

## STUDY OF THE CONTENT OF HEAVY METALS IN FOODS IN BULGARIA

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### Summary

Food safety is a priority for the European Union (EU). EU food safety policy covers all sectors of the food chain. In recent years, numerous studies have been conducted in Bulgaria on contaminants in plant and animal foods as one of the priority areas of food safety policy. Metals are part of the chemical pollutants in the environment. People are exposed to metals from the environment and food and water contaminated with metals. Researchers have found that the intake of metals with food is different and that food exposure is the primary way of metal exposure.

**Keywords:** heavy metals in food, food samples, food safety

### Introduction

It is accepted that metal elements with a relative mass larger than 40 and a relatively high density (above 0.5 g / cm<sup>3</sup>) are called heavy metals [1]. Some heavy metals (copper, zinc, iron, etc.) belong to the so-called trace elements because, in specific concentrations, they are vital for the functioning of the human body. These substances can have an adverse effect when their amount in the body exceeds what is needed. Other heavy metals (such as lead, cadmium, mercury, etc.) are not biologically necessary and have a detrimental effect on human health [2, 3]. Heavy metals can remain in the environment for a long time, as well as accumulate in organisms, which is a premise for their long-term toxic effects [4, 5, 6].

Heavy metals are chemical contaminants in the environment that occur in nature, both naturally and due to human activity. Heavy metal pollution affects not only the environment but also the organisms that inhabit it [7]. Consumption of food and drinking water is the main route for human exposure to heavy metals [8, 9, 10, 11].

Every year in Bulgaria, a national food control program for contaminants is planned and implemented, which is part of the multi-annual national control plan of the Republic of Bulgaria, prepared following the requirements of the legislation of the European Union [12].

## Aim

The study aimed to analyze the levels of heavy metals cadmium (Cd), lead (Pb), tin (Sn), and mercury (Hg) in the food products offered on the Bulgarian market.

## Methods and Materials

The study covered the period 2013-2019. We analyzed 2757 food samples for metal content (including cadmium, lead, tin, and mercury), as well as the trace elements copper and zinc. The samples were collected from 77 plant and animal foods, which had different contributions to the average daily intake of metals such as leafy vegetables, root crops, cereals, and legumes; animal foods - milk, dairy products, meat and offal used for food, fish and shellfish, special purpose foods. The samples were prepared for direct analysis in accredited laboratories.

The method of Graphite-furnace atomic absorption spectrometry (GF-AAS) was used to determine the metal content in the tested food samples (Figure 1).

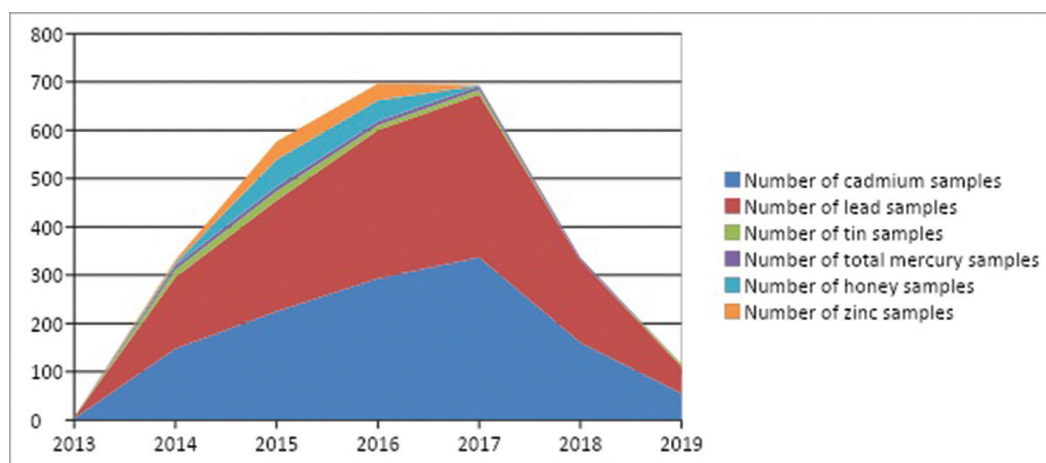
The EU has strict food contamination standards to protect consumers' health [13, 14, 15, 16, 17, 18, 20]. Regulation (EC) № 1881/2006 (as amended by Regulation (EC) 2021/1323 of the Commission of August 10, 2021) and Ordinance № 5, of February 9, 2015, defined maximum levels (mg / kg) for heavy metals in foods [20, 21].

## Results and Discussion

The analysis of the results showed that 97.71% of all tested food samples corresponded to the established maximum permissible amounts for heavy metals in food. [20, 21] Non-compliant samples accounted for 2.28% of all samples, and the discrepancy was only in terms of cadmium and lead content (Table 1).

From Table 1, it can be seen that in products of animal origin, in 97.63% of the samples, the levels of heavy metals corresponded to the statutory maximum levels. Non-compliant samples were only 2.36%.

Compared with the tested products of



**Figure 1.** Samples tested of foodstuffs for heavy metals content for the period 2013 – 2019

**Table 1.** Compliance of the tested samples with the established maximum permissible quantities for heavy metals in food

	Number of samples	Number of samples over LOQ	Number of samples with the value of the result following the maximum permissible quantities (mg/kg)	Number of samples with a result value above the maximum permissible quantities (mg/kg)
Samples of plant origin tested	1149	465	1124	25
Samples of animal origin tested	1608	1434	1570	38
<b>Total</b>	<b>2757</b>	<b>1899</b>	<b>2697</b>	<b>60</b>

vegetable origin, the percentage of non-standard samples in the products of animal origin was higher. In the tested samples of plant foods, 97.82% of them corresponded to the statutory maximum permissible amounts, while in 2.17%, it was found that these norms were exceeded (Figure 2).

Of the 77 types of food studied, 19.48% had a metal content above the maximum allowable amounts.

The simultaneous presence of several heavy metals in one food product was established. 6.49% of the studied types of food contain two metals - cadmium and lead, in a concentration above the maximum allowable amounts. It is established in apples, wild boar meat, herbs and spices, horse meat, and fresh spinach.

Only cadmium or lead was present in 12.98% of the tested foods, in concentrations above the permissible level.

In the studied types of food, the amount of mercury corresponded to the maximum regulated amount.

In the case of products of animal origin:

- 3.44% of the samples of wild boar meat;
- 5.76% of the cow's milk samples;
- 3.33% of the sheep's milk samples;
- 3.12% of the offal samples;
- 1.94% of honey samples;
- 26,31% of the samples of horsemeat;
- 11,11% of the samples of fish and other seafood did not meet the requirements of Regulation (EC) (1881/2006.

In all other tested products of animal origin, the quantities of heavy metals met the regulatory

requirements.

In the tested food products of plant origin, the share of non-standard samples was distributed as follows:

- 2.23% of the samples spinach canned/ deep-frozen;
- 2.72% of fresh spinach samples;
- 11.53% of the apple samples;
- 4.16% of the aubergine samples;
- 0.90% of carrot samples; 12.90% of the samples of herbs and spices;
- 5% of the samples Oyster mushroom sponge mushroom;
- 3.70% of the samples peppers, red.

Samples with established high concentrations of heavy metals are were single:

- lead in wild boar meat - 0.49 mg Pb / kg, standard 0.10 mg Pb / kg;

- lead in cow's milk - 0.1900 mg Pb / kg, standard 0.020 mg Pb / kg;

- lead in sheep's milk - 0.0970 mg Pb / kg, standard 0.020 mg Pb / kg;

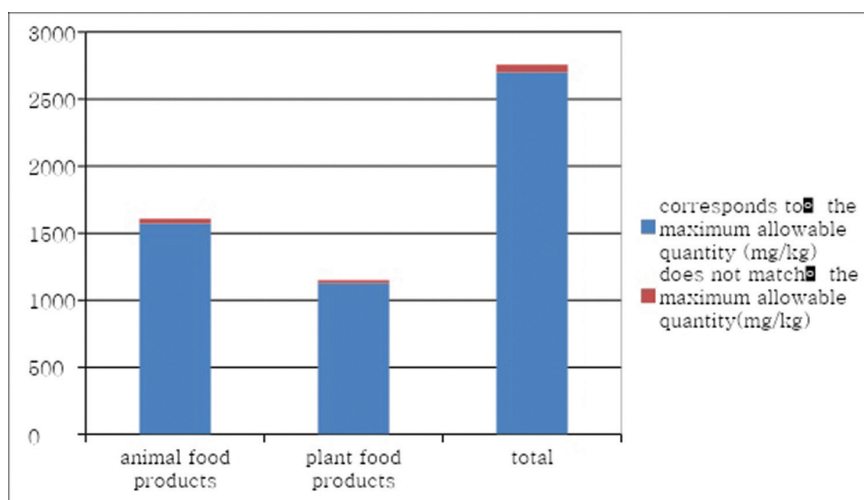
- lead and cadmium in apples - 0.4900 mg Pb / kg, standard 0.10 mg Pb / kg and 0.4400 mg Cd / kg at a rate of 0.050 mg Cd / kg;

- cadmium in fish and other seafood - 0.1170 mg Cd / kg, standard 0.050 mg Cd / kg;

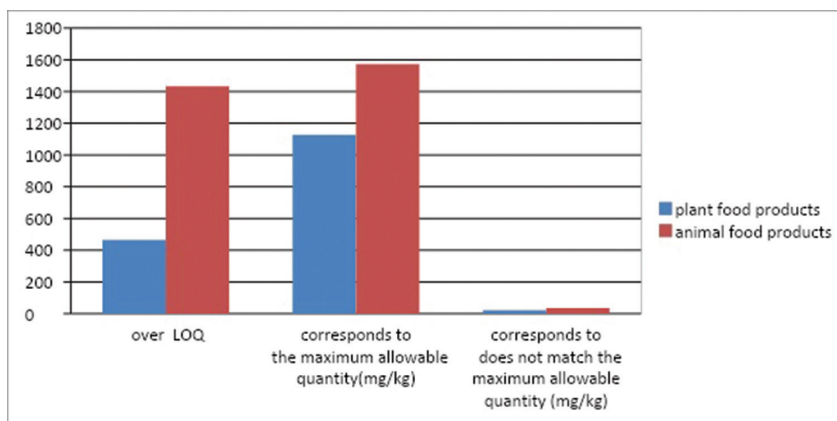
- cadmium in horse meat - 0.8990 mg Cd / kg, standard 0.20 mg Cd / kg;

- cadmium in spinach - 0.4500 mg Cd / kg, standard 0.20 mg Cd / kg.

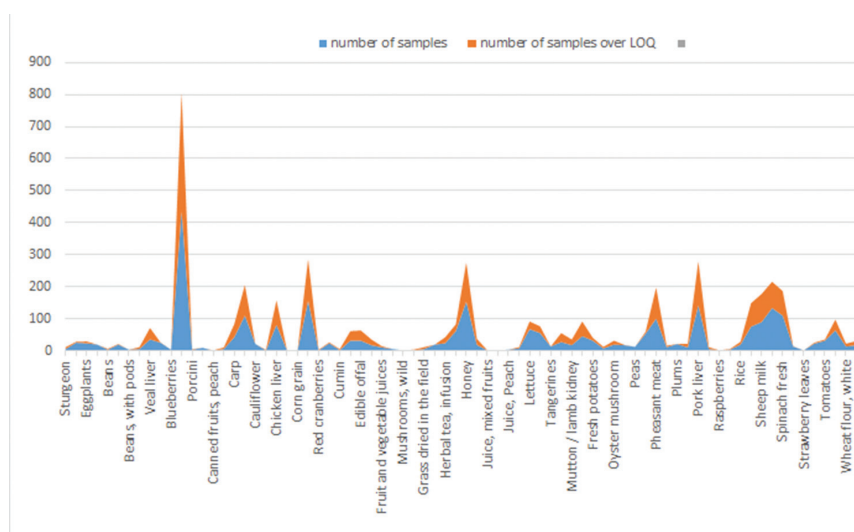
Food was found to be one of the main sources of lead exposure. In Bulgaria, the consumption of bread and pasta is high. In children of all ages, the bread consumed is half the amount of cereals



**Figure 2.** Results for the content of heavy metals in the tested food samples



**Figure 3.** Results of compliance of the tested samples with the established maximum levels for heavy metals in food and samples with a value above the limit of quantification



**Figure 4.** Results of compliance of the tested samples with the established maximum levels for heavy metals in food and samples with a value above the limit of quantification

consumed, the average daily consumption of vegetables and milk is also slightly high and that of fish. Cereals, potatoes, leafy vegetables, milk, meat, offal, game consumption are the alimentary route of lead exposure [22].

The established amounts of tin (inorganic) in canned foods, other than beverages, canned fruits (peach and pineapple), fruit and vegetable juices, tomato paste were within the maximum allowable amounts.

In the studied fish samples, the amount of mercury corresponded to the determined maximum regulated amount of mercury in food.

The limit of quantification (LOQ) is the lowest amount of a substance that can be measured with certainty by standard laboratory tests [23, 24, 25, 26]. “The LOQ is calculated from the lowest concentration point taking into

account the recovery of internal standards and sample intake” [27].

The simultaneous presence of several heavy metals in the studied foods was found within the limits of quantification. Of all tested samples, 68.9% had a value above LOQ for more than two metals and trace elements.

For the samples of animal origin, 89.17% of the results were above LOQ, and for products of plant origin, 40.46% of the results were above LOQ for more than two metals and trace elements (Figure 3).

The most significant number of samples with a value above LOQ were found in samples of game meat, meat and offal of cattle, sheep, pigs and poultry, fish and bivalve molluscs, carrots, milk, honey, leafy vegetables (spinach), infusion of herbal tea, horse meat, pheasants and rabbit

meat.

All samples of sturgeon, beef kidney, veal liver, offal of farm animals, fish and other seafood, game birds, horse meat, mussels, mutton/lamb kidney, mutton/lamb liver, pork kidney, and rabbit meat were with values above LOQ. These products represented 20.77% of the tested foods and 8.56% of all samples.

With a value above LOQ were:

- 85.28% of the samples from wild boar,
- 95.34% from the samples from carp,
- 87.27% from the samples from carrots,
- 95.06% from the samples from chicken liver,
- 83.97% from the samples from cow's milk,
- 93.75% of the samples duck liver,
- 79.16 5 of the samples herbal tea infusion,
- 79.22% of the samples honey, 35.29% of the lettuce samples,
- 37.5% of the samples potatoes, 96.03% from pheasant meat samples,
- 97.18% from pork liver samples,
- 45% from rice samples, 96.05% from salmon and trout samples salmo spp.,
- 74.44% from sheep milk samples,
- 61.19% from spinach samples,
- 53.12% of the deer meat samples (Figure 4).

## Discussion

It should be noted that children take more metals than adults, in the following order: Mn> Zn> Cu> Sr> Cr> Ni> As> Cd> Pb> Co> Sb. Lead is the most bioaccumulated metal of mussels and fish, along with cadmium. The sequence of accumulation of metals in different tissues of fish is established in the following order: liver> gills> gonads> muscles [28, 29, 30, 31, 32].

The study, covering six years, showed a generally low level of contamination of the studied foods with heavy metals.

Non-compliance with the maximum levels for heavy metals in foods for cadmium and lead alone was established.

The content of heavy metals in foods of animal origin was higher than in foods of plant origin.

More metals accumulate in wild boar meat, cow's milk, fish, and other seafood. It is important to note that frequent consumption of these foods can increase the amounts of heavy metals, which is important for public health.

In general, no significant contamination with heavy metals of the foods offered on the market in Bulgaria was established.

## Conclusion

Chronic exposure to heavy metals through food intake can cause adverse health effects.

A risk assessment should be carried out so that actions and measures can be taken to reduce exposure levels and achieve safe exposure levels.

The results of the analysis can be used to assess the risk to human health associated with the detected content of heavy metals and serve in planning the monitoring and control of heavy metals in foods.

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