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PRACA ORYGINALNA
ORIGINAL PAPER

Cadmium and lead content in gluten and gluten-free bread available on Polish market – potential health risk to consumers

Zawartość kadmu i ołowiu w pieczywie glutenowym i bezglutenowym dostępnym na polskim rynku – potencjalne ryzyko zdrowotne konsumentów

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ABSTRACT

INTRODUCTION: Grain products, especially bread, play an essential role in the nutrition of people around the world. However, due to the release of excessive amounts of pollutants into the environment, grain products may also contain some amounts of potentially toxic elements, which may have a negative effect on the human body.

MATERIAL AND METHODS: The aim of the study is to analyze the content of heavy metals in selected gluten and gluten-free breads available on the Polish market, and to assess the non-cancerous and cancerous exposure of consumers to cadmium (Cd) and lead (Pb). This assessment covered the exposure of eight age groups. The content of Cd and Pb was determined by the ET-AAS method.

RESULTS: The highest mean concentration of Cd was recorded in wheat-rye bread (0.072 mg/kg), the lowest in gluten-free bread (0.021 mg/kg). The general distribution of Cd in individual types of bread was as follows: wheat-rye > wheat > rye > gluten-free bread.

CONCLUSIONS: The estimation of exposure to Cd and Pb of the consumers of wheat and wheat-rye bread showed that a significant health risk may occur in the population of children < 11 years of age.

KEYWORDS

bread, gluten, gluten-free, lead, cadmium, health risk

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STRESZCZENIE

WSTĘP: Produkty zbożowe, zwłaszcza pieczywo, odgrywają istotną rolę w żywieniu ludzi na całym świecie. Jednak ze względu na uwalnianie nadmiernych ilości zanieczyszczeń do środowiska produkty zbożowe mogą również zawierać pewne ilości pierwiastków potencjalnie toksycznych, które mogą mieć negatywny wpływ na organizm człowieka.
MATERIAŁ I METODY: Celem pracy jest analiza zawartości metali ciężkich w wybranych chlebach glutenowych i bezglutenowych dostępnych na polskim rynku oraz ocena nienowotworowego i nowotworowego narażenia konsumentów na kadm (Cd) i ołów (Pb). Ocena ta obejmowała narażenie ośmiu grup wiekowych. Zawartość Cd i Pb oznaczono metodą ET-AAS.
WYNIKI: Najwyższą średnią zawartość Cd odnotowano w pieczywie pszenno-żytnim (0,072 mg/kg), najniższą w pieczywie bezglutenowym (0,021 mg/kg). Ogólny rozkład Cd w poszczególnych rodzajach chleba przedstawiał się następująco: pszenno-żytni > pszenny > żytni > bezglutenowy.
WNIOSKI: Oszacowanie narażenia na Cd i Pb konsumentów pieczywa pszennego i pszenno-żytniego wykazało, że w populacji dzieci w wieku < 11 lat może wystąpić istotne ryzyko zdrowotne.

SŁOWA KLUCZOWE

chleb, gluten, bezglutenowy, ołów, kadm, ryzyko zdrowotne

INTRODUCTION

Grain products, especially bread, play a vital role in the nutrition of people around the world. Therefore, in recent years, nutritionists have paid special attention to bread, taking into account modern nutritional recommendations, which consider grain products as the basis of a balanced diet. In all the schemes promoting the principles of proper nutrition (so-called food pyramids) developed to date, grain products are placed almost at the base of the triangle, which should constitute the basis of the daily food ration of a healthy person, whereby their recommended consumption differs [1]. The consumption of bread in Poland is falling from year to year. Bread consumption is constantly decreasing, although it is a product rooted in the Polish culinary tradition. In 2020, a statistical resident of Poland ate on average less than 2.75 kg of bread per month, a year earlier it was 2.98 kg [2]. However, bread is still the most frequently consumed product in the group of grain products and in the largest quantities. The regularly declining sales are mainly influenced by the trend for healthy eating, which requires the elimination of carbohydrates from the daily diet, especially white bread. The decrease in consumption is also influenced by the reduced quality of most bread, compared to the "old times", although Polish domestic bread is largely unique compared to foreign ones – it has an individual taste and is diverse. Whole grain, wheat, rye, yeast and sourdough bread are made, with the addition of, for example, bran, herbs, nuts, tomatoes and even dried fruit.
A diet based on grain products guarantees the proper development of our body because these products provide energy from the complex carbohydrates contained in them (mainly starch), proteins – an essential building component, B vitamins (in particular B1, B2, PP), minerals (calcium, iron, magnesium, zinc) and fiber [3]. Nevertheless, as a result of the release of excessive amounts of pollutants into the environment related to industrial or agricultural activities (the use of artificial fertilizers and plant protection products), the grain products that we eat on a daily basis may also contain certain amounts of potentially toxic elements, which may have negative effects on the human body. As is commonly known, both cadmium (Cd) and lead (Pb) belong to the group of elements potentially dangerous to human health, and excessive amounts accumulated in the body can cause chronic or even acute poisoning. Heavy metals, including Cd and Pb, are neither degradable nor biodegradable. Once taken into the body, they bioaccumulate in it, most often in parenchymal organs such as the kidneys or liver, and are also deposited in the bones or the brain. They demonstrate mutagenic, embryotoxic, teratogenic, hepatotoxic, nephrotoxic, neurotoxic and even carcinogenic effects [4]. According to the classification of the International Agency for Research on Cancer (IARC), Cd is an element with confirmed carcinogenic activity in the human body (group I), inorganic Pb compounds are included in group 2A (possibly carcinogenic), and Pb itself in group 2B (presumably carcinogenic) [5].
In recent years, there has been growing public interest in gluten and a gluten-free diet. A gluten-free diet has become very popular and is promoted by many celebrities. It is estimated that approximately 10–20% of US and Australian residents consume gluten-free foods [6]. Nonetheless, this is not always related to medical indications. Gluten is a protein fraction found in the seeds of cereals such as wheat (gliadin), rye (secalin) and barley (hordein) [7]. The technological properties of gluten is the reason why gluten is so common. It gives the characteristic elasticity and texture to products made of gluten cereals. It is a protein that cannot be fully digested by the human body; it contains a 33-amino acid fragment consisting of, among others, glutamine and proline, which are resistant to gastric juice and all proteolytic enzymes in the human gastrointestinal tract [7,8].

There are many disorders associated with the consumption of gluten. Celiac disease, i.e. gluten-dependent celiac disease, gluten-sensitive enteropathy, or non-tropical sprue is an inflammatory enteropathy of the small intestine of autoimmune etiology. It applies to people with a genetic predisposition and it is associated with a permanent intolerance to gluten. Contrary to popular belief, there is no way to "outgrow celiac disease". This is because celiac disease is misconceived as a food allergy. It is estimated that it affects 0.3–1% of people, but symptoms appear in about one in 3,345 people [8]. In patients diagnosed with celiac disease and Duhring's disease, the only effective method of treatment is to follow an absolutely gluten-free diet until the end of life [9].

According to the Commission Implementing Regulation (EU) No 828/2014, gluten-free products are those that do not naturally contain gluten or contain gluten in a concentration of less than 20 ppm, i.e. 20 mg/kg [10]. Only products containing less than 20 ppm gluten may bear the word "gluten-free" on the packaging, which should be next to the product name. Naturally gluten-free products should not be labeled as special dietary products, however, they may be described as naturally gluten-free, provided that the requirements for gluten-free food are met. Gluten-free products, unlike gluten ones, are characterized by a low content of B vitamins, iron, zinc, magnesium, calcium and dietary fiber. Moreover, they contain large amounts of fat (including saturated and hydrogenated fats), sucrose and starch [10]. Additionally, increasingly more studies show that people on a gluten-free diet are exposed to higher concentrations of toxic elements such as Cd, Pb, mercury and arsenic than those who do not follow such a diet. The increased exposure results, among other things, from the way of cultivating gluten-free cereals (e.g. rice), as well as the greater ability to accumulate heavy metals by these cereals [11].

The study assumes that gluten-free bread, due to its composition – a high content of rice or maize flour, will be highly contaminated with heavy metals, and therefore their consumption will constitute a significant health risk.

In order to verify the hypothesis, the aim of the study was outlined, which is to analyze the content of Cd and Pb in selected gluten and gluten-free breads available on the Polish market, and then to assess the non-cancerous and cancerous exposure of consumers of different ages to these heavy metals.

MATERIAL AND METHODS

The research material consisted of 50 bread samples in total, in which the content of Cd and Pb was analyzed. The tested breads were purchased in various cities in the Silesia Province (Poland). The collected research material was characterized by a diverse composition, mainly in terms of the type of flour used for baking. Taking into account the increasing number of people diagnosed with food intolerances (e.g. allergy to gluten or celiac disease), 10 samples of gluten-free bread were tested in the study, taking into account the availability and dissemination of the brand on the Polish market. The characteristics of the collected samples are presented in Table I.

Table I. Characteristics of analyzed samples

Kind of bread	Number of samples
Gluten-free	10
Wheat-rye	12
Wheat	20
Rye	8

The preparation of the purchased bread for analysis first of all involved thorough homogenization of the research material, and then the preparation of 1 g (± 3%) samples. All the samples were subjected to the microwave mineralization process in a closed system Magnum II mineralizer (Ertec, Poland). 8 ml of ultrapure nitric acid (V) and 1 ml of hydrogen peroxide (Merck, Germany) were used as reactants. After the completion of mineralization, the samples were poured into 25 ml volumetric flasks and made up to the required volume with ultrapure water. The parameters of the microwave mineralization process are presented in Table II.

Table II. Parameters of microwave mineralization process

Stage	Time	Power	Pressure	Temperature
I	5 min	60%	17–20 bar	295–300°C
II	5 min	80%	30–32 bar	295–300°C
III	10 min	100%	42–45 bar	295–300°C
IV (sample cooling)	10 min	–	–	–

The content of Cd and Pb in the mineralized bread samples was determined by the method of atomic absorption spectrometry with electrothermal atomization (ET-AAS) on a Savanta Sigma spectrometer by GBC (Australia). Before starting the analysis, a calibration curve was made. In order to monitor the validity of the results, during the determination of Cd and Pb in the bread samples, the analysis also included double samples and tests with the addition of a standard (Cd and Pb), the determinations of which fell within the acceptable range. Additionally, a matrix modifier was used to determine the Pb content (10 mg/ml $NH_4H_2PO_4$ & 600 µg/ml $Mg(NO_3)_2$ in 2% HNO_3). The LOD (limit of detection) and LOQ (limit of quantification) values for Cd and Pb, wavelengths and other method parameters are presented in Table III.

Table III. Method parameters

Parameter	Cd	Pb
LOD	0.0043 mg/kg	0.044 mg/kg
LOQ	0.008 mg/kg	0.08 mg/kg
Wavelength	228.8 nm	217 nm
Lamp current	0.3 mA	5.0 mA
Slit width	0.5 nm	1.0 nm

Cd – cadmium; Pb – lead; LOD – limit of detection; LOQ – limit of quantification.

Based on the obtained results, an estimated, lifetime assessment of the exposure of gluten and gluten-free bread consumers to Cd and Pb was performed. This assessment covered the exposure of eight age groups and examined non-cancerous and cancerous health risks. The assessment was made on the basis of the formulas recommended by the American Environmental Protection Agency (US EPA) [12,13]. At the outset, the average daily dose (ADD) was calculated, which was the starting value for further calculations – both non-cancerous and cancerous exposure. Then, based on the obtained ADD values, the hazard quotient (HQ) was calculated. Based on the obtained HQ values, calculated for Cd and Pb, the hazard index (HI) was calculated for both elements. Based on the slope factor (SF) and the baseline ADD value, the cancer risk (CR) and the cancer risk index (CRI) were calculated. The formulas used to calculate the individual indicators are presented below.

$$\mathbf{ADD}\ [\frac{\text{mg}}{\text{kg}}] = \frac{\mathbf{C}\ \left[\frac{\text{mg}}{\text{kg}}\right] \text{x}\ \mathbf{IngR}\ \left[\frac{\text{kg}}{\text{day}}\right]}{\mathbf{BW}\ [\text{kg}]}$$

where: ADD – average daily dose, C – concentration, IngR – ingestion rate, BW – body weight [12]. The consumption (IngR) used in the calculation was 100 g, which corresponds to an average of 4 slices of bread.

Non-cancerous exposure

$$\mathbf{HQ} = \frac{\mathbf{ADD}\ \left[\frac{\text{mg}}{\text{kg}}\right]}{\mathbf{RfD}\ \left[\frac{\text{mg}}{\text{kg}}\right]}$$

where: HQ – hazard quotient, RfD – reference dose [12].

$$\mathbf{HI = HQ1\ +\ HQ2} + \ldots + \mathbf{HQx}$$

where: HI – hazard index, HQx – the hazard quotient for a given *x* element [12].

Cancerous exposure

$$\mathbf{CR} = \frac{\mathbf{ADD}\ \left[\frac{\text{mg}}{\text{kg}}\right]}{\mathbf{SF}\ \left[\frac{\text{mg}}{\text{kg}}\right]}$$

where: CR – cancer risk, SF – slope factor [13].

$$\mathbf{CRI = CR1\ +\ CR2} + \ldots + \mathbf{CRx}$$

where: CRI – cancer risk index, CRx – cancer risk for a given *x* element [13].

The HQ, HI, CR, and CRI scores are unitless values. In the case of HQ and HI, a result of less than or equal to 1 is considered a safe level for the study population. A score of more than 1 is associated with the possibility of negative, non-cancerous health effects in the exposed population, where the amount consumed and the degree of contamination of consumed products will remain at the current level. However, it is only an indicative value, which does not mean that these effects will occur or will not occur at all [12,13].
The CR index is defined as the lifetime probability of developing cancer in an exposed population/person as a result of 24-hour exposure to a given daily amount of a carcinogen for seventy years. Acceptable CR limits are assumed to be from 1×10^{-6} to $< 1 \times 10^{-4}$ for a single carcinogen and multi-element carcinogens. For example, a CR of 10^{-4} suggests that 1 in 10,000 people is likely to develop cancer. The SF factor is defined as the risk generated by the lifetime mean amount of 1 mg/kg/day of a carcinogen and is specific to the pollutant – the element [13]. Table IV shows the values of the reference doses (RfDs) and slope factors (SFs) for Cd and Pb [14,15,16,17].

Table IV. Reference dose (RfD) and slope factor (SF) for cadmium (Cd) and lead (Pb)

Element	RfD [mg/kg/day]	SF [mg/kg/day]
Cd	0.0010	6.1
Pb	0.0036	8.5

The risk was estimated for the age groups recommended by US EPA [18], taking into account the average body weight in a given group (Table V).

Table V. Age groups and their assigned average body weight

Population	Body weight [kg]
Child 6–11 months	9.20
Child 1–<2 years	11.40
Child 2–<3 years	13.80
Child 3–<6 years	18.60
Child 6–<11 years	31.80
Child 11–<16 years	56.80
Child 16–<21 years	71.60
Adult	80.00

RESULTS

The highest average Cd concentration in the analyzed types of bread was recorded in wheat-rye bread

(0.072 mg/kg), while the lowest was in gluten-free bread (0.021 mg/kg). A Pb concentration above the limit of quantification (> LOQ) was recorded only in one wheat bread sample (Table VI). The general distribution of Cd in individual types of bread was as follows: wheat-rye bread > wheat bread > rye bread > gluten-free bread (Figure 1).

Table VI. Descriptive statistics of distribution of cadmium (Cd) and lead (Pb) concentrations in tested bread samples

Type of bread	Cd [mg/kg]			Pb [mg/kg]		
	min	max	mean	min	max	mean
Gluten-free	< LOQ	0.040	0.021	< LOQ	< LOQ	< LOQ
Wheat-rye	0.030	0.164	0.072	< LOQ	< LOQ	< LOQ
Wheat	0.022	0.090	0.050	< LOQ	0.152	–
Rye	0.008	0.088	0.041	< LOQ	< LOQ	< LOQ

LOQ – limit of quantification

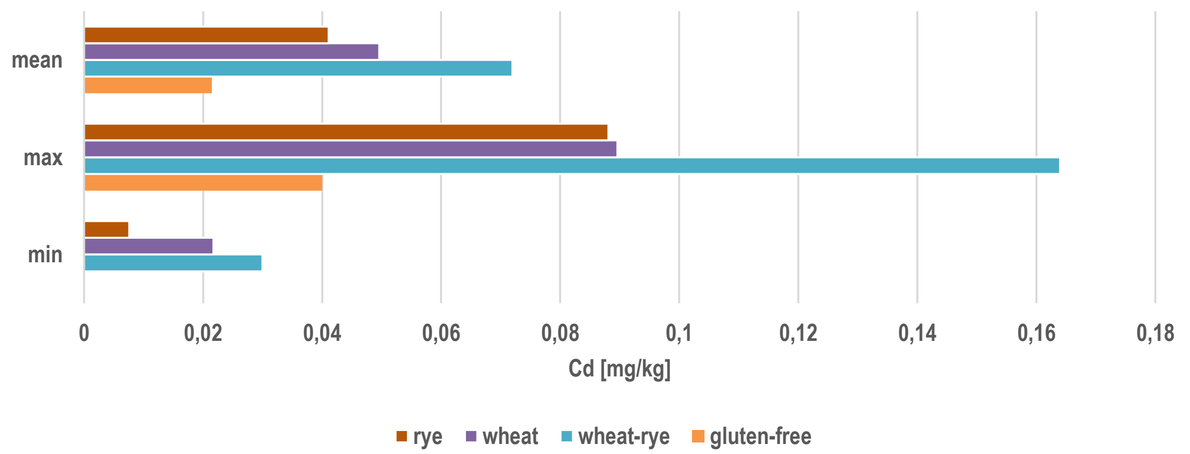

Fig. 1. Descriptive statistics of cadmium (Cd) concentration distribution in tested bread samples.

Pursuant to Regulation 2021/1323 on the maximum levels for Cd in certain foodstuffs [19], no separate limit values have been established for bread. Nevertheless, the maximum allowable level of Cd in cereals is indicated, amounting to 0.10 mg/kg of fresh weight, distinguishing rye from this group, for which the limit value is 0.050 mg/kg [19]. Similar regulations apply to the content of Pb, the limit value of which in cereals according to Regulation 2021/1317 [20] is 0.20 mg/kg fresh weight. In the only sample where the Pb content was > LOQ, which was a wheat bread sample, the maximum level was not exceeded. In the case of rye bread, the limit value of Cd was exceeded in 2 of 8 samples. In the case of the remaining samples, the permissible Cd content of 0.10 mg/kg was exceeded in one sample and it applied to wheat-rye bread.

Taking into account the hazard quotient calculated for the minimum, average and maximum Cd concentration determined in the tested bread samples, the value of HQ > 1 was recorded in 3 exposure cases, in the scenario assuming the maximum concentration of Cd determined in the tested samples of wheat-rye bread, in the population of children aged from 6 months to < 3 years (Table VII, VIII and IX).

Table VII. Average daily dose (ADD) and hazard quotient (HQ) calculated for minimum cadmium concentration determined in tested bread samples

Type of bread	Child 6–11 months	Child 1–<2 years	Child 2–<3 years	Child 3–<6 years	Child 6–<11 years	Child 11–< 16 years	Child 16–< 21 years	Adult
	ADD							
Wheat-rye	0.000326	0.000263	0.000217	0.000161	0.000094	0.000053	0.000042	0.000037
Wheat	0.000236	0.000191	0.000157	0.000117	0.000068	0.000038	0.000030	0.000027
Rye	0.000083	0.000067	0.000055	0.000041	0.000024	0.000013	0.000011	0.000009
	HQ							
Wheat-rye	0.33	0.26	0.22	0.16	0.09	0.05	0.04	0.04
Wheat	0.24	0.19	0.16	0.12	0.07	0.04	0.03	0.03
Rye	0.08	0.07	0.06	0.04	0.02	0.01	0.01	0.01

Table VIII. Average daily dose (ADD) and hazard quotient (HQ) calculated for mean cadmium concentration determined in tested bread samples

Type of bread	Child 6–11 months	Child 1–<2 years	Child 2–<3 years	Child 3–<6 years	Child 6–<11 years	Child 11–<16 years	Child 16–<21 years	Adult
	ADD							
Gluten-free	0.000234	0.000188	0.000156	0.000116	0.000068	0.000038	0.000030	0.000027
Wheat-rye	0.000782	0.000631	0.000522	0.000387	0.000226	0.000127	0.000101	0.000090
Wheat	0.000539	0.000435	0.000359	0.000267	0.000156	0.000087	0.000069	0.000062
Rye	0.000447	0.000361	0.000298	0.000221	0.000129	0.000072	0.000057	0.000051
	HQ							
Gluten-free	0.23	0.19	0.16	0.12	0.07	0.04	0.03	0.03
Wheat-rye	0.78	0.63	0.52	0.39	0.23	0.13	0.10	0.09
Wheat	0.54	0.44	0.36	0.27	0.16	0.09	0.07	0.06
Rye	0.45	0.36	0.30	0.22	0.13	0.07	0.06	0.05

Table IX. Average daily dose (ADD) and hazard quotient (HQ) calculated for maximum cadmium concentration determined in tested bread samples

Type of bread	Child 6–11 months	Child 1–<2 years	Child 2–<3 years	Child 3–<6 years	Child 6–<11 years	Child 11–< 16 years	Child 16–<21 years	Adult
	ADD							
Gluten-free	0.000435	0.000351	0.000290	0.000215	0.000126	0.000070	0.000056	0.000050
Wheat-rye	0.001783	0.001439	0.001188	0.000882	0.000516	0.000289	0.000229	0.000205
Wheat	0.000974	0.000786	0.000649	0.000482	0.000282	0.000158	0.000125	0.000112
Rye	0.000083	0.000067	0.000055	0.000041	0.000024	0.000013	0.000011	0.000009
	HQ							
Gluten-free	0.44	0.35	0.29	0.22	0.13	0.07	0.06	0.05
Wheat-rye	**1.78**	**1.44**	**1.19**	0.88	0.52	0.29	0.23	0.21
Wheat	0.97	0.79	0.65	0.48	0.28	0.16	0.13	0.11
Rye	0.08	0.07	0.06	0.04	0.02	0.01	0.01	0.01

The CR at a level equal to and higher than 10^{-4} was recorded in the exposure scenario assuming the average Cd concentration determined in the tested samples of wheat-rye bread – for the population of children from 6 months to < 2 years of age, and the maximum concentration of Cd determined in the tested samples of wheat-rye bread – for the population of children aged 6 months to < 6 years of age and in the studied samples of wheat bread – for the population of children from 6 months to < 3 years of age (Table X).

The HQ value determined for the maximum concentration of Pb in the tested sample of wheat bread for any of the analyzed age groups did not exceed a value equal to 1. However, a cancer risk exceeding 10^{-4} was reported for the population of children from 6 months to < 3 years of age (Table XI).

The non-cancer hazard index (HI) calculated for the maximum concentration of Pb and Cd determined in the samples of wheat bread exceeded a value equal to 1 in two cases – for the population of children from 6 months to < 2 years of age. The cancer risk index at a level higher than 10^{-4} was obtained for 5 of the analyzed age groups – for children aged 6 months to < 11 years (Table XII).

Table X. Cancer risk (CR) calculated for cadmium (Cd) contained in tested bread samples

Type of bread	Child 6–11 months	Child 1–<2 years	Child 2–<3 years	Child 3–<6 years	Child 6–<11 years	Child 11–<16 years	Child 16–<21 years	Adult
	CR for <u>minimal</u> concentration of Cd							
Wheat-rye	5.3×10^{-5}	4.3×10^{-5}	3.6×10^{-5}	2.6×10^{-5}	1.5×10^{-5}	9.0×10^{-6}	7.0×10^{-6}	6.0×10^{-6}
Wheat	3.9×10^{-5}	3.1×10^{-5}	2.6×10^{-5}	1.9×10^{-5}	1.1×10^{-5}	6.0×10^{-6}	5.0×10^{-6}	4.0×10^{-6}
Rye	1.4×10^{-5}	1.1×10^{-5}	9.0×10^{-6}	7.0×10^{-6}	4.0×10^{-6}	2.0×10^{-6}	2.0×10^{-6}	2.0×10^{-6}
	CR for <u>mean</u> concentration of Cd							
Gluten-free	3.8×10^{-5}	3.1×10^{-5}	2.6×10^{-5}	1.9×10^{-5}	1.1×10^{-5}	6.2×10^{-6}	4.9×10^{-6}	4.4×10^{-6}
Wheat-rye	**1.3×10^{-4}**	**1.0×10^{-4}**	8.6×10^{-5}	6.3×10^{-5}	3.7×10^{-5}	2.1×10^{-5}	1.7×10^{-5}	1.5×10^{-5}
Wheat	8.8×10^{-5}	7.1×10^{-5}	5.9×10^{-5}	4.4×10^{-5}	2.6×10^{-5}	1.4×10^{-5}	1.1×10^{-5}	1.0×10^{-5}
Rye	7.3×10^{-5}	5.9×10^{-5}	4.9×10^{-5}	3.6×10^{-5}	2.1×10^{-5}	1.2×10^{-5}	9.4×10^{-6}	8.4×10^{-6}
	CR for <u>maximum</u> concentration of Cd							
Gluten-free	7.1×10^{-5}	5.8×10^{-5}	4.8×10^{-5}	3.5×10^{-5}	2.1×10^{-5}	1.2×10^{-5}	9.2×10^{-6}	8.2×10^{-6}
Wheat-rye	**2.9×10^{-4}**	**2.4×10^{-4}**	**2.0×10^{-4}**	**1.5×10^{-4}**	8.5×10^{-5}	4.7×10^{-5}	3.8×10^{-5}	3.4×10^{-5}
Wheat	**1.6×10^{-4}**	**1.3×10^{-4}**	**1.1×10^{-4}**	7.9×10^{-5}	4.6×10^{-5}	2.6×10^{-5}	2.1×10^{-5}	1.8×10^{-5}
Rye	1.4×10^{-5}	1.1×10^{-5}	9.0×10^{-6}	6.7×10^{-6}	3.9×10^{-6}	2.2×10^{-6}	1.7×10^{-6}	1.6×10^{-6}

Table XI. Average daily dose (ADD), hazard quotient (HQ) and cancer risk (CR) calculated for maximum lead concentration determined in tested bread sample

Parameter	Child 6–11 months	Child 1–<2 years	Child 2–<3 years	Child 3–<6 years	Child 6–<11 years	Child 11–<16 years	Child 16–<21 years	Adult
ADD	0.001650	0.001332	0.001100	0.000816	0.000477	0.000267	0.000212	0.000190
HQ	0.46	0.37	0.31	0.23	0.13	0.07	0.06	0.05
CR	**1.9×10^{-4}**	**1.6×10^{-4}**	**1.3×10^{-4}**	9.6×10^{-5}	5.6×10^{-5}	3.1×10^{-5}	2.5×10^{-5}	2.2×10^{-5}

Table XII. Non-cancer hazard index (HI) and cancer risk index (CRI) calculated for maximum concentration of lead and cadmium determined in wheat bread samples

Parameter	Child 6–11 months	Child 1–<2 years	Child 2–<3 years	Child 3–<6 years	Child 6–<11 years	Child 11–<16 years	Child 16–<21 years	Adult
HI	**1.43**	**1.16**	0.95	0.71	0.41	0.23	0.18	0.16
CRI	**3.54×10^{-4}**	**2.86×10^{-4}**	**2.36×10^{-4}**	**1.75×10^{-4}**	**1.02×10^{-4}**	5.73×10^{-5}	4.55×10^{-5}	4.07×10^{-5}

DISCUSSION

In the past, only people diagnosed with celiac disease, about 1% of the population, avoided gluten in their diets. In recent years, the mass media have often misrepresented gluten-free food as a healthier choice, as a result of which ever more people say that gluten is a harmful part of the diet and should be eliminated [21]. A market study conducted in the US in 2013 by the Mintel Group found that 17% of the respondents avoided gluten, while 31% replied that a gluten-free diet is a "fad diet". Nonetheless, two years later, when asked the same question, 25% of the respondents answered that they were on a gluten-free diet, and 47% of the respondents indicated that a gluten-free diet was a diet caused by a whim. It is worrying that only 11% of the respondents on a gluten-free diet consulted their doctor or other health care professional on the decision to introduce it. It is especially dangerous because the self-exclusion of gluten may make it impossible to diagnose celiac disease later [22].

Naghipour et al. [23] examined 40 different grades of gluten bread available in Iran. In the tested samples they examined, among others, the Cd and Pb content. The obtained Pb concentrations for all the tested samples ranged from 0 to 3.3 mg/kg, while the Cd concentrations ranged from 0 to 0.4 mg/kg. The results of the Iranian researchers were higher than those obtained in the present study. Basaran [24] tested 60 samples of bread available on the Turkish market. The average Cd content in the tested material was 0.015 mg/kg, and Pb was 0.034 mg/kg. Analyzing only the Cd content, these values were lower than those obtained in the present research. In Polish studies by Hajok et al. [25], the Cd content was determined, among others, in wheat and wheat-rye bread. The obtained concentration range was as follows: wheat bread 0.01–0.07 mg/kg, wheat-rye bread 0.01–0.05 mg/kg. Comparing the concentration values obtained in the selected types of bread, in the author's own work, comparable Cd values in wheat bread and higher values in wheat-rye bread were determined. In a Nigerian study by Irogbeyi et al. [26], the Pb content was found at the level of 0.05–0.45 mg/kg.

Basaran [24] assessed the non-cancer risk of the Turkish population in his research, and the resulting HQ was lower than 1. Hajok et al. [25] also obtained a value of HQ below 1, and the assessment covered the Polish population. The assessments carried out by the researchers covered the general population and did not take into account a division into age groups, which, as shown in the present study, is crucial in estimating individual health risk. The authors are aware that for the youngest age groups (especially children from 6 months to under a year), the assumed consumption of 100 g of bread a day is high, but not impossible.

Due to the fact that a gluten-free diet is often based on rice products, people who follow it are exposed to an additional portion of arsenic. Scientists have found that such a diet can provide up to twice as much of this compound, which is a known cause of cancer. Experts also point out that the amount of mercury, another toxic chemical found in gluten-free products, is nearly 70 percent greater than in a normal diet. Researchers warn that people who do not need to follow such a diet, i.e. people who do not experience stomach problems after eating gluten-containing products, should not switch to it. Gluten-free versions of grain products, including bread and spaghetti, often contain rice flour as a wheat substitute. However, rice can contain up to ten times more arsenic than other foods because of the way it is grown [27,28,29]. In the studies by Raehsler et al. [30] it was found that those on a gluten-free diet had significantly higher levels of total arsenic in their urine and levels of mercury, Pb, and Cd in their blood than those who did not avoid gluten. The authors also emphasized that research is needed to determine the long-term effects of the accumulation of these elements in people on a gluten-free diet. The current study did not include the determination of mercury or arsenic in the tested bread samples, both gluten and gluten-free, which is a work limitation and should be supplemented with these analyses. In the study by Naghipour et al. [23] the arsenic concentrations in the tested samples of gluten bread were each time below the limit of quantification (< LOQ). In the Basaran study [24] the mean concentration of arsenic in gluten breads was 0.019 mg/kg, and the concentration of mercury was 0.0004 g/kg. No studies were found in which these elements were determined in gluten-free bread.

After examining the available scientific papers, the authors would like to point out that the presented results are innovative owing to the inclusion of gluten-free bread in the research and because of the estimated cancer and non-cancer health risks related to the consumption of Cd-contaminated bread (and in the case of one sample, also Pb).

CONCLUSIONS

1. The highest concentrations of Cd were recorded in wheat-rye bread. In turn, the content of this element was the lowest in gluten-free bread.
2. The concentrations of Cd and Pb were definitely lower in gluten-free than in gluten breads. In subsequent studies, mercury and arsenic should be included in the analyses.
3. In the study population, with a constant level of consumption and the persistent degree of Cd contamination of bread, there may be a significant non-cancer health risk. This risk concerns the group of children < 3 years of age and includes the consumption of wheat-rye bread.
4. The estimation of exposure to Cd and Pb of consumers of wheat and wheat-rye bread allowed us to conclude that with a constant level of consumption and a constant degree of contamination of these products, a significant cancer health risk may occur in the population of children < 11 years of age.

Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Declaration of originality

This manuscript has not been published elsewhere and it has not been simultaneously submitted for publication elsewhere. All the tables and figures are our original work and no permissions are required.

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Data collection – M. Gacal, K. Gut-Pietrasz, M. Ćwieląg-Drabek
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