

Urszula PANKIEWICZ<sup>1</sup>

**MONITORING OF TOTAL MERCURY LEVEL  
IN SELECTED DAIRY PRODUCTS  
FROM THE SOUTH-EAST REGIONS OF POLAND**

**MONITORING POZIOMU RĘCI CAŁKOWITEJ  
W WYBRANYCH PRODUKTACH MLECZARSKICH  
Z REGIONÓW POŁUDNIOWO-WSCHODNIEJ POLSKI**

**Abstract:** Mercury content was examined in forty-eight milk products produced by *regional milk cooperatives* (OSM) located in south and south-east Poland. In the market range of products, the mercury content was lower than the permissible level of 10 µg/kg. The lowest mercury concentration, 0.01 µg/kg of product, was determined in kefir and the highest one – in milk and it was equal 0.79 µg/kg of product. The examined products are not dangerous for the consumers because the mercury content do not exceed a boundary value recommended by FAO/WHO. They also do not point at any pollution of natural or agricultural areas by this toxic element.

**Keywords:** mercury content, milk products, non-flame atomic spectrometry absorption technique

One percent of the global emission of mercury, that is about 20 Mg (ton)/year, falls on Poland. It put our country on the second place in the EU ranking [1]. Growing environmental pollution caused consumers' fears of consumption of products containing heavy metals. These metals get inside the human body mainly by inhalation and consumption and then cumulate. Usually their toxic action do not appear at once, but after many years or even in the next generations. Heavy metals present in food even in trace amounts are very dangerous for the human health, because they cause non-infectious diseases which are characterized by the far effects of action. Polish legal act, based on EU directives, permits the following mercury contents in foodstuff: 0.01 mg/kg in milk; 0.01 mg/kg in fruits, vegetables, leguminous vegetables, potatoes; 0.02 mg/kg in oil seeds, tea, hop; and 0.01 mg/kg in other foodstuffs of plant origin.

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<sup>1</sup> Department of Analysis and Evaluation of Food Quality, Faculty of Food Science and Biotechnology, University of Life Sciences in Lublin, ul. Skromna 8; 20-704 Lublin, Poland, phone: +48 81 462 33 33, fax: +48 81 462 33 76, email: urszula.pankiewicz@up.lublin.pl

Mercury occurs in several chemical forms including elemental mercury, as well as its inorganic and organic compounds [2], which differ in their solubility, reactivity and toxicity [3]. Global mercury circulation involves both natural and anthropogenic sources [4]. The main source of mercury is natural emission originating from volcanic eruptions and evaporation from the ocean and land surfaces. Mercury poisoning caused by mercury vapour and inorganic compounds of this element are mainly connected with the development of industry, while methylmercury (MeHg) is a serious environmental pollutant [5, 6]. It is reported that limited amounts of inorganic and organic mercury can be passed from feed to cow's milk [7, 8] and cumulate in dairy products. Therefore, monitoring of mercury presence in such products is so important [9].

The aim of this work was analysis of mercury content in milk and dairy products originated from south and south-eastern Poland.

## Materials and methods

Milk and a range of dairy products originated from south and south-east regions of Poland were purchased from the local markets. The main criterion was the selection of producers from the regions differing in their degree of industrialization and, consequently, pollution of the environment. Mercury content was analyzed in the products such as: milk, buttermilk, kefir, yogurt, cream, cream cheese, cottage cheese, butter and milk powder in order to assess if degree of concentration of a raw material and food processing have an influence on its level in dairy products.

**Measurement of water content.** Water content of lyophilizates and products was determined with the use of a moisture analyzer (Radwag WPS50SW) after drying samples at 100 °C. Analysis was carried out in triplicate.

**Lyophilization.** Samples of dairy products were lyophilized in Labconco freeze dryer (Model 64132, Kansas City, MO, USA). Obtained lyophilizates were stored in an exsiccator and used for the further analyses.

**Determination of total mercury content.** Mercury content in such prepared lyophilized samples was specified using non-flame atomic spectrometry absorption technique (mercury analyzer AMA 254, Altec, Czech Republic). During the analysis in the AMA 254, samples were pre-dried in the internal oven of the analyzer and burned in oxygen (99.999 % of purity). The decomposition products were further carried to the amalgamator for selective mercury trap. After temperature stabilization (120 °C) in the amalgamator the content of trapped mercury was measured. Thus mercury was released from the amalgamator by a short heat-up and then the mercury cloud was transferred by O<sub>2</sub> carrier gas to a double measuring cuvette. Then the same quantity of mercury was measured twice using different sensitivities, resulting in a dynamic range of 0.05–600 ng Hg in single measurement. The detection limit was 10<sup>-5</sup> µg/g. The original factory calibration was still valid for the calibration of the instrument. The values were controlled regularly by calibration standard mercury solutions – NIST- traceable Hg standard solution (Accu Trace Single Element Standard; AccuStandard Inc., New Haven, Ct, USA). Each sample was analyzed in triplicate and the data are an average of

these replications. Additionally, *standard deviation* (SD) for each determination was calculated. Mercury content in lyophilizate was converted to its concentration in the base product.

## Results and discussion

Mean values of mercury content [ $\mu\text{g}/\text{kg}$ ] determined in lyophilizates and base products are presented in Table 1. Mercury levels in all the examined products were in the order of ten thousandth parts of ppm and oscillated from about 0.01  $\mu\text{g}/\text{kg}$  in natural kefir (7D, 6C), cream (5D), buttermilk (5C) and strawberry flavoured yogurt “Serduszeko” (3D) to over 0.45  $\mu\text{g}/\text{kg}$  in butter from all the producers – the highest value determined for butter 10A was equal 0.79  $\mu\text{g}/\text{kg}$ .

Variety of mercury content was observed among the products originated from the same dairy or region of Poland:

- OSM (A) – from 0.00002 ppm in kefir (2A) to 0.00030 ppm in milk powder (8A) and 0.00079 ppm in butter extra (10A);
- OSM (B) – from 0.00003 ppm in milk (1B) to 0.00010 ppm in half fat cottage cheese (8B);
- OSM (C) – from 0.00001 ppm in kefir (6C) and buttermilk (5C) to 0.00014 ppm in vanilla flavoured cream cheese (2C) and 0.00058 ppm in butter extra (10C);
- OSM (D) – from 0.00001 ppm in natural kefir (7D), cream (5D) and strawberry flavoured yogurt (3D) to 0.00011 ppm in vanilla flavoured cream cheese (8D);
- OSM (E) – from 0.00004 ppm in kefir (5E) to 0.00016 ppm in cream cheese with spring onion (4E);
- OSM (F) – from 0.00002 ppm in kefir (2F) to 0.00008 ppm in granular cottage cheese (3F) and 0.00047 ppm in butter extra (5F).

To summarize, if a certain dairy produced kefir, it contained the lowest mercury level from the whole range of products. The highest mercury content was determined in butter in the case of all the producers.

Some differences in mercury content were observed in the particular range of dairy products (Table 1). Similar products originated from the different regions of Poland revealed diversity in mercury content (Table 1), for instance milk contained from *ca* 0.03  $\mu\text{g Hg}/\text{kg}$  (OSM B from Silesia) to 0.06  $\mu\text{g Hg}/\text{kg}$  (OSMs A and F from Lublin and Podkarpackie provinces).

The lowest amounts of mercury in the whole range of dairy products were determined for kefirs – from 0.01  $\mu\text{g Hg}/\text{kg}$  (OSMs D and C) to 0.04  $\mu\text{g Hg}/\text{kg}$  (OSM E). For kefirs produced by OSMs A and F, mercury content was about 0.02  $\mu\text{g}/\text{kg}$  (Table 1).

Similar mercury concentrations, in the range of 0.01–0.09  $\mu\text{g}/\text{kg}$ , were obtained for buttermilks, yogurts and creams. Among buttermilks, that produced by OSM C contained the highest amount of mercury and the lowest value was obtained for buttermilk produced by OSM A.

Strawberry flavoured yogurt produced by OSM D contained the lowest amount of Hg in the examined range of products, whereas the highest mercury content was obtained

for natural yogurt of OSM C. Addition of flavour supplements or fruits to yogurt did not cause any increase of mercury content.

Table 1

Mercury content in dairy products and their lyophilizates.  
A, B, C, D, E, F – codes of the particular producers

No.	Code	Product	Hg content in lyophilizate (mean $\pm$ SD) [ $\mu\text{g}/\text{kg}$ ]	Hg content in product (mean $\pm$ SD) [ $\mu\text{g}/\text{kg}$ ]
1	1A	Milk	0.53 $\pm$ 0.07	0.06 $\pm$ 0.01
2	2A	Kefir	0.11 $\pm$ 0.05	0.02 $\pm$ 0.01
3	3A	Natural yogurt	0.17 $\pm$ 0.02	0.03 $\pm$ 0.00
4	4A	Natural yogurt "JOGUŚ"	0.32 $\pm$ 0.07	0.05 $\pm$ 0.01
5	5A	Cream	0.23 $\pm$ 0.09	0.06 $\pm$ 0.02
6	6A	Cream cheese (vanilla taste)	0.37 $\pm$ 0.01	0.12 $\pm$ 0.00
7	7A	Half fat cottage cheese	0.77 $\pm$ 0.02	0.23 $\pm$ 0.01
8	8A	Whole milk powder	0.31 $\pm$ 0.02	0.30 $\pm$ 0.01
9	9A	Buttermilk	0.11 $\pm$ 0.04	0.09 $\pm$ 0.03
10	10A	Butter extra	0.90 $\pm$ 0.90	0.79 $\pm$ 0.79
11	1B	Milk	0.24 $\pm$ 0.07	0.03 $\pm$ 0.01
12	2B	Vanilla flavoured cream cheese	0.12 $\pm$ 0.05	0.05 $\pm$ 0.02
13	3B	Natural cream cheese	0.27 $\pm$ 0.04	0.05 $\pm$ 0.01
14	4B	Cream	0.19 $\pm$ 0.04	0.04 $\pm$ 0.01
15	5B	Cream cheese with horseradish	0.30 $\pm$ 0.12	0.09 $\pm$ 0.04
16	6B	Cream cheese with caraway	0.14 $\pm$ 0.05	0.06 $\pm$ 0.02
17	7B	Natural buttermilk	0.49 $\pm$ 0.04	0.06 $\pm$ 0.01
18	8B	Half fat cottage cheese	0.32 $\pm$ 0.04	0.10 $\pm$ 0.01
19	1C	Natural yogurt "Nadburzański"	0.38 $\pm$ 0.08	0.07 $\pm$ 0.02
20	2C	Vanilla flavoured cream cheese	0.45 $\pm$ 0.04	0.14 $\pm$ 0.01
21	3C	Natural cream cheese	0.22 $\pm$ 0.06	0.05 $\pm$ 0.01
22	4C	Natural yogurt	0.25 $\pm$ 0.02	0.03 $\pm$ 0.00
23	5C	Buttermilk	0.13 $\pm$ 0.06	0.01 $\pm$ 0.01
24	6C	Kefir	0.08 $\pm$ 0.08	0.01 $\pm$ 0.01
25	7C	Cream	0.17 $\pm$ 0.11	0.03 $\pm$ 0.02
26	8C	Milk "Nadbuzański dzban"	0.25 $\pm$ 0.16	0.03 $\pm$ 0.02
27	9C	Half fat cottage cheese	0.13 $\pm$ 0.03	0.04 $\pm$ 0.01
28	10C	Butter extra	0.65 $\pm$ 0.29	0.58 $\pm$ 0.26
29	1D	Vanilla flavoured cream cheese "Serduszko"	0.18 $\pm$ 0.10	0.04 $\pm$ 0.02
30	2D	Yogurt "Regularis"	0.23 $\pm$ 0.15	0.06 $\pm$ 0.04
31	3D	Strawberry flavoured yogurt "Serduszko"	0.12 $\pm$ 0.06	0.01 $\pm$ 0.01
32	4D	TZATZIKI with fresh cucumber and garlic	0.11 $\pm$ 0.05	0.02 $\pm$ 0.01
33	5D	Cream	0.05 $\pm$ 0.04	0.01 $\pm$ 0.01
34	6D	Natural yogurt	0.28 $\pm$ 0.17	0.04 $\pm$ 0.02

Table 1 contd.

No.	Code	Product	Hg content in lyophilizate (mean $\pm$ SD) [ $\mu\text{g}/\text{kg}$ ]	Hg content in product (mean $\pm$ SD) [ $\mu\text{g}/\text{kg}$ ]
35	7D	Natural kefir	0.08 $\pm$ 0.03	0.01 $\pm$ 0.00
36	8D	Vanilla flavoured cream cheese	0.14 $\pm$ 0.06	0.11 $\pm$ 0.05
37	1E	Cream	0.22 $\pm$ 0.20	0.05 $\pm$ 0.04
38	2E	Natural cottage cheese	0.51 $\pm$ 0.07	0.12 $\pm$ 0.02
39	3E	Natural cottage cheese "Domowy"	0.38 $\pm$ 0.08	0.08 $\pm$ 0.02
40	4E	Cream cheese with spring onion "Twój smak"	0.44 $\pm$ 0.12	0.16 $\pm$ 0.04
41	5E	Kefir	0.29 $\pm$ 0.15	0.04 $\pm$ 0.02
42	6E	Half fat cottage cheese	0.18 $\pm$ 0.09	0.13 $\pm$ 0.07
43	1F	Cream	0.13 $\pm$ 0.02	0.03 $\pm$ 0.01
44	2F	Kefir	0.23 $\pm$ 0.13	0.02 $\pm$ 0.01
45	3F	Granular cottage cheese	0.32 $\pm$ 0.13	0.08 $\pm$ 0.03
46	4F	Half fat cottage cheese KRAJANKA	0.19 $\pm$ 0.10	0.05 $\pm$ 0.03
47	5F	Butter extra	0.55 $\pm$ 0.19	0.47 $\pm$ 0.16
48	6F	Milk	0.60 $\pm$ 0.19	0.06 $\pm$ 0.02

Mean – mean arithmetic value; SD – standard deviation.

Among examined creams, that produced by OSM D was characterized by the lowest content of mercury and the highest content of this element was obtained for cream of OSM A (Table 1). Slightly higher amounts of mercury were determined in the case of cream cheeses and cottage cheeses. Natural cream cheeses (3B and 3C) and vanilla flavoured cream cheese (2B) contained the lowest amount of mercury – 0.05  $\mu\text{g}/\text{kg}$ . Cream cheese with spring onion (OSM E) was characterized by the highest mercury content – 0.16  $\mu\text{g}/\text{kg}$  (Table 1). Cottage cheeses from the different regions of Poland contained various amounts of mercury – from 0.04 and 0.05  $\mu\text{g}/\text{kg}$  in the case of half fat cottage cheese of OSMs C and F, respectively, to 0.23  $\mu\text{g}/\text{kg}$  for cottage cheese produced by OSM A. Half fat cottage cheese from Silesia and cottage cheese produced by OSM E contained a moderate mercury level of, respectively, 0.10 and 0.13  $\mu\text{g}/\text{kg}$  of product (Table 1).

The highest level of mercury in the whole range of dairy products was obtained for butter originated from different parts of Poland. This level was as high as 0.79  $\mu\text{g}/\text{kg}$  of product in the case of butter extra produced by OSM A. The lower contents, respectively, 0.33 do 0.50  $\mu\text{g}/\text{kg}$ , were noted for butter extra from OSM F and OSM C (Table 1).

The investigations allowed to assess how environmental pollution and concentration of component in technological processes influence mercury content in dairy products representing a wide range of assortment and originated from various regions of the country. Mercury concentration varied from 0.01 to 0.79  $\mu\text{g}/\text{kg}$ , which means that the lowest concentration corresponds with a detection limit of the apparatus.

Taking into consideration the mean contents of mercury in all the products of the particular producers, with the exception of butter and whole milk powder, there were no significant difference between them. What is interesting, the products from the less industrialized regions contained higher mean concentrations of mercury than those from the regions of higher environmental pollution. Mercury can be transferred over long distances to regions where there are no significant sources of emission.

In the year 2000, participation of the particular industrial sources in mercury emission to the air was as follows: power stations – 26.5 %, individual home fireplaces – 20.4 %, cement production – 12.6 %, chlorine production – 16.9 % (technology withdrawn from UE since 2007), ferrous metallurgy – 5.2 %, zinc and lead metallurgy – 6.5 %, oil burning – 0.7 %, waste burning – 4.8 %, other – 6.4 % (*ie* crematories) [10].

In the particular groups of assortment, big differences in mercury content were observed between butter as well as whole milk powder and the other products. Milk, cream and fermented milk drinks were characterized by the low values of mean mercury concentration and the lowest value – 0.02 µg Hg/kg, was noted for kefir. Safety of these product for the consumers' health is very important as they are valuable source of calcium in diet.

A small increase in mean mercury content (to 0.11 µg Hg/kg) was observed in the case of cream and cottage cheeses. Similar interactions were reported by Anastasio et al [11] who noted the highest mercury concentration in fresh sheep cheeses.

According to JECFA (*The Joint FAO/WHO Expert Committee on Food Additives*), the *provisional tolerable weekly intake* (PTWI) for mercury from all the sources is 0.005 mg/kg of body weight including 0.0016 mg/kg for organic mercury compounds. These values are continuously verified [12, 13].

*European Commission* Regulation (EC) No. 1881/2006 of 19 December 2006 [14] setting maximum levels for certain contaminants in foodstuffs, defines the limits of mercury concentration exclusively in fish and seafood – 0.5 mg Hg/kg of wet weight (for the chosen fish species – 1.0 mg/kg). The research concerning mercury content in various foodstuffs, carried out in recent years showed that they are not dangerous for human's health. For the particular assortments of food concentrates, values of mercury concentration were as follows:

- desserts, ersatz coffee, juices and beverages – 0.001 mg/kg (0.001 ppm),
- ice-creams, broths and protein hydrolysates – 0.002 mg/kg (0.002 ppm),
- instant soups – 0.013 mg/kg (0.013 ppm), especially high mercury content was determined in boletus soup, champignon soup and cream soup and beetroot soup [12].

Analysis of bakery and confectionery flours revealed that mercury concentration was lower than 0.1 mg/kg, whereas the *maximum permissible* dose is equal 0.2 mg/kg [15]. In 2004 the monitoring research of mercury content in foodstuffs was carried out and the results were as follows: wheat and mixed bread, flours, grouts and noodles – 0.003 mg/kg; confectionery products – 0.002 mg/kg; food for infants and young children (dairy, cereal, vegetable and soy products) – 0.002 mg/kg [16]. On the other hand, herbal products contained less than 0.01 mg Hg/kg and only in one case mercury content was slightly higher than 0.02 mg/kg [17]. Results of the studies concerning

mercury content in milk and dairy products, obtained in different countries and in different years, are presented below:

- Roh et al 1975 (Korea) [18] – 3–10 ppb,
- Sell et al 1975 (North Dakota) [19] – less than 2 ppb,
- Sell and Davison 1975 (North Dakota) [20] – less than 1 ppb,
- Dobrzanski et al 2009 (Poland, Wroclaw) [21] – 0.24 and 0.27 ppb,
- Silesia (late 80s.) – 0.3–6.7 ppb,
- region of Zgorzelec and Bogatynia – 2 ppb,
- Spain, the Canary Islands – 0.09–0.61 ppb,
- China – 1.0–3.9 ppb,
- Italy, Rome surroundings – 0.9–38 ppb,
- Egypt – even 86–556 ppb [21].

The national norms specify the maximum mercury content in food which is various, depending on the country, *eg* 1 mg Hg/kg in Sweden and Japan, 1.5 mg Hg/kg in Norway and 0.5 mg Hg/kg in Poland. In the latter country, the maximum concentration of mercury in milk has been officially established at 10 µg/kg (10 ppb) [21]. Comparing the results obtained for the examined products with above-mentioned data, it can be observed that they are lower and do not exceed permissible levels. Therefore, it can be assumed, that the investigated dairy products do not pose a threat to consumers' health and can be safely consumed providing valuable nutrients.

## Conclusions

In all studied dairy products the mercury content was lower than the permissible level of 10 µg/kg. Results indicate no hazard of mercury intoxication when using dairy products of Polish production.

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**MONITORING POZIOMU RTĘCI CAŁKOWITEJ  
W WYBRANYCH PRODUKTACH MLECZARSKICH  
Z REGIONÓW POŁUDNIOWO-WSCHODNIEJ POLSKI**

Katedra Analizy i Oceny Jakości Żywności  
Uniwersytet Przyrodniczy w Lublinie

**Abstrakt:** Celem pracy było określenie poziomu stężenia rtęci całkowitej w mleku i produktach mleczarskich pochodzących z rejonów południowo-wschodniej Polski. Oznaczanie prowadzono z użyciem analizatora rtęci AMA-254. W pracy przebadano 48 produktów, począwszy od mleka spożywczego, a skończywszy na mleku w proszku, co miało na celu określenie wpływu koncentracji surowca na ewentualny wzrost stężenia rtęci w próbkach. Zawartość rtęci w badanych produktach mleczarskich nie przekracza dopuszczalnego limitu stężenia rtęci w mleku, jako produkcie spożywczym, który wynosi 10 µg/kg (10 ppb). Najmniej rtęci zawierają kefiry (0.01 µg Hg/kg produktu, Biomlek), natomiast największa jej ilość występuje w masłach (0.79 µg Hg/kg produktu, Krasnystaw). W świetle obecnego stanu wiedzy analizowane produkty nie stanowią zagrożenia dla konsumentów, jeśli chodzi o skażenie rtęcią. Uzyskane dane wskazują, że produkty mleczarskie pochodzące z Polski południowo-wschodniej nie dają jakichkolwiek niepokojących przesłanek informujących pośrednio o ewentualnym skażeniu terenów naturalnych czy rolniczych tym toksycznym pierwiastkiem.

**Słowa kluczowe:** rtęć, produkty mleczarskie, bezplamieniowa absorpcyjna spektrometria atomowa