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Pesticide residues and heavy metals levels in some selected fruits and vegetables from Ghanaian markets

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ABSTRACT

A total of 350 fruits and vegetable samples were collected from local markets in Ghana and subjected to pesticide residue and heavy metal analyses. Residues of some organochlorine pesticides (OCPs), such as gamma-HCH, methoxychlor, aldrin, dieldrin, endrin, p,p'-DDE, and p,p'-DDT, as well as synthetic pyrethroid pesticides (SPPs), such as permethrin, cyfluthrin, cypermethrin, fenvalerate, deltamethrin were found in a number of samples at different concentrations. The results obtained showed that 37 % of the fruit and vegetable samples analyzed contained no detectable level of the monitored OCPs, 22 % of the samples gave results with levels of organochlorine pesticides residues above the MRL, while 41 % of the samples showed results below the MRL. The majority of the analyzed samples contained detectable concentrations of Pb, Cd, Cu, Zn and Cr. All the metal concentrations were far below their respective limit. Daily intake limit was calculated and compared with their respective tolerable limits. Synthetic pyrethroid pesticides were detected in 66 % of the samples without violation. The daily intake of heavy metals and pesticide residues revealed that all the monitored pesticides and heavy metals were far below the recommended tolerable levels except aldrin and dieldrin in fruits and aldrin, dieldrin and endrin in vegetables suggesting a great potential for systemic toxicity particularly in children considered the most vulnerable population subgroup. Based on observations made in these studies, it is proposed that more extensive monitoring investigation covering all foodstuffs be carried out to find the exact position of heavy metals and pesticide residues.

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Introduction

Pesticide and heavy metals are persistent and non-biodegradable and they can be bioaccumulated through the biologic chains: soil-plant-food and seawater-marine organism-food (Shawi *et al.*, 1999). So, the presence in high amount of pesticide and heavy metals in environment represents a potential danger for human health and for environment due to their extreme toxicity. For this reason, accurate monitoring of their concentration plays an important role.

Population can be contaminated with organic pollutants and heavy metals by ingestion of contaminated or polluted food and water. The gravity of toxic effect depends on nature, concentration, body resistance and presence of other contaminants. The concentration of these elements in food products is varied, depending of their origin, storage conditions and processing technologies (Stanciu *et al.*, 2005).

Food safety is a major public concern worldwide. During the last decades, the increasing demand of food safety has stimulated research regarding the risk associated with consumption of foodstuffs contaminated by pesticides, heavy metals and/or toxins (D'Mello, 2003).

Farmers around the world including Ghana use pesticides as a preventive policy against the possibility of a devastating crop loss from pests and diseases. Accordingly in Ghana, for several decades now, pesticides have been employed in agriculture not only to control and eradicate crop pests but also in the public health sector for disease vector control. Nevertheless, heavy metals are also among the major contaminants of food supply and may considered the most important problem to our environment (Zaidi *et al.*, 2005). Such problem is getting more serious all over the world especially in developing countries.

During the last decades, the increasing demand of food safety has stimulated research regarding the risk associated with consumption of fruit and vegetables as they constitute major part of human diet contributing nutrients, vitamins, minerals and fibers. However, these plants contain both pesticide residues and heavy metals over a wide range of concentration. Therefore, residues of pesticides and heavy metals could affect the ultimate consumers especially when these commodities are freshly consumed. The total dietary intake of pesticides residues that remain on agricultural commodities are known as carcinogens/or toxins and therefore it is desirable to reduce these residues (Zawiyah *et al.*, 2007).

In general, food is the main exposure route. Exposure to pesticide residues and heavy metals through the diet is assumed to be five orders of magnitude higher than other exposure routes, such as air and drinking water (Juraske *et al.*, 2009). According to the World Health Organization (WHO), food consumption consists on averaged for 30 % (based on mass) of fruit and vegetables, and fruit and vegetables are the most frequently consumed food group (WHO, 2003). Furthermore, because fruit and vegetables are mainly consumed raw or semi-processed, it is expected that they contain higher pesticides residue and heavy metal levels compared to other food groups of plant origin, such

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as bread and other foodstuffs bases on cereal processing (Claeys *et al*., 2011).

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 pesticide and heavy metals contamination in foodstuffs have been carried out for decades in most developed countries (Claeys *et al*., 2011; Stanciu *et al*., 2005; Pennington *et al*., 1995a,b; Milacic and Kralj, 2003), but, fruits and vegetables of Ghana is not much investigated from heavy metals and pesticide 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 the (Bempah *et al*., 2011).

This work presents aspects regarding the pollution with pesticides (organochlorine and pyrethroid) and heavy metals (Pb, Cd, Zn, Cu and Cr) in selected fruits and vegetables sold in the local markets of Ghana and to generate awareness about the lethal effects of these pesticides and heavy metals on human beings as well as to estimate the daily intake of these pesticides and heavy metals through the consumption of fruits and vegetables.

Materials and methods

Sampling

A total of 350 samples of fruits and vegetables were purchased from several local and supermarkets in Ghana, during February 2010 and March 2011. The markets were 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 through out the cities. For the analysis only the edible portions were included, whereas bruised or rotten parts were removed. The various collected samples (item, scientific name, number) are presented in Table 1.

Sample preparation and treatment

Sample unit of fresh fruits and vegetable (2 kg) were thoroughly shredded and homogenized. Approximately 120.0 g of the sample was used for pesticide analysis while 180.0 g subsample for heavy metals determinations.

Analysis of pesticide residues

Determination of pesticide residues in fruits and vegetables were performed according to the Netherlands analytical methods of pesticide residues and foodstuffs (2006) with modifications.

Extraction and clean-up

Triplicate sub sample (40.0 g each) of fruits and vegetables were homogenized and macerated with 40 ml of ethyl acetate. Sodium hydrogen carbonate 5.0 g and anhydrous sodium sulfate 20.0 g were added to remove moisture and further macerated for 3 min using the ultra-turax macerator. The samples were then centrifuged for 5 min at 3,000 rpm to obtain the two phases. The extraction process was followed by a clean-up step using solid-phase extraction with florisil (5 g) and alumina (2 g) deactivated with Milli-Q water (5 %) as adsorbents. Pesticides in sample extract were eluted with 35 ml of hexane:ethyl acetae (80 + 20, V + V), concentrated to 1 ml using a rotary evaporator and then dried by a gentle nitrogen stream. This was dissolved in 5 ml of hexane; pesticides were then quantified by gas chromatograph equipped with electron capture detector (GC-ECD).

Quantitation

An external method was employed in the determination of the quantities of residues in the sample extracts. A standard mixture of known concentration of pesticide was run and the response of the detector for each compound ascertained. The area of the corresponding peak in the sample was compared with that of the standard. All analyses were carried out in triplicates and the mean concentrations computed accordingly.

Recovery rate and limit of detection

Carrot samples were fortifies at 0.01, 0.02 and 0.1 mg/kg by adding 5.0 ml of a mixed standard solution. Recovery and precision (expressed as relative standard deviation) were calculated for three replicate samples and the data are presented in Table 2. The table shows that the recovery rate for twelve pesticides were within acceptable range. The method is applicable for the determination of eleven pesticides in fruit and vegetable samples (Zawiyah *et al*., 2007). Percent recoveries in spiked samples ranged 87.0 % - 104.0 %, Table 2. Accordingly, the sample analysis data were corrected for these recoveries. Detection limit(s) of the method were also assessed based on the lowest concentrations of the residues in each of the matrices that could be reproducibly measured at the operating conditions of the GC; which were 0.001 mg/kg. Blank analyses were also carried in order to check any interfering species in the reagents.

Gas chromatographic determination

The residues were analyzed by Shimadzu gas chromatograph GC-2010 equipped with ^{63}Ni electron capture detector that allowed the detection of contaminants even at trace level concentrations from the matrix to which other detector do not respond. The GC conditions and the detector response were adjusted so as to match the relative retention times and response. The conditions used for the analysis were: capillary column coated with ZB-5 (30 m × 0.25 mm, 0.25 μm film thickness). Carrier gas and make-up gas was nitrogen at a flow rate of 1.0 and 29 mL/min, respectively. The injector and detector temperature were set at 280 ^{o}C and 300 ^{o}C respectively. The oven temperature was programmed as follows: 60 ^{o}C held for 1 min, ramp at 30 ^{o}C min^{-1} to 180 ^{o}C, held for 3 min, ramp at 3 ^{o}C min^{-1} to 220 ^{o}C, held for 3 min, ramp at 10 ^{o}C min^{-1} to 300 ^{o}C. The injection volume of the GC was 1.0 μL. The residues detected by the GC analysis were further confirmed by the analysis of the extract on two other columns of different polarities. The first column was coated with ZB-1 (methyl polysiloxane) connected to ECD and the second column was coated with ZB-17 (50% phenyl, methyl polysiloxane) and ECD was also used as detector. The conditions used for these columns were the same.

Analysis of heavy metals

Determination of heavy metals in fruits and vegetables were performed according to the method of Crosby, 1977 with modifications.

Sample preparation and treatment

Samples in triplicate were homogenized separately and 20 g of the fresh homogenate was transferred into a crucible and oven dried at 105 ^{o}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 furnance 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 quatz 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**

Daily intake estimate of pesticides residues and heavy metals in fruits and vegetables

Daily intake estimations were done based on an integration of pesticide and heavy metals analysis data, and food consumption rate. Food consumption rates were based on the International Food Policy Research Institute data, 2004.

Food consumption rate for fruits and vegetables in Ghana is found to be 0.064 kg/person/day and 0.137 kg/person/day, respectively. Hence, for each type of exposure, the estimated daily intake (mg/kg/day) was obtained by multiplying the residual pesticide and heavy metals concentration (mg/kg) in the food of interest times the food consumption rate (kg/day).

Results and discussion

The fruits and vegetables under study are mostly used uncooked. Pesticides and heavy metals are the part of majority of contaminants of food supply and may be considered as the most important problem to our environment. The present investigation determined the pesticide residues and heavy metals in various fruits and vegetables samples collected from different markets of Ghana and compared them with the limits set by the EC Directive (2006) and Extension Toxicology Network (2011).

Levels of pesticide residues

Table 1 gives the scientific, English and the incidence of pesticide residues in fruits and vegetables samples analyzed. Residues occurred in 58 and 49 % of all fruits and vegetables samples, respectively. The mean concentrations and range of pesticide residues (organochlorine and pyrethroid) found in fruits and vegetables sampled from the local markets in Ghana are summarized in Table 4 and 5.

Chlorinated pesticide residues in fresh fruits and vegetables

Fresh fruits and vegetables studied in this research are produced and consumed locally with no or minimal preparation which may constitute an important potential source of pesticide residues. Washing under running water is most commonly the only treatment given to many fruits and vegetables prior to consumption.

Among the detected organochlorine pesticide residues, the results showed that gamma-HCH in some commodities were between 0.008 mg/kg in okra and 0.133 mg/kg in pineapple with a range of 0.006-0.012 and 0.121-0.153, respectively (Table 4). The presence of an average mean sum of 0.055 mg/kg was recorded during the entire study. This high level might be connected with the extensive use of technical lindane (gamma-HCH) which is marketed as Gammalin 20 and used by some farmers for agricultural purposes for crop protection in Ghana. This result is comparable with mean concentration level of 0.002 and 0.004 mg/kg reported in Nigeria and India markets respectively (Bhanti and Taneja, 2007; Adeyeye and Osibanjo, 1999).

A mean value of 0.004mg/kg and 0.041 mg/kg of methoxychlor was achieved in mango and onion samples, with range of 0.004-0.006 and 0.025-0.066, respectively. A mean sum value of was 0.016 mg/kg achieved for all the tested commodities. Methoxychlor concentration in fruit and vegetable samples was a little high, perhaps being as a result of historical use of DDT of which technical methoxychlor contains about 88 % of the p,p'-isomer together with more than 50 structurally related contamination, which might have been added to the actual amount of methoxychlor present (Bempah and Donkor,2011).

The presence of an average value of 0.006 mg/kg in pineapple and 0.040 mg/kg of aldrin in tomato with range values of 0.004-0.008 and 0.080-0.015, respectively recorded in the analyzed samples declare that, there may be conversion of aldrin to dieldrin by an epoxidation in biological systems (Rumsey and Bond, 1974) and, therefore dieldrin is expected to be found in relatively higher levels than aldrin. Hence, concentration levels of dieldrin varied from 0.010 mg/kg in cucumber and 0.090 mg/kg in banana with a range of 0.005-0.013and 0.013-0.203, respectively. A mean sum value of 0.016 mg/kg and 0.037 mg/kg was recorded for aldrin and dieldrin, respectively.

Endrin was recorded in 0.004 mg/kg in pineapple with a range of 0.004-0.008 and 0.040 in carrot with a range of 0.080-0.015. Endrin recorded low mean sum value of 0.016 mg/kg in all the tested commodities. The low level of endrin was as a result of its susceptibility to volatilization, photodegradation, and heat to form metabolites of endrin (Fan and Alexeeff, 1999).

p,p'-DDE were between 0.004 mg/kg in watermelon and 0.173 mg/kg in lettuce with range of 0.004-0.008 and 0.050-0.400, respectively while p,p'-DDT contents varied from 0.006 mg/kg and 0.050 mg/kg in tomato and lettuce samples with a range of 0.004-0.008 and 0.022-0.078 in tomato and lettuce samples, respectively. The presence of mean sum values of 0.039 and 0.023 mg/kg for p,p'-DDE and p,p-DDT was recorded in all the tested fruit and vegetable samples, respectively. The low level of technical DDT might due to Dehydrochlorination and/or photodegradation to form metabolite form of DDE (Bempah and Donkor, 2011)

The results corroborate the findings of Abou-Arab and Abou Donia, 2001 who detected levels of DDT and its derivatives at levels ranging from 0.009 to 0.116 mg/kg and mean value of 0.05 mg/kg in West African city farms (Manirakiza *et al*., 2003).

Among the different organochlorine pesticide residues found in fruit and vegetable samples, p,p'-DDE was found with the highest concentration of 0.173 mg/kg in lettuce, followed by gamma-HCH (0.133 mg/kg) in pineapple, dieldrin (0.090 mg/kg) in banana, p,p'-DDT (0.050 mg/kg) in lettuce, methoxychlor (0.041 mg/kg) in onion, aldrin (0.040 mg/kg) and endrin (0.040 mg/kg) in tomato and carrot samples, respectively.

Synthetic pyrethroid pesticide residues in fresh fruits and vegetables

The levels of five pyrethroid residues in fruits and vegetables collected from local markets in Ghana are illustrated in Table 5. The results showed that, the levels of permethrin in all the commodities were between 0.006 mg/kg in pear and 0.090 mg/kg in lettuce with range of 0.004-0.008 and 0.011-0.051, respectively. Cyfluthrin contents varied from 0.008 mg/kg in watermelon and 0.020 mg/kg in pineapple with a range of 0.004-0.010 and 0.018-0.021, respectively. Residual levels of cypermethrin detected in the samples are 0.004 mg/kg in pear and 0.060 mg/kg in lettuce with a range of 0.004-0.008 and 0.030-0.080, respectively. The levels of fenvalerate detected in the commodities varied between 0.006 mg/kg in carrot and 0.002 mg/kg in papaya with range of 0.004-0.008 and 0.018-0.024, respectively while the levels of deltamethrin varied between 0.008 mg/kg in pear and 0.044 mg/kg in pineapple with range of 0.007-0.010 and 0.026-0.062, respectively.

The data in Table 5 further revealed that, permethrin was found with the highest concentration of 0.090 mg/kg in lettuce, followed by cypermethrin with concentration of 0.060 mg/kg in pineapple, deltamethrin with concentration of 0.044 mg/kg in pineapple, fenvalerate with concentration of 0.037 mg/kg in onion and cyfluthrin with a concentration of 0.020 mg/kg in pineapple.

The results of the present investigation further support the findings of the study carried out by Kumar *et al*., (2006), Andersen, (2001), Pang *et al*., 1995 who detected different levels of pyrethroid pesticides in both fruits and vegetables in India, Denmark and China markets respectively.

Levels of heavy metals

The mean concentrations and range of heavy metals found in fresh fruits and vegetables sampled from the local markets in Ghana are summarized in Table 7.

Heavy metal concentrations determined were based on sample dry weight. The results showed that, the levels of Pb in all the fruit and vegetable samples were between below detection limit in watermelon and 0.56 mg/kg in lettuce with a range of 0.48-0.63 mg/kg. Cd contents varied from no detectable amount in watermelon and banana to 0.08 mg/kg (0.02-0.12 mg/kg) in lettuce. This contamination level of Pb and Cd might have occurred due to irrigation with contaminated water and Pb aerosol resulting from emission from automobile/industrial exhaust. It is interesting to note that lettuce received the high concentration values for Pb and Cd. 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. The low level of Cd was as a result of the presence of sufficient amount of Zn which neutralizes the Cd contents. Hence low values were recorded in cabbage samples.

The concentration levels of Cu varied between 1.24 mg/kg (0.68-1.84) in mango to 6.32 mg/kg (4.54-7.48) in cucumber. The high levels of Cu content might due to increase in application of micronutrients fertilizers and copper-based fungicides. Plants generally contain Cu which is inadequate for its normal growth. However such application of micronutrients and copper-based fungicides can lead to its alarming levels which might be detrimental to human health (Radwan and Salama, 2006). Cu is necessary for the body pigmentation in addition to Fe, the maintenance of a healthy central nervous system, prevention of anemia and is interrelated with the function of Zn and Fe in the body (Akinyele and Osibanjo, 1982). Hence its residual concentration ought to be checked in order not to exceed its required levels.

Minimum concentration value for Zn (2.32 mg/kg) was found in papaya with a range of 1.46-3.22 and maximum concentration value of 21.4 mg/kg was detected in cucumber with range of 18.44-23.7. Zn is essential for plants and animals. These elevated levels of Zn will interfere with the physiological processes of plants and animals Corresponding value for Cr is no detectable amount in pineapple to 0.55 mg/kg in okra.

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).

Health risk estimates for organochlorine pesticides

Table 8 represents the estimation of organochlorine pesticide residue intake through consumption of the studied foodstuffs. Data of the average diet per person per day were collected from International Food Policy Research Institute data, 2004. The average diet per person per day of fruits and vegetables are 0.064 and 0.137 kg, respectively. If the obtained mean levels of gamma-HCH (0.051 mg/kg), methoxychlor (0.057 mg/kg), aldrin (0.010 mg/kg), dieldrin (0.040 mg/kg), endrin (0.005 mg/kg), p,p'-DDE (0.007m/kg) and p,p'-DDT (0.020 mg/kg) are consumed daily, the contribution of fruits to pesticide intake is 0.003 mg, 0.004 mg, 0.001 mg, 0.003 mg, 0.000mg, 0.000 mg and 0.001 mg, respectively. In the case of vegetables, if the consumed daily mean levels of gamma-HCH, methoxychlor, aldrin, dieldrin, endrin, p,p'-DDE and p,p'-DDT are 0.059, 0.019, 0.020, 0.031, 0.023, 0.063 and 0.010 mg/kg, respectively, the corresponding estimated daily intake will be 0.008 mg, 0.003 mg, 0.003 mg, 0.004 mg 0.003 mg, 0.009 mg and 0.001 mg, respectively. It could be concluded that our estimated daily intake for almost all the studied organochlorine pesticide residues are far below those reported by the Extension Toxicology Network who has set a limit for organochlorine pesticide residue intake based on body weight with the exception of aldrin and dieldrin in fruits and aldrin, dieldrin and endrin in vegetables, respectively. The acceptable daily intake for gamma-HCH, methoxychlor, endrin, p,p'-DDE and p,p'-DDT are 0.008 mg, 0.1 mg, 0.0001 mg, 0.02 mg and 0.02 mg, respectively while aldrin and dieldrin the permissible limit are set at 0.0001 mg (Extension Toxicology Network, 2011). Thus, the consumption of average amounts of these foodstuffs does not pose a health risk for the consumer except aldrin and dieldrin which exceeded permissible limit in fruits and aldrin, dieldrin and endrin in vegetables, respectively suggesting a great potential for systemic toxicity in consumers especially children considered the most vulnerable population subgroup.

Health risk estimates for synthetic pyrethroid pesticides

For the monitored synthetic pyrethroid pesticides, the obtained mean levels of permethrin (0.024 mg/kg), cyfluthrin (0.013 mg/kg), cypermethrin (0.016 mg/kg), and deltamethrin (0.021 mg/kg) are consumed daily, the contribution of fruits to metal intake is 0.015 mg, 0.001 mg, 0.001 mg, and 0.001 mg, respectively. In the case of vegetables, if the consumed daily mean levels of permethrin, cyfluthrin, cypermethrin and deltamethrin are 0.036, 0.012, 0.023 and 0.023 mg/kg, respectively, the corresponding estimated daily intake will be 0.005 mg, 0.002 mg, 0.003 mg and 0.003 mg, respectively. It could be concluded that our estimated daily intake for the studied pyrethroid pesticide residues are far below those reported by the who has set a Extension Toxicology Network limit for pyrethroid pesticide residues intake based on body weigh. However, permissible limit for fenvalerate was not available, hence its exclusion from the data (Table 9). The acceptable daily intake for permethrin, cyfluthrin, cypermethrin and deltamethrin are 0.05 mg, 0.02 mg, 0.05 mg and 0.01 mg, respectively (Extension Toxicology Network, 2011). Thus, the consumption of average amounts of these foodstuffs does not pose a health risk for the consumer.

Health risk estimates for heavy metals

Table 10 represents the estimation of each heavy metal intake through consumption of the studied foodstuffs. If the obtained mean levels of Pb (0.06 mg/kg), Cd (0.2 mg/kg), Cu (2.25 mg/kg), Zn (95.8 mg/kg) and Cr (0.33 mg/kg) are consumed daily, the contribution of fruits to metal intake is 0.004 mg, 0.013 mg, 0.144 mg, 0.371 mg and 0.021 mg, respectively. In the case of vegetables, if the consumed daily mean levels of Pb, Cd, Cu, Zn and Cr are 0.29, 0.04, 2.96, 11.47 and 0.74 mg/kg, respectively, the corresponding estimated daily intake will be 0.040 mg, 0.005 mg, 0.406 mg, 1.571 mg and 0.101 mg, respectively. It could be concluded that our estimated daily intake for the studied heavy metals are far below those reported by the FAO/WHO who has set a limit for heavy metals intake based on body weight for an average adult (60 kg body weight) except Cr whose permissible limit was not available. Provisional tolerable daily intake for Pb, Cd, Cu and Zn are 214 ug, 60 ug, 3 mg and 60 mg, respectively (Joint FAO/WHO Expert Committee on Food Additives, 1999). Thus, the consumption of average amounts of these foodstuffs does not pose a health risk for the consumer.

Conclusion

The survey revealed high occurrence of pesticide residues and heavy metals but low residue levels especially in synthetic pyrethroid pesticides and heavy metals in fruits and vegetables. However, most of the monitored pesticides and heavy metals were far below the prescribed International residue limits. Nevertheless, the high prevalence of contamination is worrisome considering the cumulative nature and level of persistence in most monitored pesticides and heavy metals.

Our estimated daily intake rate for all the pesticides and heavy metals were far below the tolerable daily intake rate except for aldrin and dieldrin in fruits and aldrin, dieldrin and endrin in vegetables.

Monitoring programs of pesticide residues and heavy metals in fruits and vegetables have shown that presence of these residues in the majority of analyzed samples occur at concentration below the established tolerance levels 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 regular monitoring of a greater number of samples for a long period is necessary and consumption of contaminated fruits and vegetables should be avoided in order to reduce the health risk caused by taking contaminated fruits and vegetables.

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Table 1: Names of samples and incidence of pesticide residues in fruits and vegetables from Kumasi markets

English name	Scientific name	No. of samples	% with one or more residues
Fruits			
Papaya	*Carica papaya*	20	83
Watermelon	*Citrullus lanatus*	15	50
Banana	*Musa sapientum*	25	67
Mango	*Mangifera indica*	25	58
Pear	*Pyrus communis*	20	42
Pineapple	*Ananas sativus*	15	75
Vegetables			
Tomato	*Lycopersicon esculentus*	30	67
Lettuce	*Lactuca sativa*	30	75
Cabbage	*Brassica oleracea*	25	83
Carrot	*Daucus carota*	25	67
Okra	*Hibiscus esculentus*	30	50
Green pepper	*Piper nigrum*	30	50
Onion	*Allium cepa*	30	58
Cucumber	*Cucumis sativus*	30	58

Table 2: Analytical recoveries (%) ± SD of seven organochlorine and five pyrethroid pesticides in carrot samples at 0.01, 0.02 and 0.1 mg/kg fortification levels (n=3)

Pesticides	Analytical recoveries (%) ± SD at different fortification levels (mg/kg)		
	0.01	0.02	0.1
Gamma-HCH	98.2±6.2	97.5±4.7	93.4±9.6
Methoxychlor	87.3±9.4	87.0±8.5	92.5±10.1
Aldrin	92.4±10.5	96.4±5.3	94.0±4.5
Dieldrin	97.8±5.1	88.9±10.5	91.5±7.2
Endrin	97.0±5.6	93.1±5.3	97.2±4.3
p,p'-DDE	92.3±9.8	91.6±10.4	92.8±10.6
p;p'-DDT	95.1±7.1	95.4±7.3	94.1±3.9
Permethrin	88.0±10.2	88.6±10.2	92.1±9.7
Cyfluthrin	98.3±7.1	96.4±5.5	97.4±5.4
Cypermethrin	102.3±10.3	98.1±7.3	101.3±8.7
Fenvalerate	104.0±5.5	103.8±3.8	98.7±6.4

SD- Standard deviation

Table 3: 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 5: Synthetic pyrethroid pesticide residues (mg/kg fresh wt) in fruits and vegetables

Commodity	Concentration and range of pesticides									
	Permethrin		Cyfluthrin		Cypermethrin		Fenvalerate		Deltamethrin	
	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range
Fruits										
Papaya	0.015±0.015	0.005-0.032	<LOD	-	0.035±0.005	0.030-0.040	0.020±0.002	0.018-0.024	<LOD	-
Water melon	0.040±0.033	0.080-0.025	0.008±0.002	0.004-0.010	<LOD	-	<LOD	-	0.015±0.011	0.007-0.023
Banana	<LOD[a]	-	0.010±0.010	0.006-0.022	0.012±0.006	0.007-0.012	<LOD	-	0.016±0.021	0.008-0.040
Mango	0.016±0.021	0.002-0.031	<LOD	-	0.008±0.002	0.004-0.012	0.008±0.004	0.006-0.014	<LOD	-
Pear	0.006±0.003	0.004-0.008	<LOD	-	0.004±0.002	0.004-0.008	<LOD	-	0.008±0.002	0.007-0.010
Pineapple	0.041±0.022	0.025-0.066	0.020±0.002	0.018-0.021	0.022±0.003	0.019-0.025	<LOD	-	0.044±0.018	0.026-0.062
Vegetables										
Tomato	0.020±0.002	0.018-0.021	<LOD	-	0.016±0.005	0.006-0.020	<LOD	-	0.013±0.005	0.011-0.017
Lettuce	0.030±0.009	0.011-0.035	<LOD	-	0.060±0.026	0.030-0.080	0.013±0.007	0.009-0.016	0.016±0.021	0.006-0.024
Cabbage	0.049±0.026	0.022-0.078	0.016±0.014	0.010-0.018	<LOD	-	0.011±0.010	0.003-0.017	0.010±0.010	0.009-0.020
Carrot	0.037±0.013	0.030-0.052	<LOD	-	0.014±0.002	0.010-0.018	0.006±0.002	0.004-0.008	0.040±0.033	0.015-0.063
Okra	<LOD	-	0.011±0.010	0.007-0.021	0.008±0.006	0.006-0.014	0.010±0.008	0.006-0.016	<LOD	-
Green pepper	0.007±0.003	0.005-0.013	<LOD	-	<LOD	-	0.010±0.005	0.007-0.015	0.023±0.008	0.022-0.034
Onion	<LOD	-	0.009±0.003	0.005-0.013	<LOD	-	0.018±0.006	0.012-0.022	0.038±0.030	0.012-0.045
Cucumber	0.012±0.008	0.008-0.016	<LOD	-	0.009±0.003	0.006-0.013	0.010±0.006	0.007-0.013	<LOD	-
∑ Mean level	0.030		0.012		0.019		0.017		0.022	

Each value is the mean of five samples with three determinations
[a]LOD (Limit of detection) = 0.001 mg/kg sample
SD-Standard deviation
Values designated by asterisks are higher than the EC-MRLs for the respective pesticide (see MRLs in Table 6)

Table 4: Organochlorine pesticide residues (mg/kg; fresh wt) in fruits and vegetables

Commodity	Concentration and range of pesticides													
	Gamma-HCH		Methoxychlor		Aldrin		Dieldrin		Endrin		p,p'-DDE		p,p'-DDT	
	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range	Mean ± SD	Range
Fruits														
Papaya	0.100*±0.004	0.092-0.105	0.006±0.002	0.004-0.012	0.013±0.007	0.009-0.019	0.017*±0.020	0.002-0.040	<LOD	-	<LOD	-	0.012±0.006	0.008-0.014
Water melon	0.004±0.002	0.004-0.006	<LOD	-	<LOD	-	<LOD	-	<LOD	-	0.004±0.001	0.004-0.008	0.008±0.004	0.006-0.010
Banana	<LOD[a]	-	0.008±0.004	0.004-0.012	<LOD	-	0.090*±0.103	0.013-0.203	0.006±0.002	0.004-0.012	<LOD	-	0.038±0.032	0.005-0.062
Mango	0.010±0.010	0.006-0.022	0.004±0.001	0.004-0.006	<LOD	-	<LOD	-	<LOD	-	0.010±0.004	0.005-0.011	0.020±0.002	0.018-0.021
Pear	0.009±0.003	0.006-0.012	<LOD	-	0.012±0.009	0.010-0.016	<LOD	-	<LOD	-	<LOD	-	<LOD	-
Pineapple	0.133*±0.014	0.121-0.153	0.031*±0.023	0.007-0.052	0.006±0.002	0.004-0.008	0.012*±0.008	0.007-0.018	0.004±0.002	0.004-0.008	<LOD	-	<LOD	-
Vegetables														
Tomato	0.129*±0.013	0.104-0.155	0.016*±0.005	0.010-0.017	0.040*±0.033	0.080-0.015	<LOD	-	<LOD	-	0.190*±0.051	0.016-0.020	0.006±0.002	0.004-0.008
Lettuce	0.040*±0.035	0.080-0.015	0.006±0.003	0.005-0.007	0.011±0.010	0.009-0.021	<LOD	-	<LOD	-	0.173*±0.193	0.050-0.400	0.050±0.005	0.022-0.078
Cabbage	0.100*±0.004	0.095-0.102	<LOD	-	<LOD	-	0.035*±0.013	0.030-0.052	0.007±0.003	0.005-0.009	0.008±0.004	0.006-0.010	0.032±0.010	0.030-0.040
Carrot	<LOD	-	0.008±0.004	0.006-0.012	0.010±0.021	0.008-0.040	<LOD	-	0.040*±0.035	0.080-0.015	<LOD	-	0.009±0.003	0.005-0.013
Okra	0.008±0.010	0.006-0.012	<LOD	-	<LOD	-	<LOD	-	0.023*±0.008	0.016-0.031	0.011±0.010	0.008-0.021	0.037±0.013	0.030-0.052
Green pepper	<LOD	-	0.016*±0.014	0.006-0.032	0.020*±0.002	0.018-0.021	0.058*±0.005	0.052-0.062	<LOD	-	<LOD	-	<LOD	-
Onion	0.019*±0.002	0.016-0.020	0.041*±0.022	0.025-0.066	<LOD	-	<LOD	-	<LOD	-	0.023±0.008	0.016-0.031	0.035±0.005	0.030-0.040
Cucumber	<LOD	-	0.020±0.002	0.018-0.021	<LOD	-	0.010±0.004	0.005-0.013	<LOD	-	0.070*±0.015	0.053-0.082	0.004±0.001	0.004-0.008
∑ Mean level	0.055		0.016		0.016		0.037		0.016		0.039		0.023	

Each value is the mean of five samples with three determinations

[a]LOD (Limit of detection) = 0.001 mg/kg sample

SD-Standard deviation

Values designated by asterisks are higher than the EC-MRLs for the respective pesticide (see MRLs in Table 6)

Table 6: Maximum residue levels of the tested pesticides

8	Maximum residue levels, MRLs (mg/kg)											
	Gamma-HCH	methoxychlor	aldrin	dieldrin	endrin	p,p-DDE	p,p'-DDT	Permethrin	Cyfluthrin	Cypermethrin	Fenvelerate	Deltamethrin
Fruits												
Papaya	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.02	0.05	0.02	0.05
Water melon	0.01	0.01	0.03	0.03	0.01	0.05	0.05	0.05	0.02	0.20	0.02	0.05
Banana	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.02	0.05	0.02	0.05
Mango	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.02	0.05	0.02	0.05
Pear	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.20	1.00	0.05	0.10
Pineapple	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.02	0.05	0.02	0.05
Vegetables												
Tomato	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.05	0.50	0.05	0.20
Lettuce	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.50	2.00	0.02	0.50
Cabbage	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.20	0.50	0.05	0.10
Carrot	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.02	0.05	0.02	0.05
Okra	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.02	0.50	0.02	0.20
Green pepper	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.30	0.50	0.02	0.20
Onion	0.01	0.01	0.01	0.01	0.01	0.05	0.05	0.05	0.02	0.10	0.02	0.10
Cucumber	0.01	0.01	0.02	0.02	0.01	0.05	0.05	0.05	0.10	0.20	0.02	0.10

Table 7: Heavy metals content (mg/kg) in fruits and vegetables

Food items	Concentration and range of metals									
	Pb		Cd		Cu		Zn		Cr	
Fruits	Mean±SD	Range	Mean±SD	Range	Mean±SD	Range	Mean±SD	Range	Mean±SD	Range
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	0.38±0.08	0.14-0.42
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	-
Vegetables										
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	1.17±0.11	0.55-1.93
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	0.59±0.09	0.23-0.77
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	1.07±0.07	0.03-1.15
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	1.71±0.09	1.13-2.19

Each value is the mean of five samples with three determinations
SD- Standard deviation
LOD= Limit of detection (See table 3)

Table 8: Estimation of organochlorine pesticide residue intake through consumption of fruits and vegetables in Ghana

Food stuffs	Mean consumption (kg/person/day)	Gamma-HCH		Methoxychlor		Aldrin		Dieldrin		Endrin		p,p'-DDE		p,p'-DDT	
		Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day
ruits	0.064	0.051	0.003	0.057	0.004	0.010	0.001	0.040	0.003	0.005	0.000	0.007	0.000	0.020	0.001
'egetables	0.137	0.059	0.008	0.019	0.003	0.020	0.003	0.031	0.004	0.023	0.003	0.063	0.009	0.010	0.001

Table 9: Estimation of synthetic pyrethroid pesticide residue intake through consumption of fruits and vegetables in Ghana

Food stuffs	Mean consumption (kg/person/day)	Permethrin		Cyfluthrin		Cypermethrin		Deltamethrin	
		Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)
Fruits	0.064	0.024	0.015	0.013	0.001	0.016	0.001	0.021	0.001
Vegetables	0.137	0.036	0.005	0.012	0.002	0.023	0.003	0.023	0.003

Table 10: Estimation of heavy metals intake through consumption of fruits and vegetables in Ghana

Food stuffs	Mean consumption (kg/person/day)	Pb		Cd		Cu		Zn		Cr	
		Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)	Mean level (mg/kg)	Intake (mg/day)
Fruits	0.064	0.06	0.004	0.2	0.013	2.25	0.144	5.8	0.371	0.33	0.021
Vegetables	0.137	0.29	0.040	0.04	0.005	2.96	0.406	11.47	1.571	0.74	0.101