# HEAVY METALS CONTAMINATION OF FRUITS AND VEGETABLES IN EUROPE

## CONTAMINAREA FRUCTELOR ȘI LEGUMELOR CU METALE GRELE ÎN EUROPA

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Abstract. Heavy metals are natural compounds of the soil and due the human activities and some natural causes, their levels exceed the legislative threshold values in some agricultural soils, becoming harmful for human health and environment. At the level of the European Union there are laws which regulate the threshold values of heavy metals in agriculture soils and in fruits and vegetables. Unfortunately, in markets there are still plant products of whose heavy metals (or other contaminants) content exceeds the maximum allowable levels. Contamination of food crops with heavy metals is mainly due to the crops growth in polluted soils and the excessive use of plants protection products with heavy metals. Thus, their short- or long-term consumption can pose risk to human health. In this context, the purpose of this paper is to identify, based on published studies, the content of cadmium, lead, copper and zinc in fruits and vegetables grown in different parts of Europe or sold on the markets of European Community countries. According to the results of published studies, the fruits and vegetables grown in certain areas of Europe, including in some areas of Romania, may be harmful to human health, the cadmium, lead, copper and zinc levels exceeding the maximum permitted limits. Key words: heavy metals, human health effects, food crops, soil contamination

Rezumat. Metalele grele sunt compuși naturali ai solului, dar ca urmare a activitătilor umane și a unor fenomene naturale nivelurile acestora ajung să depăşească valorile admisibile în solurile agricole și devin, astfel, un pericol pentru sănătatea umană și mediu. La nivel European există acte normative prin care sunt reglementate nivelurile admisibile a metalelor grele în solurile agricole si a continutului acestora în fructe si legume, dar pe piata de desfacere există încă produse de origine vegetală care conțin metale grele în concentrații peste limitele admisibile. Contaminarea produselor vegetale cu metale grele este cauzată în principal de creșterea culturilor în soluri poluate și de utilizarea excesivă a produselor de protecție a plantelor cu metale grele. Astfel, consumul lor pe termen scurt sau lung poate reprezenta un risc pentru sănătatea umană. În acest context, scopul acestei lucrări este de a identifica, pe baza studiilor publicate, conținutul de cadmiu, plumb, cupru și zinc din fructele și legumele cultivate în diferite părți ale Europei sau vândute pe piețele țărilor din Comunitatea Europeană. Conform rezultatelor studiilor publicate, fructele și legumele cultivate în anumite zone ale Europei, inclusiv în unele zone din

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România, pot fi dăunătoare sănătății umane, nivelurile de cadmiu, plumb, cupru și zinc depășind limitele maxime permise. **Cuvinte cheie:** metale grele, efecte asupra sănătății umane, culturi agricole, contaminarea solului

#### **INTRODUCTION**

The presence of cadmium, lead, copper and zinc in soil, water and atmosphere in various levels cause negative effects on all life forms. Due to their properties, these metals end up accumulating in plants, animals and human body (Fig. 1). Heavy metals bioaccumulation in plants and organism's tissues result in damage of cellular structures and cytoplasmic enzymes, inhibit protein synthesis and in the human body usually replace the vital nutrients from enzymes, proteins, nucleic acids, thus disrupting their biological functions. Long-term exposure to cadmium and lead can induce also genetic mutations or cancer. The highest quantity of heavy metals penetrates the human body through ingestion of contaminated food and water, so the control of heavy metals content in food and water becomes foremost for human health protection (Roșca *et al.*, 2017; 2019). In this context, the purpose of this paper is to identify, based on published studies, the content of cadmium, lead, copper and zinc in fruits and vegetables grown in different parts of Europe or sold in the markets of the European Community countries.

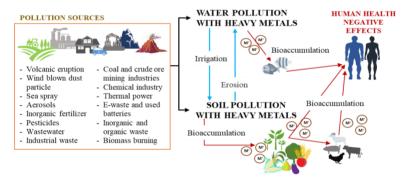


Fig. 1 Sources of water and soil pollution with heavy metals pollutants and their penetration into the human body

#### HEAVY METALS LEVELS IN EUROPEAN AGRICULTURE SOIL

Contamination of food crops with heavy metals is mainly due to the growth of plants in polluted soils and the excessive use of plants protection products with heavy metals. According to Toth *et al.* (2017) in Europe, the concentrations of cadmium, lead, copper and zinc in the soil vary from region to region. For example, Ireland and Greece are the regions with the highest cadmium concentration in soil and the agricultural lands of France, Italy, Portugal and

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Romania are particularly affected by copper pollution. In the soil samples taken from Central Italy, France, Germany and the UK, the highest values of Pb concentrations were detected and in more than 20% of the samples taken from different parts of Europe, the zinc concentrations are above the threshold values (Toth et al., 2017). In agricultural lands of Romania, based to the information provided by Dumitru et al. (2016) the highest concentrations of Cd. Pb and Cu were detected in pastures lands (1.6 mg Cd/kg, 335 mg Pb/kg and 551 mg Cu/kg) and the highest zinc concentration was found in arable lands (974 mg Zn/kg). Since the levels of heavy metals in soils vary from region to region and depend on human activities, but also on some natural causes. Table 1 shows the levels of cadmium, copper, lead and zinc detected in different areas of Romania.

Table 1

| Heavy metals levels in some areas from Romania                    |                              |                   |                   |                       |  |
|---|------------------------------|-------------------|-------------------|-----------------------|--|
| Area  | Heavy metals content (mg/kg) |                   |                   |                       |  |
|   | Cd                           | Cu                | Pb                | Zn                    |  |
| Banat area of Southern<br>Carpathians <sup>1</sup>                | 0.17-2.08                    | 24.67 <b>-231</b> | 14.77 <b>-136</b> | 130.67 <b>-359.33</b> |  |
| Ilfov region <sup>2</sup>   | 0.300-0.532                  | -                 | 9-37              | -                     |  |
| NE area of Vaslui county <sup>3</sup>                             | 0.02-0.8                     | 14- <b>300</b>    | 16-84             | 31-192                |  |
| Baia Mare area <sup>4</sup>                                       | 15.84                        | 3165.26           | 3288.98           | 1989.12               |  |
| Baia Sprie area <sup>4</sup>                                      | 26.84                        | 4020.85           | 4062.30           | 3218.88               |  |
| Simleul Silvaniei area <sup>4</sup>                               | 0.19                         | 762.52            | 8.49              | 59.99                 |  |
| In the vicinity of an Industrial area near Bucharest <sup>5</sup> | 7                            | 350               | 750               | 1300                  |  |
| Alert threshold values for<br>agriculture soil <sup>6</sup>       | 3                            | 100               | 50                | 300                   |  |
| Intervention threshold values<br>agriculture soil <sup>6</sup>    | 5                            | 200               | 100               | 600                   |  |

The bolded values exceed the intervention threshold values; <sup>1</sup>Manea et al. (2020); <sup>2</sup>Mustătea et al. (2019); <sup>3</sup>Unqureanu et al. (2017); <sup>4</sup>Bora et al. (2020); <sup>5</sup>Velea et al. (2009); Order no. 756 (1997)

# LEVELS OF HEAVY METALS IN FRUITS AND VEGETABLES

It is important to avoid as much as possible food contaminated with heavy metals since its consumption could cause negative effects to human health. Based on the state-of-the art, within Europe markets, there are still plant products of whose heavy metals content exceeds the allowable levels (Stančić et al., 2016; Cherfi et al., 2016; Rusin et al., 2021). Thus, in table 2 are shown the concentrations of cadmium, copper, lead and zinc detected in fruits and vegetables grown in different parts of Europe or sold on the markets of European Community countries. The results presented in table 2 show that in most cases the concentrations of heavy metals exceed the allowed limit values.

In order to protect human health, WHO recommend a provisional tolerable monthly intake value for Cd of 25 µg/kg bw/month and a provisional maximum tolerable daily intake of 0.5 mg/kg bw for Cu and 0.3-1 mg/kg bw for Zn. For Pb, in 1999, WHO established the value of 0.025 mg/kg bw as provisional tolerable

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weekly intake, but in 2011 this value was withdrawn and it was not established another value as health protective value (WHO, 2021). For the safe consumption of food, the European Community set maximum permissible levels (MRL) for each heavy metal in each product category. Table 3 provides the MRL of cadmium, copper, lead and zinc in fruits and vegetables. Thus, fruits and vegetables with a level of heavy metals above the MRL values, must be removed from the market and their consumption is prohibited.

Table 2

| Levels of heavy metals in fruits and vegetables from Europe             |                           |                               |                   |                    |  |  |
|---|---------------------------|-------------------------------|-------------------|--------------------|--|--|
| Food crop   | Heavy metal concentration |                               |                   |                    |  |  |
|   | Cd                        | Cu                            | Pb                | Zn                 |  |  |
| Varaždin City Market, Croatia <sup>1</sup> (mg/kg d.w.)                 |                           |                               |                   |                    |  |  |
| White potato  | 0.13-0.34                 | 0.10-0.24                     | 0.30-0.40         | 41.0-94.5          |  |  |
| Red potato  | 0.16-0.46                 | 5.4-6.4                       | 0.37-1.03         | 56.5-64.0          |  |  |
| Onion   | 0.17-0.34                 | 3.6-12.3                      | 0.16-0.18         | 28.0-47.0          |  |  |
| Carrot  | 0.27- <b>0.69</b>         | 6.0-9.4                       | 0.54-0.94         | 51.0-75.5          |  |  |
| Beans   | 0.04-0.05                 | 7.0-11.1                      | 0.08-0.52         | 58.5-91.0          |  |  |
| Lettuce   | 0.97-1.52                 | 8.1-11.8                      | 0.82-2.22         | 59.5-139.5         |  |  |
| Vegetables collected  | from different su         | permarkets in La              | Rochelle, France  |                    |  |  |
| Potato  | -                         | 4.14                          | -                 | 1.78               |  |  |
| Onion   | -                         | 6.19                          | -                 | 2.93               |  |  |
| Carrot  | -                         | 3.4                           | -                 | 3.98               |  |  |
| Tomato  | -                         | 4.62                          | -                 | 2.28               |  |  |
| Cabbage   | -                         | 4.35                          | -                 | 3.85               |  |  |
|   | inat area of Southe       |                               |                   |                    |  |  |
| Parsley leaf  | 0.03-0.09                 | 1.03-4.79                     | 0.28- <b>1.97</b> | 9.13-10.44         |  |  |
| Cabbage   | 0.01- <b>0.12</b>         | 0.45-2.77                     | 0.05- <b>0.90</b> | 3.28-16.30         |  |  |
| Lettuce   | 0.02-0.09                 | 0.76-2.22                     | 0.08- <b>0.62</b> | 5.14-14.46         |  |  |
| Carrot root   | 0.01-0.08                 | 0.73-1.77                     | 0.04- <b>2.11</b> | 2.07-4.93          |  |  |
| Parsley root  | 0.010.20                  | 1.23 <b>-6.87</b>             | 0.08- <b>2.45</b> | 3.50- <b>45.83</b> |  |  |
| Onion   | 0.01-0.06                 | 0.23-1.37                     | 0.04-0.50         | 0.77-10.90         |  |  |
| Potato  | 0.02-0.05                 | 1.94- <b>7.36</b>             | 0.25- <b>1.06</b> | 2.35-6.02          |  |  |
| Cucumber  | 0.03 <b>-0.15</b>         | 0.47-2.40                     | 0.16- <b>0.72</b> | 0.93-8.97          |  |  |
| Tomato  | 0.02-0.05                 | 0.89-4.22                     | 0.18- <b>0.85</b> | 2.09- <b>9.34</b>  |  |  |
| Peach   | 0.02-0.04                 | 1.44 <b>-8.61</b>             | 0.12- <b>0.77</b> | 1.07- <b>8.09</b>  |  |  |
| Plum  | 0.02-0.03                 | 0.48-4.68                     | 0.10-0.49         | 1.19- <b>6.08</b>  |  |  |
| Apple   | 0.02-0.04                 | 0.82-4.91                     | 0.11- <b>0.56</b> | 1.17-4.34          |  |  |
| Pear  | 0.02-0.03                 | 0.87-4.87                     | 0.11- <b>0.56</b> | 0.97-4.08          |  |  |
| Grape   | 0.02-0.03                 | 1.50 <b>-7.71</b>             | 0.12- <b>0.61</b> | 0.98- <b>7.11</b>  |  |  |
|   | ithin and nearby o        |                               |                   |                    |  |  |
| Tomato  | 0.1 <b>-0.2</b>           | 11.50-68.8                    | 0.40-3.60         | 21.7-144.4         |  |  |
| Zucchini  | 0                         | 3.7-16.1                      | 0.16-2.3          | 13.5-140           |  |  |
| Fruits and vegetables from the Polish markets <sup>5</sup> (mg/kg f.w.) |                           |                               |                   |                    |  |  |
| Fresh raspberry   | 0.003-0.021               | -                             | 0.003-0.033       | -                  |  |  |
| Fresh beetroot  | 0.022-0.67                | -                             | 0.056-0.135       | -                  |  |  |
| Fresh celery  | 0.0019-0.712              | -                             | 0.003-0.074       | -                  |  |  |
| Fresh carrot  | 0.024-0.062               | -                             | 0.02-0.041        | -                  |  |  |
|   |                           | ion <sup>6</sup> (mg/kg f.w.) |                   |                    |  |  |
| Lettuce   | 0.65-1.92                 | -                             | 0.002-10.05       | -                  |  |  |
| Spinach   | 1.66                      | -                             | < 0.002           | -                  |  |  |

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|--|--|-------------|-------------------|------------|--|--|
| Radishes   | 0.11- <b>0.26</b>  | -           | < 0.002           | -          |  |  |
| Nonferrous   | Nonferrous metallurgical industrial areas in Romania <sup>7</sup> (mg/kg f.w.) |             |                   |            |  |  |
| Potato   | 0.01-0.1   | 1.37-3.17   | 0.04-0.11         | 3.9-9.3    |  |  |
| Carrot   | 0.03- <b>0.37</b>  | 0.65-1.01   | 0.11- <b>0.54</b> | 0.01-0.93  |  |  |
| Yellow onion   | 0.01- <b>0.3</b>   | 0.51-0.55   | 0.01-0.03         | 0.03-0.78  |  |  |
| Baia Mare*, Baia Sprie** and Simleul Silvaniei*** areas, Romania <sup>8</sup> (mg/kg f.w.) |  |             |                   |            |  |  |
| Grapes*  | 0.58-0.75  | 8.49-13.30  | 4.60-6.34         | 8.52-10.13 |  |  |
| Grapes**   | 0.88-1.22  | 12.31-14.51 | 6.74-8.91         | 6.78-8.91  |  |  |
| Grapes***  | 1.09-1.30  | 2.31-3.38   | 0.40- <b>0.59</b> | 1.11-1.25  |  |  |

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<sup>1</sup>Stančić et al. (2016); <sup>2</sup>Cherfi et al. (2016); <sup>3</sup>Manea et al. (2020); <sup>4</sup>Antisari et al. (2015); <sup>5</sup>Rusin et al. (2021); <sup>6</sup>Mustățea et al. (2019); <sup>7</sup>Nedelescu et al. (2015); <sup>8</sup>Bora et al. (2020); The bolded values exceed the maximum permissible levels

Table 3

| MRL values of cadmium, copper, lead and zinc in fruits and vegetables |
|---|
| (EC Regulation 1881, 2006 and EC Regulation 629, 2008)                |

| Product   |      | MRL (mg/kg f.w.) |     |    |  |
|---|------|------------------|-----|----|--|
|   |      | Cu               | Pb  | Zn |  |
| Fresh and frozen vegetables and fruit, excluding leaf<br>vegetables | 0.1  | 5                | 0.5 | 15 |  |
| Stem vegetables and root vegetables                                 | 0.2  | -                | 0.5 | -  |  |
| Dehydrated vegetables and fruits                                    | 0.5  | 50               | 3   | 50 |  |
| Potatoes  | 0.1  | 3                | 0.3 | 10 |  |
| Fresh and frozen fruits   | 0.05 | 5                | 0.5 | 5  |  |
| Leaf vegetables   |      |                  |     |    |  |

### CONCLUSIONS

Despite the fact that considerable efforts are made at the European level to avoid the consumption of food contaminated with heavy metals, special attention still needs to be paid to the trade of food crops obtained in various parts of Europe.

According to the results presented in this study, the fruits and vegetables grown in certain areas of Europe, including in some areas of Romania, are harmful to human health, cadmium, lead, copper and zinc levels exceeding the maximum permitted limits. In order to avoid the contamination of fruits and vegetables with heavy metals, it is important to identify the polluted sites and to carry out studies in order to establish the maximum permitted concentrations in soil at which specific crops accumulate the metals in their parts below the allowed limit.

This measure is recommended because there are plants that, even if are grown in soils with heavy metals below the allowed levels, in their plant parts they accumulate quantities above the allowable limits. By setting of the allowable limits for each type of crop, the risk of heavy metals on human health can be reduced.

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