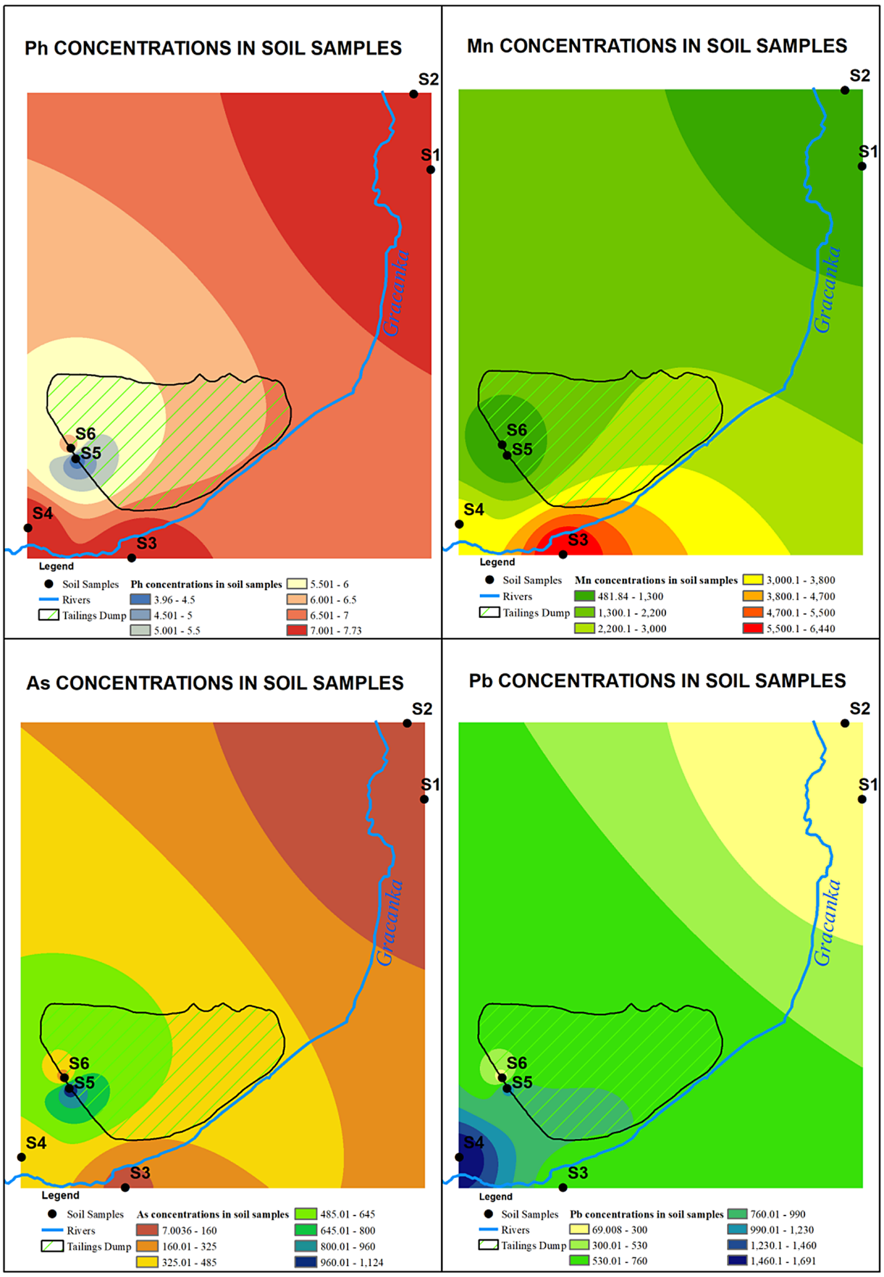

Figure 12. Spatial distribution of metal concentrations in soil samples

areas (S5, S6) may increase the mobility of heavy metals, presenting higher environmental risks. The northern and eastern parts have lower concentrations of heavy metals, indicating a lower impact from pollution sources.

The combination of data suggests that the main source of pollution is the disposal of mining waste, with the greatest impact on arsenic and lead, while for manganese the impact appears more distributed in the south.

The prospects for future research

Future research prospects are promising due to the bio accumulative nature of toxic metals, highlighting the need for government action to reduce their presence in soil and water in the Municipality of Graçanica. Key actions include renovating landfill dumps and preventing further contamination. Ongoing research should focus on the long-term mobility of heavy metals, redox processes, and their transformations in soil-water and soil-plant systems. Additionally, it is crucial to assess contamination in surface and groundwater, conduct hydrogeochemical evaluations of drinking and irrigation water, and analyze ecosystems, including rivers, Badovci Lake, and the surrounding flora and fauna. Medical studies are also needed to evaluate the impact on the local population.

CONCLUSIONS

The statistical evaluations reveal an exceptionally high accumulation of heavy metals in agricultural lands and residential areas within the municipality of Gračanica. Such an environmental ecological assessment of pollution is only possible when considering the permissible limits of heavy metal content in the soil, as outlined by national and international standards, including the Administrative Instruction (Kosovo), EU Directives, and the WHO. The high concentrations of heavy metals, such as lead, manganese, and arsenic, in residential areas near the tailings dump, due to anthropogenic impacts, raise significant concerns. All analyses clearly demonstrate that the metal content exceeds the permissible standard values by a considerable margin, posing a high risk and continuing to cause environmental pollution in the municipality. The mobility and infiltration of heavy metals beyond the tailings dump into nearby ecosystems will increase, influenced by pH levels and geochemical processes that contribute to the further breakdown of minerals, thereby significantly amplifying the ecological risks for both the environment and the residents.

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