

EVALUATION OF SURFACE WATER QUALITY IN THE BĂIȚA MINING AREA

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Abstract: The concentrations of heavy metals and the spatial distribution of Na, Ca, Cu, Zn, Ni, Co, Mg, Fe, Mn, Pb, and Cd in the waters of the Băița area were studied to assess the quality class of surface waters and identify pollution sources. The study area is located along the Băița River and its tributary, Valea Roșie, which are influenced by the Câmpurele mining gallery and mining waste dumps, used for extracting common metal ore deposits. Sampling points were selected along the adjacent path of mine water discharge from mining operations. The metal content in the analyzed surface water was compared with the standard limits for surface waters, in accordance with the five quality classes established by Normative 161/2006. The concentrations of heavy metals in the mine water samples discharged from mining pollution sources were compared with the permissible pollutant loading limits for wastewater discharge into natural receptors, as per NTPA-001/2002 regulations. The spatial distribution of heavy metal concentrations highlights mining activities as the primary source of pollution in the Băița River. The concentrations of Cu, Zn, Ni, Co, Mg, Fe, Mn, Pb, and Cd exceed the specific limits for quality classes I and II of surface waters. Excessive levels of Fe (ranging from 30 mg/L to 105 mg/L) and Mn (ranging from 11 mg/L to 28 mg/L) were recorded in the tributary (Valea Roșie) of the Băița River, associated with mine water discharge, as well as water from the mining waste dump. The exceedance of maximum allowable concentrations for Ni, Co, Pb, Cu, Cd, and Zn in the Băița River and its main tributary, Valea Roșie, leads to the classification of surface water into lower quality classes III and V. Meanwhile, sodium and calcium concentrations fall within the permissible limits for quality class I. The single-factor pollution index reflects surface water pollution levels ranging from light to severe pollution.

Keywords: Water quality, pollution sources, heavy metal, mining gallery, mining dump, Băița.

1. INTRODUCTION

Mining activity is the main source of pollution affecting surface water quality (Basri et al., 2022; Moldovan et al., 2021). Rivers in mining areas act as transport pathways for pollutants over long distances (Howladar et al., 2018). For this reason, integrated studies are necessary to assess both pollution sources associated with mining activities and the effects of pollutants on environmental factors (Liu et al., 2020). Local pollutants in rivers from mining areas can be mobilized and redistributed within fluvial systems through flooding events (Ciszewski & Grygar, 2016). Cordoș et al. (2003) conducted a study on the pollution levels of the Lăpuș and Someș Rivers following the

tailings dam accident in Baia Mare in 2000 (Macklin et al., 2003).

By definition, acid mine drainage is the result of metal solubilization caused by mining activities through the oxidation of sulphide minerals in the ore, leading to the formation of sulfuric acid. This reaction occurs when exposed ore comes into contact with oxygen and water (Lin et al., 2007). The primary mineral contributing to acid mine drainage formation during oxidation is pyrite, which releases sulfuric acid and heavy metals (Nzulu et al., 2024). As a result, effluents from metal processing facilities, tailings pond discharges, and waste dumps have very low pH levels, which allows metals to be transported in their most soluble form along with the water mass (Sheoran &

Sheoran, 2006). Pollutants specific to acid mine drainage formation areas, both in terms of their dangerous specific elements content (toxic heavy metals) and the secondary products formed through interactions with external environmental factors, have become significantly more mobile (Van der Perk, 2014). Due to their acidity and their role in increasing the concentration of suspended solids, these pollutants contribute to the mobilization of toxic metals, posing a threat to aquatic ecosystems (Ochieng et al., 2010).

The assessment of water resource pollution provides an understanding of the negative impact of mining on the quality of surface waters (Cesar Minga et al., 2023).

The purpose of this study is to assess the water quality in the hydrological system of the Băița River along the route adjacent to the mining area by classifying it into quality categories and analyzing the spatial distribution of toxic metals: Fe, Mg, Mn, Co, Ni, Pb, Zn, Cu, and Cd. The negative impact on surface waters and surrounding lands is primarily caused by the inadequate treatment of water discharged from mining galleries. In the studied area, water discharged from the Câmpurele mine is treated with lime powder in a plastic tank until a pH value of 11 is reached.

This pH level is maintained until the water passes through the Câmpurele mining waste dump. This process takes place in the presence of a nearby wastewater treatment plant, located between the gallery and the waste dump, which has been non-functional or under retrofitting for many years.

Heavy metal pollutants affect the quality of surface waters to varying degrees along the investigated route. The spatial distribution of the analyzed heavy metals is closely associated with nearby pollution sources. To determine the level of river water pollution, the pollution index was calculated in accordance with Jiao et al. (2023).

2. STUDY AREA

The locality of Băița (Figure 1) belongs to the town of Tăuții - Măgherauș, which is located in the northwestern part of Maramureș County, in the immediate vicinity of the county seat, Baia Mare. The localities of Băița and Nistru are situated in the southern part of the Igriș Massif. To the north, Băița is bordered by Pietroasa Peak (1,200 m), to the south by the Someș River, to the east by the Borcut Valley, and to the west by the Nistru Valley.

Geologically, the Baia Mare area in northwestern Romania represents a complex segment of the volcanic chain in the Eastern Carpathians. The calc-alkaline volcanism and subvolcanic intrusions date from 13.4 to 6.9 Ma and are partially

contemporaneous with the deposition of Upper Badenian and Pannonian sedimentary rocks (Pécskay et al., 1997, 2006; Seghedi et al., 2004). A large number of ore deposits are associated with this magmatic event.

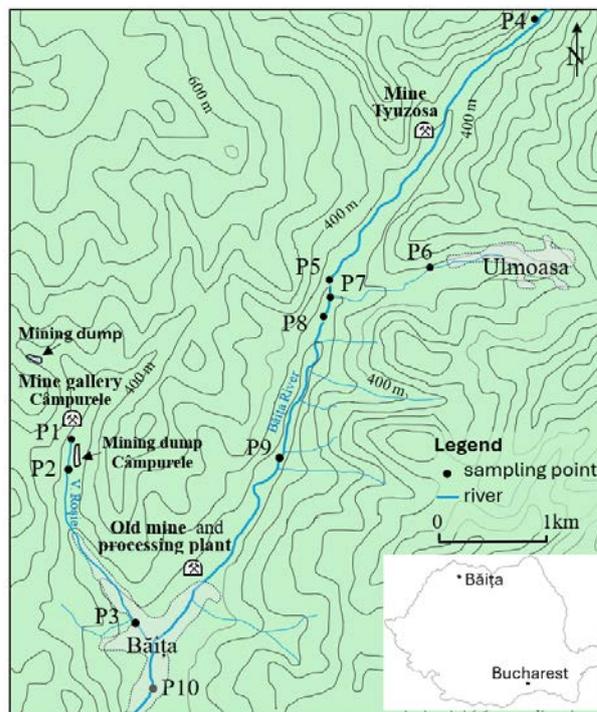


Figure 1. The hydrological basin of the Băița River (the path of acid water from the Câmpurele gallery along Valea Roșie to the confluence with the Băița River).

Several studies (Bailly et al., 1998; Grancea et al., 2002) have identified five mineralization stages in the Baia Mare area, characterized by the deposition of phases rich in: (1) Fe; (2) Cu-(Bi)-W; (3) Pb-Zn; (4) Sb; and (5) Au-Ag.

The gangue minerals include quartz-illite/muscovite (sericite)-feldspar potassium (adularia) in Au-Ag systems and quartz-calcite-rhodochrosite-rhodonite in Pb and Zn mineralizations. Within the Neogene hydrothermal polymetallic Pb-Zn-Cu-Au-Ag mineralizations of the Gutâi Mountains, the Nistru-Băița deposit forms a significant vein field located in an extensive development area of Neogene volcanics, represented by Sarmatian pyroxene andesites and Pannonian quartz andesites (Borcoș et al., 1974). The "9 Mai" and "11 Iunie" galleries in the Nistru area, as well as the "Câmpurele" gallery in the Băița area, were the sites of mining activities and have represented the main sources of significant toxic pollutant release into surface waters. Currently, without any prior treatment, intensely yellow-brown colored water, rich in Fe, flows from the closed "9 Mai" and "11 Iunie" galleries, which are directly connected to the "Câmpurele" gallery in Băița. This

contaminated water discharges directly into the Valea Roşie stream. From there, the water flows into the Băiţa River, then into the Someş River, and eventually reaches the Tisa River (Macklin et al., 2003). The hydrographically studied area is part of the Someş River basin (Damian et al., 2022).

The Băiţa River collects water from the most important valleys, namely the Nistru Valley and the Băiţa Valley, along with their tributaries.

In the studied mining area, the Băiţa River receives water from the following tributaries: Colbu, Limpedeia, Ulmoasa, Coseiu, Valea Roşie, etc. The course of the Băiţa River is well-defined until it reaches the Someş plain, where it exhibits numerous meanders, with steep slopes of the thalwegs in the upper course, and very gentle slopes in the plain. The valleys are supplied by a mixed, pluvio-nival regime, with precipitation infiltrating into the groundwater, depending on the micro and macro-relief as well as the lithological formation. The water can reach significant depths on the upper terraces (up to 10 m), while at the base of the slopes, it is found at shallower depths (2.5 m).

Mine water that is discharged into natural waters is also associated with water that ex-filtrates from dumps located downstream of the mine's entrance and represents a significant source of pollution for the environment (Mohapatra & Kirpalani, 2017). In addition to aquatic environmental pollution, in areas where mining activity intersects with settlements, contaminants also affect human activity (Minga et al., 2023).

The mining industry, through the exploitation of polymetallic ores, has negatively affected the quality of surface waters, even in areas distant from the mining operations. The Băiţa area is a clear example of the interference between mine water discharged from closed, inactive mines and flowing waters that drain the natural environment, extending to inhabited areas

(Figure 2a-b).

When it comes to managing mine waters, there have been rising concern regarding the negative impact they may have on environmental factors, especially the aquatic environment, with potential effects on human health (Singovszka et al., 2020).

Water from mining galleries located in areas closest to populated or agricultural zones, or surface waters, generates environmental risks due to the short migration paths (Modoi et al., 2014). Associated with these pollution sources, tailings ponds also play an important role, posing a long-term threat to adjacent hydrologic systems (Macklin et al., 2003).

The western part of the mining area is heavily crossed by watercourses, which, after the closure of the mines, became the discharge site for mining waters. The negative impact of mining activities on surface waters is the drainage of mine water, which originates from the Câmpurele gallery (Figure 3a).

The Câmpurele gallery drains all the water collected from the abandoned mines in the Nistru area. Associated with the water released from the mining gallery in the Băiţa area is the immense Câmpurele mining dump, which is in direct contact in the lower part with the water draining from the mining gallery.

The Câmpurele mining dump (approximately 0.3 ha) is located on the left bank of the Valea Roşie stream and was formed by the deposition of materials resulting from geological exploration works at the Nistru deposit during the years 1970-1995. The mining dump shows poor establishment of natural vegetation in the upper part, and even less in the sloped area (with an incline of approximately 40°, Figure 3b).

Therefore, the Băiţa River and its adjacent valleys have become sites with high contamination potential. River pollution has not decreased over time (Figure 4).



Figure 2. Images of the Băiţa River in the bridge areas of the Băiţa locality: a) view from upstream; b) view from downstream.



Figure 3. Images of the main pollution sources of the Băița River (tributary Valea Roșie): a) Câmpurele mining gallery; b) Câmpurele mine waste dump.

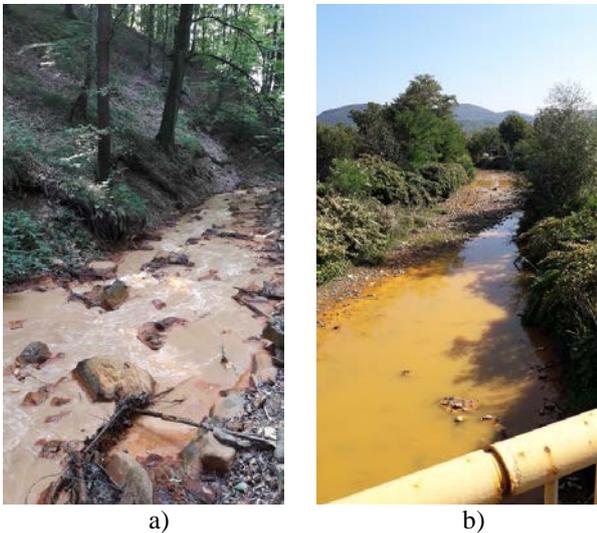


Figure 4. Images of pollution in the Băița River: a) mine water drainage on Valea Roșie, a tributary of the Băița River; b) the Băița River on its way to the confluence with the Lăpușel River.

Based on concentrations of several heavy metals, Sur et al. (2022) showed surface waters get contaminated over time while pH varies significantly. Recent data on the pollution level of sediments in the Săsar River basin, which crosses the city of Baia Mare, were highlighted through a complex geochemical study by Bereș et al. (2024).

3. MATERIALS AND METHODS

3.1. Pollution sources

The investigated water samples were collected from the main pollution sources identified in the Băița area. The most important sources of acid water pollution in the Băița River, as well as in the Valea

Roșie stream, are the mining waste dumps that extend over tens of meters in length and height, and the well-known abandoned mine entrances (Figure 5a-b).

The Câmpurele mining dump, shown in Figure 5a, is located on the Valea Roșie stream. Water that comes from precipitation and percolates through the mine dump is discharged into the Valea Roșie stream and then reaches the Băița River. It should be noted that along this stream, similar or even larger waste heaps can be found.

Another significant source of pollution is the abandoned mine galleries (mine entrances) (Iepure & Pop, 2025). In recent years, efforts have been made to seal them with rubble or, in some cases, iron grilles to prevent scrap metal theft from these sites.

3.2. Water sample collection

To determine the chemical properties of the water in the Băița area, 10 water samples were collected. In establishing the monitoring points, areas with visible intense pollution, the course of the Băița River, areas influenced by tributaries and human activities were considered.

The numbering of the sampling points was chosen as follows: 1 - 3 for the actively polluted area with mine water, and 4 - 10 for the Băița River course, from upstream to downstream. The sampling points are positioned along the path of the acid mine water from the Câmpurele gallery (P1, P2, P3) on the Valea Roșie, up to the confluence with the Băița River, and along its course throughout the entire Băița mining area: P4, P5, P6, P7, P8, P9 and P10 (Figure 1).

The collection of the 10 water samples was carried out as follows:

- the samples were collected under sterile conditions, ensuring no contamination between samples;
- the collected samples were stored in sterile plastic bottles;
- each sample was labelled with the number and location of the sampling point.

The samples were collected at the following sampling points:

Point 1 (P1) – sample collection from the Câmpurele gallery, before the mine water enters the treatment plant on the Valea Roșie stream course (Figure 6a).

Point 2 (P2) – sample collection after the treatment-plant, after the water has been discharged from the treatment plant into the Valea Roșie stream.

This sample was taken to observe the effectiveness of the treatment plant. The sampling points P1 and P2 are significant for the identification of the main pollution source.

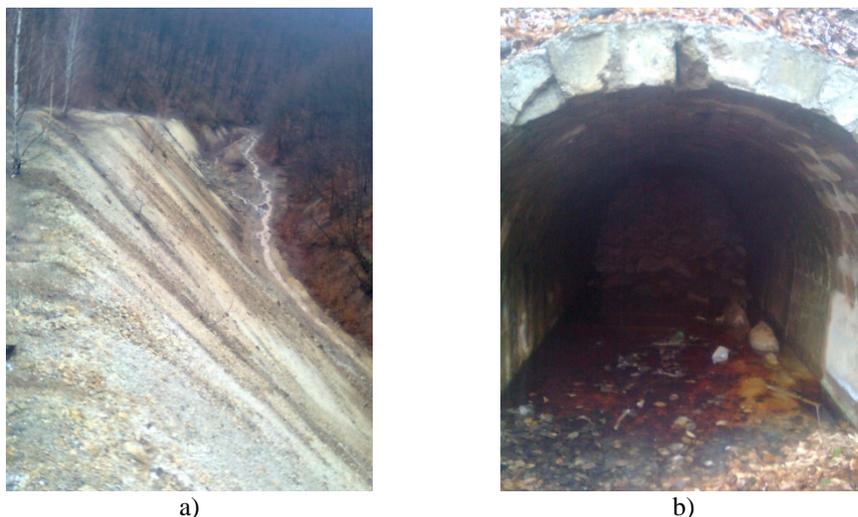


Figure 5. Pollution sources of the rivers: a) Câmpurele mine waste dump; b) abandoned mine gallery.

Point 3 (P3) – sample collection before the confluence of the Valea Roșie stream with the Băița River, to observe changes in the water caused by the population living on both sides of this valley.

Point 4 (P4) – this point corresponds to the upper course of the Băița River at the confluence of the Bacnău and Valea Mare rivers, where a water intake basin for a micro hydro power plant has recently been constructed (Figure 6b).

Point 5 (P5) – the sample was taken before the confluence of the Ulmoasa and Băița rivers. This sampling point serves two purposes: a) to assess the pollution occurring before this confluence, and b) to evaluate the impact of the village of Lomaș, which is located approximately 2 km upstream from the sampling point, along with the nearby mine entrances (Figure 6c).

Point 6 (P6) – the sample was taken from the Ulmoasa River before it reaches the village of Ulmoasa, in order to determine the pollution caused by the village's inhabitants (Figure 6d).

Point 7 (P7) – the sample was collected after the confluence of the Ulmoasa and Băița rivers. This point was chosen to monitor the accumulation of pollutants from both rivers and to assess water quality before it passes by a livestock farm located on the left bank of the Băița River (Figure 6e).

Point 8 (P8) – the objective of this sampling point is to observe potential changes in water composition due to the livestock farm, as well as to evaluate water quality before its discharge from the micro hydro power plant (Figure 6f).

Point 9 (P9) – this sample was collected after the water was discharged from the micro hydro power plant into the Băița River and before the river entered the village of Băița, in order to determine the water quality before the impact of the village's population.

Point 10 (P10) – this final sample is located

after the confluence of the Băița and Valea Roșie rivers, which also corresponds to the center of the village of Băița. At this point, all pollution sources from the entire area converge, making the results of this sample an approximate indicator of what is discharged into the Someș River from the mining area (village) of Băița, without considering other settlements (Figure 7).

3.3. Determination of metals content and water pH

The chemical characteristics (pH, heavy metals) of the water samples are essential for assessing river water quality (Normative 161/2006) and for evaluating mine water compliance with pollutant limits before discharged into natural receptors (HG 188/2002).

3.3.1 Determination of heavy metals content

The determination of metals content in water was carried out using atomic absorption spectrometry (AAS) with a Perkin Elmer Analyst 800 spectrometer (Shelton, USA) equipped with a flame and graphite furnace. The analyses were performed in a flame by burning acetylene in air.

The standards were prepared using analytical-grade reagents from Merck, and the water used was obtained with the Barnstead Easypure RoDi ultrapure water system, model D13321, England.

For the analysis of heavy metals, calibration curves were obtained based on three to four standards with standard concentrations for each metal.

Metal analysis was conducted using AAS and calibration curves based on the following standards: SR EN ISO 7980:2002 for Ca and Mg, SR 13315:1996 for Fe, SR 8662-2:1997 for Mn, SR

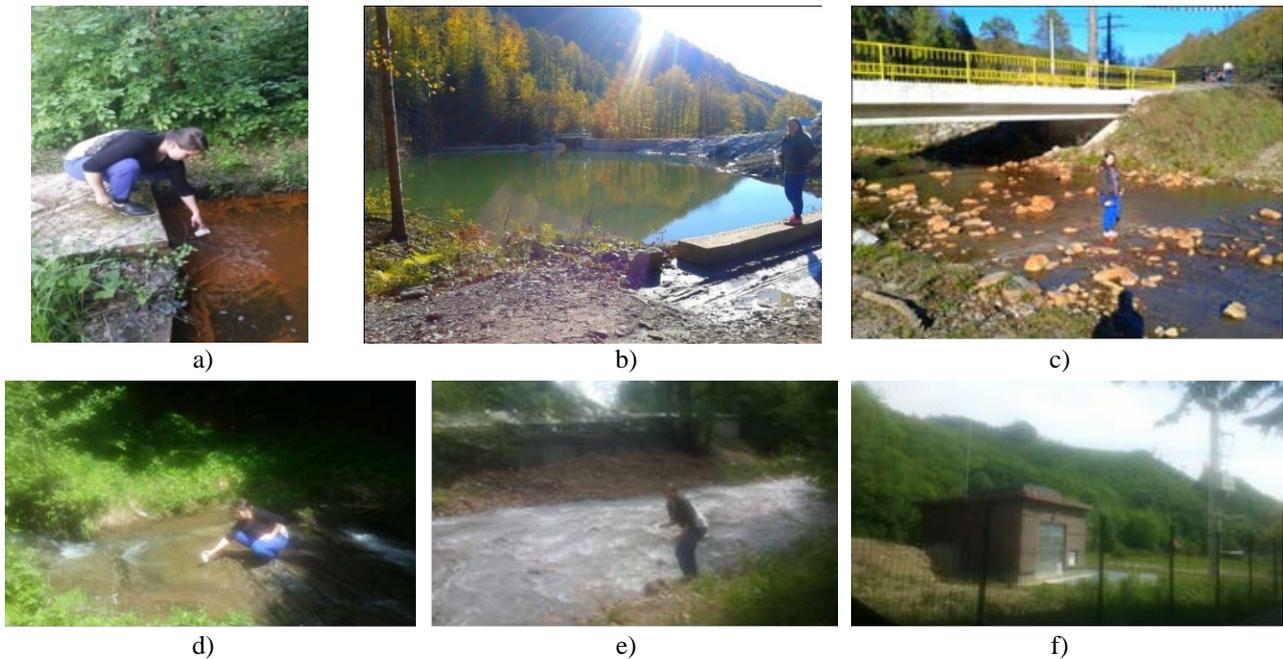


Figure 6. Images of the water sampling locations for several points: a) source of sample P1; b) source of sample P4; c) source of sample P5; d) source of sample P6; e) source of sample P7; f) source of sample P8.



Figure 7. Confluence of the Valea Roșie tributary with the Băița River (Source of sample P10).

ISO 8288:2001 for Co, Ni, Cu, Zn, Cd, and Pb; SR EN ISO 5961:2002 for Cd; ISO 9964-3:2001 for Na.

3.3.2. Determination of pH levels

The pH determination was performed using the manual Vario pH meter. The procedure for determining the pH was as follows:

- calibration of the device using calibration solutions;
- determination of the pH value by inserting the electrode into the sample.

3.3.3. Determination of the Pollution Index

According to Jiao et al. (2023), the single pollution index (P_i) was calculated using the following formula:

$$P_i = C_i / B_i$$

where:

- C_i is represented by the measured concentration of heavy metal i ;
- B_i - surface water environmental quality standard value of heavy metal.

Four pollution levels were adopted based on the calculated index value: clean (<1), light pollution (1 - 2), moderate pollution (2 - 3), and severe pollution (>3).

4. RESULTS AND DISCUSSIONS

4.1. Spatial distribution of heavy metal concentrations

The concentration values of the analyzed metals from the river water samples are presented in Table 1. The variation limits of the analyzed metal concentrations were compared with surface water quality standards according to Normative 161/2006 (Table 1). Surface water classification, from an ecological and chemical perspective, consists of five quality classes: I, II, III, IV, V (Table 1). The concentration values of the main pollutants analyzed in the mine water were compared with the concentration values from the wastewater discharge quality standards NTPA-001/2002, (Table 1).

To interpret and represent the heavy metal analysis results, spatial distribution maps were created, indicating the corresponding water quality classes at each location.

From Table 1, it can be observed that the concentrations of different heavy metals vary

significantly. At sampling points P1, P2, and P3, water is discharged from the Câmpurele gallery. The composition of samples from these three points was compared with the pollutant loading limits for industrial and urban wastewater discharged into natural receptors, in accordance with NTPA-001/2002. The remaining samples come from surface water in the Valea Roşie and Băiţa streams. The concentration values of toxic metals exceed the maximum allowed limits according to Normative 161/2006. The concentrations of Cu, Zn, Ni, Co, Mg, Fe, Mn, Pb, and Cd in the Băiţa River and Valea Roşie at all sampling points exceed the quality limits for classes I and II. The concentration of Cu, Pb, Cd are above the highest limit of V class quality of surface water.

The spatial distribution of heavy metal pollution in the Băiţa stream and its tributary Valea Roşie was mapped using a hydrological map (Figures 8–18).

Analysis of the heavy metal distribution maps reveals that the most affected areas by toxic metals correspond to specific points, primarily where mine water interacts with surface water courses.

Based on the distribution of high concentrations of Pb, Zn, Cd, Ni, and Co, the predominant surface water quality classes in the study area are classes III, IV, and V, according to the standards set by Normative 161/2006.

Copper: The high concentrations of this metal (0.24 - 0.39 mg/L) classify the water as quality class V (Figure 8). These values correspond to sections of the Băiţa River and Roşia Valey River, which interact with mining waste dumps and abandoned mine entrances, from which wastewater from the former mining industry is discharged. The distribution of Cu is also associated with the acidic pH of the analyzed water samples. Anthropogenic sources present in the hydrographic basin may contribute to Cu pollution in rivers (Charters et al., 2021).

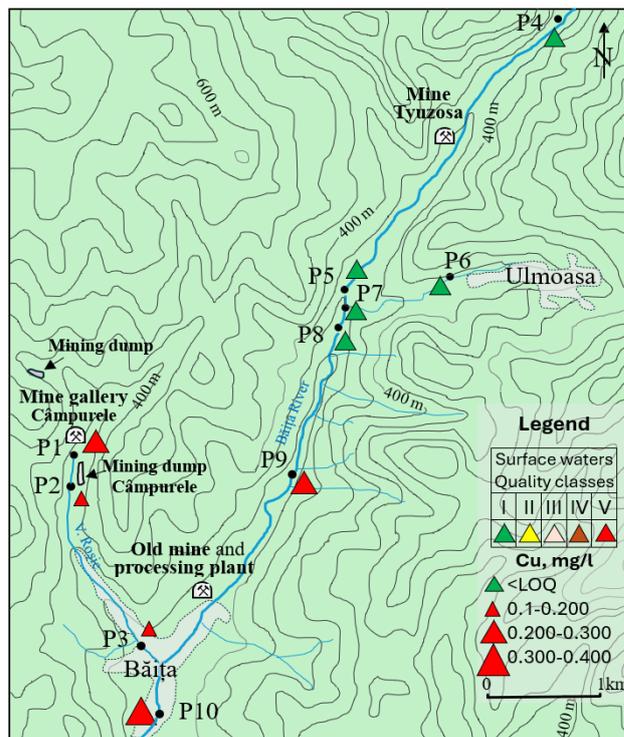


Figure 8. Spatial distribution of Cu.

Zinc: Similarly, Zn has a uniform distribution of the high values across all sampling points, with wide variations ranging from 1 to 49 mg/L, corresponding to quality classes IV and V for surface water (Figure 9). In the Băiţa River, these high Zn values are attributed to the presence of the Tyuzoşa mine, from which mine water is discharged directly into the river. The direct contamination of the river by mine water controls Zn pollution levels in the river water (Shikazono et al., 2008).

Zinc is highly mobile during alteration processes, and its easily soluble compounds are precipitated through reactions with carbonates, or they are adsorbed by mineral and organic compounds in the presence of sulphate anions (Kabata-Pendias & Mukherjee, 2007). Zinc can also originate from non-point

Table 1. Concentrations of metals measured at sampling points P1 - P10 and comparison with surface water quality classes and permissible pollutant loading limits for wastewater discharge into natural receptors (NTPA-001/2002), mg/L.

Indicator	P1 mg/L	P2 mg/L	P3 mg/L	P4 mg/L	P5 mg/L	P6 mg/L	P7 mg/L	P8 mg/L	P9 mg/L	P10 mg/L	Surface waters Quality classes, mg/L					NTPA- 001/2002 mg/L
											I	II	III	IV	V	
Calcium (Ca ²⁺)	171.7	177.7	159.5	21.1	47.3	92.2	19.95	19.02	10.46	28.47	50	100	200	300	>300	300
Sodium (Na ⁺)	11.05	8.3	7.9	3.63.6	7.2	4.8	9	6.5	5	4.6	25	50	100	200	>200	
Copper (Cu ²⁺)	0.245	0.162	0.162	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.246	0.397	0.02	0.03	0.05	0.1	>0.1	0.1
Zinc (Zn ²⁺)	49	32	30	2	2	1	4	4	2	5	0.1	0.2	0.5	1	>1	0.5
Nickel (Ni ²⁺)	0.11	0.13	0.05	0.15	0.09	0.1	0.06	0.07	0.13	0.08	0.01	0.025	0.050	0.100	>0.100	0.1-0.5
Cobalt (Co ³⁺)	0.17	0.22	0.2	<LOQ	0.11	0.073	0.023	0.006	0	0.192	0.01	0.02	0.05	0.1	>0.1	
Magnesium (Mg ²⁺)	82	52	52	6	10	8	11	11	10	15	12	50	100	200	>200	100
Iron (Fe ²⁺ +Fe ³⁺)	60	30	105	<LOQ	<LOQ	<LOQ	9	<LOQ	<LOQ	<LOQ	0.3	0.5	1.0	2	>2	5.0
Manganese (Mn ²⁺ +Mn ⁷⁺)	28	21	11	0.068	0.5	0.039	0.034	0.035	4.13	0.5	0.05	0.1	0.3	1	>1	1.0
Lead (Pb ²⁺)	0.83	0.15	2.33	1.92	1.64	0.83	1.164	1.8	2.49	1.624	0.005	0.01	0.025	0.05	>0.05	0.2
Cadmium (Cd ²⁺)	0.082	0.059	0.081	0.008	0.029	0.009	0.015	0.003	0.019	0.017	0.0005	0.001	0.002	0.005	>0.005	0.2

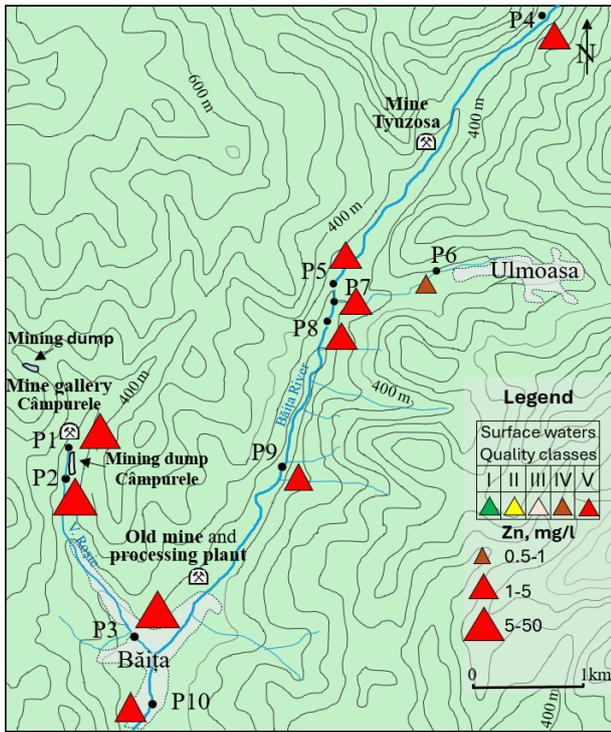


Figure 9. Spatial distribution of Zn.

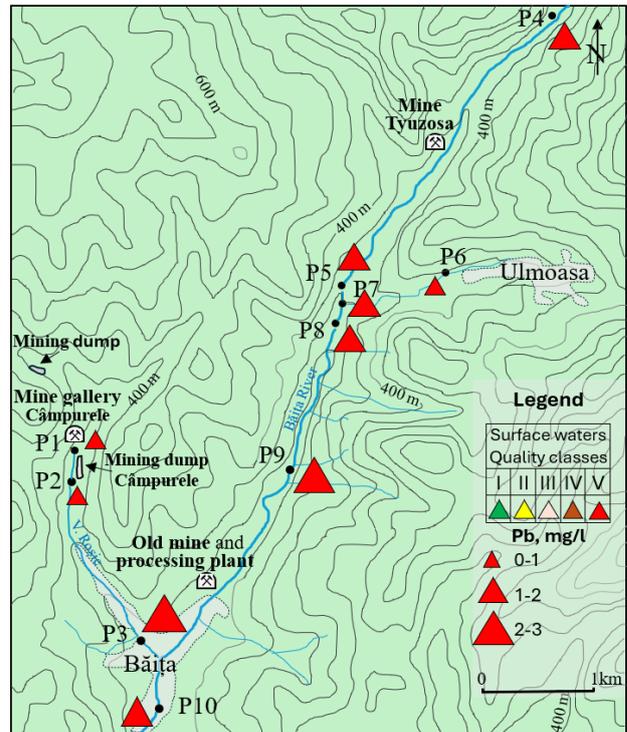


Figure 10. Spatial distribution of Pb.

anthropogenic sources (Andarani et al., 2021, Charters et al., 2021) present in the hydrographic basin.

Lead: The concentrations for Pb indicate high pollution, with variations between 0.15 and 2.49 mg/L. The high level of contamination classifies the water as class V quality according to Normative 161/2006 regarding the classification of surface waters at all 10 measurement points (Figure 10). There are also exceedances of the maximum allowed values for class V quality by up to 49.8 times (Table 1) along the entire tested course of the Băița River and its tributaries (Figure 10).

The main source for the high Pb concentrations in the Băița River is the Tyuzosa mine, where Pb and Zn were exploited.

Cadmium: Cd indicates a high degree of pollution with concentration values ranging from 0.003 to 0.082 mg/L, values corresponding to quality classes III and V. The variation in Cd concentration, according to the spatial distribution map (Figure 11), is similar to that of zinc, which could be explained by the same source of the two metals, zinc ores, where the geochemical behavior of Cd is similar to that of zinc (Shikazono et al., 2008). The cadmium concentration values exceed the maximum allowed limits by up to 16.4 times, except for one sample, which is located at a considerable distance from the pollution sources, thus classifying the water from this point (P8) into class III quality (Figure 11). Compared to zinc, which shows a high value in the same point (P8), the low Cd value can be attributed to the chemical composition of

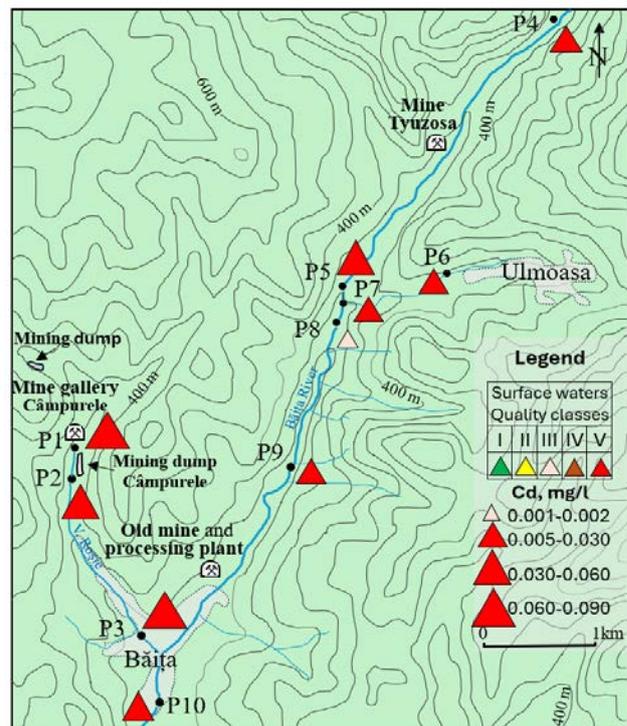


Figure 11. Spatial distribution of Cd.

zinc sulphides (Damian et al., 2021), where Cd is a minor element. However, Cd has a stronger affinity for sulfur than Zn, and its mobility in an acidic environment is greater than that of Zn. During alteration processes, Cd forms simple compounds such as CdO, Cd(OH)₂, CdCl₂ and CdF₂, which are highly mobile and follow Zn in sedimentation processes

(Kabata-Pendias & Mukherjee, 2007). Cd is easily mobile in oxidative conditions, at $\text{pH} < 8$. As the pH decreases and water flow increases, Cd mobility increases (Salminen, 2005).

Iron: Fe is present in surface waters due to natural and geological factors of the hydrographic basin, being a component of silicates in volcanic rocks, but also as a result of mining industrial wastewater.

At high concentrations, it is toxic to aquatic ecosystems, as concentrations of 0.9 mg/L have been shown to be toxic to crabs, and a value of 12 mg/L has caused mortality in pike (Doudoroff & Katz, 1953).

From the examination of iron distribution (Figure 12), high values are shown in points P1, P2, and P3, corresponding to mine water, which is associated with a very acidic pH of 2.71 - 2.84. The concentration of iron exceeds the pollutant load limits for wastewater in samples P1, P2, and P3, which are required before being discharged into natural receptors according to NTPA-001/2002.

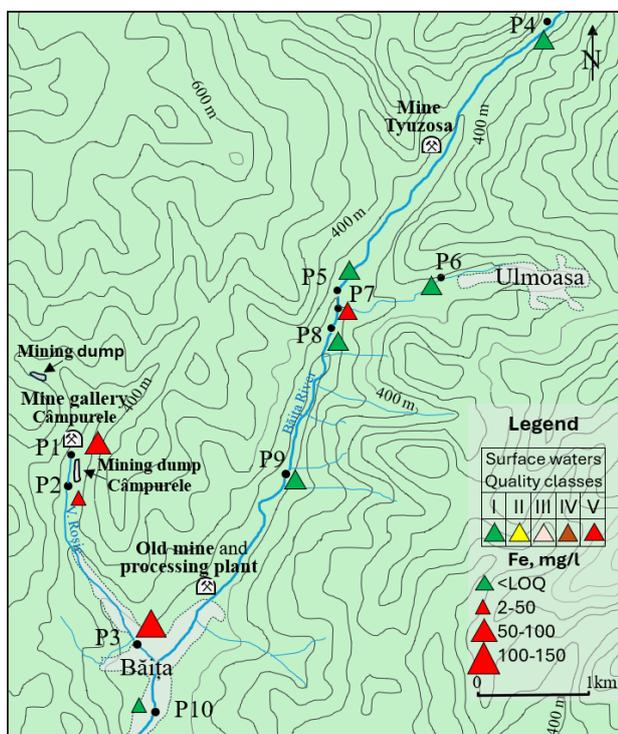


Figure 12. Spatial distribution of Fe.

In water samples from the two river courses, the iron concentration is below the standard limit of class I water quality, except for sample P7, where the iron concentration exceeds the upper limit specific to class V, and in the area adjacent to the sampling point, a mine gallery is present.

Additionally, variations in iron concentration in the presence of acidic pH imply an exchange of iron primarily between the river water and the soil in the immediate vicinity of the riverbed (Ekström et al.,

2016).

High levels of iron can also be attributed to anthropogenic factors, which can play a significant role in the contamination of freshwater resources with iron (Sarkar & Shelkhar, 2018).

Manganese: Mn is present in combination with iron in several minerals. In the studied area, Mn appears in black andesites and basaltic rocks that contain pyroxenes.

Due to the connection between Fe and Mn, the spread of manganese is also explained, with its values being higher, which, based on concentration, classifies the water in the class V water quality.

These high values of manganese concentration correspond to the samples where iron also has values classified in the same class (Figure 13).

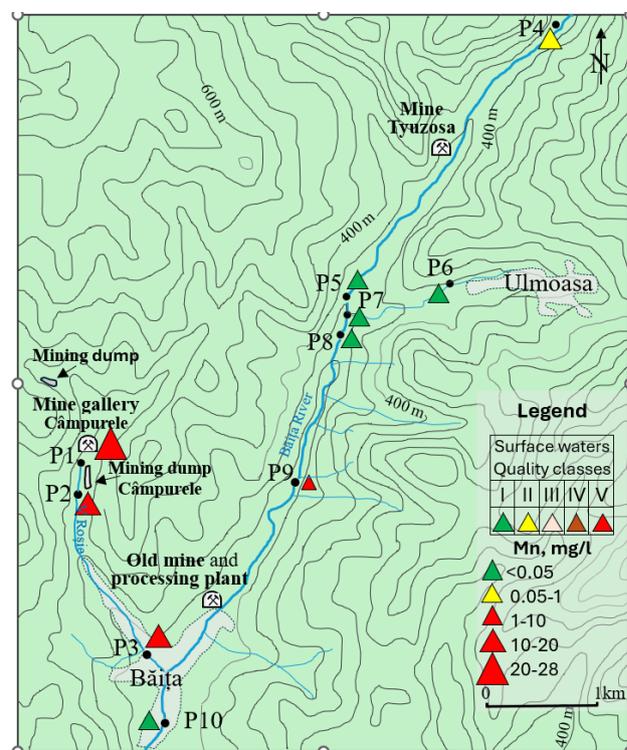


Figure 13 Spatial distribution of Mn.

Cobalt: In most samples, Co exceeds standard values, with concentrations ranging from 0.006 to 0.22 mg/L. The high cadmium concentration causes the water to be classified as quality class V in almost all sampling points (Figure 14).

The presence of this element in high concentrations in the analyzed water is due to its occurrence (Co) as a minor element in the structure of pyrite.

The high Co concentrations at the measured points are attributed to their location in infiltration zones of mining waste dumps and mine entrances.

Nickel: The variation in Ni concentrations in the analyzed water is evenly distributed across three

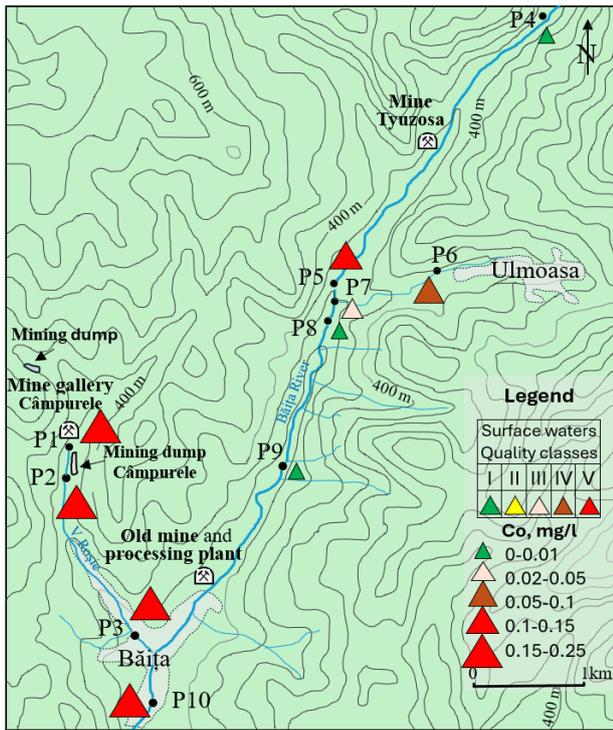


Figure 14 Spatial distribution of Co.

quality classes, ranging from 0.05 to 0.09 mg/L for class II, 0.1 mg/L for class IV, and between 0.13 and 0.15 mg/L for class V. The points where Ni concentration falls into quality classes IV and V correspond to the upper course of the Băița River, which is connected to the water drainage basin for a micro hydro power plant. It remains in quality class IV at the level of the Ulmoasa River and after the discharge of water from the micro hydro power plant into the Băița River. Upstream of the village of Băița, the Ni concentration corresponds to quality class V (Figure 15).

Based on the spatial distribution map (Figure 15), it is estimated that the source of Ni is either natural or derived from mining waste that is drained by surface waters. Nickel, similar to cobalt, is a minor element in pyrite. After alteration, Ni co-precipitates with Mn and Fe oxides, being incorporated into goethite, limonite, serpentine, as well as other Fe-containing minerals. Ni is also found associated with carbonates, phosphates, and silicates.

Water pollution with Ni is generally reflected by its accumulation in river sediments (Kabata-Pendias & Mukherjee, 2007).

Magnesium: Similar to sodium, the concentration values of magnesium place the water within the high quality classes, with the predominant classification being class I quality (Figure 16). The origin of magnesium is also attributed to natural geological formations drained by water.

In most of the samples, magnesium contents range from 6 to 11 mg/L, corresponding to surface

waters and being lower than the maximum value specified for class I water quality. A slight increase was measured in sample P10, reaching 15 mg/L, while concentrations between 52 and 82 mg/L exceed the standard for classes I and II at sampling points P1 - P3, which leads to classification in class III water quality at those points.

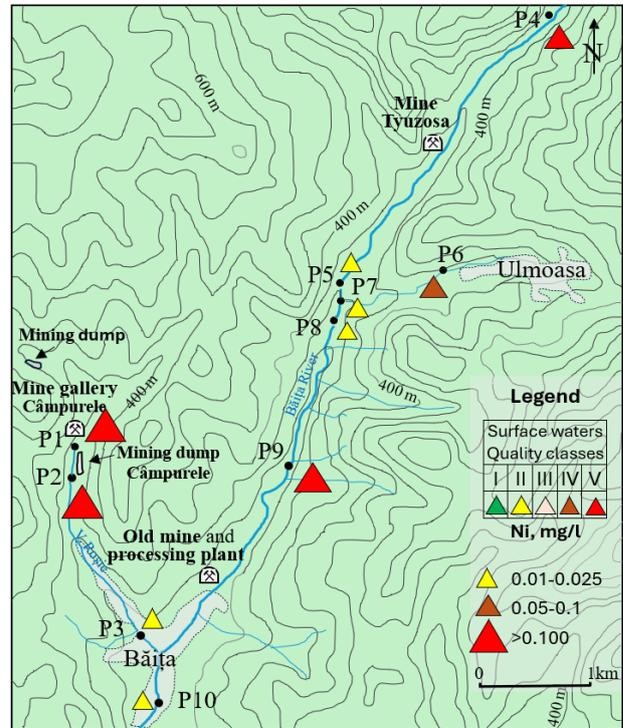


Figure 15 Spatial distribution of Ni.

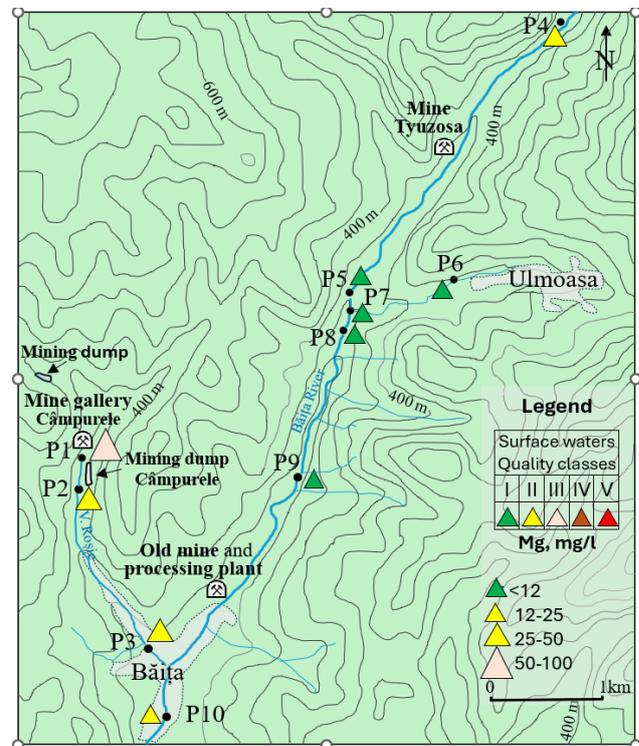


Figure 16. Spatial distribution of Mg.

Calcium: The concentration values of calcium are elevated only at the points with a significant contribution of calcium compounds, P1 - P3 (Figure 17). This is due to the mixture of mining water with lime dust from the discharge from the Câmpurele mining gallery. For calcium, an increase in concentration values up to 159.5 and 177.7 mg/L was noted in samples from points P2 and P3. However, the amount of lime that is used for water treatment is much too high, thus the mine waters after a few tens of meters distance fall into the III quality class in terms of Ca content.

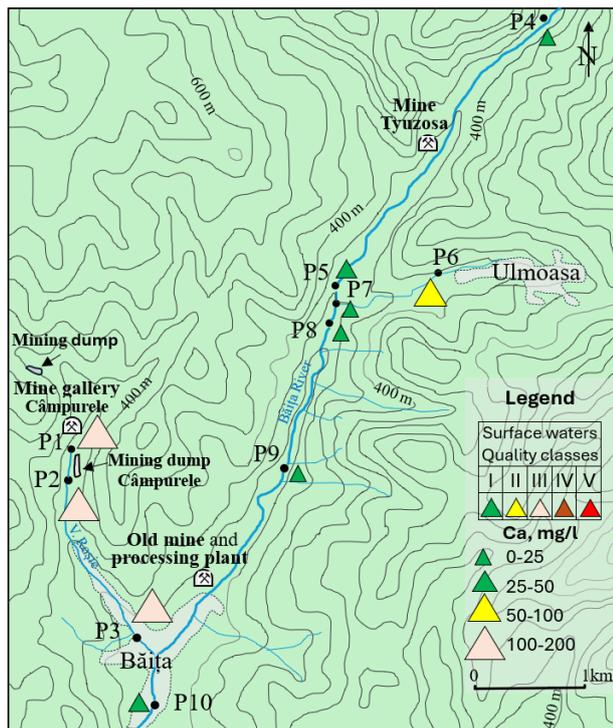


Figure 17 Spatial distribution of Ca.

At the other sampling points, except for point P6, the calcium concentration values were below 50 mg/L, which classifies the water as class I quality.

At sampling point P6, located on the Ulmoasa tributary, the calcium concentration was 92.2 mg/L, a value that classifies the water as class II quality. The higher concentration of calcium can be explained by the intake of calcium from altered rocks.

The increase in water hardness (calcium content – up to 150 mg/L CaCO₃) can lead to a decrease in the toxicity of zinc (Li et al., 2021).

Sodium: The concentration values of Na range from 3.63 mg/L to 8.3 mg/L at all sampling points of natural waters, and these concentrations correspond to class I quality (Figure 18). The highest value for Na is 11.05 mg/L and is found at point P1, where water is collected from the Câmpurele mine gallery. The source of Na is represented by natural geological formations.

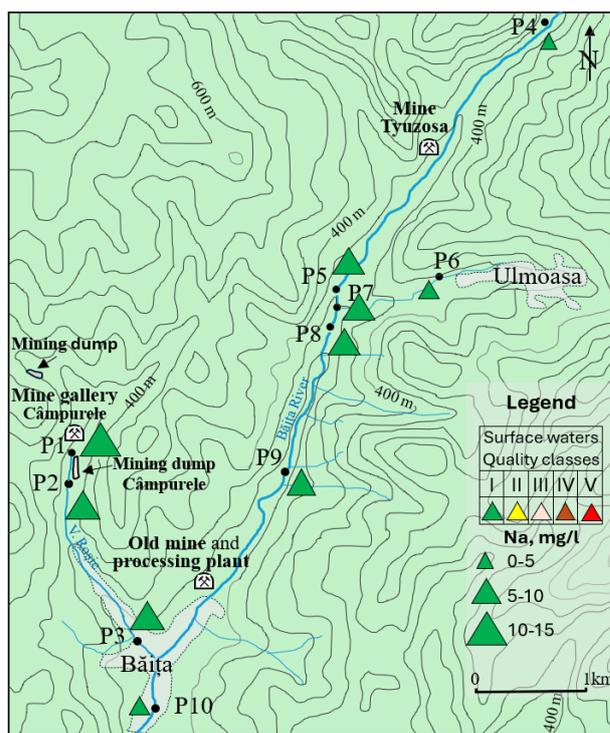


Figure 18. Spatial distribution of Na.

Analyzing each metal (measured) individually at each point on the spatial distribution maps, it is observed that all sampling points contain Pb, Zn, Cd, Ni, and Co at concentrations exceeding the maximum values specific to classes III, IV, and V of water quality according to the national standard (Normative 161/2006).

The cadmium, cobalt, and nickel contents decrease after the confluence of the Băița River with the Ulmoasa River, due to the influx of clean water from the lateral streams.

Cu, Pb, Zn and Cd have as source the polymetallic deposits, (Wei et al., 2018), or are specific for gold deposits (Ninga et al., 2011), so the mining activity represents the main source for the high concentration in all sampling points.

The concentrations of iron and copper decrease as a result of the treatment of mine water with calcium carbonate. In the Băița area, the iron concentration exceeds the maximum limit for class V quality in sampling points P3 and P7, which correspond to the interference zone of water from the Valea Roșie tributary with water from the Câmpurele mining gallery (P3) and the confluence of the Ulmoasa stream with the Băița stream (P7). The excess iron is due to both the contribution from mine water and mine tailings, as well as potential waste from the livestock farm located upstream on the left bank of the Băița River.

Other studies have highlighted the bioavailability of iron and its importance in aquatic biological activity (Viana et al., 2021).

The absence of iron in the other sampling points can also be explained by the retention of the metal in the brownish-red sludge deposited on the riverbed of the two streams, resulting from the precipitation of iron oxyhydroxides formed through abiotic neutralization (Baleeiro et al., 2018).

4.2. The results of pH determination in the water samples

The pH values measured in the 10 sampling points are shown in Table 2. The results indicate high acidity in the samples from P1, P2, P3, and P10. These low pH values are due to the acid mine waters from the former mine galleries and the water infiltrating the tailing piles, which subsequently discharge into the Valea Roşie River.

The samples from points P1, P2, P3, and P10 do not fall within the pH range specified in Normative 161/2006 for surface water classification, where the optimal pH value is between 6.5 and 8.5.

Tabel 2. pH determination results.

Sample	pH sample	pH, N161/2006	pH, NTPA-001/2002, HG 188/2002, HG.352/2005
P1	2.71		
P2	2.76		
P3	2.84		
P4	7.40		
P5	7.26		
P6	7.29	6.5-8.5	6.5-8.5
P7	7.21		
P8	7.13		
P9	7.19		
P10	4.08		

The results indicate high acidity in samples P1 and P2, which are representative of water from the Câmpurele mining gallery. Samples P3 and P10, located upstream on Valea Roşie stream and downstream on Băiţa stream, owe their acidity to the occasional discharge of mine water and runoff from mine dumps. The discharge of these polluted waters into surface water bodies alters their chemical state (Minga et al., 2023).

4.3. Pollution Index (P_i)

The method for determining the water pollution index (after Jiao et al., 2023) is based on the concentrations of metals determined through analysis, reported to the standard value of the surface water quality class according to national legislation. This

method is used to assess the level of river water pollution.

Pollution indices are generated by evaluating heavy metals and their concentration in water bodies or sediments (Islam et al., 2018) and are an indicator for assessing the ecological consequences of surface water pollution (Banu et al., 2013; Milanović et al. 2011).

Pollution indices (P_i) undergo a four-stage evaluation process, for example, selecting water quality parameters, determining sub-indices of the parameters, assigning weights to the parameters, and finally applying an aggregation function to calculate the pollution index. The accuracy of the results can be influenced by the complexity of the issue (Syeed et al., 2023).

The pollution indices calculated at points P3 - P10 for each analyzed element are presented in Table 3. The water from points P1 and P2 is considered mine water and cannot be taken into account in the classification of surface waters.

The pollution indices were calculated in accordance with the data published by Jiao et al. (2023). Similar research has been conducted by Hafsi et al. (2024) for surface water from industrial areas. Low values indicate the absence of pollution sources (Table 3). Based on the single-factor pollution index value (Jiao et al., 2023), pollution levels were determined for each metal according to the quality class in which the surface water was classified at the respective sampling point.

Copper has many values that exceed the pollution level. In some points, the pollution index falls into the Light to Severe Pollution category. These points are located on the Roşia Valley River and Băiţa River.

The pollution index values for zinc range from "Light to Severe Pollution." The highest value is recorded at the collection point where water exits the mine gallery. At the exit of the treatment plant, the index indicates "Light to Moderate Pollution." At the confluence of the Roşia River and Băiţa River, the pollution index increases, likely due to the solubilization of zinc from deposited sediments. Thus, for Zn, the pollution level is classified as "Light to Severe Pollution" for the majority of sampling points (over 50 %).

For nickel, the pollution index ranges from "Light to Moderate Pollution." The high pollution index values do not indicate a pollution source. Large amounts of nickel originate from the geological background, where nickel is frequently found in the composition of pyroxenes in andesites (Damian et al., 2022), and less from the alteration of pyrites in non-ferrous ores.

Table 3. Pollution index for each element in the P3 – P10 sampling points.

Sample	Pollution index (P _i)							
	Cu	Zn	Ni	Co	Fe	Mn	Pb	Cd
P3	1.62	30	2	2	52.5	11	4.66	16.2
P4	-	2	1.5	-	-	0.68	38.4	1.6
P5	-	2	1.125	1.1	-	10	32.8	5.8
P6	-	1	1	0.73	-	0.78	16.6	1.8
P7	-	4	2.4	0.23	4.5	0.68	23.28	3
P8	-	4	2.8	0.06	-	0.72	36	1.5
P9	2.46	2	1.3	-	-	4.13	49.8	3.8
P10	3.97	5	3.2	1.92	-	0.5	32.4	3.4
Pollution level	Light to Severe pollution	Light to severe pollution	Light to moderate pollution	Light pollution	Severe pollution	Light to Moderate pollution	Severe pollution	Light to severe pollution

For cobalt, the index indicates normal values and, to a lesser extent, "Light Pollution."

The pollution index for iron is at normal values. At the gallery exit point, the water has a pollution index classified as "Severe Pollution. As a result of the increase in pH, the iron in the mine water is deposited in the form of red precipitates, (Tabel 2).

For manganese, the evaluation index indicates "Moderate Pollution" only in two points while in the rest of the samples, the pollution index values indicate clean water.

In all investigation points, the pollution index for lead indicates "Severe Pollution."

The pollution index for cadmium ranges from "Light to Severe Pollution." The highest values are found at the mine water discharge point and along the Roşia River until its confluence with the Băiţa River. The high pollution index values along this river segment indicate that mining activity is the main pollution source. Another pollution point (P5) has a severe pollution index due to the influence of water from the Ulmoasa stream, which also collects mine drainage from this area. Thus, for Cd, in 62.5 % of the samples, the pollution level is classified as "Severe Pollution."

Even though, in some sampling points, the pollution index falls under "Clean" and "Light Pollution," we still classify the pollution as "Severe Pollution." This classification is justified because the toxic elements: cadmium, lead, zinc, and sometimes copper are categorized under "Severe Pollution."

4.4. Statistical analysis

To establish a linear relationship between two parameters of the water samples, the Pearson correlation coefficient can be used. The statistical analysis was performed by determining the correlation between elements and calculating the

Pearson coefficient (Table 4).

The index of positive correlation is dominant for heavy metals.

Copper has positive correlations with zinc, cadmium, and iron (Table 4). Moderate positive correlations indicate the same source, represented by the extraction of base metal ores and the deposits of mining waste heaps. The low values of the Pearson correlation coefficient are also caused by the presence of unpolluted water samples upstream on the Băiţa River. Even though the correlation with zinc has positive values, the correlation index varies significantly across sampling points (Figure 19a). There is no correlation with lead and nickel.

Zinc strongly correlates with cadmium because they are frequently found in the structure of sphalerite (Damian et al., 2021).

The correlation points are projected near the correlation line, which also suggests a single source for both zinc and cadmium, originating from the extraction of non-ferrous ores (base metals) rich in zinc (Figure 19a). There is also a positive correlation with iron and a moderate one with copper (Figure 19b). The high values of these correlations are due to common sources for these elements. The moderate positive correlation with cobalt and similar correlation values with iron can be explained because both iron and cobalt predominantly have the same source, represented by pyrite deposits in the mining waste. The weak positive correlation with calcium, magnesium, and manganese is explainable because these elements predominantly come from the geological substrate.

Lead correlates negatively, moderately, and weakly with the other metals except for copper. These correlations can be explained by the fact that Pb forms insoluble compounds that settle in the sludge at the treatment plant or in the river sediments. The negative correlations may also be explained by other sources from the local settlements.

Table 4. Correlation matrix for the analyzed elements at the sampling points in the Băița River hydrographic basin.

	Ca	Na	Cu	Zn	Ni	Co	Mg	Fe	Mn	Pb	Cd
Ca	1										
Na	0.616	1									
Cu	0.224	0.049	1								
Zn	0.893	0.761	0.390	1							
Ni	0.013	-0.316	0.054	0.020	1						
Co	0.777	0.422	0.564	0.693	-0.177	1					
Mg	0.889	0.764	0.418	0.998	-0.002	0.704	1				
Fe	0.776	0.583	0.258	0.797	-0.330	0.612	0.801	1			
Mn	0.861	0.721	0.407	0.966	0.220	0.632	0.962	0.651	1		
Pb	-0.524	-0.443	0.076	-0.400	-0.176	-0.402	-0.382	0.034	-0.482	1	
Cd	0.887	0.718	0.389	0.934	-0.094	0.755	0.940	0.901	0.872	-0.211	1

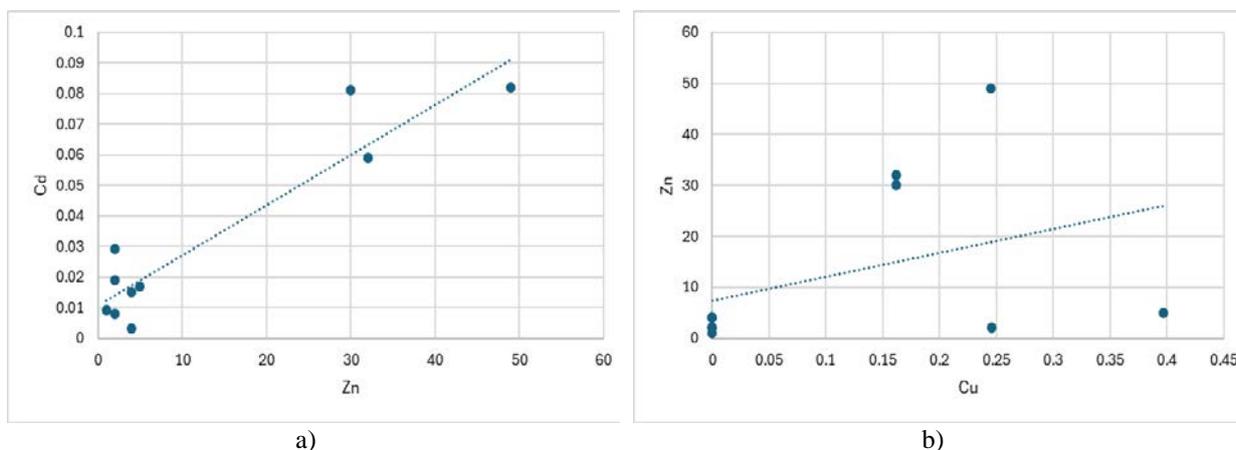


Figure 19. The correlations between: a) zinc and cadmium; b) zinc and copper.

Cadmium has moderate positive correlations with copper and strong positive correlations with other elements: Ca, Na, Mg, Mn, which explains the source of this element solely from the geological substrate (ore deposit and geological substrate) and not from an industrial pollution source.

Iron strongly correlates positively with zinc and cobalt, indicating a common source represented by the old mining operations, with zinc being commonly adsorbed by Fe-Mn oxyhydroxides (Andarani et al., 2021, Gammons et al., 2015). The positive correlations with manganese, magnesium, and calcium indicate that the source is also the geological substrate.

The statistical analysis through correlation coefficients shows that the concentrations of metals are influenced by common pollution sources and their geochemical behavior is in accordance with other information according to Ninga et al. (2011).

5. CONCLUSIONS

Ten water samples were analyzed at 10 sampling points (P1 - P10), three of which (P1 - P3)

came from the direct mine water discharge sources, while the remaining samples were from surface waters draining the mining area, moving downstream from the Băița locality.

Acid mine drainage in this area has been present since the beginning of mining activities and continues to persist even after the mines were closed. Mine waters are characterized by acidic pH and high concentrations of heavy metals. The study conducted in the Băița area provided a comprehensive view of the consequences related to the extraction of base metals, the abandonment of mines, and the mismanagement of mine waste dumps. The dumps, the brown-red color of the alluvial sediments, and the color and smell of the water in the rivers provide clear evidence of pollution.

The results obtained demonstrated the contamination of surface waters with heavy metals in the Băița locality area. The highest values were recorded for Pb, Cd, and Zn, followed by Co, Ni, and Cu. The highest concentrations in the first group of metals exceeded the upper limit of quality class V.

Mine water samples (P1 - P3) contain all the metallic indicators with concentrations exceeding the pollutant loading limits for industrial wastewater

discharged into natural recipients, in accordance with NTPA-001/2002. The excessive concentration of heavy metals is closely linked to the abandonment of mining activities. Evidence exists of the interference between the mine water discharged from the Câmpurele gallery upstream and the water from the two streams. A long-term pollution source is also represented by the immense Câmpurele mine waste dump, from which metals contained in the rock and ore residues are continuously released and are in varying stages of alteration.

The primary minerals subjected to transformation are pyrite, sphalerite, chalcopyrite, arsenopyrite, and, to a lesser extent, galena. Among these minerals, the most important pollutant element is pyrite. It undergoes complex alteration processes that took place throughout the underground mining operation and also in the waste dumps along the Câmpurele valley. New minerals, rich in Fe, such as goethite and ferrihydrite, which are poorly crystallized and may be responsible for metal adsorption, can form in this process.

Therefore, it can be observed that in the points where an acidic pH was recorded, namely points P1 - P3 and P10, the content of heavy metals (Cd, Co, Cu, Fe, Mn, Ni, Pb, and Zn) is high, leading to the classification of the surface water quality (Roşie Valley and Băița Rivers) in class V.

In areas where the pH was neutral, the analysis of heavy metals was below the detectable limit (<LOQ) for Cu and Fe, while the concentrations of Mg, Mn, and Na classified the surface water in quality class I.

Among the analyzed water samples, the standard limits were exceeded by 100 % for Pb, 90 % for Cd and Zn, 80 % for Co, and 50 % for Cu and Ni.

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