



Determining content of mercury in vitamin and mineral dietary supplements

Oznaczanie zawartości rtęci w witaminowych i mineralnych suplementach diety

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ABSTRACT

INTRODUCTION: Dietary supplements are increasingly more frequently used to increase the daily intake of essential vitamins and minerals. These preparations are classified as a special category of foods and are subject to food law, not pharmaceutical regulations. As a result, the requirements for dietary supplements are much less restrictive than to medications. The aim of study was to determine the level of mercury in multivitamin and multimineral preparations and to analyze the results.

MATERIAL AND METHODS: The study included 22 dietary supplements containing vitamins and minerals available on the Polish market. Mercury was analyzed with an AAS using AMA 254.

RESULTS: The concentrations of Hg were 0.22–5.85 µg/kg (the average content was 2.40 µg/kg). The highest average mercury content was 4.51 µg/kg, whereas the lowest one was 0.31 µg/kg. The results were also analyzed in individual groups depending on the main component and indication for use as well as pharmaceutical formulation. Furthermore, according to the manufacturer's recommendation, the dose of mercury taken with a single medicine was estimated for time of a day, a week, a month, 3 months and also a year. The findings were compared to the limits defined in the regulations of the European Union and PTWI set by the JECFA to assess the health risk. The results presented in this study proved that the amount of mercury in the tested dietary supplements is much lower than the value of valid regulations.

CONCLUSIONS: As long as they are used with the recommendation of manufacturer, the analyzed supplements do not pose a threat to human health.

KEY WORDS

vitamin and mineral dietary supplements, mercury, AAS

STRESZCZENIE

WSTĘP: Suplementy diety to produkty coraz częściej wybierane jako łatwy sposób na uzupełnienie niedoborów witamin i minerałów w codziennej diecie. Są to środki spożywcze podlegające prawu żywnościowemu a nie farmaceutycznemu, co przekłada się na znacznie mniejsze wymagania niż w przypadku leków. Również prosta i niekosztowna procedura ich rejestracji sprawia, że każdego roku zwiększa się liczba dostępnych preparatów. Celem pracy było oznaczenie zawartości rtęci w witaminowo-mineralnych suplementach diety oraz analiza uzyskanych wyników.

Received: 29.04.2019

Revised: 03.06.2019

Accepted: 04.06.2019

Published online: 25.11.2019

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MATERIAŁ I METODY: Do badania wybrano 22 suplementy diety dostępne na polskim rynku, zawierające w swoim składzie witaminy i minerały. W zebranych próbkach oznaczono zawartość rtęci przy użyciu spektrometru absorpcji atomowej AMA 254.

WYNIKI: Zawartość rtęci mieściła się w zakresie 0,22–5,85 µg/kg. Najwięcej rtęci – 4,51 µg/kg – występowało w preparacie uzupełniającym poziom cynku i miedzi. Najmniejsza zawartość wynosiła 0,31 µg/kg w preparacie witaminowo-mineralnym. Uzyskane wyniki analizowano w poszczególnych grupach suplementów diety w zależności od składnika głównego i wskazań do stosowania oraz od postaci farmaceutycznej. Obliczono także dzienne pobranie rtęci przy maksymalnych zalecanych dawkach w ciągu dnia, miesiąca, 3 miesięcy oraz roku. Uzyskane oznaczenia porównano z obowiązującym w krajach Unii Europejskiej maksymalnym dopuszczalnym poziomem zawartości rtęci w suplementach diety oraz tymczasowym tygodniowym pobraniem PTWI, ustalonym przez JECFA.

WNIOSKI: Przeprowadzona analiza wykazała, że poziom zawartości rtęci w badanych suplementach diety witaminowo-mineralnych nie został przekroczony. Stosowanie tych preparatów w ilości zalecanej przez producenta nie stwarza ryzyka dla zdrowia.

SŁOWA KLUCZOWE

witaminowo-mineralne suplementy diety, rtęć, AAS

INTRODUCTION

There has been growing interest in dietary supplements among consumers for the last several years. Their widespread availability and producers' extensive marketing activities have made the supplement market in Poland grow very dynamically, and their consumption has increased significantly. Preparations that contain vitamins and minerals are recommended as an easy way to supplement vital ingredients in the daily diet, and eliminate the effects of their deficiency [1].

According to food law, dietary supplements are food products and to be able to put them on the market, the manufacturer is only required to submit a notification to the Chief Sanitary Inspectorate [2,3]. Thus, there is insufficient control of these products and there are risks of many side effects that may occur in consumers [4]. Dietary supplements are not tested for interactions with drugs and food, nor are they observed when it comes to controlling possible side effects [4,5,6].

Dietary supplements can be a potential source of mercury, among others, introduced into the body with food, especially since in recent years an increase in the consumption of drugs and dietary supplements in developed countries such as the US, Canada and Europe has been observed. This is related to their low prices, easy accessibility, and the conviction that there are no side effects in comparison with medicines [7,8]. One of the major sources of toxic heavy metals in complementary medicines products is the deliberate introduction of some of the metals including As, Hg, and Pb as therapeutic ingredients in Ayurvedic medicines [9].

Mercury in dietary supplements can come from various sources: contaminated raw materials, reagents used in production, equipment and packaging [10]. In regulation No. 629/2008 of 2 July 2008 by the EC Commission, the maximum acceptable levels for certain pollutants (lead, cadmium, mercury) in food products related to the exposure of persons using dietary supplements were set. Due to the detection of the above-mentioned

heavy metals in some food products, the maximum permissible levels of these heavy metals were set. For mercury, the maximum level of its content in a dietary supplement available on the market is 0.10 mg/kg [11]. Mercury is one of the most toxic elements for humans, animals, plants and aquatic organisms. It has mutagenic and teratogenic properties, and can accumulate in the human body [12]. The toxic effects of mercury at the cellular level involve the induction of oxidative stress and mitochondrial activity dysfunctions. As a consequence, there is an increase in the concentration of hydrogen peroxide, a decrease in the glutathione pool in mitochondria, an increase in lipid, protein and DNA peroxidation, as well as a decrease in ATP synthesis [13,14,15]. Mercury binds to metallothionein, displacing zinc, copper and other trace elements, and also competes with selenium, which causes a decrease in the activity of metalloenzymes. Moreover, the mercury-selenium complex limits the activity of the glutathione peroxidase enzyme, which participates in the conversion of hydrogen peroxide into water, where selenium acts as a cofactor [16,17].

The number of published works on the content of mercury in herbal dietary supplements is not very large [18,19,20].

The aim of the manuscript was to determine the content of mercury in vitamin and mineral dietary supplements. An analysis of the obtained results depending on the main component and an indication for the use of specific groups of dietary supplements as well as on the pharmaceutical form in which it is available on the market was conducted. Additionally, the daily mercury intake at the maximum recommended doses during one day, one month, 3 months and one year were determined. The obtained data was compared to the maximum permissible mercury content in dietary supplements. The study included 22 dietary supplements containing vitamins and minerals. Table 1 includes information about the tested preparations, their form, composition and declared effects.



MATERIALS AND METHODS

The study included 22 dietary supplements containing vitamins and minerals available on the Polish market Table I. The supplements were hand-powdered in a mortar. Then, they were weighed and analysed using an AMA 254 atomic absorption spectrometer. Approximately 20 mg of powdered dietary supplement was weighed for each analysis and labelled. The device was suitably cleaned in air and deionized water prior to each analysis and blank tests were made.

Markings were made using the AMA 254 device. AMA 254 is an atomic absorption spectrometer that is designed to determine total mercury, regardless of the form in which mercury occurs. Mercury is easily released from its compounds (organic and inorganic) by being converted to an atomic form. This makes mercury determination simple and fast, and it is not necessary to use an atomic absorption spectrometer with a hydride generator attachment. Another advantage is that the pyrolytic mineralization process takes place inside the device and it is not necessary to use a mineralizer for mercury determination.

Mercury measurement using this device consists of three stages:

- First, the solid or liquid sample is dried and then burned in a stream of oxygen
- In the second stage, the released mercury vapours pass through the catalytic column and are collected by the amalgamator (a small glass tube containing a gold-coated ceramic material). After collecting all of the mercury from the released gases, the amalgamator is heated to about 900°C, and the mercury vapour is released to the detection system. In the case of mercury-low matrices, it is possible to concentrate it more.
- At the third stage (detection), the mercury vapour is segregated into two parts in a device called a cuvette. One part of the cuvette supplies the mercury carrier gas sample to an optical pathway conducive to low mercury concentration analysis, and the other part supplies gas to the optical pathway and is optimized for high mercury concentrations. This dual cuvette system allows the device to extend the dynamic

range for analysis results at different mercury concentrations. The cuvette is set on the pathway of a typical atomic absorption spectrometer. The spectrometer has a mercury lamp that emits light at a wavelength of 253.7 nm and a silicon UV diode as a detector for mercury quantification [21].

This method uses the radiation absorption phenomenon by free mercury atoms in a basic state. It consists in the fact that free mercury atoms absorb the radiation emitted by the mercury lamp, the hollow cathode of which is made of mercury. As a result, the initial intensity of radiation emitted by the mercury lamp is reduced, and it is recorded by the spectrometer. The magnitude of this reduction is proportional to the number of mercury atoms in released pairs of this element.

The detection limit is 0.003 ng of mercury in the marked sample. Medical or technical oxygen that provides better combustion characteristics and guarantees repeatability of the device is the carrier gas and the oxidizer.

The device is controlled by an external PC with advanced software (calibration curves, statistical analysis of results, process control with display of a current signal) running on Windows®.

The correctness of the applied method was determined using a reference material, INCT-MPH-2 Polish Herbs Mixture, in which the mercury-certified content was 0.018 ± 0.002 mg/kg. The content of mercury obtained from 5 repetitions was 0.019 ± 0.0004 mg/kg, and the recovery value was 105.6%.

The statistical analysis of the obtained results was performed using Microsoft Excel and Statistica for Windows 12 pl. The first stage of the statistical analysis of the results was to investigate the normality of mercury content distribution of the tested herbal supplements. For this purpose, the Shapiro-Wilk (W) test was used. The mercury content distribution deviated from the normal one ($p > 0.05$), and was right-side developed. Non-parametric tests were used for subsequent analyses. The values of arithmetic mean, standard deviation, change range, variation coefficient, and median were used to develop and describe the results. Statistical variability between particular groups was based on the Kruskal-Wallis test for multiple samples, and for equality between the two groups, the Mann-Whitney U test.

Table I. Composition and action of tested vitamin and mineral supplements

Tabela I. Spis badanych witaminowo-mineralnych suplementów diety

No.	Ingredients	Indication
1	2	3
1.	vitamin: C minerals: Zn, Se	supports normal immune system function
2.	vitamins: C, niacin, E, pantothenic acid, B6, riboflavin, thiamine, A, folic acid, K, D, B12 minerals : Ca, Mg, Fe, Zn, Mn, Cu, J, Se, Cr, Mo	dietary supplementation of vitamins and minerals for woman with menopause symptoms
3.	vitamins : B6, pantothenic acid minerals : Mg	recommended for adults burdened with intellectual effort, and the elderly



1	2	3
4.	vitamins: C, niacin, E, pantothenic acid, B6, riboflavin, thiamine, A, biotin, B12 minerals : Fe, Zn	skin, hair and nail regeneration
5.	vitamins: niacin, B6, folic acid, E minerals: Ca	reduces menopause symptoms, helps maintain weight and normal bone function
6.	vitamins: A, C, D, E, riboflavin, thiamine, niacin, B6, pantothenic acid, folic acid, B12, biotin minerals: Ca, Fe, J, Mg, Zn, Mn, K, P, Se, Cr, Mo	vitamin and mineral supplement designed for active people
7.	vitamin: D3 minerals: Ca, Mg, Zn	dietary supplementation of calcium, magnesium, zinc and vitamin D
8.	vitamins: niacin, E, biotin, riboflavin, B6, thiamine, A, folic acid, D, B12 minerals: Zn, Mn, Cu, J, Se, Mo, Cr	ensures optimal intake of vitamins in minerals especially in period of increased demand, convalescence, autumn and winter time, intensive physical and mental activity
9.	vitamins: E, riboflavin, thiamin, B6, niacin, folic acid, pantothenic acid, B12, C, A minerals: Ca, Mg, Zn, Fe, Se, J	dietary supplementation of vitamins and minerals in period of increased demand, intensive physical and mental activity
10.	vitamins: C, E, B6, folic acid, thiamine, riboflavin, B12, D, niacin, biotin, pantothenic acid minerals: Fe, Zn, Mg, Ca, Cu, Mn, J	ensures optimal intake of immune system boosting ingredients
11.	minerals: Zn, Cu	ensures optimal daily intake of calcium, especially with skin problems resulting from hyperactivity of sebaceous glands
12.	vitamins: E, thiamine, riboflavin, B6, B12, C, biotin, niacin, pantothenic acid folic acid minerals: Fe, Zn, J	supports maintaining good health and normal immune system function
13.	vitamin: B6 minerals: Mg	supporting normal nervous system and muscle function, reducing physical fatigue
14.	vitamins: C, niacin, E pantothenic acid, B6, riboflavin, thiamine, folic acid, K, D, B12 minerals: Ca, P, Mg, Fe, Zn, Mn, Cu, J, Cr, Se, Mo	supplements daily diet in vitamins and minerals in period of increased demand and physical fatigue
15.	vitamins: C, riboflavin, niacin, E, pantothenic acid, B6, thiamine, A folic acid, biotin, D3, B12, K minerals: Mo, Cr, J, Zn, Cu, Se, Mg, Mn, Ca, P, Fe	supplements daily diet in vitamins and minerals
16.	vitamins: thiamine, riboflavin, B6, C, E, PP, pantothenic acid, biotin, folic acid, A, D3 minerals: Fe, Zn, Mn, Cr, Cu, Mg, Se	support in period of physical fatigue, convalescence, supplementation in deficiency of vitamins and minerals connected with inappropriate diet, abnormal absorption or increasing demand
17.	vitamins: A, D, E, C, riboflavin thiamine , niacin, B6 folic acid, B12 biotin, pantothenic acid minerals: Ca, Mg, K, Zn, Fe, Mn, Cu, J, Se others: lutein, hyaluronic acid	dedicated for elderly people to help maintain proper daily intake of vitamins and minerals
18.	vitamins: A, D, E, K, thiamine, riboflavin, niacin, pantothenic acid , B6, B12, C, biotin minerals: Zn, I, Mg, Cu, Se, Fe others: panax ginseng root extract, maca root extract	designed for active people, helps maintain natural immunity and vitality
19.	vitamins: riboflavin , C, E minerals: Fe	maintains proper condition of mucous membrane and red blood cells, healthy skin, ensures proper metabolism of iron
20.	vitamins: folic acid, thiamine, riboflavin, B6, B12, niacin, B5, pantothenic acid, C, D minerals: Mo, Cu, Se	helps safeguard daily intake of nutrients during pregnancy and postnatal period
21.	vitamin: B6 minerals : Mg	reduces tiredness and fatigue, contributes to normal functioning of nervous system
22.	vitamin: C minerals: Zn	contributes to strengthening of immune system, diet supplementation in period of decreased immunity and increased flu incidence



RESULTS

The average mercury content in the tested dietary supplements was 2.40 µg/kg. In 9 preparations the content was above this value, while in 13 it was below. The preparations in which the content of mercury was at a level that is close to the average value of all the tested samples is Preparation No. 5 with a result of 2.40 µg/kg and Preparations No. 21 and 13 with an average content of mercury of 2.31 µg/kg and 2.32 µg/kg. The highest average content of mercury was determined in Preparation No. 11, which was 4.51 µg/kg. However, the lowest content of this element, 0.31 µg/kg, was found in preparation No. 12 (Tab. II). The tested dietary supplements were divided into 10 groups, depending on the indication for use and the content of the main component.

The highest average content of mercury of 4.51 µg/kg was determined in the group of preparations supplementing the diet with copper and zinc, intended for skin problems resulting from excessive activity of sebaceous glands. A high content of mercury – 4.20 µg/kg was also found in the group of preparations containing iron. In the group of folic acid supplements, the content of mercury was 3.93 µg/kg. The lowest content of mercury of 0.99 µg/kg was found in the group of preparations containing calcium and vitamin D3, and preparations supporting the functioning of the skeletal system (Tab. III). The content of mercury that is close to the average content was found in the following groups of supplements: for menopausal women, containing vitamin C, and in preparations with magnesium and vitamin B6. The Kruskal-Wallis test showed the significance of differences between the occurrence of mercury in individual groups, depending on the use and the main component, the significance level was $p < 0.05$.

With the information on the content of magnesium and vitamin C in most of the tested supplements, an analysis of the co-occurrence between the content of Mg, vitamin C and Hg was made (Tab. IV). In the tested group of vitamin and mineral supplements, statistically significant correlations occurred between the content of mercury and magnesium (-0.58) and the content of magnesium and vitamin C (0.49).

In terms of their pharmaceutical form, the tested dietary supplements were tablets, coated tablets, drops and capsules. The highest content of mercury was found in coated tablets – 3.59 µg/kg, and the lowest in drops – 0.83 µg/kg.

The significance of differences between the co-occurrence of mercury depending on the drug form was shown using the Kruskal-Wallis test. Statistically significant differences ($p = 0.005$) in the content of mercury occurred between the following: drop-coated tablet, drop-tablet, capsule-tablet, capsule-coated tablet, tablet-coated tablet (Tab. V).

Table II. Statistical analysis of content of mercury in vitamin and mineral supplements [µg/kg]

Tabela II. Analiza statystyczna zawartości rtęci w witaminowo-mineralnych suplementach diety [µg/kg]

No.	Arithmetic mean	Standard deviation	Range
1.	1.47	0.28	1.15–1.63
2.	2.13	0.04	2.09–2.16
3.	1.63	0.23	1.37–1.79
4.	1.34	0.14	1.24–1.50
5.	2.40	0.15	2.23–2.51
6.	1.00	0.04	0.96–1.04
7.	0.99	0.06	0.93–1.04
8.	3.46	0.08	3.41–3.55
9.	4.03	0.21	3.81–4.24
10.	3.32	0.36	2.93–3.62
11.	4.51	1.07	3.42–5.49
12.	0.31	0.15	0.22–0.49
13.	2.32	0.11	2.20–2.43
14.	1.46	0.07	1.38–1.52
15.	2.0	0.66	1.45–2.81
16.	0.36	0.02	0.34–0.38
17.	1.05	0.17	0.85–1.23
18.	3.48	0.09	3.42–3.58
19.	4.20	0.90	3.37–5.85
20.	3.93	0.73	3.30–4.98
21.	2.31	0.34	1.99–2.75
22.	2.74	0.05	2.69–2.79
Whole	2.40	1.33	0.22–5.85



Table III. Statistical analysis of content of mercury in vitamin and mineral dietary supplements depending on main component and indication for use [$\mu\text{g}/\text{kg}$]
Tabela III. Analiza statystyczna zawartości rtęci w witaminowo-mineralnych suplementach diety w zależności od składnika głównego i wskazania do stosowania [$\mu\text{g}/\text{kg}$]

No.	Main ingredient/indication	Arithmetic mean	Standard deviation	Range	Coefficient variability [%]
1.	vitamins C/supporting immune system	2.51	0.85	1.15–3.62	34
2.	hop cones extract/reduction of menopause symptoms	2.25	0.17	2.09–2.51	8
3.	biotin, cysteine/hair, skin and nails regeneration	1.34	0.14	1.24–1.50	10
4.	magnesium, vitamin B6/supports normal nervous system and muscle functions, decreases physical fatigue	2.11	0.40	1.37–2.75	19
5.	calcium, vitamin D3/maintenance of normal bones function	0.99	0.06	0.93–1.04	6
6.	multivitamin supplements/ensures optimal daily intake of vitamins and minerals	2.01	1.52	0.22–4.24	75
7.	zinc, copper/skin problems resulting from hyperactivity of sebaceous glands	4.51	1.07	3.42–5.49	24
8.	senior multivitamin supplements/supplements deficiencies in elderly diet	1.53	0.68	0.85–2.81	44
9.	iron/maintains proper condition of mucous membrane and red blood cells	4.20	0.90	3.37–5.85	21
10.	folic acid/supplements daily intake of nutrients during pregnancy and postnatal period	3.93	0.73	3.30–4.98	19

Table IV. Spearman's Correlation analysis

Tabela IV. Analiza korelacyjna Spearmana

	Hg [$\mu\text{g}/\text{kg}$]	Vitamin C content
Vitamin C content	0.22	
Magnesium content	-0.58*	0.49*

* correlation coefficient statistically significant at $p < 0.05$

Table V. Statistical analysis of content of mercury in vitamin and mineral dietary supplements depending on their pharmaceutical forms [$\mu\text{g}/\text{kg}$]

Tabela V. Analiza statystyczna zawartości rtęci w witaminowo-mineralnych suplementach diety w zależności od postaci farmaceutycznej [$\mu\text{g}/\text{kg}$]

Pharmaceutical forms	Arithmetic mean	Standard deviation	Range	Coefficient variability [%]
Tablet	2.33	0.34–5.49	1.18	51
Coated tablet	3.59	1.45–5.85	1.12	31
Drop-tablet	0.83	0.22–1.50	0.58	70
Capsule	1.30	0.85–1.79	0.36	28

DISCUSSION

Dietary supplements are a specific type of food that is subject to food law. Unlike medicines, they have no healing properties and are supplements to the daily diet. Despite the fact that qualitative and quantitative compositions are required, there is no need to conduct tests to confirm this information. All legal requirements for both the launching, labelling and testing of dietary supplements are much less restrictive than for medicines, or they are simply not specified [2].

There are many factors that make up the increase in consumption of dietary supplements in many countries. First, as food products, supplements are widely available, which results from legal regulations in force. Increasingly more frequently, their use is considered by consumers as necessary for proper functioning of the body and health [5]. Advertising also plays an impor-

tant role. According to a report by the Supreme Audit Office in 2015, every fourth advertisement concerned health and medicinal products of which 62% were trade messages with dietary supplements [22].

Among the numerous groups of dietary supplements, the largest one is vitamin and mineral dietary supplements. According to a report by the European Commission, the total value of the dietary supplements market in 2005 was estimated at EUR 5 billion, and half of this value concerned vitamin and mineral dietary supplements. The data shows that the Polish supplement market grew by 219% in the years 1997–2005, which is the highest result among the EU Member States [5,23,24].

Vitamin and mineral supplements that are easily available on the Polish market were selected for the study. The conducted analysis of the content of mercury in these preparations did not indicate any risk to human health. The determined concentrations of mercury were



much lower than the maximum permissible level of the content of mercury specified in the EC Commission Regulation that amounted to 100 µg/kg [11,22]. Additionally, to assess the safety of people using dietary supplements, the PTWI index was used, which assesses the intake of heavy metals with food in a given period. The European Food Safety Agency (EFSA) defines the PTWI, i.e. the provisional tolerable weekly intake, as a defined amount of substance that can be consumed within a week with food or water throughout life without significant adverse health effects. The PTWI index is usually given in units of a given substance per kilogram of body weight, and refers to chemicals that do not show immediate toxic effects and are not quickly removed from the body. They include heavy metals such as mercury, arsenic, cadmium and lead, which after a certain time accumulate in the body [25,26]. The calculated PTWI indices that determine the provisional tolerated weekly intake were lower than the values es-

tablished by the Expert Committee on Food Additives FAO/WHO (JECFA). Taking a preparation with the highest determined content of mercury in accordance with the maximum recommended daily portion for a week is 0.01% of the PTWI determined for inorganic mercury (Tab. VI).

Based on the results of the content of mercury obtained in the studied food supplements, the content of this element in 1 tablet of the preparation was calculated. Using the information on the maximum recommended portion placed by the manufacturer on the packaging of a dietary supplement, the amount of mercury absorbed with this preparation during the day was calculated. Additionally, the dose of mercury taken during the use of a given vitamin and mineral supplement for 1 month, 3 months, and a year was calculated.

The greatest amount of mercury in one tablet that amounted to 0.00135 µg was determined in preparation No. 11. When taking 3 tablets per day, which is

Table VI. Average mercury content in recommended daily portion and in monthly and annual treatment

Tabela VI. Średnia zawartość rtęci w zalecanej dziennej porcji w kuracji miesięcznej oraz rocznej

No.	Daily max dosage	Average mercury content in 1 tablet [µg]	Daily intake [µg]	Monthly intake [µg]	Annual intake [µg]
1.	2	0.00044	0.00088	0.0265	0.32
2.	1	0.00064	0.00064	0.0192	0.23
3.	1	0.00049	0.00049	0.0147	0.18
4.	2	0.00040	0.00080	0.0241	0.29
5.	1	0.00072	0.00072	0.0216	0.26
6.	1	0.00030	0.00030	0.0090	0.11
7.	1	0.00030	0.00030	0.0089	0.11
8.	2	0.00104	0.00208	0.0623	0.76
9.	1	0.00121	0.00121	0.0363	0.44
10.	1	0.00100	0.00100	0.0299	0.36
11.	3	0.00135	0.00406	0.1218	1.48
12.	1	0.00009	0.00009	0.0028	0.03
13.	1	0.00070	0.00070	0.0209	0.25
14.	1	0.00044	0.00044	0.0131	0.16
15.	1	0.00060	0.00060	0.0180	0.22
16.	1	0.00011	0.00011	0.0032	0.04
17.	1	0.00032	0.00032	0.0095	0.11
18.	1	0.00104	0.00104	0.0313	0.38
19.	1	0.00126	0.00126	0.0378	0.46
20.	1	0.00118	0.00118	0.0354	0.43
21.	1	0.00069	0.00069	0.0208	0.25
22.	2	0.00082	0.00164	0.0493	0.60



the maximum dose recommended by the manufacturer, the intake of mercury per day will be 0.00406 μg . The content of mercury consumed during a monthly treatment is 0.1218 μg , whereas in a 3-month treatment it is 0.365 μg of mercury. The use of supplement No. 11 for one year equals an intake of 1.48 μg of Hg. The smallest amount of mercury in one tablet was in Preparations No. 12 – 0.0009 μg and No. 16 – 0.0011 μg . The same intake of mercury is in a daily maximum serving, which is equal to one tablet. The monthly portion of Preparation No. 12 contains 0.0028 μg of mercury and No. 16 – 0.0032 μg , and the annual dose – 0.03 μg and 0.04 μg , respectively.

As a result of analysing the collected samples of dietary supplements, it was shown that the average content of mercury was 2.40 $\mu\text{g}/\text{kg}$. The lowest concentration of this element was 0.22 $\mu\text{g}/\text{kg}$, whereas the highest was 5.85 $\mu\text{g}/\text{kg}$. A higher content of mercury was obtained in the studies carried out by Kowalski and Frankowski [7]. The range of mercury concentration in the analysed dietary supplements available on the Polish market was 0.9–16.7 $\mu\text{g}/\text{kg}$, and the average value was 5.9 $\mu\text{g}/\text{kg}$. The highest concentrations were obtained in the groups of supplements that contained micro- and macro-elements, as well as vitamins. The highest content of mercury amounting to 16.7 $\mu\text{g}/\text{kg}$ was determined in a preparation containing vitamin C and rutin that supports the immune system [7]. In the studies by Kowalski and Frankowski [7], the highest content of mercury was 3.7 ng of Hg in one tablet, whereas in our studies the highest average content of Hg in one tablet was 1.4 ng of Hg. The amount of mercury taken during the week was 207.2 ng of Hg/week [7], while in our study it was several times smaller and amounted to 28 ng of Hg/week. According to Socha et al. [27], the average content of mercury in supplements was comparable to the obtained value and was 5.36 $\mu\text{g}/\text{kg}$. The range of Hg concentrations was higher and amounted to 0.10–7.99 $\mu\text{g}/\text{kg}$. None of the tested preparations exceeded the permissible content of mercury in food supplements. In the group of supplements that support immunity, the concentration of this element was 9.62 $\mu\text{g}/\text{kg}$, whereas in our studies, the concentration of mercury in this type of supplements was 2.51 $\mu\text{g}/\text{kg}$. What is more, a high content of mercury amounting to 3.25 $\mu\text{g}/\text{kg}$ was obtained in hair, skin and nail strengthening preparations when compared to our studies – 1.34 $\mu\text{g}/\text{kg}$. The highest dose of mercury associated with the use of a given product as recommended by the manufacturers was equal to 0.25% of PTWI, and for the tested supplements it was 0.01% of PTWI.

The content of mercury in preparations containing calcium amounting to 0.99 $\mu\text{g}/\text{kg}$ is similar to the value of 1.52 $\mu\text{g}/\text{kg}$ obtained in the study by Levine et al. [28]. According to Avula et al. [29], from among the 35 dietary supplements tested, only 2 of them contained mercury in the amount of 0.2 μg of Hg/daily dose and 2.6 μg of Hg/daily dose. In the obtained results, the average amount of mercury in the daily portion was 0.00093 μg of Hg.

A higher content of mercury was determined in vitamin and mineral dietary supplements in Lebanon in the studies carried out by Korfali et al. [30]. The average concentration of mercury was 80 $\mu\text{g}/\text{kg}$, and the range was from 10 up to 550 $\mu\text{g}/\text{kg}$. The obtained values did not exceed the standards in force either.

In the studies that included 35 supplements available on the Croatian market [31], 14 of which contained vitamins and minerals, a content of mercury in the range of 2–120 $\mu\text{g}/\text{kg}$ was determined.

The presence of mercury was also detected in Mexican dietary supplements in the studies by Garcia-Rico et al. [32], where the content of mercury ranged from 240 up to 850 $\mu\text{g}/\text{kg}$.

Despite the fact that the mercury concentrations in the studied vitamin and mineral supplements were low and did not exceed the applicable standards, activities to control the concentration of this element should be continued. Mercury shows high toxicity, even in low concentrations, its presence in the environment is a threat to all living organisms [7]. Moreover, consumers often experience the phenomenon of taking several dietary supplements simultaneously over a long period of time, which increases the number of sources of potential exposure to mercury and the risk of its accumulation. According to the applicable law, manufacturers are not obliged to present the results of tests that would confirm that there is no contamination of a given product, only randomly selected products are examined by the State Sanitary Inspection in this respect. The lack of specific requirements in terms of ensuring the proper quality of dietary supplements results in an increased risk of contamination with these substances and is dangerous to human health. Due to the growing popularity of dietary supplements, including vitamin and mineral products, which are increasingly used to supplement the daily diet with essential nutrients, the quality of these products needs to be controlled at all stages, both at the initial stages of production and in the final products [33,34].

CONCLUSIONS

The content of mercury in the studied dietary supplements averaged 2.40 $\mu\text{g}/\text{kg}$, ranging from 0.22–5.85 $\mu\text{g}/\text{kg}$.

The highest content of mercury was found in a product supplementing zinc and copper (4.51 $\mu\text{g}/\text{kg}$), whereas the lowest in a vitamin and mineral preparation (0.31 $\mu\text{g}/\text{kg}$).

The analysis of the supplements in terms of the main component and indications for use showed the lowest content of mercury in the supplements containing calcium and vitamin D3. Significant statistical differences in the occurrence of mercury were found, depending on the indication for use and the main component.

Using the correlation analysis, the existence of statistically significant interdependencies between mercury and magnesium of an antagonistic nature were found.



There were differences in the content of mercury, depending on the pharmaceutical form of the dietary supplement.

The determined content of mercury does not exceed the standards specified in the EC Commission Regulations, or the PTWI indicator set by JECFA. The consumption of the maximum recommended portions of the tested products is safe in the respect of mercury.

Author's contribution

Study design – B. Brodziak-Dopierala, A. Fischer

Data collection – P. Derkacz

Statistical analysis – B. Brodziak-Dopierala

Data interpretation – B. Brodziak-Dopierala, A. Fischer

Manuscript preparation – B. Brodziak-Dopierala, J. Stojko

Literature research – B. Brodziak-Dopierala

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ACKNOWLEDGMENTS

This work was financed by the Medical University of Silesia in Katowice (contract No. KNW-1-166/N/7/1).