

CONTAMINATION OF CEREAL PRODUCTS WITH LEAD AND CADMIUM AS A FACTOR OF A HEALTH RISK FOR PEOPLE IN PODLASKIE VOIVODSHIP

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Abstract. Due to a major threat to human health, the content of Pb (lead) and Cd (cadmium) in food products has been limited by EU (European Union) standards that are binding in Poland. Food contamination with these metals is an etiological factor of civilization diseases. The knowledge of the degree of environmental pollution with Pb and Cd, reflected in the level of these metals in food products, is extremely important from the point of view of prophylaxis. The determination of Pb and Cd in food products allows assessment of exposure to their toxic action and thus evaluation of a threat to health in the respective human population. The aim of the study was to assess the health threat in the population of Podlaskie Voivodship according to Pb and Cd contamination of cereal products.

The specimen's materials were: flour, groats, and bread, pasta, rice, bran and soya products collected in 13 administrative districts of Podlaskie Province. Pb and Cd concentrations were estimated by using AAS (atomic absorption spectroscopy) method. The contents of Pb and Cd were analyzed depending on product type and sampling period; health threat was assessed by current Polish requirements. The results were statistically analyzed using the Statistica 7.1 software.

The highest Cd level was noted in pastas (0.058 ± 0.0330 mg/kg) and the highest Pb level was in couscous (0.120 ± 0.0899 mg/kg). The lowest average Pb concentration was noted in groats (0.042 ± 0.0306 mg/kg) and Cd in wholemeal bread (0.016 ± 0.0106 mg/kg).

Average concentration of Pb and Cd in the examined cereal products did not exceed the permissible limits established by the MH (Minister of Health). The permissible level of Pb was exceeded in the 90th percentile (in couscous and soya products). The average intake Pb and Cd was within the norm of tolerable intake and it comes to be 10% PTWI (provisional tolerable weekly intake) and 12% PTWI respectively and should not threaten the health of the population in the Podlaskie Province.

Keywords: lead, cadmium, cereal products, atomic absorption spectroscopy.

GRŪDINIŲ KULTŪRŲ UŽTERŠTUMAS ŠVINU IR KADMIU – RIZIKOS VEIKSNYS PALENKĖS VAIVADIJOS GYVENTOJŲ SVEIKATAI

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Santrauka. Dėl pavojaus žmonių sveikatai Europos Sąjunga (ES) nustatė leistiną Pb (švino) ir Cd (kadmio) koncentracijas maisto produktuose. Šie reikalavimai taikomi ir Lenkijoje. Maisto užteršimas šiais metalais yra viena priežasčių, sukeliančių vadinamąsias civilizacijos ligas. Žinios apie aplinkos užterštumą Pb ir Cd, kurį rodo šių metalų koncentracija maisto produktuose, yra ypač svarbios ligų profilaktikai. Nustačius Pb ir Cd koncentraciją maisto produktuose, galima įvertinti tikėtiną toksinį metalų poveikį ir numatyti pavojų tam tikros gyventojų grupės sveikatai. Šio darbo tikslas – nustatyti galimai švinu ir kadmiu užterštų grūdinių kultūrų keliamą pavojų Palenkės vaivadijos gyventojų sveikatai.

Buvo ištirti miltai, kruopos, duona, makaronai, ryžiai, sėlenų ir sojų produktai iš trylikos administracinių Palenkės provincijos rajonų. Pb ir Cd koncentracija nustatyta atominės absorbcijos spektroskopijos (AAS) metodu. Gautos vertės buvo analizuojamos atsižvelgiant į produkto rūšį ir pavyzdžių paėmimo laiką. Poveikis sveikatai vertintas pagal Lenkijoje galiojančius reikalavimus. Tyrimų rezultatai statistiškai apdoroti programa „Statistica 7.1“.

Didžiausios Cd reikšmės nustatytos makaronuose ($0,058 \pm 0,0330$ mg/kg), o Pb – kuskuse ($0,120 \pm 0,0899$ mg/kg). Mažiausia Pb koncentracija rasta kruopose ($0,042 \pm 0,0306$ mg/kg), o Cd – nesijotų miltų duonoje ($0,016 \pm 0,0106$ mg/kg).

Vidutinė Pb ir Cd koncentracija tirtose grūdinėse kultūrose neviršijo leistinų Sveikatos ministerijos (SM) nustatytų verčių. Pb koncentracija viršijo leistiną ribą 90 procentilėje (kuskuse ir sojų produktuose). Vidutinės suvartojamų Pb ir Cd reikšmės atitiko toleruotinos vertės ribas – atitinkamai 10 proc. ir 12 proc. Tokia jų koncentracija neturėtų kenkti Palenkės vaivadijos gyventojų sveikatai.

Raktažodžiai: švinas, kadmio, grūdinių kultūros, atominės absorbcijos spektroskopija.

Introduction. Lead (Pb) and cadmium (Cd), highly toxic elements, undergo quick absorption from the alimentary tract, easily pass through biological barriers and accumulate in internal organs. Even tiny amounts of Pb and Cd may cause metabolic disorders (Kozielec et al., 2002; Malara et al., 2002; Wojciechowska-Mazurek et al., 2008; Medyńska et al., 2009; Dobrzański et al., 2009; Winiarska – Mieczan, 2009). Pb is the cause of many diseases, including cancers of the stomach, ovaries, kidneys and leukemia; it also causes irreversible damage to the nervous system. Cd causes decalcification and deformation of bones, myatropy and anosmia, impotence and hypertension, and has been classified as carcinogenic to humans by IARC (International Agency for Research on Cancer) (Kozielec et al., 2002; Ryselis S., et al., 2007; ATSDR (Agency for Toxic Substances and Diseases Registry), 2007; 2008; Fortier et al., 2008; EFSA (European Food Safety Authority), 2009; 2010; Trojanowski et al., 2010; Kaniuczak et al., 2011; Nowak et al., 2011).

Due to a major threat to human health, the content of Pb and Cd in food products has been limited by EU standards that are binding in Poland. Food contamination with these metals is an etiological factor of civilization diseases. The knowledge of the degree of environmental pollution with Pb and Cd, reflected in the level of these metals in food products, is extremely important from the point of view of prophylaxis. The determination of Pb and Cd in food products allows assessment of exposure to their toxic action and thus evaluation of a threat to health in the respective human population. Nowadays, due to environmental pollution all food products are contaminated with Pb and Cd. Now that Poland joined the European Union, only these two metals are taken into consideration in the assessment of food contamination degree (Wojciechowska-Mazurek et al., 2003; Malakauskas et al., 2003; Wójcik-Stopczyńska, 2003; Bączek-Kwinta et al., 2011).

The aim of the study was to assess the health threat in the population of Podlaskie Voivodship according to Pb and Cd contamination of cereal products.

Material and methods

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 Podlaskie Province, as part of the monitoring of health quality of food products and utilitarian articles.

Sample collection procedures followed the binding standards. Validated analytical methods were applied, complying with the EU legal criteria (Commission Directive 2001/22/EC of 8 March 2001; Directive of Minister of Health of 30 April 2004. Dz. U. 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, respectively 4.2% and 3.6%. Pb and Cd concentrations were estimated by using atomic absorption spectroscopy (AAS) method.

The samples were dry mineralized in quartz crucibles in a muffle furnace at 400°C. The ash was solubilized in 6

mol/l hydrochloric acid and quantitatively transferred to laboratory flasks. If mineralization was incomplete, oxidation in 10% HNO₃ was additionally performed. The elements studied were transferred into complex bindings with ammonium 1-pyrrolidine dithiocarbamate in citrate buffer and then extracted to obtain 4-methyl-2-pentanone (MIBK). Determinations were carried out using an apparatus Pye Unicam SP-192 at a wavelength of 217 nm – Pb and 228.8 nm – Cd.

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 Dz. U. No 37 item 326.). Intake of these metals with cereal products was assessed according to the data of the Statistical Office in Białystok concerning the consumption of the respective groups of products (GUS, 2008). Health threat due to Pb and Cd was evaluated by comparing their mean intake with the provisional tolerable weekly intake (PTWI) (Commission Regulation (EC) no 1881/2006 of 19 December 2006).

The results were statistically analyzed using the Statistica 7.1 computer program. 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 considered to be $p \leq 0.05$.

Results and discussion

Tables I and II present the mean content, ranges, standard deviations, median and the 90th percentile values for Pb and Cd in the respective cereal products from Podlaskie Province.

The level of Pb in Podlaskie Province ranged from 0.013 mg/kg to 0.275 mg/kg depending on the type of cereal product. The mean Pb content was 0.080±0.0545 mg/kg, median – 0.061 mg/kg; 90% of results did not exceed 0.125 mg/kg.

The cereal products from Podlaskie Province contained from 0.000 mg Cd/kg to 0.100 mgCd/kg. The mean Cd content was 0.028±0.0217 mg/kg, median – 0.025 mg/kg; 90% of results did not exceed 0.059 mg/kg.

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 ± 0.0899), with 90% of results not exceeding 0.250 mg/kg. The level of Pb was the lowest in barley groats (0.042 mg/kg ± 0.0306) and only 10% of results exceeded 0.085 mg/kg. These values were statistically significant (Fig. 1).

The lowest mean amounts of Cd were noted in wholemeal bread and rye flour (0.016 mg/kg ± 0.0106, 0.018 mg/kg ± 0.0099, respectively), and only 10% of results were higher than 0.025 mg/kg. Statistical analysis revealed (Fig. 1) a significantly higher Cd content in pasta (0.058 mg/kg ± 0.0330) and bran (0.039 mg/kg ± 0.0181) as compared to other cereal products. In 10% of the examined pasta samples the Cd content was 0.100 mg/kg.

The presented 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 studies of other Polish authors (Kot, 2003; Kot & Zaręba, 2007; Wojciechowska-

Mazurek et al., 2008; Kot et al., 2009; Medyńska et al., 2009).

Smoczyńska et al. (1999) obtained higher levels of Pb

and Cd in flours from southern Poland (0.104 mg/kg and 0.034 mg/kg, respectively), which was consistent with the data reported in the 1980s (Zawadzka et al., 1985).

Table 1. **Pb content in respective cereal products from Podlaskie Province (mg/kg)**

Cereal products	Number of samples	$\bar{x} \pm SD$	Median	Range	90th-percentile
Groats	11	0.042±0.0306	0.025	0.013-0.097	0.085
Couscous	11	0.120±0.0899	0.120	0.043-0.250	0.250
Pasta	17	0.089±0.0460	0.125	0.013-0.125	0.125
Wheat flour	22	0.076±0.0448	0.064	0.017-0.125	0.125
Rye flour	11	0.089±0.0395	0.120	0.032-0.125	0.125
Bran	11	0.075±0.0249	0.087	0.013-0.087	0.087
White bread	16	0.09±0.0567	0.120	0.017-0.206	0.184
Wholemeal bread	13	0.083±0.0473	0.120	0.025-0.125	0.125
Soya products	18	0.067±0.0745	0.035	0.013-0.275	0.225
Rice and its preparations	18	0.068±0.0440	0.060	0.013-0.125	0.125
All cereal products	148	0.080±0.0545	0.061	0.013-0.275	0.125

Table 2. **Cd content in respective cereal products from Podlasie Province (mg/kg)**

Cereal products	Number of samples	$\bar{x} \pm SD$	Median	Range	90th-percentile
Groats	11	0.022±0.0155	0.021	0.007-0.061	0.030
Couscous	11	0.031±0.0125	0.025	0.022-0.050	0.050
Pasta	17	0.058±0.0330	0.044	0.021-0.100	0.100
Wheat flour	22	0.025±0.0125	0.025	0.005-0.068	0.035
Rye flour	11	0.018±0.0099	0.025	0.003-0.025	0.025
Bran	11	0.039±0.0181	0.033	0.028-0.086	0.060
White bread	16	0.019±0.0112	0.016	0.003-0.048	0.029
Wholemeal bread	13	0.016±0.0106	0.025	0.003-0.025	0.025
Soya products	18	0.022±0.0223	0.008	0.002-0.069	0.059
Rice and its preparations	18	0.025±0.0186	0.025	0.000-0.067	0.042
All cereal products	148	0.028±0.0217	0.025	0.000-0.100	0.059

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 (Scientific Co-operation on Questions Relating to Food), 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 Cd content was the highest in wheat flour. This finding is confirmed by other authors in various parts of Europe and Poland (Brüggemann & Kumpulainen, 1995; Brüggemann et al., 1996; Buliński et al., 1990; 1992; Kot & Zaręba 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 & Zaręba, 2007). This is explained by the effect of the technological process and addition of other ingredients during the production process. 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 & Zaręba, 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 Dz. U. No 37 item 326.). Results have been presented in Fig. 2.

It was only in the 90th percentile, that the content of Pb in couscous (125%) and soya products (112.5%) exceeded the standard limit. In 10% of the pasta samples the level of Cd was on the border 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 and 0.007 mg/kg 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 Fig. 3.

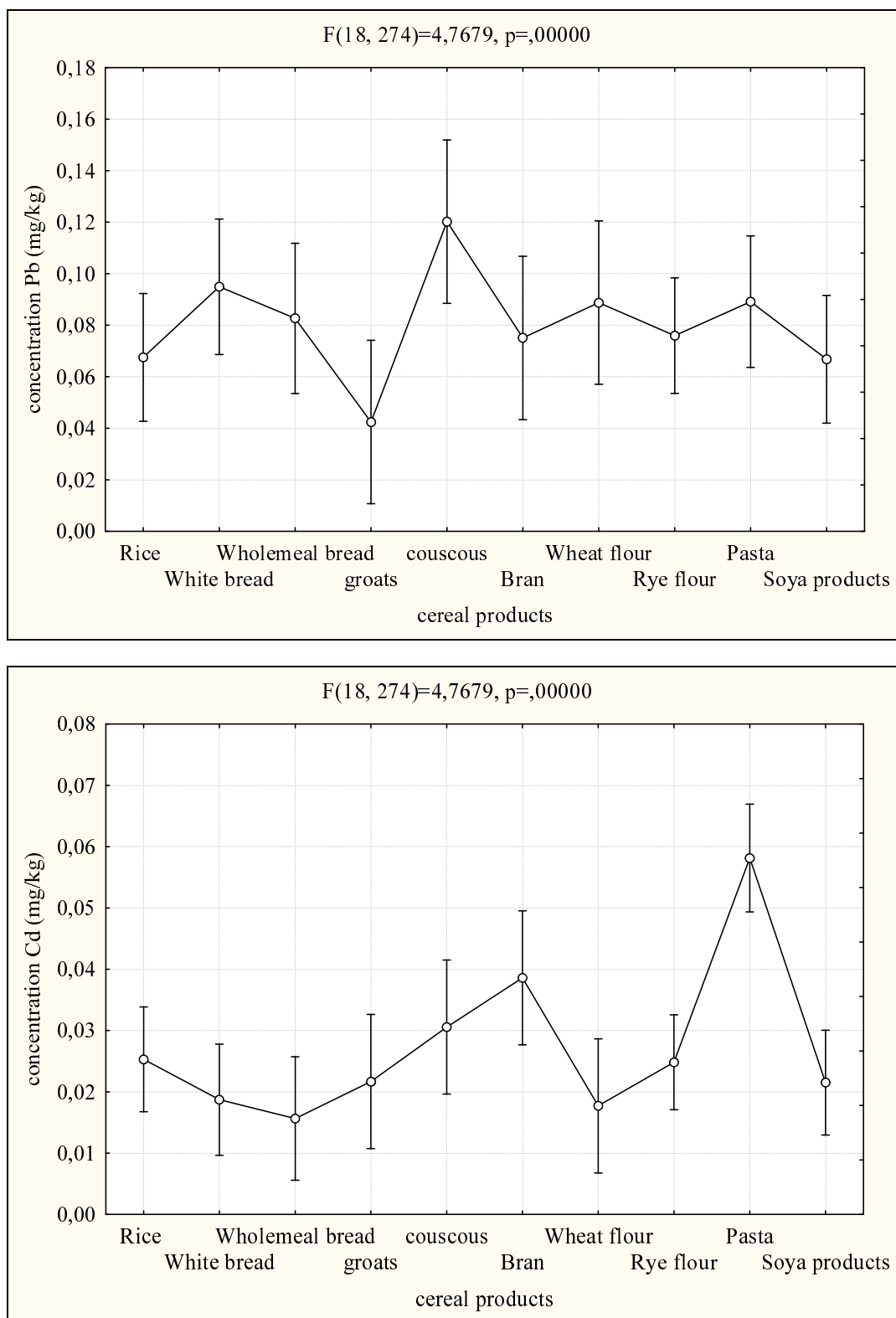


Figure 1. Results of Anova analysis for the Pb and Cd content in cereal products (mg/kg)

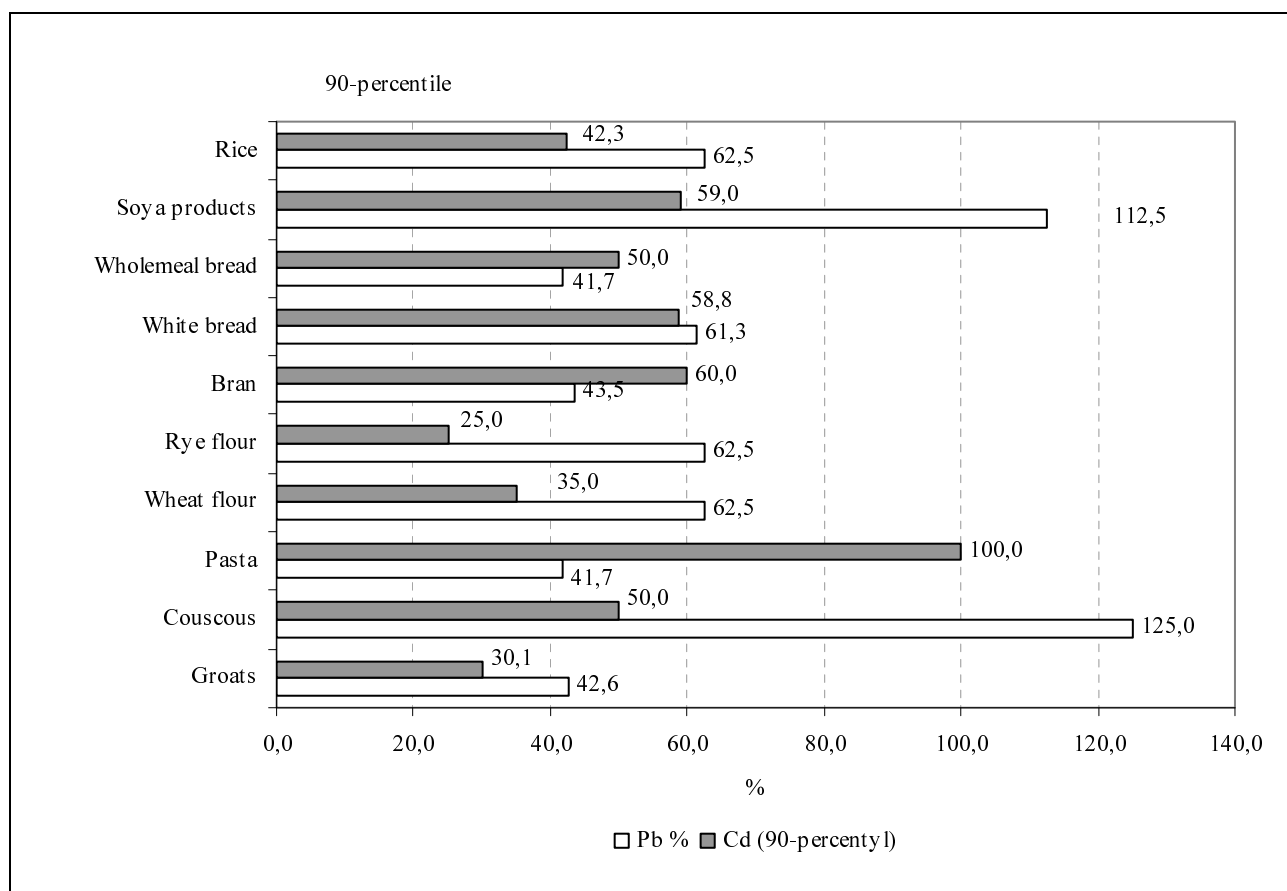
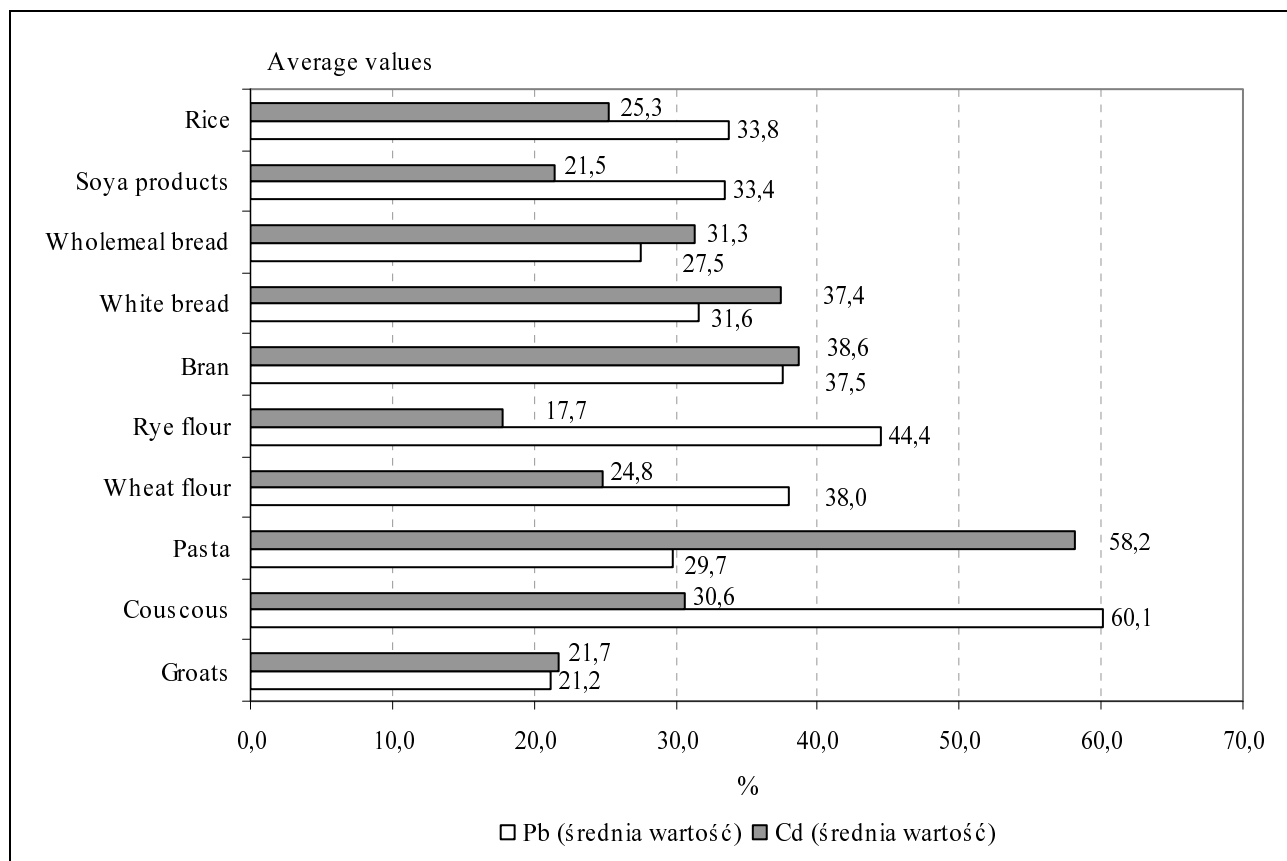


Figure 2. Mean content and the 90th percentile values for Pb and Cd in respective cereal products from Podlasie Province with reference to the standards binding in Poland

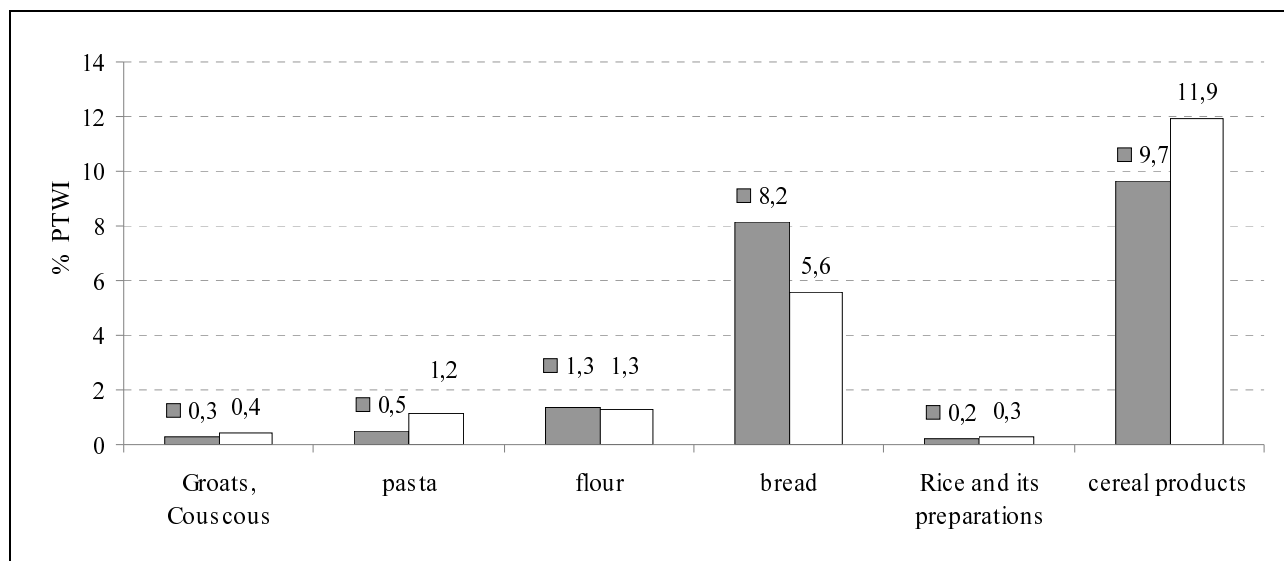


Figure 3. **Pb and Cd in respective cereal products from Podlasie Province with reference to PTWI**

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.

Conclusions

1. The mean levels of Pb and Cd did not exceed the limit values. Pb content was found to be higher than the standard limit only in 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 Podlaskie Province.

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