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Data on heavy metals (Pb, Ni, Zn) in soil and biota (common nettle and Roman snail) around the power plant TC Kosova A in Obiliq (Kosovo)

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Abstract

The heavy metal content of Pb, Zn, and Ni in soil, nettle plants (rhizomes, stems, and leaves), and snail shells was investigated in this study. Samples were collected during the summer-autumn period of 2020 from the vicinity of the TC Kosova A power plant in Obiliq (Prishtina), Kosovo, and compared to samples from the control site, Brezne-Opoja. Soil samples were dried at 105°C for 48 hours and treated with a mixture of 69% HNO3 and HClcc in a 1:3 ratio. Similarly, biota samples were dried at 105°C for 48 hours and treated with a mixture of 69% HNO3 and H2O2cc in a 1:3 ratio. All sample types were digested in the microwave at 200°C for 45 minutes and analyzed using a flame type absorber (Analyticyena Contra AAA).

The results indicate significantly higher concentrations of Pb, Ni, and Zn in the Obiliq area compared to Opoja, in both soil and biota (nettle and snail shells), suggesting potential environmental impact from the nearby coal-burning plant, heavy traffic, or other urban services. The average values generally fell below the limits specified by the UK (1989) or Germany (1992) standards (Directive 2008/50/EC). However, the Zn content in all soil samples exceeded the German standard. Pb and Zn were found to be extensively absorbed by nettle plants (U. dioica) and garden snails (H. pomatia), whereas Ni showed no significant bioaccumulation in either species, despite its high concentration in the soil. Nettle plants exhibited heavy bioaccumulation of Pb, reaching levels up to 105.6 mg/kg dw, surpassing the German standard limit. Nettle plants show promise in the phytoremediation and soil amendment processes for heavy metal pollution, particularly for Pb and Zn, but not for Ni. Garden snails can serve as bioindicators for assessing heavy metal pollution and its impact on transfer processes within the food chain, as well as the resulting effects on biochemical and physiological processes in living organisms, specifically in relation to oxidative stress. The contamination of soil and biota with heavy metals in the Obiliq area should be regarded as a significant concern. Therefore, regular monitoring of heavy metal content in agricultural, garden, livestock, and poultry activities is necessary to mitigate the associated health risks to humans.

Keywords: TC Kosova A, heavy metals, stinging nettle, snail, bioaccumulation

1. Introduction

The electricity sector in Kosovo heavily relies on coal-fired power plants, particularly Kosova A and Kosova B, which have been operational since 1983. Located in Kastriot (Obilić), just 13 km away from Prishtina, the capital of Kosovo, these power plants are known to be major sources of pollution in Europe, emitting significant amounts of particulate matter and sulphur dioxide. The nearby lignite mines in Bardh and Mirash villages supply the fuel for these plants. As the harmful effects of heavy metals (HM) as environmental pollutants become more apparent and present a growing concern for their presence in soil and their potential to penetrate the food chain, leading to bioaccumulation in plants, animals and ultimately to humans. Various human activities, including mining, traffic, and intensive agriculture, can contribute to the contamination of soil and air with particulate matter, especially during dry and windy weather conditions. The concentrations of heavy metals in the soil are influenced by factors such as the chemical properties of the soil and the distance from the contamination source. The amount of heavy metals absorbed by biota depends on the type and concentration of the metals, as well as the specific plant and animal species involved.

Nettle plants (*Urtica dioica L.)* and land snails (*Helix pomatia L.)* are examples of organisms that naturally accumulate heavy metals, particularly Pb, Zn, and Ni. In industrial areas and along urban settlements and roads, these metals tend to accumulate in snail shells and tissues. Nettle plants, characterized by their erect green stems and stinging hairs, are herbaceous perennials that grow up to 2 meters tall during the summer and die off in winter. They are commonly found in Kosovo, particularly in moist and nutrient-rich soils near human settlements. The young

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shoots of nettle plants are traditionally valued for their medicinal properties and as a food source.

The Roman or garden snail, on the other hand, is a large land gastropod mollusk that prefers habitats with calcareous substrate, high humidity, lower temperatures, and soft soil for egg-laying and hibernation. Feeding primarily on fresh green plant parts, garden snails can live up to 8 years in controlled environments but have a shorter lifespan in the wild. Both nettle plants and garden snails are considered Least Concern species on the IUCN Red List, although habitat disturbance and unsustainable harvesting pose threats to their populations.

The Obiliq area, where the TC Kosova A power plant is located, has long been exposed to industrial operations, making it susceptible to heavy metal pollution. The landscape has been significantly altered due to open-cast mining activities, resulting in large dumps of overburden and solid wastes such as ash and sludge. These industrial activities contribute to high levels of dust, sulphur dioxide, and nitrogen oxides, with lead, cadmium, mercury, and arsenic being prevalent contaminants in the area.

In this study, we aim to investigate the impact and distribution of heavy metals (Pb, Zn, Ni) in soil, nettle plant parts (rhizomes, stems, and leaves), and snail shells collected around the TC Kosova A power plant in Obiliq, Kosovo. A comparison will be made with samples collected from Brezne-Opoja, serving as a control site. The goal is to provide an updated assessment of the environmental conditions in the vicinity of TC Kosova A, specifically focusing on heavy metal concentrations in the soil and their potential mobility to different parts of nettle plants and snail shells.

2. Materials and methods

Soil samples, as well as samples of common nettle (U. dioica) and Roman or garden snail (H. pomatia) (Fig. 3), were collected for the purpose of analyzing heavy metal concentrations, specifically Pb, Zn, and Ni. The sampling took place during the summer-autumn period of 2020 in two locations: around the TC Kosova A power plant in Obiliq, Kosovo, and in Brezne-Opoja, a mountainous plain in the south of Kosovo situated at an elevation of 1380 m above sea level, which is considered an unpolluted control site (Fig. 1).

The area surrounding TC Kosova A was divided into four geographical regions: northwest, northeast, southeast, and southwest, each encompassing imaginary circles at 1 km, 2 km, and 5 km radii. A total of 12 sampling stations were selected, with three stations in each region (Fig. 2 above). At each sampling site, 10 material samples were collected, including soil, nettle plants, and snails, resulting in a total of 120 samples. Additionally, in the control site of Brezne-Opoja, which was also divided into four geographical regions mirroring those in Obiliq (Fig. 2 below), 10 samples of soil, nettle plants, and snails were collected separately, amounting to 30 samples in total.

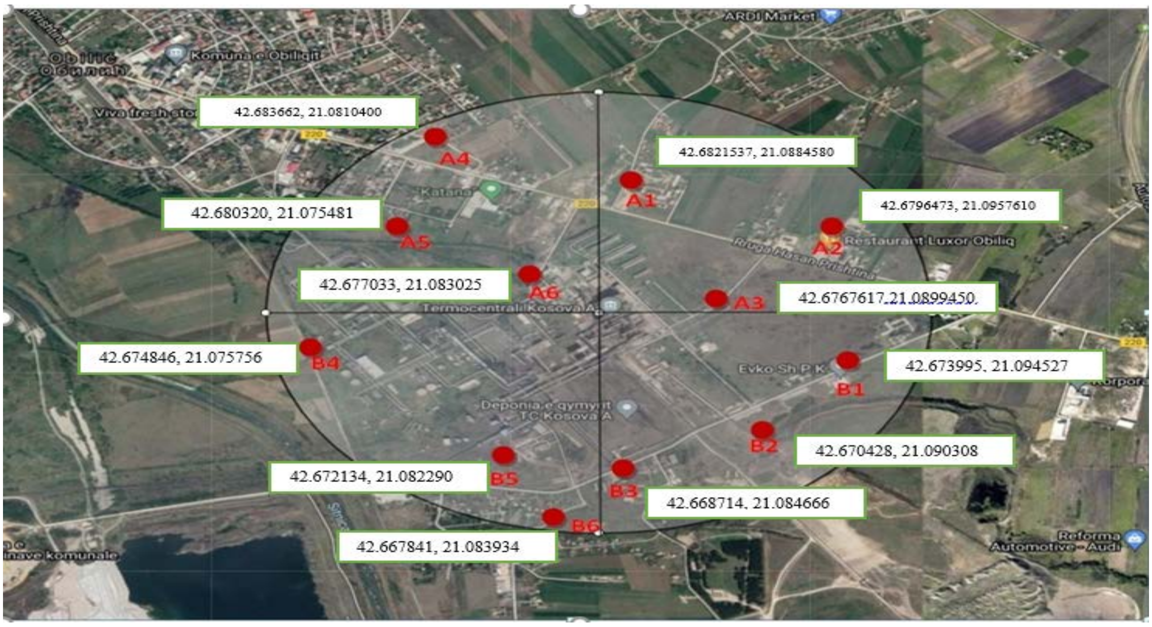

Figure 1.Kosovo map where the two sampling places are shown: TC Kosova A in Obiliq and Brezne-Opoja (control site) (elaborated in Google Earth 2015).

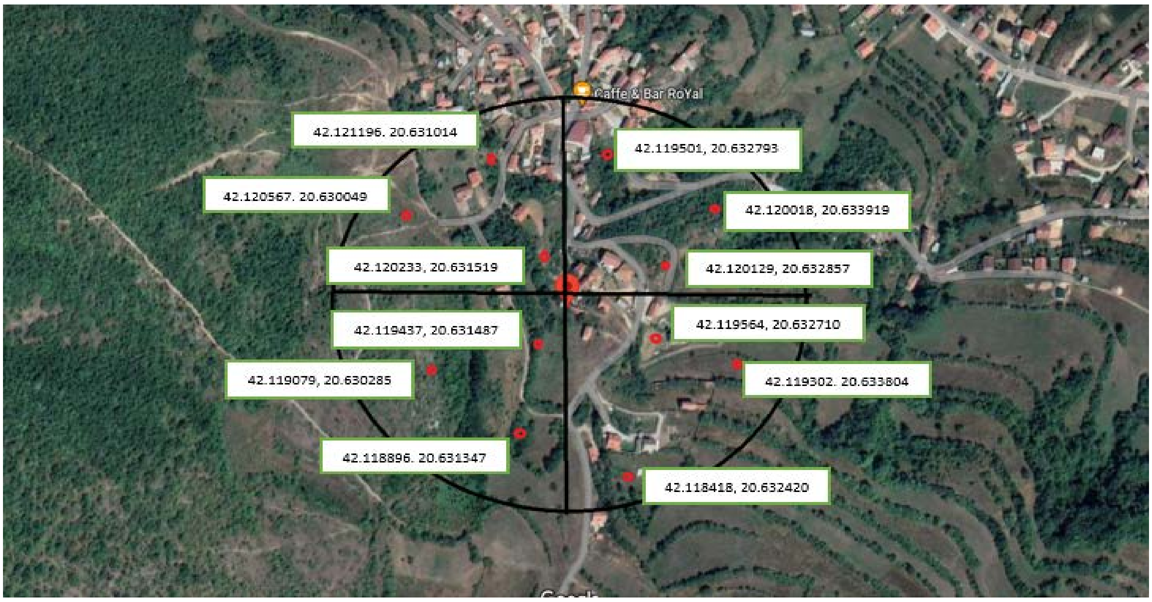

Figure 2. Schematic representation of the sample collection at locality TC Kosova A, Obiliq (above) and at the unpolluted locality Brezne-Opoja (below).

Figure 3. Photo of common nettle (U. dioica) from Obiliq: vegetative shoots and an individual of garden snail (H. pomatia) (white arrow) in it (© M. Bici).

Soil samples were collected manually using a hand probe at a depth of 15 cm from undisturbed areas, following the methods described by Kluge and Wessolek (2012) and ISO11466 (1995). The collected soil samples were then ground using a soil mill and placed in glass cups, after which they were dried in a thermostat at 105°C for 48 hours to eliminate moisture. A 0.3 g portion of the dried soil sample (based on dry weight) was subjected to treatment with a mixture of 69% HNO3 and HClcc (Merck Millipore) in a 2:6 ratio, using Teflon columns. The samples were digested in a microwave instrument (Analyticyena TOPwave) at 200°C for 45 minutes. After filtration, the contents were transferred to 50 ml glass containers using distilled water. Analysis of metals Pb, Zn, and Ni was conducted using a flame type absorber (Analyticyena Contra AAA) with the application of Merck Millipore ICP multi-element standard solution 111355.

Samples of nettle plants and snails (4-6 years old) were collected from the same habitats. The vegetative parts of the nettle plants (rhizomes, stalks, and leaves) were separated and washed with distilled water. They were then dried at 105°C for 24-48 hours and ground using a Philips kitchen mixer. A 0.5 g portion of the sample was treated with ultra-pure nitric acid (HNO3) of 69% purity and Lachner hydrogen peroxide (H2O2) of 30% purity in a 1:3 ratio. The samples were digested in a microwave instrument at 200°C for 45 minutes. After filtration, the contents were transferred to 50 ml glass containers and normalized with distilled water. Analysis of metals Pb, Zn, and Ni in the samples was performed using a flame absorber (Analyticyena Contra AAA).

Bioaccumulation factor (BCF) and transfer factor (TF) calculations were carried out to assess the movement of heavy metals from the soil to different plant parts (rhizomes, stems, and leaves), as well as from leaves to snail shells in the Obiliq area at distances of 1 km, 2 km, and 5 km. The BCF was determined using the formula: BCF = Cbiota / Csoil, where Cbiota represents the concentration of the metal in plant or animal tissues (dry weight) and Csoil represents the concentration of the metal in the soil (mg/kg dry weight). The TF within plant parts, such as from rhizomes to stems or leaves, and from leaves to snail shells, was calculated using the formulas: TF = Cshoot/leave / Crhizome and Cshells / Cleaves, respectively. Here, Cshoot, Cleave, Crhizome, and Cshells represent the concentrations of heavy metals in the plant shoot, leaves, rhizomes, and snail shells, respectively (Galal and Shehata, 2015).

The statistical analysis of the results was performed using Minitab® 19 software, including the Tukey-Kramer Test and ANOVA software for Excel.

3. Results

Table 2 presents the average concentrations of heavy metals (Pb in mg/kg, Ni in µg/kg, and Zn in mg/kg) reported as dry weight values for all sample types, including soil, nettle parts (rhizomes, stems, and leaves), the overall nettle plant, and snail shells collected from Obiliq and Opoja. The data is also visually represented in Figures 4 and 5. Due to the low levels of nickel observed in the vegetative organs, the concentration is reported in µg/kg for both nettle and shell samples in all cases.

Additionally, Table 2 provides information on the concentrations, bioaccumulation factors (BCFs), and transfer factors (TFs) of heavy metals from the soil to rhizomes, from rhizomes to stems and leaves, and from leaves to shells in the Obiliq area at distances of 1 km, 2 km, and 5 km.

Table 1. Average data of heavy metals (Pb, mg/kg, Ni, µg/kg, Zn, mg/kg), reported as dry weight values, in all sample types: soil, nettle parts (rhizomes, stems and leaves), overall nettle plant, and snail shells from Obiliq and Opoja.

HMs / Sample	Soil	SD	Rhizome	SD	Stem	SD	Leave	SD	Nettle	Snail	SD
Obiliq 1 km											
Pb, mg/kg	32.6	0.4	22.5	1.9	16.8	2.72	15.4	2.3	18.23	11.6	1.4
Ni, µg/kg	186.7	5.1	0.06	5.1	0.093	5.2	0.015	1.2	0.06	2.8	2.4
Zn, mg/kg	161.5	3.3	44.7	10.9	13.9	6.27	11.4	1.3	23.33	44.5	5.6
Obiliq 2 km											
Pb, mg/kg	77.3	1.3	17.7	6.65	10.8	2.48	12.7	2.2	13.73	11.1	1.38
Ni, µg/kg	214.9	0.4	0.065	0.95	0.084	4.1	0.017	1.3	0.06	1.58	2.5
Zn, mg/kg	230.8	13.2	25.3	12.3	16.1	9.15	6.6	1.1	16.00	40.7	4.3
Obiliq 5 km											
Pb, mg/kg	90.4	1.4	56.1	0.46	99.3	5.25	105.6	5.4	87.00	10.3	1.29
Ni, µg/kg	252.2	0.5	0.06	3.05	0.014	2.3	0.032	2.5	0.04	2.3	2.1
Zn, mg/kg	290.9	9.6	33.5	3.05	19.8	9.71	23.5	1.4	25.60	40.9	6.34
Obiliq area, overall average											
Pb, mg/kg	66.77	1.03	32.10	3.00	42.30	3.48	44.57	3.30	39.66	11.00	1.36
Ni, µg/kg	217.93	2.00	0.06	3.03	0.06	3.87	0.02	1.67	0.05	2.23	2.33
Zn, mg/kg	227.73	8.70	34.50	8.75	16.60	8.38	13.83	1.27	21.64	42.03	5.41
Opoja											
Pb, mg/kg	9.23	0.9	0.16	0.12	0.078	0.035	0.012	0.09	0.08	0.09	0.02
Ni, µg/kg	25.1	1.3	0.021	0.019	0.034	0.013	0.054	0.026	0.04	0.011	0.05
Zn, mg/kg	46.2	0.62	0.042	0.021	0.026	0.018	0.045	0.017	0.04	0.025	0.09

Table 2. Concentration, bioaccumulation factor (BCF), transfer factor (TF) of heavy metals from soil to rhizomes, from rhizomes to stems and leaves, and from leaves to shells in Obiliq (1 km, 2 km and 5 km).

Sample	C	BCF	TF	C	BCF	TF	C	BCF	TF
Obiliq 1 km				Obiliq 2 km			Obiliq 5 km		
Pb (mg/kg)									
Soil	32.6			90.3			77.4		
Rhizome	22.5	0.69		17.7	0.2		56.06	0.72	
Stem	16.8	0.52	0.75	10.8	0.12	0.61	99.25	1.28	1.77
Leaf	15.3	0.47	0.68	12.7	0.14	0.71	105.6	1.36	1.88
Shell	11.7	0.36	0.76	11.1	0.12	0.87	10.33	0.13	0.1
Zn (mg/kg)									
Soil	162			231			290.9		
Rhizome	44.7	0.28		25.3	0.82		33.51	0.12	
Stalk	14	0.08	0.31	16.1	0.07	0.64	19.84	0.07	0.59
Leaf	11.4	0.07	0.25	6.62	0.03	0.26	23.47	0.08	0.7
Shell	44.5	0.28	3.89	40.7	0.18	6.15	40.92	0.14	1.74
Ni (µg/kg)									
Soil	187			215			252.2		
Rhizome	0.08	0.04		0.06	0.03		0.05	0.02	
Stalk	0.09	0.05	0.01	0.08	0.04	0.01	0.014	0.05	0.03
Leaf	0.02	0.08	0.02	0.02	0.08	0.03	0.032	0.01	0.06
Shell	8.17	0.04	0.06	1.58	0.07	0.94	2.3	0.09	7.2

Significant differences were observed between the samples collected from Opoja (Op) and those from Obiliq ($p<0.001$; $n=10$) as depicted in Figures 4 and 5. Table 1 and Figures 3a, c, and e demonstrate that the heavy metal (HM) content in the soil was generally higher compared to the biota. Nettle plants showed a greater affinity for absorbing Pb compared to Ni or Zn. On the other hand, the garden snail exhibited a higher bioaccumulation of Ni and Zn in its shell than Pb (Figures 4b, d, and f). Notably, higher HM values were recorded at a distance of 5 km, followed by lower values at 2 km, and the lowest values at 1 km in the Obiliq area. While the HM content in the soil increased with the distance from TC Kosova A, this trend was not consistently observed in plant and shell samples. For instance, Ni content in plant vegetative organs and shell did not show a similar increase with distance (Figure 5).

Overall, the concentrations of HMs in both soil and biota were generally below the limits set by the UK (1989) and Germany (1992) standards (Directive 2008/50/EC).

However, the average Zn content in the soil samples from Obiliq at 2 km, 5 km, and Obiliq itself exceeded the German standard (highlighted in red in Table 1). Lead, which is bioaccumulated in nettle plants up to 105.6 mg/kg dry weight, surpassed the limit set by the German standard. Furthermore, when compared to the control site in Opoja, both soil and biota samples consistently exhibited higher HM content (Table 1), indicating pollution in the area, possibly resulting from the coal-burning plant, heavy traffic, or other urban activities.

The relatively high content of Pb absorbed by nettles raises concerns for animal health, particularly for grazing animals and human consumption. The young shoots of nettle plants are traditionally harvested and used in various food preparations, such as 'burek,' 'lakror,' or 'pispilit.' Additionally, nettle leaves are commonly traded as medicinal plants. Similar considerations apply to garden snails, as their preferential bioaccumulation of Ni and Zn can have detrimental effects on other biota higher up in the food chain (e.g., birds and mammals) as well as on human health. Garden snails are often harvested and consumed as food, and they are also traded in the pharmaceutical and cosmetic industries.

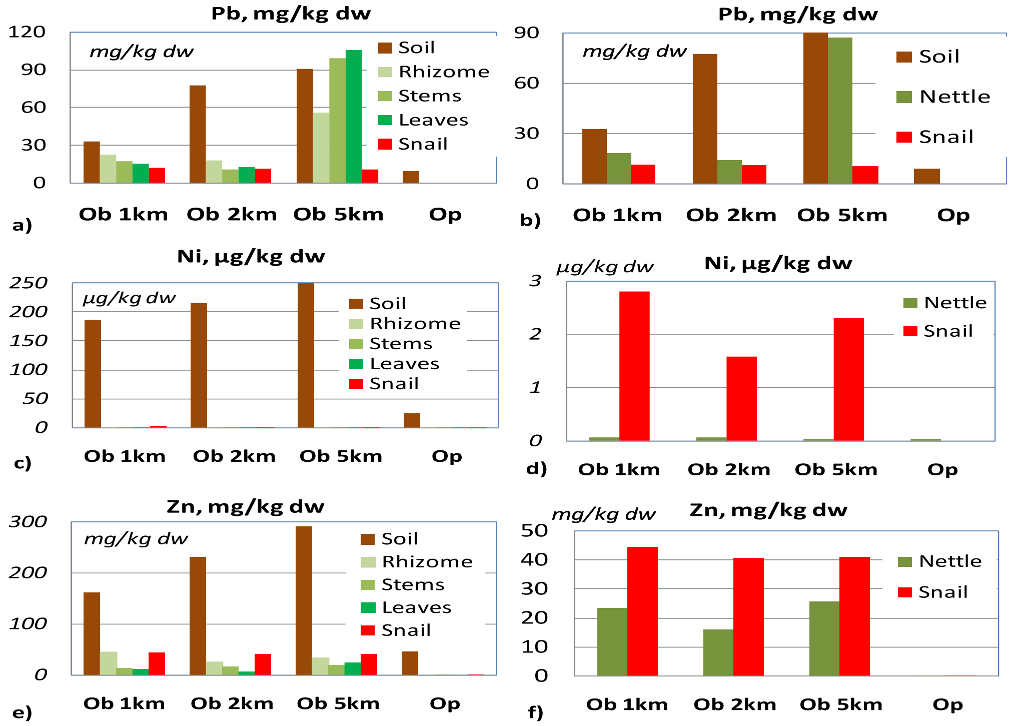

Figure 4. Average content of heavy metals considering the distance from TC Kosova A (1 km, 2 km dhe 5 km) and Opoja, in all sample types (soil, rhizomes, stems, leaves, snail shells) (a, c, e), and in soil, whole plant (nettle) and snail shells (b), and only nettle and snail (d, f). Ob, Obiliq; Op, Opoja.

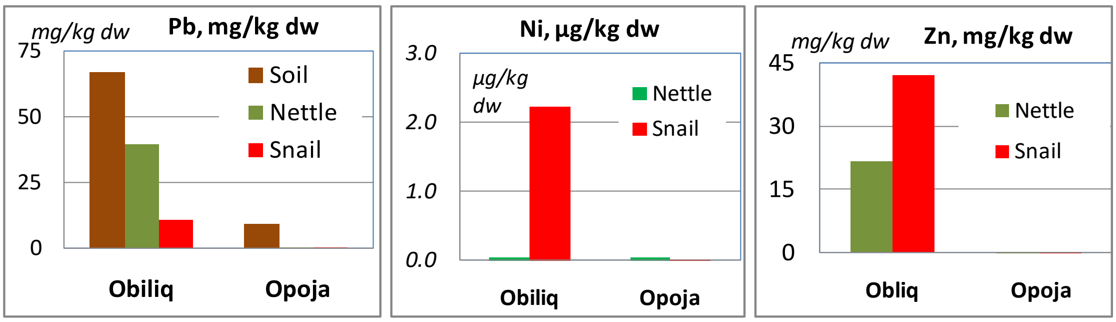

Figure 5. Average content of heavy metals in Obiliq and Opoja; for Pb in soil, whole plant (nettle) and snail shells; and for Ni and Zn in nettle plant and snail shells.

4. Discussion

The overall contamination of heavy metals (HMs) in Obiliq poses a serious threat not only to the indicated species but also to other biota, plants, animals, and human life itself. Similar to the nettle and snail, other plants and animals living in the area, including cultivated vegetables, crops, fruit orchards, and livestock and their products, may be affected. Bislimi et al. (2013) reported data on hemocyanin (Hc) and transaminase activity (AST and ALT) in garden snails (H. pomatia) from the Obiliq Power Plant area. Bislimi et al. (2021) provided data on heavy metal concentrations (Pb, Ni, Cd, Cu, Fe) in soils and plants (U. dioica) around the Kishnica mineral deposit. Additionally, Çarkaj et al. (2022) reported data on heavy metals (Pb, Zn, Cu, Cd) in garden snails collected around the Trepça smelter and Vernica village (control area) in

Mitrovica. Demaku et al. (2022) presented data on heavy metals (Pb, Fe, Cd, Zn, Ni, Al, Cu, Mn, Cr) in water, sediment, and soil around the Sitnica River, which flows through the Kosovo A landfill, during April, July, and October of 2018. According to their findings, the order of HMs in almost all soil samples was Pb > Fe > Ni > Cd > Mn > Al > Cu > Zn and > Cr, which is consistent with our results from soil samples in Obiliq: Pb 0.7 mg/kg > Ni 0.17 mg/kg > Zn 0.25 mg/kg.

The relatively high nickel content in Obiliq, particularly at a distance of 5 km in the southern area, indicates the polluted state of the zone. The source of nickel, lead, and zinc is likely the power plants, resulting from the combustion of lignite and substandard oil during turbine ignition for energy production, as well as from industrial and municipal waste. Factors such as ash dumps, wind rosettes, and heavy coal-carrying vehicles likely contribute to their distribution in the southwest part of the area. The Pb content in soil samples at 1 km was 38.5 mg/kg in the southern part and 26.8 mg/kg in the northern part. The Ni content was 162.2 µg/kg in the northern part and 211.2 µg/kg in the southern part, while the Zn content was 151 mg/kg in the northern part and 179.9 mg/kg in the southern part. At a distance of 2 km, the soil content was: Pb 35.4 mg/kg in the northern samples and 52.9 mg/kg in the southern part; Ni was 213.5 µg/kg in the north and 216.3 µg/kg in the south; Zn was 162.3 mg/kg in the north and 188.1 mg/kg in the southern samples. At a distance of 5 km, the respective average values were: Pb 58.6 mg/kg, Ni 215 µg/kg, Zn 218.3 mg/kg in the north soil samples, whereas Pb 96.3 mg/kg, Ni 288.7 µg/kg, Zn 280.1 mg/kg in the south soil samples in the polluted area (Table 1).

These findings align with the fact that nickel is a widely dispersed transition element present in the environment, including air, water, and soil. It can originate from both anthropogenic activities and natural sources (Genchi et al., 2020). Altıkulaç et al. (2022) confirmed that the concentrations of Ti, V, Cr, Mn, Fe, Cu, Zn, As, Sr, Hg, and Pb in fly ash are higher compared to those in slag. Furthermore, highly toxic heavy metals such as As and Hg are significantly enriched in coal compared to the average Earth's crust. Some heavy metals may leach from ash and slag heaps, contaminating agricultural areas, soil, surface water, and groundwater.

Table 2 displays the bioaccumulation values of heavy metals from soil to plants or animals. In some cases, such as different parts of the nettle plant (rhizomes, stems, leaves) and shells, the bioconcentration factor (BCF) for Pb and Zn is <1, indicating biomagnification along the food chain. These results are consistent with those of other studies (Nica et al., 2012; Salih et al., 2021). In other cases, BCF is close to 1, while very low translocation values were recorded for Ni.

The concentration of heavy metals in soil and plant species is influenced by various factors such as bioavailability, cation exchange capacity, pH, vegetation season, climatic conditions, age, and nutrition of animals like snails, mammals, and humans (Gardiner et al., 1995). Our data challenges the claim by Bislimi et al. (2013) that snails (H. pomatia) are super bioindicators of environmental degradation due to their sensitivity to heavy metals and other pollutants. Çarkaj et al. (2022) also emphasizes that garden snails can accumulate relatively high concentrations of heavy metals and survive, making them a good model for biomonitoring heavy metals in the environment.

The mining industry, including soil, groundwater, and air pollution, is often responsible for ecological and environmental problems (Zhang et al., 2019). Heavy metal contamination has become a significant environmental issue in mining regions, affecting the quality of agricultural goods and ultimately impacting human health. Therefore, soil and water-related environmental challenges play a crucial role in agriculture (Linhua and Songbao, 2019). HMs, such as Cd, Pb, Ni, and Cr, may have long-term effects on human health through the consumption of apple fruit and other plants, with the possibility of these elements entering the food chain (Imeri et al., 2019). Pb and Zn frequently exceed the maximum permissible limits set by various countries and are responsible for most of the potential ecological impact in the studied sites (98.64%) (Yahya et al., 2021).

Pb, Cd, and Ni are examples of non-essential elements that tend to accumulate in vegetative parts and biomagnify from roots to stems and leaves with translocation factor values >1 (Bislimi et al., 2021). Furthermore, their potential bioavailability in products near contaminated soils is found to be extremely high (Zogaj et al., 2014). The nettle plant (U. dioica) has been shown to bioaccumulate and translocate heavy metals, making it a potential candidate for phytoremediation and soil amendment processes, particularly for Pb (Bislimi et al., 2021). Heavy metals, particularly Pb, Zn, and Ni, bioaccumulate in the shells and tissues of snails, and this bioaccumulation is more common in industrial areas and along urban highways (Salih et al., 2021). To fully understand the human risks associated with heavy metal pollution, further research is needed on other plant species (cereals, fruits, vegetables), different locations, and routes of metal exposure, such as the consumption of animal foods (meat, milk, eggs), drinking water, or air contact (Filimon et al., 2021).

Our results demonstrate that the heavy metal content (Pb, Ni, and Zn) in the Obiliq area is significantly higher compared to Opoja, both in soil and biota (nettle and snail shells), indicating the environmental impact likely from coal-burning plants, heavy traffic, or other urban services. While the average values generally fall below the limits set by standards like those of the UK (1989) or Germany (1992) (Directive 2008/50/EC), Zn content in all soil samples exceeds the German standard, and Pb is heavily absorbed by nettle from the soil, with concentrations as high as 105.6 mg/kg dw in its leaves, surpassing the limit of the German standard.

5. Conclusions

The observed soil pollution by heavy metals (HMs) in Obiliq is likely attributed to the emissions from the chimneys of the Kosovo A power plant, as well as the dispersion of ash and its widespread distribution across the area. The presence of wind rosettes contributes to a higher dispersion in the southern part compared to the northern part.

Pb and Zn show a higher affinity for uptake by both the nettle plant (U. dioica) and the garden snail (H. pomatia), whereas Ni does not exhibit significant bioaccumulation in either species despite its high concentration in the soil. The nettle plant shows potential for effective phytoremediation and soil amendment processes to address heavy metal pollution, particularly for Pb and Zn, but not for Ni. The garden snail can serve as a valuable bioindicator for assessing heavy metal pollution and its impact on transfer processes within the food chain, as well as its effects on the biochemical and physiological processes of living organisms.

The contamination of soil and biota with HMs in the Obiliq area is a matter of serious concern. It is crucial to regularly monitor the HM content in agricultural, garden, livestock, and poultry products from this region to prevent potential risks to human health.

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