

# Dietary exposure to cadmium, lead and nickel among students from south-east Poland

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## Abstract

The dietary intake of cadmium, lead and nickel was determined among students from three universities in the city of Lublin in south-east Poland to assess the levels of exposure to these contaminants, compared to PTWI and TDI values. The study was performed in 2006–2010 and involved 850 daily food rations of students. The technique of 24-hour dietary recall and diet duplicates was used. Cadmium, lead and nickel complexes with ammonium-pyrrolidindithiocarbamate were formed and extracted to the organic phase with 4-methylpentan-2-one – MIBK, in which their content was measured by flame atomic absorption spectrometry. The highest intake of the elements studied was observed in 2008. The data show that in none of the cases, the level of intake reached 70% of PTWI/TDI values, and thus the risk of developing diseases related to high exposure to these toxic metals absorbed from foodstuffs was low. The parameters of methods were checked during determinations by adding standard solutions to the samples before mineralization and by using two reference materials: Total diet ARC/CL HDP and Bovine muscle RM NIST 8414.

The dietary exposure to lead and cadmium has significantly decreased in recent years, whereas the exposures to nickel remains on a stable level.

## Key words

cadmium, lead, nickel, diets, students, health hazard

## INTRODUCTION

According to current knowledge, heavy metals such as cadmium and lead do not have any positive effects on the human organism. Moreover, the intake of the above elements, even during a short exposure, results in their bioaccumulation. Toxic effects of exposure to cadmium and lead can be observed once their concentrations in tissues are critical. Cadmium accumulates primarily in the kidneys and may induce kidney dysfunction, skeletal changes and reproductive disorders. In 1993, the International Agency for Research on Cancer (IARC) classified cadmium and cadmium compounds as human carcinogens (Group I) [1]. The most severe form of chronic cadmium poisoning caused by prolonged ingestion is Itai-Itai disease, for which the clinical symptoms include renal malfunction, and bone damage in the form of osteomalacia combined with osteoporosis [2]. Lead causes toxic effects in the haematopoietic and central nervous systems. It inhibits the synthesis of haemoglobin and shortens erythrocyte life expectancy, resulting in anaemia [3, 4]. The essentiality of nickel in humans has not been proved and its beneficial effects can only be implicated by extrapolation from animals. Nickel is reported to be the most frequent cause of contact allergy, a very common disorder affecting up to 30% of the population with a rising incidence, especially in females who have previously had skin piercing [5]. In 1990, the IARC classified all nickel compounds as carcinogenic to humans (Group I) [6]. Cadmium, lead and nickel can accumulate in tissues and have toxic effects after

long-term chronic exposure. Environmental concern has led to attempts to replace Cd via the replacement of NiCd batteries with lithium ion and hydride batteries. This can eventually lead to an efficient decrease in Cd consumption. The ban on tetra-alkyl Pb in petrol and the ban on Pb in paint also significantly decreased lead diffusion in the environment [7]. However, despite these legal actions, the levels of toxic metals in the environment remain high. Since the main source of cadmium, lead and nickel for the professionally unexposed population is food, including drinks, their intake with daily food rations (DFRs) should be as low as possible. The international agencies, FAO and WHO, have set the limits of heavy metals intake to reduce the incidence of toxic effects in the general population. The aim of the presented study was to assess the risk related to cadmium, lead and nickel exposure among students from the south-east region of Poland. Assessment was based on the intakes of the metals listed above from the DFRs in relation to maximum permissible intake limits.

## MATERIAL AND METHODS

**Preparation of analogue duplicates of diets.** The study was conducted in 2006–2010 and involved 850 randomly chosen students, males and females living in the south-eastern region of Poland – the city of Lublin and its surrounding province. All students were volunteers whose their lifestyles were characterized by moderate physical activity. The participants studied at three universities in the city: the Medical University, University of Life Sciences and the Catholic University. The study was carried out using the 24-hour dietary recall technique. The analogue duplicates of diets were prepared using the information concerning qualitative

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and quantitative parameters of diets provided by the students. All the products used to prepare food rations were from the retail market of the Lublin region. Since this is an agricultural region, over 90% of the most important products – cereals, milk and its products, meat and its products, eggs, vegetables and fruit – were of local origin. The daily rations consisted of portions of main meals as consumed by a particular individual, and all other foodstuffs and beverages consumed daily. For each group (depending on gender, university and year), 6 averaged duplicates of diets were prepared according to commonly accepted culinary techniques. The non-edible parts (bones, fish bones) were removed and the whole diet was homogenized (titanium blades) and placed in plastic containers (treated with 20% nitric (V) acid of Suprapur grade for 24 h, and then rinsed with deionised water). From each mean freshly-prepared daily diet, a sample four 50-gram portions were weighted into quartz crucibles and prepared for determination of heavy metals. In total, there were 264 samples for each gender in which cadmium, lead and nickel were determined. Statistically analysis was made using MS Excell 2007 and StatSoft Statistica 6.0 PL.

**Determination of total cadmium, lead and nickel contents in daily food rations samples.** Cadmium, lead and nickel contents were evaluated using flame atomic absorption spectrometry after sample mineralization by ashing at 450 °C in a muffle furnace. The ashing process was accelerated using 30% water solution of nitric (V) acid of Suprapur grade, which was then evaporated and the samples re-heated at 250 °C. The ash was dissolved in hydrochloric acid of Suprapur grade and diluted with deionised water (1+1 v/v). Complexes of all elements with ammonium pyrrolidindithiocarbamate (APDC) were formed, extracted to organic phase of 4-methylpentan-2-one – MIBK, and assayed directly in Pye Unicam SP 192 and a Thermo Elemental Solaar M5 spectrometers. Standard solutions of cadmium, lead and nickel with the initial concentration of 1 mg/cm<sup>3</sup>, Titrisol Standardlosung, acids Suprapur and MIBK for extraction analysis, were obtained from Merck (Darmstadt, Germany), and APDC from Fluka (Buchs, Switzerland). Each batch of determinations contained two reagent blanks prepared in the same way as the diet blanks. No contamination of the reagent blanks was observed.

## RESULTS

**Quality assurance of analytical parameters.** Parameters of the methods were checked during determinations by adding standard solutions to the samples before mineralization, and by using two reference materials: Total diet ARC/CL HDP and Bovine muscle RM NIST 8414. Contents of the elements are shown in Table 1. The results indicated the good agreement between the certified and the determined values. The average values of recovery were found to be: 95.1% for cadmium; 94.9% for lead and 96.5% for nickel. The relative standard deviations (n =10) were: 7.4% for cadmium, 9.3% for lead and 9.4% for nickel (Tab. 2). The cadmium, lead and nickel intake in the DFRs was additionally calculated using Food Software, designed in the National Food and Nutrition Institute, based on the Polish National Database published in 1992. Calculations were performed using the same questionnaires applied in the analytical section.

**Table 1.** Analytical results for cadmium, lead and nickel content in certified reference materials (n=10)

Certified reference materials	Cadmium content		Lead content		Nickel content	
	Certified value	Value found	Certified value	Value found	Certified value	Value found
Total diet ARC/CL HDP ng/g	21±3	22±4	43±8	46±7	271±38	268±35
Bovine muscle RM NIST 8414 mg/kg	0.013±0.011	0.015±0.004	0.38±0.24	0.36±0.04	0.05±0.04	0.05±0.03

**Table 2.** Analytical performance of target analytes

Analyte	Matrix	LOD (µg kg <sup>-1</sup> )	LOQ (µg kg <sup>-1</sup> )	Recovery range (%)	RSDr (%)	Measurement uncertainty N=10
Total Cadmium	Daily Diet	0.2	0.5	91.0–101.0	5.3	11.7%
Total Lead	Daily Diet	0.8	2.5	88.0–103.0	8.1	17.8%
Total Nickel	Daily Diet	0.8	2.5	90.0–106.0	7.5	16.4%

## DISCUSSION

**Weekly dietary intake of cadmium, lead and nickel.** Tables 3, 4, 5 and 6 present the weekly dietary intakes of cadmium and lead, as well as the daily intake of nickel among students of the south-eastern region of Poland in 2006–2010, determined analytically and assessed for the variety of risks associated with these toxic metals in relation to PTWIs/TDI, as determined by the FAO and WHO experts [8, 9, 10]. Statistically significant differences between the studied universities in particular years are shown in the Tables. Data obtained using calculations were intentionally omitted from the Tables and Discussion as they were over-estimated. In general, the average intake of cadmium, lead and nickel in the years studied was higher in males compared to females –  $p < 0.05$ . The higher energy in the total daily diets of the males may account for such findings. The analysis showed that the average cadmium intake varied from 76.4–241 µg/person/week in the female group, which corresponds to an environmental exposure of 18–57% of the PTWI (420 µg/person/week). In the male group, the average cadmium weekly intake varied from 93.7–330 µg/person/week, which constituted 19–67% of the PTWIs (7 µg/kg b.wt./week).

In females, the lowest intake of lead (104 µg/person/week) was observed in 2009, and the highest (337 µg/person/week) in 2008. In males, the weekly intake of lead ranged from 139–522 µg/person. The health hazard from the dietary lead intake, measured as the element content compared to the PTWI (25 µg/kg b.wt./week), ranged from 6.9–22%, and from 7.9–30% in females and males, respectively.

The DFR nickel intake among females varied from 101–152 µg/day, which constituted 34–51% of the TDI value (300 µg/day). In the male group, the daily intake of nickel was significantly higher and ranged from 139–204 µg/day, which corresponds with an exposure from 40–58% of the TDIs (350 µg/day for males).

**Cadmium.** The lowest intake of this element for both genders was observed in 2010 (76.4 and 93.7 µg/week in females and males at the Catholic University, respectively), whereas the highest was found in 2008 among students of the Medical

**Table 3.** Cadmium, lead and nickel intake with student's diets in 2006 and 2007

Year	Females		Males	
	2006	2007	2006	2007
Cd [ $\mu\text{g}/\text{week}$ ]	177 $\pm$ 34.4	173 $\pm$ 32.3	227 $\pm$ 29.9	215 $\pm$ 29.2
% PTWI	42	41	46	44
Pb [ $\mu\text{g}/\text{week}$ ]	254 $\pm$ 43.3 <sup>a</sup>	303 $\pm$ 60.3 <sup>b</sup>	423 $\pm$ 74.9	387 $\pm$ 80.2
% PTWI	17	20	24	22
Ni [ $\mu\text{g}/\text{day}$ ]	122 $\pm$ 43.4	145 $\pm$ 51.6	182 $\pm$ 66.9	160 $\pm$ 53.5
% TDI	41	48	52	46

a-b – differences significant at  $p < 0.05$ **Table 4.** Cadmium, lead and nickel intake with daily food rations among students from three Lublin Universities in 2008

Year	Females			Males		
	Medical University	University of Life Sciences	Catholic University	Medical University	University of Life Sciences	Catholic University
Cd [ $\mu\text{g}/\text{week}$ ]	241 $\pm$ 32.9	225 $\pm$ 43.3	209 $\pm$ 53.5	330 $\pm$ 44.4 <sup>a</sup>	286 $\pm$ 33.7	239 $\pm$ 45.7 <sup>b</sup>
% PTWI	57	54	50	67	58	49
Pb [ $\mu\text{g}/\text{week}$ ]	337 $\pm$ 39.8	290 $\pm$ 57.5	312 $\pm$ 67.5	484 $\pm$ 110 <sup>a</sup>	522 $\pm$ 129 <sup>a</sup>	379 $\pm$ 82.1 <sup>b</sup>
% PTWI	22	19	21	28	30	22
Ni [ $\mu\text{g}/\text{day}$ ]	152 $\pm$ 51.0	136 $\pm$ 41.2	137 $\pm$ 33.0	171 $\pm$ 51.0	193 $\pm$ 69.9	177 $\pm$ 53.1
% TDI	51	45	46	49	55	51

a-b – differences significant at  $p < 0.05$ **Table 5.** Cadmium, lead and nickel intake with daily food rations among students from three Lublin Universities in 2009

Year	Females			Males		
	Medical University	University of Life Sciences	Catholic University	Medical University	University of Life Sciences	Catholic University
Cd [ $\mu\text{g}/\text{week}$ ]	108 $\pm$ 27.0	111 $\pm$ 30.0	83.0 $\pm$ 20.2	169 $\pm$ 31.4	143 $\pm$ 30.1	132 $\pm$ 19.9
% PTWI	26	26	20	35	29	27
Pb [ $\mu\text{g}/\text{week}$ ]	263 $\pm$ 77.3 <sup>a</sup>	104 $\pm$ 30.3 <sup>b</sup>	134 $\pm$ 39.1 <sup>b</sup>	330 $\pm$ 85.1 <sup>c</sup>	211 $\pm$ 68.8 <sup>d</sup>	188 $\pm$ 33.8 <sup>d</sup>
% PTWI	18	6,9	8,9	19	12	11
Ni [ $\mu\text{g}/\text{day}$ ]	141 $\pm$ 49.8	102 $\pm$ 37.9	106 $\pm$ 44.6	166 $\pm$ 68.1	204 $\pm$ 59.7	153 $\pm$ 54.0
% TDI	47	34	35	47	58	44

a-b – differences significant at  $p < 0.05$   
c-d – differences significant at  $p < 0.05$ 

University (241 and 330  $\mu\text{g}/\text{week}$  in females and males, respectively). These data suggest that the cadmium intake is well correlated with the total energy of daily food rations, because the lowest was observed in 2010 and the highest in 2008 among Medical University students, both females and males. The results obtained in 2006 and 2007 were close to the countrywide data from the previous years 1990–2002 [11, 12, 13]. This confirms that the intake of cadmium in Poland has been maintained at a similar level throughout recent years. The results obtained in 2008, especially among students of the Medical and Life Sciences Universities, were considerably higher due to extremely high energy intakes, which are rarely observed in the general population. Yet even in this case, the cadmium intake translated to PTWI did not exceed 70% of that exposure indicator. It should be also emphasized that the

**Table 6.** Cadmium, lead and nickel intake with daily food rations among students from three Lublin Universities in 2010

Year	Females			Males		
	Medical University	University of Life Sciences	Catholic University	Medical University	University of Life Sciences	Catholic University
Cd [ $\mu\text{g}/\text{week}$ ]	83.9 $\pm$ 26.7	85.5 $\pm$ 23.1	76.4 $\pm$ 18.4	98.3 $\pm$ 30.4	102.1 $\pm$ 22.7	93.7 $\pm$ 28.1
% PTWI	20	20	18	20	21	19
Pb [ $\mu\text{g}/\text{week}$ ]	199 $\pm$ 65.1 <sup>a</sup>	124 $\pm$ 69.1 <sup>b</sup>	132 $\pm$ 52.2 <sup>b</sup>	176 $\pm$ 88.5	139 $\pm$ 68.8	139 $\pm$ 86.9
% PTWI	13	8.3	8.8	10	7.9	7.9
Ni [ $\mu\text{g}/\text{day}$ ]	111 $\pm$ 52.9	118 $\pm$ 41.5	101 $\pm$ 44.5	174 $\pm$ 79.5	158 $\pm$ 53.9	139 $\pm$ 36.0
% TDI	37	39	34	50	45	40

a-b – differences significant at  $p < 0.05$ 

significantly lower intake of cadmium in 2009 and 2010, in comparison to the previous years, may have been influenced not only by a significantly lower energy intake, but also by a much lower amount of this contaminant in the products used to preparation of diets duplicates. This could suggest a diminishing concentration of this element in the environment.

In comparison to other countries, the average weekly cadmium intakes presented in this study are higher than or close to those in Belgium (data from 2010) – 114  $\mu\text{g}/\text{week}$ , Denmark – 112  $\mu\text{g}/\text{week}$ , Finland – 66  $\mu\text{g}/\text{week}$ , Portugal – 116  $\mu\text{g}/\text{week}$  or the United Kingdom – 85  $\mu\text{g}/\text{week}$ ; similar to the Netherlands – 176  $\mu\text{g}/\text{week}$  or Italy – 141  $\mu\text{g}/\text{week}$  [14], but considerable lower than in Greece – 315–392  $\mu\text{g}/\text{week}$  [15] or Spain – 385  $\mu\text{g}/\text{week}$  [16].

**Lead.** The lowest intake of lead among females was observed in 2009 among women from the University of Life Sciences (only 104  $\mu\text{g}/\text{week}$ ) and in males in 2010 in the group of students from the University of Life Sciences and the Catholic University – in both investigated groups the average intake was 139  $\mu\text{g}/\text{week}$ . The highest intakes of lead in females was observed in 2008 among students from Medical University (337  $\mu\text{g}/\text{week}$ ) and in males in 2008 in the group from University of Life Sciences (522  $\mu\text{g}/\text{week}$ ). These findings confirm that the intake of toxic elements correlates well with the energy of the daily food rations, similar to the case of cadmium, as DFRs of both genders from Medical and Life Sciences Universities in 2008 were characterized by the highest energy intakes. The results presented in this study, especially for 2009 and 2010, are considerable lower compared to those reported in other studies [12, 13], and suggest a constant reduction in lead emission into the environment. Moreover, it should be noticed that even the highest intake translated to PTWI constitutes only 30% of that value, and the lowest less than 7%, which demonstrates a decreasing risk of lead exposure. A similar tendency is observed in the USA [17], Germany [18], India [19], China [20] and the UK [21]. Furthermore, in recent years, low intakes of lead, presented as a percentage of PTWI value, have been observed in other European countries – Belgium: 18%, Denmark 7%, Finland 2.5%, Greece 10%, Norway 8% and Sweden 2%. Extremely low exposure to lead in the Nordic countries and relatively high intakes of this element in Portugal – 931  $\mu\text{g}/\text{week}$  (which corresponds to 54% of PTWI) [14], are worth stressing. The data presented in this study, especially those observed



in 2006–2008, are similar to those observed in France – 397.6 µg/week: 24% of PTWI, Germany – 329 µg/week: 19% and Italy – 364 µg/week: 22% [14, 22].

**Nickel.** The intake of nickel was estimated per day to compare it with the TDI value.; Thus, the lowest intake was observed in 2010 among both females and males from the Catholic University (101 and 139 µg/day, respectively). As in the case of lead and cadmium, very high intakes were found in 2008 in females from the Medical University (152 µg), and in males from the University of Life Sciences (193 µg), which was associated with the highest energy intakes observed in those two groups. Although the highest intake of nickel among males was found in 2009 in the group of students from University of Life Sciences. The intakes presented do not pose any health hazard once compared to TDIs – they constituted no more than 51 and 58% of the safe level value in females and males, respectively. The data obtained in 2010 are even safer: 34–39% in females and 40–50% of the TDI in males. In comparison to other Polish studies conducted in previous years [23], the intake of nickel remains at a similar level, which is higher or near to that observed in other countries: Brazil – 89 µg/day [24], India – 108 µg/day [19], Sweden – 110 µg/day [25], UK – 120 µg/day [21], Spain – 144 µg/day [26] and the USA – 190 µg [27]. Moreover, there are studies reporting much more higher intakes of nickel: Italy – 216 µg/day [22], Sweden – 410 µg/day [28], Spain – 600 µg/day [29] and China – 799 µg/day [30].

## CONCLUSIONS

The dietary exposure to lead and cadmium has significantly decreased in recent years, whereas the exposure to nickel remain on a stable level. The results of the presented study indicate that there is no risk of developing any toxic effects resulting from high bio-accumulation of heavy metals, since exposure to the contaminants did not exceed 70% of the PTWI/TDI values in all groups examined throughout the study period. It was revealed that the exposure to heavy metals in foodstuffs was well correlated with the total energy value of the daily food rations. Moreover, the presented study revealed a considerable differences between the results of analyses and the calculations, the latter being substantially higher. A new Polish National Database pertaining to heavy metals in food should be introduced since the database currently in use has not been updated since the 1990s. However, the levels of toxic metals in foodstuffs are now significantly lower than those observed 20 years ago.

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