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Spectral Analysis Method for Distinguishing Heavy Metals Pollution in the Pioneer Vegetation of Landfills Located within the Prikarpatian Geobotanical District of Ukraine

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ABSTRACT

In Ukraine, the tourism and recreation industry is widespread. These are mostly resort complexes with health boarding houses, hotels and restaurants. However, in Ukraine, the issue of solid household waste processing and active landfills, located near recreation facilities, is practically not addressed. Undoubtedly, this situation has a negative effect on the processes of recovery, because, as is known, landfills are depositing dangerous substances and compounds, which, due to geochemical flows, get into all components of the environment, which seem to be clean. The aim of this investigation is to determine the heavy metals content in the vegetation of a landfill near a places for recreation. The object of present research is the analysis of heavy metals content in the roots of trees on the Bronytsya landfill, located within the tourist and recreational complex of the Lviv region of Ukraine. It was established that tree roots are contaminated with Pb components and partially with Cd and Co components. Among the toxic chemicals, we can highlight the presence of Pb with a significant difference on the eastern side of the landfill in samples of *Acer negundo* L. (4 times higher than the MAC) and *Populus nigra* L. (7.1 times higher than the MAC), on the southern side – *Fagus sylvatica* L (1.1 times higher than the MAC) and *Malus sylvestris* Mill. (7 times higher than the MAC), on the west side – *Salix cinerea* L. (2.5 times higher than the MAC) and *Carpinus betulus* L. (6 times higher than the MAC), on the northern side of the landfill – *Malus sylvestris* Mill (2.5 times higher than the MAC) and *Prunus spinosa* L. (3 times higher than the MAC) and in the central part – *Populus nigra* L. (1.5 times higher than the MAC) and *Salix alba* L. (2 times higher than the MAC) in accordance with other samples on the investigated object. Cd exceeds the allowable concentration from the south side in *Fagus sylvatica* L. (1 times higher than the MAC) and *Malus sylvestris* Mill. (2 times higher than the MAC), from the west side in the root of *Carpinus betulus* L. (1 times higher than the MAC), from the center of the *Salix alba* L. (8 times higher than the MAC), from the south side of *Malus sylvestris* Mill. (11 times higher than the MAC), and on the eastern side *Acer negundo* L. (4 times higher than the MAC) and *Populus nigra* L. (8 times higher than the MAC). The content of Co was higher in the roots of trees from the west side of *Salix cinerea* L. (1 times higher than the MAC), from the west side of *Carpinus betulus* L. (1.7 times higher than the MAC), from the south side – *Malus sylvestris* Mill. (2 times higher than the MAC), and on the eastern side – *Populus nigra* L. (2 times higher than the MAC). An excess of Cu content was recorded on the western side of the landfill in the root of *Carpinus betulus* L. (1 times higher than the MAC). The determination of the chemical content in the roots of trees is important because it is possible to define the pollution rate of the environment in the health resort regions.

Keywords: landfill, heavy metals, environmental safety, phytomelioration.

INTRODUCTION

Heavy metals are one of the most dangerous environmental pollutants for human and animal health. They do not decompose in the environment, but migrate and accumulate in the tissues of living organisms. Penetrating into plants, heavy metals can have a negative effect on metabolic processes, which ultimately leads to a decrease in yield and the threat of toxicant contamination of all links of the food chain (microcenosis, zoocenosis, micromycetes, macromycetes). Absorption of toxic doses of heavy metals by plants causes inhibition of growth and development, disruption of photosynthesis, respiration and other biochemical processes. In addition, their entry into plant cells leads to oxidant stress due to the formation of active forms of oxygen (Ganeval G. et al., 2007).

An investigation (Adamcova D. et al., 2017) was conducted to monitor plants growing in potentially contaminated areas of a landfill to determine their potential to accumulate heavy metals. For the plant-soil system, correlations were used to determine environmental pollution from the landfill in terms of heavy metal accumulation. Based on the average concentrations, it was found that the components of heavy metals in the soil comply the following order: iron > manganese > chromium > nickel > copper > zinc > cobalt > lead > cadmium > mercury. Fe reached the highest values among all analyzed metals in the samples, i.e. in stems (103.4–6564.6 mg/kg dry matter), roots (6563.6–33036.6 mg/kg dry matter), leaves (535.1–11.275 mg/kg of dry matter) and soil (12389–39381.9 mg/kg of dry matter). The highest concentrations were determined in 2013 for Fe, Mn and Zn. The highest concentration of Fe was achieved in 2013 and 2014. The results of Pearson's correlation analysis showed that there are significant positive relationships between the content of Pb, Hg, Zn in the soil and the content of Zn, Pb, Cu in plant matter.

The research (Oka M. et al., 2017) shows the removal of heavy metals from landfill filtrate using *Phragmites australis* and *Juncus effusus*. These plants were found to show high removal rates of heavy metals such as Zn, Cr, Ni, Cd, Fe and Pb, but not Mn. *Juncus effusus* showed slightly lower removal capacity of Cr, Ni, Mn and Cd, although *Phragmites australis* showed lower removal capacity for Mn. It was concluded that the presented plants are effective absorbents of heavy metals from the bottom of filtrate ponds.

In (Akanchise T. et al., 2020) the heavy metals content and the degree of soils contamination from abandoned landfills in Kumasi, Ghana were investigated. The average metals concentrations in the soils were in the following order: Zn (166 mg/kg). Cr (67 mg/kg) > Cu (32 mg/kg), Ni (22 mg/kg) > Pb (11 mg/kg) Cd (8.9 mg/kg) > As (4.2 mg/kg) > Hg (0.04) mg/kg) for crown; and Zn (558 mg/kg), Cu (347 mg/kg), Pb (288 mg/kg) > Cr (77 mg/kg) > Ni (35 mg/kg) > As (11 mg/kg) > Cd (3.0 mg/kg) > Hg (0.19 mg/kg) for Amacom. Contamination indices (geoaccumulation, pollution coefficient, pollution load and potential environmental risk) showed very high arsenic, cadmium and lead pollution at Kronum and arsenic, cadmium, copper, lead and zinc at Amakoma. Mercury content was at the lowest contamination level for both landfills. The authors concluded that regular monitoring of these abandoned landfills is necessary, and reclamation programs should be implemented.

The long-term distribution, mobility and phytoavailability of heavy metals in anthropogenic soils was investigated in (Businelli D. et al., 2009) by collecting soil samples at different depths during a 10-year chronological sequence after the top soil layer of the landfill was changed. It was established that metals were mainly retained in the compost-amended soil horizon, however, over time, their vertical distribution led to a moderate enrichment of the underlying mineral horizons, without the direct influence of compost changes. Analysis of plant tissues that grew spontaneously in the landfill showed that the phytoavailability of the metal was limited and generally depended on the species composition.

In (Gautam M. et al., 2019) the research was conducted to assess the phenotype and distribution of metals in herbaceous species in forests and abandoned red mud landfill sites. The soil in the red mud site was characterized by high alkalinity, salinity, and poor nutritional status. In addition, the contents of Fe, Zn, Cu, Mg, Mn, Ni, Co, Cr, Cd, and Pb in soil were higher in the red mud, but their phytoavailability was lower compared to the forest soil samples. The herbaceous communities at both sites were dominated by the families *Poaceae* and *Asteraceae*. Dominant species, e.g. *Brachiaria mutica, Cynodon dactylon, Dactyloctenium aegyptium, Digitaria ischaemum, Digitaria longiflora, Eragrostis cynosuroides, Launaea asplenifolia, Parthenium hysterophorus, Sporobolus diander* and *Stylosanthes scabra* highly tolerant to metals showed their tendency towards a certain selection strategy in response to the change of soil properties in the red area mud. Thus, in (Gautam M. et al., 2019), scientists propose a potential way to phytomanagement of abandoned red sludge landfills by recultivation with dominant and metal-resistant plant species.

In (Kasassi A. et al., 2008) heavy metal content in the soil of abandoned landfill located northwest of Thessaloniki, Northern Greece, was investigated. Samples were taken by drilling to different depths (2.5–17.5 m). Chemical analysis showed that the content of metals varied in a wide range: from 0.50 to 18.75 mg/kg for Cd, 3.88–171.88 mg/kg for Cr, 8.13–356.25 mg/kg for Cu, 5.63–63.75 mg/kg for Ni, 2.50–92.50 mg/kg for Pb and 6.38–343.75 mg/kg for Zn. The highest indicators were found in three out of six wells, at depths of more than 2.5 m. Despite the fact that the territory is heavily industrialized, the presented results showed that the local industry is not a significant source of metal pollution.

The research presented in (Mukhopadhyay S. et al., 2020) confirmed the use of the NixPro™ sensor and PXRF for the rapid characterization of soil pollution by heavy metals in agricultural soils in the area affected by landfills. Mn concentrations were relatively high in that area, ranging from 493 to 3889 mg/kg. The average Pb content was 378 mg/kg, ranging from 115 to 1003 mg/kg with significant spatial variation within the investigated area. The objectives of the research (Oziegbe O. et al., 2021) were to evaluate the bioremediation potential of the investigated native bacteria and the effect of carbon source and pH on the enhancement of the bioremediation process. *Pseudomonas aeruginosa, Klebsiella edwardsii* and *Enterobacter cloacae* were selected based on their heavy metal tolerance for the remediation process. *Pseudomonas aeruginosa* showed the highest bioremediation potential among bacterial isolates with 58.80 and 33.67 percent remediation at 50 mg Cd/l and 300 mg Pb/l. However, a higher percentage of remediation (79.87 and 92.41) was observed with *Klebsiella edwardsii* due to the addition of carbon source (5 g/L) and change in pH (6) in heavy metal-contaminated medium. The results of this research indicate that the efficiency of native bacteria in the remediation process can be increased by adding a carbon source and increasing the pH for effective remediation of contaminated soil.

In (Pastor J. et al., 2012), heavy metals, salts, and organic compounds were investigated in soil and surface water samples taken from 15 landfills in the Madrid region. The impact of landfill soil cover on nematodes and plant diversity was also assessed. These analyzes continue to detect the presence of heavy metals (Zn, Cu, Cr, Ni, Pb, Cd) in soils, as well as salts (sulfates, chlorides and nitrates) in soils and surface waters. Organic compounds, mainly aromatic and aliphatic hydrocarbons, often occured in very high concentrations, and high levels of insecticides such as gamma-HCH (lindane) were also found in the soils. About 50% of the collected water samples showed chemical oxygen demand values of more than 150 mg/l. Paper (Pastor J. et al., 2012) updates the current situation and discusses the risks to the health of ecosystems, people, domestic animals and wildlife living near these landfills. Rock dumps cause significant technogenic pressure on the environment, in particular on the vegetation development (Popovych V. et al., 2020). It was established that mine rock dumps have high acidity, a significant content of various salts and sulfate ions. This high mineralization is caused by the movement to the catchment points and the interaction of such water with rock outcrops. Wastewater is also enriched with the destruction products of the mine rocks. Research on rock dumps is relevant, as it provides an opportunity to assess the toxicity of the territory and danger it may pose to the environment.

In (Karabyn V. et al., 2019), patterns of horizontal and radial distribution of petroleum products in landscapes were investigated. The highest concentration of pollutants is set at up to 1750 mg/kg. The research results highlighted in the article confirm and develop the theory of geochemistry of technogenesis, the theory of geochemical landscapes, creating a reliable experimental base for improving scientific methods of environmental and crisis monitoring. Thus, after analyzing a number of cross-border investigations of the physical and chemical composition of the vegetation cover of landfills, it can be stated that constant monitoring of their content is extremely relevant.

MATERIALS AND METHODS

Lviv region is located in the western part of Ukraine, and borders the European Union state - Poland. The region occupies the southwestern edge of the Eastern European Plain and the western part of the northern macroslope of the Ukrainian Carpathians. The territory of Lviv region is 21.8 thousand km^2 (3.6% of the territory of Ukraine), divided into 7 districts, each of which is divided into territorial communities (Korol K.A., 2022). The Bronnytsia landfill is located in the Drohobych district of Lviv region, near the Bronnytsia village. Geographic coordinates 49.429954, 23.435612. Figure 1 shows the location of experimental plots from which tree roots samples were taken for further analysis. The

Figure 1. Location of the sampling areas on landfill in Lviv region (Ukraine)

area of the landfill is 3.48 hectares (Fig. 1). The entrance is equipped with a road, a bypass ditch for leachate is partially built, there is no fence and no leachate collection and removal system. It is closed since 2018. There is no accumulation of waste. Reclamation works are not carried out. In this area, south-easterly winds prevail, and north-westerly winds are somewhat rarer. The temperature distribution shows the predominance of positive daytime temperatures during the sampling (10–25 °C), which alternated on certain days with nighttime cold temperatures (3–15 °C). The spread of the wind is more concentrated in the eastern side, carrying the remains of substances into the surrounding forest massif.

The purpose of this research is to determine chemical content of the landfill vegetation near the rest area. The object of our research is the determination of the heavy metals content in the roots of trees on the Bronnytsia landfill, located within the tourist and recreational complex of Lviv region, Ukraine. In order to determine the heavy metals content in the tree roots during the summer period (in June 2022) 10 samples were taken. Root samples were taken from 4 sides of the horizon and the central part of the landfill. The samples were dried, ground and labeled accordingly. The research was conducted in the laboratory of industrial toxicology of the Danylo Halytsky Lviv National Medical University (Lviv, Ukraine) (Certificate No. RL 086/17 dated 06/26/2017 on compliance of the measurement management system in accordance with DSTU ISO 10012:2005). Determination of heavy metals content in landfill plants was carried out at the C-115 M1 atomic adsorption spectrophotometer, part number 01-2015L (calibration certificate (UA/37/210916/001412 dated September 16, 2021). Toxic elements of the first (Pb, Zn, Cd) and second (Co, Cu) hazard class were determined in samples of tree roots from the landfill (Table 1).

Tree root samples of *Salix cinerea* L. and *Carpinus betulus* L. (west side); *Prunus spinosa* L. and *Malus sylvestris* Mill. (north side); *Acer negundo* L. and *Populus nigra* L. (east side), *Fagus sylvatica* L. and *Malus sylvestris* Mill. (south side) and *Populus nigra* L. and *Salix alba* L. (central part of the landfill) were taken.

RESULTS

The analysis of the Cu content in tree roots showed that 0.1 mg/kg (*Malus sylvestris* Mill.) and 0.2 mg/kg (*Prunus spinosa* L.) accumulates on the northern side of the Bronnytsia landfill, which does not exceed the allowable concentration. The MAC for Cu in accordance with (Order of the

Table 1. Assessment of heavy metal contamination of the vegetation cover of the Bronnytsia landfill

Hazard class	Metals	
Toxic	1 class	2 class
	Pb, Zn, Cd	Co, Cu

Ministry of Health dated of Ukraine…, 2013) is 5 mg/kg for plants. Zn content in the roots of trees was about 0.25 mg/kg (*Malus sylvestris* Mill.) and 0.29 mg/kg (*Prunus spinosa* L.) from the southern side, which does not exceed the allowable concentration of 10 mg/kg. The Pb content was 1.29 mg/kg (*Malus sylvestris* Mill.) and 1.58 mg/kg (*Prunus spinosa* L.), which exceeds the allowable MAC concentration for Pb three times according to (Order of the Ministry of Health dated of Ukraine…, 2013) for tree species 0.5 mg/kg (Figure 2). The highest Cd content in the investigated tree roots corresponded to the measurements for *Malus sylvestris* Mill. – 0.03 mg/kg, which is within the normal range and 0.07 mg/kg for *Prunus spinosa* L., which is twice the allowable concentration (Order of the Ministry of Health dated of Ukraine …, 2013). The MAC for Co (Order of the Ministry of Health dated of Ukraine …, 2013) is 1 mg/kg for plants, which in this research showed a content within the normal range of 0.05 mg/kg of a sample of *Malus sylvestris* Mill. and 0.16 mg/kg of *Prunus spinosa* L. root sample, which slightly exceeds the allowable concentration.

The content of heavy metals in the roots of *Salix cinerea* L. and *Carpinus betulus* L. trees will be considered separately for each metal. Cu from the western side of the Bronnytsia landfill was 0.39 mg/kg (*Salix cinerea* L.) and 5.57 mg/kg (*Carpinus betulus* L.), which exceeds the allowable concentration for Cu in the roots of *Carpinus betulus* L. trees in accordance with (Order of the Ministry of Health dated of Ukraine…, 2013). The Zn content in the roots of trees was 0.17 mg/kg (*Salix cinerea* L.) and 8.15 mg/kg (*Carpinus betulus* L.) on the western side, which does not exceed the allowable concentration. The Pb content in accordance with (Order of the Ministry of Health dated of Ukraine…, 2013) exceeds the allowable concentration, in our case it was 1.27 mg/kg for *Salix cinerea* L. and 2.95 mg/kg for *Carpinus betulus* L. The highest Cd content in the roots of the trees corresponded to the measurements for *Salix cinerea* L. – 0.02 mg/kg, which is within the normal range and 0.33 mg/kg for *Carpinus betulus* L., which exceeds the allowable concentration by 10 times (Figure 3). The Co content was within the normal range of 1 mg/kg in the sample of *Salix cinerea* L. and 1.7 mg/kg of a root sample of the *Carpinus betulus* L., which showed that the allowable concentration was exceeded.

The obtained data of the heavy metals content in the roots of *Populus nigra* L. and *Salix alba* L. in the central part of the landfill was as follows: the Cu content was 0.09 mg/kg (*Populus nigra* L.) and 0.22 mg/kg (*Salix alba* L.), which does not exceed the allowable concentration in accordance with (Order of the Ministry of Health dated of

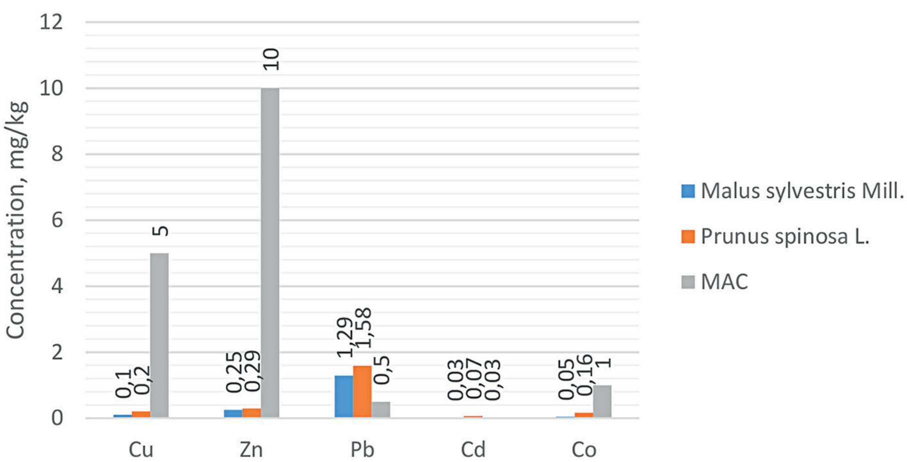

Figure 2. The metals content in the root system of *Malus sylvestris* Mill and *Prunus spinosa* L. trees on the northern side of the landfill

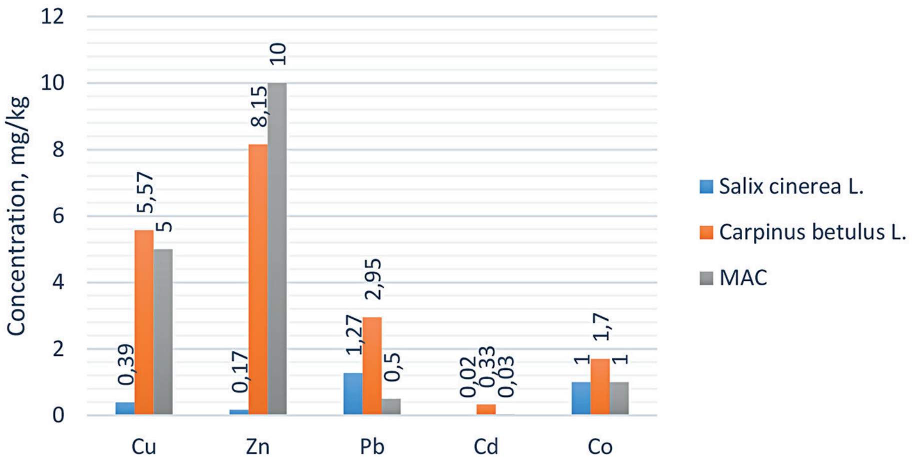

Figure 3. Metals content in the root system of *Salix cinerea* L. and *Carpinus betulus* L. from the western side of the landfill

Ukraine…, 2013). The present content of Zn in the roots of trees was 0.6 mg/kg (*Populus nigra* L.) and 0.23 mg/kg (*Salix alba* L.), which does not exceed the allowable level. The content of Pb was 0.84 mg/kg (*Populus nigra* L.) and 1.03 mg/kg (*Salix alba* L.), which exceeds the allowable concentration. The highest content of Cd (0.03 mg/kg) in the investigated tree roots of *Populus nigra* L. was 0.02 mg/kg, which is within the normal range and 0.23 mg/kg in the roots of *Salix alba* L., which exceeds allowable concentration. Co content was 0.05 mg/kg in root samples of *Salix alba* L. and *Populus nigra* L., which does not exceed the allowable concentration (Figure 4).

The obtained data of the heavy metals content in the roots of *Fagus sylvatica* L. and *Malus sylvestris* Mill was as follows: Cu from the southern side of the Bronnytsia landfill was 0.04 mg/kg (*Fagus sylvatica* L.) and 0.59 mg/kg (*Malus sylvestris* Mill.), which does not exceed the allowable concentration. The Zn content in the tree roots from the southern side of the landfill was 0.08 mg/kg (*Fagus sylvatica* L.) and 0.74 mg/kg (*Malus sylvestris* Mill.), which does not exceed the allowable concentration. The maximum Pb content was 0.61 mg/kg (*Fagus sylvatica* L.) and 3.55 mg/kg (*Malus sylvestris* Mill.), which exceeds the allowable MAC concentration for Pb. The highest content of Cd in the roots was defined in the sample of *Fagus sylvatica* L., which did not exceed the allowable concentration, for *Malus sylvestris* Mill. it was 0.34 mg/kg, which exceeds the MAC of 0.02 mg/kg (Figure 5). The maximum content of Co in this research was within the normal range of 0.05 mg/kg in the root sample of *Fagus sylvatica* L., and in the roots of *Malus sylvestris* Mill. it was 2 mg/kg, which exceeded the MAC.

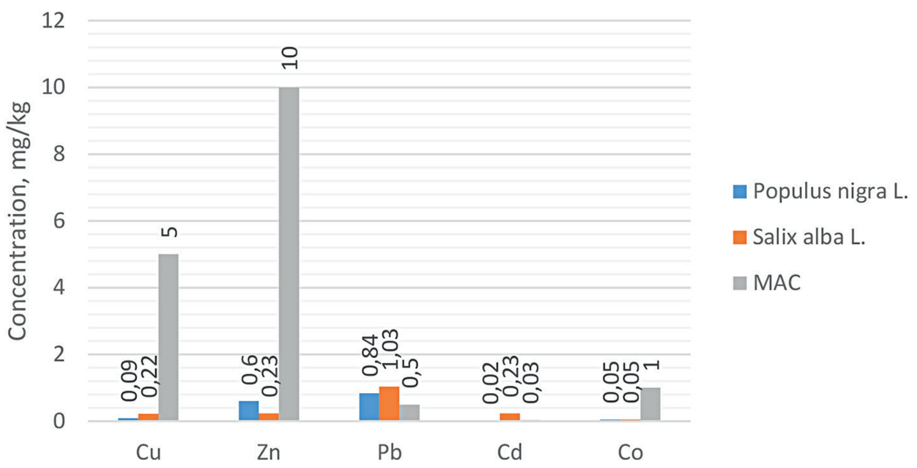

Figure 4. The heavy metals content in the root system of *Populus nigra* L. and *Salix alba* L. from the central part of the landfill

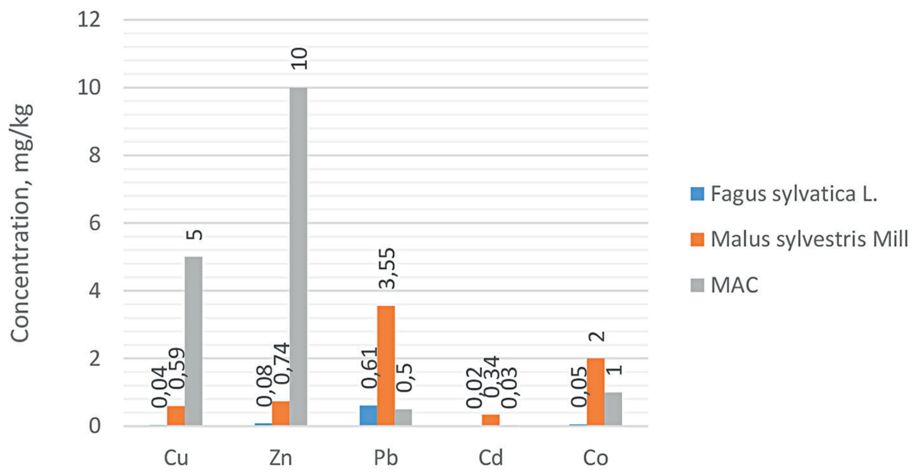

Figure 5. Heavy metals content in the root system of Fagus sylvatica L. and Malus sylvestris Mill. on the southern side of the landfill

The obtained data of the heavy metals content in the roots of *Acer negundo* L. and *Populus nigra* L. Was as follows: the Cu content from the eastern side of the Bronnytsia landfill was 0.13 mg/kg (*Acer negundo L.*) and 0.33 mg/kg (*Populus nigra* L.), which does not exceed the allowable concentration for Cu according to (Order of the Ministry of Health dated of Ukraine…, 2013). On the eastern side the Zn content was 0.86 mg/kg (*Acer negundo* L.) and 0.48 mg/kg (*Populus nigra L.*), which does not exceed the allowable concentration. The content of Pb is 2.05 mg/kg for *Acer negundo* L. and 3.12 mg/kg for *Populus nigra* L., which significantly exceeds the allowable concentration for Pb in accordance with (Order of the Ministry of Health dated of Ukraine …, 2013). The highest Cd content in the investigated tree roots of *Acer negundo* L. was 0.11 mg/kg and of *Populus nigra* L*,* – 0.25 mg/kg, which exceeds the allowable concentration (Order of the Ministry of Health dated of Ukraine …, 2013). The maximum Co content was within the normal range of 0.27 mg/kg in the roots of the *Acer negundo* L. Sample. In *Populus nigra* L. sample Co content of 1.99 mg/kg exceeded the allowable concentration. The analysis of the heavy metals content showed a significant excess of Pb content in all samples of the root system of tree species growing on the territory of the Bronnytsia landfill

CONCLUSIONS

The research analyzed the content of 5 heavy metals (Cu, Zn, Pb, Cd, Co) in the root system of trees of the Bronnytsia landfill in the Lviv

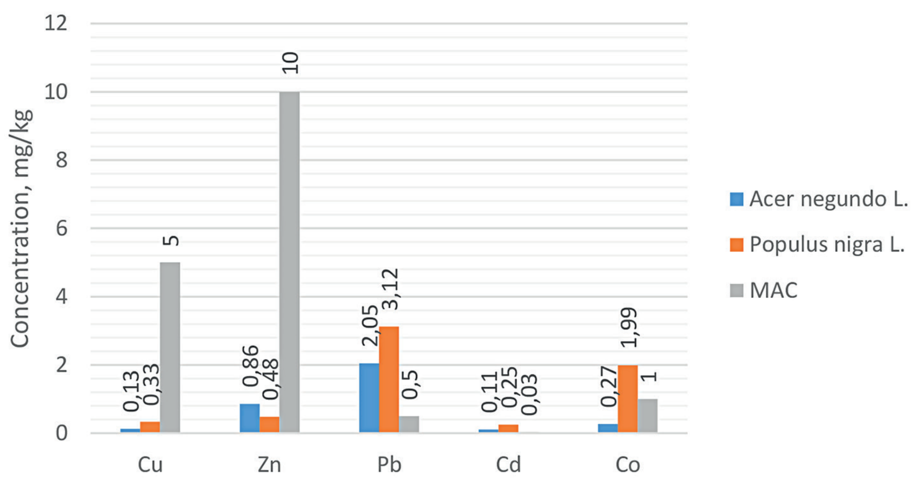

Figure 6. The heavy metal content in the root system of *Acer negundo* L. and *Populus nigra* L. from the eastern side of the landfill

region of Ukraine. The landfill is located on the territory of a tourist and recreational zone, which causes interest in investigation the impact of its hazardous components on the quality of the environment in the recreation area.

It was established that tree roots are chemically contaminated with Pb and partially with Cd and Co compounds. Among the toxic chemical elements, we highlight Pb with a significant difference on the eastern side of the landfill in samples of *Acer negundo* L. (4 times higher than the MAC) and *Populus nigra* L. (7.1 times higher than the MAC); on the south side – *Fagus sylvatica* L. (1.1 times higher than the MAC) and *Malus sylvestris* Mill. (7 times higher than the MAC); on the western side – *Salix cinerea* L. (2.5 times higher than the MAC) and *Carpinus betulus* L. (6 times higher than the MAC); on the northern side of the landfill – *Malus sylvestris* Mill (2.5 times higher than the MAC) and *Prunus spinosa* L. (3 times higher than the MAC) and in the central part – *Populus nigra* L. (1.5 times higher than the MAC) and *Salix alba* L. (2 times higher than the MAC) in comparison with other samples at the investigated object.

Cd exceeds the allowable concentration on the southern side in *Fagus sylvatica* L. (1 times higher than the MAC) and *Malus sylvestris* Mill. (2 times higher than the MAC), on the western side in the root of *Carpinus betulus* L. (1 times higher than the MAC), in the center of the landfill in the sample of *Salix alba* L. (8 times higher than the MAC), on the southern side in *Malus sylvestris* Mill. (11 times higher than the MAC), and on the eastern side in *Acer negundo* L. (4 times higher than the MAC) and *Populus nigra* L. (8 times higher than the MAC). The content of Co was higher in the roots of trees on the western side of *Salix cinerea* L. (1 times higher than the MAC), on the western side of *Carpinus betulus* L. (1.7 times higher than the MAC), on the southern side – *Malus sylvestris* Mill. (2 times higher than the MAC), and on the eastern side – *Populus nigra* L. (2 times higher than the MAC). An excess Cu content was detected on the western side in the root of *Carpinus betulus* L. (1 times higher than the MAC).

The determination of the chemical content of the tree roots is important because it is possible to see the analysis and spread of pollution. Plants growing on the polluted territory of the Bronytsia landfill completely absorb the investigated heavy metals. An important indicator of the ecological state of the investigated landfill was the exploring of assortments of wood species, which, due to their physiological characteristics, have an increased absorption capacity. The roots of trees accumulate heavy metals as much as possible, which causes the cleaning of the substrate at this object and due to this, the migration chain of pollution is shortened. In order to minimize the accumulation of heavy metals in landfill substrates and, as a result, plant organs, engineering measures for environmental protection, in particular, bioplateau constructions and phytomelioration, should be implemented.

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