

## Distribution of Heavy Metals in the Coal Mine Waste Dumps Based on Statistical Analysis

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**Summary:** At the UN Climate Change Conference in Glasgow in autumn 2021 (COP26), world leaders and participants decided to phase out coal energy and gradually replace fossil fuels. At COP26, Ukraine has committed to shutting down state-owned coal-fired power plants by 2035, reducing methane emissions by 30% by 2030 and halting deforestation. The waste dumps are also a detrimental factor in coal mining. The determination of the heavy metals content in the waste dumps of coal mines is a topical issue today, as the results of such studies are part of the monitoring of the environmental safety of mining regions. This scientific article considers the results of research on the heavy metals content in the waste dumps and their distribution in one of the largest coal mines in the Lviv-Volyn coal basin – ‘Chervonohradska’ (Ukraine). 2.9 million m<sup>3</sup> of rock have accumulated in the dump since 1971. Every year, 40,000 m<sup>3</sup> of fresh rock is dumped. Samples were taken uniformly from all sides of the dump in places with no vegetation cover. Note that exceeding the maximum allowable concentrations of the studied heavy metals is observed in all areas except for Zn. Statistical analysis of the results of semi-quantitative spectral studies of waste dumps rock at the Chervonohradska mine site was performed using the Statistica 8 applied statistics package. According to the results of more than 50 samples taken at a depth of 0.3 m, the statistical distribution of heavy metals Mn, Pb, Ni, Cu, Zn and Co in the mine dumps was determined, and a correlation analysis was performed. It was found that the distributions of Cu and Zn in dumps are closest to normal. The nonparametric Spearman coefficient (rs) revealed the average level of correlation of the content of heavy metals in the pairs Mn and Ni (rs = 0.46), Mn and Zn (rs = 0.52), Ni and Zn (rs = 0.57), Cu and Zn (rs = 0.49). Chaotic and uneven discharge of rock onto the open area caused uneven substrate deposition of certain chemical elements in the profile of the dump.

### Introduction

The coal industry causes a significant technogenic impact on the environment, affects human health and transforms living conditions regardless of the place or country. The most dangerous factor of environmental degradation is waste dumps. Investigation of

the impact of waste dumps on the human body and biota is conducted in almost all industrialized countries and countries where mineral mines are operated.

At the UN Climate Change Conference in Glasgow in the autumn of 2021 (COP26), world leaders and participants decided to phase out coal energy and gradually replace subsidized fossil fuels. As part of the event, more than 40 countries, including large coal consumers such as Poland, Vietnam and Chile, have agreed to abandon its use. The Glasgow Climate Pact calls on all countries to present more effective national action plans next year, rather than 2025, as previously envisaged. The countries reaffirmed the goal of the Paris Agreement is to keep global average temperatures well below 2 °C and make efforts to limit the temperature rise to 1.5 °C. At COP26, Ukraine has committed to shutting down state-owned coal power plants by 2035, reducing methane emissions by 30% by 2030 and halting deforestation.

Coal waste dump sites, despite their thermal activity, are spontaneously inhabited by vegetation, but how it gets and how it covers the dump depends largely on the shape of the site, the method of construction, exposure and development of surrounding areas, and thermal processes. The sites were dominated by ruderal and meadow species belonging to the classes *Artemisieta* and *Molinio-Arrhenatheretea*, which belong to the transformed biotopes. The investigation has shown that in the case of burning waste dumps, reclamation has no effect if the object is not properly protected from fire (Abramowicz et al. 2021). During self-heating, some chalcophilic elements, such as Hg (mainly present as HgS), Pb, Zn, can be enriched and released, or various organic pollutants such as phenols (derived from vitrinite particles), various PAHs with alkyl substitutes, chlorinated PAHs or sulfur form a heterocycles. Remote sensing techniques have helped to locate and control hotspots with different temperature ranges (Nadudvari et al. 2021).

Nowadays, there are many methods of measuring the temperature of waste heaps. The newest and most popular are pyrometric and remote methods. Fieldwork on the selected coal heap in the Upper Silesian Coal Basin was carried out using pyrometric (point measurements) and remote sensing (thermal imaging) (Abramowicz et al. 2019). Fieldwork has shown that aerial photography helps to predict the direction and speed of fire (Abramowicz et al. 2020).

Scientists (Kribek et al. 2021) found that at a temperature of + 500 °C in the solid residue of heap rock coke and semi-coke appeared. At temperatures <+ 500 °C, the amount of In, Sb, Tl, Zn, As, Mo, Sn, Pb, Se, Hg and Cd in the pyrolysis residue decreased. The melting of sulfides during pyrolysis may be one of the reasons for the high volatility of chalcophilic elements (Silveira et al. 2022).

Analysis of As and potentially toxic metals (Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb and Zn) in the surface soil of an old gold mining site in Meldon, Victoria, Australia was performed to determine the current metal concentration. The results revealed the average concentrations of metals from highest to lowest in the following order: Mn > Zn > As > Cr > Cu > Pb > Ni > Co > Hg > Cd. Site status was assessed directly by comparing the metal concentration in the study area with the known average soil levels in Australia

and Victoria and the sanitary study levels established by various organizations (Abraham et al. 2018).

The results of studies (Chen et al. 2021) showed that biocarbon media showed a markedly high ability to bind metals and neutralize acidity, support significantly better plant growth and mitigate the transfer of metal from plant roots to shoots. The addition of organic liquid waste (domestic wastewater and plant straw hydrolysis mixture) stimulated the recovery of nutrient sulfate after 40 days of adaptation to effectively remove many heavy metals from mountain wastewater.

The aim of the research of Woch et al. (2018) was to evaluate the relationships between vegetation, physicochemical and microbial properties of the substrate at the sites of coal ash dump and sludge. The properties of ash and sludge negatively affected microbial biomass and enzymatic activity, as evidenced by significant negative correlations between alkaline/alkaline earth metals, heavy metals and macronutrients with enzymatic activity and/or microbial biomass, and positive correlations of these parameters with metabolic coefficient ( $qCO_2$ ).

The research (Popovych et al. 2019a, 2019b; Bosak et al. 2020) considers the features of the Lviv-Volyn coal basin as a center of industrial development in Western Ukraine. Dumps of the mines have high acidity and a significant content of various salts and sulfate ions. The high mineralization of wastewater is due to the movement to water collection points and the interaction of such water with waste heaps.

In Kalybekov et al. (2020) the technological scheme of bulldozer dump formation is substantiated taking into account the suitability of rocks for reclamation purposes, which reduces the negative impact of mining on the environment. In (Linhares et al. 2019) a mathematical model of the process of pollutants filtering through the soil is developed and recommendations for environmentally friendly storage and processing of phosphogypsum waste are provided.

The analysis of the influence of mechanical equipment for reclamation works on the quality of the lands, which pertains to reproduction in the case of open mining operations was carried out by Terekhov et al. (2021). A methodical approach to the choice of mechanization of land reclamation works according to the quality of technogenic agricultural lands as a factor of their monetary value and the level of costs for their reproduction is proposed.

The results of research (Karimaei et al. 2020) have shown that untreated coal waste particles can potentially be reused in the production of concrete aggregates. It was also confirmed that choosing the right amount of replacement can help improve the properties of concrete and also offer an environmentally friendly solution for the reduction of untreated coal waste.

'Recommendations for substantiation of parameters of pond accumulative capacity recovery using hydro mechanization' and 'Methods for calculating parameters of hydro transportation of highly concentrated hydro mixtures' are developed (Blyuss et al. 2020), which can be useful for design organizations and mining and metallurgical enterprises to provide additional volumes of raw materials and increase the service life of tailings.

In Welch et al. (2021) five important elements that should be included in the study of best practice are declared. There are climatic conditions, physical characteristics of the dump, geochemical processes, water regime and environmental impact over time, as water emissions of COIs from coal dumps occur over decades or centuries. Key considerations have been identified for each of these elements to guide best practice.

The analysis of scientific sources on the study of other types of dumps in different climatic zones showed that all of them cause man-made pressure on the environment. Also, the results of research by scientists indicate the need for reclamation of disturbed areas.

Thus, the investigation of heavy metal content in the rock of coal mines is a topical issue today, as the results of such research are part of the monitoring of the environmental safety of mining regions. In this paper, we are to consider the content of heavy metals in the heap and their distribution in one of the largest coal mines in the Lviv-Volyn coal basin – Chervonohradska.

### **Materials and methods**

Samples of waste heaps edaphotops were taken following the State Standard of Ukraine (DSTU ISO 10381-1: 2004).

The research was carried out in the Research Laboratory (RL) of Environmental Safety, which operates at the Lviv State University of Life Safety (Lviv, Ukraine). Certificate of conformity of the measurement management system RA091/21 dated 30.11.2021, valid until 29.11.2026, issued by SE 'Lvivstandardmetrologiya'. Regulations on RL are developed based on the normative document: 'Procedure for voluntary assessment of the measurement management system. General requirements and procedure. COY 43.01-04725912-001.2016' (order of the State Enterprise 'Lvivstandardmetrologiya' dated 21.03.2016 No. 648). The premises and environment of the laboratory meet the sanitary norms, rules and requirements of labor protection. Also in the Central Research Laboratory and Laboratory of Industrial Toxicology of Lviv National Medical University named after Danylo Halytsky (Lviv, Ukraine) the content of heavy metals in the rock of coal mines was studied (Certificate No. RL 086/17 dated 26.06.2017 on the compliance of the measurements control system to the requirements of DSTU ISO 10012:2005). Testing and auxiliary equipment, measuring equipment and materials of the laboratory of environmental safety meet the requirements of regulatory documentation, and are certified following DSTU 3215-95, DSTU 2708: 2006. The content of heavy metals in the samples was determined using an atomic adsorption electro-spectrophotometer.

Statistical analysis of the results of semi-quantitative spectral measurements of waste heap rocks at the Chervonohradska mine was performed using the Statistica 8 applied statistics package. According to the analysis of more than 50 samples taken at a depth of 0.3 m, the statistical distribution of heavy metals content (Mn, Pb, Ni, Cu,

Zn, Co) in the mine dumps was determined, and a correlation analysis was also performed. Samples were taken uniformly from all sides of the dump in places with no vegetation cover.

### Climatic conditions of the environment

The region of Maly Polissya is characterized by low atmospheric pressure and high humidity. The predominant Atlantic air masses, combined with the influence of many meteorological factors, cause frequent, sometimes heavy rains, rapid weather changes and unstable snow cover. It has been established (Bosak et al. 2020) that as a result of precipitation and heavy rains, chlorides, hydrocarbons, sulfates and other pollutants are leached from the heaps of coal mines, which pose a significant environmental hazard to the environment.

Chervonohradska Mine (until 2001 – Chervonohradska Mine No. 2) – occupies part of the Mezhyrichansk and Zahidny Bug deposits of the Lviv-Volyn coal basin. The mine is located in the city of Chervonohrad, Lviv region (Ukraine) (Biletsky 2020). At a distance of 550 m north of the mine on the forest-like loams of the Volyn upland and the slope with absolute marks of 205–210 m there is a concentrated heap with an area of 142,000 m<sup>2</sup> and a height of 10 m to 33 m (Figure 1.).

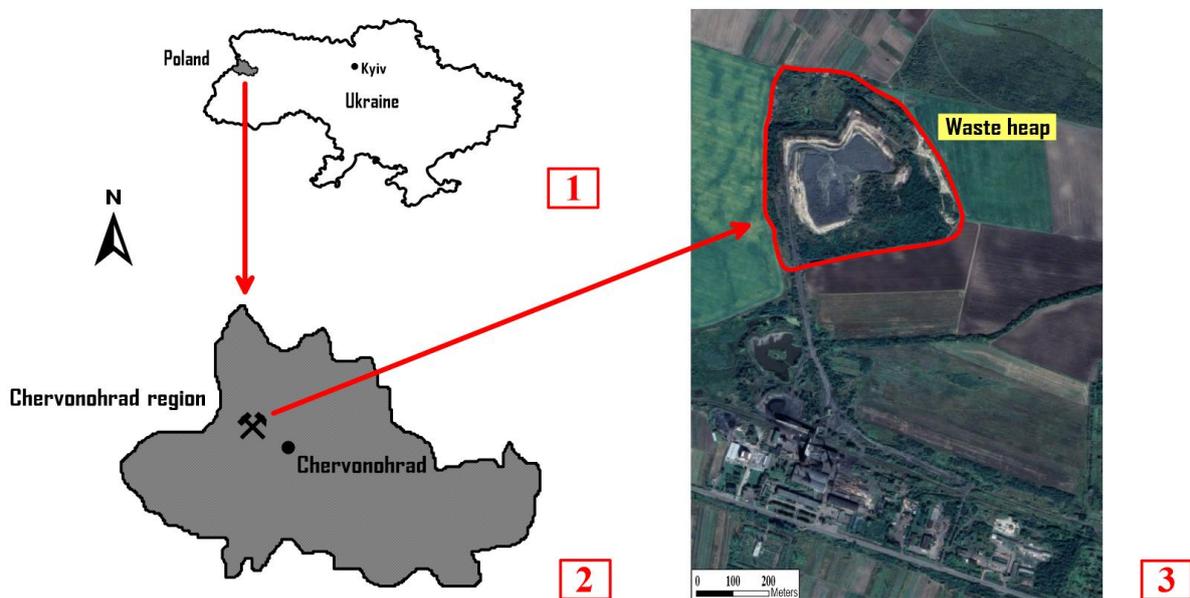


Figure 1 Layout of the Chervonohradska coal mine and waste heap within it: 1 – location of the Chervonohrad mining district on the map of Ukraine; 2 – designation on the map of the mining area of the Chervonohradska mine; 3 – image of the waste heap of the mine using Google Maps

1. ábra. A cservonohradszkai szénbánya és a benne lévő hulladékhegy elrendezése: 1 – a cservonohradi bányakerület elhelyezkedése Ukrajna térképén; 2 – elhelyezkedése a Chervonohradska bánya bányaterületének térképén; 3 – a bánya szemétdombjának képe a Google Maps segítségével

Atmospheric precipitation from the heap through the network of brooks falls directly into the streams and reclamation canals of the Buh and Solokia rivers (Lviv region). The heap is partially reclaimed (Karabyn et al. 2019).

## Results and Discussion

The following heavy metals were selected for normal distribution studies: Mn, Pb, Ni, Cu, Zn and Co. The maximum allowable concentrations (MAC) of gross forms of heavy metals are Mn 1500 mg/kg, Pb 32 mg/kg, Ni 85 mg/kg, Cu 55 mg/kg, Zn 100 mg/kg, Co 50 mg/kg (Kuraeva et al. 2012). Note that exceeding the maximum allowable concentrations of the studied heavy metals is observed in all areas except for Zn. Statistics of heavy metals content in the dumps of the Chervonohradska mine are shown in Table 1.

Table 1 Summary statistics of heavy metals content in the dumps of Chervonohradska mine

1. táblázat Összefoglaló statisztikák a Chervonohradska bánya lerakóinak nehézfém-tartalmáról

Variable	Descriptive statistics					
	Valid N	Mean	Median	Minimum	Maximum	Std. Dev.
Mn	52	1294.506	1175	360	3800	681.125
Pb	52	45.773	39.93	1.95	154	33.587
Ni	52	51.237	50.7	11.2	154	25.706
Cu	52	77.313	69.5	6.1	214.4	44.446
Zn	52	40.869	37.9	25.5	77	11.477
Co	52	1320.186	47.6	9.3	17100	4323.271

Abnormally high and uneven Co content is observed, for which data range from 9.3 mg/kg to 17100 mg/kg. This uneven content of Co in the rock may be caused by the fact that the rock extracted from the mine at depths of 800 m to 1100 m is dumped chaotically in different parts of the dump, namely, according to the principle – filling landslides and faults in the waste heap. The migration of Co in the thickness of the waste heap depends on such indicators as pH, organic matter content, free Fe, and total Mn. The investigation of Co and its migration is described in detail in (Luo et al. 2010), establishing the bioavailability of cobalt and its transfer from soil to vegetables and rice. Among the 312 soils harvested from vegetable and rice fields in suburban areas of some large cities in Fujian Province, southeast China, the total Co content in soil ranged from 3.5 mg/kg to 21.7 mg/kg, indicating a small accumulation compared to the background value of the province.

An imbalance of essential elements, like Co, in the soil can affect the health of grazing plants and animals, and thus humans. In the specific geological context of the Azores the assessment of cobalt concentration in volcanic soils has been conducted to predict the risk of cobalt deficiency in animals and humans (Linhares et al. 2019).

Thus, the uneven distribution of Co is inherent in natural landscapes of volcanic origin, especially pastures, as it is noted in China and Spain.

In our case, descriptive statistics and Kolmogorov-Smirnov and Shapiro-Wilko tests give grounds for rejecting the assumption of normal distribution of heavy metals in waste heaps under investigation. The distributions of Cu (Figure 2.) and Zn in dumps (Figure 3.) are closest to normal.

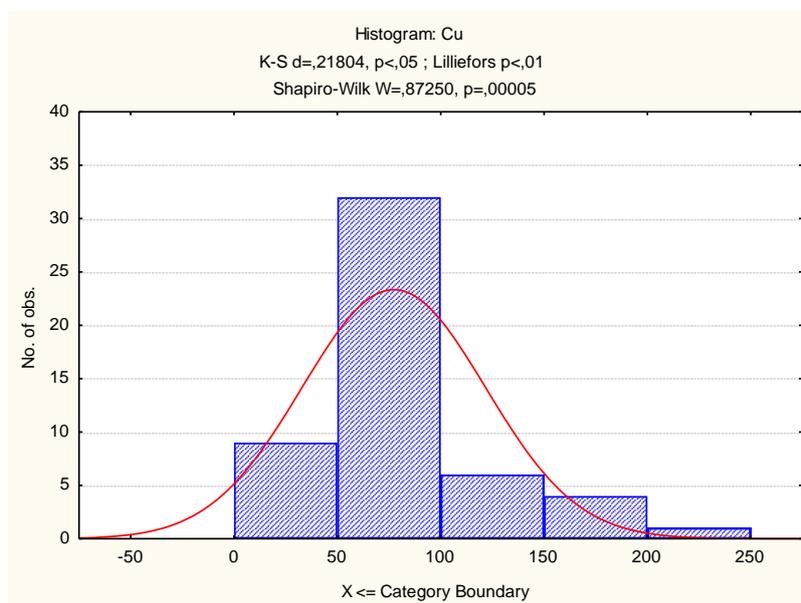


Figure 2 Approximation to the normal distribution of Cu in the waste heap of the Chervonohradaska mine

2. ábra. A Cu normál eloszlásának közelítése a hulladékhegyben Chervonohradaska bányá

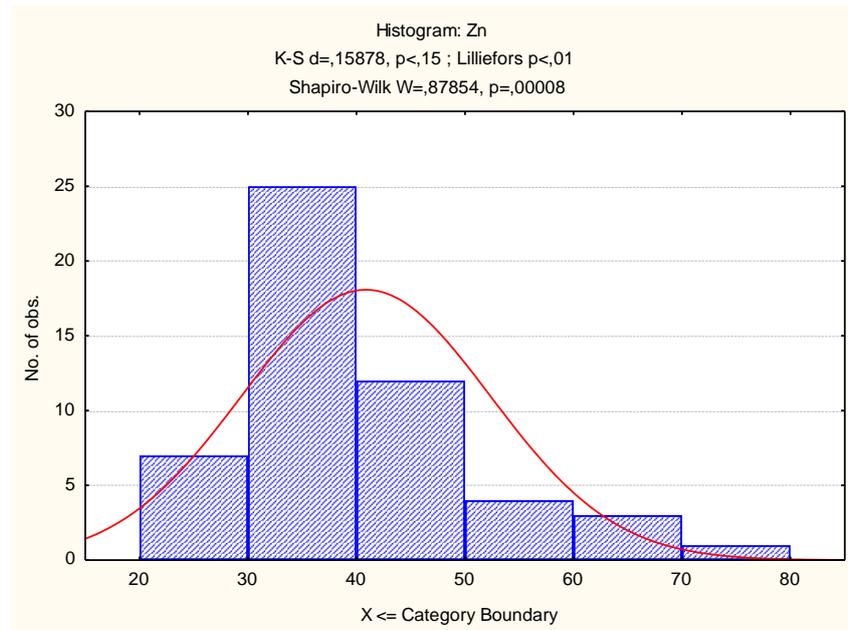


Figure 3 Approximation to the normal distribution of Zn in the waste heap of the Chervonohradka mine

3. ábra. A Zn normál eloszlásának közelítése a hulladékhegyben Chervonohradka bányá

The nonparametric Spearman coefficient ( $r_s$ ) revealed the average level of correlation of the heavy metals content in the pairs Mn and Ni ( $r_s = 0.46$ ), Mn and Zn ( $r_s = 0.52$ ), Ni and Zn ( $r_s = 0.57$ ), Cu and Zn ( $r_s = 0.49$ ) (Table 2.).

Table 2 Correlation coefficients of the investigated heavy metals in the waste heap of the Chervonohradka mine

2. táblázat. A vizsgált nehézfémek korrelációs együtthatói a Chervonohradka bányá hulladékhegyében

Variable	Mn	Pb	Ni	Cu	Zn	Co
Mn	1.00	0.12	0.46	0.33	0.52	0.11
Pb	0.12	1.00	0.43	0.37	0.41	0.09
Ni	0.46	0.43	1.00	0.69	0.58	0.26
Cu	0.33	0.37	0.69	1.00	0.49	0.06
Zn	0.52	0.41	0.58	0.49	1.00	-0.07
Co	0.11	0.09	0.26	0.06	-0.07	1.00

The highest level of correlation is in the pair of Ni and Cu ( $r_s = 0.69$ ) (Figure 4.).

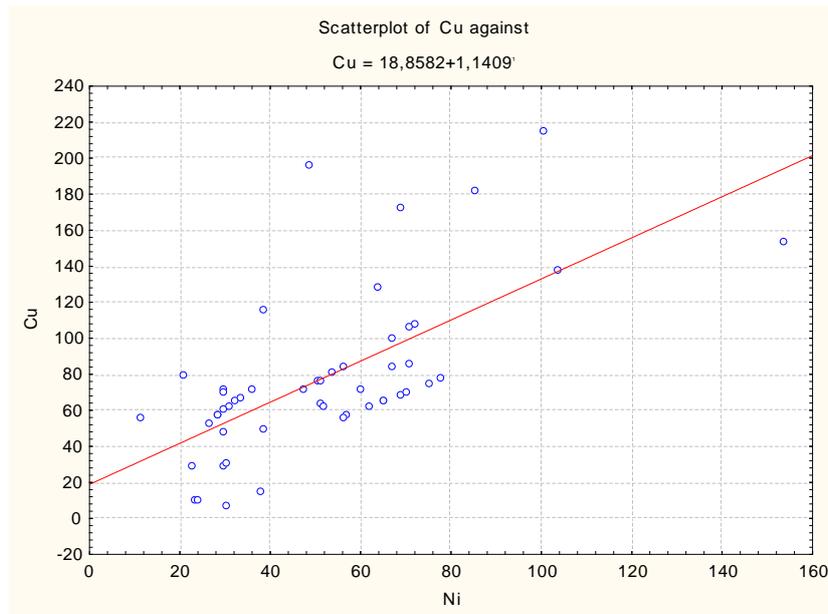


Figure 4 High correlation between Ni and Cu content ( $r_s = 0.69$ ) in the dump rock

4. ábra. Magas korreláció ( $r_s = 0,69$ ) a Ni- és Cu-tartalom között a lerakó kőzetben

Heterogeneity of normal distribution and concentration of heavy metals in the rock was observed (Álvarez et al. 2003). Concentrations of various forms of heavy metals (Fe, Mn, Zn, Cu, Cr, Ni, Cd and Pb) were determined (Álvarez et al. 2003) in the dump matter, rich in chalcopyrite. Concentrations of different forms of heavy metals were compared with concentrations in natural vegetation colonizing the landfill.

In general, the concentrations of heavy metals in soils decrease exponentially with distance from the dump, mainly due to water scattering and topography (Jung et al. 1996). Extractive soils with a high content of heavy metals have negatively affected the growth and yield of radishes (*Raphanus sativus* L.), while the adjustment to organic fertilizers has reduced the availability of heavy metals, increased radish growth and minimized risks to human health. Among the selected organic fertilizers, vermicompost was more effective and reduced the absorption of Cd, Cr, Pb and Mn by 32.5, 50.25, 44.50 and 42.25%, respectively, improved radish growth, food quality and reduced health risks (Alam et al. 2020).

In similar research (Wahsha et al. 2012), the total concentration of six potentially toxic metals (Cd, Cr, Cu, Pb, Zn and Fe) in soil and plant samples of the three dominant willow species (*Salix purpurea* L., *Salix caprea* L. and *Salix eleagnos* Scop.) was determined. Samples were collected from abandoned dumps of mixed sulfides (Imperina Valley, northeastern Italy). The results show that there is a growing need for further research projects, focusing mainly on the mechanisms by which such willows can survive in contaminated soils.

In our case, the average correlation between Ni and Mn ( $r_s = 0.46$ ), Zn and Mn ( $r_s = 0.52$ ), Zn and Ni ( $r_s = 0.58$ ), Cu and Zn and  $r_s = 0.49$ ) (Figure 5–8) was detected, which is evidence of the chaotic dumping of the rock and the likely impact of precipitation, pH of the substrate, microclimate on the concentration of heavy metals, as it was described earlier.

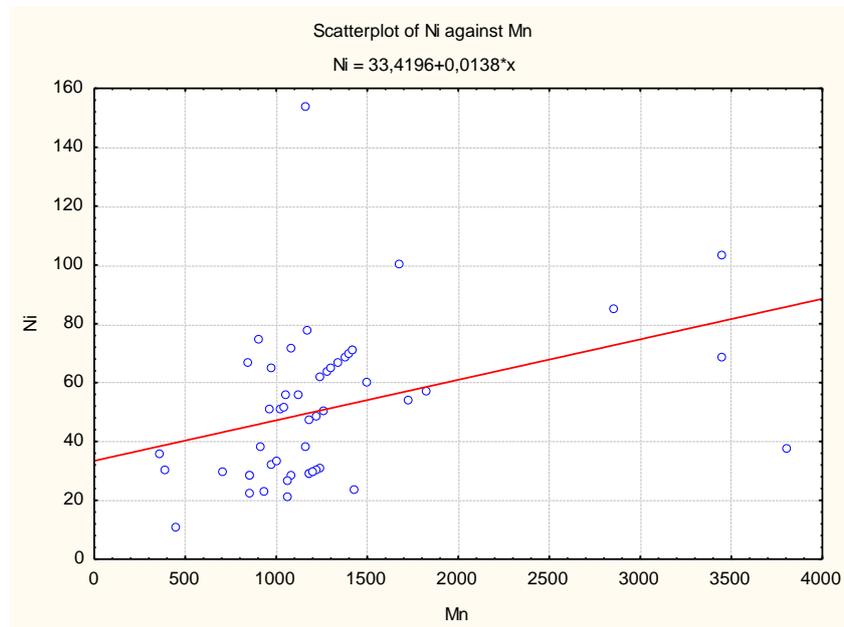


Figure 5 The average correlation between the content of Ni and Mn ( $r_s = 0.46$ ) in the dump rock

5. ábra. Az átlagos korreláció a kőzet nikkell- és mangántartalma között ( $r_s = 0,46$ )

In general, in our case, the correlation between heavy metals in the rock is below average value, and in 1/3 of cases, there is no trace at all.

Obtained values of correlation analysis coincide with the research (Terekhov et al. 2021), which is devoted to the distribution and mobility of heavy metals (Fe, Mn, Cu, Zn and Cd) in the surrounding soils of the mine and their transfer to wild flora. Soils and plants were sampled from a mining valley in northwestern Madrid, Spain; total and extracted heavy metals were analyzed.

High Cd and Zn concentrations were found in the aerial parts of *Hypericum perforatum* (Cd), *Salix atrocinerea* (Cd, Zn) and *Digitalis thapsi* (Cd, Zn). The authors declare that the article (Moreno-Jiménez et al. 2009) is the first report on the ability of the two latter species of plants to accumulate heavy metals.

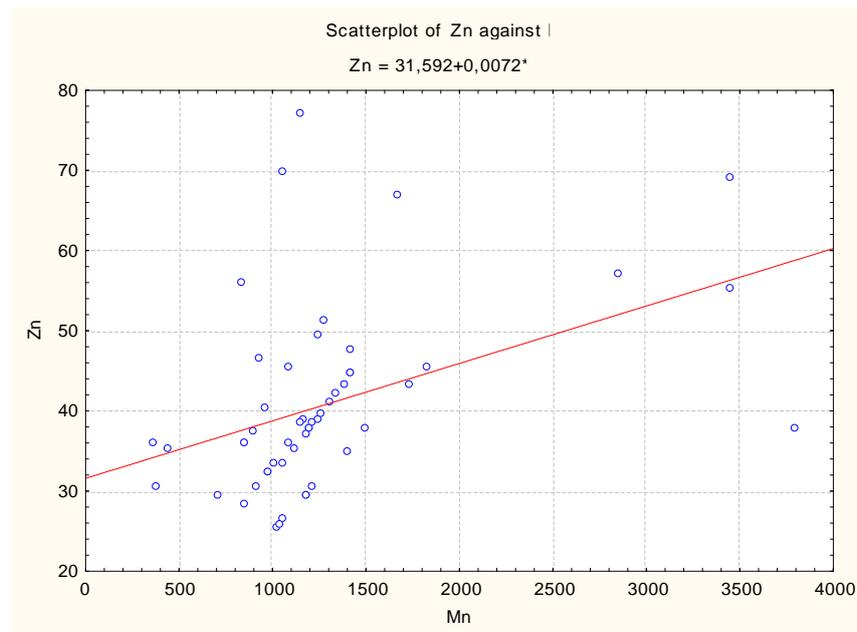


Figure 6 The average correlation between the content of Zn and Mn ( $r_s = 0.52$ ) in the dump rock

6. ábra. Az átlagos korreláció a Zn- és Mn-tartalom között ( $r_s = 0,52$ ) a kőzetben

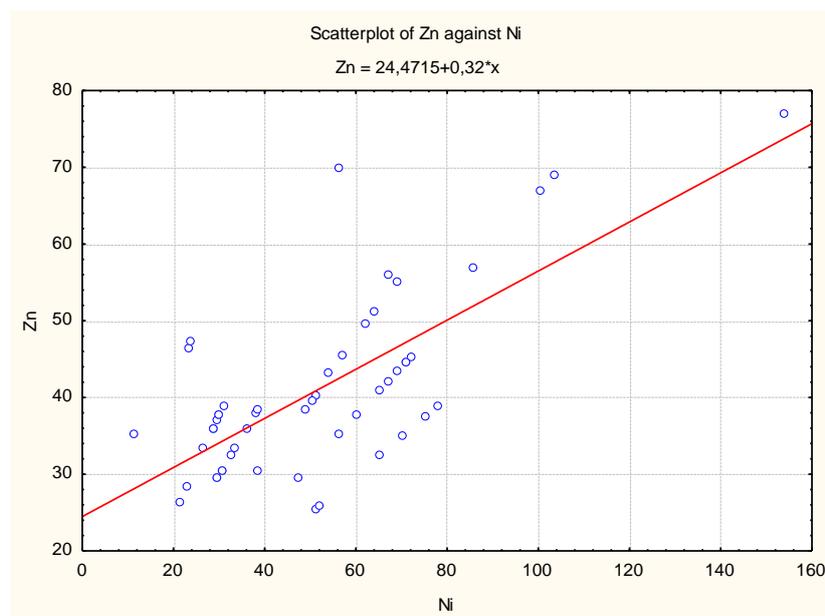


Figure 7 The average correlation between the content of Zn and Ni ( $r_s = 0.58$ ) in the dump rock

7. ábra. Az átlagos korreláció a Zn- és Ni-tartalom között ( $r_s = 0,58$ ) a lerakó kőzetben

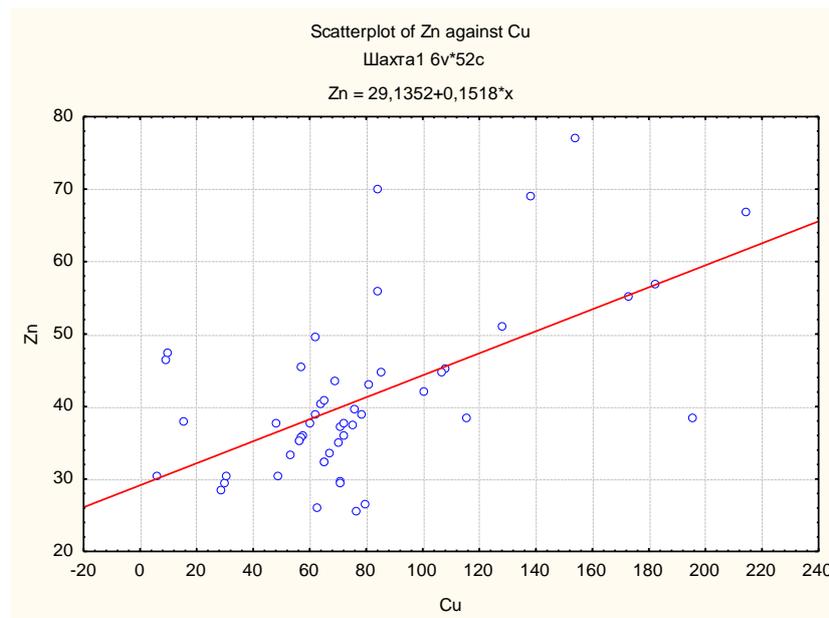


Figure 8 The average correlation between the content of Cu and Zn ( $r_s = 0.49$ ) in the dump rock

8. ábra. Az átlagos korreláció a réz- és cink-tartalom között ( $r_s = 0,49$ ) a lerakó kőzetben

The phytoremediation capacity of *S. atrocinerea* for Cd and Zn was estimated, obtaining time intervals that could be considered suitable for phytoextraction of contaminated soils. So far, in our case, the determination of the heavy metals content in vegetation is in the future.

## Conclusions

The content of Mn, Pb, Ni, Cu, Zn and Co in the heap rock of the Chervonohradaska coal mine of the Lviv-Volyn coal basin (Ukraine) was studied. The average content of Pb, Ni and Co exceeded the MAC. Abnormally high and uneven Co content was detected, for which data range from 9.3 mg/kg to 17100 mg/kg. It was found that the Cu and Zn distributions in dumps are closest to normal. The nonparametric Spearman coefficient ( $r_s$ ) revealed the average level of correlation of the content of heavy metals in the pairs Mn and Ni ( $r_s = 0.46$ ), Mn and Zn ( $r_s = 0.52$ ), Ni and Zn ( $r_s = 0.57$ ), Cu and Zn ( $r_s = 0.49$ ).

The determination of the heavy metals content in the rock of coal mine dumps is important in terms of the selection of vegetation for reclamation work, which will improve the quality of the environment and increase the regional environmental safety of coal mining complexes.

The obtained results of heavy metals distribution should be compared to the content of these elements in the vegetation developing in the research areas. Also, the data of correlation analysis will allow us to describe statistically the level of heavy metals absorption by vegetation from the substrate. That is the aim of our further research.

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## Nehézfémek elosztása a szénbányák hulladéklerakóiban statisztikai elemzés alapján

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**Kulcsszavak:** hulladéklerakó, szénbánya, nehézfémek, statisztikai elemzés, elpusztított táj, tájökológia

**Absztrakt:** Az ENSZ 2021 őszi Glasgow-i Klímaváltozási Konferenciáján (COP26) a világ vezetői és résztvevői úgy döntöttek, hogy fokozatosan megszüntetik a szénenergiát, és fokozatosan felváltják a fosszilis tüzelőanyagokat. A COP26-on Ukrajna vállalta, hogy 2035-ig leállítja az állami tulajdonú széntüzelésű erőműveket, 2030-ra 30%-kal csökkenti a metánkibocsátást, és megállítja az erdőirtást. A hulladéklerakók a szénbányászatban is káros tényezőt jelentenek. A szénbányák hulladéklerakóiban a nehézfém-tartalom meghatározása napjainkban is aktuális téma, hiszen az ilyen vizsgálatok eredményei a bányavidékek környezetbiztonságának monitorozásának részét képezik. Ez a tudományos cikk a hulladéklerakók nehézfém-tartalmával és azok eloszlásával kapcsolatos kutatások eredményeivel foglalkozik a Lviv-Volyn szénmedence egyik legnagyobb szénbányájában, a „Chervonohradskában” (Ukrajna). 1971 óta 2,9 millió m<sup>3</sup> kőzet halmozódott fel a lerakóban. Évente 40 000 m<sup>3</sup> friss kőzet kerül lerakásra. A mintákat egyenletesen vettük a szemétkerakó minden oldaláról, növénytakaró nélküli helyekről. Megjegyzendő, hogy a vizsgált nehézfémek megengedett maximális koncentrációjának túllépése a Zn kivételével minden területen megfigyelhető. A Chervonohradka bánya területén található hulladéklerakók kőzeteinek szemikvantitatív spektrális vizsgálatának eredményeinek statisztikai elemzését a Statistica 8 alkalmazott statisztikai csomag segítségével végeztük. Több mint 50; 0,3 m mélységben vett minta eredménye alapján meghatároztuk a bányatárolókban található nehézfémek Mn, Pb, Ni, Cu, Zn és Co statisztikai eloszlását, valamint korrelációs elemzést végeztünk. Megállapítást nyert, hogy a réz és a cink eloszlása a szemétkerakókban van a legközelebb a normálhoz. A nem-parametrikus Spearman-koefficiens (rs) feltárta a nehézfém-tartalom átlagos korrelációs szintjét a Mn és Ni (rs = 0,46), Mn és Zn (rs = 0,52), Ni és Zn (rs = 0,57), valamint a Cu és Zn (rs = 0,49) párokban. A kaotikus és egyenetlen kőzetkibocsátás a szabad területre bizonyos kémiai elemek egyenetlen szubsztrátumlerakódását okozta a lerakó profiljában.

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