J. Elem. s. 391–401 DOI: 10.5601/jelem.2013.18.3.04

LEAD CONTENT IN CEREAL PRODUCTS AS A POPULATION HEALTH THREAT MARKER: A CASE STUDY IN THE PROVINCE OF PODLASIE*

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Abstract

Heavy metals, which are considered basic industrial and environmental pollutants, pose a threat to the human health if found in food products even in trace amounts. Lead may cause such conditions as functional disorders of the kidneys, liver or the circulatory system. It can also impede the biosynthesis of heme and cause disturbances in the metabolism of vitamin D and the following microelements: Fe, Cu, Zn Se and Ca.

The aim of the study was to assess the health threat and estimate risks from the presence of Pb in cereal products available in retail shops in the Province of Podlasie. The material consisted of samples of cereal products (flour, groats, bakery products, pasta and rice) taken at random from retail shops in Podlasie in 2004-2005 and 2010-2011. The lead concentration was estimated by using the atomic absorption spectrometry method (AAS). The content of Pb was analyzed depending on a product type and sampling period; the health threat was assessed with reference to the current Polish requirements. The results were statistically analyzed using Statistica 7.1 software, Duncan's test and Wilcoxon test.

The results revealed that the average content of Pb in the examined cereal products did not exceed the norms. During the analyzed period, a decrease in the Pb concentration in cereals was found. This may indicate an improvement in the quality of the environment, which can be measured by the content of Pb in cereal products. At the same time, owing to the significant role of cereal products in human nutrition, the lead content may also serve as a marker of health exposure.

Key words: lead, cereal products, atomic absorption spectrometry, health threat.

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* I am a fellow in the project „Studying, researching, marketing – UMB doctoral support program", Sub-8.2.1 Human Capital Operational Programme co-financed by the European Union under the European Social Fund.

ZAWARTOŚĆ OŁOWIU W PRODUKTACH ZBOŻOWYCH JAKO MARKER NARAŻENIA ZDROWOTNEGO POPULACJI NA PRZYKŁADZIE WOJEWÓDZTWA PODLASKIEGO

Abstrakt

Metale ciężkie, zaliczane do podstawowych zanieczyszczeń przemysłowych i środowiskowych, obecne w żywności nawet w ilościach śladowych stanowią zagrożenie dla zdrowia ludzi. Pb może powodować m.in. zaburzenia funkcjonowania nerek, wątroby, układu krążenia, a także hamować biosyntezę hemu oraz zakłócać metabolizm witaminy D i mikroelementów, tj.: Fe, Cu, Zn Se i Ca.

Celem pracy była ocena zagrożenia zdrowotnego oraz oszacowanie ryzyka wynikającego z obecności Pb w produktach zbożowych dostępnych w sprzedaży detalicznej w woj. podlaskim oraz ocena dynamiki zmian zawartości tego pierwiastka w czasie.

Materiał do badań stanowiły próbki produktów zbożowych (mąki, kasz, pieczywa, makaronu i ryżu) pobranych losowo w placówkach handlu detalicznego w woj. podlaskim w latach 2004-2005 i 2010-2011. Stężenie Pb oznaczano metodą atomowej spektrometrii absorpcyjnej (ASA).

Analizowano zawartość Pb w badanych próbkach w zależności od rodzaju produktu i okresu pobrania próby oraz oceniano zagrożenie zdrowia, opierając się na obowiązujących w Polsce normach. Wyniki poddano analizie statystycznej z użyciem programu komputerowego Statistica PL 7.1, testu Duncana i testu kolejności par Wilcoxona.

Stwierdzono że średnia zawartość Pb w badanych produktach zbożowych nie przekraczała przyjętych norm. Średnia zawartość Pb w latach 2010-2011 była istotnie niższa w porównaniu z zawartością tego pierwiastka w latach 2004-2005. Wyniki te mogą świadczyć o poprawie stanu środowiska naturalnego, czego miernikiem może być zawartość Pb w produktach zbożowych. Jednocześnie ze względu na istotną rolę tej grupy produktów w żywieniu człowieka może on stanowić również marker narażenia zdrowotnego.

Słowa kluczowe: ołów, produkty zbożowe, atomowa spektrometria absorpcyjna, zagrożenie

INTRODUCTION

As a result of various human activities, heavy metals are widespread in the environment and permeate into food products, exposing people to their toxic effects. Heavy metals, which are considered basic industrial and environmental pollutant, pose a threat to the human health if found in food products even in trace amounts. This group includes both elements that are necessary for the body (e.g. microelements: Cu, Fe, Zn, Ni, Cr) and elements of an unknown physiological role (e.g. Cd, Hg, Pb, As) (Wojciechowska-Mazurek et al. 2008 , Dufault et al. 2009, Winiarska-Mieczan 2009). Lead (Pb) is considered as particularly harmful for human health. As a microelement, it can be found in all types of biological material: soil, plants, water and live organisms. It is characterized by a high accumulation factor in the body. Moreover, it is quickly absorbed from the digestive tract and easily permeates across biological barriers. Lead is absorbed into the human body mainly by the digestive tract and respiratory system. As regards food in the

digestive tract, it is absorbed in approximately 10% by adults and up to 50% by children. In the human body, lead from food first permeates into internal organs and then is accumulated in the osseous tissue, most commonly in joints, teeth, hair and the liver. The toxic effects of lead on the body include mainly functional disorders of the cardiovascular system, kidneys and nervous system (Tong et al. 2000, ATSDR 2007, Medyńska et al. 2009, EFSA 2010, Trojanowski et al. 2010, WHO 2011, Cielecka, Dereń 2011, Nowak et al. 2011).

The presence of free lead in tissues causes significant metabolic disturbances: retarded activation of certain enzymes, disturbances of protein synthesis and other metallic element balance. For example, lead may have an antagonistic effect or can compete with Se, Fe, Ca, Cu or Zn, which may disturb the cell's functions and impair the cellular antioxidative defense. Moreover, it has been shown that exposure to lead under experimental conditions as well and occupational or environmental exposure may lead to a decrease in the supplies of various antioxidative vitamins, especially ascorbic acid or a-tocopherol. A high intake of calcium and phosphorus in the diet causes a decrease in the amount of lead absorbed in the intestines. In contrast, ascorbic and citric acids intensify this process (Hsu, Guo 2002, Stohs, Bagchi 1995, Antonowicz et al. 1998, Peraza et al. 1998, Kozielec et al. 2002, ATSDR 2007, EFSA 2010, WHO 2011, Cielecka, Dereń 2011).

Cereal products play a crucial role in human nutrition, being a staple element of everyday diet. However, they are also one of the main sources of lead intake (approximately 33% of daily dose of this element) (Kot et al. 2011). Therefore, attempts to assess the level of lead contamination in these products are undertaken. Research provides data on amounts of metals in food products and their changes in time. The analysis of lead in cereal products enables assessment of consumers' exposure to its toxic effects.

The aim of the study was to assess the health threat and to estimate the lead presence in cereal products available in retail shops in the Province of Podlasie as well as to evaluate the dynamics of changes in the lead content in time.

MATERIAL AND METHODS

The material consisted of samples of cereal products collected randomly in retail shops in the Province of Podlasie in 2004-2005 and 2010-2011. The study included 251 cereal products: flour (wheat and rye), groats, brans, bakery products (white and wholemeal), pasta and rice.

After the mineralization of samples, levels of lead were determined by electrothermal AAS in a graphite cuvette with the Zeeman background correction, at 283.3 nm wavelength, using a Z-5000 Hitachi apparatus. The

accuracy of determinations was verified on certified reference material NSC ZC73009 (flour). The recovery was approximately 97%, at the variation coefficient of 4.1%.

The lead content in samples was analyzed depending on a type of product and time of sampling. The level of lead contamination of food products was assessed according to the Polish norms (*Commission Regulation (EC) no 1881/2006 of 19 December 2006, Commission Regulation (EC) no 420/2011 of 29 April 2011*). Based on the data of the Statistical Office in Białystok (GUS 2007, 2011) on consumption of particular groups of products, the intake of the analyzed metal with cereal products was estimated. The health threat was estimated by comparison of the average lead intake with the values of the Provisional Tolerable Weekly Intake (PTWI) and Benchmark Dose Lower Confidence Limit; in children $BMDL_{01}$ = 0.50 μg kg^{-1} bw day^{-1} (neurotoxic effect), in adults $BMDL_{10}$=0.63 μg kg^{-1} b.w. day^{-1} (nephrotoxic effect) and $BMDL_{01}$ = 1.50 μg kg b.w. day^{-1} (cardiovascular disorders) (*Commission Regulation (EC) no 1881/2006 of 19 December 2006*, EFSA 2010, WHO 2011).

The results were statistically analyzed by means of Statistica PL software. The analysis of changes in the lead content in the examined products depending on a type of product and time of sampling was performed by univariate analysis of variance (Anova) with the use of Duncan test and Wilcoxon signed rank test. Statistical significance was considered as $p \leq 0.05$.

RESULTS AND DISCUSSION

Table 1 presents the mean content of lead in cereal products, ranges, standard deviations, medians and 90^{th} percentile values in particular years.

The products collected in Podlasie contained between 0.009 mg Pb kg^{-1} and 0.299 mg Pb kg^{-1}, depending on the type of cereal products and collection time.

The results show an evident dispersion of lead values within each group of products. In 2004, the highest mean lead content was observed in pasta (0.094 ± 0.04 mg kg^{-1}), in 2005 – in rye flour (0.125 ± 0.00 mg kg^{-1}), in 2010 and 2011 – in bran (0.256 ± 0.03 mg kg^{-1} and 0.211± 0.02 mg kg^{-1} respectively).

In 2004, the lowest mean lead content was observed in wholemeal bakery products (0.043± 0.00 mg kg^{-1}), in 2005 – in groat (0.037 ± 0.03 mg kg^{-1}), while in 2010 and 2011 – in white bakery products (0.025 ± 0.01 mg kg^{-1} and 0.28 ± 0,01 mg kg^{-1} respectively).

A decrease in the lead contamination in cereal products was observed during the analyzed period of time. The samples collected in 2004 and 2005 contained approximately twice as much lead as the samples from 2010 and

Table 1

Pb content in cereal products from the Province of Podlasie (mg kg^{-1})

Cereal products		2004 $N = 52$	2005 $N = 67$	2010 $N = 64$	2011 $N = 68$
Groats	$\bar{x}$±SD	0.053±0.03	0.037±0.03	0.069±0.11	0.026±0.02
	median	0.052	0.025	0.023	0.021
	range	0.022-0.085	0.013-0.097	0.011-0.375	0.012-0.067
	90^{th}-percentile	0.085	0.097	0.179	0.041
Pasta	$\bar{x}$±SD	0.094±0.04	0.086±0.05	0.060±0.06	0.056±0.07
	median	0.125	0.125	0.046	0.036
	range	0.042-0.125	0.013-0.125	0.009-0.287	0.007-0.299
	90^{th}-percentile	0.125	0.125	0.102	0.162
Wheat flour	$\bar{x}$±SD	0.089±0.04	0.054±0.04	0.053±0.09	0.067±0.06
	median	0.12	0.04	0.02	0.064
	range	0.022-0.125	0.017-0.120	0.009-0.228	0.018-0.157
	90^{th}-percentile	0.125	0.12	0.228	0.157
Rye flour	$\bar{x}$±SD	0.075±0.04	0.125±0.00	0.062±0.00	0.087±0.02
	median	0.057	0.125	0.062	0.087
	range	0.032-0.120	0.125-0.125	0.060-0.064	0.073-0.102
	90^{th}-percentile	0.12	0.125	0.064	0.102
Bran	$\bar{x}$±SD	0.085±0.00	0.063±0.03	0.256±0.03	0.211±0.02
	median	0.085	0.087	0.256	0.197
	range	0.084-0.087	0.013-0.087	0.234-0.278	0.197-0.240
	90^{th}-percentile	0.087	0.087	0.278	0.240
White bread	$\bar{x}$±SD	0.083±0.04	0.107±0.07	0.025±0.01	0.028±0.01
	median	0.091	0.123	0.027	0.025
	range	0.028-0.120	0.017-0.206	0.013-0.038	0.013-0.044
	90^{th}-percentile	0.12	0.206	0.037	0.041
Wholemeal bread	$\bar{x}$±SD	0.043±0.00	0.086±0.05	0.044±0.02	0.044±0.02
	median	0.043	0.123	0.046	0.046
	range	0.043-0.043	0.025-0.125	0.020-0.062	0.020-0.062
	90^{th}-percentile	0.043	0.125	0.062	0.062
Rice and rice products	$\bar{x}$±SD	0.074±0.06	0.066±0.04	0.035±0.03	0.056±0.06
	median	0.074	0.06	0.026	0.032
	range	0.025-0.125	0.013-0.125	0.012-0.089	0.014-0.218
	90^{th}-percentile	0.123	0.114	0.089	0.153
All cereal products	$\bar{x}$±SD	0.081±0.04	0.081±0.04	0.058±0.07	0.055±0.06
	median	0.084	0.084	0.029	0.034
	range	0.022-0.125	0.022-0.125	0.009-0.375	0.007-0.299
	90^{th}-percentile	0.125	0.125	0.158	0.162

2011. The lead concentration in wholemeal bakery products and rye flour did not change in the years of sample collection. A considerable decrease (approximately two-fold) in the lead content was observed in pasta, white bakery products (statistically significant dependency) and rice. Statistical analysis (Figure 1) showed that the time of sample collection significantly affected the lead content in cereal products. The analysis of the results from particular cereal products (Figure 1) showed that the lead content in bran in 2010 and 2011 was significantly higher compared to other cereal products.

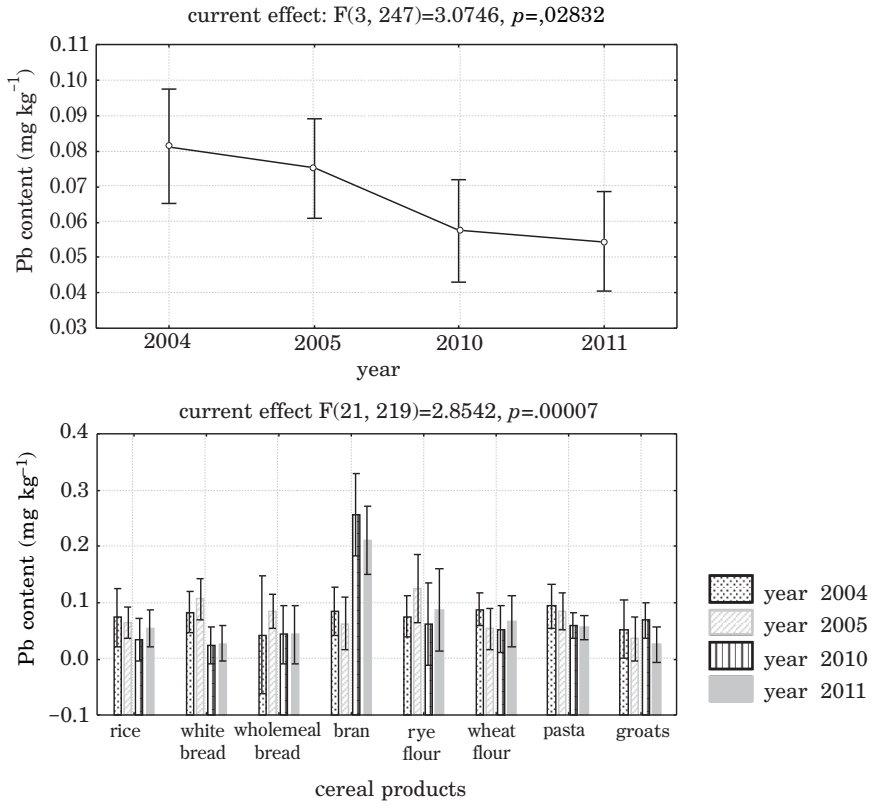

Fig. 1. Results of Anova analysis for the Pb content in cereal products (mg kg^{-1})

These results may suggest that the natural environments is improving, and the rate of improvement could be measured by the lead content in cereal products. The lead content in cereal products could also be a marker of the health threat because cereals are a staple food.

The results are not significantly different from the data obtained in the 1990s by Buliński et al. (1990, 1992), Falandysz et al. (1987) and Ilow et al. (1999) or published later by other Polish authors (Kot 2003, Kot 2003, Kot,

ZARĘBA 2007, WOJCIECHOWSKA-MAZUREK et al. 2008, MEDYŃSKA et al. 2009, KOT et al. 2011, ZAJĄC 2011).

In the south of Poland, for example, SMOCZYŃSKA et al. (1999) obtained higher results, as the reported mean lead content in flour reached 0.104 mg kg^{-1} compared to the present 0.034 mg kg^{-1}, and was in accordance with studies carried out in the 1980s (NABRZYSKI, GAJEWSKA 1984, ZAWADZKA et al. 1985).

The above results are higher than found in Germany (BRÜGGEMANN, KUMPULAINEN 1995, BRÜGGEMANN et al. 1996), Finland (TAHVONEN, KUMPULAINEN 1994), the UK (Food Standards Agency 2004, Food Standards Agency 2007), or in most countries participating in the SCOOP programme (2004) as well as the data obtained by the Polish nationwide food monitoring in 2004 (WOJCIECHOWSKA-MAZUREK et al. 2008).

Our assessment of the lead content in cereal products (Figure 2) showed an excess of the Polish norms on two occasions, namely bran in 2010 and 2011 (*Commission Regulation (EC) no 1881/2006 of 19 December 2006, Commission Regulation (EC) no 420/2011 of 29 April 2011*). As regards the other cereal products, the mean lead content was below 63% of the threshold limit.

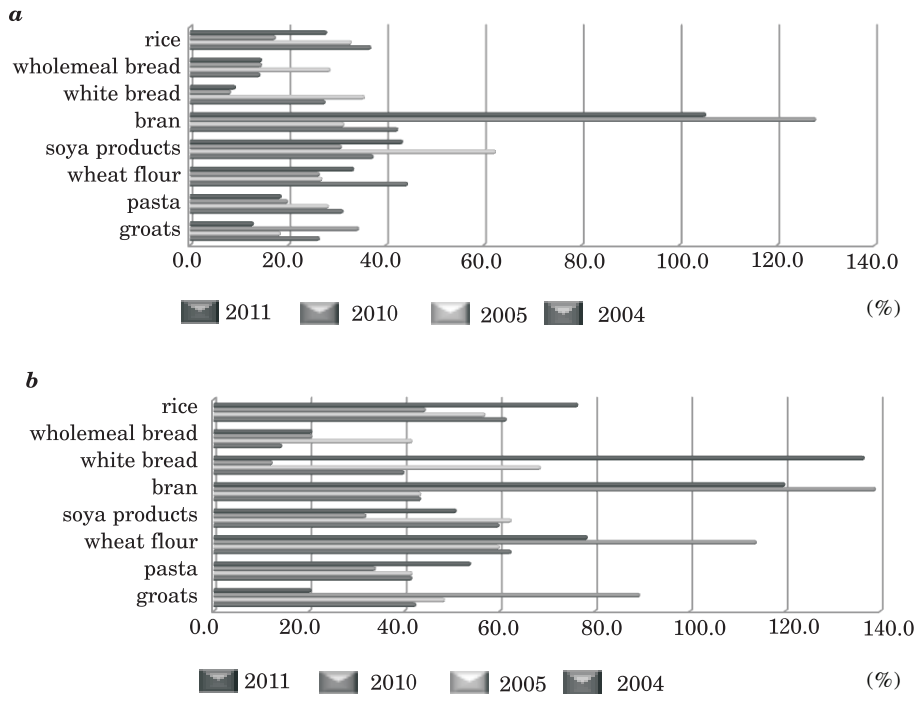

Fig. 2. Mean content (*a*) and the 90th percentile values (*b*) for Pb in cereal products from the Province of Podlasie with reference to the standards

The provisional tolerable weekly intake (PTWI) of lead from all the sources tolerated by the human body, established by the FAO/WHO Expert Committee, is 0.025 mg kg^{-1} of body weight (Commission Regulation (EC) No 1881/2006 of 19 December 2006). A weekly intake of these metals by an adult person weighing 60 kg was estimated based on both the mean lead content in particular cereal products and the data on their intake. The results are presented on Figure 3.

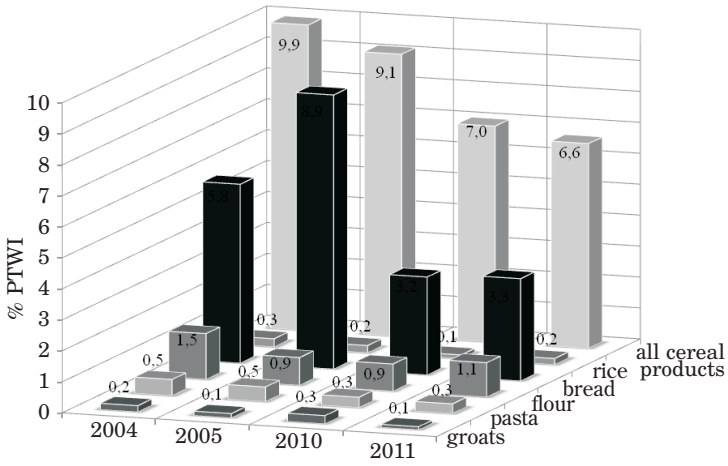

Fig. 3. Pb in cereal products from the Province of Podlasie with reference to the PTWI

According to the calculations, the lead intake with cereal products was within the amounts tolerable by the human body, that is less than 9.9% of the PTWI, of which 50% originated from bakery products.

In 2010, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) cancelled the weekly lead content tolerable by the human body (PTWI) of 0.025 mg kg^{-1} b.w. on the ground that it had failed to ensure health safety. Lower doses were determined, such as the Benchmark Dose Lower Confidence Limit (BMDL), with respect to a clearly determined effect on the human body (EFSA 2010, WHO 2011). The daily lead intake was below 56% of the $BMDL_{10}$ and 24% of the $BMDL_{01,}$ and thus the risk of developing functional disorders by the kidneys or cardiovascular system is very low. However, considering both the high intake of cereal products during the whole life and accumulation of the metal in the body, the level of lead may be estimated as being higher albeit not threatening the human health yet.

CONCLUSIONS

1. The mean lead content in the examined products (except bran in 2010-2011) was within the acceptable limits.

2. The lead intake with cereal products is within the limits tolerable by the human body (10% of the PTWI) and the lowest benchmark dose (56% $BMDL_{10}$ and 24% $BMDL_{01}$), which poses no threat to the health of residents of the Province of Podlasie.

3. A significant variety of the lead content in the examined cereal products, depending on the time of sample collection and type of product, was observed.

4. The mean lead content in 2010-2011 was significantly lower compared to 2001-2005.

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