Original Research

Noxious Elements in Milk and Milk Products in Poland

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> Received: 23 April 2010 Accepted: 21 January 2011

Abstract

Results of investigations for content of lead, cadmium, mercury, and arsenic in milk and milk products sampled from trade and directly from manufacturers throughout Poland in 2006-07 with participation of sanitary-epidemiological stations were presented. Results were obtained from all 16 Voivodships. Laboratories used accredited validated analytical methods to satisfy performance criteria specified in EU regulations on official control of foodstuffs.

Average reported contents for milk and milk products, respectively, were: lead 0.008 and 0.017 mg/kg, cadmium 0.001 and 0.002 mg/kg, arsenic 0.005 and 0.009 mg/kg, and mercury 0.001 and 0.002 mg/kg. Intake of the above elements noxious to human health, based on investigation results and reported consumption of milk and milk products, does not pose a threat to human health.

Keywords: milk, milk products, lead, cadmium, mercury, arsenic, monitoring studies, risk assessment

Introduction

Laboratories of the Department of Food and Consumer Articles Research, the National Institute of Public Health – National Institute of Hygiene (NIH) and sanitary-epidemiological stations have for many years investigated contamination of foodstuffs with elements noxious to human health. Monitoring allows systematic assessment of contaminants in individual foodstuff groups, enables evaluation of population exposure to the toxicity of heavy metals consumed with food, permits undertaking preventive measures should threatening contamination levels be reported, and evaluates efficiency of actions taken to mitigate such threats.

In the 1980s and '90s such research was performed in line with FAO/WHO guidelines within the GEMS/Food

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system and, subsequently, within the FAO European Cooperative Research Network on Trace Elements, Natural Antioxidants and Contaminants [1, 2]. Also, whole day diets of various population groups were investigated [3].

The prevailing structure involves preparing annual plans for official control and monitoring safety parameters of foodstuffs by the NIH, to be implemented by laboratories of the sanitary-epidemiological stations and the NIH laboratory as the national reference laboratory.

Heavy metals, being basic industrial and environmental pollutants, pose a threat to human health even at trace levels in food, causing non-contagious diseases characterized by future consequences. Exposure of a fetus and infants to lead, cadmium, and methylmercury compounds causes irreversible changes in the central nervous system [4]. These elements tend to accumulate within the human organism.

Provisional tolerable intake of those elements from all sources specified by the Joint FAO/WHO Expert

Committee on Food Additives (JECFA) is subject to regular review based on risk assessment. Until recently, the Provisional Tolerable Weekly Intake (PTWI) of those elements was as follows: Pb 0.025 mg/kg body weight (mg/kg b.w.), Cd 0.007 mg/kg b.w., As 0.015 mg/kg b.w., and Hg 0.005 (MeHg 0.0016) mg/kg b.w. These threshold values were verified by JECFA in the first half of 2010. PTWI for lead was withdrawn, concluding that intake of this element at the level of then applicable PTWI of 0.025 mg/kg b.w. does not ensure health safety and causes at least a 3-point decrease in intelligence quotient in children and an increase in systolic blood pressure in adults [5]. The European Food Safety Authority (EFSA) specified the Benchmark Dose Lower Confidence Limits: for developmental neurotoxicity $BMDL_{01}$ blood lead level (B-Pb) 12 $\mu g/l$ corresponding to daily lead intake with food and negligible exposure from other sources of 0.50 µg/kg b.w., for increase in systolic blood pressure in adults BMDL₀₁ 36 μg/l, corresponding to daily intake of 1.50 µg/kg b.w., and for effects on kidney in adults $BMDL_{10}$ 15 $\mu g/l$, corresponding to daily intake of $0.63~\mu g/kg$ b.w. [6]. JECFA stated BMDL_{0.5} for inorganic arsenic causing a 0.5% increase in lung cancer incidence at $3.0 \mu g/kg b.w./day (2-7 \mu g/kg b.w./day) [7]$. The withdrawn PTWI value for inorganic arsenic remained within the range. The PTWI value for total mercury was replaced by PTWI for inorganic mercury: 0.004 mg/kg b.w., retaining the existing PTWI for methylmercury [7]. In the case of cadmium, taking into account the exceptionally long biological half-life of this element (10-30 years), it was considered appropriate to determine the provisional tolerable monthly intake (PTMI) [5]. Cadmium intake at the PTMI level specified by JECFA at 0.025 mg/kg b.w. is just slightly lower than the equivalent so far PTWI. Risk assessment made in June 2010 by JECFA is not in compliance with EFSA risk assessment justifying adoption as critical a much lower cadmium level in urea of 1 µg/g creatinine in 95% population after 50 years of exposure. An EFSA opinion issued on 30.01.2009 states a 2.8-times lower tolerable weekly intake (TWI) for cadmium of 2.5 µg/kg b.w. [8], taking into consideration also cadmium dietary exposure – biokinetics models [9, 10]. The European Commission moved to appoint a working group with participation of EFSA and JECFA experts to resolve these differences this

Content of those toxic elements in foodstuffs has been limited in Poland since 1971, and the setting of maximum levels for individual foodstuff groups, as well as their review, were based, among other things, on monitoring results and risk assessment.

Presently applicable noxious metals limits: lead and cadmium in various foodstuff groups, mercury in fish, seafood and food supplements, and tin in products in metal packaging, are regulated by Commission Regulation (EC) No. 1881/2006, as amended [11].

Approximately 8-9% of the Rapid Alert System for Food and Feed (RASFF) notifications over the last few years referred to hazardous levels of metals noxious to human health, primarily mercury, cadmium, and lead [12].

Milk and milk products are not frequently encountered in RASFF communications, but their consumption is high, and milk is the principal component in products for infants and small children, a population group particularly susceptible to toxic action of heavy metals.

Milk and milk products belong to foodstuff groups that are versatile as to their nutrition value, constituting a source of well assimilative, whole protein, calcium, potassium, magnesium, zinc, cobalt, and manganese, as well as vitamins of the A, B, D, and E groups. Milk products contain probiotic acidophilic bacteria. Calcium and vitamin D intake in Poland is low due to recent falling milk consumption. The Central Office of Statistics data shows that milk and milk product consumption has been decreasing gradually from 260 g daily (95 kg per annum) in 1999 to 194 g daily (70 kg per annum) in 2009 [13]. EFSA Concise European Food Consumption Database reports average daily milk and dairy-based product consumption in Poland at approximately 180 g for the general population [14]. Milk and milk product consumption in a number of European countries is higher than in Poland, 1.5 times on average. Average consumption in Europe is approximately 300 g daily in the UK 328 g, and in Finland 437 g [8].

Recommended high milk and milk products consumption should not lead to high intake of noxious substances. The maximum level of lead in liquid milk equals 0.020 mg/kg according to Commission Regulation (EC) No. 1881/2006, as amended, and no limits have been imposed in this regulation for other elements in milk [11].

According to Regulation (EC) No. 396/2005 of the European Parliament and of the Council, as amended, the maximum residual level of mercury compounds expressed as mercury equals 0.01 mg/kg (limit of analytical method determination) [15]. For dried, diluted, processed, and compound foodstuffs the specific concentration and dilution factors shall be stated. Community legislation does not provide for maximum cadmium and arsenic levels in milk. General food safety requirements set in Regulation (EC) No. 178/2002 of the European Parliament and of the Council have to be fulfilled [16].

Experimental Procedures

Milk and milk product samples were taken in 2006-07 from all voivodships of Poland by sanitary-epidemiological stations, mainly from trade, with some samples taken also from domestic producers in line with the sampling plan prepared by the NIH.

Samples were taken in adherence to principles set forth in Regulations of the Minister of Health implementing Commission Directive 2001/22/EC and, starting with 1.06.2007, in accordance with Commission Regulation (EC) 333/2007 of 28.03 2007 [17] and specified in the Methodology Publications of the National Institute of Hygiene [18].

A total of 483 milk and milk product samples were investigated in 2006-07, with 92% of the samples taken

from domestic production and 8% from foodstuffs produced in other EU member states.

227 samples were investigated in 2006, in that 75 of milk and 152 of liquid processed milk products (81 samples of fermented milk products – kefir, yogurt, buttermilk, curds, and deserts based on such products, and 71 samples of non-fermented milk products – cream, coffee cream, condensed milk, and milk deserts). Samples of domestic products dominated, with products from other EU member states comprising just 5% of the total.

A total of 256 processed milk product samples were investigated in 2007 (66 maturing cheeses, 71 cottage cheeses, 61 butter, 58 ice cream samples) with the majority of them made in Poland, and those from the EU contributing just 10% to the total. However, 1/3 of maturing cheese samples were made in other EU member states, primarily Germany and the Netherlands. Also, some 10% of all butter samples were taken from products imported from EU member states.

Content of metals noxious to human health was determined in the integrated voivodship sanitary-epidemiological station laboratories in all 16 voivodships.

Both those laboratories, as well as the NIH reference laboratory, use validated, accredited analytical methods satisfying criteria set forth in Regulation (EC) No. 882/2004 of the European Parliament and of the Council [19] and Commission Regulation (EC) No. 333/2007 [17]. Commission Regulation No. 333/2007 provides for the requirements for methods applied in monitoring investigations, including limit of detection, limit of quantification, precision, recovery, and specificity. The methods applied should allow determinations to be made with the detection limit equal to less than one-tenth of the maximum level set forth in Regulation No. 1881/2006 [11] (if the limit for Pb is less than 0.1 mg/kg – less than one-fifth of that value) and quantification limit equal to less than one-fifth of the maximum level set forth in Regulation 1881/2006 (if the limit for Pb is less than 0.1 mg/kg – not more than two-fifths of that value); e.g. the maximum level for liquid milk is 0.02 mg/kg and the required limits of detection and quantification are less than 0.004 mg/kg and 0.008 mg/kg, respectively.

Laboratories participating in monitoring studies are accredited according to EN ISO/IEC 17025 and apply internal quality control procedures. Quality control also involves the use of certified reference materials, e.g. CRM 8435 Whole milk powder (NIST) and CRM BCR 150 Spiked Skim Milk Powder (BCR).

The laboratories are involved in regular proficiency test rounds, with the sanitary-epidemiological stations laboratories participating in tests organized by the NIH – National Reference Laboratory [20]; the NIH Reference Laboratory itself participates in proficiency tests organized by the UK Food Science Laboratory (FAPAS), the Swedish National Food Administration (trace elements in food) and the Community Reference Laboratory, Joint Research Centre – Institute for Reference Materials and Measurements, Belgium.

Lead and cadmium were determined using flame atomic absorption spectrometry (FAAS) or graphite furnace

atomic absorption spectrometry (GFAAS); arsenic was determined using hydride generation atomic absorption spectrometry (HGAAS) and mercury was determined using the "cold vapor" method (CVAAS).

Results

Results of studies for lead, cadmium, arsenic, and mercury content in milk and milk products are presented in Figs. 1-4.

To enable comparison with data from other EU member states participating in the SCOOP (Scientific Cooperation on Questions Relating to Food) program, if the result was below detection limit, calculations were made taking one-half of the range (e.g. for a result of <0.01 mg/kg calculations were made taking 0.005 mg/kg) [21, 22].

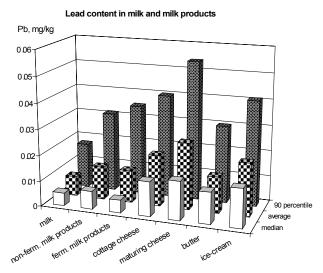


Fig. 1. Content of lead in milk and milk products tested in food monitoring studies in 2006-07, mg/kg.

Cadmium content in milk and milk products

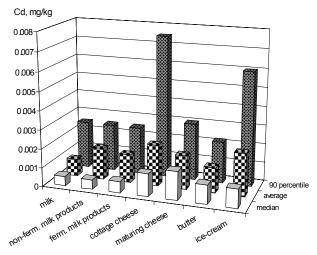


Fig. 2. Content of cadmium in milk and milk products tested in food monitoring studies in 2006-07, mg/kg.

Arsenic content in milk and milk products

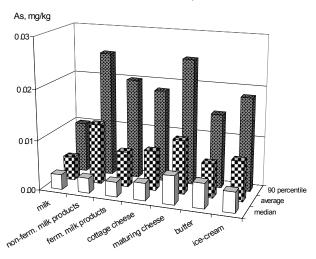


Fig. 3. Content of arsenic in milk and milk products tested in food monitoring studies in 2006-07, mg/kg.

Mercury content in milk and milk products

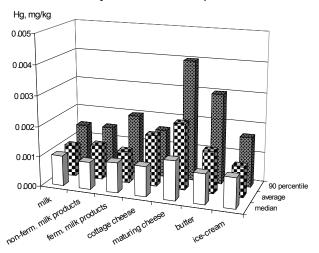


Fig. 4. Content of mercury in milk and milk products tested in food monitoring studies in 2006-07, mg/kg.

Content of investigated elements in the majority of milk and liquid processed milk product samples was low, with average content as follows: 0.011 mg/kg for lead, 0.001 mg/kg for cadmium, 0.008 mg/kg for arsenic, and 0.001 mg/kg for mercury.

Average lead content in Polish liquid milk is 0.008 mg/kg, much lower than the highest permissible level. 90% of the results remain below 0.017 mg/kg; two samples of milk did not meet the 0.020 mg Pb/kg threshold provided for in the legislation; the highest result was 0.05 mg/kg.

In the case of liquid processed milk products, average lead content was higher: 0.013 mg/kg in domestic products and 0.015 mg/kg in products imported from other EU member states. The 90th percentile values were 0.033 mg/kg and 0.031 mg/kg, and the highest values 0.072 mg/kg and 0.059 mg/kg, respectively.

Cadmium content in domestic milk and liquid milk products, i.e. average 0.001 mg/kg; 90th percentile 0.003 mg/kg (highest result in milk 0.007 mg/kg) do not raise concerns. The highest reported results were 0.020 mg/kg in processed products of domestic origin and 0.039 mg/kg in processed products of other EU member states.

Mercury and arsenic contamination in this product group is very low: arsenic – average 0.008 mg/kg, 90th percentile – 0.019 mg/kg; mercury – average 0.001 mg/kg, 90th percentile – 0.002 mg/kg; highest results – Hg 0.010 mg/kg, As 0.08 mg/kg.

No significant differences were reported in element content between fermented and non-fermented processed milk product groups.

Content of investigated elements in the great majority of cottage and maturing cheeses, butter, and ice cream samples was also low. Average lead content was 0.020 mg/kg, 90th percentile 0.043 mg/kg, and highest values reported in domestic salami cheese 0.220 mg/kg, and 0.147 mg/kg in rural cottage cheese and ice cream.

Cadmium, mercury, and arsenic contents in this group of milk products were only slightly higher than in investigated liquid products: Cd average 0.002 mg/kg, 90% results below 0.004; As average 0.009 mg/kg, 90% results below 0.020 mg/kg; Hg average 0.001 mg/kg, 90% results below 0.003 mg/kg.

Highest content of elements under study were reported in maturing cheeses, where averages and 90th percentile values were: Pb 0.026 mg/kg and 0.054 mg/kg; Cd 0.002 mg/kg and 0.003 mg/kg; As 0.011 mg/kg and 0.025 mg/kg; Hg 0.002 mg/kg and 0.004 mg/kg. The lowest contamination levels were reported for butter: average content of lead 0.014 mg/kg and cadmium 0.001 mg/kg.

Discussion of Results

Contamination of trade samples of milk is usually comparable with results obtained by the state veterinary inspection for raw milk with lead and arsenic content in trade samples of milk being slightly higher [23, 24]. The average lead content reported in pasteurized milk in trade as reported in this paper (0.008 mg/kg) was lower than the average value reported for raw milk in the 1990s (0.012 mg/kg) [25], but was twice higher than that reported for raw milk in 2002 and 2006 (0.004 mg/kg) [23, 24]. The average arsenic content in milk available in trade (0.005 mg/kg) was slightly lower than for raw milk in 2006 (0.006 mg/kg) [24], but higher than reported in the investigations of 1996 and 2002 [23, 25]. Cadmium and mercury contents in milk were similar to those reported in investigations conducted by veterinary inspection [23-25]. In the case of milk products such as cottage cheese and mature ng cheese, results for lead and arsenic content were twice higher, for cadmium slightly higher, and for mercury three times lower than the results reported in the 1990s by veterinary inspection [25]. Some regions of Poland reported higher milk contamination especially with cadmium. Król et al. reported average cadmium content of 0.006 mg/kg in raw milk from the

Lublin region, with contamination twice and three times lower reported for Beskid Średni and Bieszczady areas, respectively [26], whereas Radzymińska et al. reported higher contamination with both cadmium and lead, which attained levels of 0.002 mg/kg (Cd) and 0.012 mg/kg (Pb) for milk and milk products (cream, cottage cheese and butter) originating from Central Poland [27]. Slightly higher contents of those metals were observed in milk products originating during the summer season [28]. Lower milk contamination was reported in the agricultural Podlasie region, attaining an average lead level of 0.003 (range 0.001-0.013) mg/kg and cadmium 0.0004 (range 0.0001-0.0012) mg/kg [29]. Milk from industrial regions of Poland contains much higher levels of arsenic (average 0.035 mg/kg in Upper Silesia and 0.029 mg/kg in Lower Silesia) [30] and slightly higher mercury levels with average of 0.0017 mg/kg in Lower Silesia) [31]. A significantly lower mercury level was reported for the Wrocław region with an average of 0.00025 mg/kg [32].

Comparisons of results obtained during Polish monitoring studies for this product group with results obtained in other countries is difficult due to differences in analytical methods used, detection limits, and non-uniform treatment of results below the detection limit (which constituted even up to 100% of results in certain countries).

The majority of countries compared reported lower lead contents in the investigated group of milk products, whereas the average content in EU member states reported by EFSA in 2010 for milk is similar to the value given here for Poland at 0.008 mg/kg, 0.025 mg/kg for milk products, and 0.031 mg/kg for cheeses, i.e. higher as compared with domestic data [6]. Investigations undertaken by European countries in 2003 within the Scientific Co-operation on Questions Relating to Food (SCOOP) program reported lead content in liquid milk at levels of from 0.001 mg/kg (United Kingdom) through 0.002 mg/kg in France and Scandinavia, 0.004 mg/kg in Belgium, 0.007 mg/kg in Italy, and up to 0.012 mg/kg in Greece [22]. The average lead content in cheese ranged from 0.002 mg/kg (Norway) to 0.058 mg/kg (Denmark). UK monitoring studies in 2006 reported lead content in milk below 0.001 mg/kg, and 0.003 mg/kg, and more, in milk products [33, 34], monitoring in France in 2000 reported values from 0.003 mg/kg in milk, through 0.004 and 0.007 mg/kg in liquid products and butter, respectively, and up to 0.016 mg/kg in cheese [35]. Spanish data: 0.01 mg/kg in milk and 0.027 mg/kg in products [36] and German data reported in 1995-2002: average lead content in milk at 0.0108 mg/kg, and in cheese at 0.0413 mg/kg [37], are higher than reported for products from the domestic market investigated in this research.

There are also many works limited solely to milk and milk products. Lead content in milk and cheese in various regions of Romania varied from below detection limit to 0.051 mg/kg and 0.080 mg/kg, respectively [38]; results for raw milk in France gave 0.009-0.126 mg/kg, Comte cheese 0.002-0.925 mg/kg [39] and milk 0.005 mg/kg [40]; results in Italy were from below 0.0001 to 0.0099 mg/kg [41]. Very low lead contamination of raw milk was reported in Lithuania: average 0.00047 (range 0.00017-0.001) mg/kg

and 0.00054 (range 0.00006-0.00176) mg/kg, in the winter and summer seasons, respectively [42], and for milk and milk products in Finland: milk 0.0017 mg/kg, cheese 0.017 mg/kg [43]. Lithuania canned condensed milk had lead content of 0.06-0,47 mg/kg [44]. High lead contamination levels of yogurt, curds and cheese at 0.055-0.065 mg/kg were reported from the Slovakian market [45].

Monitoring studies in USA and New Zealand reported lead contamination at lower levels as compared with Poland. Most recent FDA data give levels of 0.001 mg/kg in milk and liquid products, and 0.001-0.002 mg/kg in cheese, except for single cottage cheese products (0.030 mg/kg) [46, 47]. New Zealand reports average lead levels in liquid milk at 0.0002 mg/kg, 0.0003 mg/kg in yogurts and 0.0050 mg/kg in butter and cheese [48]. Contamination levels lower than in Poland were reported also for Saudi Arabia (0.0035 mg/kg in fresh milk) [49], and in Lebanon – 0.003 mg/kg in milk and 0.018 mg/kg in cheese [50]. Much higher lead contamination of milk has been reported in publications from China and Japan, 0.034 mg/kg and 0.013 mg/kg, respectively [51], in industrial regions of India – up to 0.85 mg/l [52], and in Nigeria 0.531 mg/l [53].

Cadmium content reported by the majority of European countries is, in turn, higher than in Poland, with the average for milk and liquid milk products of 0.0030±0.0043 mg/kg and 0.0065±0.0084 mg/kg for cheese, as published by EFSA [8]. SCOOP data give average cadmium content in milk at from 0.0002 mg/kg in the UK, 0.0003 mg/kg in France and 0.0004 mg/kg in Belgium, and up to 0.001 in Scandinavia, below 0.004 mg/kg in Italy, and up to 0.034 mg/kg for Italian cheese [22]. The UK Food Standards Agency report of 2009 states contents of cadmium in milk and milk products at below 0.001 mg/kg and 0.003 mg/kg, respectively [34]; French monitoring data give 0.0004 mg/kg [35], Spanish data 0.002 mg/kg in milk and 0.006 in milk products [36], and German data for cheese report levels of 0.0040 mg/kg [37] - considerably higher than data reported in this work for products on the domestic market. In turn, data from Greece and Finland are comparable, i.e. milk 0.0006 mg/kg, cheese 0.002-0.003 mg/kg [54] and 0.001 mg/kg [43], respectively. Rubio reports very low cadmium content of milk in the Canary Islands - from 0.000015 mg/kg in milk, and up to 0.0006 mg/kg in cheese [55]. Cadmium content in various regions of Romania in milk and cheese varies from below the detection limit to 0.005 mg/kg and 0.004 mg/kg, respectively [38]. Cd content in raw milk in France is 0.0003-0.001 mg/kg, in Comte cheese 0.0007-0.011 mg/kg [39], and 0.001 mg/kg in milk [40], with data for Italy of less than 0.00001 mg/kg up to 0.023 mg/kg in milk [41]. Average cadmium content in raw cow milk received from Lithuanian producers was at 0.00018 (range 0.00011-0.00023) mg/kg and 0.00037 (0.00025-0.00049) mg/kg in the summer and winter season, respectively [42]. Cadmium contamination of cheese, yogurt, and curds on the Slovakian market is in the range of 0.002-0.004 mg/kg [45]. Cadmium levels reported in the USA are comparable or slightly lower as compared with those reported domestically [46, 47], and are lower in New Zealand (milk 0.0002 mg/kg, butter and cheese 0.001

mg/kg) [48]. Reported cadmium contamination in Saudi Arabia is higher as compared with the domestic market (0.0047 mg/kg in fresh milk) [49], and similarly in Lebanon at 0.003 mg/kg in milk and 0.0014 mg/kg in cheese [50]. Higher cadmium contamination of milk has been reported in publications from China and Japan at 0.004 mg/kg, and 0.002 mg/kg, respectively [51], high in industrial regions of India – up to 0.23 mg/l [52], and in Nigeria 0.257 mg/l [53].

Arsenic content reported by the majority of European countries is comparable to that reported in Poland. Average arsenic contents in EU member states as published by EFSA of 2010 reach 0.0065 mg/kg for milk and milk-derived beverages, 0.013 mg/kg for milk products and 0.013 mg/kg for cheese, i.e. are higher than those reported for Poland [56]. SCOOP (2003) data give average arsenic content in milk at from 0.0004 mg/kg in the UK, below 0.005 mg/kg in Finland, up to 0.007 mg/kg in Denmark; with the average arsenic content in cheese being from 0.002 mg/kg (UK) through 0.004 mg/kg (Germany) to 0.021 mg/kg (Denmark) [22]. The UK Food Standards Agency report of 2009 states 0.001 mg/kg and 0.003 mg/kg arsenic content in milk and milk products, respectively [34].

French data of 0.003 mg/kg arsenic in milk, 0.003 mg/kg in milk products and cheese, and 0.051 mg/kg in butter – are lower [35], German data of 1995-2002: average arsenic content in cheese at 0.010 mg/kg – are similar [37], and Spanish data: 0.005 mg/kg in milk and 0.020 mg/kg in milk products [36] – are higher than data reported in this paper for the domestic market. Arsenic content in milk and cheese in various regions of Romania varied from below 0.003 mg/kg to 0.010 mg/kg and 0.004 mg/kg, respectively [38]. In Italy arsenic content in raw milk was on average 0.038 mg/kg (range from below 0.00015 mg/kg to 0.684 mg/kg) [41].

Studies undertaken by European countries in 2003 within the SCOOP program reported mercury content in liquid milk at from 0.0004 mg/kg (UK) through 0.00053 mg/kg in Belgium, to 0.0010 mg/kg in Denmark, whereas average content of mercury in cheese ranged from 0.0009 mg/kg w Denmark, through 0.002 mg/kg (UK) up to 0.003 -0.004 mg/kg in Germany [22]. The UK Food Standards Agency report of 2009 states content of mercury in milk and milk products at below 0.0005 mg/kg and below 0.0002 mg/kg, respectively [34]. French data give mercury content of 0.003 mg/kg in milk, milk products, cheese, and butter [35]. German data of 1995-2002 give average mercury content in cheese at 0.006 mg/kg [37], Spanish data give mercury content in milk at 0.002 mg/kg, in milk products at 0.008 mg/kg, and are all higher than values reported in this paper for the Polish market for the same products [36]. Spanish data for the Canary Islands gives mercury content lower than reported here for the Polish market, i.e. 0.00025 mg/kg in milk, 0.00037 mg/kg in cheese, 0.00026 mg/kg in yogurt, and 0.0003 mg/kg in milk products [57]. Romanian data are similar and range from below the detection limit up to 0.001 mg/kg in milk [38].

Monitoring studies performed in USA and New Zealand gave arsenic and mercury contents lower than those reported in Poland. FDA data of 2007 give contents

of those elements in milk, yogurt and cheese below the detection limit [46, 47]. New Zealand reports average arsenic content in liquid milk at below 0.001 mg/kg, in yogurt and cream below 0.002 mg/kg, for cheese and butter below 0.010 mg/kg, and of mercury in liquid milk at below 0.001 mg/kg, for cheese, cream, and butter at 0.001 mg/kg and for yogurt below 0.002 mg/kg [48]. Milk contamination with arsenic and mercury lower than reported for the domestic market have been stated in milk from China and Japan, at 0.004 mg/kg and 0.002 mg/kg, and below 0.00006 mg/kg and 0.000015 mg/kg, respectively [51].

Taking into consideration average and 90th percentile content of metals in foodstuff groups discussed in this paper, combined with Central Office of Statistics data on consumption of such products in Poland in 2009 [13], an assessment was made of metals intake with milk and milk products by a 60 kg adult. The data below present average intake (with 90th percentile in parentheses), and the square bracket amounts present cadmium intake with consideration of the lower TWI value stated by EFSA [8]:

Milk: Pb -0.936 (1.989) in μ g/person/day; Cd -0.117 (0.351) in μ g/person/day; As -0.585 (1.170) in μ g/person/day; Hg -0.117 (0.176) in μ g/person/day.

Liquid, fermented and non-fermented milk products: Pb - 0.468 (1.188) in μ g/person/day; Cd - 0.036 (0.108) in μ g/person/day; As - 0.324 (0.792) in μ g/person/day; Hg - 0.040 (0.068) in μ g/person/day.

Cheese: Pb - 0.713 (1.488) in $\mu g/person/day$; Cd - 0.062 (0.155) in $\mu g/person/day$; As - 0.310 (0.682) in $\mu g/person/day$; Hg - 0.047 (0.093) in $\mu g/person/day$.

Butter: Pb -0.140 (0.310) in μ g/person/day; Cd -0.10 (0.20) in μ g/person/day; As -0.070 (0.170) in μ g/person/day; Hg -0.010 (0.030) in μ g/person/day.

Total daily intake of the investigated elements with milk and milk products is as follows: Pb -2.257 (4.975) μ g/person/day; Cd -0.225 (0.634) μ g/person/day; As -1.289 (2.814) μ g/person/day; and Hg -0.214 (0.367) μ g/person/day.

Total dietary intake of lead with milk and milk products is of the order of 1.1 (2.3)% of the newly applicable PTWI and 6.1 (13.4)% BMDL₁₀ (renal effects), of which more than 40% is contributed by milk.

Total dietary intake of cadmium with milk and milk products is of the order of 0.4 (1.1)% of the newly applicable PTWI, 0.4 (1.3)% PTMI and 1.1 (3.0) TWI EFSA, of which more than one-half is contributed by milk.

Total dietary intake of arsenic with milk and milk products is of the order of 1.0 (2.2)% of the newly applicable PTWI and 0.7 (1.6)% BMDL $_{0.5}$ (lung cancer), of which about 45% is contributed by milk.

Total dietary intake of mercury with milk and milk products is of the order of 0.5 (0.9)% of the newly applicable PTWI for total mercury, 0.6 (1.1)% PTWI for inorganic mercury and 1.6 (2.7) PTWI for methylmercury, of which more than 55% is contributed by milk.

Total dietary intake of the investigated elements with milk and milk products is for Pb -6.1%, Cd -1.4%, As -6.2%, Hg -4.5% of total intake of those elements in Poland [3, 58].

Contribution of this product group to total lead intake with diet in the UK and France is quite similar, and equal to 6 and 4.7% of total Pb intake, respectively. The share of milk and milk products in cadmium and mercury intake in those countries is higher in those countries as compared with Poland, i.e. Cd - 4% and 2.4 %; Hg - 9% and 6%, respectively, and in case of arsenic is lower: <2% and 1.3%, respectively [34, 35]. Similar data have been published by Llobet for Spain (Cd - 6%; Hg - 9%) [36]. In turn, Rubio reports a lower share of milk and its products in total mercury and cadmium intake, at 2% and 0.3%, respectively [55, 57]. The SCOOP program reported average share of milk and milk products in total daily diet intake of these elements in the European Union at: Pb - 3.7%, Cd - 3.5%, As -1%, and Hg -4%, i.e. this group contributes on average more cadmium, and mercury, and less lead and arsenic as compared to Poland [22]. Recently formulated reports of EFSA point to similar shares of milk and milk products in overall lead intake in EU counties as compared with Poland: average 7% [6], higher in overall cadmium intake: average 5.7% [8], lower in total arsenic intake: average 2.1% [56].

Metals intake with foodstuffs expressed as % PTWI (PTMI, BMDL) is much higher in the case of low bodyweight children.

EFSA data collected from most of the European countries show that high consumption of milk and milk products by children aged 0.5 to 6 years cause those products to comprise approximately 15% of total cadmium intake with food, whereas the same factor for adults is 5.7% on average [8].

Taking as reference the previously applied PTWI values, intake of metals noxious to human health with milk and milk products group assuming contamination levels at the 90th percentile would comprise from 0.9% (for total mercury) to 2.3% (for lead) of intake tolerated by organism, so it should not pose any threat to human health.

When the newest toxicological assessments for those elements established by JECFA and EFSA are taken into account, lead intake comprising more than 13% BMDL₁₀, the threshold dose causing nephrotoxic action in 10% adults, cannot be disregarded. Such products could pose a much higher neurotoxic action threat to children. Assuming child bodyweight of 20 kg, lead intake with milk and milk products contaminated with lead at the 90th percentile level could constitute 50% of the benchmark dose lower confidence limit for neurotoxic action. Even though environmental pollution with lead has decreased significantly over the last few years, it is still required to undertake further actions to lower milk contamination with this element.

Conclusions

- 1. Content of metals noxious to health in milk and milk products is lower than legal limits.
- The majority of countries reported in milk and milk products lower lead, but higher cadmium contents than in Poland; mercury and arsenic levels are comparable.

3. Intake of cadmium, mercury, and arsenic with milk and milk products is low in Poland and does not pose a health hazard. Lead intake comprising more than 13% BMDL₁₀, the threshold dose causing nephrotoxic action in 10% of adults, could pose a potential health hazard. Those products could pose a much greater health hazard for infants and children. Even though environmental pollution with lead has decreased significantly over the last few years, it is still required to undertake further actions to lower milk contamination with this element.

Acknowledgements

Department of Food and Consumer Articles Research, National Institute of Public Health – National Institute of Hygiene acknowledges the cooperation of sanitary-epidemiological stations in performance of the research program.

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