

## MONITORING THE LEAD CONTAMINATION OF FOOD PRODUCTS OF NON-ANIMAL ORIGIN IN DIFFERENT REGIONS FROM ROMANIA IN 2019

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### Abstract

*The monitoring of lead contamination of food products of non-animal origin started in Romania in 2006, at the same time with entry into force of The European Commission Regulation (EC) no. 1881/2006 which regulates the maximum allowed values for this contaminant. In this study, samples that were collected and analyzed to determine lead contamination in 2019 have been analyzed by graphite furnace atomic absorption spectrometry (GFAAS). The samples came from Bucharest and from 12 other counties of the country. The cereals, wine, fruit, vegetables and mushrooms samples present a weaker lead contamination, compared to the fruit juice samples. Considering the results, all of the analyzed samples framed within recommended values regarding the lead contamination, and the average values falls within those of the European Union.*

**Key words:** lead, food safety, contamination.

### INTRODUCTION

Lead is a contaminant that commonly appears in the environment as a result of different types of industrial activity (Tiwari and Tripathi, 2012; Tiwari et al., 2013).

The accumulation of lead in water and soil depends on many factors such as pH, mineral composition and the type of organic material in which these contaminants are found (Khan and Ghouri, 2011). The lead found in soil, based on natural cycle, may be transferred in different types of food (vegetables, fruit, cereals, etc.)

Since 2006, The International Agency for Research on Cancer (IARC) has put lead in the 2A group, meaning the group of substances which probably have a carcinogenic effect on humans.

People's exposure to lead is made through food, cigarettes, water, soil, garbage and air

(WHO, 1996; Russell, 1989; Philip and Gerson, 1994).

Lead is found under both forms: organic and anorganic lead. In the environment it is found especially under the form of anorganic lead. The fact that organic forms are much more toxic than the anorganic ones is well-known (EFSA, 2012). According to IARC, the organic lead compounds are converted partially in anorganic lead compounds. (EFSA, 2012).

Lead accumulates especially in the bone tissue, being gradually released from there into the bloodstream during physiological or pathological periods of demineralisation such as gestation, lactation and osteoporosis (EFSA 2013). Lead can be transferred from mother to fetus/baby during both pregnancy and lactation. Lead affects the functioning of all systems in the body, from the circulatory system to the immune and reproductive systems (Araki et al., 1986; Papanikolaou et al., 2005).

Food is a source of lead exposure, and the main technique of determination is represented by the atomic absorption with flame or graphite furnace.

The absorption of lead in the digestive tract depends in particular on the physiological characteristics of the host as well as the characteristics of the material that can be ingested or inhaled. The absorption of lead by inhalation is higher. The absorbed lead is transported through the bloodstream and initially reaches the soft tissues such as the liver, kidneys and later the bone tissue where it accumulates with age. The half-life of lead in the blood is about 30 days and in the bone tissue it can reach up to 10 years (Landrigan et al., 1990; De Haro et al., 2001; EFSA, 2013; CodexSTAN, 1995). The lead excretion is mainly through urine and feces.

During the last years, a decrease in lead food contamination has been observed, most likely due to the increasing use of unleaded fuels as well as due to various environmental protection actions (EFSA, 2012). Regarding food, the highest concentration of lead is found in organs, then in beverages, bread and vegetables.

It is already known that the packaging or the packaging system, in gas mixtures used for packaging in controlled atmosphere are used, can influence the degree of contamination of food products (Petcu et al., 2014; Vişoescu et al., 2015). Studies are available on monitoring food contamination with mercury, iron, antibiotic residues, mycotoxins, deoxynivalenol, pesticides, genetically modified organisms) (Marin et al., 2013; Goncearov et al., 2015; Petcu et al., 2019; Pogurschi et al., 2015). The monitoring of lead contamination in our country is performed according to the Surveillance and Control Program in the field of Food Safety approved by the Order of the President of ANSVSA No. 35/2016 with subsequent amendments and completions. The monitoring of food safety is needed in order to be able to guarantee the food safety (Savu et al., 2002; Petcu, 2006).

This study aims to monitor the lead contamination of various foods of non-animal origin in 2019. The samples were collected from different counties of the country and

Bucharest, being analyzed in an accredited laboratory in Romania.

## MATERIALS AND METHODS

In 2019, a number of 288 samples represented by non-animal food products (cereals, fruit, vegetables, mushrooms, fruit juices and wine) were analyzed by GFAAS in an accredited laboratory in Romania (Figure 1).

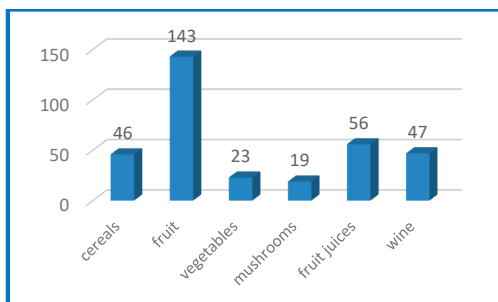


Figure 1. Types of analyzed samples in order to determinate lead residues - 2019

The samples came from Bucharest and from 8 other counties of the country.

Sampling and methods of analysis play a decisive role in determining lead contamination of foodstuff. The sampling and analysis criteria were first established in the European Union (EU) by EC Directive no. 2001/22/EC which was subsequently replaced by EC Regulation no. 333/2007 on sampling and analysis methods for the official control of lead, cadmium, mercury, 3MCPD (3-monochloropropane-diol) and bezopiren in foodstuff.

In order to protect public health, at the level of the European Union maximum limits have been established for lead in various foods, both of animal and non-animal origin by European Commission Regulation no. 1881/2006, with subsequent amendments and completions.

The maximum permitted limits have been set by reference to the edible part of a product, while the product undergoes or transforms by processing, dilution, etc., this aspect can be taken into account while reporting and interpreting the results.

All maximum permissible limits for lead are expressed in milligrams/kilogram (mg/kg), and the performance criteria and analysis

methodology are in EC Regulation no. 333/2007, can be modified and completed afterwards.

The analytical technique used was atomic absorption with graphite furnace (GFAAS), and the performance criteria corresponded to those of EC Regulation no. 333/2007 with subsequent amendments and completions, respectively: detection limit (LOD), quantification limit (LOQ), recovery, reproducibility, uncertainty.

The following definitions are used under this Regulation:

*The limit of detection* is the lowest measured content, from which the presence of the analyte can be deduced with reasonable statistical certainty. The limit of detection is numerically equal to three times the standard deviation of the mean of the control determinations ( $n > 20$ ).

The limit of quantification is the lowest analyte content that can be analyzed with reasonable statistical certainty. If both the precision and the accuracy are constant for a range of concentrations around the limit of detection, then the limit of quantification is numerically equal to six to ten times the standard deviation of the mean of the control determinations ( $n > 20$ ).

*The repeatability* is the value below which the absolute difference between the results obtained in the individual tests under repeatability conditions (for example, the same sample, the same operator, the same apparatus, the same laboratory and in a short period of time) is expected to be within a certain probability interval (usually 95%) and therefore  $r = 2.8 \times sr$ .

"Sr" - the standard deviation calculated from the results generated under repeatability conditions.

"RSDr" - the relative standard deviation calculated from the results generated under repeatability conditions.

"sR" - the standard deviation calculated from the results generated under reproducibility conditions.

"RSDR" - the relative standard deviation calculated from the results generated under reproducibility conditions.

*The reproducibility* is the value below which the absolute difference between the results obtained in individual tests under

reproducibility conditions (for example, on identical material obtained by operators in different laboratories using the standardized test method) is expected to be within a certain range of probability (usually 95%);  $R = 2.8 \times sR$ .

*The uncertainty* is the extended measurement uncertainty, using a coverage factor 2 gives a confidence level of approximately 95% ( $U = 2u$ ).

The reagents used (hydrogen peroxide, nitric acid, matrix modifier) are represented by ultrapure reagents in which the concentration of heavy metals is very low so as not to influence the results, in the sense of the appearance of false positive results.

Some studies have highlighted the existence of lead contaminants in fish samples (Hristov and Kirin, 2014).

## RESULTS AND DISCUSSIONS

### *Results and discussions on lead contamination of grain samples*

The monitored cereals regarding lead contamination are those provided in the Surveillance and Control Program in the field of food safety approved by the Order of the President of ANSVSA no. 35/2016 and for which maximum limits are set according to EC Regulation no. 1881/2006 with subsequent amendments and completions.

In 2019, a number of 46 wheat samples were analyzed.

For wheat, the maximum limit allowed according to EC regulation no. 1881/2006 is 0.2 mg/kg.

Out of a total of 46 samples, a number of 18 samples were reported with undetectable results, a number of nine samples resulted in 0.01 mg/kg, for three wheat samples. The results are: 0.02 mg/kg, two samples had the value of 0.03 mg/kg, one sample had the value of 0.04 mg/kg, and 13 samples had values lower than the quantification's limit of the method, respectively 0.007 mg/kg.

The results obtained from the analysis of lead residues coming from the wheat samples are presented in Figure 2.

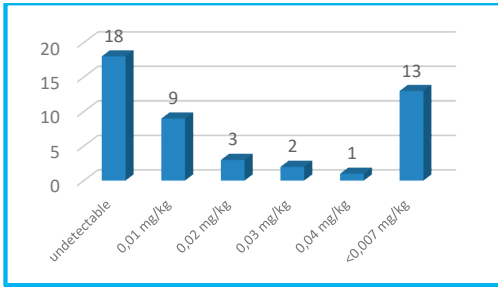


Figure 2. Results of lead contamination of cereals samples

*Results and discussions on lead contamination of fruit samples*

In 2019, 143 apple samples were analyzed in the study. A number of 76 samples had the result expressed as undetectable, while a number of 32 samples had the value of 0.01 mg/kg. For 18 samples the value of 0.02 mg/kg was registered, for nine samples the value of 0.03 mg/kg was registered, for two samples the value of 0.04 mg/kg was registered, and for six samples the value was < 0.007 mg/kg.

According to Reg. Ec. 1881/2006, the maximum allowed limit for lead residues in fruits is 0.1 mg/kg.

In the Figure 3 are presented the results obtained in the present study, for the determination of lead residues in apple samples.

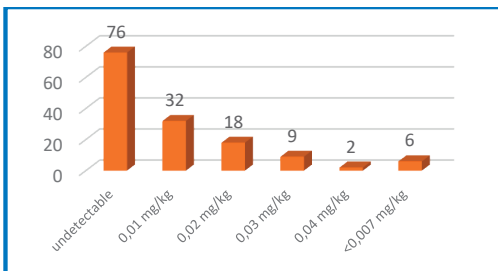


Figure 3. Results of lead contamination of apple samples

*Results and discussions on lead contamination of vegetables samples*

A number of 23 samples of fresh vegetables were analyzed, represented by carrots, eggplants, potatoes, cucumbers, cabbage and

celery. For a number of eight samples, the results were reported with undetectable values, in ten samples the value of 0.01 mg/kg was recorded, in one sample the value of 0.02 mg/kg was recorded, in one sample the value of 0.03 mg/kg was recorded, and in two samples the value of 0.05 mg/kg was registered. A value of <0.007 mg/kg, the limit of quantification of the method, was recorded in one sample.

The maximum permissible limit for lead, according to Reg. Ec. 1881/2006 for vegetables, except *Brassica*, fresh leafy vegetables and herbs is 0.1 mg/kg, and for *Brassica* and leafy vegetables is 0.3 mg/kg. For potatoes, the maximum level applies to peeled potatoes.

Figure 4 shows the graphical distribution of the results of lead contamination of the vegetable samples analyzed during the study.

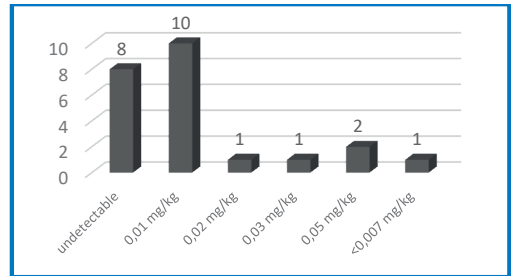


Figure 4. Results of lead contamination of vegetables samples

*Results and discussions on lead contamination of mushroom samples*

In the EC Regulation no. 1881/2006, subsequently amended and supplemented, the maximum permitted limits for cultivated mushrooms are set at a maximum level of 0.3 mg/kg wet weight.

In 2019, a number of 19 mushroom samples were analyzed, represented in about 90% of mushroom cases. For a number of nine samples, undetectable values of lead residues were obtained, and for the rest of the samples different values were reported according to Figure 5.

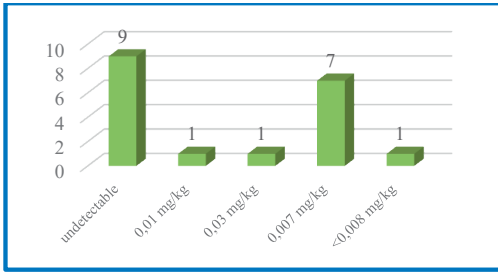


Figure 5. Results of lead contamination of mushrooms samples

*Results and discussions on lead contamination of fruit juice samples*

The maximum permitted limit provided in EC Regulation no. 1881/2006, with subsequent amendments and completions, is different depending on the types of fruit used. Thereby, for fruit juices, reconstituted concentrated fruit juices and fruit nectars, the accepted value is a maximum of 0.05 mg/kg.

Out of the total 56 samples of fruit juices analyzed in 2019, a number of 15 samples were reported with undetectable values, and the rest of the samples registered different values according to Table 1 and Figure 6.

Table 1. Results of lead residues from fruit juice samples

Year	Samples with undetectable results	Samples with 0.01 mg/kg	Samples with 0.02 mg/kg	Samples with 0.05 mg/kg	Samples with 0.007 mg/kg
2019	15	20	13	1	7

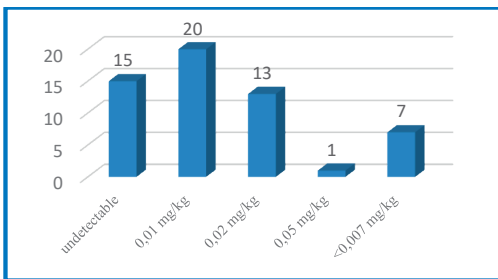


Figure 6. Results of lead contamination of fruit juice samples

*Results and discussions on lead contamination of wine samples*

Out of the total 47 wine samples analyzed during the study, for 29 of them the result was undetectable. The highest value was recorded in three samples for which the value of the

results obtained was 0.05 mg/kg, within the limits of legal acceptability.

In Table 2 and Figure 7 present distribution of the results concerning lead contamination of the analyzed grape wine samples.

Table 2. Results of lead residues from the wine samples

Year	Samples with undetectable results	Samples with 0.01 mg/kg	Samples with 0.02 mg/kg	Samples with 0.03 mg/kg	Samples with 0.05 mg/kg	Samples with 0.007 mg/kg
2019	29	6	2	2	3	5

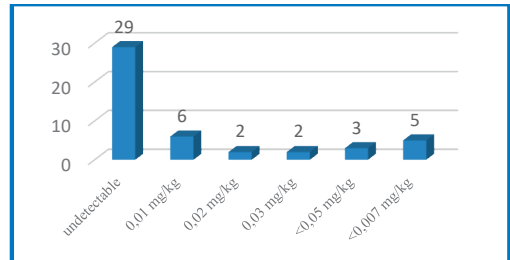


Figure 7. Results of lead contamination of wine samples

The maximum allowed limit provided in the EC Regulation no. 1881/2006, with subsequent amendments and completions, is 0.2 mg/kg for wines obtained from the fruit harvested during 2001 and 2015 and 0.15 mg/kg for wines obtained from the harvest starting with 2016. In the European regulation are defined 2 wine categories for which maximum permitted limits are set.

**CONCLUSIONS**

From the total samples of cereals analyzed, 39.13% had results expressed as undetectable, and the rest of the samples identified values that are much lower than the maximum allowed value.

Undetectable values were obtained at the analysis of 53.15% of apple samples, and at a percentage of 1.39% samples had two times lower results compared to the maximum allowed value, and 5.59% samples recorded values 10 times lower than the maximum allowed limit.

In the analysis of vegetables, 34.78% of the samples did not contain lead residues at a detectable level by the analysis method performed, and 43.48% of the samples recorded 10

times lower values compared to the maximum allowed limit.

Almost half of the analyzed mushroom samples (47.37%) recorded an undetectable level.

Regarding the fruit juice, the lowest number of samples with undetectable level was recorded, in 26.78% of samples.

61.70% of the wine samples had undetectable content, and for some samples, even 10 times lower values than the maximum accepted average was identified.

The samples of cereals, wine, fruits, vegetables and mushrooms showed lower lead contamination compared to the samples of fruit juice.

All of the analyzed samples corresponded in terms of lead contamination.

Finally, we could say that the level of lead contamination of non-animal food products in Romania falls within the provisions of current legislation is linked to the increasingly present policy at national and international level to reduce the degree of lead contamination by effectively applying various social and economical measures. One of the most well-known measures is the gradual giving up on the usage of fuels based on lead. The fact that lead contamination has as an important source a series of industries (battery production, extraction of various ores, burning of fuels containing lead) causes pollution of water, atmosphere and different crops of cereals, vegetables and fruits which are the main sources of food.

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