

EXPOSURE TO LEAD AND CADMIUM RELEASED FROM CERAMICS AND GLASSWARE INTENDED TO COME INTO CONTACT WITH FOOD

Małgorzata Rebeniak* Maria Wojciechowska-Mazurek, Monika Mania, Tomasz Szynal, Agnieszka Strzelecka, Krystyna Starska

Department of Food Safety, National Institute of Public Health– National Institute of Hygiene, Warsaw, Poland

ABSTRACT

Background. The dietary intake of harmful elements, particularly lead and cadmium constitutes a health threat and essential measures should be undertaken to reduce consumer exposure. The latest risk assessments by the European Food Safety Authority (EFSA) and Joint FAO/WHO Expert Committee on Food Additives (JECFA) have indicated that the Provisional Tolerable Weekly Intake (PTWI) for lead and cadmium do not ensure health safety and their review had to be undertaken. Migration from ceramics and glassware intended for food contact is an important source of lead and cadmium intake.

Objectives. To study the release of lead and cadmium from ceramics and glassware (including decorated products) intended for food contact that are available on the Polish market and to assess the resulting health risk to the consumer.

Materials and Methods. Ceramics and glassware (mainly decorated) were sampled from the Polish market during 2010-2012 throughout the country by staff of the Sanitary-Epidemiological Stations in accordance with monitoring procedures and guidelines designed by the National Institute of Public Health-National Institute of Hygiene. Migration of lead and cadmium was measured by incubating the samples with 4% acetic acid for 24 hours at a temperature of 22±2°C in the dark. Flame Atomic Absorption Spectrometry (FAAS) was used to measure these elements in food simulant according to a validated and accredited method (PN-EN ISO/IEC 17025).

Results. 1273 samples of ceramics and glass wares were analysed in 2010-2012. Lead and cadmium release were usually found to be below analytical detection limits. Permissible migration limits (as prescribed by the legislation) of these metals were rarely exceeded and were reported mainly in articles imported from outside the EU. Two imported and decorated ceramic flat plates released lead at 0.9 and 11.9 mg/dm² (limit 0.8 mg/dm²) and 5 imported deep plates gave migration values of 4.7 mg/L, 4.9 mg/L, 5.6 mg/L, 6.1 mg/L, 8.6 mg/L (limit 4.0 mg/L). Lead migrations from ceramic ware rims above the 2.0 mg per product limit (as established in Polish Standard PN-B-13210:1997 [16]) were observed in 4 samples, at 2.1, 3.7, 4.2 and 14.4 mg per product, respectively. Migrations of cadmium from the ceramic samples' rims were within permissible limits. Majority of high migration results were obtained for decorated rims of glass vessels for beverages. The highest migration from the rim of an imported glass mug was reported at 163.8 mg/product for lead and at 8.96 mg/product for cadmium. Risk assessment indicated that exposures to lead and cadmium released from ceramic wares based on the migration limits set by the EU legislation lead to human intake close to, or exceeding reference doses. For a 20 kg b.w. child the lead BMDL₀₁ value could thus be exceeded by over 30-fold and the cadmium TWI value 4-fold.

Conclusions. Review of EU legislation applicable to lead and cadmium migration limits from ceramics is necessary with an intention to lower such limits. The limits applied to the rims of ceramics and glassware intended for beverages should be included. The release of lead and cadmium at the maximum permissible levels for ceramics may lead to uptakes becoming hazardous to human health. Appropriate measures are thus necessary to reduce sources of exposure.

Key words: *lead, cadmium, ceramic food contact articles, glass food contact articles, lead migration, cadmium migration, lead exposure, cadmium exposure, food contact articles, risk assessment*

STRESZCZENIE

Wprowadzenie. Pobranie pierwiastków szkodliwych dla zdrowia, szczególnie ołowiu i kadmu z żywnością stwarza zagrożenie dla zdrowia i niezbędne jest podejmowanie działań mających na celu obniżenie narażenia konsumentów. Najnowsze oceny ryzyka dokonane przez Europejski Urząd ds. Bezpieczeństwa Żywności (EFSA) i Połączony Komitet Ekspertów FAO/WHO ds. Dodatków do Żywności (JECFA) wykazały, że wartości tymczasowego tolerowanego tygodniowego pobrania (PTWI) ołowiu i kadmu nie zapewniały bezpieczeństwa zdrowotnego i doprowadziły do ich weryfikacji. Jednym z istotnych źródeł pobrania ołowiu i kadmu z żywnością jest ich migracja z wyrobów ceramicznych i szklanych przeznaczonych do kontaktu z żywnością.

*Corresponding author: Małgorzata Rebeniak, Department of Food Safety, National Institute of Public Health - National Institute of Hygiene, 24 Chocimska street, 00-791 Warsaw, Poland, phone: +48 22 54 21 362, fax: +48 22 54 21 392,e-mail: mrebeniak@pzh.gov.pl

© Copyright by the National Institute of Public Health - National Institute of Hygiene

Cel. Zbadanie uwalniania ołowiu i kadmu z wyrobów ceramicznych i szklanych w tym dekorowanych przeznaczonych do kontaktu z żywnością dostępnych w obrocie w Polsce oraz ocena ryzyka dla zdrowia konsumenta.

Materiał i metody. Materiał do badań stanowiły wyroby ceramiczne i szklane, głównie dekorowane, pobierane z obrotu handlowego w latach 2010-2012 na terenie całego kraju przez przedstawicieli Stacji Sanitarno-Epidemiologicznych w ramach monitoringu, zgodnie z planem i wytycznymi opracowanymi przez Zakład Bezpieczeństwa Żywności Narodowego Instytutu Zdrowia Publicznego – Państwowego Zakładu Higieny. Migrację ołowiu i kadmu z próbek wyrobów ceramicznych i szklanych przeprowadzano do 4% kwasu octowego przez 24 godziny w temperaturze 22±2°C (w ciemności). Zawartość ołowiu i kadmu w płynie modelowym po migracji oznaczano metodą płomieniowej atomowej spektrometrii absorpcyjnej (FAAS), zwalidowaną i akredytowaną zgodnie z PN-EN ISO/IEC 17025.

Wyniki. W latach 2010-2012 zbadano 1273 próbki wyrobów ceramicznych i szklanych. Uwalnianie ołowiu i kadmu z naczyń ceramicznych i szklanych było przeważnie poniżej granic wykrywalności stosowanych metod analitycznych. Przekroczenia limitów migracji tych metali z naczyń ceramicznych określonych w ustawodawstwie stwierdzano rzadko, głównie z wyrobów z importu (spoza UE). Z 2 naczyń ceramicznych płaskich zdobionych (z importu) ilości uwalnianego ołowiu wynosiły 0,9 mg/dm² i 11,9 mg/dm² (limit 0,8 mg/dm²). Z 5 naczyń ceramicznych głębokich (z importu) migracja ołowiu powyżej limitu (4,0 mg/L) wynosiła: 4,7 mg/L; 4,9 mg/L; 5,6 mg/L; 6,1 mg/L; 8,6 mg/L. Migrację ołowiu z obrzeży naczyń ceramicznych w ilościach przekraczających limit (2,0 mg/wyrób), podany w polskiej normie PN-B-13210:1997 [16], stwierdzono z 4 wyrobów i wynosiła ona: 2,1 mg/wyrób 3,7 mg/wyrób, 4,2 mg/wyrób, 14,4 mg/wyrób. Nie stwierdzono migracji kadmu z obrzeży badanych naczyń ceramicznych w ilości przekraczającej limit. Największą migrację ołowiu wynoszącą 163,8 mg/wyrób i kadmu 8,96 mg/wyrób stwierdzono z obrzeża kubka szklanego, z importu. Ocena narażenia na ołów i kadm wykazała, że uwalnianie ołowiu i kadmu na poziomie najwyższych dopuszczalnych ilości, podanych w przepisach UE, prowadzi do pobrania tych metali bliskiego bądź przekraczającego dawki referencyjne. W przypadku dziecka o masie ciała 20 kg przekroczenie dawki BMDL₀₁ dla ołowiu może być ponad 30-krotne, a przekroczenie TWI dla kadmu ponad 4-krotne.

Wnioski. Konieczne jest obniżenie obowiązujących w UE limitów migracji ołowiu i kadmu z naczyń ceramicznych oraz wprowadzenie limitów dla obrzeży naczyń ceramicznych przeznaczonych do napojów, a także dla wyrobów szklanych. Uwalnianie ołowiu i kadmu na poziomie najwyższych dopuszczalnych ilości zgodnych z aktualnymi przepisami dla wyrobów ceramicznych może prowadzić do pobrania tych metali zagrażającego zdrowiu. Niezbędne jest podjęcie działań eliminujących źródła zagrożenia.

Słowa kluczowe: ołów, kadm, naczynia ceramiczne, wyroby szklane do kontaktu z żywnością, migracja ołowiu, migracja kadmu, narażenie na ołów, narażenie na kadm, ocena ryzyka

INTRODUCTION

Human dietary exposure to chemical elements that are harmful to health, especially in children, is still significant although it has been decreasing in recent years. This drop has been mainly due to actions taken to reduce uptake of such contaminants from the environment, like the curtailing of industrial emissions, using lead-free petrol or withdrawing packaging soldered with lead containing binders.

International organisations such as JECFA and EFSA have recommended taking measures for reducing exposure to lead and cadmium. These are backed by the latest risk assessment results based on toxicological and epidemiological studies that change tolerable intake levels of many metals. Absence of evidence for existence of a threshold for critical lead effects is of particular importance. The Committee, therefore, concluded that the PTWI could no longer be considered health protective, and it was withdrawn.

Lead toxicity is one of the causes of cardiovascular diseases as well as of disorders in heme biosynthesis, vitamin D metabolism, renal and hepatic functioning. In 2010, the JECFA decided to withdraw the 0.025 mg/kg body weight (b.w.) value for the PTWI because it did not confer a guarantee of health safety [7]. However, EFSA has provided a Benchmark Dose Lower Confidence Limit (BMDL₀₁) for lead in children set at $0.5 \ \mu g/kg b.w.$ daily (based on neurotoxic effects) and a BMDL₁₀ level of 0.63 $\ \mu g/kg b.w.$ daily in adults (based on nephrotoxic effects), together with a BMDL₀₁ of 1.5 $\ \mu g/kg b.w.$ daily (based on cardiovascular disorders) as reference doses when performing risk assessment [6].

Cadmium is carcinogenic (Group I according to the International Agency for Research on Cancer - IARC), nephrotoxic, neurotoxic to the Central Nervous System (CNS) during development, causes cardiovascular diseases, osteomalacia, liver damage and deficiencies of iron, copper and zinc. Due to such toxic effects of cadmium, EFSA pointed out the need to reduce the currently set PTWI level by 2.8-fold from 0.007 mg/kg b.w. to 0.0025 mg/kg b.w. [21, 25]. Furthermore, JECFA has replaced the PTWI with the Provisional Tolerable Monthly Intake (PTMI) set at 0.025 mg/kg b.w. [7].

Ceramic articles and glassware coming into contact with foodstuffs may release metals in amounts significant for affecting health, present either as contaminants or as constituents in the glaze and majority of ceramic colours [10, 12, 15, 30]. The durability of enamel and decorations depends on how such coatings are made and products manufactured, including the time and temperature of firing, along with the conditions of using the ceramic product, e.g. pH of the foodstuffs concerned and the duration and temperature of contact [8, 12, 13]. In the case of sodium-calcium-silicate or borosilicate glassware, the chances of any metals being released into foodstuffs are small. However, these increases in crystal glass, which contains around 7% - 32% lead oxides, or in decorated glass. [9].

Legally binding regulations have been introduced in the EU and several other countries which limit the migration of metals from ceramics and glassware. The process of firing, when done under uncontrolled conditions (e.g. when temperatures are too high or too low, uneven temperature distribution in ovens, etc.) still results in non-durable metal bonding [23, 28], as is particularly reported in the smaller scale undertakings. The most frequent examples of this quoted in the literature refer to Mexico, where there are a number of family-run manufacturers that for centuries have produced ceramic cooking vessels which contain lead enamelling, fired at low temperatures [5, 22, 27]. This creates a significant health problem for the Spanish community living in the USA. Over 50% of tested vessels of such origin reported migration of lead in excess of the acceptable levels [13]. A number of cases have also been reported of lead being released into juices and wine from ceramic vessels originating from the Mediterranean regions [23, 28]. A further and significant health risk is posed by the decorative rims of glassware intended for beverages. The increasing import of such ceramics and glassware from all over the world is becoming a significant health concern and, thus, needs to be placed under effective control [14, 30].

Obligatory limits and methods for measuring lead and cadmium migration from ceramics are set down in EU legislation (Directive 84/500/EEC [3] amended by Directive 2005/31/EC [2]), which has been incorporated into the appropriate Polish Ministry of Health regulation [20]; these requirements are presented in Table 1. Nevertheless, such provisions do not set limits for lead or cadmium migration from the rims of ceramics that comes into direct contact with the mouth or from the inside surface and rims of glassware.

Polish Standard PN-B-13210:1997 [16] has set a migration limit for lead and cadmium from the rims of vessels for beverages at: 2.0 mg/product and 0.2 mg/ product, respectively.

The European Commission Working Group on Food Contact Materials is currently working on introducing significantly lower migration limits into the legislation for both lead and cadmium as well as on setting migration limits of these metals from the rims of vessels for beverages [31].

Manufacturers of ceramics and glassware are obliged to comply with the requirements of the Commission Regulation (EC) No. 2023/2006 on good manufacturing practice for materials and articles intended to come into contact with food [4].

The aim of this study was to determine lead and cadmium migration from ceramics and glassware (including decorated products) intended to contact with food that are available on the Polish market. In this regard, the risk to consumer health was also assessed.

MATERIALS AND METHODS

The sampled test materials were both ceramic products (deep and flat dishes and vessels for beverages), with the majority of which had decorations on surfaces contacting with foodstuffs, as well as glassware decorated on the rim and crystal glassware. These samples were taken from all regions throughout Poland by representatives of the Sanitary-Epidemiological Stations, based on plans and guidelines designed by the Department of Food Safety at the NIPH-NIH that took into account (amongst other factors) the number of inhabitants in regions and the ratio of imports and domestic-made products.

The study undertaken during 2010, 2011 and 2012 consisted of:

- (1) In 2010: 281 decorated ceramic samples including 114 vessels for beverages, 99 deep dishes and 68 flat dishes together with 142 samples of glassware for beverages, of which 108 were decorated on the rim (mugs, beverage glasses and vodka/wine glasses) and 34 were crystal glass (mainly vodka/ wine glasses).
- (2) In 2011: 225 decorated ceramic samples including 105 vessels for beverages, 65 deep dishes and 55 flat dishes together with 167 samples of glassware for beverages, of which 131 were decorated on the rim (mugs, beverage glasses and vodka/wine glasses) and 36 were crystal glass (mainly vodka/ wine glasses).
- (3) In 2012: 245 decorated ceramic samples including 112 drinking vessels, 69 deep dishes and 64 flat dishes together with 213 samples of glassware for beverages, of which 154 were decorated on the rim (mugs, beverage glasses and vodka/wine glasses) and 59 were crystal glass (mainly vodka/wine glasses).-

The test samples were either domestic products and from the EU or mainly from Asian imports.

The laboratory sample for testing migration to the food simulant constituted of 4 individual identical pro-

ducts from the same manufacturer, made of the same material, with the same decoration, shape, dimensions and from the same batch.

Determining lead and cadmium migration from the samples was performed according to the following standard procedures: PN-EN 1388-1:2000 [17] and PN-EN 1388-2:2000 [18]. Article surfaces were subjected to 24 hour incubation in 4% acetic acid (food simulant) at 22°C±2 °C in the dark, with test solution subsequently analysed by Flame Atomic Absorption Spectrometry (FAAS). The analyses were performed in laboratories of the Provincial Sanitary-Epidemiological Stations, possessing the accreditation according PN-EN ISO/IEC 17025 [19]. Levels of metals were measured by validated methods performing criteria set out in Directive No 2005/31/EC [2].

RESULTS AND DISCUSSION

In all, 1273 samples of ceramics and glassware were analysed, of which over half were vessels for beverages having decorations on their rims that made contact with the mouth; majority of the products originated from non-EU countries.

 Table. 1
 Permissible limits of lead and cadmium release from ceramic wares [3]

Category of ceramic wares	Pb	Cd	Unit
<i>Category 1</i> Articles which cannot be filled and articles which can be filled, the internal depth of which, measured from the lowest point to the horizontal plane passing through the upper rim, does not exceed 25 mm	0.8	0.07	mg/dm ²
<i>Category 2</i> All other articles which can be filled	4.0	0.3	mg/L
Category 3 Packaging and storage vessels having a capacity of more than three liters	1.5	0.1	mg/L

Flat ceramic dishes

These consisted of 197 samples in which lead and cadmium migration were mostly below the method's detection limits. There were only two instances where lead concentrations exceeded the permissible limit of 0.8 mg/dm², i.e. at 0.9 and 11.9 mg/dm². Both these samples comprised imported and decorated ceramic plates. However, migration of cadmium remained within permissible levels; never exceeding the limit of 0.07 mg/dm². The proportion of samples where lead and cadmium fell within the analytical range of measurement was 17% and 7%, respectively.

Deep ceramic dishes (excluding vessels for beverages)

Of the 233 samples, 66% and 87% results were below the limits of detection of analytical methods for

lead and cadmium, respectively. Measurable concentrations below permissible lead levels were reported for 75 products (of which 92% were imported goods) and in case of cadmium for 31 products (97% were imported goods). Of the 5 imported samples with lead migration above the permissible limit (i.e. > 4.0 mg/L) the reported results were 4.7 mg/L, 4.9 mg/L, 5.6 mg/L, 6.1 mg/L and 8.6 mg/L, respectively.

Ceramic vessels for beverages

331 samples (mugs) were tested. Migration of lead and cadmium from inner surfaces of these samples did not exceed permissible limits. Measurable levels were detected for lead and cadmium in 8 and 6 samples, respectively, only. The releases of lead and cadmium from the rims of vessels for beverages in most instances were below the analytical detection limits. Measurable levels of lead migration from the rims was observed in 46 samples, of which 4 were imports with results of 2.1 mg, 3.7 mg, 4.2 mg and 14.4 mg per product that exceeded the 2.0 mg/product level set in PN-B 13210:1997 [16]. Cadmium migration from rims of ceramic vessels for beverages did not exceed the 0.2 mg/product limit. Only 18 out 303 samples had detectable levels of cadmium migration.

Results are shown in Tables 2 and 3.

Glasses for beverages

393 samples of glasses for beverages were tested. Lead and cadmium migration from the inner surfaces were for the most part analytically undetectable. Only 5% of samples gave detectable levels of lead (glasses for beverages, vodka/wine glasses) whilst there were only 2% reported in the case of cadmium (glasses for beverages). All results reported for cadmium migrating from vodka/wine glasses samples were below detection limits of analytical methods used.

Of the ceramic and glassware products tested, majority of high migration results were reported for richly decorated rims of glass vessels for beverages.

Migration of lead above limits set in Polish standard, was observed in 57 (20%) and 10 (11%) samples, respectively, of the 275 glasses for beverages and 95 vodka/wine glasses with decorated rims tested. Cadmium limits (0.2 mg/product) were exceeded in 52 (19%) beverage glasses and 7 (7%) vodka/wine glasses. The majority of non-compliant items came from Asian countries. As a rule, these transgressions were very high, with the greatest levels seen in the rim of an imported glass mug with a lead migration of 163 mg/product and cadmium migration of 8.96 mg/product. Detectable, but nevertheless permissible levels of lead migration were found in 27 (7.3%) samples and for cadmium in for 41 (11%) samples. Results are presented in Figures 1 and 2.

Product	Year	Origin*	No of samples	Distributions of samples						
					be	above permissible level	Unit			
				< 0.01	0.01-0.02	>0.07				
		т	45		1	0.02-0.035	0.035-0.07	20.07		
	2010	I EU	45 6	44	-	-	1	-		
	2010	K	17	6 16	-	-	- 1	-		
		total	68	66	-	-	1 2	-		
les		I	38	35	2	-	1	-		
Flat dishes	2011	I EU	3	35	2	-	1	-	mg/dm ²	
		K	14	13	-	-	- 1	-		
		total	55	51	2	-	2	-		
		I	53	48	2	2	1	-		
		EU	2	2	2	2	1	-		
	2012	K	9	7	2	-	-	-		
		total	64	57	4	2	1			
		totai	04	<0.01	0.01-0.05	0.05-0.15	0.15-0.30	>0.30		
		Ι	64	54	7	2	1	>0.50		
	2010	EU	13	54 13		-		-	- mg/L	
	2010	K	22	21	-	-				
Deep dishes **		total	99	88	8	2	- 1	-		
		I	56	47	7	2	-			
ish	2011	EU	1	47	/	2	-	-		
ib c		K	8	8	_	_	_			
Jee		total	65	56	7	2	_	_		
П		I	46	35	7	4	_		-	
		EU	6	6	-	-	_			
	2012	K	17	17	_	_	_	_		
		total	69	58	7	4	_	_		
		totur		< 0.01	0.01-0.05	0.05-0.15	0.15-0.30	>0.30		
		Ι	91	88	3	-	-	-		
	2010	EU	9	9	-	_	_	_		
es	2010	K	14	14	-	-	-	_		
Vessels for beverages (internal surface)		total	114	111	3	-	-	-	- mg/L	
eve Irfa		I	90	87	3	_	_	_		
r be l su		EU	6	6	-	_	_	_		
s fo rna	2011	K	9	9	-	-	-	_		
sels nte		total	105	102	3	-	-	-		
Ves (i		Ι	91	91		-	-	_		
-		EU	4	4	-	-	-	-		
	2012	K	17	17	-	-	-	-		
		total	112	112	-	-	-	-		
				< 0.01	0.01-0.05	0.05-0.10	0.10-0.20	>0.20		
		Ι	73	70	2	-	1	-		
_		EU	8	8	-	-	-	-		
(sm		K	12	11	-	1	-	-		
els (rin		total	93	89	2	1	1	-	mg/product	
	2011	I	83	76	4	2	1	-		
ess		EU	10	9	1	-	-	-		
Drinking vessels (rims)		K	9	9	-	-	-	-	Or-Suudi	
		total	102	94	5	2	1	-		
		I	90	84	5	1	-	-		
	2012	EU	-	-	-	-	-	-		
		K	18	18	-	-	-	-		
		total	108	102	5	1	_	_		

Table 2. Migration of cadmium from ceramic vessels

* - I –import (outside EU); EU - European Union; K - domestic; ** - without vessels for beverages

Table 3. Migration of lead from ceramic vessels

Product	Year	Origin*	No of samples	Distributions of samples					Unit
				below permissible level above permi					
				< 0.1	0.1-0.2	0.2-0.4	0.4-0.8	>0.8	
Flat dishes		Ι	45	38	6	1	-	-	
	2010	EU	6	5	1	-	-	-	
		K	17	16	-	1	-	-	
		total	68	59	7	2	-	-	
	2011	Ι	38	26	4	7	1	-	
		EU	3	3	-	-	-	-	mg/dm ²
	2011	К	14	12	1	1	-	-	
		total	55	41	5	8	1	-	
		Ι	53	44	5	2	-	2	
	2012	EU	2	2	-	-	-	-	
		K total	9 64	7 53	- 5	1 3	1	- 2	
		totai	04	<0.1	0.1-0.5	0.5-2.0	2.0-4.0	>4.0	
		т	C A						
	2010	I EU	64 13	34 13	25	2	1	2	
	2010	K	22	13	2	- 1	-	-	
*		total	99	66	27	3	1	2	
Deep dishes **			56	35	15	4		2	
dish		I EU	1	1	15	4	-	2	mg/L
eb e	2011	K	8	8		_	_	_	L L
De		total	65	44	15	4	-	2	
	2012	Ι	46	23	16	5	1	1	
		EU	6	6	-	-	-	-	
	2012	Κ	17	14	3	-	-	-	
		total	69	43	19	5	1	1	
				<0.1	0.1-0.5	0.5-2.0	2.0-4.0	>4.0	
lal	2010	Ι	91	87	4	-	-	-	
terr		EU	9	9	-	-	-	-	
(in		K total	14 114	13 109	1 5	-	-	-	mg/L
ges (-	-	-	
beverag		Ι	90	88	1	1	-	-	
bev sur	2011	EU K	6 9	6 9	-	-	-	-	
for		total	105	103	- 1	- 1	-	-	
Vessels for beverages (internal surface)		I	91	91	-	-	_	-	
Vest	2012	EU	4	4	-	-	-	-	
-	2012	Κ	17	16	1	-	-	-	
		total	112	111	1	-	-	-	
				< 0.1	0.1-0.5	0.5-1.0	1.0-2.0	>2.0	
		Ι	73	58	10	1	2	2	
Drinking vessels (rims)	2010	EU	8	8	-	-	-	-	
		K	12	11	1	-	-	-	
		total	93	77	11	1	2	2	
	2011	Ι	83	67	10	3	2	1	mg/product
		EU	10	10	-	-	-	-	ing/product
kin		K total	9 102	9 86	- 10	- 3	- 2	- 1	
)rin		I	90	77	9	2	1	1	
Ц		EU	-	-	-	-	-	-	
	2012	K	18	17	1	-	-	-	
		total	108	94	10	2	1	1	

* - I - import (outside EU); EU - European Union; K - domestic; ** - without vessels for beverages

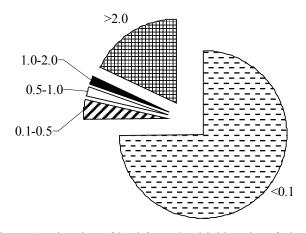


Figure 1. Migration of lead from the drinking rim of glass vessels, mg/product.

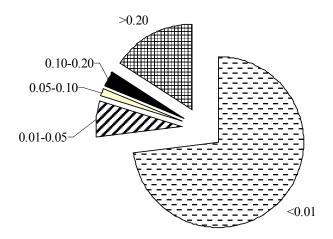


Figure 2. Migration of cadmium from the drinking rim area of glass vessels, mg/product.

Vessels for beverages made of crystal glass

Of the 129 samples tested (96% being manufactured in Poland or other EU countries) there was only one instance (0.447 mg/L) where the release of cadmium exceeded the permissible limit of 0.3 mg/L. Levels of cadmium in the other samples were below the detection limits. No exceeding of the lead release limit (4.0 mg/L) was observed. In 45% cases, the levels of detectable lead ranged between 0.5 mg/L to 2.0 mg/L, and results below detection limit were received for 18.6% samples.

During 2010-2012, there were 80 notifications made to the Rapid Alert System for Food and Feed (RASFF) concerning health threats due to high migrations of lead and cadmium from ceramics and glassware coming into contact with food [26]. A significant number of these originated from Poland and were mainly found in glassware with decorated rims. Most of the notified products were manufactured in China although some were from EU Member States. The highest migration levels observed were 163.8 mg/product for lead and 8.96 mg/product for cadmium [26].

Current regulations permit a lead migration of 2.0 mg from a deep plate of 0.5 L volume and 2.4 mg from

a flat plate with a 20 cm diameter. Intake of such levels of lead for an adult of 70 kg body b.w. results in the BMDL₀₁ (cardiovascular disorders) being exceeded by 2.5-fold for deep plates and over three-fold for flat plates. In the case of 20 kg b.w. children, exceeding of the BMDL₀₁ dose (neurotoxic effects) becomes 30-fold and 34-fold, respectively.

For cadmium, a permissible migration from a deep plate of 0.5 L volume is of 0.15 mg, and from flat plate of 20 cm diameter - 0.21 mg. Intake of cadmium for a 70 kg b.w. adult is equivalent to 86% and 120%, of the Tolerable Weekly Intake (TWI) respectively. In the case of a 20 kg b.w. child, the TWI levels would be exceeded 3-fold and 4.2-fold. When the PN-B-13210:1997 [16] limits for lead and cadmium (2 mg/product and 0.2 mg/product, respectively) are adopted as a reference point for their migration from the rims of vessels for beverages, the lead $BMDL_{01}$ levels are exceeded more than 2.5-fold for adults and about 30-fold in children. For cadmium, the intakes would be from equal to the TWI in adults to a 4-fold exceeding of the TWI in children. Such intakes would occur if the vessels for beverages were used once weekly. In the extreme case of daily use, the intakes would obviously be seven times more.

Studies on the migration kinetics of these metals from ceramics show that migration occurs over many consecutive extractions and that its decrease is not always linear [24, 32].

The following should however be accounted for:

- lead intakes from ceramics and glassware under real conditions of use will probably be lower than that resulting from migration into the food simulant;
- the real food contact time with plates or beverage glass or mug with the mouth is shorter than that during the tests;
- lead and cadmium release decreases upon repeated use of the dishware items so that the real intake of these elements by users will be lower than for the extreme cases presented.

When assessing exposures, the intake of lead and cadmium from other sources should also be considered, such as from the contamination of food, water and air. In adults, for cadmium this on average constitutes 70% of the TWI, whilst for lead it constitutes 35% of the BMDL₀₁ for cardiovascular disorders and about 80% of the BMDL₀₁ in nephrotoxicity. For children, the corresponding intakes are: for cadmium 150% TWI and for lead 200% BMDL₀₁ [1, 11, 29].

CONCLUSIONS

 In most of the ceramic and glassware samples, intended to come into contact with food that were placed on the Polish market lead or cadmium released did not exceed legally established limits.

- 2. Because the permissible levels of lead and cadmium release (set in EU legislation) cause human intakes of these metals either to be very close to, or exceeding reference doses, a review of these limits is necessary to ensure human health safety.
- 3. Both the results of these investigations, as well as RASFF notifications in relation to lead and/ or cadmium migration from rims of ceramic and glass products for beverages at levels posing health hazard, point to the urgent need of including such parameters into the applicable legislation.

Acknowledgements

The authors would like to thank the staff of the Sanitary-Epidemiological Stations for their assistance and cooperation throughout this research programme.

Conflict of interest

The authors declare no conflict of interest.

REFERENCES

- Cadmium dietary exposure in the European population. EFSA Journal 2012; 10(1): 2551.
- Commission Directive No 2005/31/EC amending Council Directive 84/500/EEC as regards a declaration of compliance and performance criteria of the analytical method for ceramic articles intended to come into contact with foodstuffs. OJ L EU L 110/36, 30.04.2005.
- Council Directive No 84/500/EEC of 15 October 1984 on the approximation of the laws of the member states relating to ceramic articles intended to come into contact with foodstuffs. Off J Eur Union L 277/12, 20.10.1984.
- Commission Regulation No 2023/2006 of 22 December 2006 on good manufacturing practice for materials and articles intended to come into contact with food. OJ L EU L 384/75, 29.12.2006.
- Demont M., Boutakhrit K., Fekete V., Bolle F., Van Loco J.: Migration of 18 trace elements from ceramic food contact material: Influence of pigment, pH, nature of acid and temperature. Food and Chemical Toxicology 2012; 50:734-743.
- EFSA Panel on Contaminants in the Food Chain (CON-TAM). Scientific Opinion on Lead in Food. EFSA Journal 2010; 8(4):1570.
- Evaluation of certain contaminants in food. Seventy-third report of the Joint FAO/WHO Expert Committee on Food Additives. WHO Technical Report Series. 2011;960.
- Gonzalez-Soto E., Gonzalez-Rodriquez V., Lopez-Suarez C., Castro-Romero J.M., Perez-Iglesias J., Fernandez-Solis J.M.: Migration of lead and cadmium from ceramic materials used in food preparation. Bull Environ Contam Toxicol 2000; 65:598-603.
- 9. Guadagnino E., Gambaro M., Gramiccioni L., Denaro M., Feliciani R., Baldini M., Stacchini P., Giovannangeli S., Carelli G., Castellino N., Vinci F.: Estimation of lead

intake from crystalware under conditions of consumer use. Food Addit Contam 2000; 17(3):205-218.

- Jorhem L., Fjeldal P., Sundstrom B., Svensson K.: Lead extracted from ceramics under household conditions. Livsmedels Verket, National Food Administration, Sweden, Raport 19, 2007.
- 11. Lead dietary exposure in the European population. EFSA Journal 2012;10(7):2831.
- 12. *Lehman R.L.*: Lead glazes for ceramic foodware. The International Lead Management Center, USA 2002.
- Lynch R., Elledge B., Peters Ch.: An assessment of lead leachability from lead-glazed ceramic cooking vessels. J Environ Health 2008;70(9):36-40.
- 14. *Omolaoye J.A., Uzairu A., Gimba C.E.:* Heavy metals assessment of some ceramic products imported into Nigeria from China. Arch Appl Sci Res 2010;2:120-125.
- Opoka M., Wojciechowska-Mazurek M., Starska K., Mania M. Biernat U.: Wykorzystanie techniki AAS w badaniach wyrobów ceramicznych przeznaczonych do kontaktu z żywnością – badania monitoringowe. Materiały XIV Konferencji 'Zastosowanie metod AAS, ICP--OES i ICP-MS w analizie środowiskowej', Warszawa, 2009;R-5:16.
- 16. Polish Standard PN-B-13210:1997. Glass and ceramic articles intended to contact with food. Permissible quantities of lead and cadmium release (in Polish).
- 17. PN-EN 1388-1:2000. Materials and articles in contact with foodstuffs Silicate surfaces Part 1: Determination of the release of lead and cadmium from ceramic ware.
- PN-EN 1388-2:2000. Materials and articles in contact with foodstuffs - Silicate surfaces - Part 2: Determination of the release of lead and cadmium from silicate surfaces other than ceramic ware.
- PN-EN ISO/IEC 17025:2005. General requirements for the competence of testing and calibration laboratories, as amended AC 2007 and Ap1 2007.
- 20. Regulation of the Polish Minister of Health of 15 January 2008 on the list of substances, the use of which is permitted in the process of manufacturing or processing materials and products from materials other than plastics, intended to contact with foodstuffs. Journal of Laws of 2008, No 11, item 113 (in Polish).
- 21. 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 2009; 980: 1-139.
- Schnaas L., Rothenberg S.J., Flores M-F., Martinez S., Hernandez C., Osorio E., Perroni E.: Blood Lead Secular Trend in a Cohort of Children in Mexico City (1987-2002). Environ Health Perspect 2004; 112: 1110-1115.
- Selden A.I., Bergström B-E., Gunnarsson L-G.: Lead exposure from tourist earthenware: A pilot survey: Int J Sci Eng 2008; 211: 587-590.
- 24. *Sheets R.W.*: Extraction of lead, cadmium and zinc from overglaze decorations on ceramic dinnerware by acidic and basic food substances. Sci Total Environ 1997; 197: 167-175.
- 25. Statement on tolerable weekly intake of cadmium. EFSA Journal 2011; 9(2): 1975
- 26. The Rapid Alert System for Food and Feed (RASFF). Annual report 2010-2012. Available from:

http://ec.europa.eu/food/safety/rasff/docs/rasff_annual_report_2010_en.pdf http://ec.europa.eu/food/safety/rasff/docs/rasff_annu-

al_report_2011_en.pdf http://ec.europa.eu/food/safety/rasff/docs/rasff_annual_report_2012_en.pdf

- Valadez-Vega C., Zuniga-Perez C., Quintanar-Gomez S., Morales-Gonzales J.A., Madrigal-Santillan E., Villagomez-Ibarra J.R. Sumaya-Martinez M.T., Garcia-Peredes J.D.: Lead, cadmium and cobalt (Pb, Cd and Co) leaching of glass-clay containers by pH effect of Food. Int J Mol Sci 2011; 12: 2336-2350.
- VKM. Norwegian Scientific Committee for Food Safety: Risk assessment of health hazards from lead and other heavy metals migrated from ceramic articles 2004; 04/403-10.
- 29. Wojciechowska-Mazurek M., Starska K., Brulińska-Ostrowska E., Plewa M., Biernat U., Karłowski K.: Monitoring of contamination of foodstuffs with elements noxious to human health. Part I. Wheat cereal products, vegetable products, confectionery and products for in-

fants and small children (2004 year). Rocz Panstw Zakl Hig 2008; 59(3):251-266 (in Polish).

- Wojciechowska-Mazurek M., Starska K., Mania M., Brulińska-Ostrowska E., Opoka M., Biernat U., Karlowski K.: Ceramic articles intended to come into contact with foodstuffs – safety assessment of lead and cadmium migration. Bromat. Chem. Toksykol. 2009; 42(3): 475-480 (in Polish).
- 31. Working paper of European Commission. Discussion of options and questionnaire to prepare the possible update and extension of the legislation on ceramic food contact materials – possibly including Glass and other Glazed materials. Version number 0.6.1, date 4.03.2011.
- Zawadzka T., Brulińska E.: Kinetics of lead and cadmium migration from porcelain dishes. Rocz Panstw Zakl Hig 1986; 37(3): 210-218 9 (in Polish).

Received: 28.07.2014 Accepted: 16.10.2014