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ANALYSIS OF POTENTIAL CARCINOGENIC HEAVY METALS IN FRUITS AND VEGETABLES WITH IMPACT ON THE MARKETS OF BUCHAREST-ILFOV REGION

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Abstract

Bucharest-Ilfov region is formed by Bucharest – Romania's capital and Ilfov County, being located in the southern of Romania, in the center of Romanian Plain, representing the largest economical agglomeration of the country. Bucharest-Ilfov region is a place where vegetables are intensively grown, in 2014, in here being cultivated 2.25% of whole country vegetables. Heavy metals contamination of plant origin food is made through soil and atmosphere, especially when cultures are nearby enterprises, municipal waste waters, high traffic streets, etc.

Analysis of vegetables and fruits commonly consumed in the Bucharest-Ilfov region, with direct impact on the markets in this area, especially in Bucharest was followed. Fruits and vegetables cultivated in Ilfov county as well as surrounding counties (Dambovita, Giurgiu, Ialomita) where purchased from food markets in the southern of Bucharest (3 types of fruits: apples, plums, pears - 6 samples and 9 types of vegetables: potatoes, tomatoes, cabbage, carrot, onion, red onion, yellow bell pepper, red bell pepper and green bell pepper - 15 samples). For these products was analyzed the content of potential carcinogenic heavy metals: Pb, Cd, Cr, As and Ni, by inductively coupled plasma mass spectrometry (ICP-MS).

Obtained results were compared with maximum levels stated by (EC) Regulation 1881/2006, Codex STAN 193-1995 (FAO and WHO) as well as GB 2762-2012 Standard. Lead, cadmium and arsenic content of all analyzed samples was very low, most of the values being below the detection limit of the equipment. Chromium content was higher compared to other metals; however, the method used determined the total chromium content, which excludes the hypothesis of a possible hexavalent chromium (Cr(VI)) contamination. Regarding nickel, the values obtained are higher for vegetables compared to fruits; however, there are no imposed limits.

Bucharest-Ilfov is a highly populated area of Romania and lot of producers came in this area. The conclusion of this research was that tested fruits and vegetables are not containing potentially carcinogenic heavy metals (Pb, Cd, Cr, As or Ni) above the imposed limits.

Key words: *Fruits, Vegetables, Heavy metals, Carcinogenicity.*

1. Introduction

Fruits and vegetables are important components of human diet as they are rich in proteins, carbohydrates, minerals, vitamins and fibers. Therefore, contamination of these products in general, and heavy metals contamination in particular, are ones of the most important aspects of the food quality assurance [1 - 5].

Main concern for human health regarding heavy metals contamination of fruits and vegetables is related to their capacity to accumulate in the edible parts of the plants and then enter in the food chain causing adverse toxicological effects for the consumers [6]. Plants take up heavy metals by absorbing them from contaminated soils, as well as from the part of the plants that came into contact with polluted air [5, 7 - 12].

During the last years, a large number of studies had shown heavy metals as important contaminants of fruits and vegetables [13 - 23]. All studies revealed that the main source of heavy metals contamination is their grown media (soil, air and nutrient solutions).

Arsenic (As) is a ubiquitous element that can pollute soil, water, plants including other compartments of the ecosystem and the environment, and ultimately affects human health [24]. Arsenic and inorganic arsenic compounds are class one carcinogens in International Agency for Research on Cancer (IARC) classification.

Cadmium (Cd) occurs naturally in the environment in its inorganic form, and anthropogenic sources have further contributed to background levels of cadmium in soil, water and vegetables as well [25]. Cadmium is a class one carcinogen in IARC classification.

Lead (Pb) also occurs naturally in the environment from anthropogenic activities, such as: mining, smelting and battery manufacturing [6]. Lead is a class two B carcinogen in IARC classification.

Nickel (Ni) is omnipresent and is vital for the function of many organisms, but concentrations in some areas from both anthropogenic release and naturally varying levels may be toxic to living organisms [26]. Nickel compounds are class one carcinogens in IARC classification.

Chromium (Cr) is an important element especially in metallurgical/steel or pigment industry. The main source of Cr pollution is considered to be from dyestuffs and leather tanning when wastes are discharged directly into waste streams [27]. Chromium (VI) compounds are class one carcinogens in IARC classification.

The European Union (EU) has set maximum levels (MLs) for lead and cadmium in fruits and vegetables based on produce class (Commission Regulation (EC) no. 1881/2006 with amendments). For lead the maximum level for vegetables (excluding leafy brassica, salsify, leaf vegetables & fresh herbs, fungi, seaweed and fruiting vegetables) and fruits (excluding cranberries, currants, elderberries and strawberry tree fruit) is 0.10 mg/kg wet weight while for leafy brassica, salsify, leaf vegetables excluding fresh herbs, fungi (*Agaricus bisporus*, *Pleurotus ostreatus*, *Lentinula edodes*) is 0.3 mg/kg wet weight. Codex Alimentarius - International Food Standards has set also maximum levels (MLs) for lead and cadmium in fruits and vegetables (Codex STAN 193-1995 - General Standard for Contaminants and toxins in food and feed, with amendments).

Bucharest-Ilfov region is the largest economic agglomeration of Romania, where are presented all industrial activities. The economic environment of Bucharest-Ilfov region is particularly attractive due to existing institutional structure, qualified workforce and the communication system more developed than in other regions, so that Foreign Direct Investments (FDI) attracted represents almost two thirds of all FDI nationwide. With this in mind, the purpose of this study was to analyze fruits and vegetables samples grown in Ilfov county and neighboring counties, commonly consumed in the Bucharest-Ilfov region, with direct impact on the markets in this area, particularly in Bucharest, in terms of content in potentially carcinogenic heavy metals, namely Pb, Cd, Cr, As and Ni.

2. Materials and Methods

2.1 Study area

The present study was carried out in Bucharest-Ilfov region, which is part of South-Muntenia region of Romania (number 3 on the map presented in Figure 1), by analyzing fruits and vegetables samples grown in Ilfov county and in the following neighboring counties: Dambovita, Ialomita and Giurgiu (Figure 1). Samples were purchased from 3 markets from south part of Bucharest, namely: Berceni Oltenitei, Berceni Sudului and Progresul market.

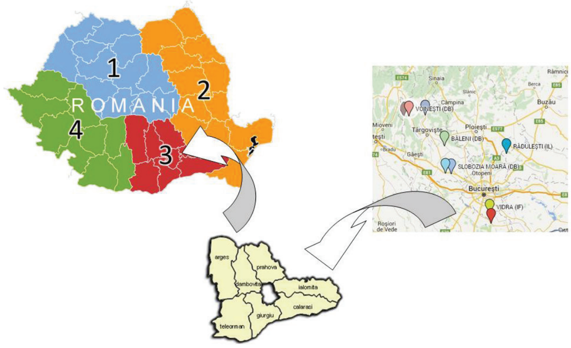

Figure 1. Study area of fruits and vegetables samples`

2.2. Sampling and analysis

Fruits (3 types of fruits: apple, plum and pear - 6 samples) and vegetables (6 types of vegetables: potato, tomato, cabbage, carrot, onion and bell pepper - 15 samples) samples were analyzed in terms of potential carcinogenic heavy metals content, namely Pb, Cd, Cr, As and Ni (Table 1).

Samples of fruits and vegetables (0.4 - 0.5 g each) were placed into 100 mL TFM (fluoropolymer) vessel and 9 mL of nitric acid (65%) and 1 mL hydrogen peroxide (30%) were added. The vessels was sealed and heated in a microwave digester (Ethos Easy Millestone) according to the program presented in Table 2.

For all samples, Pb, Cd, Cr, As and Ni contents were measured by inductively coupled-mass spectrometry (ICP - MS), using a quadruple inductively coupled plasma mass spectrometer NexION 300Q (Perkin Elmer, USA), equipped with low liquid uptake nebulizer, free-running RF plasma generator, automated X, Y, Z torch positioning, Triple Cone Interface and patented PlasmaLok® technology. All calibration curves showed good linearity (r > 0.998).

2.3. Quality control

Samples were carefully handled to avoid contamination. Deionized water was used for this study. Glassware used was properly cleaned and all reagents were of analytical grades. For the evaluation of the reliability

Table 1. Sample list

Sample		Grown place	Market
FRUITS			
Plum		Voinesti (Dambovita)	Berceni Oltenitei
		Pucioasa (Dambovita)	
Apple		Voinesti (Dambovita)	
		Candesti (Dambovita)	
Pear		Voinesti (Dambovita)	
		Candesti (Dambovita)	
VEGETABLES			
Potato		Radulesti (Ialomiţa)	Berceni Sudului
		Slobozia Moara (Dambovita)	
Tomato		Falastoaca (Giurgiu)	
		Vidra (Ilfov)	Progresul
Cabbage		Vidra (Ilfov)	
		Lunguletu (Dambovita)	Berceni Sudului
Carrot		Vidra (Ilfov)	Progresul
		Baleni (Dambovita)	Berceni Sudului
Onion		Vidra (Ilfov)	Progresul
		Baleni (Dambovita)	Berceni Sudului
Bell pepper		Baleni (Dambovita)	
		Vidra (Ilfov)	Progresul
		Falastoaca (Giurgiu)	Berceni Sudului
		Vidra (Ilfov)	Progresul
		Falastoaca (Giurgiu)	Berceni Sudului

Table 2. Sample digestion program

Temperature (^{o}C)	Power (W)	Time (min)	Reagents volume (ml)		Sample weight (g)
			HNO_3	H_2O_2	
T_1 - 200	P_1 - 1800	15	9	1	0.4 - 0.5
T_2 - 200	P_2 - 1800	15			
Cooling 15 min.					

of the analytical method were used two Standard Reference Materials: BCR®679 "White cabbage" and ERM®-BC084a "Tomato paste". Double samples were analyzed for each Standard Reference Material. The recovery rates ranged between 77.5% and 78% for Cd, 94% and 96% for Pb, 102% and 107% for Ni, and 107% and 115% for Cr.

3. Results and Discussion

3.1 Heavy metals content in fruits (apple, pear and plum)

The concentrations of lead in fruits samples were all below limit of detection (Table 3). The maximum level set by Commission Regulation (EC) no. 1881/2006 (last amended in April 2016) for lead in fresh fruits is 100 µg/kg.

The concentrations of cadmium in fruits samples (Table 3) ranged from a minimum below limit of detection to a maximum 7.25 µg/kg. All values were bellow maximum limit of 50 µg/kg set by Commission Regulation (EC) no. 1881/2006. The highest cadmium values were obtained for pear samples.

The chromium content of fruits was higher than all other elements. Two facts are to be taken into account: chromium was determined as total chromium content (thus higher values can be explained), and there are no maximum levels from chromium in the EU regulation.

Arsenic content in fruits ranged between 0.7 µg/kg and 3.3 µg/kg while the nickel content ranged between 85 µg/kg and 381 µg/kg. For these elements there are no maximum levels stated.

The overall content of the 5 analyzed heavy metals decrease in the following order: apple > plum > pear.

3.2 Heavy metals content in root vegetables (potato, carrot and onion)

The values for heavy metals in root vegetables (Table 4), as expected, were higher than in fruits and all other analyzed vegetables.

For lead and cadmium contents, all values were bellow limits of detection (100 µg/kg). One of the carrot samples had the lead content closed to the maximum level (98.29 µg/kg). Chromium content was higher than 500 µg/kg value, which is the maximum level stated by China Standard GB 2762-2012. The same mention as previous should be done: the values obtained are for total chromium. Arsenic content was also low, at levels below maximum level of 500 µg/kg stated by the same Chinese standard. Nickel content was higher than in fruits, for one the potato samples the value being of 1 mg/kg which may suggest a possible contamination.

The overall content of the 5 analyzed heavy metals decrease in the following order: potato > carrot > onion.

3.3 Heavy metals content in fruit and leaf vegetables (tomato, bell pepper and cabbage)

The concentrations of lead in fruit and leaf vegetables samples ranged from a minimum below limit of detection (Table 5) to a maximum 62.16 µg/kg. The highest values of lead content are observed for bell peppers. Two samples had a lead content higher than 50 µg/kg maximum level set by Codex STAN 193-1995 (last amended in 2015). However, Commission Regulation (EC) no. 1881/2006 set a maximum level for lead content in bell peppers of 100 µg/kg.

Cadmium content was slightly higher than in fruits, but lower than in root vegetables. All values were bellow maximum levels stated by Commission Regulation (EC) no. 1881/2006.

Table 3. Heavy metals content in fruits samples

No.	Sample	Grown place (Origin)	Lead content				Cadmium content				Chromium content				Arsenic content				Nickel content			
			Pb (µg/kg)	Maximum levels (µg/kg)			Cd (µg/kg)	Maximum levels (µg/kg)			Cr (µg/kg)	Maximum levels (µg/kg)			As (µg/kg)	Maximum levels (µg/kg)			Ni (µg/kg)	Max. levels (µg/kg)		
				EU	Cod	GB		EU	Cod	GB		EU	Cod	GB		EU	Cod	GB		EU	Cod	GB
P1	Plum	Voinesti (DB)	< LOD	100	100	100	0.71 ± 0.04	50	-	50	973.83 ± 61.97	-	-	-	3.24 ± 0.65	-	-	-	158.60 ± 8.73	-	-	-
P2		Pucioasa (DB)	< LOD				< LOD				978.24 ± 40.59				3.14 ± 0.63				118.09 ± 11.42			
P3	Apple	Voinesti (DB)	< LOD	100	100	100	1.46 ± 0.41	50	-	50	1247.66 ± 11.59	-	-	-	1.02 ± 0.21	-	-	-	32.19 ± 15.72	-	-	-
P4		Candesti (DB)	< LOD				1.05 ± 0.21				581.05 ± 9.76				1.05 ± 0.21				380.97 ± 2.54			
P5	Pear	Voinesti (DB)	< LOD	100	100	100	6.64 ± 0.41	50	-	50	926.75 ± 12.65	-	-	-	0.72 ± 0.20	-	-	-	85.43 ± 6.53	-	-	-
P6		Candesti (DB)	< LOD				7.25 ± 0.41				874.22 ± 1.23				1.45 ± 0.41				259.80 ± 21.32			

LOD – limit of detection for: Pb: 2.0 µg/kg; Cd: 0.2 µg/kg;

EU – Commission Regulation (EC) no. 1881/2006; **Cod** – Standard Codex STAN 193-1995; **GB** – China Standard GB 2762-2012

Table 4. Heavy metals content in root vegetables samples

No.	Sample	Grown place (Origin)	Lead content: Pb (µg/kg)	Maximum levels (µg/kg) EU	Cod	GB	Cadmium content: Cd (µg/kg)	Maximum levels (µg/kg) EU	Cod	GB	Chromium content: Cr (µg/kg)	Maximum levels (µg/kg) EU	Cod	GB	Arsenic content: As (µg/kg)	Maximum levels (µg/kg) EU	Cod	GB	Nickel content: Ni (µg/kg)	Max. levels (µg/kg) EU	Cod	GB
P7	Potato	Radulesti (IL)	12.96 ± 2.29	100	100	200	17.28 ± 1.53	100	100	100	539.03 ± 10.31	-	-	500	3.37 ± 0.57	-	-	500	1035.68 ± 52.30	-	-	-
P8		Slobozia Moara (DB)	2.02 ± 0.41				11.28 ± 0.41				774.06 ± 2.04				8.67 ± 0.82				537.82 ± 13.09			
P13	Carrot	Vidra (IF)	74.38 ± 0.40	100	100	100	9.90 ± 1.20	100	100	100	730.77 ± 20.00	-	-	500	6.79 ± 0.80	-	-	500	572.40 ± 20.00	-	-	-
P14		Baleni (DB)	98.29 ± 4.85				26.29 ± 0.81				651.43 ± 4.04				14.86 ± 1.62				591.71 ± 18.99			
P15	Onion	Vidra (IF)	23.26 ± 0.77	100	100	100	7.84 ± 1.15	100	50	100	570.37 ± 1.15	-	-	500	2.03 ± 0.19	-	-	500	153.88 ± 3.44	-	-	-
P16		Baleni (DB)	< LOD				22.24 ± 0.40				478.32 ± 1.20				3.66 ± 0.40				280.12 ± 3.58			

LOD – limit of detection for: Pb: 2.0 µg/kg; Cd: 0.2 µg/kg;

EU – Commission Regulation (EC) no. 1881/2006; **Cod** – Standard Codex STAN 193-1995; **GB** – China Standard GB 2762-2012

Table 5. Heavy metals content in fruit and leaf vegetables samples

No.	Sample	Grown place (Origin)	Lead content: Pb (µg/kg)	Maximum levels (µg/kg) EU	Cod	GB	Cadmium content: Cd (µg/kg)	Maximum levels (µg/kg) EU	Cod	GB	Chromium content: Cr (µg/kg)	Maximum levels (µg/kg) EU	Cod	GB	Arsenic content: As (µg/kg)	Maximum levels (µg/kg) EU	Cod	GB	Nickel content: Ni (µg/kg)	Max.levels (µg/kg) EU	Cod	GB
P9	Tomato	Falastoaca (GR)	4.26 ± 0.86	100	50	100	6.09 ± 0.86	50	50	50	359.10 ± 23.69	-	-	500	4.42 ± 0.65	-	-	500	232.09 ± 43.94	-	-	-
P10		Vidra (IF)	8.09 ± 1.11				7.05 ± 0.37				398.91 ± 14.03				2.87 ± 0.37				306.50 ± 2.22			
P11	Cabbage	Vidra (IF)	< LOD	300	100	300	15.61 ± 1.30	200	50	200	337.99 ± 13.85	-	-	500	3.83 ± 0.65	-	-	500	110.52 ± 16.89	-	-	-
P12		Lunguletu (DB)	2.14 ± 0.43				2.14 ± 0.43				615.06 ± 6.92				4.44 ± 0.65				305.69 ± 0.43			
P17	Bell pepper	Baleni (DB)	62.16 ± 5.81	100	50	100	7.12 ± 0.77	50	50	50	650.33 ± 7.36	-	-	500	2.05 ± 0.19	-	-	500	707.01 ± 5.42	-	-	-
P18		Vidra (IF)	8.84 ± 1.26				1.49 ± 0.42				529.85 ± 2.52				4.16 ± 0.84				957.72 ± 6.31			
P19		Falastoaca (GR)	14.51 ± 1.71				4.53 ± 0.43				574.37 ± 3.42				2.12 ± 0.43				219.77 ± 16.67			
P20		Vidra (IF)	43.78 ± 5.13				5.58 ± 0.79				534.80 ± 10.25				2.51 ± 0.39				674.77 ± 22.87			
P21		Falastoaca (GR)	57.97 ± 3.95				4.03 ± 0.44				385.66 ± 5.26				4.19 ± 0.22				7501.55 ± 154.77			

LOD – limit of detection for: Pb: 2.0 µg/kg; Cd: 0.2 µg/kg;

EU – Commission Regulation (EC) no. 1881/2006; **Cod** – Standard Codex STAN 193-1995; **GB** – China Standard GB 2762-2012

Chromium and nickel contents were also higher. Most of the samples had chromium values higher than 500 µg/kg levels stated by GB 2762-2012 standard. Regarding nickel content, one of bell pepper samples had a value of 7.5 mg/kg, an extremely high content, which showed a contamination. Arsenic content was low, as in all other samples.

The overall content of the 5 analyzed heavy metals decreases in the following order: bell pepper > cabbage > tomato.

The overall content of the 5 analyzed heavy metals in all samples (Figure 2) decreases in the following order: bell pepper > potato > carrot > apple > plum > pear > onion > cabbage > tomato.

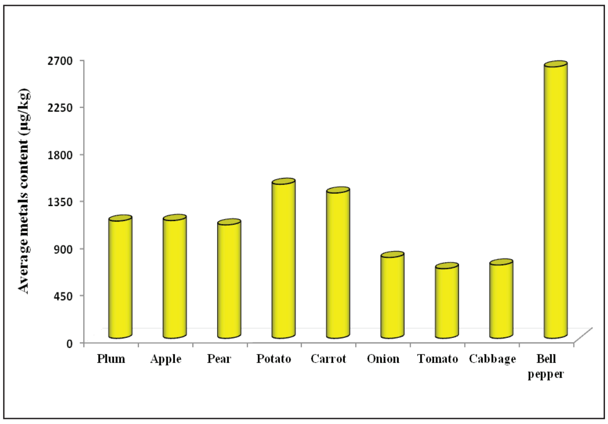

Figure 2. Average metals content (Sum of Pb + Cd + Cr + As + Ni)

4. Conclusions

- The aim of this study was to analyze fruits and vegetables samples grown in Ilfov county and neighboring counties, commonly consumed in the Bucharest-Ilfov region, with direct impact on the markets in this area, particularly in Bucharest, in terms of content in potentially carcinogenic heavy metals, namely: Pb, Cd, Cr, As and Ni. All samples were analyzed by ICP-MS technique after wet digestion on a microwave oven.

- Heavy metal concentrations varied among different vegetables, which may be attributed to differential absorption capacity of vegetables for different heavy metals. The highest overall average content of heavy metals was found in bell peppers, mainly due to the higher values of nickel content, while the lowest overall average content was found in tomatoes.

- For lead and cadmium, which have maximum levels at European level, all samples had values bellow the levels stated by Commission Regulation (EC) no. 1881/2006 (last amended in 2016).

- Arsenic content was at low levels, bellow 15 μg/kg; there is no maximum level for this metal stated at European or international level.

- Chromium content was higher than all other metals, most of them being above maximum level set by China Standard GB 2762-2012. The obtained values are for total chromium content, and not for chromium (VI) content which is classified as carcinogenic by IARC.

- A special case is represented by nickel. Although nickel compounds are class one carcinogens in IARC classification, there is no maximum allowed level for this metal. Some of the values obtained are higher, two of them, for potato and bell pepper, being so high that suggests a possible contamination.

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