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Exposure and Risk Assessment of Selected Chemical Hazards in Cabbage and Lettuce

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Abstract

The objective of this study was to determine health risks associated with consumption of cabbage and lettuce sampled in an urban industrialized study area. Samples were initially digested in Teflon vessels using HNO_3 and H_2O_2, homogenized in Na_2SO_4, agitated in a mixture of hexane/acetone, cleaned-up and analyzed in ICP - MS in order to quantify toxic heavy metals. The PAH content was also quantified in GC-MS against reference PAH congeners. Food frequency questionnaire designed based on the US EPA format, was used to collect elements of chronic daily intake and information on consumption characteristics of the selected samples. The data was processed in Palisade@Risk-based Microsoft Excel, where distributions were fitted for the elements of chronic daily intake. Exposure quantifications of hazards based on the US EPA guidelines, presented hazard quotients and risks, relative to the respective reference doses and cancer slopes factors (CSF). Incremental lifetime cancer risks (ILTCR) were determined as the product of the CSF of the carcinogens and their chronic exposures. The modal and median mercury exposures were low but the exposures of total PAHs (expressed as benzo[a]pyrene -BaP) were extremely high. The hazard quotients revealed that cadmium and mercury had negligible risks (HQ ≤ 1). Though the median and modal ILTCR showed lead to have low health risk concerns (≤ 10^{-6}), the uncertainty ranging from 0 to 3 out-10-thousand consumers revealed insidous risk unacceptability. Similarly, even though the modal and median values of ILTCR of arsenic appeared acceptable (≤ 10^{-6}), the risk uncertainty ranging from 0 to 6 out-1-thousand consumers, still raise health concerns. The median and modal ILTCR values of BaP suggested both a cautiously acceptable risk (≥ 10^{-6} ≤ 10^{-4}) and a barely negligible risk (≤ 10^{-6}) levels respectively. Thus, every effort must be made to mitigate the seriously high risks lurking in the study area.

Keywords

Heavy metals, PAH, Hazard quotient, Incremental litime cancer risk, Vegetables

Introduction

Many health-conscious consumers are increasingly making deliberate choices for plant-based diets because probably they think they were safer. Majority of these consumers who dwell in cities therefore demand leafy vegetables for their daily consumption. The demand has created the opportunity for many urban dwellers to join the business of vegetable cultivation using every space available. Urbanization is associated with industrialization, and with it comes the attendant anthropogenic hazards. Subsequently, these activities contaminate the soil with toxic heavy metals such as arsenic (As), cadmium (Cd), mercury (Hg) and lead (Pb) [1,2]. Although, some studies indicate low concentration of such hazards in vegetables [3,4], their bioaccumulation is likely to pose significant threat due

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to bioaccumulation [5]. There is also vehicular exhausts and industrial incinerators that discharge organic pollutants (PAHs) into the atmosphere that eventually enter our food chain.

The principles of hazard identification explain how toxic metals and PAHs manifest disease end-points after consumption. Cadmium and lead have been implicated in renal failure, reproductive system dysfunctions, cardio vascular system distress and demineralization agent of bone [6-10]. Mercury has also been documented as damaging the central nervous system and also causing allergic contact dermatitis [11,12]. On the other hand, polyaromatic hydrocarbons (PAHs), arsenic and lead have been reported to be carcinogenic in humans and experimental animals [7,13,14]. Indeed, the Environmental Protection Agency's Integrated Risk Information System (IRIS) and other standard organizations [15,16] have documented the reference doses and slope factors of such hazards. Such databases enable researchers to characterize health risks of ingested such hazards using indices as hazard quotients (HQ), margin of exposure (MoE) and incremental lifetime cancer risks (ILTCR).

There are procedurally straight-forward guidelines that show how exposures of hazards must be quantified. Average daily dose (ADD) has been explained as the quantities of hazards ingested per body weight of consumers per day [17]. Thus, when the ADD is integrated with the consumption level (the product of exposure frequency (EF) and exposure duration (ED) per averaging time (AT)), the exposure, called chronic daily intake (CDI) is obtained. It is when the exposures exceed the reference doses of the hazards (HQ > 1), that it is said that consumers are at adverse health risk. Studies have shown that there is no acceptable exposure to a carcinogen [18]. Thus, when the exposures of the hazard (carcinogen) is multiplied by the cancer slope factor (potency factor), then the ILTCR is obtained [17]. These risk values are the expressions of the theoretical maximum number of cancer cases that are expected to develop when consumers are exposed to carcinogens. It has been indicated that when the ILTCR value exceeds 10^{-4} (1 case out-of-10 thousand consumers) it poses health concerns; thus, it is regarded as unacceptable [19]. Comparatively, risks below the de minimis value of 1×10^{-6} (1 case out-of-1 million consumers) are considered to pose no significant adverse health risks. Those values in-between 1×10^{-4} and 1×10^{-6} are regarded as a cautiously acceptable since the risk largely depend on the population of consumers and also on the circumstances of exposure [19]. PAHs are often bonded to particulate matter thus, air current carry them and contaminate cultivated food products. Many of the congeners of PAH pose health risks because they are usually non-biodegradable [20,21]. It is therefore, important to investigate the levels of toxic heavy metals and PAHs in these vegetable samples. The objective of the study therefore sought to estimate exposures and risks of some selected toxic dietary heavy metals (As, Cd, Pb, Hg) and PAHs through cabbage and lettuce consumption.

Materials and Methods

Materials

Sample collection: The sampling sites for the selected samples consisted of vegetable farms, market centers and eateries such as street food vending sites, restaurants and cafeterias. Suburbs within the sampling sites included Atonsu, Chirepatre, Tek, Tanoso, Ayigya, Kejetia, Kwadaso, Santasi, Aboabo, Asafo and Bantama. Others were Adum, Airport Roundabout, Fante New Town and Suame. Fresh cabbage (*Brassica oleracea*) and lettuce (*Lactuca sativa*) were randomly sampled from selected farms and market centers in Kumasi. Ready-to-eat cabbage-lettuce salads were also randomly sampled from selected street food vending sites and cafeterias within the same sites. The raw samples were washed and subsequently stored in labeled zip-lock bags kept refrigerated at 4 °C until analyzed.

Standards and reagents: The digestion reagents; nitric acid and hydrogen peroxide were purchased from BDH Chemical Ltd., UK. Analytical grade heavy metals standards were purchased from Sigma Aldrich (St. Louis, Missouri, USA). Standards of PAH congeners were purchased from Panreac (Barcelona, Spain).

Methods

Survey: Food frequency questionnaire was designed and used to collect data from consumers of the selected samples in the study area based on the USEPA recommended elements of chronic daily intake. Information on mass of the selected samples consumed (obtained as amounts purchased) and the body weights of consumers were collected. Information relating to the consumption levels, defined as the product of the exposure frequency (EF) within a year and the exposure duration (ED) in years, per averaging time (AT) of consumption of the selected samples were collected. Subsequently, the consumption data relating to exposures to hazards from the selected samples was determined.

Sample preparation: Five hundred grams (500 g) of lettuce and cabbage each obtained from farms and market centers were weighed and blended together in a 50:50 ratios according to how they were served to customers. Only edible parts of these vegetables were used for the analysis according to the usual preparation practices. Similarly, five hundred grams (500 g) of the already prepared samples obtained from the eateries, were weighed and blended.

Determination of heavy metals in the selected samples

In this study, method 29 of the US geological survey

digestion procedure [22] was used. The samples (1.0 g) were weighed directly into Teflon digestion vessels and 4 mL of nitric acid (HNO_3) and 2 mL of hydrogen peroxide (H_2O_2) added in sequence. The mixture was subsequently subjected to Murphy Richards Microwave (MM820CXN-PM0F, UK) digestion system and held at a period of 25 min to reach 200 °C. The resulting digested solutions, together with the reagent blank digests, were diluted to 10 mL with de-ionized water. The samples were then analyzed in Inductively Coupled Plasma - Mass Spectrometry (ICP-MS) as previously described [23]. Digested Certified Reference Material (CRM) was used as reference material. The limits of detection (LOD) of the samples were 0.25 µg/g for arsenic; 0.024 µg/g for cadmium; 0.350 µg/g for lead and 0.010 µg/g for mercury.

Determination of polycyclic aromatic hydrocarbon in the selected samples

For the polycyclic aromatic hydrocarbons, a digestion method as described by Ciecierska and Obiedzinski [24] was used. Ten grams (10 g) each of the samples were homogenized with anhydrous sodium sulphate (20 g) and transferred into an Erlenmeyer flask. A mixture of hexane: acetone (60:40, v/v) (100 mL) was added and the flask agitated in Grant ultrasonic bath (XB3, England) for 30 min. Subsequently, the extract was filtered, and the filtrate evaporated and the residue dissolved in 5 mL cyclohexane: ethyl acetate (50:50 v/v). After filtering again through polytetrafluoroethylene (PTFE) (pore size of 1 µm), a clean-up was carried out in gel permeation chromatography (GPC) to obtain the purified PAHs. To screen the PAH against the 16 PAH congeners, injector volumes (1.0 µL) of the extracts were subsequently analyzed in Hewlett-Packard Varian GC-MS (CP-3800, Germany), with positive-ion electron ionization option used in the selected ion monitoring mode. The GC-MS analysis was equipped with fused silica capillary column 30 m × 0.25 mm internal diameter, coated with VF-1701 ms (0.25 µm film). The reference PAH congeners used included acenaphthene, anthracene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(k) fluoranthene, benzo(a)pyrene, benzo(g,h,i) perylene and chrysene. Others were fluoranthene, fluorene, indenol (1,2,3-c,d)pyrene, naphthalene, 1-methylnaphthalene, 2- methylnaphthalene, phenanthrene and pyrene. The column oven temperature was programmed to run from 60 °C to 180 °C then to 210 °C and finally to 340 °C respectively, at rates of 8.5 and 2 °C/min and held at 2 min. Purified nitrogen gas was used as carrier gas at a flow rate of 1.0 mL/min and as the make-up gas of 29 mL/min. The injector and detector temperatures were maintained at 210 °C and 340 °C respectively. For the mass spectrometer, the settings used were electronic ionization (EI) positive ion, closed EI for ion volume and 250 °C for ion source temperature. Emission current was 50 µA with full scan in range m/z 45-650 using 0.15 as scan width. PAHs were quantified using MS within a single chromatographic run while simultaneously hydrocarbon profile was monitored in a full scan in the GC. All reagents used during the analysis were conditioned to the same extraction procedures. Solvents used were run to verify any interfering substances within the runtime. Reagent blanks and spiked samples were used for quality control checks. Fortification level of 0.05 µg/g was chosen based on the limit of determination. LOD and LOQ values which were determined based on calibration standards presented values ranging between 0.002 and 0.98 µg/g. The RSD values were less than 4.7%, and the calculated r^2 values were above 0.998. Recoveries were calculated from the differences in total amounts of each PAH between the spiked and unspiked samples and were between 80-120%.

Data analysis and risk assessment

The data obtained from the local food frequency questionnaire survey and the determinations of the hazards, were captured into Microsoft Excel (2014). Initially, the Palisade@Risk software [25] was used to fit distributions for the hazards, contact rate (CR) (total mass of samples consumed per day), exposure frequency (EF), exposure duration (ED) and body weights (BW) of the respondents. Based on their distributions, all the variables were integrated in the [26] recommended Equations 1 and 2, from where the dietary exposure-chronic daily intake (CDI) and the ILTCR, were run in the risk software, iterating at 100,000 times.

$$\mathrm{CDI} = \frac{\mathrm{C} \times \mathrm{CR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \quad (1)$$

$$\mathrm{ILTCR} = \frac{\mathrm{C} \times \mathrm{CR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \times \mathrm{PF} \quad (2)$$

$$\text{Hazard Quotient} = \frac{\mathrm{CDI}}{\mathrm{R_f D}} \quad (3)$$

Recommended averaging times (AT) of 30 and 70 years respectively, were used for the toxic and carcinogenic hazards based the epidemiological projected periods of disease end-points [17]. The screening of the selected samples revealed several types of PAHs thus, a reported Toxic Equivalent Factors (TEF) values (Table 1) were used to sum their concentrations into the recommended benzo[a]pyrene (BaP) additive values [27].

To characterize the carcinogenic risks of arsenic, lead and BaP, their respective oral potency factors (PF) of 1.5 $(mg/kg\text{-}day)^{-1}$ [26]; 0.0085 $(mg/kg\text{-}day)^{-1}$ [28] and 7.3 $(mg/kg\text{-}day)^{-1}$ [29] were used. Similarly, to characterize

Table 1: Toxic equivalent factors used for converting PAHs congeners in vegetables into benzo[a]pyrene.

PAH (Peak name)	Toxic Equivalency Factors (TEFs)
Benzo(b)fluoranthene	0.1
Benzo(k)fluoranthene	0.1
Indenol(1,2,3-c,d)pyrene	0.1
Benzo(g,h,i)perylene	0.01

Table 2: The concentrations (µg/g) of toxic heavy metals in vegetable samples (n = 16) collected from the study area.

Sampling sites	Suburbs	Toxic heavy metals			
		As	Cd	Pb	Hg
	Atonsu	1.998	0.836	4.707	< LOD
Farms	Chirepatre	2.938	0.084	3.809	< LOD
	KNUST	2.585	0.191	4.002	< LOD
	Tanoso	0.294	0.275	2.870	< LOD
	Ayigya	1.410	0.191	2.943	0.020
Markets	Kejetia	1.293	0.287	3.661	< LOD
	Kwadaso	3.114	0.191	3.419	< LOD
	Santasi	1.293	0.550	2.585	< LOD
Street Food Vending Sites	Aboabo	2.125	0.270	1.872	0.010
	Asafo	1.184	0.367	1.687	< LOD
	Bantama	3.812	0.741	2.882	< LOD
	KNUST	1.036	0.330	2.315	< LOD
	Adum	2.187	0.151	6.337	< LOD
	Airport Roundabout	4.101	0.028	1.417	< LOD
	Fante New Town	1.100	0.160	4.190	< LOD
Cafetaria	Suame	3.451	0.253	2.814	< LOD

< LOD = Below detectable limits.

the hazard quotients (HQ) of cadmium and mercury, their respective reference doses (R_fD) of 0.001 mg/kg-day (US EPA, 1993) [30] and 0.0003 mg/kg-day [26] were used to run Equation 3, and iterating at 100,000 times.

Results and Discussion

Exposure assessment

Four hundred and six (406) consumers from different groups along the selected vegetable food chain, responded positively to the survey. From the interview schedule, majority of the consumers (56%) who patronize the selected samples in the study area were within the 16 to 30 years' age group and 42% were above 30 years. The results revealed that females (51.23%) consume relatively greater portions of the selected samples than males. However, a non-parametric Levine's test showed a homogeneity of variance ($p > 0.05$) between the gender, indicating no significant statistical difference between males and females that consume the selected samples. The results were not consistent with a previous study conducted in the study area which concluded that males consumed relatively more of the selected samples [31]. In their studies, respondents only centered around the eateries (cafeterias and vending sites) where as this study rather included farmers and vegetable traders. The different approaches used in these two separate study might probably be the cause of the differences.

The consumption of the selected samples in this study area averaged 18.60 g per meal per day and this value appears to be smaller relative to the amount reportedly consumed in China which ranged between 231.5 to 301 g per person per day [21]. This suggests that consumers in the study area were likely to ingest hazards though it would take a long period of time in order to bio accumulate. A total of 87.44% consumers claimed they consumed the selected samples once daily. However, 2.96% respondents consumed the sample vegetables three times daily and the remaining consumers (9.60%) consumed them twice daily. The study further revealed that over 29.80% of the respondents consumed the selected samples once weekly, relative to 16.50% who consumed them twice weekly. However, 13.05% consumed the selected samples once every two weeks, relative to 11.83% who consumed them once every month. A rather small number of consumers (2.71%) consumed them every day.

Toxic heavy metal in levels in selected samples

Apart from mercury which was detected in only 12.50% of the samples analyzed; arsenic, lead and cadmium were detected in all of the samples (Table 2). Heavy metal concentrations detected in samples from farms and market places were in the order; Pb > As > Cd > Hg. A similar trend was observed for cafeterias except for those located in Aboabo, Bantama and other vending sites (Airport roundabout and Suame). These sites rather had the toxic metals concentration order of As > Pb > Cd > Hg. A two way-ANOVA test, applying Tukey's multiple comparisons test, was used to study the variance of heavy metal concentrations in the selected samples. The test showed that, at a confidence interval of 95%, the concentrations of the four heavy metals were the same among the selected samples collected in the sectors of the study area, indicating there is no significant statistical difference ($p > 0.05$) among the four heavy metals in the selected samples analyzed.

Lead is a geochemical or an environmental pollutant in urban areas, usually resulting from unregulated anthropogenic activities [32]. Table 3 presents the quantities of lead determined in the study area. The lead content was inconsistent and followed no pattern. However, the statistical distribution followed "Extvalue" (0.00266226, 0.00097673), presenting a range of min-max values of 1.42 and 6.34 µg/g respectively. These

Table 3: The statistical distributions of the concentrations (µg/g) of hazards and the elements of chronic daily exposures in vegetables collected in the study area.

Variables		Statistical distribution	Distribution metrics						
			Min	Max	Mean	Mode	Median	5th	95th
Hazards	As	Uniform (0.0000402, 0.0043548)	0.29	4.10	2.120	1.257	1.998	0.294	4.100
	Cd	Extvalue (0.00021341, 0.00014842)	0.03	0.84	0.307	0.181	0.253	0.028	0.836
	Pb	Extvalue (0.00266226, 0.00097673)	1.42	6.34	3.220	2.900	2.88	1.420	6.340
	Hg	Expon (1.875×10^{-6}, -1.17188×10^{-7})	0.00	0.02	0.002	0.000	0.000	0.000	0.020
	BaP	Pareto (0.40436, 0.0002)	0.20	41.00	8.487	0.200	3.10	0.200	40.000
Mass of food (g)		Loglogistics (7.9453, 10.207, 2.4105)	8.33	150	21.90	10.00	20.00	10.00	50.00
EF (days)		Triang (0, 0, 390.62)	1.71	365	125.74	0	104.29	1.72	260.71
ED (years)		Gamma (2.5135, 5.3073, -0.41536)	0.08	40.00	12.93	2.00	10.00	3.00	30.00
BW (kg)		Logistic (68.1095, 5.8916)	23.00	107.00	68.07	53.00	69.00	50.00	85.00

observations could possibly be accounted for in terms of the wastewater that might have been used for irrigating the selected samples or aero deposition of contaminants and subsequent absorption from the soil [33]. Other reports also explain that significant lead contamination do occur, especially in vegetables cultivated along highways [34] and also during transportation of vegetables to market centres [35]. Lead is considered a carcinogen, however, based on a cardiovascular reference point [36], exposures of 1.5 µg/g-day (0.105 mg/g for 70 kg man), the mean, mode and median values of respectively 3.22, 2.9 and 2.88 µg/g (Table 3), is considered very low compared to the reference point. This information must not be regarded as satisfactory because bioaccumulation of lead is reported to occur especially when divalent ions in the diet is low [37,38].

The results also show that though the most likely (modal) amount of the selected samples consumed was 10 g, there was wide uncertainty ranging from a minimum of 8.33 and maximum of 150 g. It is also informative to know that 50% of the consumers eat the selected samples up to 104 days (exposure frequency) in year. On the other hand, the most likely (modal) number of years consumers in the study area have been eating the selected samples was 2 years though as high as 40 years was recorded for some consumers. However, 50% of the consumers reported of consuming the selected samples in the study area for at least the past 10 years.

Cadmium levels in selected samples

The statistical distribution of cadmium in the study area (Table 3) followed the "Extvalue" pattern and presented min-max levels of respectively 0.03 µg/g and 0.84 µg/g. However, the most frequent (modal) cadmium level of 0.181 µg/g was observed, whereas 50% (median) of the samples presented cadmium level of 0.253 µg/g. With a mean cadmium level of 0.307 µg/g, the results show higher concentrations in the selected samples relative to 0.073 µg/g, determined in vegetables in other studies [39]. However, cadmium concentrations were lower, compared to the range of cadmium contaminations (0.68-1.78 µg/g) observed in similar samples grown on waste sites [4]. The seemingly unpatterned nature of cadmium accumulation observed is explained that, accumulation is dependent on factors such as the vegetables involved, the soil pH and the fertilizer application [39,40]. It is also believed that flood prone areas coupled with the high mobility property of cadmium ions results in higher accumulation [41].

Mercury levels in selected samples

The fitted distribution of mercury concentrations in the selected samples in the study area (Table 3) presented "Expon" (1.875×10^{-6}, -1.17188×10^{-7}). This showed a min-max range of between 0 and 0.02 µg/g, though a mean of 0.002 µg/g was obtained. There has been a reported study [42] that show a positive correlation between distance (D1 < D2 < D3 < D4) from a coal-fired power plant and the respective decreases (21.03 µg/g > 19.41 µg/g > 9.17 µg/g > 7.23 µg/g) in mercury content in vegetables cultivated. This observation suggest that mercury contamination varies, depending on the distance from the source of pollution. Compared to their study, it was evident that the mean mercury content obtained in this (0.002 µg/g) is lower. The selected vegetables for this study are usually consumed uncooked therefore, this observation presents another challenge since cooking has been reported to be the cause of the reduction of mercury content up to 10% in mushroom [43].

Apart from anthropogenic actions such as coal-fired power plant, other sources of mercury pollution could be coming from wastewaters sweeping such areas as mechanical and electronic workshops, e-waste dumpsites and incinerators. Unfortunately, the study area is dotted with such sources of mercury pollutants, suggesting that contamination may occur in the selected samples from either way; from the air and also from the roots of the selected samples through absorption [44,45]. The presence of mercury in the selected samples is particularly worrying because it is above what is permitted in foods.

Arsenic levels in selected samples

The statistical distribution of arsenic in the study area (Table 3) was "Uniform" (0.0000402, 0.0043548) with 50% (median) of samples presenting a value of 1.998 µg/g. The mean was however, 2.12 µg/g though

Table 4: The concentrations (µg/g) of congeners of PAHs in vegetable samples (n = 16) collected from the study area.

Sampling Sites	Suburbs	Concentration level (µg/g)			
		B(b)F	B(k)F	I(1,2,3-cd)P	B(g,h,i)P
	Atonsu	< LOD	< LOD	0.4	< LOD
Farms	Chirepatre	0.011	0.011	< LOD	< LOD
	KNUST	< LOD	< LOD	0.234	< LOD
	Tanoso	0.004	0.028	< LOD	0.835
	Ayigya	< LOD	0.103	< LOD	< LOD
Markets	Kejetia	< LOD	< LOD	< LOD	< LOD
	Kwadaso	< LOD	0.031	0.187	< LOD
	Santasi	0.003	0.031	0.205	< LOD
Street Food Vending Sites	Aboabo	< LOD	< LOD	< LOD	< LOD
	Asafo	0.004	< LOD	< LOD	< LOD
	Bantama	0.002	< LOD	0.408	< LOD
	KNUST	0.002	< LOD	< LOD	< LOD
	Adum	0.002	< LOD	< LOD	< LOD
	Airport Roundabout	< LOD	< LOD	< LOD	< LOD
	Fante New Town	< LOD	< LOD	0.04	0.414
Cafetaria	Suame	0.002	< LOD	< LOD	0.342

B(b)F = Benzo(b)fluoranthene; B(k)F = Benzo(k)fluoranthene; I(1,2,3-cd)P = Indenol(1,2,3-c,d)pyrene; B(g,h,i)P = Benzo(g,h,i) perylene.

< LOD = Below detectable limits.

the most frequently (modal) arsenic contamination was 1.257 µg/g. However, the uncertainty could range from a minimum of 0.29 µg/g to a maximum of 4.14 µg/g. The International Agency for Research into Cancer has classified arsenic as a group 1 carcinogen following several reports of its carcinogenicity to humans [46]. Arsenic contamination of the selected samples is a serious matter, however, the levels reported in this study suggest lower exposures and risks compared to what is prevailing in Bangladesh and India. In Bangladesh, for instance, a range of arsenic in home grown vegetables have been reported as ranging between 19 µg/g and 489 µg/g [47]. Though other studies in India reported low mean arsenic levels (20.9 µg/g and 21.2 µg/g) respectively in two districts [48], high levels ranging between 70 and 3990 µg/g have also been reported in Bangladesh [49]. Thus, the levels of arsenic reported in this study is significantly lower compared to the reported cases in Bangladesh.

There is certainly variation in the levels of arsenic in the selected samples and the differences have been assigned as variations in the kinetics of the uptake of the contaminants from soil, apart from the waste or irrigation water [50]. Again, studies have shown that cooking tends to lower arsenic levels [51] suggesting that caterers must consider cooking as an added detoxification process. The source of arsenic in the selected samples have been shown to be coming from the physicochemical quality of the soil such as pH, cation exchange properties and even the bacterial niches of the field on which these the vegetables were grown [52,53]. Though soil samples in the study area were not studied, there are reports [54] that higher soil pH leading to metal precipitation regulates complex formation with arsenic and control its availability to accumulation in vegetables.

Polycyclic aromatic hydrocarbon levels in selected samples

Four out of the sixteen PAH congeners screened were detected in the vegetable samples (Table 4). Thus, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(g,h,i)perylene and indenol(1,2,3-c,d)pyrene were detected in 50%, 31.25%, 18.75% and 37.5% of the samples respectively. Three PAH congeners were detected in samples from Tanoso farms and Santasi markets. The other sampling sites had no PAHs or very few congeners in the samples. While indenol(1,2,3-c,d) pyrene and benzo(k)fluoranthene were prevalent in samples from farms and markets, benzo(b)fluoranthene and benzo(g,h,i)perylene were prevalent in samples from the eateries. A two way-ANOVA test, using Tukey's multiple comparisons showed that, at a confidence interval of 95%, the concentrations of the four PAH congeners detected were the same among the various vegetable food chain groups.

In Table 3, it is seen that concentrations ranging from as low as 0.2 µg/g-day to as high as 41 µg/g-day were delivered through meals. Though a high level of BaP determined (Table 3) in this study (40 µg/g) was indicated for the top 5% of the heavy consumers of the population, 50% of the consumers were exposed at 3.1 µg/g. This chronic exposure is still greater than the maximum value determined in cabbages (1.56 ng/g) grown in urban Romania [55]. Though the congeners in these studies are different, their toxicological equivalence factors provide a tool to sum up and express them in terms of BaP, in order that, their carcinogenic effect can be evaluated summatively. On this basis therefore, consumers in the study area were exposed to a higher risk of carcinogenesis. The levels of BaP might be marginal, but no quantities of

carcinogens are tolerated in principle. The quantity of the selected samples consumed under this study is also a pointer to how much consumers are exposed to BaP per day. Indeed, in this study, the most likely mass of the selected samples consumed was recorded as 10 g, however, the amount consumed by 50% of the consumers was reported as 20 g. This is similar to the observation that the selected vegetables under study, contributed relatively small quantities when matched with other dietary sources of BaP [56,57].

Benzo[a]pyrene has been reportedly ranging from 0.17 to 0.43 µg/g in The Netherlands, relative to another study that ranged between 2.5 to 5.7 µg/g for vegetarians in the United States [58,59]. However, some samples used for this study were collected along highways and semi-industrialized areas. Thus, it is not surprising that they contain significant amounts of BaP relative to other studies where samples were collected from rural areas [55]. The observation that there were significant increases in quantities of PAHs in vegetables relative to their surface area [60] is a cause for concern because bioaccumulation of PAH along the food chain. Indeed, one study reported PAHs in vegetables ranging from 0.07 µg/g to 1.1 µg/g in industrialized site relative to those sampled from rural areas [29]. Studies have shown that the exposure pathway for contamination of the selected samples of vegetables with PAHs is by the gaseous route and subsequently, particle deposition. However, there are cases where significant quantities were absorbed by the roots from the soil [61]. There are reports that PAH congeners on leafy vegetables can be removed by washing with cleaning solutions [62]. Thus, food service operators must be trained in the proper ways of cleaning in order to control careless ingestion of such hazards. In spite of their marginal contamination and consumption from fruits and vegetables, there is a global trend of PAH contamination of vegetables [63].

Exposures and risks of hazards in selected samples

Table 5 present the indices of the chronic exposure of the hazards, as calculated by Equation 1, with inputs from the study area. All the hazards presented a minimum exposure of 0 mg/kg showing that there were instances where consumers were not exposed to the hazards. This information is corroborated by the extremely small values (in the order of $\times 10^{-6}$) recorded as the 5th percentiles. However, there were significant maximum exposures for all the hazards, ranging from 1.15×10^{-5} mg/kg in mercury to 4.0×10^{8} mg/kg in BaP. The maximum and mean exposures as obtained for BaP in the survey appears to be isolated cases since the most frequently or modal occurring exposures of BaP (1.80×10^{-6} mg/kg) and the 95th percentile (4.0×10^{-3} mg/kg) were low. Though these low prevailing exposures could translate into low health risk, it is important to note that as the numbers of consumers eating the selected samples increase, a greater number of people would be as risk.

Health risk hazard quotient in selected samples

Table 6 presents the estimated health risk indices for mercury and cadmium through the oral route of the selected vegetables in the study area. The mean, modal and median level of the hazard quotient reinforce the observation that mercury and cadmium ingested area poses lower risk levels. However, cadmium recorded a maximum range value with HQ greater than 1, suggesting that there could be sectors in the study area where ingestion of cadmium could be dangerously high.

There are other reports, here in Ghana that indicate low health concern for cadmium in the selected samples studied [64]. In China, studies have shown acceptable limits (HQ < 1) at certain sites with hazard quotient of cadmium in vegetables ranging from 0.2 to 0.49 [61]. However, there are also observations contrary to these afore-mentioned reports. For instance, Muchuweti, et al. [65] reported that *Tsunga*, a popular local vegetable consumed in Zimbabwe, had several times cadmium higher than acceptable limits according to reference for water standards (0.001 mg/kg) [66] and also

Table 5: Statistical distribution metrics of the chronic exposures (mg/kg-day) of lead, cadmium, arsenic mercury and *BaP of the vegetables consumed in the study area.

Distribution metrics of exposure							
Hazards	Min	Max	Mean	Mode	Median	5th	95th
Pb	0.00	4.00×10^{-3}	7.00×10^{-5}	2.04×10^{-6}	3.80×10^{-5}	2.30×10^{-6}	2.80×10^{-4}
Cd	0.00	1.20×10^{-3}	1.70×10^{-5}	2.04×10^{-8}	7.00×10^{-6}	2.20×10^{-7}	6.30×10^{-5}
As	0.00	3.80×10^{-3}	5.00×10^{-5}	1.53×10^{-7}	2.20×10^{-5}	8.00×10^{-7}	2.00×10^{-4}
Hg	0.00	1.15×10^{-5}	10.00×10^{-8}	0.00	2.80×10^{-8}	0.00	4.10×10^{-7}
BaP	0.00	4.40×10^{8}	4.70×10^{3}	1.80×10^{-6}	1.20×10^{-4}	4.20×10^{-6}	4.00×10^{-3}

*BaP was determined as the sum of all the congeners of PAHs present in the vegetables.

Table 6: Statistical distribution metrics of health risk hazard quotients of cadmium and mercury content of vegetables consumed in the study area.

Distibution metrics of hazard quotient							
Hazards	Min	Max	Mean	Mode	Median	5th	95th
Cd	0.00	1.2	1.60×10^{-2}	2.60×10^{-6}	7.00×10^{-5}	2.00×10^{-4}	6.30×10^{-2}
Hg	0.00	0.04	3.30×10^{-4}	0.00	9.20×10^{-6}	0.00	1.40×10^{-3}

Table 7: Statistical distribution metrics of the incremental lifetime cancer risks of arsenic, lead and *BaP in the study area.

Distibution metrics of incremental lifetime cancer risk							
Hazards	Min	Max	Mean	Mode	Median	5^{th}	95^{th}
As	0.00	5.60×10^{-3}	7.80×10^{-5}	2.30×10^{-7}	3.30×10^{-5}	1.20×10^{-6}	3.00×10^{-4}
Pb	0.00	3.35×10^{-5}	6.45×10^{-7}	1.75×10^{-8}	3.20×10^{-8}	2.00×10^{-8}	2.30×10^{-6}
BaP	0.00	1.50×10^{10}	1.50×10^{5}	1.80×10^{-6}	1.20×10^{-4}	4.00×10^{-6}	3.70×10^{-2}

*Benzo[a]pyrene was determined as the sum of all the congeners of PAHs present in the sampled vegetables.

according to the Irish standards (0.1 mg/kg) [67]. This contrasting observations suggest that the absorption and accumulation of these divalent cations depend on a variety of factors, which may include soil quality, edaphic factors, plant types and their genetic make ups. The mercury content of vegetables in the study area was low, and it is similar to what was reported in another study in Lagos, Nigeria, where the mercury levels recorded in fresh and boiled Amaranthus was less than 0.8 µg/g [68]. Thus, with respect to mercury, the health risk in the selected vegetables was negligible (HQ < 1).

Cancer risk assessment

Presented in Table 7 are the results showing ILTCR for the three hazards; lead, arsenic and BaP. De minimus for BaP is 10^{-6} (US EPA, 2000) [26] whereas that for the toxic heavy metals; cadmium and lead is 10^{-4} [19]. Arsenic presented median (50%) risk of consumers of the selected samples in the study area as 3 out-of-100-thousand consumers. However, the most frequent (modal) risk was 2 out-of-10-million consumers is below the de minimis of 1 out-of-one-million consumers, meaning risk is negligible. This observation must be treated with caution because the uncertainty ranged from 0 to a risk of 6 out-of-thousand consumers, which is unacceptable. Again, the results indicated that the median, modal and mean risks recorded for lead, were remarkably below the de minimis risk (10^{-6}). Once again, this information must be treated with caution because the uncertainty ranged from 0 to 3 out-of-100-thousand consumers, which is a cautiously acceptable risk is dependent on the circumstances of exposures of the hazard. In the case of BaP, the modal risk was unacceptable because a risk of 2 out-of-1-million is higher than the recommended de minimis of 1 out-of-1 million. This is so because, the USEPA considers ILTCR greater than 1 in a million (1×10^{-6}) to be serious and of high priority for adverse health problems [69]. Thus, consumers are at risk of manifesting conditions that could lead to the manifestation of cancer [26].

Conclusion

The exposures of mercury at the modal and median levels were very low but PAHs exposures were extremely high. The low hazard quotients (HQ < 1) of cadmium and mercury revealed that they have low risks, meaning health concerns of these hazards are negligible in the study area. The high median and modal ILTCR of lead show that consumers are at high risk and indeed the risk uncertainty ranging from 0 to 3 out-of-100-thousand consumers is unacceptable. Though the modal and median ILTCR of arsenic seem to be acceptable, again, the risk uncertainty ranging from 0 to 6 out-1-thousand consumers still brings the risk acceptability of arsenic into question. BaP contamination was quite high since the median and modal ILTCR fall within both unacceptable risk ($\geq 10^{-6} \leq 10^{-4}$) and barely negligible risks ($\geq 10^{-6}$) ranges respectively. Though some consumers may not be exposed to these hazards at all, others are variously and significantly exposed, and this raises health concerns. It is still difficult to comprehend why regulatory organizations seem not to be rising to the challenge of regulating the cultivation of vegetables in the urban centers in this study area.

Compliance with Ethical Standards

Conflict of interest

The authors declare that they have no conflict of interest.

Ethical approval

This article does not contain any studies with human or animal subjects.

Informed consent

Informed consent was not applicable.

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