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TEMPORAL TRENDS OF HEAVY METALS AND GEOCHEMICAL PARAMETERS IN RAMURA 1, 2, 3 AND 4 PETRILA TAILINGS DUMP

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Abstract: *Mining activities in the Jiu Valley coal basin have generated substantial volumes of waste materials, and Ramura 1, 2, 3 and 4 tailings dump in the Petrila area represents a continuously evolving geochemical system. This study examines the temporal trends of heavy metals (Cr(total), Cu, Ni, Zn, Co, Pb) and associated geochemical parameters (pH, Eh, moisture, organic matter, electrical conductivity) over the 2022–2024 period, using standardised determinations and year-to-year comparative analysis. The results indicate progressive increases in Cr (total), Cu and Ni (3–7% annually), while Zn remains consistently elevated but relatively stable. Geochemical parameters reveal gradual acidification (pH 6.72 → 6.31) and increasingly oxidising conditions (Eh 312 → 342 mV), both favouring the mobilisation of redox-sensitive metals. Comparison with Romanian Regulation Order 756/1997 shows no exceedance of intervention thresholds; however, several metals surpass normal values or approach alert thresholds. Overall, the findings confirm the dynamic nature of Ramura 1–4 tailings dump and highlight the need for continued long-term monitoring to assess environmental risks and to support informed post-mining management strategies.*
Keywords: *heavy metals, temporal evolution, tailings dump, oxidation processes, geochemical parameters*

1. Introduction

Mining activity carried out for more than a century in the coal-bearing basin of the Jiu Valley has generated substantial quantities of waste material originating from both underground extraction and coal processing. These materials were deposited near mining sites, mainly along slopes or in natural accumulation areas, forming the characteristic tailings dumps of the region. In Petrila area, Ramura-type tailings dumps developed through successive deposition stages, resulting in elongated and parallel bodies adapted to the topography of secondary valleys. Today, these deposits represent significant potential sources of soil and groundwater contamination, as active geochemical processes can mobilise heavy metals and other toxic elements derived from carboniferous waste.

Tailings dumps are widely recognised in international literature as open systems undergoing persistent physic-chemical transformations, including sulphur oxidation, acid drainage formation, carbonate dissolution, metal solubilisation and colloidal transport [1, 2, 3]. These processes are strongly governed by environmental parameters such as pH, Eh, moisture, temperature, waste texture and oxygen exposure. Under oxidising conditions—common in tailings dumps exposed to the atmosphere—the mobility of metals such as Fe, Mn, Ni, Cu and Cr increases substantially, enhancing their impact on soils and adjacent environmental compartments.

Studies conducted in mining regions from Spain, Poland, the Czech Republic, Germany and China demonstrate that non-rehabilitated tailings dumps exhibit significant temporal variations in heavy metal concentrations over intervals of 3–5 years. Research from the Rio Tinto district in Spain [4, 5] and from the Silesia region in Poland [6] reports progressive increases in mobile metal fractions, correlated with intensified redox processes and climate-related fluctuations, including increased precipitation and freeze–thaw cycles. Additional studies emphasise that the temporal dynamics of heavy metals constitute a direct indicator of the geochemical instability of mining deposits [7, 8].

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In Romania, research addressing the multi-year evolution of heavy metals in tailings dumps remains limited. Most studies have focused on static contamination assessments or one-time ecological risk evaluations. In the Jiu Valley, investigations have identified elevated concentrations of Cr, Ni, Cu, Mn, Pb and Fe at Ramura-type deposits [9], yet their temporal evolution and relationship with key geochemical parameters have seldom been analysed systematically. This information gap is relevant, as redox-driven mineral alteration processes can modify metal mobility over relatively short periods.

In this context, multi-year monitoring of Ramura 1, 2, 3 and 4 tailings dump is essential for understanding the mechanisms governing metal mobilisation and for evaluating potential ecological risks. The proximity of these deposits to residential areas and watercourses further underscores the need to characterise their geochemical evolution and environmental implications.

Therefore, this study aims to:

- investigate the temporal evolution of heavy metal concentrations within Ramura 1–4 tailings dump over the 2022–2024 period;
- assess the influence of key geochemical parameters (pH, Eh, moisture and organic matter content) on metal mobility;
- identify dominant geochemical processes controlling the behaviour of these elements; and
- evaluate the medium-term geochemical stability of the waste material.

By integrating multi-year monitoring, geochemical interpretation and comparison with international research, this study contributes important insights into the evolution of carboniferous tailings dumps in the Jiu Valley and supports future monitoring, management and rehabilitation strategies.

2. Materials and Methods

2.1 Study area

The study area is represented by Ramura 1, 2, 3 and 4 tailings dump within Petrila mining perimeter, a depositional structure resulting from coal preparation activities at the former Petrila Coal Processing Plant. Deposition on the first three Ramura segments remained active until 1970, when mining operations were restructured and the tailings dumps were gradually placed into conservation. Ramura 4, developed later for waste derived from underground extraction, continued to operate until 2015, when Petrila mine was permanently closed. Altogether, the four Ramura units cover approximately 80.04 ha and contain an estimated 5.105 million m³ of waste material, making them a key element in assessing environmental impact in the Jiu Valley [10, 11].

The general configuration of the tailings dump, together with the distribution of the sampling points, is illustrated in Fig. 1.

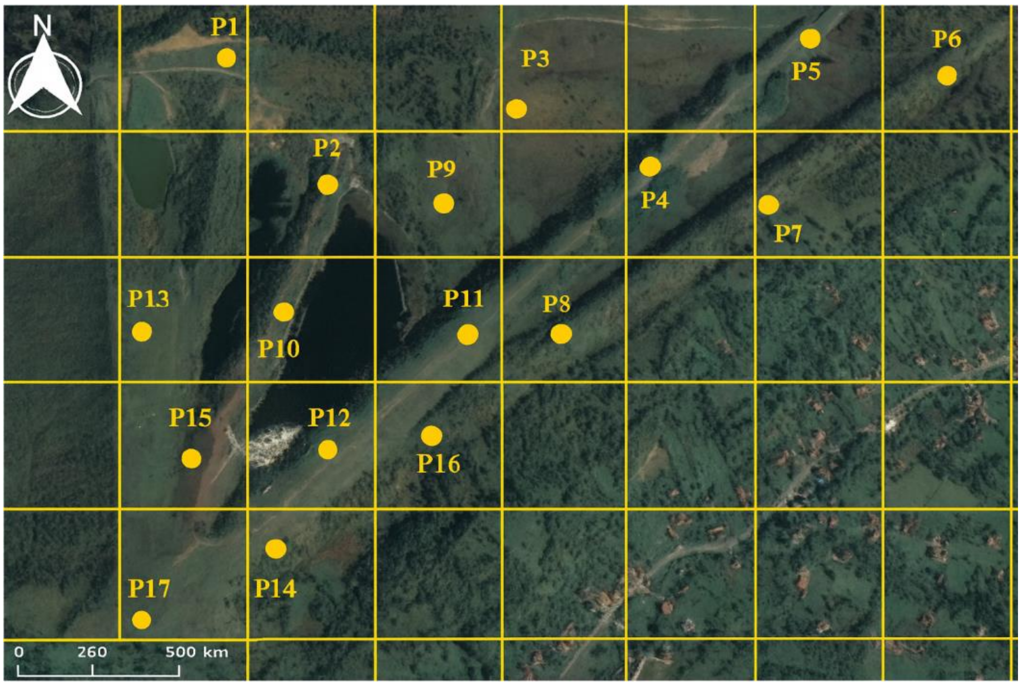

Fig. 1. Map of the study area and distribution of sampling points across Ramura 1, 2, 3 and 4 tailings dump

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The deposited waste originates mainly from coal extraction and processing and has a heterogeneous composition dominated by clay and sandy-clay rocks, shales, carbonaceous fragments and minerals associated with alteration processes. The morphology of the dump has been shaped by local topography, forming elongated bodies arranged parallel to the valley, with moderate to steep slopes along the margins.

Approximately 20 m downslope from the base of Ramura 1–4 lie three anthropogenic lakes formed through compaction and subsidence of the deposited materials, followed by surface water accumulation. These water bodies influence the local microclimate and can enhance metal mobilisation by maintaining higher moisture levels at the base of the dump.

During recent decades, the surface of the tailings dump has undergone both spontaneous and assisted revegetation. Natural colonisation includes species typical of disturbed environments (bramble, dog rose, and hawthorn), while anthropogenic interventions involved planting black locust (Robinia pseudoacacia) and Scots pine (Pinus sylvestris), species known for their tolerance to degraded soils and slope-stabilising role. Although vegetation reduces erosion, mineralogical alteration and metal mobilisation processes remain active within the waste material.

Thus, Ramura 1–4 tailings dump represents a complex and dynamically evolving geochemical system, influenced by operational history, lithological characteristics and local climatic factors—factors which together justify the need for multi-year monitoring.

2.2. Sample collection

Sampling campaigns were carried out over three consecutive years (2022, 2023 and 2024), using the same twelve fixed sampling points (P1–P12) uniformly distributed across Ramura 1–4 tailings dump. This ensured coverage of both horizontal and vertical contamination gradients. Soil samples were collected from the 0–20 cm layer, considered the most sensitive to atmospheric exposure and the most relevant for metal mobility.

All sampling campaigns were conducted during the same season (late spring), in order to minimise the influence of seasonal variability on geochemical parameters and metal mobility.

Soil sampling was carried out using consistent and internationally accepted methodologies for soil investigations, and samples were stored in sealed containers to avoid physic-chemical alteration prior to analysis.

2.3. Laboratory analyses

Determinations of pH and redox potential (Eh) were performed in situ using standardised portable electrodes immediately after sampling. pH measurements were conducted with a Hanna HI-98129 portable multiparameter meter, calibrated before each field campaign. Redox potential (Eh) was measured using a Hanna HI-8314 electrode equipped with a platinum sensor and an Ag/AgCl reference system, ensuring stability and accuracy under field conditions.

Moisture content was quantified by oven drying at 105°C, while organic matter content was determined by calcination at 550°C. Concentrations of heavy metals (Cr, Cu, Ni, Zn, Pb, Mn, Fe) were analysed using atomic absorption spectrometry (AAS) or ICP-OES following acid digestion in accordance with ISO 11466 methodology [12, 13].

2.4. Evaluation of temporal trends

Data interpretation was performed by comparing annual values of heavy metals and geochemical parameters using the following indicators: annual mean, standard deviation, year-to-year percentage variation, linear regression trends.

These analyses made it possible to identify the direction and magnitude of metal evolution and to assess the geochemical stability of the tailings dump during the 2022–2024 monitoring period.

3. Results

3.1. Heavy metal concentrations during 2022–2024

The analysis of soil samples collected from Ramura 1, 2, 3 and 4 tailings dump shows clear increasing trends for most heavy metals throughout the 2022–2024 period. The annual mean concentrations and corresponding standard deviations (SD) are presented in Table 1.

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Table 1. Annual mean concentrations and standard deviations (SD) of heavy metals during 2022–2024

Indicator (mg/kg)	2022 (Mean ± SD)	2023 (Mean ± SD)	2024 (Mean ± SD)
Cr (total)	35.53 ± 8.01	38.37 ± 8.65	41.05 ± 9.29
Cu	53.05 ± 7.97	55.17 ± 8.29	57.11 ± 8.57
Ni	84.07 ± 14.95	87.44 ± 15.55	90.49 ± 16.13
Zn	124.54 ± 44.12	124.54 ± 44.12	127.03 ± 44.82
Co	22.76 ± 9.84	23.67 ± 10.24	24.50 ± 10.63
Pb	19.03 ± 2.46	19.80 ± 2.55	20.30 ± 2.61

Chromium shows the most pronounced year-to-year increase, reaching 41.05 ± 9.29 mg/kg in 2024, representing a ~6.8% rise compared to 2023. Copper and nickel also show moderate but continuous increases, while zinc remains the metal with the highest absolute concentrations, although with minimal inter-annual variation. Cobalt and lead display small but steady upward trends, with low variability reflected by narrow SDs.

The temporal evolution of mean concentrations for the analysed metals is illustrated in Fig. 2, highlighting consistent upward trends during the 2022–2024 monitoring interval.

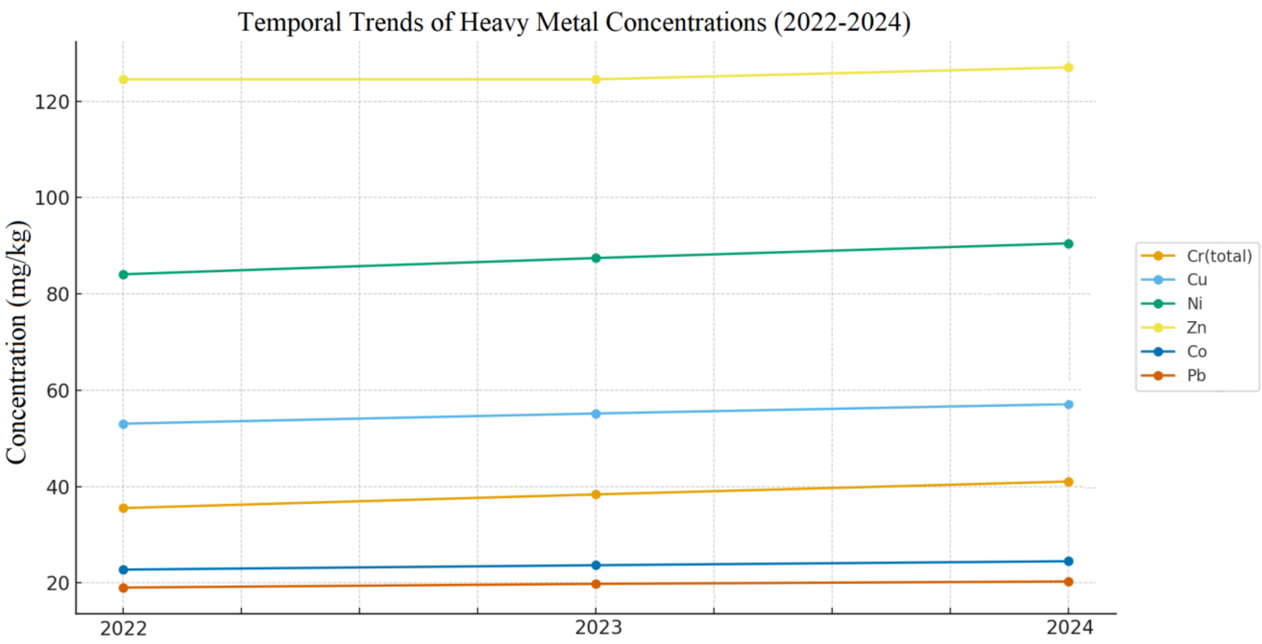

Fig. 2. Annual evolution of mean heavy-metal concentrations (2022–2024)

Figure 2 shows that all analysed metals follow a gradual upward trend over the three-year period, with Cr(total), Cu and Ni exhibiting the most pronounced increases. Zinc remains the metal with the highest absolute concentrations, although its temporal variation is minimal. Cobalt and lead display only subtle increases, confirming relatively stable but slowly rising concentration levels across the tailings dump.

To complement these trends, Figure 3 shows the annual distribution of concentrations for each metal, providing additional insight into variability among sampling points.

Figure 3 confirms that Cr (total), Ni, and Cu show the most pronounced upward trends across the monitoring period, reflected by progressively higher medians and expanded upper quartiles. Zinc maintains the highest concentration range overall, with only minor interannual variability. Cobalt and lead display comparatively stable distributions, although both follow a subtle but consistent increasing trajectory.

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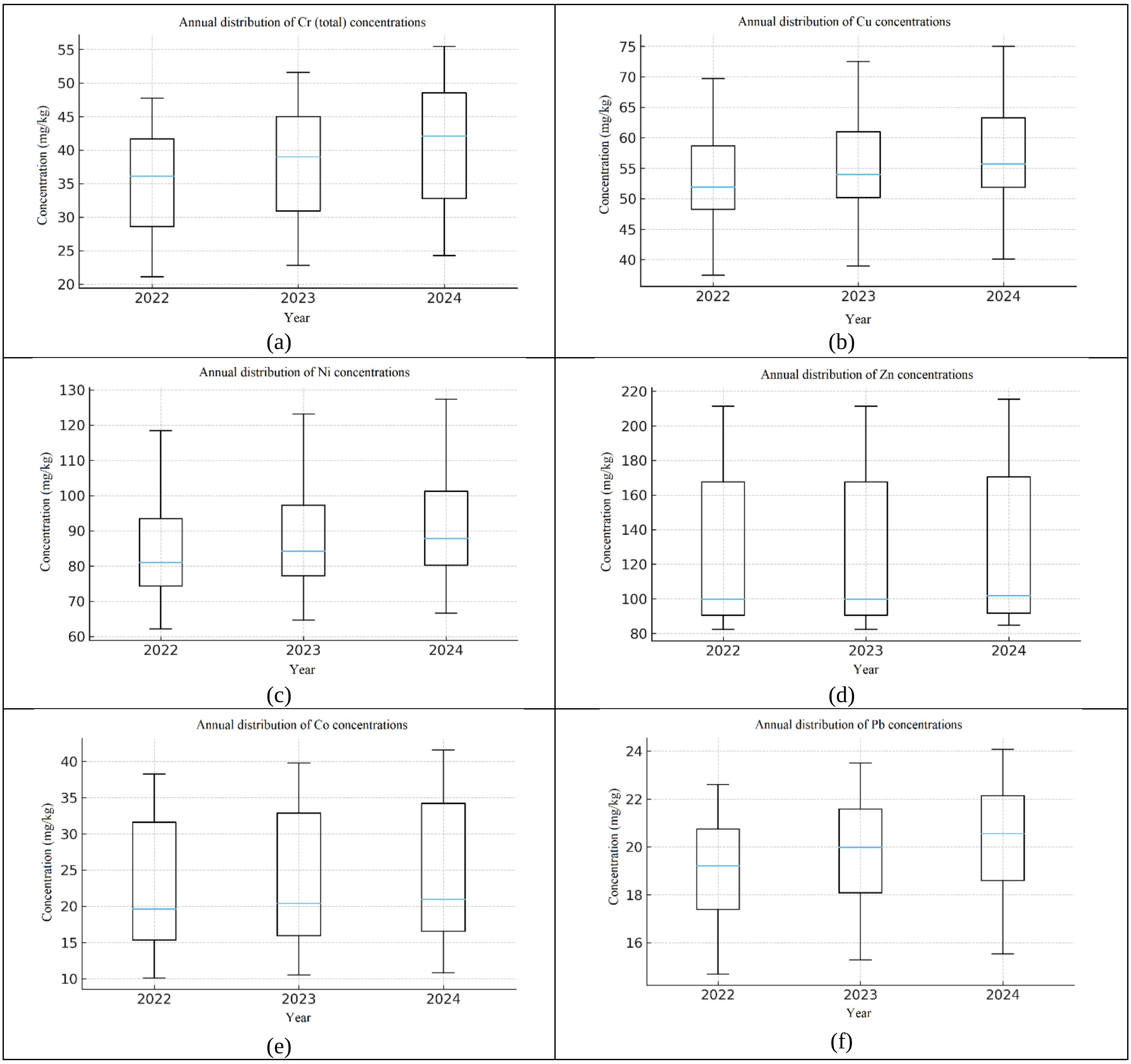

Fig. 3. Annual distribution of heavy-metal concentrations: (a) Cr(total), (b) Cu, (c) Ni, (d) Zn, (e) Co, and (f) Pb

3.2. Geochemical parameters of Ramura 1, 2, 3 and 4 tailings dump

To support the interpretation of heavy metal evolution, several geochemical parameters essential for understanding mobilisation processes were measured annually. The values for pH, Eh, moisture, organic matter, electrical conductivity (EC) and soil temperature are summarised in Table 2.

Table 2. Geochemical parameters of Ramura 1, 2, 3 and 4 tailings dump during the 2022–2024 period

Analysed parameter	**Unit of measurement**	**Study years**		
		2022	**2023**	**2024**
pH	–	6.72	6.48	6.31
Eh	mV	312	328	342
Moisture	%	14.8	16.3	18.9
Organic matter	%	2,41	2.56	2.73
Electrical conductivity (EC)	μS/cm	482	515	549
Soil temperature	°C	12.3	12.8	13.1

The results indicate a progressively more acidic environment (pH 6.72 → 6.31) and increasingly oxidising conditions (Eh 312 → 342 mV). Moisture content and organic matter also exhibit gradual increases, consistent with the presence of nearby water bodies and partial revegetation of the dump surface.

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Electrical conductivity follows a similar upward trend, reflecting increasing dissolved ion content associated with ongoing mineral alteration.

3.3. Comparison of results with the threshold values established by Order 756/1997

To assess contamination levels, the measured metal concentrations were compared with the threshold values set by Order 756/1997 for soils classified under "less sensitive use" [14-16]. These reference values are presented in Table 3.

Table 3. Threshold values for heavy metals according to Order 756/1997 (land use category: less sensitive)

Element	Normal values (mg/kg)	Alert threshold (mg/kg)	Intervention (mg/kg)
Cr (total)	30	300	600
Cu	20	200	500
Ni	20	150	500
Zn	100	600	1500
Co	15	50	250
Pb	20	100	1000

None of the investigated metals exceed the intervention thresholds. However:

- Zn consistently exceeds the normal value (100 mg/kg), although it remains well below the alert threshold.
- Cr (total), Cu and Ni show increasing tendencies and are approaching their respective alert thresholds, though they remain within acceptable ranges for the analysed period.
- Co and Pb remain within the limits of normal to moderately elevated values.

These observations indicate a moderate but evolving level of geochemical loading within the Ramura 1–4 tailings dump.

4. Discussion

The evolution of heavy metal concentrations and the behaviour of geochemical parameters during the 2022–2024 period reveal a continuously transforming system in which oxidative processes, moisture dynamics and the specific mineralogy of carboniferous tailings control metal mobility [17–19]. The consistent annual increases in Cr (total), Cu, Ni, Co and Pb indicate that Ramura 1–4 tailings dump remains geochemically active, with progressive alteration of the waste material strengthening from year to year.

Chromium displays the most pronounced temporal increase, a behaviour strongly associated with redox-sensitive geochemical environments [20, 21]. The upward trend in Cr (total) corresponds with the rise in Eh values observed in the field, suggesting intensified oxidative reactions. Such conditions may favour the transformation of Cr (III) into more mobile and environmentally persistent forms. Although the present study quantifies only total chromium, the trend underscores the potential for enhanced chromium mobility in future years if oxidative conditions persist.

Copper and nickel show steady but moderate increases across the monitoring period. This behaviour is consistent with the slow disintegration of clay–carbonaceous material typical of tailings deposits in the Jiu Valley, where mineral weathering progressively releases metals retained within fine-grained matrices. In contrast, zinc exhibits minimal variation, suggesting low short-term mobility. This stability is likely due to the association of Zn with more resistant mineral phases or secondary precipitates, limiting its reactivity under present geochemical conditions.

The geochemical parameters measured in situ support the interpretation of metal variability. The gradual decrease in pH reflects the progressive acidification of the waste material, a trend well documented in the evolution of mining–impacted soils [22–24]. Simultaneously, the increase in Eh values confirms the development of an increasingly oxidising environment—conditions favourable for the mobilisation of redox-sensitive metals such as Cr and Mn. Moisture content also increased significantly in 2024, likely intensifying ionic transport and accelerating mineral alteration processes that facilitate the release of metals from the waste matrix. The slight rise in organic matter may create localised reductive microenvironments [25], but not to an extent that counterbalances the overarching oxidative trend observed across the dump surface.

Comparison of the measured concentrations with regulatory values defined by Order 756/1997 shows that none of the metals exceed intervention thresholds for soils classified as "less sensitive." However, several elements—particularly Zn—consistently exceed normal values, and others such as Cu, Ni and Cr are

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gradually approaching alert thresholds. These trends highlight a moderate but evolving contamination pattern that warrants continuous monitoring. The joint evolution of decreasing pH and increasing Eh further suggests that conditions favouring metal mobilisation may intensify over time.

Overall, the results indicate that Ramura 1–4 tailings dump is undergoing active geochemical evolution driven by the mineralogical composition of the waste material and the influence of local climatic and hydrological conditions. While no immediate high-risk scenarios are indicated by current values, the observed trajectories point to the possibility of increased metal mobility over the long term. These findings reinforce the importance of sustained multi-year monitoring to anticipate future shifts in geochemical behaviour and to support effective environmental management and post-mining land-use planning in the Jiu Valley.

5. Conclusions

The multi-year monitoring of Ramura 1–4 tailings dump demonstrates that the deposit remains an evolving geochemical system in which oxidation processes, moisture dynamics, and mineralogical alteration progressively influence heavy-metal mobility. Between 2022 and 2024, Cr (total), Cu, Ni, Co, and Pb exhibited consistent upward trends, whereas Zn maintained the highest overall concentrations but showed only limited interannual variation. These trends align with the gradual acidification (pH 6.72 → 6.31) and increasingly oxidising conditions (Eh 312 → 342 mV), both of which are known to enhance the release and transport of redox-sensitive metals from carboniferous waste materials.

Although none of the analysed metals exceeded the intervention thresholds established by Order 756/1997, several elements surpassed normal values or have begun progressing toward alert thresholds, indicating a moderate but evolving contamination pattern. The combined trajectories of decreasing pH, increasing Eh, and rising electrical conductivity suggest an intensification of geochemical processes capable of enhancing metal mobility in the medium and long term.

These findings confirm that Ramura 1–4 tailings dump remains geochemically reactive and unlikely to stabilise naturally in the near future. Continuous, long-term monitoring is therefore essential for tracking the evolution of metal mobility, identifying early-warning signals of environmental risk, and supporting the development of science-based post-mining management and rehabilitation strategies. The dataset produced in this study provides a valuable baseline for future assessments and contributes to a better understanding of the geochemical behaviour of carboniferous tailings in the Jiu Valley.

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