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Original Research

Ecological Risk Factor, Contamination Factor and Bioconcentration Factor of Heavy Metals in Vegetables Grown on Polluted Soils in Kosovo

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Abstract

The accumulation of heavy metals in plants is a serious environmental and health problem, especially in regions near industrial facilities, smelters, and mines. The aim of this study is to assess the environmental risks of cadmium (Cd), nickel (Ni), and lead (Pb) contamination in different types of vegetables. For this purpose, the bioconcentration factor (BCf), contamination factor (Cf), and ecological risk factor (Erf) were analyzed in vegetable samples collected from regions close (Zvecan and Frasher) and further (Polac) from the smelter. Heavy metal analysis was performed with inductively coupled plasma mass spectrometry (ICP-MS) in onion, garlic, and potatoes from these areas. The concentration of Cd and Pb in the analyzed vegetables from the Zvecan and Frasher is high and above the allowable limit, while in the Polac region, Cd and Pb were not detected. The highest concentrations of Cd and Ni were determined in garlic, fresh weight (0.134 mg/kg; 14.32 mg/kg; 5.982 mg/kg and 32.951 mg/kg, respectively), and for Pb in potatoes, fresh weight (0.555 mg/kg). According to BCf, onion accumulates the most Cd, garlic accumulates the most Ni, and potatoes accumulate the most Pb. According to the contamination factor (Cf) in the areas of Zvecan and Frasher, there is a very high level of contamination by Cd and Pb. Significantly, very high ecological risk for Cd is in the regions of Frasher and Zvecan, and for Pb in the region of Zvecan. The results of the concentration of the analyzed metals in vegetables correspond to the results of contamination from environmental risks.

Keywords: ecological risk factor, contamination factor, bioconcentration factor, heavy metals

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Introduction

Vegetables provide essential vitamins and minerals for humans. They could be in a variety of colors, which is evidence of numerous phytochemicals present in vegetables. These phytochemicals help the human body to fight against various diseases and reduce the incidence of new ones. The metabolic activities of the human body are enhanced when vegetables are consumed in large quantities. They can be eaten in a raw or cooked form. In general, they are low in fat and carbohydrates, and rich in vitamins, minerals, and dietary fiber [1].

Onion (*Allium cepa*, L.) is one of the most important vegetable crops. Based on the color of the onion cover, there are three varieties of onion: red, yellow/brown, and white [2]. The main root part of the onion is the one that is consumed and is characterized by a strong taste and pungent odor [3]. The main reason for the wide consumption of onions globally is that it is the richest source of quercetin. The typical taste and aroma of onion are due to volatile organosulfur compounds that make it an excellent ingredient in most of the foods to which it is added in the form of essential oil, powder, paste, flakes, or juice [4].

Moreover, garlic (*Allium sativum* L.) originates from Central Asia, i.e. Kazakhstan, and is rich in volatile compounds, which usually are used for flavor, and non-volatile compounds such as phenolic compounds (sapogenins, flavonoids, proteins, and saponins), amides, nitric oxides, minerals (Se, P, and K), vitamins (B and C), and antioxidants. The root part of garlic is rich in carbohydrates and contains significant amounts of protein, while fat is present in lower concentrations [5].

On the other hand, potato (*Solanum tuberosum*) is a major basic food for humans and the fourth largest crop grown in the world after rice, wheat, and maize. The annual global potato production and the millions of hectares devoted to its production have prompted researchers to analyze its various nutritional and health aspects [6]. Potatoes contribute key nutrients to the diet, including vitamin C, potassium, and dietary fiber [7].

These vegetables are valuable sources of nutrients essential for human health, but only if they are grown on clean agricultural land. If the soil is contaminated with heavy metals, vegetables can accumulate these elements and, instead of being desirable, be dangerous for human health.

Recently, there has been an increasing concern about environmental pollution. Human exposure to heavy metals has increased due to the use of these metals in industry, agriculture, and households [8]. Moreover, soil around industrial areas, mines, and smelters usually contains high amounts of various heavy metals [9-11]. Vegetables grown in areas contaminated with heavy metals could absorb heavy metals through the roots, allowing the heavy metals to reach the edible parts of the plant (Fig. 1) [12-13]. According to the review by Bouida et al. [14], heavy metals taken up by plants can easily enter the food chain, creating a serious risk for the environment and human health. Their accumulation in plants depends on several factors, such as the type of vegetable, soil properties, pH, organic matter, as well as climate conditions.

To quantify the contamination of vegetables that grow in polluted areas with heavy metals, different quantification factors have been proposed, such as the bioconcentration factor, contamination factor, and ecological risk factor (Fig. 1) [15-17]. These values help explain how much of the metals move from the soil into the plants, how polluted the soil is, and what kinds of environmental risks these metals may cause [18-20].

The bioconcentration factor (BCf) is the concentration of heavy metals present in plant biomass divided by the concentration of the corresponding heavy metal in the soil on which the corresponding plants are grown [18]. If the value is higher than 1, it means the plant absorbs more metal than what is present in the soil, and if it is less than 1, the plant absorbs less [21, 22]. BCf depends on many factors, such as the type of vegetable, the type of soil, pH, climate, and the form of the metal in the soil [23, 24]. For example, Aljahdali and Alhassan [25] showed that plants growing in polluted marine areas had low BCf for lead (Pb), likely because lead sticks strongly to soil and does not move easily into the plant. Another quantification of the contamination factor (Cf) is the ratio of the actual concentration and the background concentration of the individual heavy metal in soil. It is used to assess the impact of pollution of individual toxic elements in soils [19, 26]. In places close to mines or smelters, very high Cf values have been found, especially for metals like cadmium (Cd) and lead (Pb), which can stay in the soil for a long time [27, 28]. In addition, the ecological risk factor (Erf) quantitatively expresses the potential ecological risk of an individual contaminant [20]. It includes both how polluted the soil is (Cf) and how toxic the metal is. For example, cadmium (Cd) is considered more dangerous than other metals because of its high toxic response factor of 30 [20]. Many studies have shown that Cd and Pb are often the main metals that cause high ecological risk in soils near smelters and other industrial areas [25, 26, 29].

Heavy metal toxicity can increase the risk of cardiovascular and neurotoxic diseases, brittle bones, damage to the lungs, kidneys, and other vital organs [13]. Long-term exposure can lead to the gradual progression of physical, muscular, and neurological degenerative processes that mimic diseases such as multiple sclerosis, Parkinson's disease, Alzheimer's disease, and muscular dystrophy [30].

Many studies show high concentrations of heavy metals in various vegetables that exceed the permissible limits according to FAO and WHO standards [31-33]. Recently, awareness of the consumption of fresh, minimally processed, and/or natural foods without additives has increased, and therefore, the demand for health-promoting foods has increased. A two-year research in China on the potential risk of Cd indicates that surveillance should focus on reducing environmental risks and protecting the safety of agricultural products rather than concentrating on health risks [34].

Therefore, this study aims to assess the accumulation and ecological risk of Cd, Pb, and Ni in selected vegetables from polluted soils in Kosovo using BCf, Cf, and Erf indices.

Materials and Methods

Three types of vegetables were analyzed – potato (*Solanum tuberosum*), onion (*Allium Cepa*), and garlic (*Allium sativum*), as well as the soil where these vegetables were grown. The analyzes were made at the technological and nutritional maturity of the vegetables (harvest 2023). The vegetable samples were taken from three areas in the region of Kosovska Mitrovica (42°53'55"N, 20°51'48"E) in Kosovo. In the vicinity of the two areas Zvecan (42°54'40.62"N, 20°50'15.06"E) and Frasher (42°51'21.66"N, 20°53'15.08"E), there is a lead and zinc smelter, and the third region is Polac (42°44'4.84"N, 20°49'9.15"E), which is at a greater distance from the smelter. Fig. 2 illustrates the geographical location of the study areas in Kosovo, including Zvecan, Frasher, and Polac, on a map.

All samples for analysis were dried to constant mass in a drying oven (SLN 15, Wodzisław Śląski, Poland) for a period of 24-30 hours, depending on the type of vegetable. The dried samples were ground into a powder suitable for making vegetable digests.

Determination of Heavy Metals Cd, Ni, and Pb

Determination of the concentration of Cd, Ni, and Pb in the selected vegetable species was performed according to an accredited method MKC EN ISO/IEC 17025:2018 using microwave digestion and inductively coupled plasma mass spectrometry ICP-MS (model 7500cx, Agilent USA).

Cd, Ni, and Pb in the soil were determined using methods ISO 11464:2006(E) and ISO 14869-1:2001 by use of the ICP-MS technique (ISO 17294-2:2009).

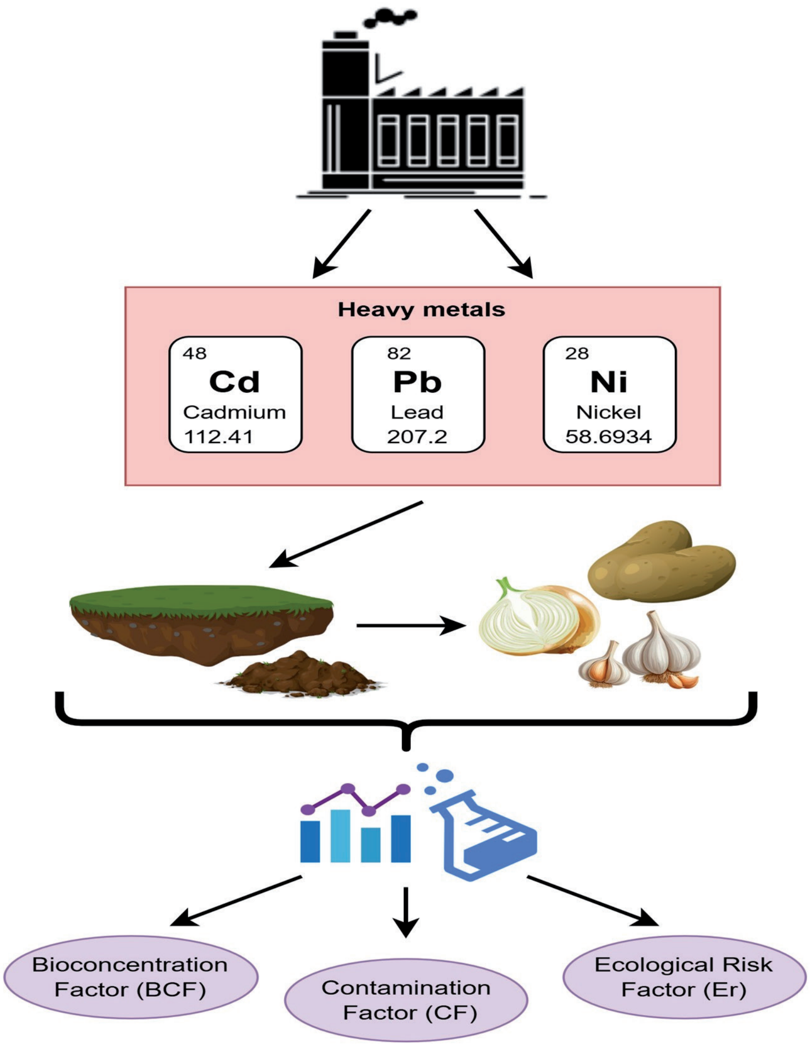

Fig 1. Schematic visualization of heavy metal contamination pathway and factors of contamination.

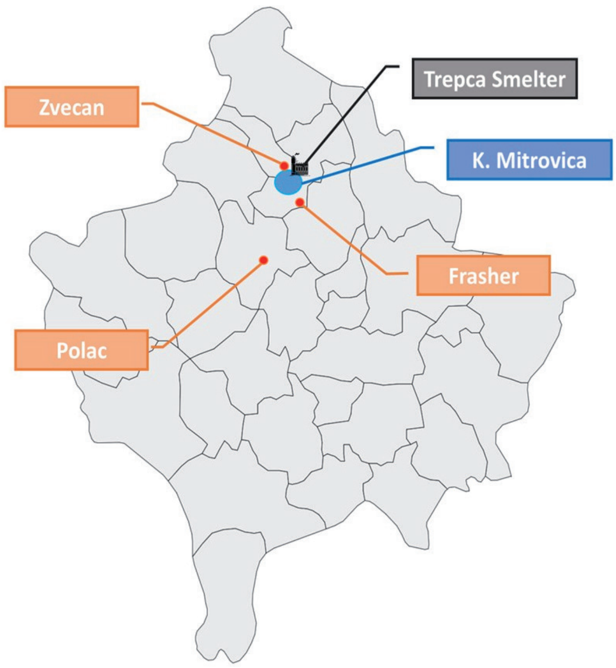

Fig. 2. Geographical Distribution of Sampling Regions in Kosovo.

Determination of Bioconcentration Factor (BCf)

BCf is the ratio of plant to soil metal concentration; it is calculated by use of Equation (1) [21, 22]:

$$\mathrm{BCf} = \frac{C_{vegetables}}{C_{soil}} \quad (1)$$

Where: $C_{vegetables}$ and C_{soil} represent the heavy metal concentrations in the vegetables and soils, respectively, on a dry-weight basis [21, 32].

Contamination Factor (Cf)

Contamination factor (Cf) is calculated using Equation (2) [19]:

$$\mathrm{Cf} = Cs/C_{background} \quad (2)$$

Where: C_s is the concentration of metal in the sample soil, $C_{background}$ is the concentration of metal in the background soil. The concentrations of potential toxic elements in the background soil value are: Cd-0.3; Ni-68; Pb-20 ppm [35]. The contamination is classified as [36]: Cf<1: low degree; 1≤Cf<3: moderate degree; 3≤Cf<6: considerable degree; and Cf≥6: very high degree.

Ecological Risk Factor (Erf)

The ecological risk factor is calculated by Equation (3) [20]:

$$\mathrm{Erf} = \mathrm{Tf} \times \mathrm{Cf} \quad (3)$$

Where: Tf is the toxic response factor of the individual element (Cd-30; Ni-1; Pb-5) and Cf is the contamination factor [20]. The classification criteria of Erf proposed by Hakanson [36] and Zhang [20] are: Erf<40 low potential ecological risk; 40≤Erf<80 moderate potential risk; 80≤Erf<160 considerable potential risk; 160 Erf<320 high potential risk; and Erf≥320 significantly very high.

Statistical Data Processing

Statistical data processing was performed using Microsoft Excel 2016. Correlation was calculated on the concentration of Cd, Ni, and Pb in onion, garlic, and potatoes. Statistical analysis of the data was performed using the "Pearson correlation coefficient" [37, 38].

Results and Discussion

Heavy metal concentration in the analyzed soil is presented in Table 1. The analysis showed much higher concentrations of Cd and Pb in the soil from the areas closer to the smelter (Zvecan and Frasher) compared to the permissible limits stated in the European Commission guidelines (EC-DG, 2009) [33]. For example, the Cd concentration in Zvecan is 5.6 times higher than the EU limit, and the Pb concentration is 13.4 times higher. In Frasher, Cd is 5.3 times and Pb is 9.81 times higher than the allowable limits. High levels of Cd and Pb have also been found in other areas near old mines and smelters, mainly due to long-term industrial deposition and the low mobility of lead in soil [39].

The values of the contamination factor and the environmental risk factor are given in Table 2. In this study, Cf<1 for the Ni in all three regions, which means that in these regions, there is a low level of contamination for these metals. Moderate contamination is found in the Polac region for Pb (Cf = 2.14). There is a very high level of contamination for Cd in all three regions and for Pb in Zvecan and Frasher. The Zvecan region has the highest contamination for Cd (Cf = 28.00), which is almost close to the results of Raj and Maiti, and for Pb (Cf = 67.00), which is much higher than those of Raj and Maiti's research [19]. Similar results showing high contamination and environmental risk from cadmium and lead have been found in mangrove areas and mining sites, showing that these metals can cause harm in many different places [23, 25].

The ecological risk factor was <40 for Ni in all three regions and for Pb in Polac, indicating a low ecological risk for these elements. The Polac region has a high ecological risk factor for Cd, and the Frasher region has a high ecological risk factor for Pb. A significantly high ecological risk for Cd is presented in the Frasher and Zvecan regions, and for Pb in the Zvecan region.

Table 1. Heavy metal concentration in the analyzed soil.

Region	Cd (mg/kg)	Ni (mg/kg)	Pb (mg/kg)
Zvecan	8.4	50.5	1340
Frasher	7.9	66.1	981
Polac	1.9	48.8	42.7
European Commission (EC-DG) (2009) [39]	1.5	70	100

Table 2. Contamination factor and environmental risk factor.

	Regions	Cd	Ni	Pb
Cf	Zvecan	28.00	0.74	67.00
	Frasher	26.33	0.97	49.05
	Polac	6.33	0.72	2.14
Erf	Zvecan	840.00	0.74	335.00
	Frasher	790.00	0.97	245.25
	Polac	190.00	0.72	10.68

Heavy metal concentration in the analyzed vegetables (dry mass) of onion, garlic, and potato is presented in Table 3.

Of all the analyzed metals, the lowest concentrations were determined for Cd. In the Polac region, which is the furthest from the smelter plant, Cd and Pb were not detected, and in the regions that are closer to the smelter, the concentration of Cd is similar in the three types of vegetables - it coincides with the previously reported data presented in Table 4.

Garlic from the Frasher region showed the highest Ni value.

Furthermore, Pb as well as Cd were not detected in any of the analyzed vegetables in the farthest area from the smelter, Polac. The highest values for Pb were detected in potato, and the lowest in garlic. These values for Pb concentrations in examined vegetables are higher than the permitted limits, but are in accordance with literature data Table 4. In places affected by factories and industry, root vegetables like potatoes often collect more lead than the leafy ones, raising concerns about potential health risks [21].

The bioconcentration factor, which is the ratio of the metal concentration of the vegetables, dry mass, to the soil, dry mass, is given in Table 5.

Table 3. Heavy metal concentration in the analyzed vegetables (dry weight).

Vegetables	Region	Cd (mg/kg)	Ni (mg/kg)	Pb (mg/kg)
Onion	Zvecan	0.394	14.8	1.269
	Frasher	0.252	25.5	n.d.
	Polac	n.d.	15.2	n.d.
Garlic	Zvecan	0.353	29.1	0.474
	Frasher	0.218	92.3	0.533
	Polac	n.d.	27.9	n.d.
Potato	Zvecan	0.303	16.2	2.364
	Frasher	0.236	20.2	2.536
	Polac	n.d.	12.9	n.d.
Allowable limit *		0.05-0.2	10	0.1-0.3
FAO/WHO (2001) [40]		0.2	-	0.3

n.d. - not detected (Cd<0.0001 mg/kg; Pb<0.001 mg/kg); *Allowable heavy metals limit (mg/kg) in vegetables as described by Gebeyehu and Bayissa [41].

Table 4. Heavy metal concentration in vegetables (dry weight) from previous research.

Vegetables	Cd (mg/kg)	Ni (mg/kg)	Pb (mg/kg)	References
Onion	4	28	13	[42]
	0.00-0.30	0.13-1.66	0.19-2.21	[43]
	0.76	18.37	10.29	[44]
	0.97	-	8.7	[45]
	0.05	-	0.33	[32]
	0.36	-	1.62	[46]
Garlic	0.19-0.21	1.23-9.85	0.26-0.38	[47]
	0 -1.211	-	0.039-0.757	[33]
Potato	30	27	43	[42]
	0.17-0.21	3.37-6.04	0.26-0.49	[47]
	0.61	-	3.62	[46]
	0.88	10.74	4.50	[44]
	0.005	2.115	0.472	[48]

Table 5. Bioconcentration factor.

Bioconcentration factor				
Heavy metals	Region	Onion	Garlic	Potatoes
Cd	Zvecan	0.047	0.042	0.036
	Frasher	0.032	0.028	0.030
	Polac	n.d	n.d	n.d
Ni	Zvecan	0.293	0.576	0.321
	Frasher	0.386	1.396	0.306
	Polac	0.311	0.572	0.264
Pb	Zvecan	0.001	0.000	0.002
	Frasher	n.d,	0.001	0.003
	Polac	n.d.	n.d.	n.d

*n.d. - not detected.

The bioconcentration factor is an important parameter in phytoremediation and indicates the absorption of heavy metals into plant tissues from the soil. Values >1 indicate greater absorption of heavy metals into plants, and for values <1, the concentration of heavy metals in the soil is greater than in plants [18]. Table 7 shows that the bioconcentration factor for nickel in garlic grown in the village of Fraşer is greater than 1 (1.396), indicating that the concentration of nickel in this region is greater in the plant itself than in the soil where the plant is grown. The bioconcentration factor for Cd is much higher than the bioconcentration factor for Pb, but it is still less than 1. In Ba's research, the bioconcentration factor for all elements in several plant species was <1 [24], however, Anişoara's research showed BCf>1 for Cd in wheat [22]. Al-Kahtany highlighted that metal uptake by plants varies based on both the specific element and its interaction with soil properties, particularly in the case of nickel, zinc, and cadmium. These findings align with the trends observed in this study [28].

All analyzed vegetables have the lowest BCf values for Pb, i.e., Pb is the metal that is most difficult to absorb in vegetables. The mobility of Pb in the soil is poor because once it enters the soil, it is very easily adsorbed by organic matter or minerals. Then, Pb forms insoluble substances with the organic matter, which reduces its absorption by plants [21].

The results showed that the BCf values in onion for Cd are lower than those reported by Bedasa [45].

Table 6. Heavy metal concentration in the analyzed vegetables (fresh weight).

Vegetables	Region	Cd (mg/kg)	Ni (mg/kg)	Pb (mg/kg)
Onion	Zvecan	0.056	2.102	0.180
	Frasher	0.033	3.341	n.d.
	Polac	n.d.	1.900	n.d.
Garlic	Zvecan	0.134	11.087	0.181
	Frasher	0.078	32.951	0.190
	Polac	n.d.	9.486	n.d.
Potato	Zvecan	0.052	2.770	0.404
	Frasher	0.052	4.424	0.555
	Polac	n.d.	2.477	n.d.
FAO/WHO (2023) [53]		0.05* 0.1**	-	0.1

n.d. - not detected; *onion and garlic; **potato.

In potato and garlic, the BCf values for Cd are lower than those obtained by Mahmud and Malik [47]. On the other hand, Ni has the highest BCf values of the determined metals. In all analyzed regions, garlic has the highest BCf values for Ni; compared to potato and onion, it has the greatest ability to extract Ni from the soil and accumulate it within itself. Similar findings were reported by Zeneli [49], who observed elevated concentrations of Ni in fruits collected from the same region, indicating a broader pattern of heavy metal accumulation in local agricultural produce.

The concentration of heavy metals in fresh vegetables is calculated using Equation (4) (EPA, 2018) [50].

$$C_{ww} = C_{dw}\left[\frac{100 - W}{100}\right] \quad (4)$$

Where: C_{ww} is the concentration of the metal in fresh vegetable mass; C_{dw} is the concentration of the metal in the dry mass of vegetables; W is the water content in %. The water content (W) was determined in each vegetable from each region. It ranged from 85.8% to 87.5% in onions, from 61.9% to 66% in garlic, and from 78.1% to 82.9% in potatoes [51, 52].

Table 6 presents the heavy metal concentrations in vegetables (fresh weight) compared to the Maximum Allowed Concentration (MAC) of the Codex Alimentarius [53]. The concentration of Cd and Pb in onion from the Zvecan area and garlic from the Zvecan and Frasher areas is higher than MAC. Cd in potato from Zvecan and Frasher, although at high concentrations, is still within the permissible limits; however, Pb from these areas is above the MAC. The highest concentrations of Ni were measured in garlic, and the lowest in onion.

Table 7. Correlation coefficient between the content of heavy metals (Cd, Ni and Pb) in tested vegetables.

	Onion	Garlic	Potatoes
Onion	1		
Garlic	0.9978305	1	
Potatoes	0.9863005	0.9474403	1

The results indicate a strong positive correlation between the concentrations of heavy metals in all three vegetables. The highest correlation was between onion and garlic (0.9978), followed by onion and potato (0.9863), and the lowest (but still strong) correlation was between garlic and potato (0.9474). These findings suggest a common source of contamination, likely from the nearby smelter.

Conclusions

Analyzed vegetables from regions near the smelter (Zvecan and Frasher) showed high concentrations of cadmium (Cd) and lead (Pb). The only exception was lead in onions from Frasher. In contrast, in the Polac area, the furthest from the smelter, these metals were not detected at all. The concentration of Cd in onion from Zvecan, garlic from Zvecan and Frasher, and Pb in all fresh vegetable samples were above the MAC. Garlic has been found to have the highest concentrations of Ni compared to other examined vegetables. Soil analyses showed a difference between the areas near the smelter (Zvecan and Frasher) and the more distant Polac area, especially for Cd and Pb, which are above the concentrations for these metals according to EU regulations.

The highest BCf value of 1.396 was determined for garlic from Frasher, while all other BCf values were <0. BCf values for Cd were highest for onion, for Ni for garlic, and for Pb for potato. Another analyzed factor, Cf, indicates very high soil contamination with Cd and Pb in the regions of Zvecan and Frasher near the smelter. Also, a very high ecological risk factor – Erf for Cd in Zvecan and Frasher, while for Pb in Zvecan. These values are in line with the obtained high values for Cd and Pb determined in vegetables from these regions as well.

The accumulation of Cd, Ni, and Pb in the soil, in onion, garlic, and potato, from these areas, represents a significant environmental and health problem for human beings. This study will contribute to further monitoring of heavy metal pollution and their distribution near the industrial zones, as well as near mines and smelters, not just in the examined regions, but also in other parts of the country and region.

Conflict of Interest

The authors declare no conflict of interest.

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