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RISK ELEMENTS' INPUT INTO THE FOOD CHAIN IN OLD LOADED LOCALITIES

METALE CIĘŻKIE W ROŚLINACH UPRAWNYCH UPRAWIANYCH W POBLIŻU DAWNYCH ŹRÓDEŁ ZANIECZYSZCZEŃ

Abstract: The aim of this work is to evaluate the potential influence of three various sources on soil hygiene and plant production in their vicinity. In the soil of the first observed area in the vicinity of the former Nickel Smelter in Sered, the contents of Cd, Ni, Cu and Co were higher than the background values. Agricultural production grown in this area does not pose any risk to human organism with the exception of Cd in barley grain grown in one locality. The soil of the second observed area in the vicinity of Iron Ore Mines in Rudnany in loaded area of Middle Spis, can be evaluated as highly contaminated with As, Cu and Hg. Potatoes are at the highest risk in this area, but determined contents of risky metals in cereals are also higher than the legislative limits of the Food Codex (FC) of the Slovak Republic. In the third observed locality, Zahorska Lowland, in the vicinity of 5 municipal landfills, the determined total Cu, Cd and Ni soil contents were higher than the background values and the metal contents in grain of cereals were higher than the legislative limits. The results suggest that the old loaded localities present potential danger of risky elements input into the human food chain.

Keywords: old loaded areas, soil contamination, risky elements, human food chain

The old loaded localities present potential danger of risky elements input into the human food chain. The old mines, scrapheaps, landfills, industrial and municipal waste are important sources of environmental contamination. The aim of this study is to evaluate the potential influence of three various sources on soil hygiene and plant production in their vicinity. In the past these localities were intensively contaminated by metallic emissions from metallurgical factories as a dominant source of environmental contamination.

The first locality in the wider vicinity of the former Nickel Smelter in Sered is situated in the region of Lower-Povazie, one of the 8 loaded regions in the Slovak Republic [1]. The Nickel Smelter Sered began its activity in 1964. It was oriented at electrolytic nickel and cobalt production, using special steel and nickel salts produced

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mainly for the chemical industry. The Albanian iron-nickel ore was the basic raw material for this production. The nickel production appeared to be inefficient because of low quality and high cost of Albanian ore treatment [2]. The nickel production ended in February 1993 and the cobalt production ended in June 1993 [3]. During its activity, the Nickel Smelter produced waste which was cummulated in the form of leached scrap (mineral residuum of nickel ore in nickel and cobalt production) in the neighbourhood of the factory. The scrapheap is to be found in Sered and Dolna Streda territory on the right riverbank of the Vah. It has triangular configuration, its extent is about 32 hectares. According to the calculations, the scrapheap's cubature is 3 950 000 m^3, the weight equals 84 million Mg, the height is estimated to be 20 m. The weight of NH_4^+–N in the scrapheap measures aroud 1 848 000 kg. [4]. The black colouring of the leach scrap caused waste overheating, especially in the summer. Because of the second dusting this waste is the source of potential risky input of heavy metals into the soil near the factory. The environmental risk includes the potential contamination of the underground water in the alluvial gravel of the river Vah. The ash from the heating of the Nickel Smelter is another source of environmental contamination. The waste dump is situated in the territory Dolna Streda in the area of approximately 26 hectares. Both dumps can be the potential sources of underground water contamination by the enhanced ammonium and sulphate ion content.

The leach scrap composition is presented in Tables 1 and 2 [5].

Table 1

Chemical composition [%] of the Albanian leach scrap

Fe_{total}	Fe^{2+}	Fe^{3+}	Fe_{metal}	SiO_2	Al_2O_3	CaO	MgO	Cr_2O_3	NiO
45.89	17.60	26.29	1.32	15.03	4.80	3.54	2.21	1.06	0.17

Table 2

Mineralogical composition [%] of the Albanian leach scrap

Magnetite	Quartz	Wustite	Calcite	Ferrochromiumpicotite
54.09	13.15	8.02	6.32	5.51

Currently, the experiments with biological recultivation of scrapheap are realised. To prevent dusting of scrap with wind water is applied on the surface of the dumping site. Wetting of the black mud surface lowered the dusting, however, it is not sufficient for the elimination of emission transportation by wind. Gradual recultivation has been developed on the hills of the dump or in the vicinity of this area with the aim to hide the view on the dump and to prevent dust dispersion, waste spreading, hill fixing and erosion processes. The aim of the recultivation is to utilize the recultivated area for agricultural purposes and afforestation. Agricultural utilization requires the elimination of dump impact on crops, ie the safety of surface waste layers and cover, degassing, dewatering of the dump and the leachate from the dump. Currently, there is about 1/3 of dump covered with grass and weeds, the attempts to recultivate the whole area have not been very successful.

The second locality where the survey was conducted was the wider vicinity of Iron Ore Mines in Rudnany. In the past, this enterprise belonged to one of the main sources of emission contamination of the environment in loaded area of Middle Spis. The first record about the copper ore deposit in the mentioned locality comes from 1332. The year 1874 made a breakthrough in the mining period. Mining for iron siderite ores had begun after mining for copper and silver ores. Precise records about ore mining have existed in Rudnany since 1899. In that year, the mining activities represented $114.7 \cdot 10^9$ g. The great progress of mining in Rudnany was recorded at the end of the 18^{th} century and in the first decade of the 19^{th} century, when copper and silver ore minig were in their maximum. As the consequence of the world economic crisis in 1932–1933 the mining was stopped. After the liberation of Slovakia and especially after 1948, there was a big boom of mining and Iron Ore Mines Rudnany became the biggest ore enterprise in the former Czechoslovak Republic and the biggest exporter of mercury. Maximal ore mining of $968.5 \cdot 10^9$ g was achieved in 1978. Gradually, the reserves of siderite, chalcopyrite, tetraedrite with the mixture of mercury, silver and antimony had been running out. After 1989, all mining activities declined. In 1992, mining of siderite ore was finished as the consequence of this social phenomenon.

Waste presents a complex of ecological, economic and also social problems. Waste disposal takes place in all stages of production and consumption cycle. At present new possibilities of waste utilization beside classical disposal in various areas of economy are studied [6]. Out of the total volumes of generated waste in the Slovak Republic, 43 % of waste was disposed, which in absolute numbers means 6 185 272 Mg of waste. Dominance of landfill waste is a historical rule, with a 91 % share of total waste disposal. As of December 31, 2006, there were 160 landfills operating in Slovakia [7].

In the third observed locality, Zahorska Lowland, the influence of the 5 former or present landfills on soil and plant heavy metal contents in their vicinity is estimated. The landfill Mokry Haj began its activity in 1992 with the assumed closure in 2013. Biela Jama arose in 1975 as the material hole. Despite its recultivation in the 1990s, at present this place is utilized as municipal landfill. The landfills in Skalicke hory and Hrudy were built in the 1970s. The first of them was utilized for dangerous industrial waste, the second for municipal waste. After recultivation of landfill Hrudy agricultural plants are grown in this area. The landfill in the vicinity of the machinery factory in Skalica was localized aside the sewage tank. Currently this area is utilized for agricultural production.

Materials and methods

The observation of risky elements in soils and in cultivated crops were realized on plots in the cadasters of the villages: Zavar, Krizovany, Vlckovce, Hoste, Mala Maca and Velka Maca, in the distance of 5 to 9 km in the northern and north-western part and in cadasters of villages Velka Maca, Sintava and Vinohrady nad Vahom in the distance of 2 km south-west, 2.8 km east and 3.5 km north-east of former emission source of the Nickel Smelter Sered.

In the Middle Spis region, the soil samples and cultivated agricultural products were taken in cadaster of Markusovce in the distance of 10–12.5 km north-west, Matejovce –

7.6 km north, Chrast nad Hronom – 10 km north-east and Poráč – 6 km east of former emission source in Rudnany.

In the third observed locality, Zahorska Lowland, the soil samples and cultivated crops in 5 observed areas were taken.

The soil samples from the surface soil layer were taken from the experimental sites with pedological probe. Samples of soil ground of fine soil I. (average 2 mm) were analysed and from this fine soil the representative sample was taken and sieved through the sieve with an average of 0.2 mm (fine soil II). The total content of risky elements was determined using the AAS method in soil extract gained after total decomposition of soil using the wet way with the mixture of acids HF + HNO_3 + $HClO_4$.

The plant material was taken in full ripeness from the same localities as the soil. The samples of plant material were dried and homogenized before the analysis. The content of risky metals in plant material was determined using the AAS method after its previous mineralization using the dry way.

Results

Table 3 presents the values of selected heavy metals soil contents in the surveyed localities in wider vicinity of the former emission source, the Nickel Smelter Sered. As regards the risky metal contents, these soils can be considered as relatively "clean", in spite of moderately enhanced contents of Cd, Cu, Ni and Co. There was no content value indicating analytical proof of soil contamination. The Cd content was in the range from 1.6 to 2.5-fold of background value in all observed plots. The Ni content exceeded the background value in all plots with the exception of the locality of Vinohrady nad Vahom, while it was enhanced from 1.1 to 1.4-fold. The copper content was increased in the localities with the exception of cadasters of Zavar and Velka Maca (1.03 to 1.93-fold of background value). The enhanced soil content of Co was observed in villages Zavar, Hoste, Velka Maca and Sintava (1.06 to 1.22-fold of background value).

Table 3

Total content of heavy metals [$mg \cdot kg^{-1}$] in soil samples in the vicinity of emission source Nickel Smelter Sered

Locality No.	Cr	Cd	Cu	Fe	Mn	Pb	Zn	Ni	Co
1	68.4	2.00*	46.4*	24900	664.8	43.2	86.0	40.0*	18.8
2	67.2	1.88*	46.4*	22440	790.0	34.4	102.4	38.8*	18.4
3	62.0	1.40*	28.4	24128	662.0	48.4	88.8	41.2*	22.8*
4	71.2	1,90*	43.0*	27720	556.0	33.2	86.0	49.2*	20.0*
5	70.4	1.68*	37.2*	25880	636.4	36.4	91.6	41.2*	19.2
6	66.0	1.28*	26.8	25088	687.2	51.2	86.0	44.0*	21.6*
7	34.0	1.92*	25.2	24640	775.6	35.6	85.2	42.8*	21.2*
8	31.2	1.32*	44.4*	22440	1062.0	33.6	63.2	38.4*	24.4*
9	22.4	1.40*	69.6*	19880	743.2	32.4	63.6	30.4	17.6

* Exceeded the background soil content of the element.

All determined values were deeply below the hygienic limit, which in the case of enhancing means soil contamination.

The situation in the wider vicinity of the second surveyed site of the former emission source was diametrically different (Table 4).

Table 4

Total content of heavy metals in soil samples in the vicinity of emission source (ES) Rudnany

Locality No.	Cr	Cd	Cu	Fe	Mn	Pb	Zn	Hg	As
10	2.60	0.25	90.00*	2168	352.15	12.65	17.55	20.64***	25.00
11	1.50	0.24	73.27*	2348	324.30	12.15	12.85	38.00***	43.40**
12	2.40	0.26	29.30	2447	339.20	14.10	8.35	19.06***	15.00
13	1.40	0.45	11.40	1014	294.60	16.30	14.80	1.50*	12.00
14	1.70	0.35	16.85	1499	298.60	15.85	8.75	3.16**	22.60
15	1.90	0.23	13.75	1886	315.25	12.35	10.25	3.00**	23.70
16	2.30	0.38	14.65	1776	288.10	14.65	12.70	2.00**	18.40
17	3.40	0.25	19.30	2875	573.10	14.55	17.00	1.50*	12.00
18	3.20	0.24	14.30	2949	421.80	14.60	19.60	0.18	9.50
19	1.80	0.62	28.95	1392	918.50	25.75	27.05	9.30**	27.00
20	2.70	0.44	114.65**	3442	986.80	16.20	25.70	39.10***	32.00**
21	2.30	0.19	9.30	1890	249.60	14.70	10.25	0.40*	12.00

* Exceeded the background soil content of the element; ** Analytical proof of soil contamination by a certain element; *** Need of soil sanation with regard to the extremely high soil content of the element.

The enormly high content of Hg was found in cadaster of Markusovce in the locality Olsanske Pole, which was even exceeding the hygienic limit determining the sanation of the soil (1.9 to 3.8-fold). Similarly the extremely high content of Hg was determined in cadaster of Porac village, where the 3.2-fold higher exceeding of this limit value was determined. The contamination of soil by mercury was obvious also in other observed localities: Pod Horky, Zemkovske (Markusovce) and Na Strani (Matejovce), while the exceeding of limit value indicates analytical proof of soil contamination, and it was exceeded 4.65-fold.

Similarly, the soil contamination was proved in case of arsenic in cadasters of Markusovce village (locality Olsanske Pole), where the limit value was exceeded 1.45-fold and Porac (locality Pasienky) with 1.06-fold of limit. In Porac, the soil contamination by copper was evident (1.15-fold of limit).

The determined content of risky elements in soil samples of the observed localities in Zahorska Lowland are deeply below background values with the exception of Cd content in all localities, Cu content in Hrudy and Skalica and Ni content in Hrudy, Mokry Haj, Skalicke Hory and Skalica (Table 5). The problem of enhanced Cd content in soil is evident in many localities of the Slovak Republic and the potential risk of its input into the food stuffs of plant origin is high [7–9]. As far as other risky metal contents are concerned, the soils in the observed locality can be considered as relatively "clean".

Table 5

Total content of heavy metals [mg · kg^{-1}] in soil samples in the vicinity of landfills in Zahorska Lowland

Locality No.	Cr	Cd	Cu	Fe	Mn	Pb	Zn	Ni	Co
22	17.6	0.97*	11.3	6843	220.5	20.3	19.7	16.7	9.5
23	73.6	1.01*	42.7*	26592	486.5	33.2	70.0	51.1*	18.5
24	55.4	0.80*	32.1	21328	755.9	32.1	53.6	38.0*	16.9
25	78.3	1.16*	35.9	29437	529.3	30.3	79.3	54.0*	18.7
26	63.5	0.95*	41.7*	21019	527.9	54.8	66.4	35.9*	15.3

* Exceeded the background soil content of the element.

The total content includes all forms of heavy metals in soil. The human health risk is determined by the bioavailable forms of the element and by the plant species and crop grown on the metal loaded soil.

The values of determined contents of risky elements in crops cultivated in the surveyed localities are presented in Tables 6–8. The content of risky metals in overground biomass (straw) was evaluated according to the valid legislation for forage, the content of metals in edible parts (grain) of crops and in foodstuffs of plant origin was evaluated according to the Food Codex of the Slovak Republic (FC SR).

Despite the relatively "clean" soils, the agricultural production grown in the investigated localities in wider vicinity of former emission source of Nickel Smelter Sered presents a potential human organism risk from the point of Cd content in barley grain grown in Vlckovce (1.25-fold exceeding of hygienic limit) and Pb in all the crop samples used for human nutrition (Table 6). The overground biomass from the point of its utilization as forage showed the enhanced content of Cr (1.54-fold) in wheat and Co (1.3-fold) in sunflower from Velka Maca in comparison with defined hygienic limits.

The situation in the second observed locality is entirely different. The cereals are characterised by a high content of metals: grain of barley from Markusovce Cd and As (the content on the level of the limit value), Hg 1.8 to 2-fold and Pb even 2.65-fold of hygienic limit; grain of rye from Markusovce Hg 1.6-fold and grain of wheat from Chrast Cd 1.5-fold and Hg 3-fold of the maximum acceptable amount for foodstuffs (Table 7). From the crops grown in wider vicinity of former emission source Iron Ore Mines Rudnany, potatoes cultivated in Markusovce (Olsanske Pole) are the most risky. The determined contents of Cd, Cu, Pb, Zn and Hg were 1.4-fold, 3.1-fold, 1.7-fold, 1.43-fold and 6-fold, respectively, higher than hygienic limits defined in the Food Codex of the Slovak Republic. The Hg content was higher also in the overground biomasss of barley from Markusovce, where it reached even 4-fold and in grassland in Porac, where the 2-fold of hygienic limit defined for forage was found.

In Table 8 the results of determined heavy metal contents in crops grown in the vicinity of landfills in Zahorska Lowland are presented. The contents of risky heavy metals in overground biomass of grown crops were deeply under the hygienic limits given by the legislation for forage. From all the tested crops the Pb content in grain in the observed localities was higher than the hygienic limit given by the Food Codex of

Table 6

Heavy metal content [mg · kg^{-1}] in samples of crops grown in the vicinity of ES Nickel Smelter Sered

No.	Crop		Cr	Cd	Cu	Fe	Mn	Pb	Zn	Ni	Co
1	barley	s	1.60	0.260	7.10	50.4	24.9	0.70	18.5	1.20	0.80
	barley	g	1.25	0.125**	7.14	41.8	16.3	0.50**	36.9	0.29	0.22
2	barley	s	1.90	0.300	6.40	49.2	46.5	1.50	20.4	1.10	1.10
	barley	g	1.40	0.055	8.50	49.4	20.8	0.50**	38.3	0.40	0.30
3	maize	s	2.00	0.220	9.40	85.1	25.2	2.00	40.4	0.90	0.90
4	wheat	s	3.39	0.197	10.61	66.1	26.4	0.98	13.4	0.77	0.77
	wheat	g	0.50	0.085	4.10	43.6	34.3	0.35**	29.2	0.35	0.35
5	sunflower	s	2.40	0.560	23.10	112.8	59.3	3.90	42.2	2.60	2.60*
6	wheat	s	4.63*	0.375	11.25	60.9	45.9	3.53	13.0	1.10	1.32
	wheat	g	0.45	0.055	1.75	56.2	36.6	0.20**	33.2	0.35	0.20
7	paprika		0.25	0.055**	0.81	17.1	2.3	0.85**	2.3	0.08	0.04
	tomatoes		0.13	0.022	0.49	6.7	0.9	0.31**	1.5	0.13	0.05
	carrot		0.04	0.025	0.48	4.4	1.3	0.22**	2.2	0.11	0.07
	parsley		0.43	0.031	1.80	12.8	3.4	0.59**	3.9	0.27	0.09
8	sugar beet		0.04	0.035	0.91	20.7	4.8	0.12	4.0	0.20	0.08

* Exceeded hygienic limit for forage (s = straw); ** Exceeded hygienic limit for foodstuffs of plant origin (g = grain).

Table 7

Heavy metal content [$mg \cdot kg^{-1}$] in samples of crops grown in the vicinity of ES Iron Ore Mines Rudnany

No.	Crop	Cr	Cd	Cu	Fe	Mn	Pb	Zn	Hg	As
10	colza	0.75	0.20	7.9	88.6	34.0	2.16	37.7	0.05	0.20
11	barley s	1.22	0.22	7.4	92.6	10.9	2.97	13.2	0.40*	0.30
	barley g	0.74	0.10**	7.0	54.3	14.4	0.90**	25.7	0.10**	0.20**
12	rye s	0.13	0.01	5.5	33.8	33.9	0.85	28.5	0.09	0.20
	rye g	0.13	0.01	4.8	31.3	33.6	0.73**	24.4	0.08**	0.15
13	grassland	1.11	0.19	6.6	161.9	36.2	1.69	21.1	0.05	0.40
14	clover	1.27	0.27	13.1	120.8	28.7	2.29	20.9	0.06	0.40
15	barley s	0.64	0.09	5.9	66.9	14.1	1.32	20.2	0.10*	0.16
	barley g	0.26	0.06	4.9	31.6	34.8	0.77**	22.3	0.09**	0.12
16	barley s	1.11	0.17	6.4	49.9	10.1	0.72	20.2	0.20*	0.30
	barley g	0.59	0.07	4.6	40.9	15.4	2.65**	23.3	0.09**	0.10
17	potatoes	0.50	0.14**	9.4**	28.8	5.5	1.70**	14.3**	0.12**	0.10
18	grassland	1.27	0.27	13.8	136.3	26.0	2.51	23.5	0.05	0.30
19	grassland	1.31	0.21	8.3	127.2	44.0	1.47	22.2	0.05	0.40
20	grassland	1.00	0.23	11.6	168.0	44.0	1.53	39.6	0.10*	0.30
21	wheat g	0.43	0.15**	4.7	43.6	55.1	0.94**	29.4	0.15**	0.15

* Exceeded hygienic limit for forage (s = straw); ** Exceeded hygienic limit for foodstuffs of plant origin (g = grain).

Table 8

Heavy metal content [mg · kg^{-1}] in samples of crops grown in the vicinity of landfills in Zahorska Lowland

No.	Crop		Cr	Cd	Cu	Fe	Mn	Pb	Zn	Ni	Co
22	barley	s	1.70	0.270	2.00	53.8	18.1	1.60	14.1	0.80	0.70
	barley	g	1.20	0.065	10.15**	42.3	14.9	0.60**	32.4	0.35	0.35
23	wheat	s	1.90	0.250	3.40	88.7	10.3	0.90	33.7	1.00	0.80
	wheat	g	1.05	0.160	4.05	208.2	36.0	0.35**	29.8	0.95	0.30
24	barley	s	2.30	0.160	2.60	106.9	21.5	1.20	13.6	1.10	0.40
	barley	g	1.50	0.135**	4.55	55.8	14.0	0.45**	25.1	0.40	0.30
25	wheat	s	2.50	0.230	2.70	85.0	16.1	1.80	12.3	1.10	0.90
	wheat	g	0.70	0.120	4.65	54.9	14.2	0.45**	28.8	0.75	0.20
26	maize	s	1.80	0.140	8.60	88.9	80.9	1.30	24.5	1.10	1.00
	maize	g	0.60	0.035	1.45	26.2	6.50	0.30**	18.6	0.55	0.10

** Exceeded hygienic limit for foodstuffs of plant origin (g = grain).

the Slovak Republic. The grain of barley from the locality Mokry Haj contained an enhanced content of Cd and the same crop from the locality Biela Jama contained enhanced content of Cu in comparison with the hygienic limits**.** The negative influence of waste deposition in the observed landfills can be the potential cause of enhanced heavy metal contents in agricultural plants cultivated near the landfills.

The heavy metal mobility in soil depends on the soil reaction, the content of organic matter, the soil sorption capacity and it can be affected by the change of these factors [10]. The transfer of risky elements from the soil into the agricultural plants is influenced by soil properties as well as by plant species or variety [11, 12].

Conclusion

The results suggest that the investigated soils in the area of the former Nickel Smelter and in the area of the former or present landfills in Zahorska Lowland can be considered as "clean" from the point of heavy metal contents. In these areas, only weakly enhanced soil contents of some risky elements in relationship to their background values were found. Despite this fact, a high Pb content in the grown crops was confirmed. In this respect, lead can be considered as the most risky heavy metal with possible negative influence on human health.

Our results also suggest causal connection between the enhanced content of risky heavy metals, especially Hg, in soils and crops grown in the vicinity of former emission source in Rudnany and the former metallurgical activity in this area. Residual metal load of soil and possible contamination of the food chain in the area of Middle Spis presents a serious potential risk to human health. Therefore, it is important to control the waste treatment, to strictly follow the legislative directives and to monitor the risky matters input into the environment with the aim to inhibit the non-controlled pollution of the environment and to ensure food safety.

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METALE CIĘŻKIE W ROŚLINACH UPRAWNYCH UPRAWIANYCH W POBLIŻU DAWNYCH ŹRÓDEŁ ZANIECZYSZCZEŃ

Abstrakt: Celem pracy było zbadanie wpływu trzech różnych źródeł zanieczyszczeń na jakość gleby oraz produkcję roślinną. Zaobserwowana została zwiększona zawartość Cd, Ni Cu i Co w glebie w okolicy byłej huty niklu. Uprawy rolne na tym terenie nie stanowią jednak zagrożenia dla zdrowia ludzi z powodu stosunkowo małej zawartości metali ciężkich. Wyjątek stanowiło ziarno jęczmienia zawierające wysoki poziom kadmu. Na drugim badanym stanowisku – Kopalnia Rudy Żelaza Rudnany, Środkowy Spisz – stwierdzono dużą zawartość As, Cu i Hg w glebie. Największą ilość metali na tym terenie kumulowały ziemniaki. Ponadto w ziarnach zbóż przekroczone zostały obowiązujące w Republice Słowackiej normy zawartości metali ciężkich. Ostatni z badanych terenów to Nizina Zahorska. Na obszarze tym zbadano zawartość metali w glebie pól położonych w pobliżu pięciu składowisk śmieci. Uzyskane wyniki wskazały zwiększoną zawartość Cu, Cd i Ni w glebie oraz przekroczone limity dla zawartości tych metali w ziarnie zbóż. Przeprowadzone badania wykazały, że dawna emisja metali ciężkich do środowiska nadal stanowi źródło zanieczyszczenia żywności.

Słowa kluczowe: nieaktywne źródła zanieczyszczeń, zanieczyszczenie gleby, czynniki ryzyka, żywność