



Assessing potential dietary intake of heavy metals in some selected fruits and vegetables from Ghanaian markets

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ABSTRACT

Heavy metals contamination was carried out with the aim to assess the levels of lead (Pb), cadmium (Cd), copper (Cu), zinc (Zn) and chromium (Cr) in various fruits and vegetables sold in Ghanaian markets. A total of 350 fruits and vegetable samples were tested and analyzed by Atomic Absorption spectrometry. The test results showed that the average concentrations detected were ranged from below detection limit to 0.56, below detection limit to 0.08, 1.24 to 6.32, 2.32 to 21.4 and below detection limit to 1.06 mg/kg for Pb, Cd, Cu, Zn and Cr, respectively. The highest mean levels of Pb and Cd were detected in lettuce, Cu and Zn in cucumber and Cr in carrot samples. The daily intakes of Pb, Cd, Cu, Zn and Cr through fruits and vegetables have also been estimated. The levels of metals were all below the permissible limits by WHO/FAO. For there were heavy metals present in the fruits and vegetable samples, consumption of fruits and vegetables there was no zero risk. But the exposure of consumer did not exceed the acceptable daily intake (ADI). This study suggests that a yearly monitoring program for heavy metal in foodstuffs is necessity.

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Introduction

Heavy metals are important environmental pollutants, particularly in area with high anthropogenic pressure. Their presence in the atmosphere and water, even in traces, can cause serious problems to all organisms. Heavy metal accumulation in soils is of concern in agricultural production due to the adverse effects on food quality. Heavy metals are among the major contaminants of food supply and may considered the most important problem to our environment (Radwan and Salama, 2006). Heavy metals are persistent and non-biodegradable, have a long biological half-lives and they can be bioaccumulated through the biologic chains: soil-plant-food and seawater-marine organism-food leading to unwanted side effects (Shawi *et al.*, 1999). So, the presence in high amount of heavy metals in environment represents a potential danger for human health and for environment due to their extreme toxicity. Although some of the heavy metals such as Zn, Mn, Ni and Cu act as micro-nutrients at lower concentrations, they become toxic at higher concentrations.

Fruits and vegetables accumulate heavy metals in their edible and non edible parts. Food safety issues and potential health risk make this as one of the most serious environmental concerns (Singh *et al.*, 2010). During the last decades, the increasing demand of food safety has stimulated research regarding the risk associated with consumption of foodstuffs contaminated by heavy metals, pesticides and /or toxins (D'Mello, 2003).

Heavy metal contamination may be occurred due to irrigation with contaminated water, the addition of fertilizers and metal-based pesticides, industrial emissions, transportation, harvesting process, storage and/or sale. Crops and vegetables grown in soils contaminated with heavy metals have greater accumulation of heavy metals than those grown in

uncontaminated soil (Marshall *et al.*, 2007; Sharma *et al.*, 2006, 2007). Plants take up metals by absorbing them from contaminated soils as well as from deposits on parts of the plants exposed to the air from polluted environment (Khairiah *et al.*, 2004; Chojnacha *et al.*, 2005).

In general, food is the main exposure route and human beings are encourage to consume more fruits and vegetables, which are a good source of vitamins, minerals, fiber and also beneficial to their health. Unfortunately, these foodstuffs contain both essential and toxic metals over a wide range of concentrations. (Radwan and Salama, 2006). It has been reported that nearly half of the mean ingestion of lead, cadmium and mercury through food is due to plant origin (fruit, vegetables and cereals). Moreover, some population groups seem to be exposed, especially vegetarians, since they absorb more frequently 'tolerable daily doses' (Islam *et al.*, 2007).

Dietary intake of heavy metals through contaminated fruits and vegetables may lead to various chronic diseases. Duruibe *et al.*, 2007 cited by Radwan and Salama, 2006 suggested that bio-toxic effects of heavy metals depend upon the concentrations and oxidation states of heavy metals, kind of sources and mode of deposition. Lead and cadmium are among the most abundant heavy metals and particularly toxic. The excessive content of these metals in food is associated with etiology o a number of diseases, especially wit cardiovascular, kidney, nervous as well as bone diseases. (WHO, 1992, 1995; Steenland and Boffeta, 2000; Jarup, 2003; Radwan and Salama, 2006). Severe exposure of Cd may result in pulmonary effects such as emphysema, bronchiolitis and alveolitis. Renal effects may also result due to subchronic inhalation of Cd (EU, 2002; Young 2005).

Other metals such as Zn and Cu are essential for important biochemical and physiological functions and necessary for maintaining health throughout life, however, higher

concentrations of Zn can cause impairment of growth and reproduction (Nolan, 2003). Zinc deficiency results in a variety of immunological defects whereas copper deficiency is characterized by anemia, neutropenia and skeletal abnormalities (Prentice, 1993; Linder and Azam, 1996).

Keeping in view of the potential toxicity, persistent nature and cumulative behavior as well as the consumption of fruits and vegetables, there is necessary to test and analyzed these food items to ensure that the levels of these contaminants meet the agreed international requirement. Although heavy metals contamination in foodstuffs have been carried out for decades in most developed countries (Intawongse and Dean, 2006; Pennington *et al.*, 1995a,b; Milacic and Kralj, 2003), but, fruits and vegetables of Ghana is not much investigated from heavy metals contamination point of view. Everyday people are being encouraged to consume more vegetables and fruits. Thus there has been escalation in the growth of vegetables and fruits in the urban and rural areas of Ghana to meet the domestic consumption as well as international markets, hence, a large population at risk (Bempah *et al.*, 2011).

The present work deals with the quantification of heavy metals (lead, cadmium, copper zinc and Chromium) concentrations in selected fruits and vegetables sold in local markets of Accra, the national capital of Ghana.

The data enable us to estimate heavy metal intakes in citizen and to compare them with acceptable daily intake (ADIs) of the food and Agricultural Organization (FAO) as well as with the World Health Organization (WHO), so as to assess potential health hazards.

Materials and methods

Sampling

A total of 350 samples of fruits and vegetables were purchased from several local and supermarkets in Accra metropolitan city of Ghana, during February 2010 and March 2011. The markets where these foodstuffs were purchased include open markets, supermarkets, roadside grocery shops and peddlers. From each market, a sample size of at least one kg for small and medium sized of fresh product was purchased.

The minimum weight for large sample sizes was 2 kg, where the unit was generally more than 250 g (Codex Alimentarius Commission, 2000) which was quite representative since the markets from where these food stuffs examined were scattered throughout the cities. For the analysis only the edible portions were included, whereas bruised or rotten parts were removed.

Sample preparation and treatment

Sample unit of fresh fruits and vegetable (2 kg each) were taken at random from a composite sample and were thoroughly clean. Only edible portions of the test fruits and vegetables were used for the analysis. The fruits and vegetable samples were transferred into a crucible and oven dried at 105 °C for 24 h.

The dried samples were poured in a mixer grinder taking care not to overheat the sample. 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 h. The ash was left to cool and then decompose using concentrated nitric acid (10 ml).

The ash suspension was filtered into a 25 ml volumetric flask using Whatman filter paper No. 41 and the solution was completed to the mark using deionised water.

Atomic Absorption Spectrophotometer determination

Analysis of heavy metals of interest was performed using a Varian model AA 240 FS Atomic Absorption Spectrophotometer. Measurements were made using a hollow cathode lamp for Pb, Cd, Cu, Zn, and Cr at wavelengths of 217.0, 228.8, 324.7, 213.9, and 357.9 nm respectively. The slit width was adjusted for all metals at 0.2 nm. The calibration curves were prepared from standards by dissolving appropriate amounts of the metal salts in purified nitric acid, diluting with deionised water and storing as stock solutions in a quartz flask. Fresh working solutions were obtained by serial dilution of stock solutions.

Quality assurance

Appropriate quality assurance procedures and precautions were carried out to ensure reliability of the results. The recovery and reproducibility of the method was carried out by spiking and homogenizing several already analyzed samples with varied amounts of standards solutions of the metals and processed as previously described. Reagent blank determinations were used to correct the instrument readings and the limit of detection (LOD) of the analytical method for each metal was calculated as double the standard deviation of a series of measurement of a solution, the concentration of which is distinctly detectable above, but close to blank absorbance measurement (US EPA, 1983). The limit of quantification (LOQ) was determined by preparing two solutions of each sample and three separate readings were made for each solution according to International Accreditation Criteria for Laboratories Performing Food Chemistry Testing Method, 1999. Table 3 shows the LODs and LOQs obtained for each element by AAS (Table 1).

Calculation of estimated daily intake

For preliminary assessment of consumer's exposure to heavy metals in fruits and vegetables, the calculated estimated daily intake (EDI) is expressed as percentages of the ADI. Hence, for each type of exposure, the estimated daily intake (mg/kg/day) was obtained by multiplying the residual heavy metals concentration (mg/kg) in the food of interest times the food consumption rate (kg/day).

In this study, fruits and vegetables consumption data were referred by the International Food Policy Research Institute data, 2004. Food consumption rate for fruits and vegetables in Ghana is found to be 64 g/person/day and 137 g/person/day, respectively.

Results and discussions

A total of 350 samples of fruits and vegetables were analyzed for heavy metals from the period February, 2010 to March, 2011. Table 2 shows the number of samples surveyed and those containing heavy metals in each kind of fruit and vegetable sample. Overall, 15.0 % of papaya, 46.7 % of watermelon, 28.0 % of banana, 12.0 % of mango, 25.0 % of pear, 13.3 % of pineapple, 26.7 % of tomato, 6.7 % of lettuce, 16.0 % of cabbage, 12.0 % of carrot, 46.7 % of okra, 53.3 % of green pepper, 13.3 % of onion and 6.7 % of cucumber samples contained no detectable heavy metal residues. There are no detected residues of Pb in watermelon, Cd in watermelon and banana as well as Cr in watermelon, pineapple, okra and green pepper. However, Zn and Cu were detected in all the monitored fruits and vegetable samples surveyed. Lead (Pb), cadmium (Cd), copper (Cu), zinc (Zn) and chromium (Cr) were detected in 88 (25.1 %), 79 (22.6 %), 215 (61.4 %), 218 (62.3 %), and 45 (12.9 %) fruits and vegetables, respectively. Hence the predominate residues are Zn, followed by Cu, Pb, Cd and Cr.

Vegetables had higher detection rates of heavy metals than fruits. The detection rates are greater than 40.0 % in all the monitored fruit and vegetable samples. For vegetables, the detection rate is between 46.7 % in green pepper and 93.3 % in lettuce and cucumber while that of fruits ranges between 53.3 % in watermelon and 88.8 % in mango samples.

The average heavy metal concentrations in fruits and vegetables are given in Table 3. The amount of heavy metal contents are related to the crop, collected site of sample, age of fruiting bodies and distance from the source of pollution (Kelac et al., 1991). The heavy metal concentrations in the samples are hardly affected by pH or organic matter content of the soil (Isildak et al., 2004). The trace element contents of the samples depend on the ability of the crops to extract elements from the soil and on the selective uptake and deposition of elements in tissues (Demirbas, 2001; Sesli and Tuzan, 1999). The uptake of heavy metal ions in vegetables is higher than in fruits. For this reason, the concentration variations of heavy metals could be considered due to the nature of the crops and their ecosystems.

Among the fruits and vegetable samples, the highest Pb content determined was 0.56 ± 0.03 mg/kg in lettuce with a range of 0.48-0.63 mg/kg whereas the lowest concentration was in watermelon (below detection limit) (Fig.1).

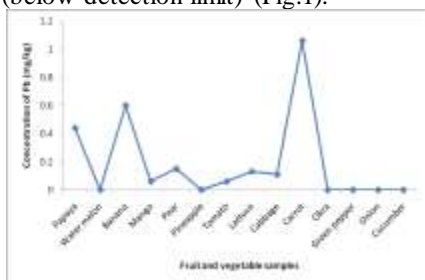


Fig. 1. Distribution of Pb content in fruits and vegetables

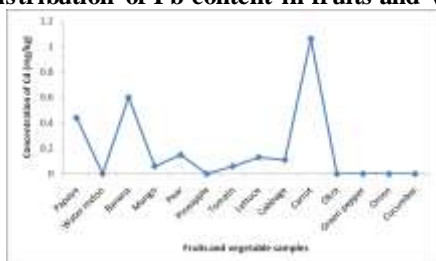


Fig.2. Distribution of Cd content in fruits and vegetables

This result therefore corroborate the findings of Yu et al, (2006) who reported that out of eight species of vegetables analyzed, non leafy vegetables were less contaminated than the leafy vegetables cultivated in the same area. Miller et al, 2004 and Lacatusa et al., in 1996 found that Pb concentration in lettuce is higher than onions and carrots, suggesting that the accumulation effect stringly depends on the crop physiological properties.

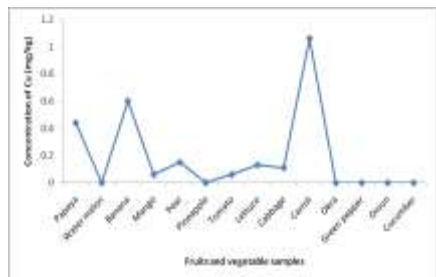


Fig.3. Distribution of Cu content in fruits and vegetables

The levels of Cd in the samples ranged from no detectable amount in watermelon and banana to 0.08 ± 0.04 mg/kg (0.02-

0.12 mg/kg) in lettuce (Fig. 2). The Cd levels are in agreement with literature values (Cibulka et al., 1996; Vetter, 1994).

Among the fruits and vegetable samples, the greatest concentration of Cu was obtained in the cucumber (6.32 ± 0.68 mg/kg), green pepper (5.31 ± 0.19 mg/kg) and pineapple (4.23 ± 0.16 mg/kg). For other samples in this study, Cu concentrations were between 1.24 ± 0.08 mg/kg and 3.23 ± 0.13 mg/kg (Fig.3). Cu value has been reported to be 10-70 mg/kg (Falandysz et al., 1994; Vetter, 1994).



Fig.4. Distribution of Zn content in fruits and vegetables

Minimum concentration value for Zn (2.32 ± 0.06 mg/kg) was found in papaya with a range of 1.46-3.22 and maximum concentration value of 21.4 ± 2.62 mg/kg was detected in cucumber with range of 18.44-23.7 (Fig.4). Zn is widespread among living organisms, due to its biological significance.

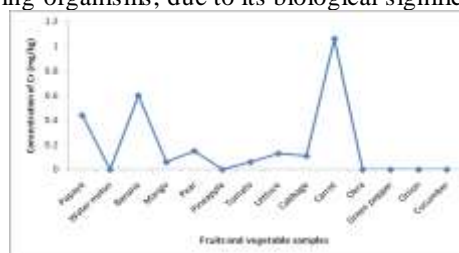


Fig.5 Distribution of Cr content in fruits and vegetables

Zn content in fruits and vegetables ranges from 1.36-20.9 mg/kg (Radwan and Salama, 2006). Hence zinc content in fruits and vegetables are in agreement with previous studies (Radwan and Salama, 2006; Islam et al., 2007).

Minimum and maximum values for Cr were obtained for pineapple, watermelon, okra, green pepper, onion and cucumber (below detection limit) and carrot (1.06 ± 0.06 mg/kg) (Fig 5). Cr results were close to those found in literature (Ray et al., 2010; Isildak et al., 2004; Singh et al., 2010)

Overall, heavy metals contamination was high in vegetables than in fruits. This can be attributed partly to the fact that vegetables are often grown in polluted and degraded environmental conditions in the peri-urban (or urban fringe) zone and are subjected to further pollution from vehicles and industries during marketing. They are often irrigated with contaminated water, as well as the addition of fertilizers and metal-based pesticides to boost production (Radwan and Salama, 2006).

Although the residue levels of these heavy metals are below the maximum permissible intake (calculated by ADI multiply body weight 60 kg) of Codex Committee on Pesticide Residues (CCPR), and some developed countries (Dogheim et al., 1996), there was no zero risk because there were heavy metals present in the fruit and vegetable samples. In order to take account of the maximum risk of consumer's exposure in fruit and vegetables, the estimated daily intake (EDI) residues was calculated with maximum residue levels.

According to the data from International Food Policy Research Institute data, 2004, the mean consumption of fruits

and vegetables in Ghana is found to be 64 g/person/day and 137 g/person/day, respectively. Table 4 shows the results of calculation of preliminary EDIs of heavy metals and the corresponding ADIs by adults in Ghana. The exposure did not exceed the ADI certainly.

When the present concentrations of metals were compared with permissible limits by WHO/FAO (WHO/FAO, 2007), it was found out that all the levels were within the safe limit (Table 5).

Conclusion

It was realized that heavy metals are environmentally hazardous and many developed and developing countries have been continuing to monitor the trends of residues in fruits and vegetables. But maximum residue limit (MRL) for heavy metal in foodstuffs is currently not in force in most countries. This study shows that there have been some heavy metals in fruits and vegetables in Ghana. It indicates that the present monitoring program that has been conducted only for heavy metals in fruits and vegetables is insufficient. Although heavy metals in fruits and vegetables do not pose any immediate risk to human health so far, but continues consumption of such fruit and vegetables even with moderate contamination level can accumulate in the receptors body and may prove fatal for human population in the long term. Thus a yearly monitoring program for heavy metals in foodstuffs is necessary.

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Table 1: Average recovery, Limit of detection (LOD) and quantitation (LOQ) of elements assayed by AAS.

Metal	Average Recovery±SD	LOD	LOQ
Lead (Pb)	93.5±6.3 %	0.001	0.003
Cadmium (Cd)	89.6±10.2 %	0.002	0.007
Copper (Cu)	94.2±5.8%	0.001	0.003
Zinc (Zn)	91.6±8.9%	0.002	0.007
Chromium (Cr)	94.7±4.8%	0.001	0.003

SD- Standard deviation; LOD- Limit of detection; LOQ- Limit of quantitation

Table 2: Number of samples surveyed and contained detectable heavy metals content in fruits and vegetable samples

Food items	surveyed samples	Total no. of samples with residues	Detection rates (%)	No. of samples with residues				
				Pb	Cd	Cu	Zn	Cr
<i>Fruits</i>								
Papaya	20	17	85.0	4	8	11	18	4
Water melon	15	8	53.3	0	0	9	12	0
Banana	25	18	72.0	6	0	18	21	3
Mango	25	22	88.0	3	6	17	18	6
Pear	20	15	75.0	4	2	13	16	5
Pineapple	15	13	86.7	5	5	11	13	0
<i>Vegetables</i>								
Tomato	30	22	73.3	12	6	21	14	9
Lettuce	30	28	93.3	9	11	19	17	7
Cabbage	25	21	84.0	16	13	17	11	7
Carrot	25	22	88.0	18	9	14	12	5
Okra	30	16	53.3	3	2	12	9	0
Green pepper	30	14	46.7	2	6	17	19	0
Onion	30	26	86.7	2	4	21	17	0
Cucumber	30	28	93.3	4	7	15	21	0

Table 3: Heavy metals content (mg/kg) in fruits and vegetables

Food items	Concentration and range of metals									
	Pb		Cd		Cu		Zn		Cr	
	Mean±SD	Range	Mean±SD	Range	Mean±SD	Range	Mean±SD	Range	Mean±SD	Range
<i>Fruits</i>										
Papaya	0.07±0.01	0.01-0.09	0.02±0.01	0.002-0.04	1.44±0.20	0.18-1.62	2.32±0.06	1.46-3.22	0.44±0.01	0.21-0.62
Water melon	<LOD	-	<LOD	-	1.90±0.07	1.73-2.22	13.6±1.42	10.4-18.3	<LOD	-
Banana	0.01±0.01	0.01-0.03	<LOD	-	3.23±0.13	2.43-4.20	6.42±1.23	4.26-8.32	0.60±0.03	0.04-0.84
Mango	0.08±0.00	0.02-0.12	0.04±0.02	0.008-0.06	1.24±0.08	0.68-1.84	2.44±0.06	1.68-4.22	0.06±0.00	0.02-0.12
Pear	0.10±0.04	0.09-0.14	0.01±0.00	0.007-0.03	1.48±0.28	0.80-2.20	4.01±0.66	2.44-8.88	0.15±0.07	0.03-1.23
Pineapple	0.06±0.02	0.03-0.11	0.01±0.00	0.003-0.05	4.23±0.16	3.62-4.88	6.03±1.04	5.33-9.13	<LOD	-
<i>Vegetables</i>										
Tomato	0.21±0.11	0.14-0.28	0.02±0.01	0.004-0.06	2.31±1.40	1.64-2.86	8.42±0.17	6.46-8.42	0.06±0.01	0.02-0.08
Lettuce	0.56±0.03	0.48-0.63	0.08±0.04	0.02-0.12	1.92±0.09	1.12-2.44	10.38±1.53	8.09-13.6	0.13±0.07	0.09-0.17
Cabbage	0.43±0.07	0.22-0.53	0.06±0.02	0.006-0.08	2.32±0.08	1.84-3.40	8.62±0.18	6.86-14.3	0.11±0.09	0.03-0.13
Carrot	0.16±0.03	0.12-0.23	0.03±0.01	0.01-0.05	1.71±0.07	1.14-2.33	8.64±0.14	8.22-10.6	1.06±0.06	0.06-1.22
Okra	0.18±0.06	0.14-0.26	0.03±0.01	0.004-0.06	2.31±0.06	1.64-2.66	7.44±0.09	5.48-11.4	<LOD	-
Green pepper	0.52±0.09	0.42-0.61	0.05±0.02	0.03-0.09	5.31±0.19	4.22-6.30	16.4±0.54	14.56-22.6	<LOD	-
Onion	0.08±0.02	0.02-0.14	0.02±0.01	0.008-0.004	1.44±0.07	0.86-2.44	10.43±0.88	8.08-12.6	<LOD	-
Cucumber	0.17±0.02	0.13-0.26	0.05±0.03	0.03-0.09	6.32±0.68	4.54-7.48	21.4±2.62	18.44-23.7	<LOD	-

Each value is the mean of five samples with three determinations

SD- Standard deviation

LOD= Limit of detection (See table 1)

Table 4: Estimated dietary intakes (EDIs) of heavy metals by adults in Ghana and acceptable daily intake (ADIs)

Heavy metals content	EDI (g/kg body weight/day)		EDI % of ADI		ADI, mg/kg body weight/day
	Fruits	Vegetables	Fruits	Vegetables	
Pb	0.068	0.662	0.32	3.09	0.214
Cd	0.021	0.098	0.35	1.63	0.060
Cu	2.400	6.759	0.80	2.25	3
Zn	6.187	26.189	0.10	0.44	60
Cr	0.334	0.776	2.23	5.17	0.15

Table 5: Safe values for Pb, Cd, Cu, Zn and Cr in fruits and vegetables recommended by WHO/FAO (2007)

Element	Maximum allowable limits of elements in fruits and vegetables (mg/kg dry weight)
Pb	5
Cd	0.2
Cu	40
Zn	60
Cr	2.3