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RESEARCHES ON THE MOBILITY AND TOXICITY OF HEAVY METALS FROM BALOMIR TAILINGS DUMP – URICANI JIU VALLEY

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Abstract: *Coal mining is a vital industrial activity but often associated with negative environmental effects, including the accumulation of waste containing heavy metals in tailings dump. These heavy metals, such as lead, chromium, copper, zinc, cadmium, mercury, and others, pose significant risks to the environment and human health. The mobility of these heavy metals is a major concern, as they can be released into soil and water during rain or erosion processes, contaminating water sources and affecting aquatic and terrestrial life. These metals can be taken over by plants and eventually end up in the trophic chain, including in humans. The toxicity of heavy metals is well known, with these substances having harmful effects on human health. In this paper, the main ways of transferring heavy metals into environmental components and their spatial distribution from sterile holes to water and soil environment components. As a case study, the sterile Balomir hole, an inactive hole for about 9 years resulting from the exploitation of the hule from the Uricani mine located in the Jiu Valley.*
Keywords: *contamination, mobility, toxicity, heavy metals, tailings dump, soil*

1. Introduction

Mining activity has had and continues to have a significant impact on the environment, generating one of the most notable problems related to heavy metal contamination, the tailings stored in dumps. These deposits, composed mainly of mineral residues, are the result of the process of coal extraction and processing and may contain significant concentrations of heavy metals such as lead, cadmium, arsenic, zinc, copper, cobalt, chromium, nickel, and others. [1]

In their initial state, these sterile materials contain heavy metals bound in inert mineral structures, which, over time, can become mobile under the influence of environmental factors, erosion, and leakage processes, as well as by interaction with living organisms. This phenomenon is all the more worrying as heavy metals are known for their toxic potential on terrestrial ecosystems and human health.

The mobility and toxicity of heavy metals in soil in sterile holdings are influenced by a number of physical, chemical, and biological factors, and understanding these interactions is essential for managing and reducing the negative impact of heavy metal pollution on the environment. Compared to natural soils, soil formed on sterile hales may present different characteristics and conditions, which can amplify or diminish the impact of heavy metals. [2]

Balomir sterile pool is located in the Western part of the Jiu Valley carboniferous basin. It resulted from the underground mining of coal, respectively, at Uricani mine. The location of the dump is in the valley of Balomir River, in the North-Eastern part of the mine. It is a hill free of technological loads, having been inactive since 2015. Currently, it is in private ownership. It is covered on the taluses with wooden vegetation and in the plateau area with herbaceous vegetation. The average annual temperature falls between 7-9°C, precipitation is between 600 and 800 mm, the predominant direction of the wind is in the South-Western direction. Figure 1 shows the location of Balomir tailings dump in the depression of the Jiu Valley.

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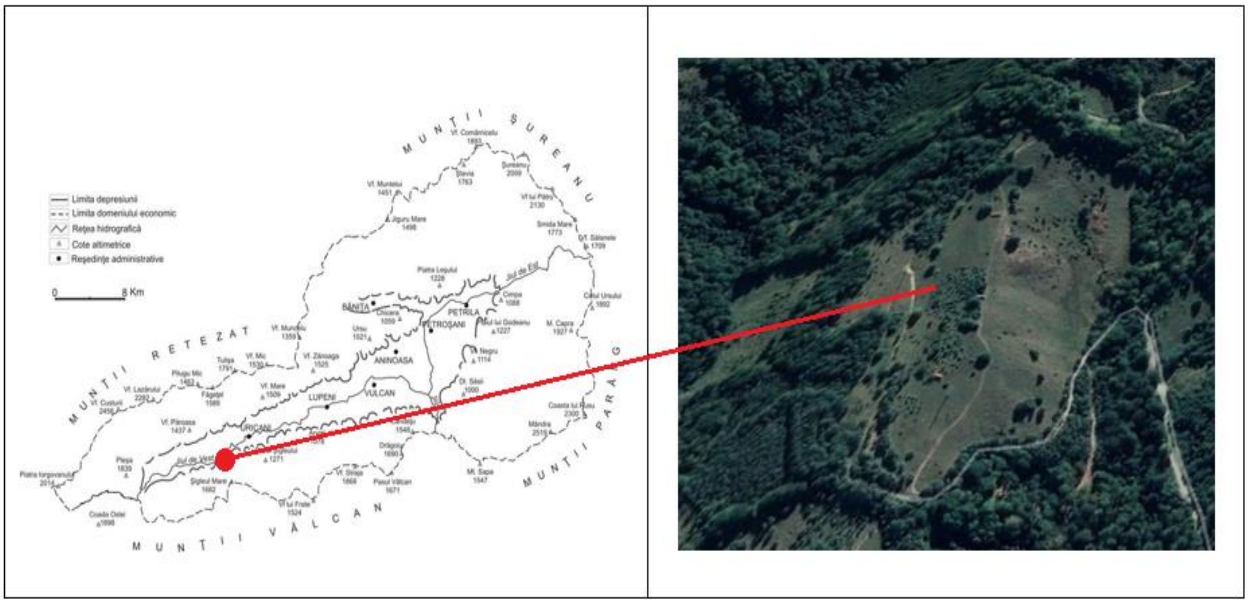

Fig 1. The location of Balomir tailings dump in the coal basin of the Jiu Valley

Factors favouring the mobility and toxicity of the heavy metals in the tailings dump

The main factors influencing the mobility and toxicity of heavy metals in tailings dump soils are:

1. The composition and structure of the material in the tailings dump can influence the retention and mobility of heavy metals. The presence or absence of specific organic or mineral materials can affect the soil's ability to bind heavy metals and make them less mobile.

2. The oxidation and reduction processes present in the tailings dumps are manifested from the moment the material is stored in the dumps and contribute to the intensification of the solubility of heavy metals. These processes are influenced by local environmental conditions such as humidity, temperature, and soil pH.

3. Microbial activity has an essential role through the appearance of microorganisms in the soil and can have an important role in the biogeochemical cycle of heavy metals in tailings dumps. Certain species of microorganisms may be resistant to heavy metals and may have the ability to transform or reduce their toxicity.

4. The degree of soil compaction, an important physical indicator of the soil, can influence the porosity and permeability of the waste material in the landfill, which can affect the transport and retention of heavy metals. Less compacted soils may have a greater capacity to retain heavy metals and make them less mobile.

5. Exposure to meteorological conditions leads to the physical-mechanical disaggregation of waste rock stored in dumps. Being exposed to varied meteorological conditions, such as intense precipitation and cyclic freeze-thaw phenomena, these conditions can have an influence on the processes of erosion, leaching, and mobilization of heavy metals from soil in tailings dumps.

6. Management and remediation have a primary role in reducing the phenomenon of soil contamination in the areas adjacent to these tailings deposits. Applying appropriate management and remediation techniques will significantly reduce the mobility and toxicity of heavy metals in tailings dumps. Among these, some technologies can be mentioned, such as phytoremediation, bioremediation, and phyto-stabilization, which could contribute to changing the behaviour of heavy metals in soil from tailings dumps. [3]

2. Mobility and toxicity of the heavy metals in Balomir tailings dump

From the analyses carried out on the soil sampled from Balomir tailings dump, the presence of heavy metals Cr (total), Cu, and Ni was identified, which exceeded the concentrations of normal values in the soil in most of the sampling points. From the specialized literature, we know that the presence of some heavy metals, even in low concentrations, can have a high toxicity depending on the favouring factors and the physical properties of the deposited material. In this sense, a brief description of the mobility and toxicity potential of the heavy metals identified in the soil samples from Balomir tailings dump will be made.

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2.1. Mobility and toxicity of total chromium

The mobility and toxicity of Cr (total) present in tailings dumps are important issues in managing the risks associated with mining activities. Cr (total) represents the sum of all forms of chromium (trivalent and hexavalent) and can have a significant impact on the environment and public health.

1. **Mobility of total chromium**

Cr (total) present in tailings dumps may become mobile due to processes of erosion, runoff, and infiltration of water from precipitation or through underground seepage processes. The mobility of Cr (total) in the environment depends on several factors, including soil pH, organic matter content, soil texture, and local hydrologic conditions.

2. **Total chromium toxicity**

The toxicity of Cr (total) is mainly determined by the presence of the hexavalent form (Cr(VI)), which is more mobile and more toxic than the trivalent form (Cr(III)). It can negatively affect human health and the environment, being associated with a number of adverse effects, including skin and respiratory irritation, lung damage, gastrointestinal disorders, and even cancer.

3. **Interaction with the environment**

Total chromium, Cr (total), can interact with environmental components such as soil, water, and air. In soil, it can be retained or become mobile, depending on soil properties and environmental conditions. In water, Cr (total) becomes soluble and can reach surface or groundwater, where it can affect biodiversity and water quality.

2.2. Mobility and toxicity of copper

In the industrial and mining environment, the mobility and toxicity of Cu are essential aspects in waste management and the protection of the environment and public health. Although Cu is an essential element for many biological functions, its excess presence in the environment can have significant negative consequences. In particular, in tailings dumps resulting from mining activities, Cu can be mobile and dispersed in the environment, affecting the soil, water and air around mining sites.

1. Mobility of copper

Copper can become mobile in tailings dumps through various processes, such as soil erosion, storm water infiltration, and underground runoff. The mobility of copper in the environment is influenced by soil pH, organic matter content, soil texture and local hydrological conditions.

2. Copper toxicity

Copper is an essential trace element for many organisms, but at high concentrations it can become toxic. The toxicity of copper can vary depending on its chemical form and the organism to which it is exposed. In general, soluble forms of Cu, such as free Cu ions, are more toxic than insoluble forms. Cu can affect a wide range of organisms, including plants, aquatic and terrestrial animals, and soil microorganisms.

3. Interaction with the environment

Cu can interact with environmental components such as soil and water in complex and varied ways. In soil, copper can be retained by solid particles or released into solution, depending on pH and organic matter content. In water, copper can affect water quality and aquatic life, especially in areas of high concentrations.

2.3. Mobility and toxicity of nickel

In light of the growing concerns regarding the impact of mining activities on the environment and public health, the study of the mobility and toxicity of nickel in tailings dumps is a topic of particular interest to the scientific community and to those involved in the management of natural resources. Nickel, a heavy metal naturally present in the earth's crust, becomes a source of concern when it is dispersed into the environment as a result of mining activities. Its mobility in soil and water, as well as its toxicity to the environment and human health, are key issues to consider in the management and remediation process of contaminated sites.

1. Mobility of nickel

The nickel present in tailings heaps resulting from mining activity becomes mobile following the processes of soil erosion, water runoff resulting from precipitation, and groundwater infiltration. The mobility of nickel in the environment is influenced by soil pH, organic matter content, soil texture, and hydrological conditions.

2. Nickel toxicity

Ni is considered a heavy metal that is toxic to many organisms, including humans. The toxicity of Ni can vary depending on its chemical form and concentration. It can adversely affect our human health and cause a variety of ailments, including skin and eye irritation, lung damage, and even cancer.

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3. Interaction with the environment

It can interact with soil, water, and air in different ways depending on local conditions and environmental characteristics. In soil, Ni can be retained or become mobile, depending on soil properties and environmental conditions. Its presence in water can affect water quality and aquatic life, making it toxic to many aquatic organisms. [4]

Figure 2 shows the mobility of heavy metals in soil, water, and air due to environmental factors.

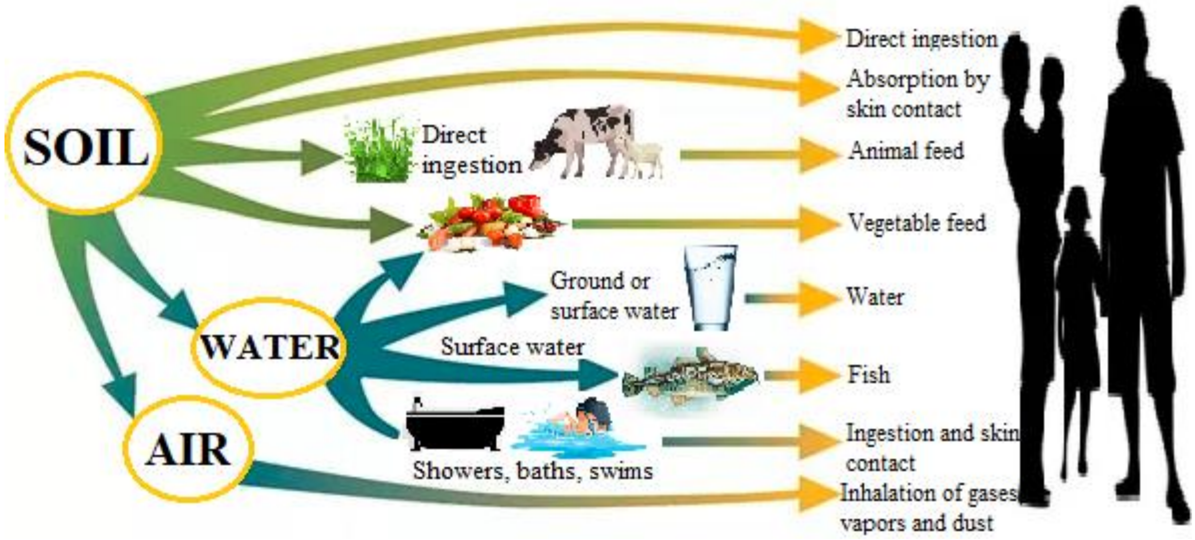

Fig.2. Mobility of heavy metals in environmental factors

3. Analysis of concentration isolines on Balomir tailings dump

In the context of analysing soil contamination from Balomir tailings dump, concentration isolines represent an effective way of visualizing and evaluating the spatial distribution of pollutants and their degree of impact on the environment. Concentration isolines are imaginary lines or isolines that connect points with the same concentration of a substance, namely heavy metals or other pollutants, in the soil. These lines are drawn on maps or charts to represent the spatial distribution of the respective concentrations in a given area. [5]

Concentration isolines are used to highlight areas of higher or lower concentrations of pollutants in soil and to identify areas of high potential for contamination or risks to the environment and human health. They provide a visual and quantifiable way to assess the extent of soil contamination and guide decision-making regarding tailings dump management and remediation. [6]

Analysis of concentration isolines can provide essential information about the extent of contamination in soil and the potential routes of pollutant dispersion in the environment. It can also help identify priority areas for pollution management interventions or the implementation of remedial measures. [7]

Table 1 shows the values determined for heavy metals at the 17 sampling points. These determined values were related to the normal values of the traces of chemical elements in the soil, according to Ord. 756/1997, which in most sampling points exceeded the normal value in the soil. After carrying out a mapping of Balomir tailings dump, we plotted the isolines of the concentrations of heavy metals present in the soil samples. From their graphic representation, it is possible to see the spatial distribution of these heavy metals in the tailings dump, and at the same time, it is possible to determine the transfer path to the environment.

On Balomir landfill, the transfer path is the aquatic environment, as can be seen from figure 3, where the isolines of heavy metal concentrations are drawn. The transport factor is the rainwater that flows from the northern side of the tailings dump to the South-Western side.

Among the analysed indicators, soil pH represents the essential factor favouring the mobility of heavy metals. Mobility refers to the ability of a heavy metal to move through soil and reach other media, such as groundwater or surface water. Soil pH is a measure of the degree of acidity or alkalinity of the soil and can range from acidic (pH < 7) to alkaline (pH > 7).

The soil pH determined from Balomir tailings dump varies from slightly neutral to alkaline. This soil pH affects the ways in which heavy metals are present in the soil. In acidic soils, heavy metals are more soluble because they are present in ionic or complex forms, which make them more mobile and more susceptible to transport. In alkaline soils, heavy metals are tightly bound to soil particles and become less mobile. [8]

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Table 1. Heavy metal concentration and pH determined at the sampling points

Sampling points	Analysed indicators						
	Cr (total)		Cu		Ni		pH
	Determined value (mg/kg d.s)	Normal value (mg/kg d.s)	Determined value (mg/kg d.s)	Normal value (mg/kg d.s)	Determined value (mg/kg d.s)	Normal value (mg/kg d.s)	
P1	26.51	30	47.59	20	57.68	20	8.3
P2	35.91	30	56.46	20	101.42	20	8.1
P3	35.85	30	56.20	20	90.62	20	7.0
P4	48.55	30	72.40	20	107.55	20	6.7
P5	46.77	30	74.69	20	108.84	20	6.4
P6	32.40	30	54.93	20	70.93	20	7.1
P7	24.44	30	49.81	20	57.36	20	8.2
P8	33.43	30	61.50	20	104.33	20	7.0
P9	49.76	30	75.09	20	109.37	20	7.4
P10	31.31	30	56.20	20	89.00	20	6.6
P11	23,94	30	51.85	20	72.78	20	7.3
P12	44.46	30	50.97	20	102.70	20	6.9
P13	29,47	30	48.98	20	58.72	20	8.1
P14	31.00	30	53.18	20	73.42	20	7.5
P15	35.60	30	58.51	20	81.74	20	7.8
P16	33.40	30	56.12	20	72.22	20	7.7
P17	35.73	30	56.09	20	81.49	20	7.2

Metal ion complexation is influenced by soil pH and can influence metal ion speciation. Thus, in acidic soils, metal ions form soluble complexes, which make them more mobile and can migrate more easily into groundwater or surface water. On the other hand, in alkaline soils, they form insoluble or sedimentable complexes that contribute to the reduction of metal mobility.

Adsorption of heavy metals on soil particles is also influenced by soil pH, which can increase or decrease its capacity to adsorb heavy metals. In soils with a low pH, the soil particles may have a higher positive electrical charge, which favours the adsorption of metal ions. In contrast, in alkaline soils, adsorption may be less, which may lead to higher mobility of heavy metals. [9]

Figure 3 shows the mobility of the analysed heavy metals, Cr (total), Cu, and Ni, as well as the chemical compounds that are formed depending on the pH of the soil.

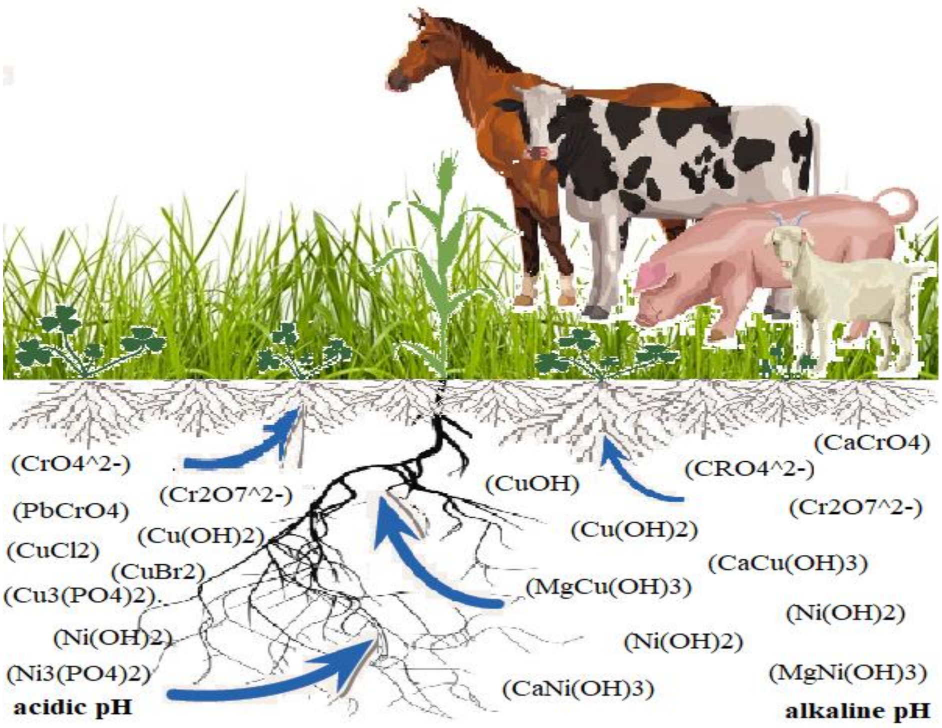

Fig.3 Mobility of chemical compounds of heavy metals as a function of soil pH

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Soil pH can influence the behaviour and bioavailability of heavy metal compounds such as chromium (Cr), copper (Cu), and nickel (Ni) in several ways.

pH affects the solubility of heavy metal compounds in soil. In general, in soils with an acidic pH, the solubility of these metals can be increased, and in soils with an alkaline pH, the solubility can be reduced. For example, Cr and Cu compounds may be more soluble in soils with an acidic pH, while Ni may be more soluble in soils with an alkaline pH.

pH can influence the chemical speciation of heavy metals in soil. Cr can exist as Cr (III) or Cr (VI), and pH can affect the balance between these forms. Cr and Ni can also be present in different ionic forms depending on pH, which can influence their mobility and toxicity in soil.

pH can affect the interactions of heavy metals with solid soil components such as clay and organic matter. For example, in acidic soils, clay and organic matter may have a greater negative charge, which may favour the adsorption of heavy metals and reduce their mobility and bioavailability.

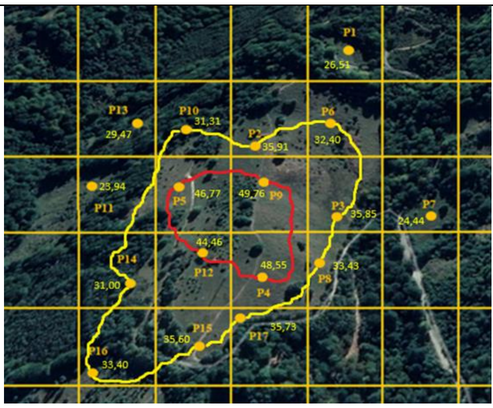
pH can influence the toxicity of heavy metals to plants and other organisms. Although pH itself does not alter the intrinsic properties of heavy metals, it can affect their availability and chemical form, which can influence their degree of toxicity. An acidic pH can increase the toxicity of Cr and Cu compounds to plants, while an alkaline pH can make Ni more toxic. [10]

Heavy metals can be present in soil as soluble or insoluble compounds. Depending on their solubility and soil chemical conditions (such as pH), these metals can be taken up by plant roots.

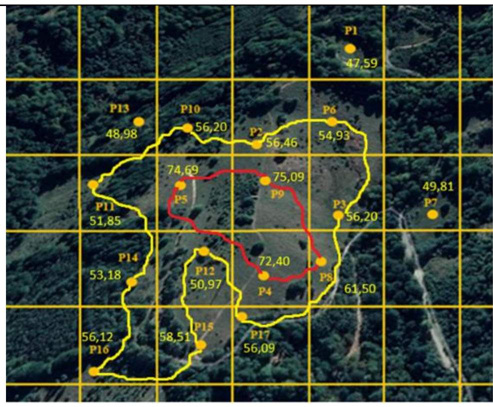
Plants can absorb heavy metals from the soil through their roots. This uptake can vary with plant species, soil pH, organic matter content, and other soil characteristics. Heavy metals absorbed by plants can accumulate in different parts of the plant, such as leaves, stems, or fruit.

Herbivores, insects, or other invertebrates can accumulate heavy metals by consuming contaminated plants. These heavy metals can accumulate in the tissues of plant-eating organisms and be transferred further up the food chain. [11]

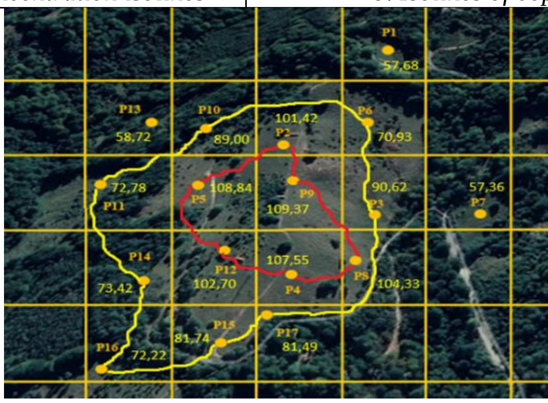
Organisms that feed on contaminated primary consumers can also accumulate heavy metals in their tissues. This can lead to the bioaccumulation and bio-magnification of heavy metals as we move up the food chain, meaning that heavy metal concentrations can increase as we move from lower to higher levels of the food chain.



a. Total chromium concentration isolines



b. Isolines of copper concentrations



c. Isolines of nickel concentrations

Fig.4 Concentration isolines of heavy metals Cr (total), Cu and Ni on Balomir tailings dump

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Organisms higher in the food chain, such as carnivorous mammals or birds of prey, can be exposed to high concentrations of heavy metals by consuming contaminated lower organisms. These heavy metals can have toxic effects on the health of these organisms, including the nervous system, reproductive system, and other organs. [12]

Factors such as time, soil type, climate, and precipitation regime are critical in determining how heavy metals are dispersed and accumulated in the environment. Soils, in particular, play a key role in the retention and dispersion of heavy metals, thus influencing the degree of pollution and the risks to the ecosystem and human health.

Figure 4 (a, b, and c) shows the concentration isolines of heavy metals Cr (total), Cu, and Ni determined in Balomir tailings dump. As can be appreciated in figure 4 (a, b, and c) in all three images, the lack of vegetation contributes to the accumulation and higher concentration of heavy metals Cr (total), Cu, and Ni in the soil of Balomir tailings dump. It is also observed that in the sampling points where the pH is slightly acidic to neutral, the concentrations of heavy metals are higher. Conversely, at points where the pH ranges from neutral to slightly alkaline, adsorption is less, which indicates a greater mobility of heavy metals in the soil. Another factor that contributed to an increase in the concentration of heavy metals in the soil sampled from Balomir tailings dump was the oxidation process.

Over time, these inactive dumps create favourable conditions for the release, mobilization, and accumulation of heavy metals in the soil. From the chemical analyses carried out, it is observed that where there is tree vegetation, the heavy metals Cr (total), Cu, and Ni are in lower concentrations but exceed the normal value allowed in the soil.

The variation in the concentration of heavy metals in the soil can provide information about the degree of contamination and its extent in a certain area. The greater the variation, the greater the potential for exposure to high levels of heavy metals, which can adversely affect the health of ecosystems and human beings. [13]

Knowing the variation in heavy metal concentration in the soil on the tailings dump is essential for planning and implementing phytoremediation measures. This can help identify areas with greater contamination problems and prioritize their phytoremediation. From tracing the concentration isolines of the heavy metals present in the tailings dump, two areas of different concentrations were identified: the first area is that of very high concentrations marked in red in Fig. 4, and the second area has high concentrations marked in yellow in Fig. 4. Both areas of these isolines exceed the normal values of the concentration of heavy metals in the soil, but in the area marked in red, the concentration of heavy metals exceeds Cr (total) 1.57 times, Cu 3.54 times, and Ni 5.28 times the normal value allowed in the soil.

The calculation of the variation in the concentration of heavy metals in soil can be used to monitor soil quality over time and to evaluate the effectiveness of management and remedial actions. By comparing the variation over different time periods or in different areas, trends and changes in the level of contamination can be highlighted. [14]

To calculate the variation in the concentration of heavy metals in the soil, the variation percentage is used, which was calculated with formula (1), and the obtained results can be found in table 2.

$$\%\Delta C = \frac{C_{\max} - C_{min}}{C_{med}}\ x\ 100\% \,, \tag{1}$$

where:

$\%\Delta C$ - percentage variation of heavy metal concentration in soil;
C_{max} - the highest heavy metal concentration in soil;
C_{min} - the lowest heavy metal concentration in soil;
C_{med} - average heavy metal concentration in soil [15]

Table 2. Calculation of the variation of the concentration of heavy metals in Balomir tailings dump

Analyzed indicators	**Percentage of determined values**
Cr (total)	11,18
	14,50
Cu	19,16
	13,69
Ni	7,52
	24,91

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To calculate the variation of the heavy metal concentration in Balomir tailings dump, the two representations of the concentration isolines for each analysed heavy metal were taken into account, as shown in Figure 4 (a, b, and c).

So, following the determination of the variation in the concentration of the analysed heavy metals, it turned out that the variation percentage of Cr (total) compared to the average concentration is 11.18% and 14.50%, respectively; the variation percentage of Cu is 19.16% and 13.69% compared to the average concentration; and the variation percentage of Ni compared to the average concentration is 7.52% and 24.91%, respectively. These results tell us, in percentage (%), that the higher the concentration of heavy metals at the isolines compared to the average concentration in the sample.

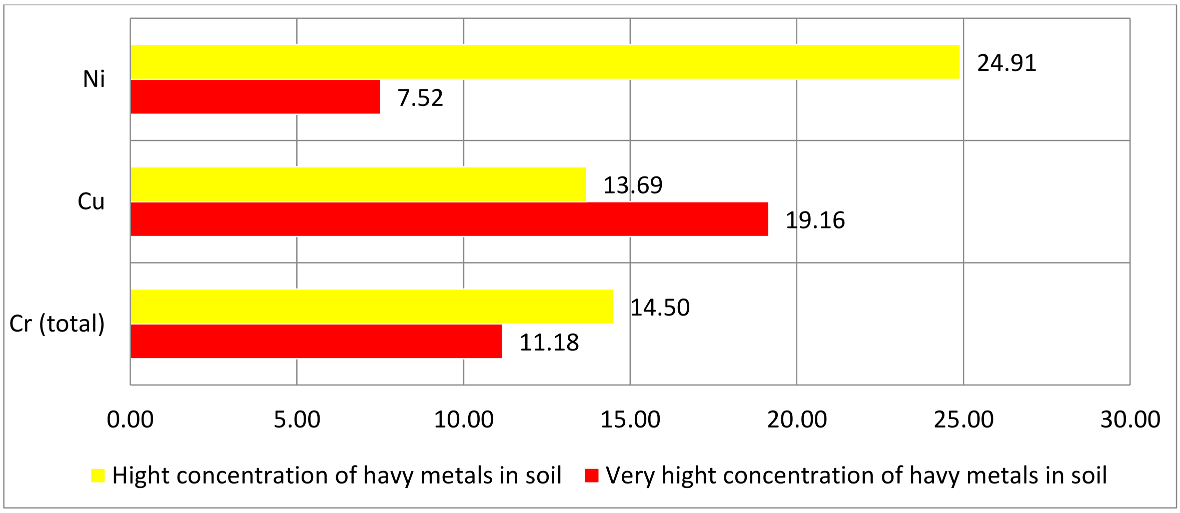

Fig.5 Percentage variation of heavy metal concentration in soil

Using the method of concentration isolines, the very large amounts of heavy metals in the body of the tailings dump and the high concentrations of heavy metals that exceed their normal concentration in the soil were determined.

Figure 5 shows the percentage variation of the three heavy metals, Cr (total), Cu, and Ni, from which it can be seen that the percentage of the very high concentration is Cu, and for the high concentrations of heavy metals analysed, Ni has the highest percentage.

5. Conclusions

Research on the mobility and toxicity of heavy metals in soils at Balomir tailings dump, resulting from coal mining activity, has revealed a serious soil pollution problem.

- Thus, it is found that the allowed concentrations for chromium (total), copper, and nickel in all 17 sampling points exceed 1.15, 2.9, and 4.23 times the normal values allowed in soil.

- Heavy metals can have negative consequences on the environment and human health, as they are toxic and can have migration paths from the soil to underground and surface waters, affecting terrestrial and aquatic ecosystems with a high potential for bioaccumulation in the food chain.

- The mobility of heavy metals in the soil on Balomir waste dump and their accumulation are favored by the lack of vegetation, the pH value of the waste material, and the oxidation processes that take place in the dump and on its surface.

The reduction of heavy metal pollution in the soil produced by Balomir tailings dumps will require immediate phytoremediation measures. These measures will include continuous monitoring of soil and surface water quality, improvement of mining waste management practices, and continuous monitoring of vegetation development for soil stabilization.

By applying phytoremediation technologies to soils polluted with heavy metals, the degree of contamination of natural ecosystems in the vicinity of these tailings dumps will be reduced.

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