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Short Communication

Cadmium Content in Plant Products Cultivated Near a Copperworks

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Abstract

The contents of cadmium were determined in plant products cultivated in the neighborhood of the Głogów (n=127) and Legnica (n=18) copperworks. The contents were recorded using the atomic absorption spectrometry (AAS) method after dry mineralization, and cadmium was extracted to isopropylacetone. The results were compared with the acceptable levels established in Europe. The content of cadmium ranged between 0.0-76.0% of allowable values. The exceeded content of cadmium was found only in one sample.

Keywords: plant products, contaminants, cadmium, AAS

Introduction

Cadmium is expanded in the crust of the earth and its content ranges between 0.15-0.20 ppm. The main mineral that contains this metal is cadmium sulphide, which occurs both in zinc and copper ore deposits. In the soil of industrial areas, particularly in the neighborhood of non-ferrous metal plants, a large quantity of cadmium can be accumulated. Additionally, the soil contamination also can be caused by soil fertilization using fertilizers produced with contaminated sludge or the long-time application of phosphate fertilizers. Salts of cadmium and its complexes with organic compounds are easily solved in water, which helps with migration of this element in the environment. High cadmium absorption occurs mainly in "sour" soil, which is connected with excessive emission of sulphur to the environment. In plant products the allowable concentration of this element ranges between 0.05-0.2 mg/kg and is within the scope resulting from the physiological conditions of cadmium absorption by separate species of plants [1]. The accumulation of cadmium in plants, particularly in their vegetative parts, depends on soil reaction and its sorption properties. Also, the level of cadmium in the plant directly depends on the climatic conditions of cultivation and indirectly on agro-technical conditions [2].

Based on the analysis of new data in 2009, The European Food Safety Authority's Panel on contaminants in the food chain has set a reduced tolerable weekly intake (TWI) for cadmium of 2.5 micrograms per kilogram of body weight (μg/kg bw). A panel analyzed data (from 2003 to 2007) of cadmium occurrence in various foods consumed in 20 different countries alongside national dietary surveys and EU-wide consumption data collected by EFSA. It stated that locally produced food in highly contaminated areas may lead to high exposure levels [3]. The Legnica-Głogów region (western Poland) is one of the biggest copper-producing areas in the world. The KGHM Polska Miedź SA (Copper Mining and Smelting Combine, Poland – KGHM) is the world's sixth largest refined copper supplier. Copper mining and smelting causes the emission of about 400 noxious substances into the environment. The most harmful are metal compounds, including cadmium. Copper mining in the Legnica area caused dynamic industrial development of this region in the 1960s. At the same

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time, the air and soil contamination with trace elements increased as a result of particulate emissions, contaminated plant sewage, and flotation wastes [4]. The increased emission of sulphur dioxide from Legnica and Głogów smelters caused acid rain and sour soils. As a consequence of environment degradation, a protective zone was designated around Legnica and Głogów smelters, and almost all the inhabitants in the zones were resettled. Only a few farms remained in the Głogów smelter. In 2006 Polish law eliminated the conception of protective zones. Current law states that smelters are obligated to limit environmental impact to their own property, and therefore the land comprising the former protective zones remains in the possession of the smelters. Directly in environmental protection, the modernization of plants and investments (e.g. modern technologies of particulates elimination) considerably reduced the release of metals to the environment. In 1985-2002, the release of particulates to the atmosphere from the Copper Plant Głogów was reduced by 94% [5].

The complete elimination of the copper industry in the Legnica-Głogów area is not possible, even with the priority of environmental protection. Trace elements accumulated to the soils for many years caused the penetration of toxic elements to the cultivated plants and linked the food chains, particularly in the case of acid soil management as the lack of organic fertilization and liming. Based on the State Institute of Environmental Protection investigations, and according to the Decree by the Ministry of the Environment, in 2004 and 2007 the soil in the Głogów region was classified as contaminated from level I (elevated contents) to III (average contamination), including cadmium contents [6].

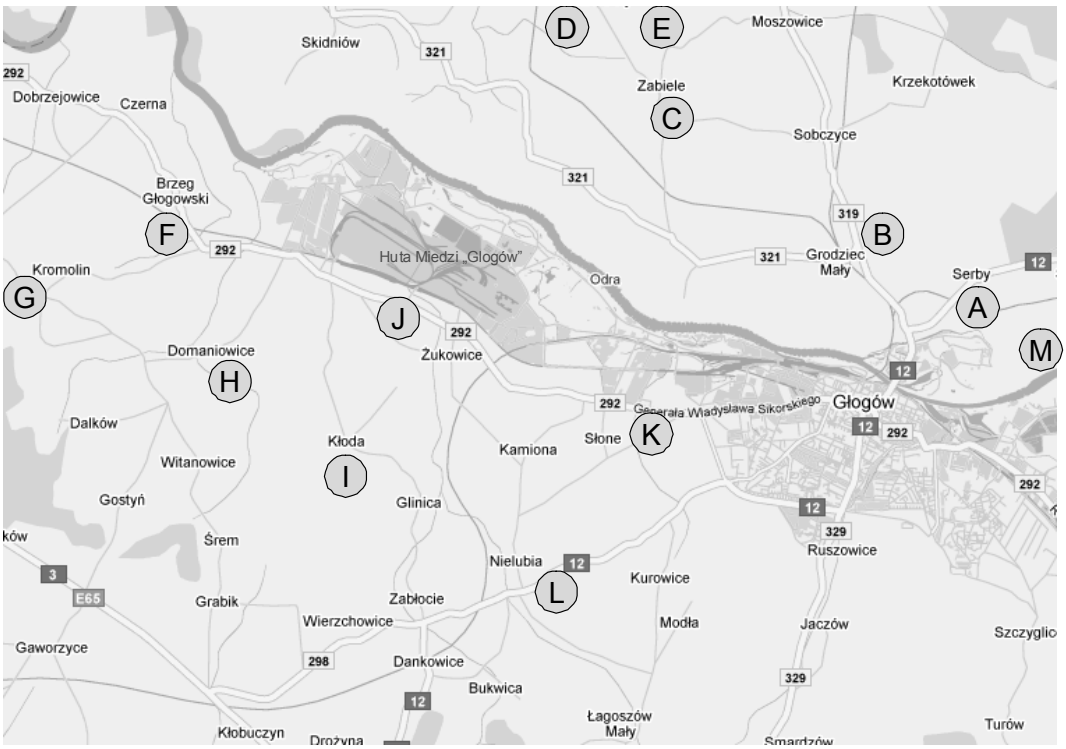
Plant products are a significant part of toxic trace elements of a daily food ration. It is estimated that for people who are not exposed to cadmium, and plant products are the source of 50-60% total amount of cadmium intake [7, 8]. The continuous monitoring of the foodstuff contamination on the area affected the copper industry is the key for countermeasures taken on the field of health and ecological policy. Following the changes of contamination associated with investments of environmental protection, it is necessary for proper evaluation and the possible follow-up actions.

The aim of these examinations was to evaluate the cadmium contamination in food products cultivated in the neighborhood of the Głogów and Legnica copperworks against the background of significant improvement to the environmental condition caused by reduction of particulate emissions by copper plants in past years.

Material and Methods

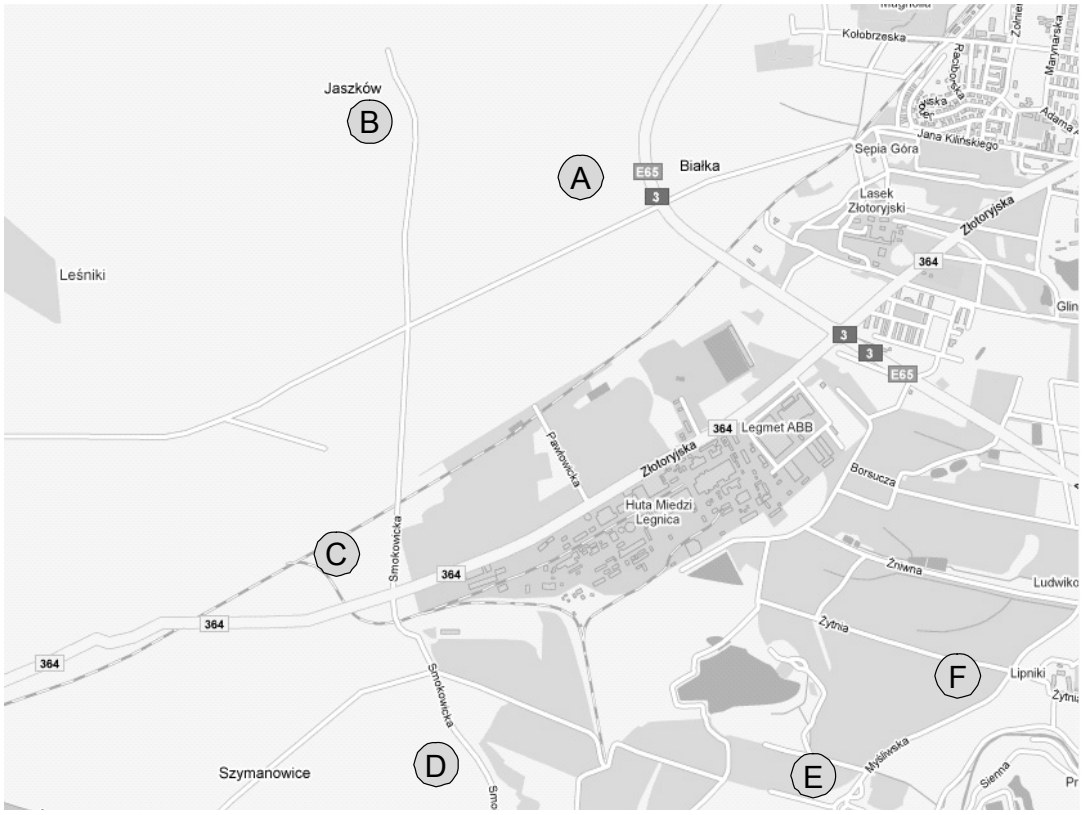
Cereal grains (wheat, barley, wheat-rye, rye), potatoes, vegetables (carrots, beetroots, cabbage, parsley tomatoes) and fruits (apples, pears) were examined. Cadmium contents were analyzed in 145 samples collected between September and October 2005 from farms located in the area around the Głogów (Fig. 1) and Legnica (Fig. 2) copperworks.

Products were collected directly from crop fields, gardens or storage places (cereals) both in the protection zone and the area outside the zone of the Głogów copperworks. Potatoes, vegetables, and fruit originated mainly from small



A – Serby
B – Grodziec Mały
C – Zabiele
D – Kozie Doły
E – Kotla
F – Brzeg Głogowski
G – Kromolin
H – Domaniowice
I – Kłoda
J – Żukowice
K – Słone
L – Nielubia
M– Klucze

Fig. 1. Places of samples colletion in Głogów smelter area (1 cm -approx. 2 km).



A – Polowice
B – Jaszków
C – Pawłowice Małe
D – Smokowice
E – Czerniewice
F – Lipniki

Fig. 2. Places of samples colletion in Legnica smelter area (1 cm -approx. 1 km).

growings were designed for local market and domestic use. Samples from the protection zone of Legnica copperworks were collected from gardens that had remained after farm liquidation or as wild berries.

Samples were collected by a staff from the Institute of Human Nutrition in the presence of farm owners and copperworks representatives. Products were taken from equally placed points depending on the size of the crop. Then material was put together into one crop sample of 1-3 kg. Products (except cereals) were processed in the same way as for consumption. Potatoes and root vegetable samples were washed, dried, peeled, and crumbled. After removing damaged leaves, vegetables were washed, dried, and crumbled. Fruits were washed, dried, and crumbled without peeling, but after removing the seed pockets. The part of a crumbled field sample (500-800 g) was prepared as an average laboratory sample, frozen and stored at -18ºC for further analysis. 500 g cereal grains samples were ground and kept at room temperature after removing the impurities.

To determine cadmium contents, 20 g samples were weighed and dry-mineralized in a muffle furnace at 450ºC for ca. 24 h. Incinerated samples were dissolved in 2% HNO_3(V), evaporated, and re-combusted at 450ºC. The obtained ash was dissolved in 25 mL of 2% HNO_3(V). The cadmium contents were determined with the application of atomic absorption spectrometry (AAS) and the use of a Varian AA240FS spectrometer. The determination of cadmium was carried out after treating the sample with ammonium pyrrolidyldithiocarbamate (APDC), and the extraction of isobutyl methyl ketone (MBIK) in accordance with European standards EN 14084:2004 and EN 13804:2002. Determination limit was specified as 0.0009 μg/ml in accordance with a regulation issued by the Polish Ministry of Health. The accuracy of determinations ranged between 3-7%. The average recovery of cadmium reached 98%.

Results and Discussion

The determined cadmium contents in the food products cultivated in the neighborhood of copperworks Głogów and Legnica were presented in Tables 1 and 2, respectively. Available data of cadmium contamination of investigated products published in the last decade in different Polish areas, as well as in other countries, are presented in Table 3.

Cadmium is an environmental contaminant because of increasing exposure and its established carcinogenicity. The anthropogenic mobilization index for cadmium is high due to the widespread use of the element in a number of industrial processes. Increased concentrations of this element in soil may occur close to industrial emission sources, particularly those of nonferrous mining and metal refining industries. The cadmium uptake in a specific plant species is proportional to the cadmium content of the soil and depends on other factors such as soil pH, content of clay, organic material and agricultural practice. Determined content of toxic metals in food products depends on the degree of environmental pollution. The copper mining region of Głogów and Legnica is highly polluted with metals, and increased levels

Table 1. Contents of cadmium in plant products from the area of Głogów copperworks in mg/kg and % of permissible values.

No.	Plant product	*n*	Places of sample collection marked in Fig. 1	Contents of Cd [mg/kg]		Permissible value [mg/kg]	Contents of Cd [% of permissible value]	
				$\bar{x}$±SD	min-max		$\bar{x}$	min-max
1	wheat	12	A, B, C, D, E, F, G, H, I, 2xJ, K, L, M	0.027±0.0053	0.019-0.037	0.2	13.5	9.5-18.5
2	rye	3	H, I, J	0.019±0.0125	0.01-0.033	0.1	19	10.0-33.0
3	wheat-rye	5	C, E, F, L, M	0.046±0.0393	0.016-0.114	0.1	46	16-114.0
4	barley	7	B, C, E, H, I, J, L	0.02±0.0051	0.011-0.025	0.1	20	11.0-25.0
5	potato	16	A, 2xB,C, D, E, F, G, H, 2xI, 2xJ, K, L, M	0.009±0.0041	0.004-0.021	0.1	9	4-21
6	carrot	13	B, C, D, F, G, H, 2xI, 2xJ, K, L, M	0.014±0.0128	0.004-0.05	0.1	14	4.0-50.0
7	beetroot	11	A, B, C, D, F, G, H, I, J, L, M	0.011±0.0107	0.004-0.039	0.1	11	4.0-39
8	cabbage	8	B, C, G, I, 2xJ, L, M	0.003±0.0012	0.002-0.005	0.2	1.5	1.0-2.5
9	parsley (root)	11	B, C, D, F, G, H, 2xI, J, L, M	0.007±0.0022	0.003-0.01	0.1	7	3.0-10
10	parsley (green)	12	2xB, C, D, F, G, H, 2xI, J, L, M	0.017±0.009	0.009-0.041	0.2	8.5	4.5-20.5
11	tomato	10	A, 2xB, C, D, F, G, I,2xJ	0.005±0.0019	0.003-0.009	0.05	10	6.0 -18.0
12	apple	12	A, B, C, D, F, G, H, I, J, K, M	0.002±0.0008	0.000-0.003	0.05	4	0.0-6.0
13	pear	6	A, B, C, G, H, I	0.004±0.0043	0.002-0.012	0.05	8	4.0-24.0

Table 2. Contents of cadmium in plant products from the area of Legnica copperworks in mg/kg and % of permissible values.

No.	Plant product	*n*	Places of sample collection marked in Fig. 2	Contents of Cd [mg/kg]		Permissible value [mg/kg]	Contents of Cd [% of permissible value]	
				$\bar{x}$±SD	min-max		$\bar{x}$	min-max
1	apple	6	A, 2xB, 2xC, D	0.003±0.001	0.002-0.004	0.05	5.3	4.0-8.0
2	pear	6	A, C, D, 2xE, F	0.007±0.001	0.004-0.008	0.05	13.3	8.0- 16.0
3	blackberry	3	D, E, F	0.01±0.007	0.005-0.018	0.05	20	10.0-36.0
4	hip (dog-rose fruit)	3	2xD, E	0.028±0.017	0.008-0.038	0.05	56	16.0-76.0

of mainly lead and copper in products from this area have appeared in previous investigations by our team as well as in other authors' works [9-11]. In our work, products were collected from areas affected by copper-smelting activities. Due to technical and logistical reasons, it was impossible to analyze other factors that might influence the product contamination (fertilization, soil properties, agro-technical conditions).

Data from reliable analyses performed in several countries indicate that most foodstuffs have a cadmium concentration in the range between 0.005-0.100 mg/kg [3, 12].

In determined products, excluding one sample, the cadmium content was rather low and ranged between 0.000-0.050 mg/kg. The contamination of determined cereals was found within allowable limits, except one sample of wheat-rye, for which the 114% of allowable quantity was determined. This sample was originated from one of a few farms remaining in the Głogów smelter protection zone. The average cadmium contents in wheat did not exceed 20% of allowable value. The contamination of other cereals was similar and ranged between 16-40% of allowable value. The estimated contamination of toxic metals in plant prod-

Table 3. Contents of cadmium in plant products from the different areas of the world.

Plant product		Contents of Cd [mg/kg]		Country (aera)	References
		$\bar{x}$	min- max		
Cereals	cereals grains	0.035		Poland (neighborhood of Głogów copperworks)	[9]
	wheat grains	0.148		Poland (southern Poland)	[16]
			0.04-0.07	Poland (other regions)	
		0.045		Poland (whole country)	[13]
		0.054		Poland (western Poland)	
		0.083		Poland (southeastern Poland)	
	wheat, barley, rye	<0.001		Finland	[14]
	wheat grains and flour	0.03		European countries	[3]
Potatoes	potatoes		0.004-0.049	Poland (neighborhood of Głogów copperworks)	[10]
		0.27		Poland (Legnica region)	[11]
			0.004-0.109	Poland (Kielce province)	[16]
			0.016-0.044	Poland (Tarnobrzeg industrial zone)	[15]
			0.001-0.024	Poland (Tarnobrzeg rural zone)	
			0.04-0.106	Poland (southern Poland)	[16]
			0.018-0.048	Poland (other regions)	
		0.009		Poland (north - eastern)	[13]
		0.017		Poland (north - western)	
		0.026		Poland (central)	
		0.009		Greece	[20]
		0.021		European countries	[3]
Root vegetables	carrots	0.016		Poland (neighborhood of Głogów copperworks)	[10]
			0.003-0.73	Poland (Kielce province)	[16]
		0.52		Poland (Płock Refinery region)	[18]
		0.011		Greece	[19]
		0.005		Greece	[20]
	beetroot	0.015		Poland (neighborhood of Głogów copperworks)	[10]
			0.008-0.038	Poland (Tarnobrzeg industrial zone)	[15]
			0.005-0.018	Poland (Tarnobrzeg rural zone)	
			0.003-0.163	Poland (Kielce province)	[16]
		0.27		Poland (Płock Refinery region)	[18]
		0.014		Greece	[19]
	parsley root		0.016-0.033	Poland (Tarnobrzeg industrial zone)	[15]
			0.009-0.024	Poland (Tarnobrzeg rural zone)	
			0.016-0.049	Poland (Legnica region)	[11]
		0.017		Poland (neighborhood of Głogów copperworks)	[11]
			0.005-0.122	Poland (Kielce province)	[16]
	root vegetables	0.0205		European countries	[3]

Table 3. Continued.

Plant product		Contents of Cd [mg/kg]		Country (aera)	References
		$\bar{x}$	min-max		
Leafy vegetables	parsley leaves	0.34		Poland (Płock Refinery region)	[18]
			0.011-0.099	Poland (Silesian region)	[17]
		0.016		Poland (neighborhood of Głogów copperworks)	[10]
			0.02-0.03	Greece	[19]
		0.006		Greece	[20]
	leafy vegetables	0.0231		European countries	[3]
Other vegetables	tomatoes		0.009-0.03	Poland (Tarnobrzeg industrial zone)	[15]
		0.011		Poland (Tarnobrzeg rural zone)	
		0.003		Greece	[20]
	cabbage		0.001-0.005	Poland (neighborhood of Głogów copperworks)	[10]
		0.006		Greece	[19]
		0.005		Finland	[14]
	other vegetables	0.0068		European countries	[3]
Fruits	apples		0.002-0.015	Poland (Neighborhood of Głogów copperworks)	[10]
		0.0093		Poland (Tarnobrzeg industrial zone)	[16]
		<0.001		Greece	[20]
		<0.001		Finland	[14]
	fruits	0.0039		European countries	[3]

ucts from the neighborhood of Głogów copperworks was performed in 1995-97 [9, 11]. In the majority of the cereals grain samples examined at that time, the quantity of cadmium was within 5-75% of allowable values and the average contents were 0.035 mg/kg. But it should be highlighted that introducing EU regulations in Poland has resulted in increasing the allowable values of cadmium in foodstuffs [4]. The determination of cadmium contamination in plant products ordered by the Ministry of Agriculture and Rural Development was carried out in Poland. The average cadmium contents in wheat grains recorded that they were found in higher amounts than obtained in our work. According to the Central Statistical Institute of Poland (GUS), the contents of cadmium in wheat grains from northern, western and southeasern parts of Poland were high [12]. Lower levels of cadmium in wheat, barley, and rye were assayed by Ekholm et al. [14] in Finland.

Potatoes are an important part of the Polish diet and are nursed in rural areas. On account is that potatoes cultivated in contaminated soils can be an important source of cadmium in the diet. In determined samples of potatoes, the recorded content of cadmium was below 21% of allowable value. In the analysis of potatoes cultivated in the neighborhood of Głogów copperworks conducted in 1995-97, higher levels of cadmium were reported [11]. The Szymczak et al. examinations [28] demonstrated higher average cadmium contents in potatoes from the Legnica area than those obtained in this work. In comparison to the results of our study, higher contents of cadmium have also been reported in potatoes from other regions of the country (both industrialized and rural) [11, 22]. From 1995 to 1998, cadmium contamination in Polish plant products showed higher contents of this element in potatoes from southern Poland than from northwestern and eastern parts of the country [7, 27].

Similar quantities of cadmium in potatoes were also found in Santiago, Chile [17]. The cadmium contents in the examined vegetables from Głogów smelter area were below 50% of allowable value. Higher average contents of cadmium were observed in carrots, beetroots, and parsley leaves, and lower in cabbage and tomatoes. In the analyzed roots (1995-97), the slightly higher levels of cadmium were reported in parsley, with similar results for carrots and beetroots [9]. The similar or higher values of cadmium contents were determined by Kocjan et al. [15] in parsley roots from rural regions in the neighborhood of Stalowa Wola, and from regions affected by the Tarnobrzeg Industrial Zone, respectively. The determinations of foodstuff from Legnica market conducted in 1994-96 showed higher cadmium contents in parsley roots [10]. Rembiałkowska et al. [16]

demonstrated that the concentration of cadmium in beetroots, parsley roots, and carrots from organic farming were statistically higher than cadmium concentrations recorded in root vegetables from standard crop. Higher contents of cadmium in parsley leaves and roots, carrots, and beetroots were determined in samples collected in the Mazovian Refinery and Petrochemical Works of the Płock region [17]. Investigations carried out in Greece compared the contents of metals in vegetables growing in two regions of the northwestern part of the country, characterized by low pollution (Preveza city) and high industrialization (Ioannina) [19]. In samples of parsley from both areas, the average contents of cadmium were similar to those obtained in the present work.

In analysis of carrot by Stalikas et al. [18] in Greece, the mean cadmium concentration was slightly lower than these presented in this paper. The average cadmium content in tomatoes was approx. 11% of allowable quantities. In comparison with this study, cadmium contents in tomatoes for industrial and rural areas of the country were similar or slightly higher [15]. The contents of cadmium in tomatoes analyzed by Karavoltsos et al. [19] were similar to the results obtained in the reported study. The average concentration of cadmium in cabbage was approx. 2% of allowable value and was found at the same level as that determined in this area in 1995-97 [9]. The content of cadmium in cabbage from the Greek market was similar to results obtained in the reported study. Similar levels of cadmium in cabbage were also assayed by Ekholm et al. in Finland [14]. The contents of cadmium in apples and pears from different locations in the neighborhood of the Głogów copperworks were just 2% to 6% of allowable quantity. Determined quantities of cadmium did not exceed 0.012 mg/kg, and were similar to that determined in 1995-97 in this region [9]. Examinations of fruits from the Tarnobrzeg Industrial Zone showed similar average cadmium contents in apples and pears for industrial and check areas [11]. Analyzed contents of cadmium in apples from the Greek market were lower than results obtained in the reported study [9]. Lower levels of cadmium in apples were assayed also by Ekholm et al. in Finland [5].

The contents of cadmium were determined on 18 product samples from the protection zone around the Legnica copperworks excluding consumption growing. The examinations covered apples and pears from former home gardens and wild blackberries and rosehips as well. The cadmium content in apples from the neighborhood of Legnica copperworks ranged between 0.002-0.004 mg/kg and did not exceed 8% of allowable quantities, and in the case of pears ranged between 8-16% of allowable quantity. In blackberries and rosehips, the quantities of cadmium ranged between 0.002-0.038 mg/kg, which corresponds to 4-7% of allowable values.

The presence of chemical contaminations including toxic metals is one of the basic criteria for evaluating food safety. The legal regulations for metal limits in foodstuffs are usually compromised between expected values from the health point of view, and it is actually possible to obtain satisfactory produce with low contamination levels.

EU Commission Regulation No. 466/2001 [4], which is referred to in Polish standards for maximum allowable contamination levels when the study was performed, recommends establishing maximum allowable cadmium contents as low as can practicably be achieved.

The obtained results show that cadmium contents in the investigated vegetables and fruits from the area of copperworks were rather low, and mostly comparable to those reported in other regions of Poland and other countries.

Conclusions

1. The examined crops grown in the neighborhood of Głogów copperworks fulfilled the requirements of current regulations of cadmium concentrations (except one sample of grain).
2. Cadmium concentrations in the samples of fruit from the neighborhood of Legnica copperworks were low and did not exceed the limits established in Europe.

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