# CONTAMINATION OF CEREAL PRODUCTS WITH LEAD AND CADMIUM AS A RISK FACTOR TO HEALTH OF THE POPULATION IN THE PROVINCE OF PODLASIE (WOJEWÓDZTWO PODLASKIE)\*

Joanna Filon<sup>1</sup>, Jolanta Ustymowicz-Farbiszewska<sup>1</sup>, Jan Górski<sup>2</sup>, Jan Karczewski<sup>1</sup>

> <sup>1</sup>Department of Hygiene and Epidemiology <sup>2</sup>Department of Physiology Medical University of Białystok

### Abstract

Lead and cadmium pose a serious threat to human health, hence their content in food products is regulated by the EU standards, also binding in Poland. Food contamination with these metals is an etiological factor of civilization diseases. In order to prevent such disorders, it is extremely important to assess the degree of environmental pollution with Pb and Cd, reflected by the level of these metals in food products. Determination of Pb and Cd in food products demonstrates exposure to the toxic effect of these metals and enables evaluation of the threat to human health in a given population. The aim of this study has been to estimate the health threat to the population of the Province of Podlasie (województwo podlaskie) due to Pb and Cd contamination of cereal products.

The following foodstuffs were analyzed: flour, groats, bread, pasta, rice, bran and soya products. They were collected in 13 administrative districts of the Province of Podlasie while monitoring health quality of food products. Pb and Cd concentrations were determined with the AAS method.

The highest Cd level was noted in pastas  $(0.058\pm0.0330~mg~kg^{-1})$  and the highest Pb level was determined in cuscus  $(0.120\pm0.0899~mg~kg^{-1})$ . The lowest average Pb concentration

mgr Joanna Filon, Department of Hygiene and Epidemiology, Medical University of Białystok, Mickiewicza 2c, 15-089 Białystok, tel/fax: (85) 7485560, e-mail: joanna@umb.edu.pl

<sup>\*</sup> 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.

tion was found in groats  $(0.042\pm0.0306~mg~kg^{-1})$  and Cd was the lowest in wholemeal bread  $(0.016\pm0.0106~mg~kg^{-1})$ .

The average concentrations of Pb and Cd in cereal products did not exceed the permissible limits established by the Minister of Health. The permissible level of Pb was not surpassed until the level of the 90<sup>th</sup> percentyl (in couscous and soya products). An average intake of Pb and Cd was within the tolerated amount, corresponding to 10% PTWI and 12% PTWI, respectively. Thus, no threat to the health of the population in Podlasie was detected.

Key words: lead, cadmium, cereal products, atomic absorption spectrometry.

# ZANIECZYSZCZENIE PRODUKTÓW ZBOŻOWYCH PB I CD JAKO CZYNNIK RYZYKA ZDROWOTNEGO LUDNOŚCI WOJEWÓDZTWA PODLASKIEGO

### Abstrakt

Pb i Cd stanowią istotne zagrożenie dla zdrowia ludzkiego, i w związku z tym ich zawartość w żywności jest limitowana przez normy Unii Europejskiej obowiązujące w Polsce. Skażenie żywności tymi metalami jest czynnikiem etiologicznym chorób cywilizacyjnych. Z punktu widzenia profilaktyki tych schorzeń bardzo istotne jest poznanie stopnia skażenia środowiska Pb i Cd, czego odzwierciedleniem jest poziom tych metali w produktach spożywczych. Badanie zawartości Pb i Cd w produktach spożywczych umożliwia ocenę stopnia narażenia na ich toksyczne działanie i określenie zagrożenia zdrowia badanej populacji ludzkiej. Celem pracy była ocena zagrożenia zdrowia ludności woj. podlaskiego produktami zbożowymi zanieczyszczonymi Pb i Cd.

Materiał do badań stanowiły próbki mąki pszennej i żytniej, kasz, pieczywa białego i razowego, makaronów, otrębów, ryżu i produktów sojowych pobrane w 13 powiatach woj. podlaskiego w ramach monitoringu jakości zdrowotnej środków spożywczych i przedmiotów użytku. Stężenie Pb i Cd oznaczano metodą AAS.

Najwyższą zawartość Cd odnotowano w makaronach (0,058 $\pm$ 0,0330 mg kg $^{-1}$ ), natomiast Pb – w kaszy kuskus (0,120 $\pm$ 0,0899 mg kg $^{-1}$ ). Najniższą średnią zawartość Pb stwierdzono w kaszach (0,042 $\pm$ 0,0306 mg kg $^{-1}$ ), a Cd w pieczywie razowym (0,016 $\pm$ 0,0106 mg kg $^{-1}$ ).

Stwierdzono, że średnia zawartość Pb i Cd w badanych produktach zbożowych nie przekraczała limitu ustalonego w Rozporządzeniu Ministra Zdrowia. Przekroczenie dopuszczalnej zawartości Pb stwierdzono dopiero na poziomie 90-percentyla (w kaszy kuskus i produktach sojowych).

Wykazano, że średnie pobranie Pb i Cd mieści się w granicach pobrania tolerowanego przez organizm i wynosi odpowiednio 10% PTWI i 12% PTWI, co nie stwarza zagrożenia dla zdrowia ludności woj. podlaskiego.

Słowa kluczowe: ołów, kadm, produkty zbożowe, atomowa spektrometria absorpcyjna.

# INTRODUCTION

Lead (Pb) and cadmium (Cd) are highly toxic elements, which are quickly absorbed from the alimentary tract. Afterwards, they easily pass through biological barriers and accumulate in internal organs. Even small amounts

of Pb and Cd may cause metabolic disorders (Kozielec et al. 2002, Malara et al. 2002, Wojciechowska-Mazurek et al. 2008, Medyńska A. et al. 2009, Dobrzański et al. 2009, Winiarska-Mieczan 2009). Lead is the cause of many diseases, including cancer of the stomach, ovaries, kidneys and leukemia; it also causes irreversible damage to the nervous system. Cadmium is responsible for decalcification and deformation of bones, myatrophy and anosmia, impotence and hypertension; it has also been classified as carcinogenic to humans by the IARC (Starska et al. 1996, Kozielec et al. 2002, ATSDR 2007, 2008, Fortier et al. 2008, Iavicoli et al. 2009, EFSA 2009, 2010, Trojanowski et al. 2010, Kaniuczak et al. 2011, Nowak et al. 2011).

Lead and cadmium pose a serious threat to human health, hence their content in food products is regulated by the EU standards, also binding in Poland. Food contamination with these metals is an etiological factor of civilization diseases. It is extremely important for prophylaxis to know the extent of environmental pollution with Pb and Cd, reflected in the levels of these metals in food products. Determination of Pb and Cd in food products enables one to estimate the exposure to their toxic action and therefore to assess the health risk in particular human populations. Nowadays, due to environmental pollution, all food products are contaminated with Pb and Cd. Since Poland accessed the European Union, only these two metals have been taken into consideration for the assessment of food contamination (Wojciechowska-Mazurek et al. 2003, Wójcik-Stopczyńska 2003, Baczek-Kwinta et al. 2011).

The objective of this study has been to determine whether consumption of cereal products by residents of Podlasie threatens their health due to Pb or Cd contamination.

## MATERIAL AND METHODS

The material for analysis included 148 samples of cereal products (wheat and wholemeal flour, cereals, groats, bran, white and wholemeal bread, pasta and soya products) collected in 13 districts of the Province of Podlasie while monitoring health quality of food and other household products.

The sample collection procedures complied with the binding standards. Validated analytical methods were applied, in line with the EU legal criteria for the methods used for official food testing (Commission Directive 2001/22/EC of 8 March 2001, Directive of Minister of Health of 30 April 2004. Journal of Law No 120 item 1257). Certified reference materials were used for quality control. The recovery was 98% for Pb and 96% for Cd, and the coefficient of variation was 4.2% and 3.6%, respectively. Concentrations of Pb and Cd were determined by atomic absorption spectrophometry (AAS).

The contamination of cereal products with Pb and Cd was determined with reference to the binding Polish norms (*Directive of Minister of Health of 13 January 2003*). The intake of these metals with cereal products was estimated according to the data published by the Statistical Office in Białystok concerning consumption of all the analyzed groups of products (GUS 2008). The risk to human health posed by Pb and Cd in cereal products was evaluated by comparing their mean intake with the provisional tolerable weekly intake (PTWI).

The results were statistically analyzed using the software Statistica 7.1. The Duncan's test was applied to compare the significance of differences between the means depending on the type of cereal product. The level of significance was  $p \leq 0.05$ .

### RESULTS AND DISCUSSION

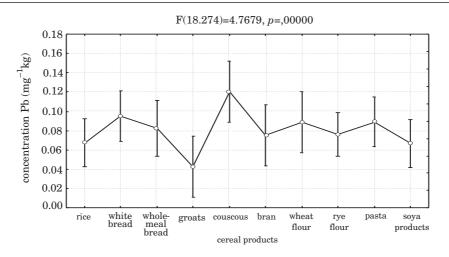
The level of Pb in cereal products available in Podlasie ranged from  $0.013~\rm mg~kg^{-1}$  to  $0.275~\rm mg~kg^{-1}$ , depending on the type of foodstuff. The mean Pb content was  $0.080\pm0.0545~\rm mg~kg^{-1}$ , median  $-0.061~\rm mg~kg^{-1}$ ; 90% of results did not exceed  $0.125~\rm mg~kg^{-1}$ .

The cereal products from Podlasie Province contained from 0.000 mg Cd kg $^{-1}$  to 0.100 mg Cd kg $^{-1}$ . The mean Cd content was 0.028±0.0217 mg kg $^{-1}$ , median - 0.025 mg kg $^{-1}$ ; 90% of results did not exceed 0.059 mg kg $^{-1}$ .

The results show distinct dispersion of the values of the elements studied within one assortment. The highest Pb content was noted in couscous (0.120 mg kg<sup>-1</sup>  $\pm$  0.0899), with 90% of results not exceeding 0.250 mg kg<sup>-1</sup>. The level of Pb was the lowest in barley groats (0.042 mg kg<sup>-1</sup>  $\pm$  0.0306) and only 10% of results exceeded 0.085 mg kg<sup>-1</sup>. These values were statistically significant (Figure 1).

The lowest mean amounts of Cd were noted in wholemeal bread and rye flour (0.016 mg kg<sup>-1</sup>  $\pm$  0.0106, 0.018 mg kg<sup>-1</sup>  $\pm$  00099, respectively), and only 10% of results were higher than 0.025 mg kg<sup>-1</sup>. Statistical analysis revealed (Figure 1) a significantly higher Cd content in pasta (0.058 mg kg<sup>-1</sup>  $\pm$  0.0330) and bran (0.039 mg kg<sup>-1</sup>  $\pm$ 0.0181) as compared to other cereal products. In 10% of the examined pasta samples, the Cd content was 0.100 mg kg<sup>-1</sup>.

The above findings are similar to those reported in the 1990s by Buliński et al. (1990, 1992), Ilow et al. (1999) and Falandysz et al. (1987). The results are also consistent with later research by other Polish authors (Kot 2003, Kot, Zaręba 2007, Wojciechowska-Mazurek et al. 2008, Kot et al. 2009 Medyńska et al. 2009).



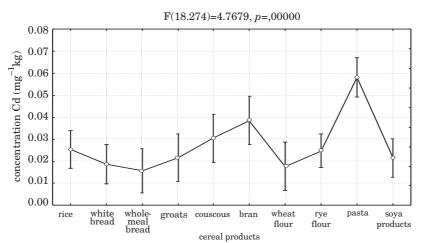


Fig. 1. Results of Anova analysis for the Pb and Cd content in cereal products (mg kg<sup>-1</sup>)

Smoczyńska et al. (1999) obtained higher levels of Pb and Cd in flours from southern Poland (0.104 mg kg $^{-1}$  and 0.034 mg kg $^{-1}$ , respectively), which was consistent with the data reported in the 1980s (Zawadzka et al. 1985).

The contents of toxic metals in Polish food products do not differ significantly from those reported from other countries. Only cereal products from Germany and Finland have lower Pb and Cd concentrations (Brüggemann, Kumpulainen 1995, Brüggemann et al. 1996, Tahvonen, Kumpulainen 1994, SCOOP 2004, Food Standards Agency 2004, 2007, EFSA 2009, 2010). By far, lower Pb levels were also revealed by the Polish food monitoring of 2004 supervised by the Department of Food Research, National Institute of Public Health – National Institute of Hygiene (NIZP-PZH) (Wojciechowska-Mazurek et al. 2008).

Rye flours contained more Pb as compared to wheat flours, whereas the Cd content was the highest in wheat flour. These results are supported by other authors in various parts of Europe and Poland (Brüggemann, Kumpulainen 1995, Brüggemann et al. 1996, Buliński et al. 1990, 1992, Kot, Zareba 2007, Kot et al. 2009).

The mean Pb content in bread was similar to or higher than the Pb level in the flours from which the bread was produced, which is consistent with observations of other authors (Brüggemann, Kumpulainen 1995, Kot 2003, Kot, Zareba 2007). This is explained by the effect of the technological process and addition of other ingredients during the production process. The Cd content in pasta was almost twice as high as in wheat flour. Worthy of note is the fact that white bread had more Pb and Cd than wholemeal bread. Most authors confirm higher concentrations of these elements in wheat bread (Buliński et al. 1990, 1992, Kot 2003, Kot, Zareba 2007).

Contamination of cereal products with Pb and Cd was determined based on standards binding in Poland (*Directive of Minister of Health of 13 January 2003*). Results have been presented in Figure 2.

It was only at the 90<sup>th</sup> percentile that the content of Pb in couscous (125%) and soya products (112.5%) exceeded the standard limit established in the *Directive of the Minister of Health of 13 January 2003*. In 10% of the pasta samples, the level of Cd was on the borderline of the permitted limit. In other cereal products, the mean levels of Pb and Cd as well as the 90th percentile values did not exceed 62% of the permitted limit.

The temporary PTWI doses of Pb and Cd from all sources tolerated by a healthy human, established by the Joint FAO/WHO Expert Committee, are respectively 0.025 mg kg<sup>-1</sup> and 0.007 mg kg<sup>-1</sup> body weight (*Commission Regulation (EC) no 1881/2006 of 19 December 2006*). Taking into account the mean concentrations of Pb and Cd in the respective cereal products and their intake-related data, a weekly intake of these metals was determined for an adult person of 60 kg body weight. Results have been presented in Figure 3.

The calculated intake of Pb and Cd with cereal products is within the limits of tolerable intake, being 9.7% PTWI and 12% PTWI respectively, of which 47% (for Cd) and 85% (for Pb) come from bread.

Taking into account high intake of cereal products, especially bread, the levels of Pb and Cd should be considered elevated yet not health-threatening.

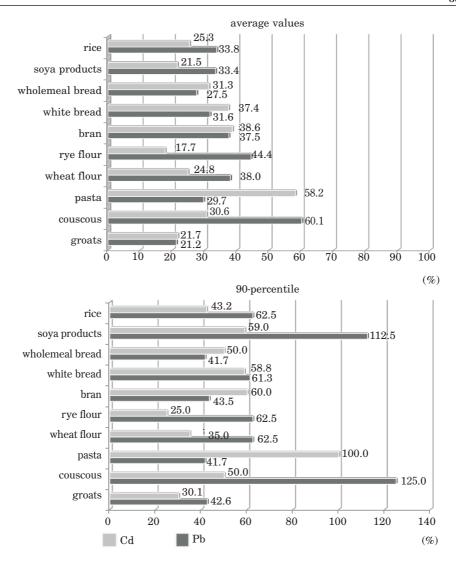


Fig. 2. Mean content and the 90<sup>th</sup> percentile values for Pb and Cd in respective cereal products from Podlasie Province with reference to the standards of the Ministry of Health

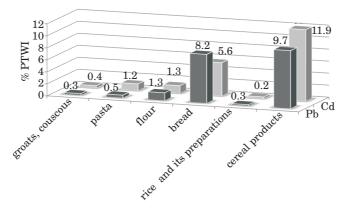


Fig. 3. Pb and Cd in respective cereal products from Podlasie Provincee with reference to PTWI

# **CONCLUSIONS**

- 1. The mean levels of Pb and Cd did not exceed the limit values. The Pb content was found to be higher than the standard limit only at the 90th percentile (in couscous and soya products).
- 2. The intake of Pb and Cd with cereal products is within the limits of tolerable intake (10% PTWI and 12% PTWI, respectively) and does not threaten the health of the population in Podlasie Province.
- 3. Significant variations were found in Pb and Cd levels in the cereal products studied depending on the type of the product.

### REFERENCES

Agency for Toxic Substances and Diseases Registry (ATSDR). 2007. Toxicological profile for lead. U.S. Department of Health and Human Services. Public Health Service. http://www.atsdr.cdc.gov/toxprofiles/tp13.pdf (accessible 08.11.2007)

Agency for Toxic Substances and Diseases Registry (ATSDR). 2008. *Draft toxicological profile for cadmium*. U.S. Department of Health and Human Services. Public Health Service. http://www.atsdr.cdc.gov/toxprofiles/tp5.pdf (accessible 26.02.2009)

Baczek-Kwinta R., Bartoszek A., Kusznierewicz B., Antonkiewicz J. 2011. *Physiological response of plants and cadmium accumulation in heads of two cultivars of white cabbage*. J. Elem., 16(3): 355-364.

Brüggemann J., and Kumpulainen J. T. 1995. Spurenelementgehalte in Deutschen Grundnahrungs-Mitteln aus Brotgetreide. Getreide Mehl Brot., 49: 171-177.

Brüggemann J., Dörfner H.H., Hecht H., Kumpulainen J.T., Westermair T. Status of trace elements in sample foods from Germany 1990-1994. 1996. REU Technical Series (FAO, Rome), 49: 5-58.

- Buliński R., Kot A., Błoniarz J. 1992. Study on some trace elements content in home food products. Part XII. Contamination of home bakery products with harmful metals. Bromat. Chem. Toksykol., 25(2): 193-196. (in Polish)
- Buliński R., Kot A., Błoniarz J., Wyszogrodzka-Koma L. 1990. Studies on some trace elements content in food-stuffs of home growth. Part XII. Evaluation of contamination with harmful metals of corn products. Bromat. Chem. Toksykol., 23(3-4): 105-108. (in Polish)
- Commission Directive 2001/22/EC of 8 March 2001 laying down the sampling methods and the methods of analysis for the official control of the levels of lead, cadmium, mercury and 3-MCPD in foodstuffs. O.J. of the EC L 77 of 16.03.2001.
- Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs. O.J. of the EC L 345/5 of 20.12.2006.
- Directive of Minister of Health of 13 January 2003. Dz. U. No 37 item 326.
- Directive of Minister of Health of 30 April 2004. Dz. U. No 120 item 1257.
- Dobrzański Z., Skiba M., Brożyńska A., Kowalska-Góralska M., 2009. The evaluation of milk selected trace elements levels in ruminants (cows and goats) reared in industrial and nonpolluted areas. Acta Sci. Pol., Med. Vet., 8 (1): 3-14. (in Polish)
- European Food Safety Authority (EFSA). 2009. Scientific Opinion of the Panel on Contaminants in the Food Chain on a request from the European Commission on cadmium in food. The EFSA Journal, 980: 1-139.
- European Food Safety Authority (EFSA). 2010. Panel on Contaminants in the Food Chain (CONTAM); Scientific Opinion on Lead in Food. The EFSA Journal, 8(4): 1570.
- Falandysz J., Lorenc-Biała H., Centkowska D. 1987. The content of metals in certain foods products. Rocz. PZH, 38 (4-5): 344-346. (in Polish)
- Food Standards Agency. 2004. 2000 Total Diet Study of 12 elements. FSIS 48/04.
- Food Standards Agency. 2007. Survey of metals in a variety of foods. FSIS 01/07.
- Fortier M., Omara F., Bernier J., Brousseau P., Fournier M. 2008. Effects of physiological concentrations of heavy metals both individually and in mixtures on the viability and function of peripheral blood human leukocytes in vitro. J. Toxicol. Environ. Health, A 71: 1327-1337
- Central Statistical Office of Poland (GUS). 2008. Household budget surveys in 2006. (in Polish)
- Ilow R., Regulska-Ilow B., Szymczak J. 1999. Estimated intake of cadmium, lead, and Merkury by the population of the lrgnica-Głogów copper mining area. Part III. Daily food rations and analytical determinations of cadmium and lead contents of selected food products in assessing the intake of those metals. Bromat. Chem. Toksykol., 33(3): 239-245. (in Polish)
- Kaniuczak J., Hajduk E., Właśniewski S. 2011. Effect of liming and mineral fertilization on cadmium content in grain of spring barley (hordeum vulgare l.) and winter wheat (triticum aestivum l.) cultivated on loessial soil. J. Elem., 16(4): 535-542.
- Kot A. 2003. Cereal products as a source of cadmium and lead. Żyw. Człow. Metab., 30 (3/4): 1097-1103. (in Polish)
- Kot A., Zareba S. 2007. Cadmium and lead content in cereal products. Żyw. Człow. Metab. 34 (3/4): 889-895. (in Polish)
- Kot A., Zareba S., Wyszogrodzka-Koma L. 2009. Assessment of cadmium contamination in cereals, cereal products and potatoes. Bromat. Chem. Toksykol., 42(3): 537-542. (in Polish)
- Kozielec T., Sałacka A., Późniak J., Sałacki A., Karakiewicz B. 2002. Cadmium and zinc concentration in adults from the Szczecin area. J. Elem., 7(2): 141-145. (in Polish)

- Kozielec T., Strecker D., Durska G. 2002. Lead concentration in hair and some hematological parameters in rheumatoid arthritis patients. J. Elem., 7(1): 5-14. (in Polish)
- Malara P., Kwapuliński J., Drugacz J., Malara B. 2005. Impact of environmental exposure on coexistence of cadmium and zinc in teeth. J. Elem., 10(4): 959-965. (in Polish)
- Medyńska A, Kabała C, Chodak T, Jezierski P. 2009. Concentration of copper, zinc, lead and cadmium in plants cultivated in the surroundings of "Żelazny Most" copper ore tailings impoundment. J. Elem., 14(4): 729-736.
- Nowak L., Dzieżyc H., Piotrowski M. 2011. Content of bioelements and toxic metals in honey of various botanical orgin from Lower Silesia. J. Elem., 16(3): 437-444.
- Scientific Co-operation on Questions Relating to Food (SCOOP). 2004. Assessment of dietary exposure to arsenic, cadmium, lead, mercury of the population of the European Union member states. SCOOP Report of experts participating in Task 3.2.11. March 2004.
- Smoczyńska K., Stasiek M., Ciecierska Z., Smoczyński S. 1999. The content of lead, cadmium, macro and microelements in wheat seeds from Tczew and a chosen after-flood region in south-western Poland. Biul. Magnezol., 4(1): 177-180. (in Polish)
- Tahvonen R., Kumpulainen J. 1994. Lead and cadmium contents in Finnish breads. Food Addit. Contam., 11: 621-631.
- Trojanowski P., Trojanowski J., Antonowicz J., Bokiniec M. 2010. Lead and cadmium content in human hair in central pomerania (northern poland). J. Elem., 15(2): 363-384.
- Winiarska-Mieczan A. 2009. Assessment of infant exposure to lead and cadmium content in infant formulas. J. Elem., 14(3): 573-581.
- Wojciechowska-Mazurek M., Starska K., Brulińska-Ostrowska E., Karłowski K., Grudzińska B. 2003. Assessment of the intake of harmful metals with Total daily diets for children and teenagers in certain voivodships. Bromat. Chem. Toksykol., 36 (supl): 267-274. (in Polish)
- Wojciechowska-Mazurek M., Starska K., Brulińska-Ostrowska E., Plewa M., Biernat U., Karłowski K. 2008. Monitoring of contamination of foodstuffs with elements noxious to human health. Part I. Wheat cereal products, vegetable products, confectionery and products for infants and children (2004 year). Rocz. PZH, 59(3): 251-266. (in Polish)
- Wójcik-Stopczyńska B. 2003. Estimated content of some heavy metals in minimally processed vegetable salads. J. Elem., 8(4): 289-295. (in Polish)
- Zawadzka T., Kamińska M., Brzozowska B. 1985. Comparison of atomic-absorption spectrophotometry and calorimetry in determination of lead, zinc and copper levels in cereal products. Rocz. PZH, 34(2): 109-112. (in Polish)