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Heavy metals (Cd, Pb) and trace elements (Cu, Zn) contents in some foodstuffs from the Egyptian market

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Abstract: Determination of heavy metals; cadmium (Cd) and lead (Pb) as well as trace elements; copper (Cu) and zinc (Zn) contents in legumes, cereals, cereal products and fried potatoes purchased from the Egyptian market were carried out using atomic absorption spectrometry. The heavy metal amounts detected in legumes ranged from 0.010 to 0.178 $mg.kg^{-1}$ of Cd and 0.013 to 0.281 $mg.kg^{-1}$ of Pb. However, the trace elements ranged from 2.839 to 8.012 $mg.kg^{-1}$ of Cu, and 6.111 to 15.861 $mg.kg^{-1}$ of Zn. In the case of cereals, they ranged from 0.091 to 0.142 $mg.kg^{-1}$ of Cd, 0.116 to 0.398 $mg.kg^{-1}$ of Pb, 0.241 to 1.962 $mg.kg^{-1}$ of Cu, and 4.893 to 15.450 $mg.kg^{-1}$ of Zn. The highest values of Cd, Pb, Cu and Zn in cereal products were observed in popcorn (0.194 $mg.kg^{-1}$), pasta (0.299 $mg.kg^{-1}$), salted biscuits (1.386 $mg.kg^{-1}$) and pizza crust (13.70 $mg.kg^{-1}$), respectively. Fried potatoes content of the studied metals were found to be ranged from 0.054 to 0.10 of Cd, 0.065 to 0.159 of Pb, 1.626 to 1.992 of Cu and 3.837 to 6.844 $mg.kg^{-1}$ of Zn. Overall, the present study showed that the levels of Cu and Zn are generally below the permissible levels. However, the levels of Pb in broad beans, common bean, lupine, fenugreek, rice, wheat, spaghetti and pasta were found to be above the permissible levels. On the other hand, Cd levels found in most samples were above the acceptable levels as established by the regulatory organizations. The levels compare well with those reported for similar foodstuffs from some other parts of the world.

Key words: heavy metal contaminants, cereals, legumes, cereal products, fried potatoes, Egyptian market.

محتوى بعض الأغذية المجمعة من الأسواق المصرية من العناصر الثقيلة (الكادميوم والرصاص) والعناصر النادرة (النحاس والزنك)

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ملخص: تم تقدير مستوى بعض العناصر الثقيلة مثل الكادميوم والرصاص وبعض العناصر النادرة مثل النحاس والزنك في البقوليات والحبوب ومنتجات الحبوب والبطاطس المقلية والمجمعة من الأسواق المصرية باستخدام جهاز الامتصاص الذري للعناصر. وقد أظهرت النتائج أن مستوى الكادميوم في البقوليات يتراوح بين ٠،٠١ – ٠،١٧٨ مجم/كجم، ومستوى الرصاص يتراوح بين ٠،٠١٣ – ٠،٢٨١ مجم/كجم، وعلى الجانب الآخر كان مستوى النحاس في البقوليات يتراوح بين ٢،٨٣٩– ٨،٠١٢ مجم/كجم، ومستوى الزنك يتراوح بين ٦،١١١– ١٥،٨٦١ مجم/كجم. أما عن مستوى تلك العناصر في الحبوب فكانت التركيزات تتراوح بين ٠،٠٩١ – ٠،١٤٢ مجم/كجم للكادميوم، ٠،١١٦ – ٠،٣٩٨ مجم/كجم للرصاص، ٠،٢٤١ – ١،٩٦٢ مجم/كجم للنحاس، ٤،٨٩٣ – ١٥،٤٥٠ مجم/كجم للزنك، أما بالنسبة لمنتجات الحبوب فقد سجلت أعلى قيم للكادميوم والرصاص والنحاس والزنك في الفشار (٠،١٩٤ مجم/كجم)، العجائن (٠،٢٩٩ مجم/كجم)، البسكويت المملح (١،٣٨٦ مجم/كجم)، البيتزا (١٧،٧٠ مجم/كجم) على الترتيب. أما مستوى العناصر في عينات البطاطس المقلية فكان يتراوح بين ٠،٠٥٤- ٠،١٠ مجم/كجم للكادميوم، ٠،٠٦٥ – ٠،١٥٩ مجم/كجم للرصاص، ١،٦٢٦-١،٩٩٢ مجم/كجم للنحاس، ٣،٨٣٧ – ٦،٨٤٤ مجم/كجم للزنك. وأوضحت النتائج بشكل عام أن مستويات النحاس والزنك كانت أقل من الحدود المسموح بها بينما مستويات الرصاص في الفول البلدي واللوبيا والترمس والحلبة والأرز والقمح والمكرونة والعجائن كانت أعلى من المستويات المسموح بها وعلى الجانب الآخر كانت مستويات الكادميوم في معظم العينات أعلى من المستويات المسموح بها دوليا بواسطة الهيئات المنظمة- وقد تم مقارنة هذه المستويات بأخرى لنفس الأغذية في بلدان أخرى من العالم.

كلمات مفتاحية: العناصر الثقيلة، الحبوب، البقوليات، منتجات الحبوب، البطاطس المقلية، الأسواق المصرية.

Introduction

Heavy metals are potential environmental contaminants with the capability of causing human health problems if present to excess in the food we eat. They are given special attention throughout the world due to their toxic effects even at very low concentrations (Das, 1990). Several cases of human disease, disorders, malfunction and malformation of organs due to metal toxicity have been reported (Jarup, 2003). Legumes, cereals and cereal products are essential foodstuffs for human diet. Many studies pointed out that legumes and cereals contaminated with different levels of heavy metals.

The level of heavy metals in foodstuffs have been reported around the world; from East Asia (Wu Leung and Butrum, 1972), Sweden (Jorhem and Sundstroem, 1993), USA (Pennington et al., 1995 a,b), Egypt (Hussein and Bruggeman, 1997), China (Zhang et al. 1998), Nigeria (Onianwa et al., 1999), Italy (Conti et al., 2000), and Turkey (Saracoglu et al., 2004). In many developing countries such data are not readily available.

Trace heavy metals are significant in nutrition, either for their essential nature or their toxicity. Copper and zinc are known to be essential and may enter the food materials from soil through mineralization by crops or environmental contamination with metal-based pesticides. The adult human body contains about 1.5 to 2 ppm of copper (Kies, 1989) and 33 ppm of zinc (Fairweather-Tait, 1988). Excessive intake of either copper or zinc has been reported to be toxic (Somer, 1974; Graham and Cordano, 1976; Prasad, 1976; Fairweather-Tait, 1988).

Exposure of consumers to heavy metals and related health risks are usually expressed as percentage intake of provisional tolerable weekly intake (PWTI), a reference value established by WHO (1992, 1995). According to this, the weekly intake of metals from all sources should not exceed 0.05 and 0.075 $mg.kg^{-1}$ body weight for lead and cadmium, respectively. Cadmium and lead are among the most abundant heavy metals and are particularly toxic. The excessive content of these metals in food is associated with etiology of a number of diseases (WHO, 1992, 1995). Cadmium compounds are used as color pigment and in re-chargeable nickel-cadmium batteries. It is also present as a pollutant in phosphate fertilizers. Jarup et al. (1998) pointed out that cadmium is present in most foodstuffs, but concentrations vary greatly. Cadmium exposure may cause kidney damage and/or skeletal damage (WHO, 1992; and Jarup et al., 1998).

Airborne lead can be deposited on soil, water and plants thus reaching human via the food chain. Lead is accumulated in the skeleton and cause renal tubular damage and may also give rise to kidney damage (WHO, 1995). International Agency for Research on Cancer (IARC) classified cadmium and lead as human carcinogen (IARC, 1993; Steenland and Boffetta, 2000).

Owing to serious health risks, the levels of heavy metals (Cd, Pb) and trace elements (Cu, Zn) were determined in some Egyptian foodstuffs which contribute to the infant and young children foods such as legumes, cereals, cereal products and fried potatoes.

Materials and Methods

Sampling

Foodstuff samples of legumes (broad beans, cowpea, chick-pea, common bean, lupine, fenugreek, split lentil, and whole seed lentil), cereals (rice, maize, barely and wheat), cereal products (spaghetti, sweet biscuits, salted biscuits, pasta, pizza crust and popcorn) and fried potatoes sold as brand names (*Chipsy* and *Lays*) were purchased from various markets in Alexandria city, Egypt. The markets where these foodstuffs were purchased included those of East, West, Middle, Amriya and Montazah districts. Sampling (2 x 5 kg, for each foodstuff) was quite representative since the districts from where foodstuffs examined were scattered throughout Alexandria city, Egypt.

Sample preparation

Sub-samples (1 kg, each) were taken at random from the composite sample (10 kg) and were processed for analysis by the dry-ashing method. Wet solid samples were first dried in oven at 105 °C for 24 hrs and then ground while solid samples were ground directly. The ground solid samples (5g each) were placed in crucibles and few drops of concentrated nitric acid were added to the solid as an ashing aid. Dry-ashing process was carried out in a muffle furnace by stepwise increase of the temperature up to 550 °C and then left to ash at this temperature for 4 hrs (Crosby, 1977). The ash was left to cool and then rinsed with 1M nitric acid. The ash suspension was filtered and the filtrate made up to the volume of 25 ml with 1M nitric acid.

Spectroscopic analysis

The sample solutions were subsequently analyzed for heavy metal and trace element contents, as dry weight basis, using a Thermo-elemental type SOLAAR atomic absorption spectrometer, model NC9423-400-30042. Measurements were made using the hollow cathode lamps for Cd, Pb, Cu, and Zn at the proper wavelength and the slit width were adjusted and other AAS conditions employed in these determinations are summarized in Table 1. The flame type used for all elements was air-acetylene. Working solutions were prepared by dilution just before the use of standard solutions for atomic absorption spectroscopy (1000 ppm). For the determination, two solutions were prepared for each sample and three separate readings were made for each solution. The means of these figures were used to calculate the concentrations.

Quality assurance

Appropriate quality assurance procedures and precautions were carried out to ensure reliability of the results. Samples were generally carefully handled to avoid contamination. Glassware was properly cleaned, and the reagents were of analytical grade. Double distilled deionized water was used throughout the study. Reagents blank determinations were used to correct the instrument readings. A recovery study of the analytical procedure was carried out by spiking and homogenizing several already analyzed samples with varied amounts of standard solutions of the metals. The spiked samples were processed for the analysis by the dry ashing method and reanalyzed as described above. Average recoveries obtained were 87.44 ± 9.60, 91.35 ± 8.73, 100 ± 0.0, and 95.21 ± 4.78% for Cd, Pb, Cu, and Zn, respectively.

Results and Discussion

The detection limits for each metal were calculated as double the standard deviation of a series of measurements of a solution, the concentration of which is distinctly detectable above, but close to blank absorbance measurement (US-EPA, 1983). The values for Cd, Pb, Cu and Zn were presented in Table 1. The concentrations of the two heavy metals, Cd and Pb as well as the two trace elements, Cu and Zn in some foodstuffs from the Egyptian market analyzed as dry weight basis are presented in Table 2. The levels of the toxic metals, Cd and Pb were generally low, being much less than or just about 0.4 $mg.kg^{-1}$ in almost all samples. Cu and Zn, on the other hand, were higher than the corresponding levels of Cd and Pb in each sample. The mean detected levels of Cd and Pb in legumes ranged from 0.010 to 0.178 and 0.013 to 0.281 $mg.kg^{-1}$, respectively. The highest values of Cd and Pb in legumes were found in common bean and broad beans, respectively. On the other hand, the average concentrations of the studied heavy metals in cereals ranged from 0.091 to 0.142 $mg.kg^{-1}$ of Cd and 0.116 to 0.398 $mg.kg^{-1}$ of Pb. The highest levels of Cd and Pb in cereals were found in maize and wheat, respectively. In the case of cereal products, the peak values of Cd and Pb were observed in popcorn (0.194 $mg.kg^{-1}$) and pasta (0.299 $mg.kg^{-1}$), respectively.

Table 1. Standard conditions used in determination of different elements and their detection limits using Atomic Absorption Spectrometer.

Element	Wavelength (nm)	Slit width (nm)	Burner height Mm	Fuel flow L/min	Detection limit (ppm)
Cadmium	228.8	0.5	7.0	1.0	0.002
Lead	217.0	0.5	7.1	1.1	0.001
Copper	324.7	0.5	7.0	1.0	0.001
Zinc	213.9	0.5	7.0	1.0	0.002

Heavy metal contents in fried potatoes were found to be ranged from 0.054 to 0.10 of Cd and 0.065 to 0.159 $mg.kg^{-1}$of Pb. The levels of Cd in all samples are higher than the permissible limits (Walker, 1988), except cowpea, lentil and sweet biscuits. However, the levels of Pb in broad beans, common bean, lupine, fenugreek, rice, wheat, spaghetti and pasta are higher than the permissible limits set by Codex Alimentarious Commission (CAC, 2003).

In fact, Cu and Zn have numerous functions in the human body and they are essential elements for human health. Cu serves as an antioxidant and helps the body to remove free radicals and prevent cell structure damage and Zn function as a cofactor for many enzymes of the body. The mean concentrations of trace elements (Cu, Zn) in legumes ranged from 2.839 to 8.012 and 6.111 to 15.861 $mg.kg^{-1}$, respectively. The highest concentrations of Cu and Zn in legumes were found in broad beans and lupine, respectively. However, the corresponding average levels of these trace elements in cereals ranged from 0.241 to 1.962 and 4.893 to 15.450 $mg.kg^{-1}$, respectively. The peak values of Cu and Zn in cereals were found in barely and maize, respectively.

Table 2. Concentrations of cadmium, lead, copper, and zinc in legumes, cereals, cereals products and fried potatoes.

Food item	Mean concentrations in $mg.kg^{-1}$ dry weight ± S.E.			
	Cd	Pb	Cu	Zn
Legumes:				
Broad beans	0.063 ± 0.05	0.281 ± 0.26	8.012 ± 0.59	14.534 ± 4.05
Cowpea	0.010 ± 0.00	0.132 ± 0.01	3.165 ± 0.01	12.455 ± 0.29
Chick-pea	0.056 ± 0.04	0.143 ± 0.03	6.193 ± 2.78	10.764 ± 0.37
Common bean	0.178 ± 0.04	0.217 ± 0.09	4.254 ± 0.49	12.940 ± 1.51
Lupine	0.150 ± 0.04	0.218 ± 0.20	2.839 ± 0.35	15.861 ± 3.03
Fenugreek	0.150 ± 0.11	0.264 ± 0.12	7.252 ± 0.29	14.605 ± 4.60
Whole seed Lentil	0.013 ± 0.00	0.013 ± 0.00	3.676 ± 0.36	9.102 ± 1.26
Split Lentil	0.024 ± 0.00	0.024 ± 0.00	3.406 ± 0.19	6.111 ± 2.46
Cereals:				
Rice	0.091 ± 0.07	0.239 ± 0.02	0.241 ± 0.04	4.893 ± 0.70
Maize	0.142 ± 0.03	0.116 ± 0.06	0.458 ± 0.08	15.45 ± 2.94
Barely	0.136 ± 0.02	0.131 ± 0.06	1.962 ± 0.08	9.244 ± 3.62
Wheat	0.131 ± 0.02	0.398 ± 0.18	1.776 ± 0.66	12.02 ± 4.70
Cereal products:				
Spaghetti	0.155 ± 0.02	0.245 ± 0.14	0.403 ± 0.06	3.395 ± 0.64
Sweet biscuits	0.013 ± 0.00	0.126 ± 0.06	0.787 ± 0.05	2.347 ± 0.01
Salted biscuits	0.122 ± 0.07	0.127 ± 0.08	1.386 ± 0.18	4.749 ± 0.16
Pasta	0.127 ± 0.04	0.299 ± 0.13	0.816 ± 0.01	5.127 ± 0.29
Pizza crust	0.123 ± 0.06	0.176 ± 0.09	0.889 ± 0.17	13.70 ± 2.08
Popcorn	0.194 ± 0.09	0.152 ± 0.11	0.633 ± 0.02	5.218 ± 0.60
Fried potatoes				
Chipsy	0.054 ± 0.02	0.065 ± 0.05	1.626 ± 0.27	3.837 ± 0.78
Lays	0.100 ± 0.08	0.159 ± 0.10	1.992 ± 0.47	6.844 ± 1.39

The permissible limits for lead in cereal and legumes: 0.2 $mg.kg^{-1}$ (CAC., 2003).
The permissible limits for copper in all foods: 10 $mg.kg^{-1}$ (CAC, 1993).
The permissible limits for cadmium in all foods: 0.05 $mg.kg^{-1}$ (Walker, 1988).
The permissible limits for zinc in grains and beans: 50 and 100 $mg.kg^{-1}$, respectively (USDA, 2003).

In the case of cereal products, the average levels of Cu and Zn ranged from 0.403 to 1.386 and 2.347 to 13.70 $mg.kg^{-1}$, respectively. The highest values of Cu and Zn in cereal products were detected in salted biscuits (1.386 $mg.kg^{-1}$) and pizza crust (13.70 $mg.kg^{-1}$), respectively. Fried potatoes content of the studied trace elements were found to be ranged from 1.626 to 1.992 of Cu and 3.837 to 6.844 $mg.kg^{-1}$ of Zn. The current data exhibited that the levels of Cu and Zn were found to be below the permissible limits in all the tested samples analyzed (CAC, 1993; USDA, 2003).

The heavy metal levels found in the present study are comparable with those of other countries (Table 3). However, they agree with the average amounts of Cd and Pb that found in cereals, they do not seem to agree with data relevant to the Cd and Pb content of foodstuffs from the Egyptian market (wheat, maize and beans) which appear to be higher (Table 3). Concerning Cu and Zn contents, they appear to be in agreement with those found in Egyptian cereals. However, Cu was found to be lower in wheat, maize, rice, biscuits, legumes and higher in beans. Zn contents was found to be lower in wheat, biscuits, and legumes (Table 4).

Overall, our study showed that the levels of Cu and Zn are generally below the safe limits. However, the levels of Pb in broad beans, common bean, lupine, fenugreek, rice, wheat, spaghetti and pasta were found to be above the permissible levels. On the other hand, Cd levels found in most samples analyzed were above the acceptable levels according to international food standards for heavy metals.

It is concluded that atmospheric deposition from urban and agricultural areas may play an important role in the enrichment of agricultural produce from Cd and/ or Pb. The use of fertilizers and metal based pesticides in agriculture are also responsible for the contamination with Cu and Zn.

The results of this study supply valuable information about the metal contents in foodstuffs in Egypt. Moreover, these results can also be used to test the chemical quality of foodstuff in order to evaluate the possible risk associated with their consumption by humans.

Table 3. Levels of cadmium and lead in some foodstuffs from our study compared with those reported from other countries.

Foodstuff	Heavy metal concentrations ($mg.kg^{-1}$)								
	Our study		Greece [a]	Nigeria [b]		China [c]		Italy [d]	
	Cd	Pb	Cd	Cd	Pb	Cd	Pb	Cd	Pb
Cereals	0.091 – 0.142	0.116 – 0.398	-	0.005 – 0.260	0.08 – 0.40	0.009	0.031	-	-
Wheat	0.131	0.398	-	-	-	-	-	0.033 – 0.040	0.014 – 0.016
Rice	0.091	0.239	0.0052 - 0.0062	-	-	-	-	-	-
Maize	0.142	0.116	-	-	-	-	0.035	-	-
Biscuits	0.013 – 0.122	0.126 – 0.127	0.0126 - 0.0143	-	-	-	-	-	-
Broad beans	0.063	0.281	0.0005 - 0.0039	-	-	0.024 – 0.056	0.025 – 0.031	-	-
Common bean	0.178	0.217	0.0069 - 0.0140	-	-	-	-	-	-
Lentil	0.013 – 0.024	0.013 – 0.024	0.0007 - 0.0039	-	-	-	-	-	-
Chick-peas	0.056	0.143	0.0002 - 0.0029	-	-	-	-	-	-

[a]Karavoltsos et al 2002
[b]Onianwa et al 1999
[c]Zhang et al 1998,
[d]Conti et al 2000

Table 4. Comparison of copper and zinc levels in similar foodstuffs between Egypt and other countries.

Foodstuff	Heavy metal concentrations ($mg.kg^{-1}$)														
	Our study		Nigeria [a]		Italy [b]		Sweden [c]		East Asia [d]		USA [e]		Turkey [f]		Egypt [g]
	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Zn
Cereals	0.241 – 1.962	4.893 – 15.445	0.14-3.88	1.15-24.6	-	-	-	-	-	-	-	-	-	-	-
Wheat	1.776	12.020	-	-	3.2 - 3.4	32 - 33	-	-	-	-	-	-	-	-	-
Maize	0.458	15.445	2.33	33.4	-	-	-	-	1.6	14.0	0.48	6.1	-	-	4.39
Rice	0.241	4.893	1.53	-	-	-	-	-	-	-	-	-	-	-	9.35
Biscuits	0.787 – 1.386	2.347 – 4.749	-	-	-	-	-	-	-	-	-	-	<1 - 4.2	3.1 - 16.1	-
Legumes	2.839 – 8.012	6.111 – 15.861	4 - 12.5	14 - 42.7	-	-	-	-	-	-	-	-	-	-	-
Beans	8.012	14.534	6.87	42.7	-	-	5.6	27	8.8	11.0	2.72	14.0	-	-	29.1

[a]Onianwa et al 1999,
[b]Conti et al 2000,
[c]Jorhem and Sundstroem 1993,
[d]Wu Leung and Butrum 1972,
[e]Pennington et al 1995 a & b,
[f]Saracoglu et al 2004,
[g]Hussein and Bruggeman 1997

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