

Article

Restoration Potential of Vegetation: Soil Nutrient Responses and Heavy Metal Distribution in Coal Mine Tailings

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Abstract

Coal mining leaves behind extensive tailing dumps that pose long-term ecological and soil degradation challenges. This study evaluates the restoration potential of vegetation on coal mine tailings in the Jiu Valley, Romania, focusing on soil nutrient dynamics and the heavy metal distribution. Field sampling was conducted across three vegetation types—unvegetated (UV), herbaceous (HV), and arboreal (AV, *Robinia pseudoacacia*)—at two intervals: three and six years post-plantation. Soil samples were analyzed for their pH, organic carbon, macronutrients, micronutrients, and heavy metals using standardized spectrometric and titrimetric methods. Between 2021 and 2024, AV plots showed a 9.5% increase in total nitrogen and a 5.2% rise in organic carbon, alongside a 6.9% reduction in soil pH. HV plots exhibited a 10.6% increase in magnesium availability and a 2.8% decrease in copper concentrations, indicating active nutrient cycling. In contrast, UV soils retained 68% higher total potassium and 24% more zinc than vegetated plots, likely due to limited biological uptake. Lead concentrations remained below the EU threshold of 60 mg kg^{−1}, while nickel levels exceeded recommended limits across all variants, peaking at 76.08 mg kg^{−1}. The vegetation type significantly influenced nutrient mobilization and metal stabilization, with arboreal cover demonstrating the most consistent ameliorative effects. These findings underscore the role of targeted revegetation—particularly with *Robinia pseudoacacia*—in improving soil quality and mitigating ecological risks in post-mining landscapes.

Keywords: soil; nutrients; heavy metals; metals; revegetation/vegetation; mine waste/tailings



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1. Introduction

Coal mining has long driven industrial growth, yet its legacy—waste dumps and tailings—continues to degrade ecosystems and soil health well beyond site closure. With sustainability now a global priority, restoring these landscapes has become increasingly urgent [1]. Surface mining severely disrupts the soil structure and microbial communities, impairing nutrient cycling and ecological resilience. Effective reclamation must rebuild soil fertility and biodiversity to support future land use [2]. Beyond ecological recovery, the integration of tailing dumps into agricultural use offers a dual benefit: mitigating mining’s environmental impact while promoting sustainable development with lasting economic, ecological, and social gains.

In Romania, mining activities have affected roughly 17,000 hectares, leaving behind 137 waste dumps and six tailing ponds containing nearly 2 billion cubic meters of residual

material. Yet only a fraction—about 2000 hectares—has been reclaimed. The Jiu Valley region exemplifies both the scale of the degradation and the promise of restoration, with 64 dumps covering over 250 hectares and holding approximately 37 million cubic meters of waste [3]. Sites like the dormant E.M. Uricani dump underscore the pressing need for ecological rehabilitation, especially as Romania advances toward sustainable land use and a green economy [4,5].

Tailing dumps are typically marked by a poor structure, low biological activity, and severe nutrient deficiencies, despite containing macronutrients (N, P, K, Ca, and Mg) and micronutrients (Fe, Cu, Mn, Mo, and Zn) alongside heavy metals that complicate reclamation efforts [6]. Their coarse texture, high bulk density, and limited water infiltration hinder natural regeneration, which can take several generations—nitrogen levels alone may require up to 200 years to match those of native forest soils [7]. Reclaimed soils often lose up to 80% of their organic carbon and 50% of their total nitrogen compared to undisturbed counterparts [8], though long-term studies show that soil properties can improve over time, following polynomial recovery trajectories influenced by the substrate composition, topography, micro-climate, and reclamation strategy [9–11]. Technical reclamation typically involves land reshaping, an amendment application, and vegetation establishment to stabilize substrates and initiate ecological succession [12]. Vegetation enhances soil development through root growth, litter deposition, and exudates that stimulate microbial communities essential for nutrient cycling and detoxification [13,14]. *Robinia pseudoacacia*, a leguminous species widely used in post-mining restoration, combines a nitrogen-fixing capacity [9,15] with a high tolerance to poor and metal-contaminated soils [16]. Its symbiosis with *Rhizobium* bacteria and arbuscular mycorrhizal fungi (AMF) enhances nutrient availability and immobilizes heavy metals, reducing toxicity and promoting plant establishment [16]. Fast-growing and deep-rooted *R. pseudoacacia* improves the soil structure, boosts organic matter and phosphorus mobilization [17,18], and contributes to carbon sequestration. Field studies confirm its role in enhancing soil fertility, biodiversity, and reducing metal bioavailability in reclaimed coal mining areas [19]. While spontaneous succession supports long-term ecological recovery through biodiversity and the natural attenuation of contaminants [20,21], *R. pseudoacacia* offers a rapid and effective strategy for initiating vegetation cover and accelerating soil rehabilitation.

To evaluate the agricultural viability of reclaimed tailings, detailed soil chemical analyses are essential for tracking improvements in fertility, structure, and contaminant attenuation. Long-term revegetation efforts in Transylvania have demonstrated measurable gains in soil quality [22], and broader studies confirm that both spontaneous and targeted vegetation enhance organic matter and nutrient cycling and reduce toxic metal bioavailability [12,15]. Misebo et al. (2021) [23] showed that the vegetation type influences the bulk density and water retention, with grasses and forbs improving the topsoil and trees benefiting deeper layers. Hu et al. (2020) [24] found that *Elymus nutans* restoration increased organic matter and nitrogen while lowering the pH and stabilizing cadmium, chromium, and lead. In tropical settings, Saidy et al. (2024) [25] reported that vegetation combined with organic amendments reduced exchangeable aluminum and improved nutrient retention, supporting plant growth and limiting metal uptake.

The aim of this study is to evaluate the effectiveness of vegetation in restoring coal mine tailings, with a focus on the soil nutrient dynamics and heavy metal distribution. By comparing soils without vegetation, soils with spontaneous herbaceous cover, and soils planted with *Robinia pseudoacacia*, this research investigates how the vegetation type influences the substrate quality over time. Soil parameters were assessed at two key intervals—3 and 6 years after plantation—to capture the early and intermediate stages of amelioration. Through integrated field sampling, laboratory analysis, and ecological

evaluation, this study seeks to identify viable strategies for transforming degraded mining landscapes into ecologically stable and productive environments.

2. Materials and Methods

2.1. Experimental Site

The experimental site, presented in Figure 1 and located at GPS coordinates $45^{\circ}19'50.93''$ N, $23^{\circ}9'6.08''$ E, is situated on the waste dump of the Uricani mine in the Jiu Valley, Hunedoara County, Romania, characterized by an annual precipitation level of 878 mm and an annual average temperature of 6.6°C .

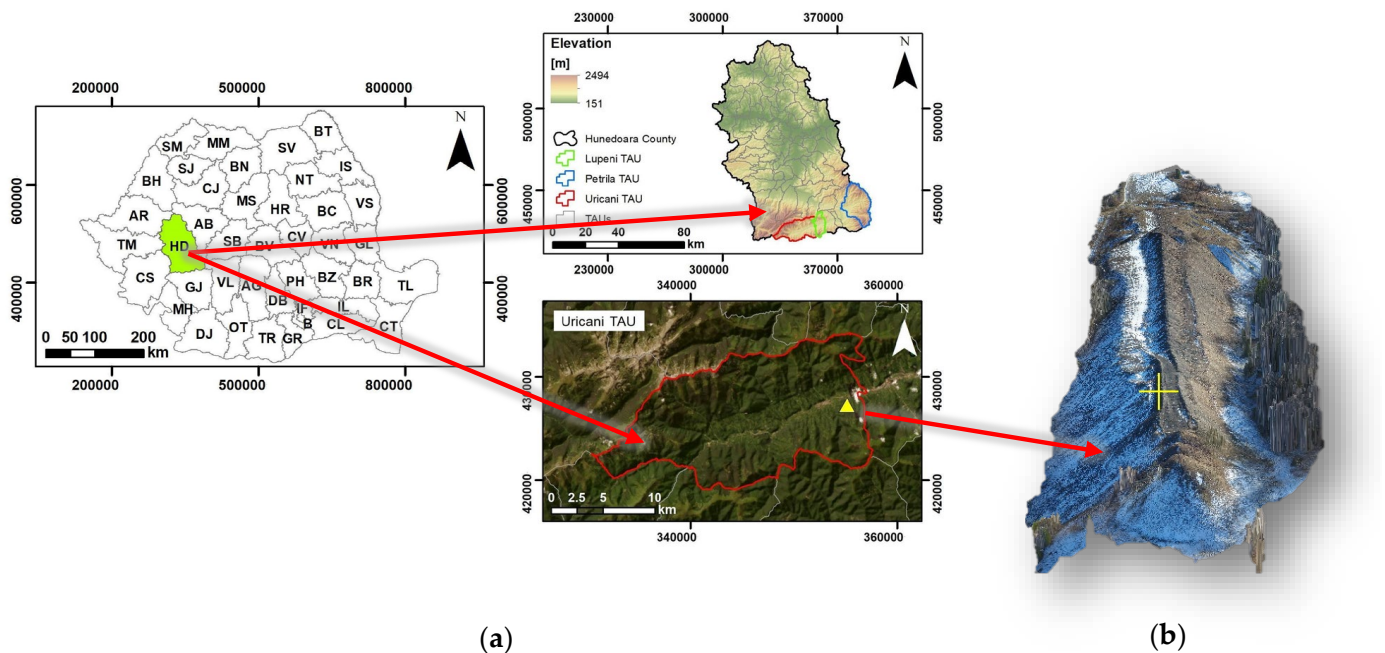


Figure 1. Experimental site (a) and dump before reclamation (b).

The dump spans approximately $27,000\text{ m}^2$ and contains an estimated $550,000\text{ m}^3$ of nutrient-poor material, with a mineralogical composition dominated by quartz (50%) and a mixture of potassium feldspar, biotite, and calcite (50%) [26]. In 2018, an ecological rehabilitation program was initiated, involving the afforestation of part of the dump with *Robinia pseudoacacia*. As a result, the site now comprises three distinct vegetation zones: (i) areas afforested with *Robinia pseudoacacia* (arboreal vegetation, AV), (ii) areas colonized by spontaneous herbaceous vegetation (HV), and (iii) barren, unvegetated surfaces (UV), which served as the control. These zones represent the experimental variants illustrated in Figure 2.

2.2. Soil Sampling

To assess the impact of vegetation on soil quality, sampling was conducted along a diagonal transect across all three variants at two time points: in 2021 (three years post-afforestation) and in 2024 (six years post-afforestation). These intervals were selected based on evidence that biochemical and structural changes in mine substrates emerge gradually through sustained plant–soil interactions [27]. Early improvements in total nitrogen and organic matter typically occur around year 3, while by year 6, cumulative effects become more pronounced, including enhanced base cation levels, increased phosphorus and potassium availability, and improved soil structure [28].

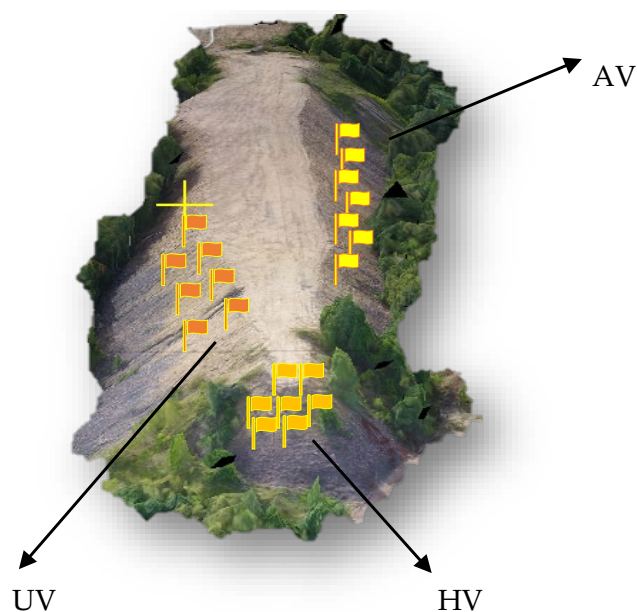


Figure 2. Experimental variants: UV—unvegetated, HV—herbaceous vegetation, and AV—arbooreal vegetation.

At each vegetation zone, composite samples were prepared from seven subsamples collected within a 10×10 m area at a depth of 0–25 cm, to ensure representativeness and minimize small-scale heterogeneity. Subsamples were placed in labeled polyethylene bags, air-dried at ambient temperature, and sieved to 2 mm prior to analysis. A total of 42 soil samples (7 samples per treatment $\times$ 2 time intervals $\times$ 3 vegetation types) were analyzed, with 16 physicochemical parameters determined for each sample. These included pH, organic carbon (OC), macronutrients (total nitrogen—TN, total potassium— K_T , total phosphorus— P_T , available potassium— K_{AL} , available phosphorus— P_{AL} , available calcium— Ca_{av} , and available magnesium— Mg_{av}), pseudo-total concentrations of micronutrients (manganese—Mn, zinc—Zn, iron—Fe, and copper—Cu), and pseudo-total concentrations of heavy metals (nickel—Ni, lead—Pb, and chromium—Cr). In total, 672 individual determinations were performed.

2.3. Soil Analysis

2.3.1. pH Determination

Soil pH was determined using a 1:2.5 soil-to-water ratio. Measurements were performed with a Mettler Toledo digital pH meter (Wien, Austria) equipped with a combined glass electrode, previously calibrated with buffer solutions at pH 4, 7, and 10 (analytical grade, Merck, Bucharest, Romania).

2.3.2. Determination of Organic Carbon (OC) Content

Organic carbon (OC) content was determined using the Walkley–Black wet oxidation method. Briefly, 1 g of air-dried and sieved soil is treated with 10 mL of $K_2Cr_2O_7$ 1 N, 20 mL of concentrated H_2SO_4 (Merk grade), and 200 mL of distilled water. After the completion of the reaction, 3–4 drops of o-phenanthroline indicator are added, and the excess $K_2Cr_2O_7$ is back-titrated with a ferrous ammonium sulphate solution 0.5 N. The difference between the initial and the remaining amount of $Cr_2O_7^{2-}$ provides an indirect measure of organic carbon.

2.3.3. Determination of Total Potassium K_T and Pseudo-Total Metal Content—Aqua Regia Extractable, Microelements (Mn, Zn, Fe, Cu), and Pseudo-Total Heavy Metals (Ni, Pb, Cr)

Pseudo-total metal content was determined using atomic absorption spectrometry (AAS) with a VARIAN 240FS (Palo Alto, CA, USA) spectrophotometer. A total of 0.5 g of the dried samples was subjected to mineralization with a 3:1 HCl:HNO₃ mixture in a TOPWAVE Analytic Jena microwave digester for 60 min at 360 °C to introduce the metal ions into solution. After cooling and filtration, the samples were moved to a 50 mL volumetric flask with deionized water. Merck-grade acids were used. For calibration, standard solutions were used, with a concentration ranging from 0.3 to 3 µg L⁻¹, which were prepared from multielement ICP Standard solution 1000 mg/L (Merck). Working conditions were as follows: air–acetylene ratio 13.50:2; nebulizer uptake rate—5 L min⁻¹; K (404 nm, 4 mA, 0.8 nm); Mn (279.5 nm, 4 mA, 0.5 nm); Fe (248.3 nm, 5 mA, 0.2 nm); Zn (213 nm, 5 mA, 1 nm); Cu (324.8 nm, 4 mA, 0.5 nm); Ni (232 nm, 4 mA, 0.2 nm); Pb (283.3 nm, 10 mA, 1.2 nm); and Cr (357.9 nm, 7 mA, 0.2 nm).

2.3.4. Determination of Total Phosphorus P_T Content

Total phosphorus was quantified from the 50 mL aqua regia digest (Section 2.3.3) by UV–Vis spectrophotometry using a Cintra spectrophotometer (GBC, Edinburgh, UK). An aliquot of the digest was reacted with an acidic solution of ammonium molybdate and stannous chloride, resulting in the formation of ammonium phosphomolybdate, which was subsequently measured calorimetrically at 715 nm. Calibration was achieved with standard solutions in the range of 2–60 µg 100 mL⁻¹, prepared from a 1000 mg L⁻¹ multielement ICP stock solution. All reagents employed were of analytical grade (Merck).

2.3.5. Determination of Available Phosphorus P_{AL} and Available Potassium K_{AL}

Available phosphorus (P_{AL}) and available potassium (K_{AL}) were determined by extracting 5 g of air-dried, sieved soil with 100 mL of ammonium acetate–lactate solution (pH 3.8 ± 0.05) under continuous shaking for 120 min to ensure transfer of the soluble fractions into solution. The suspension was subsequently filtered, and the extract was analyzed for P_{AL} using the Egner–Riehm–Domingo method, based on colorimetric measurement at 715 nm (Section 2.3.4).

Potassium availability (K_{AL}) was quantified in the same extract by atomic absorption spectrometry at 766 nm (Section 2.3.3).

2.3.6. Determination of Available Calcium (Ca_{av}) and Magnesium (Mg_{av})

Available Ca (Ca_{av}) and Mg (Mg_{av}) were determined in 0.1 M BaCl₂·2H₂O soil extracts by atomic absorption spectrometry—using VARIAN 240FS (Palo Alto, CA, USA) spectrophotometer. Soil samples (2 g, <2 mm) were repeatedly extracted with 20 mL of 0.1 M BaCl₂·2H₂O to obtain a final volume of ~100 mL. Analytical-grade reagents (Merck) were used. After filtration and volume adjustment, Ca_{av} and Mg_{av} concentrations were measured with a Varian 240 FS atomic absorption spectrophotometer (Palo Alto, CA, USA) under the following conditions: air–acetylene ratio = 13.50:2, nebulizer uptake = 5 mL min⁻¹, Ca (422.7 nm, 3 mA, 1.2 nm), and Mg (282.5 nm, 1.5 mA, 1.2 nm). Calibration employed standard solutions (0.3–3 mg L⁻¹) prepared from a 1000 mg L⁻¹ multielement ICP standard solution (Merck).

2.4. Statistical Analyses

Statistical analyses were carried out using Microsoft Excel and OriginPro 2025b. The effects of the presence of different types of vegetation, or their absence, were evaluated through analysis of variance (ANOVA) followed by Tukey test, with statistical significance set at $p < 0.05$. Relationships between macronutrients, micronutrients, heavy metals, and

soil parameters (pH, OC) were assessed by regression analysis. To reduce dimensionality and obtain a concise representation of the dataset, principal component analysis (PCA) with varimax rotation was applied.

3. Results

Table 1 summarizes the temporal dynamics of soil macronutrients across three vegetation types—arboreal (AV), herbaceous (HV), and unvegetated (UV)—over the monitoring period atop coal mine tailings.

Table 1. Dynamics of macronutrients in soil across distinct vegetation types—arboreal (AV), herbaceous (HV), and unvegetated (UV).

	Year	UV	AV	HV
TN (%)	2021	0.163 ± 0.031 ^{b3}	0.210 ± 0.020 ^{ab1}	0.250 ± 0.026 ^{a2}
	2024	0.170 ± 0.025 ^{B3}	0.230 ± 0.036 ^{AB1}	0.247 ± 0.050 ^{A2}
OC (g kg ^{−1})	2021	14.51 ± 0.40 ^{b3}	21.51 ± 0.81 ^{a1}	20.26 ± 0.71 ^{a2}
	2024	15.63 ± 0.40 ^{B3}	22.63 ± 0.25 ^{A1}	21.13 ± 0.90 ^{A2}
pH	2021	8.57 ± 0.036 ^{a4}	8.13 ± 0.072 ^{b1}	8.05 ± 0.062 ^{b2}
	2024	8.52 ± 0.093 ^{A4}	8.07 ± 0.0911 ^{B1}	7.89 ± 0.081 ^{B3}
P _T (mg kg ^{−1})	2021	393 ± 26.10 ^{a4}	448 ± 29.54 ^{a1}	314 ± 13.32 ^{b2}
	2024	432 ± 39.61 ^{AB4}	467 ± 26.35 ^{A1}	378 ± 10.26 ^{B3}
P _{AL} (mg kg ^{−1})	2021	0.226 ± 0.0235 ^{b3}	0.024 ± 0.0015 ^{a1}	0.013 ± 0.0015 ^{a2}
	2024	0.260 ± 0.0458 ^{B3}	0.029 ± 0.003 ^{A1}	0.017 ± 0.0021 ^{A2}
K _T (mg kg ^{−1})	2021	1025 ± 71 ^{c3}	635 ± 14 ^{a1}	520 ± 10 ^{b2}
	2024	1127 ± 31 ^{C3}	670 ± 19 ^{A1}	515 ± 14 ^{B2}
K _{AL} (mg kg ^{−1})	2021	140 ± 16.24 ^{a3}	149 ± 13.53 ^{a1}	137 ± 16.24 ^{a2}
	2024	144 ± 12.50 ^{A3}	166 ± 10.97 ^{A1}	160 ± 20.01 ^{A2}
Ca _{av} (mg kg ^{−1})	2021	1298 ± 47 ^{b3}	2854 ± 80 ^{a1}	1196 ± 91 ^{b2}
	2024	1563 ± 140 ^{B4}	3062 ± 159 ^{A1}	1347 ± 58 ^{B2}
Mg _{av} (mg kg ^{−1})	2021	323 ± 33 ^{b5}	758 ± 46 ^{a1}	682 ± 57 ^{a3}
	2024	503 ± 37 ^{B6}	869 ± 52 ^{A2}	810 ± 21 ^{A4}

Means that do not share a letter or a number are statistically significant difference at $p < 0.05$. Lowercase letters are used to denote statistically significant differences between groups within year 2021. Uppercase letter indicates statistically significant differences between groups within year 2024, and numbers indicate significant differences within the same group in different years. AV—arboreal vegetation; HV—herbaceous vegetation; and UV—unvegetated.

The soil pH remained predominantly alkaline across all plots in both years, with the vegetation type influencing its dynamics (Table 1). UV surfaces consistently showed the highest pH, while AV and HV plots exhibited greater temporal variation. Between 2021 and 2024, a statistically significant decline was observed in all plots, most notably in HV soils (−0.16 units), suggesting vegetation-driven acidification through biological activity.

Total nitrogen (TN) concentrations followed a stable vegetation-dependent pattern. AV and HV soils consistently showed higher TN levels than UV plots, with increases of 0.017 and 0.020 units, respectively, indicating active nitrogen cycling. The minimal change in UV soils (0.007 units) reflects the limited biological input in the absence of vegetation.

Parallel trends were observed in the organic carbon (OC) content, with the vegetation presence strongly influencing carbon accumulation. In 2021, HV soils exhibited the highest OC levels (21.51 g kg^{−1}), followed closely by AV soils (20.26 g kg^{−1}). UV soils remained significantly lower (14.51 g kg^{−1}), consistent with the minimal organic matter input. In 2024, both HV and AV variants sustained elevated OC concentrations (22.63 and

21.13 g kg⁻¹, respectively), while UV soils showed only a slight increase (15.63 g kg⁻¹), reinforcing the role of vegetation in enhancing soil carbon stocks through litter deposition and microbial activity.

Figure 3 captures notable temporal variations in the organic carbon to total nitrogen (OC/TN) ratio across different vegetation types, underscoring the distinct influence of plant cover on soil organic matter dynamics.

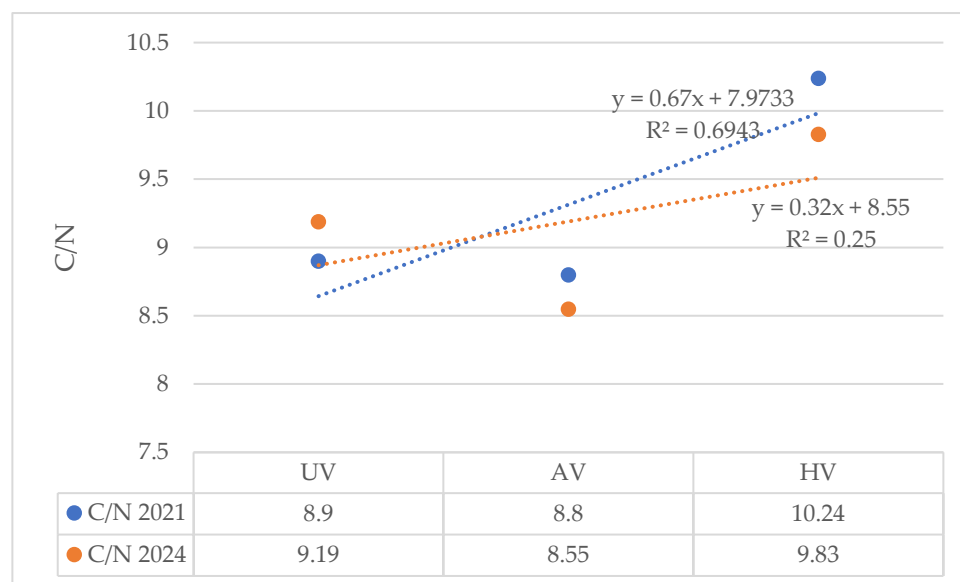


Figure 3. OC/TN ratio dynamics across vegetation types and years.

Across both sampling years, OC/TN ratios exhibited a positive linear trend with increasing vegetation complexity, consistently rising from UV to HV plots. This pattern highlights the influence of plant structural traits on the soil organic matter composition. In 2021, the regression model yielded a higher coefficient of determination ($R^2 = 0.6943$), indicating a strong predictive relationship between the vegetation type and OC/TN ratio. This suggests a relatively uniform litter quality and microbial processing across plots. In contrast, the 2024 dataset ($R^2 = 0.25$) showed a flatter slope and weaker correlation, pointing to increased variability in organic matter dynamics—potentially driven by climatic fluctuations, disturbance regimes, or transitional vegetation states. Plot-level comparisons further support this trend. HV plots consistently recorded the highest OC/TN ratios, reflecting carbon-rich, nitrogen-poor inputs typical of woody vegetation. These substrates are associated with slower decomposition rates, favoring carbon stabilization and long-term soil structural improvement. In contrast, UV plots exhibited the lowest ratios, indicative of nitrogen-rich, labile inputs that undergo rapid mineralization, enhancing short-term nutrient availability but limiting carbon retention.

The moderately alkaline pH observed across the study area provided a stable geochemical setting that supports mineral preservation while permitting biologically mediated nutrient transformations. Soils under arboreal vegetation (AV) consistently exhibited the highest concentrations of both exchangeable (P_{AL}) and total phosphorus (P_T) (Table 1), with values rising from 0.024 mg kg⁻¹ and 448 mg kg⁻¹ in 2021 to 0.029 mg kg⁻¹ and 467 mg kg⁻¹ in 2024. These elevated levels are likely attributable to deep rooting systems and substantial organic matter inputs from tree biomass, which enhance phosphorus mobilization even under alkaline conditions. Unexpectedly, unvegetated (UV) soils showed markedly higher levels of P_{AL} —0.226 mg kg⁻¹ in 2021 and 0.260 mg kg⁻¹ in 2024—alongside moderate P_T concentrations (403–432 mg kg⁻¹). This pattern may reflect surface-level accumulation and limited biological uptake due to the absence of vegetation.

Herbaceous vegetation (HV) plots recorded the lowest concentrations of both P_{AL} and P_T , indicating restricted phosphorus mobilization—likely due to a shallower root architecture and reduced organic inputs.

Potassium concentrations followed a similar trend, with AV soils showing an enrichment in both exchangeable and total forms. K_{AL} ranged from 149 to 166 mg kg^{-1} , while K_T varied between 635 and 670 mg kg^{-1} . Although K_T was slightly lower than in UV soils, the presence of vegetation suggests active nutrient cycling and uptake, promoting turnover rather than passive accumulation. K_{AL} in HV soils was marginally lower than that in AV plots, while total potassium remained comparable, suggesting minimal variation in long-term accumulation. In contrast, UV soils exhibited the highest K_T concentration in 2024 (1127 mg kg^{-1}), likely resulting from the continued weathering of potassium-bearing minerals such as feldspar, in the absence of biological export mechanisms.

As shown in Table 1, soils under arboreal vegetation (AV) exhibited exceptionally high available calcium (Ca_{av}) concentrations (2854–3062 mg kg^{-1}), indicative of accelerated calcite dissolution likely driven by biological activity. The magnesium availability in these soils was also elevated (758–869 mg kg^{-1}), suggesting active biotite weathering facilitated by root exudates and microbial interactions typical of tree-dominated systems. In contrast, soils under herbaceous vegetation (HV) displayed reduced calcium and magnesium levels relative to both AV and unvegetated (UV) soils, pointing to subdued mineral weathering and diminished nutrient retention. Mg_{av} concentrations in HV soils showed a modest increase in 2024 (810 mg kg^{-1}), potentially reflecting the seasonal variation or enhanced biotite decomposition during that period. Unvegetated soils presented intermediate Ca_{av} (1298–1563 mg kg^{-1}) and the lowest magnesium concentrations among all vegetation types (323–503 mg kg^{-1}). These findings suggest limited biotite degradation and poor nutrient retention dynamics in the absence of biological mediation.

Manganese concentrations ranged from 392 to 543 mg kg^{-1} (Figure 4a). In 2021, HV plots exhibited significantly higher Mn levels (543 mg kg^{-1} , $p < 0.05$) compared to AV and UV, a trend that persisted in 2024 (413 mg kg^{-1} in HV vs. lower values in UV). Statistically significant temporal declines were observed in HV (−130 mg kg^{-1}) and AV (−23 mg kg^{-1}), while the slight increase in UV (+6 mg kg^{-1}) was not significant. These findings highlight the role of herbaceous vegetation in Mn mobilization and its sensitivity to temporal shifts.

Pseudo-total Zn concentrations ranged from 127.03 to 161.20 mg kg^{-1} (Figure 4b), with peak values recorded in HV (2021) and UV (2024). Although AV and UV plots showed slight increases and HV showed a reduction (−11.94 mg kg^{-1}) over time, no statistically significant differences were observed between vegetation types or across years. These results indicate Zn stability under the tested conditions.

Iron concentrations ranged from 2144 to 2508 mg kg^{-1} (Figure 4c). AV plots consistently showed the highest Fe levels in both years, likely due to interactions between deep root systems and Fe-bearing minerals such as biotite. UV plots exhibited the lowest concentrations, reflecting reduced stabilization in the absence of vegetation. Although all variants showed declines from 2021 to 2024—most notably in UV (−106 mg kg^{-1})—none of these changes were statistically significant. These results suggest that Fe dynamics remained relatively unaffected by the vegetation type or sampling year.

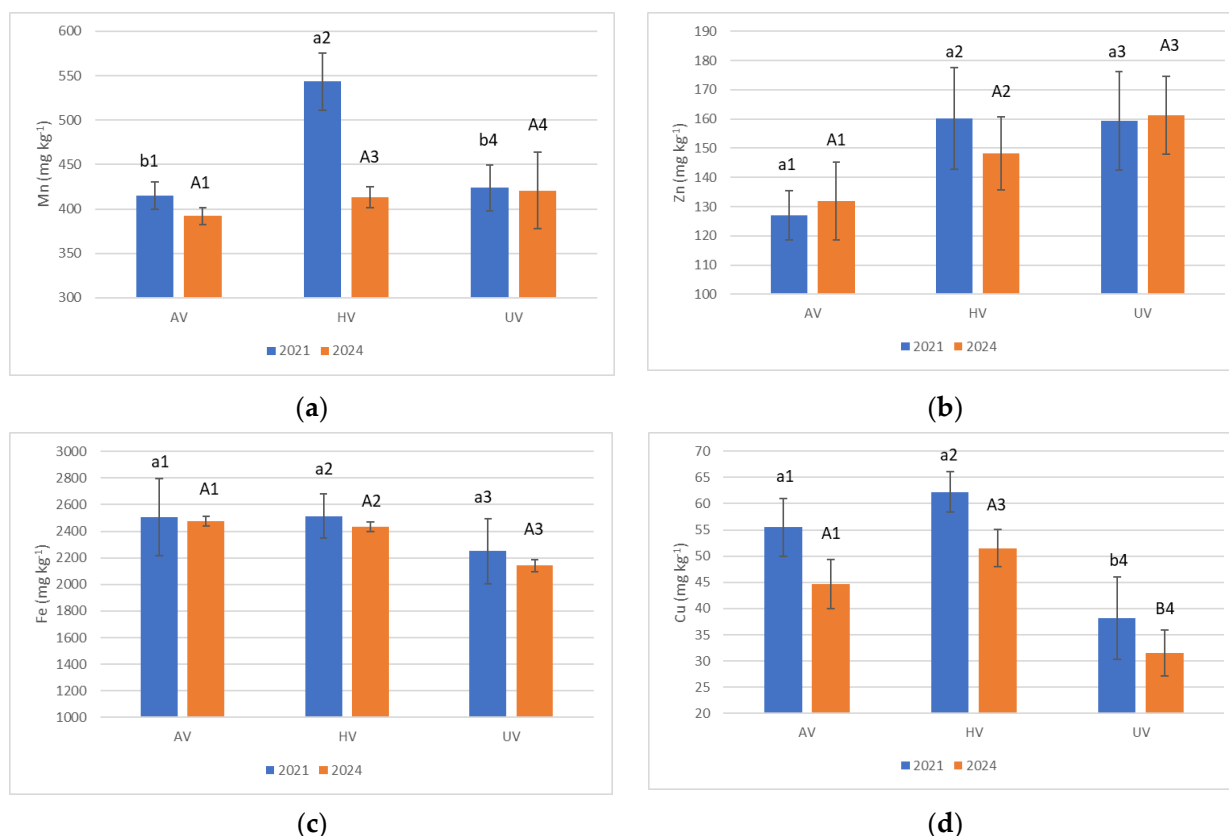


Figure 4. Microelement content: Mn (a), Zn (b), Fe (c), and Cu (d). Means that do not share a letter or a number are statistically significantly different at $p < 0.05$. Lowercase letters are used to denote statistically significant differences between groups within year 2021. Uppercase letter indicates statistically significant differences between groups within year 2024, and numbers indicate significant differences within the same group in different years. AV—arboreal vegetation; HV—herbaceous vegetation; and UV—unvegetated.

Pseudo-total copper (Cu) concentrations (Figure 4d) were generally higher in 2021 than in 2024. HV plots consistently exhibited the highest Cu levels in both years, significantly exceeding those in UV plots ($p < 0.05$), reflecting active Cu cycling associated with dense root systems and rapid organic matter turnover. Statistically significant temporal reductions were observed in HV ($-10.66 \text{ mg kg}^{-1}$) and AV, while UV maintained the lowest concentrations throughout, reaching a minimum of 31.52 mg kg^{-1} in 2024. These findings underscore the influence of the vegetation type on Cu dynamics and the element's susceptibility to leaching or biological uptake over time.

Pseudo-total lead (Pb) concentrations (Figure 4) exhibited year-to-year variation across vegetation types, with statistically significant reductions observed in HV soils ($p < 0.05$) from 2021 to 2024. In contrast, Pb levels increased in AV and UV plots over the same period, though these changes were not statistically significant ($p > 0.05$), indicating a limited influence of the vegetation type on Pb accumulation dynamics in those variants.

Ni and Cr concentrations generally rose in AV soils, while HV and UV plots exhibited slight declines or stabilization. These shifts suggest that vegetation cover influences metal retention and mobility over time.

To contextualize the heavy metal concentrations reported in Figure 5, soil data from 2021 to 2024 were compared against European agricultural soil thresholds and guideline values established by Tóth et al. (2016) [29]. Across all samples, Pb concentrations ranged from 16.47 to 34.56 mg kg^{-1} , remaining below the EU threshold of 60 mg kg^{-1} and well under both the lower (200 mg kg^{-1}) and upper (750 mg kg^{-1}) guideline values.

These findings suggest a minimal immediate risk to food safety. However, the elevated Pb levels observed in HV plots—particularly the 2021 peak—may warrant continued monitoring due to potential seasonal accumulation and root-zone interactions. Nickel concentrations, ranging from 76.08 to 94.62 mg kg⁻¹, exceeded the EU threshold of 50 mg kg⁻¹ in all variants. Several samples, especially those from HV and UV plots, approached the ecological risk guideline of 100 mg kg⁻¹. These elevated values raise concerns regarding potential phytotoxicity and the risk of leaching into groundwater, particularly in areas lacking deep-rooted vegetation that could buffer metal mobility. Chromium concentrations varied between 79.17 and 103.37 mg kg⁻¹, hovering near the EU threshold of 100 mg kg⁻¹. UV soils slightly exceeded this benchmark in 2024, suggesting enhanced surface accumulation in the absence of vegetative stabilization. Although Cr levels remained below the ecological risk guideline of 200 mg kg⁻¹, their proximity to critical thresholds—combined with the potential toxicity of hexavalent Cr—calls for cautious interpretation and further investigation. Both metals pose multiple ecological risks. Ni and Cr can be absorbed by plant roots, especially under acidic conditions or in species with high metal affinity, leading to impaired photosynthesis, root development, and nutrient uptake in sensitive vegetation [30]. In HV plots, deeper-rooted species may sequester metals in belowground biomass, potentially reducing mobility but increasing the risk of trophic transfer if contaminated tissues are consumed. UV plots, characterized by sparse root systems and limited evapotranspiration, offer a minimal buffering capacity, increasing the likelihood of Ni leaching into groundwater—particularly under fluctuating redox conditions or intense rainfall [31]. Cr(VI), though less common in natural soils, remains highly mobile and toxic, and its formation under oxidizing conditions cannot be excluded. Elevated Ni and Cr concentrations may also disrupt microbial enzymatic activity, inhibiting organic matter turnover and nitrogen cycling [32]. This microbial suppression could compromise soil fertility and delay restoration progress, especially in plots with a low organic carbon content and limited buffering capacity.

The Pearson correlation analysis revealed consistent patterns across both years, highlighting strong interdependencies among soil chemical properties. In 2021 (Figure 6), organic carbon (OC) showed strong positive correlations with TN ($R = 0.91$), P_{AL} ($R = 0.88$), and Ca_{av} ($R = 0.85$), indicating that nutrient accumulation is closely linked to organic matter enrichment. Soil pH was positively correlated with Ca_{av} ($R = 0.76$) and Mg_{av} ($R = 0.72$), suggesting that higher pH levels favor the availability of base cations. In contrast, heavy metals such as lead (Pb), nickel (Ni), and chromium (Cr) exhibited moderate to strong negative correlations with pH ($R = -0.67$, -0.70 , and -0.74 , respectively), implying increased metal mobility under acidic conditions. Notably, Pb also showed negative associations with OC ($R = -0.62$) and TN ($R = -0.59$), reinforcing the antagonistic relationship between a heavy metal presence and soil fertility indicators.

By 2024, the Pearson correlation analysis, presented in Figure 7, revealed strong interrelationships among soil chemical parameters, indicating early signs of substrate transformation. Organic carbon (OC) exhibited a robust positive correlation with total nitrogen (TN, $R = 0.93$), available phosphorus (P_{AL} , $R = 0.89$), and exchangeable calcium (Ca_{av} , $R = 0.86$), suggesting that organic matter accumulation is tightly linked to nutrient availability. The soil pH showed moderate positive correlations with Ca_{av} ($R = 0.74$) and Mg_{av} ($R = 0.71$), while negatively correlating with P_{AL} ($R = -0.62$) and heavy metals such as Fe ($R = -0.69$), Ni ($R = -0.66$), and Cr ($R = -0.68$), indicating that a lower pH may enhance metal solubility and limit phosphorus retention. Notably, Pb and Cr displayed negative associations with OC ($R = -0.61$ and -0.64 , respectively), reinforcing the antagonistic relationship between the heavy metal presence and organic matter buildup. These early-

stage correlations highlight the role of vegetation and organic inputs in improving soil fertility and buffering contaminant mobility in degraded substrates.

The PCA was performed in order to explore the relationships among the measured soil chemical parameters and to identify the main factors contributing to their variability.

The PCA biplot for 2021 (Figure 8) reveals distinct relationships among soil chemical variables and vegetation treatments, with the first two principal components (PC1 and PC2) accounting for 80.46% of the total variance (PC1: 51.91%, PC2: 28.55%). PC1 captures the primary gradient in the soil composition, separating samples enriched with organic carbon (OC), total nitrogen (TN), copper (Cu), iron (Fe), and magnesium (Mg_{av})—predominantly associated with herbaceous (HV) and arboreal (AV) vegetation—from those characterized by elevated available phosphorus (PAL), potassium (KT), chromium (Cr), and a lower pH, which cluster with the unvegetated (UV) variant. This axis reflects a transition from biologically active, nutrient-rich soils to more acidic, P–K-dominated substrates with limited organic input. PC2 further distinguishes the vegetation types by contrasting AV samples, which show higher calcium (Ca_{av}) and total phosphorus (P_T), with HV samples associated with trace metals such as manganese (Mn), nickel (Ni), and lead (Pb). This component highlights differences in the metal accumulation and nutrient partitioning between deep-rooted arboreal systems and shallow-rooted herbaceous cover.

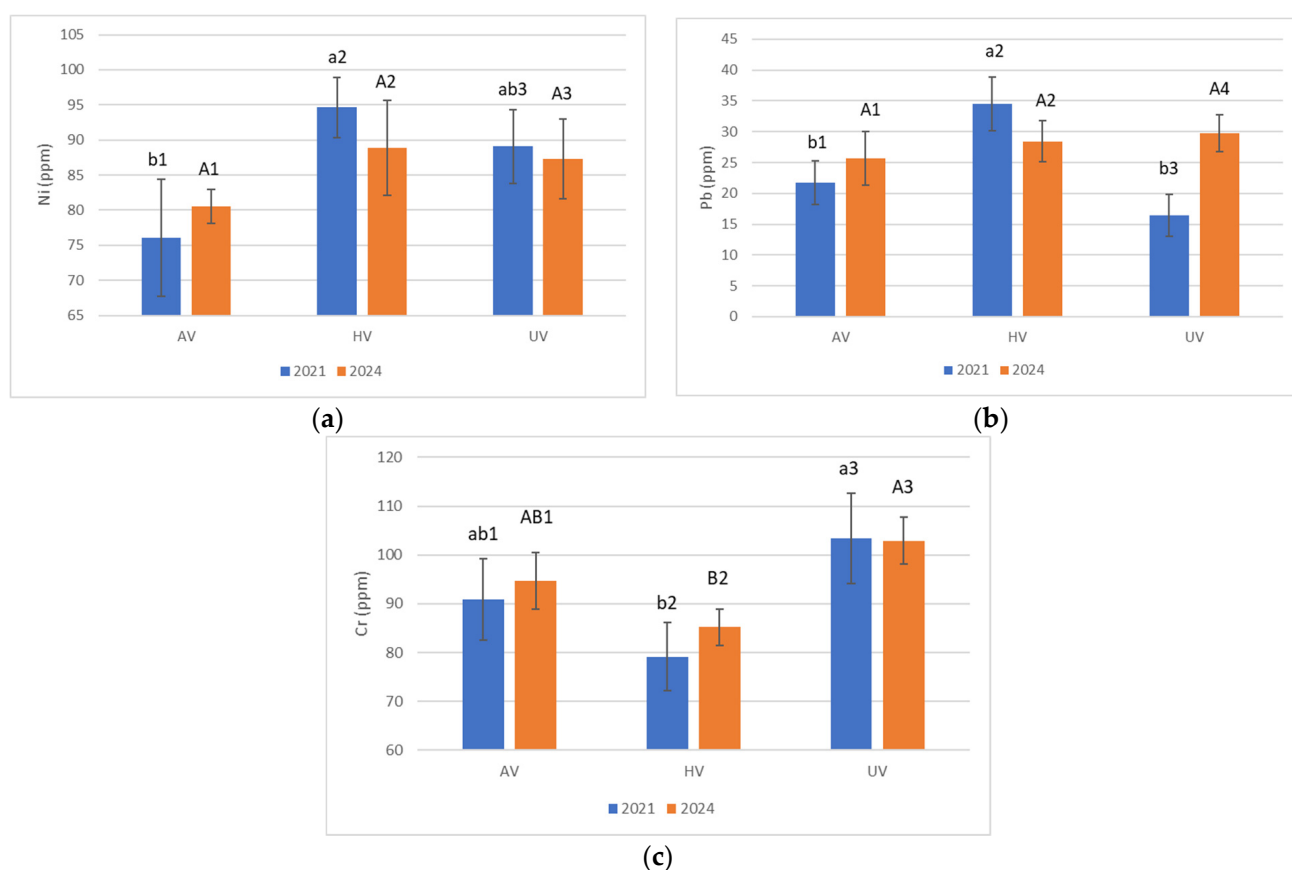


Figure 5. Variations in the pseudo-total heavy metal content of soil ((a)-Ni, (b)-Pb, (c)-Cr), covered with different types of vegetation—arboreal (AV), herbaceous (HV), and non-vegetative (UV). Means that do not share a letter or a number are statistically significant different at $p < 0.05$. Lowercase letters are used to denote statistically significant differences between groups within year 2021. Uppercase letter indicates statistically significant differences between groups within year 2024, and numbers indicate significant differences within the same group in different years.

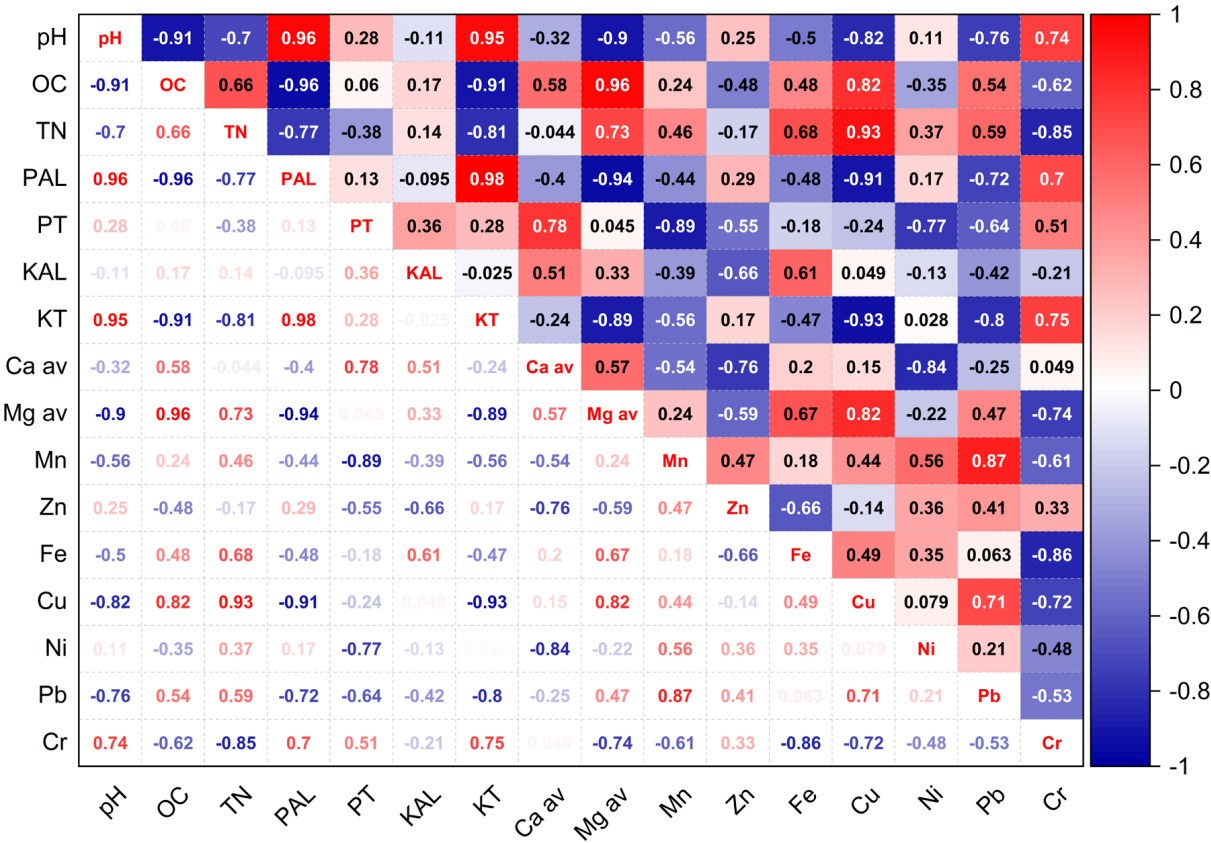


Figure 6. Pearson correlation matrix in 2021.

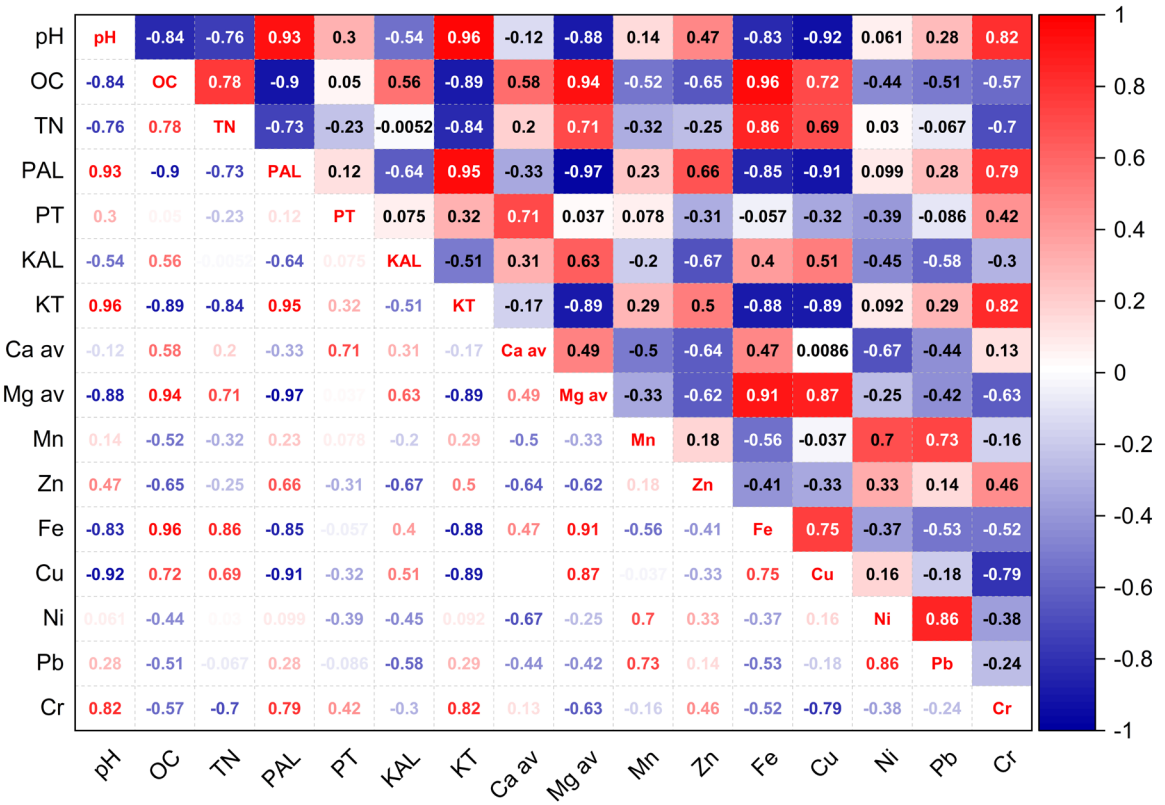


Figure 7. Pearson correlation 2024.

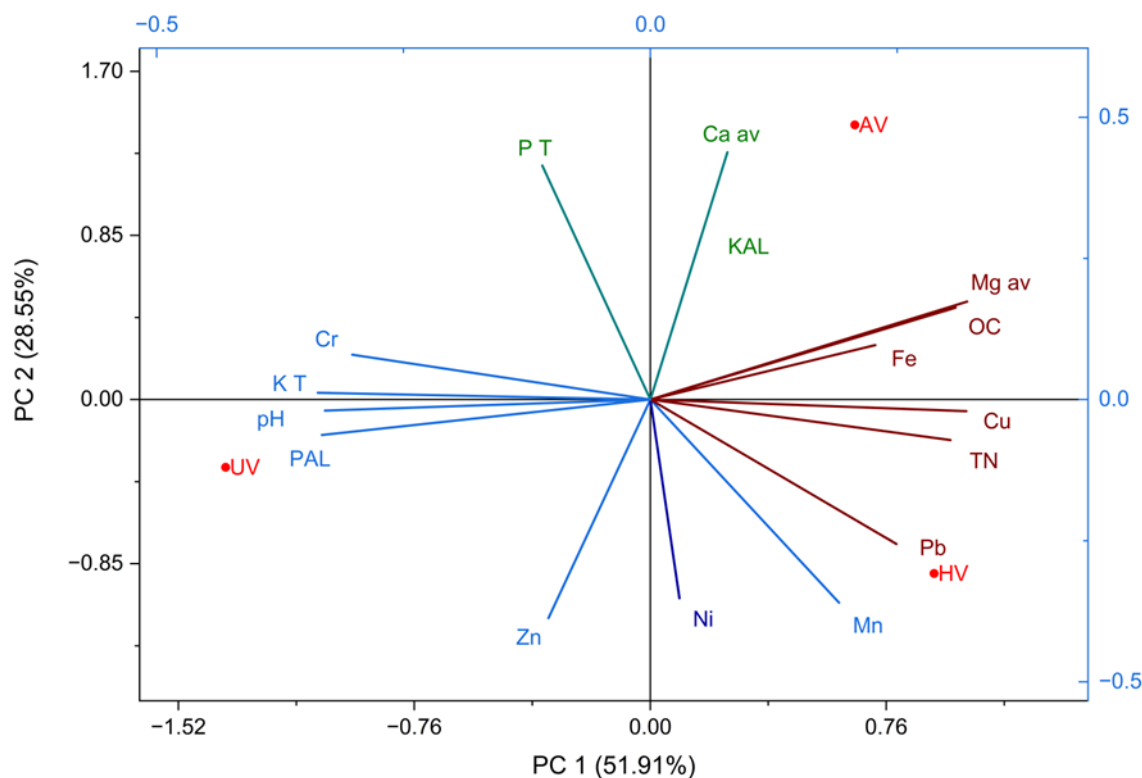


Figure 8. PCA biplot—2021.

The PCA biplot for 2024 (Figure 9) effectively captures the multivariate structure of soil chemical properties, with the first two principal components (PC1 and PC2) explaining 76.99% of the total variance (PC1: 54.33%, PC2: 22.66%). PC1 represents the dominant gradient in soil fertility. Samples positioned on the positive side of this axis—primarily AV and HV—are associated with elevated levels of organic carbon (OC), total nitrogen (TN), magnesium (Mg_{av}), copper (Cu), iron (Fe), and calcium availability (Ca_{av}), indicating nutrient-enriched conditions. In contrast, samples on the negative side—mainly UV—exhibit higher concentrations of available phosphorus (P_{AL}), potassium (K_T), chromium (Cr), and a lower pH, reflecting more acidic, P–K-dominated soils with limited organic input. PC2 further differentiates the samples based on nutrient and trace metal associations. AV samples are positively aligned with calcium and total phosphorus (P_T), while HV samples show stronger associations with manganese (Mn), nickel (Ni), and lead (Pb), suggesting distinct metal accumulation patterns linked to vegetation type. The distribution of samples along these two components highlights pronounced contrasts between vegetated (AV and HV) and unvegetated (UV) soils. Vegetated plots cluster in the nutrient-rich quadrant, while UV samples remain chemically distinct, characterized by lower fertility and elevated acidity.

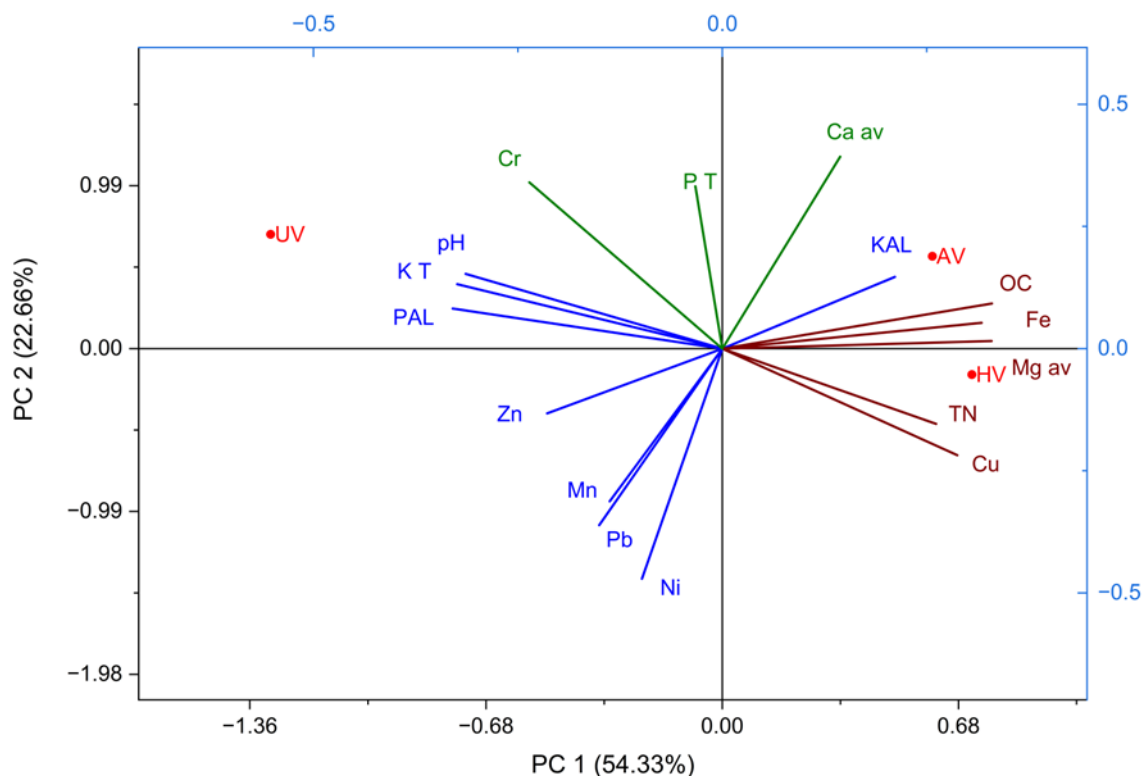


Figure 9. PCA biplot—2024.

4. Discussion

Across both sampling years, the soil pH remained alkaline due to the buffering influence of the calcite-rich coal mining waste. However, a gradual decline in pH over time—particularly in vegetated plots—signals an active biogeochemical transformation [33]. The herbaceous vegetation (HV) induced the most pronounced acidification, likely through fibrous root systems that stimulate microbial respiration and organic acid production. These findings align with studies conducted by Sun et al. (2019) [34], highlighting the acidifying role of root exudates and microbial turnover in reclaimed substrates. In contrast, unvegetated (UV) plots retained higher pH values, underscoring the dominance of abiotic buffering in the absence of biological mediation [22].

The vegetation type exerted a decisive influence on total nitrogen (TN) and organic carbon (OC) dynamics through root–soil–microbe interactions. Arboreal vegetation (AV) plots showed the highest TN levels, likely due to deep root systems and slower biomass turnover that promote nitrogen retention and microbial stabilization [35]. The herbaceous vegetation (HV) enhanced TN through rapid litter decomposition and active microbial cycling, consistent with early pedogenic processes such as the organic matter accumulation and microbial colonization observed between 2021 and 2024 [36,37]. Unvegetated (UV) plots remained nitrogen-deficient due to the lack of organic inputs and minimal microbial mediation, despite the buffering effect of the alkaline pH. Comparable limitations in nitrogen enrichment under biologically inactive conditions were also reported by Roman et al. (2018) [38] and Cântar et al. (2024) [39].

OC patterns mirrored TN trends, with HV plots leading due to dense fibrous roots and fast organic turnover, while AV plots supported gradual carbon stabilization via woody biomass and mycorrhizal associations. UV soils lacked organic inputs and microbial colonization, resulting in persistently low OC levels despite favorable pH conditions [40]. The concurrent rise in OC and TN across vegetated plots reflects advancing soil development

and aligns with reclamation trajectories described by Chan et al. (2014) [41], underscoring the role of vegetation in catalyzing biogeochemical recovery.

OC/TN ratio trends highlight how the vegetation structure governs organic matter turnover through plant inputs and microbial mediation. In 2021, steeper regression slopes and higher R^2 values reflected strong vegetation control over soil organic dynamics, while the increased variability in 2024 suggests evolving biotic–abiotic interactions. HV plots consistently showed elevated C/N ratios, driven by lignified, carbon-rich residues that resist decomposition and promote carbon stabilization—findings consistent with Nickels et al. (2021) [42]. AV plots exhibited intermediate ratios, indicating gradual organic accumulation via woody biomass. UV plots had the lowest ratios, dominated by nitrogen-rich, labile substrates that favor rapid mineralization and early nutrient release, as also observed by Sun et al. (2024) [43]. These gradients affirm the OC/TN ratio as a robust indicator of restoration progress, linking the vegetation type to soil fertility and resilience.

Phosphorus, potassium, calcium, and magnesium dynamics in reclaimed mining soils are shaped by the interplay between the mineral composition and vegetation-driven biological processes. In vegetated plots, the microbial activity and root exudation enhanced nutrient mobilization—particularly for phosphorus, calcium, and magnesium—while unvegetated (UV) soils showed nutrient persistence and surface accumulation due to limited biological uptake [44]. Arboreal vegetation (AV) plots exhibited the highest levels of bioavailable and total phosphorus, reflecting intensified microbial turnover and organic matter cycling, consistent with the findings of Wulandari et al. (2020) [45]. The elevated P_{AL} in UV soils likely resulted from surface accumulation in the absence of plant uptake, a pattern also observed by Więckol-Ryk et al. (2021) [6]. Rutkowska (2013) [46] highlighted biotite as a slow-release phosphorus source, supporting long-term fertility in mineral-rich substrates. Potassium concentrations peaked in UV soils due to the abiotic weathering of potassium feldspar, echoing Radulov and Goian (2004) [47]. In contrast, AV and HV plots maintained moderate K levels through active uptake and biotite-mediated exchange, with biotite contributing more readily to the exchangeable K pool than feldspar [48].

Calcium availability (Ca_{av}) was highest in AV soils, driven by calcite dissolution facilitated by root exudates and microbial acidification—mechanisms supported by Więckol-Ryk et al. (2021) [6] and Tomaszewicz et al. (2022) [49]. UV soils showed lower Ca_{av} , reflecting reduced carbonate breakdown in the absence of vegetation. Magnesium (Mg_{av}) concentrations followed a similar trend, with AV and HV plots showing elevated levels due to intensified biotite weathering and organic matter decomposition. These synergistic root–microbe interactions enhance Mg mobilization, as previously reported by Więckol-Ryk et al. (2021) [6] and supported by microbial–mineral coupling mechanisms described by [50].

In 2024, microelement concentrations declined in vegetated plots due to sustained plant uptake, microbial mediation, and the reduced geogenic input from tailing weathering. Vegetation acts as both a sink and stabilizer, modulating nutrient availability through root absorption, organic turnover, and rhizosphere interactions—mechanisms well-documented by Mendez and Maier (2007) [51] and Colombo et al. (2014) [52]. High annual precipitation ($\sim 878 \text{ mm year}^{-1}$) further intensified leaching and ion mobility, enhancing nutrient cycling in vegetated soils while promoting surface accumulation and erosion in unvegetated (UV) areas. The mineral matrix—dominated by quartz, K-feldspar, calcite, and biotite—offered limited nutrient release. Quartz and feldspar remained inert, while calcite buffered the pH and influenced metal solubility. Biotite, under biologically active conditions, contributed Fe and Mg via root exudate-driven weathering and microbial processes, as shown by Aznar-Sánchez et al. (2018) [53] and Yan et al. (2020) [54].

Manganese (Mn) peaked in HV plots, where fibrous roots and microbial mobilization promoted solubilization and uptake. AV plots showed moderate Mn levels due

to deeper root buffering, while UV soils remained Mn-poor, reflecting minimal biological mobilization—consistent with Tripathi et al. (2018), Fomina et al. (2005), Vink et al. (2010) [55–57]. The zinc (Zn) distribution varied spatially. HV plots maintained elevated Zn levels through rhizosphere acidification and active cycling; AV plots showed lower Zn levels due to canopy-mediated immobilization; and UV plots accumulated Zn via surface leaching and limited uptake. Aznar-Sánchez et al. (2018) [53] further noted Zn stabilization via sorption onto quartz and feldspar surfaces in unvegetated substrates.

Iron (Fe) concentrations were highest in AV plots, where deep roots accessed biotite-bound Fe, enhanced by organic exudates and microbial solubilization [52,54]. HV plots showed slightly lower Fe levels, likely due to competitive uptake, while UV soils retained minimal Fe due to biological inactivity. Copper (Cu) was lowest in UV soils, likely immobilized in non-bioavailable forms. AV plots showed intermediate Cu levels, stabilized by the root depth, and moderated uptake. HV plots recorded the highest Cu concentrations, driven by rhizosphere acidification and microbial cycling—findings aligned with McManus et al. (2018) [58].

Vegetation cover plays a pivotal role in microelement retention and heavy metal distribution through root–soil–microbe interactions [59,60]. Herbaceous species (HV) enhanced Mn, Zn, Cu, and Pb concentrations via dense root mats and rapid organic turnover, which stimulate microbial mobilization and rhizosphere acidification—mechanisms supported by Mendez and Maier (2007) [51], Tripathi et al. (2018) [55], and McManus et al. (2018) [58]. Arboreal vegetation (AV), with deeper root systems and mineral buffering capacity, stabilized Fe and reduced Pb and Cr levels, limiting erosion and metal redistribution, consistent with the phytoremediation findings of Pulford and Watson (2003) [61].

In contrast, unvegetated (UV) plots lacked biological uptake, resulting in greater metal mobility and surface accumulation, especially under hydrological stress. Between 2021 and 2024, pseudo-total concentrations of Pb, Ni, and Cr increased across all vegetation types, driven by geochemical weathering, seasonal variation, and possible anthropogenic inputs. High annual precipitation (~870 mm) likely facilitated leaching and vertical migration, particularly in UV soils with poor infiltration resistance. The substrate mineralogy further influenced metal behavior. Quartz and K-feldspar, being chemically inert and with a weak cation exchange capacity, contributed to metal mobility [62]. In contrast, biotite adsorbs metals via interlayer exchange, and calcite buffers pH and promotes metal precipitation as carbonates under alkaline conditions—mechanisms described by Kabata-Pendias & Mukherjee (2007) [63] and McBride (1994) [64].

Although pseudo-total measurements encompass both bioavailable and residual fractions, the environmental risk remains uncertain without speciation data. Nonetheless, observed concentrations approach or exceed thresholds that may impair plant growth and pose food safety risks through bioaccumulation [65,66]. Elevated Ni and Cr levels in HV and UV plots highlight emerging ecological concerns that warrant targeted investigation and remediation.

To address ecological risks posed by elevated Ni and Cr concentrations, integrated vegetation and soil management strategies are vital. Planting deep-rooted, metal-tolerant species like *Robinia pseudoacacia* and *Festuca* in HV and AV plots can enhance phytostabilization by immobilizing metals in the rhizosphere and limiting vertical migration. Complementary soil amendments—such as compost, biochar, or manure—have been shown by Vischetti et al. (2022) [31] to increase the cation exchange capacity and bind heavy metals, reducing their bioavailability and leaching potential. In UV plots, maintaining a near-neutral pH through liming is crucial, as acidification may intensify metal solubility and mobility. The ongoing monitoring of soil and plant tissue is recommended to track metal dynamics and detect early signs of bioaccumulation or leaching.

Pearson correlation matrices from 2021 and 2024 highlight vegetation as a key driver of nutrient cycling and soil chemical coherence. In unvegetated soils, weak and erratic correlations among nutrients and heavy metals reflected limited biological activity and poor substrate structures. The establishment of herbaceous cover introduced more consistent relationships—particularly among organic carbon, nitrogen, and macroelements—indicating early microbial stabilization and nutrient integration. *Robinia pseudoacacia*, with its nitrogen-fixing capacity and deep rooting, further strengthened positive correlations among organic matter, base cations, and nutrient availability, signaling enhanced soil development. Concurrently, stronger negative associations between pH and heavy metals in vegetated plots suggest improved buffering and reduced metal mobility. These evolving patterns affirm the role of vegetation—especially leguminous trees—in accelerating soil functional recovery and mitigating contamination risks, consistent with findings by Mukhopadhyay et al. (2016) [60].

Principal component analyses (PCAs) conducted in 2021 and 2024 consistently highlight the ecological influence of vegetation cover on soil chemical trajectories atop coal waste substrates. While both biplots share a similar structural framework, the evolution of the sample clustering and variable associations over time reveals the dynamic nature of soil–vegetation interactions. In both years, PC1 delineates a clear fertility gradient, separating nutrient-enriched soils—characterized by elevated organic carbon (OC), total nitrogen (TN), magnesium (Mg_{av}), copper (Cu), and iron (Fe)—from more acidic, phosphorus–potassium-dominated substrates with higher P_{AL} , K_T , and Cr levels and a lower pH. This axis consistently distinguishes vegetated plots (AV and HV) from unvegetated (UV) soils. PC2 captures the secondary variation, contrasting calcium and total phosphorus (P_T) enrichment in AV plots with trace metal accumulation (Mn, Ni, Pb) more closely associated with HV.

In 2021, the PCA captured the early-stage differentiation among treatments. UV samples clustered distinctly, reflecting the minimal biological influence and persistent acidity. HV plots aligned with elevated TN, Cu, and Pb, indicative of spontaneous colonization and active nutrient cycling. AV samples, associated with *Robinia pseudoacacia*, showed an emerging influence over Ca_{av} , P_T , and K_{AL} —suggesting initial nutrient accumulation driven by litter input, root exudation, and microbial stimulation. By 2024, the PCA reveals a more pronounced separation of AV samples from both the HV and UV, signifying cumulative improvements in soil quality under arboreal cover. The shift in the AV cluster positioning reflects the sustained enrichment in Ca_{av} and P_T , alongside the stabilization of trace metals. This progression underscores the transformative role of *Robinia pseudoacacia* in enhancing soil fertility and buffering contaminant mobility over time. Notably, the 2024 biplot displays sharper contrasts among sample groups, likely amplified by the documented decline in Mn and Cu concentrations. These shifts reinforce the distinction between active revegetation (AV) and passive colonization (HV), with AV exerting a more consistent and targeted influence on soil amelioration.

5. Conclusions

This study demonstrates the critical role of vegetation in improving the ecological resilience of coal mine tailings. Both arboreal and herbaceous cover significantly influenced soil chemistry—modifying pH, enhancing nutrient availability, and reducing heavy metal mobility. *Robinia pseudoacacia* notably increased calcium levels and maintained the highest phosphorus concentrations across both years, while the herbaceous vegetation showed lower phosphorus levels but contributed to the organic carbon enrichment and Mn and Cu availability. In contrast, unvegetated plots remained chemically unstable, with poor nutrient retention and elevated leaching risks. The principal component analysis revealed

progressive differentiation among vegetation types, indicating cumulative improvements in the soil quality and soil–plant interactions over time. Although lead concentrations remained within safe limits, elevated nickel and borderline chromium levels in unvegetated and herbaceous plots suggest potential ecological risks. Arboreal systems showed a greater capacity for contaminant immobilization and fertility enhancement, reinforcing their value in phytoremediation. These findings support restoration strategies that combine deep-rooted arboreal species (e.g., *Robinia pseudoacacia*) with fast-growing herbaceous cover to enhance nutrient cycling, organic matter input, and contaminant stabilization. Effective reclamation should prioritize native or adaptive species with complementary root systems and proven phytoremediation potential. To ensure long-term success, we recommend the biennial monitoring of key soil parameters (pH, OC, TN, P_{AL}, trace metals), alongside metal fractionation studies every 3–5 years to assess bioavailability. Integrating mineralogical assessments with ecological indicators will enable adaptive, site-specific management and promote sustainable reclamation practices.

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