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Heavy metal content in mixed and unmixed seasonings on the Ghanaian market

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Human exposure to some heavy metals through consumption of various seasonings in some Ghanaian markets was evaluated. The heavy metals considered were iron (Fe), zinc (Zn), copper (Cu), cadmium (Cd), lead (Pb) and mercury (Hg). The levels of iron (Fe) , zinc (Zn), copper (Cu), cadmium (Cd) and lead (Pb) in a total of twenty two (22) mixed and unmixed seasonings were determined using flame atomic absorption spectrometry whereas the mercury levels were determined by cold vapour atomic absorption spectrometry. In unmixed seasonings, Fe content ranged from 19.4 to 971.40 mg/kg, Zn from 2.40 to 34.60 mg/kg, Cu from 0.9 to 10.10 mg/kg, Cd from below detection limit (0.01) to 0.9 mg/kg and Pb ranged from 0.6 to 1.8 mg/kg. In mixed seasonings, concentration ranged from 83.36 to 480.82 mg/kg for Fe, 1.72 to 26.78 mg/kg for Zn, 1.73 to 7.70 mg/kg for Cu and 0.63 to 1.39 mg/kg for Pb and from below detection limit (0.01) to 0.06 mg/kg for Cd. Hg was below the detection limit (0.01) in all the seasonings. The results indicated that Fe, Zn and Cu were below permissible levels whereas Pb and Cd were above permissible levels.

Key words: Toxic metals, seasonings, consumption, spectrometry, Kumasi.

INTRODUCTION

Heavy metals have bio-importance as trace elements but the biotoxic effects of many of them in human biochemistry are of great concern. They enter our bodies via food, drinking water and air (Lenntech, 2008). Iron, zinc and copper are essential metals whereas cadmium, lead and mercury have no bio-importance (Divirikli et al., 2006).

Heavy metals contamination in plants, animals and humans are due to environmental pollution through air emissions from automobile exhaust, pesticides leaching into water bodies, smelters and process wastes from mining and other industries (Ansari et al., 2004, Duruibe et al., 2007; Opuene and Agbozu, 2008).

Spices which are dried plants parts (Satter et al., 1989) can easily be contaminated by heavy metals from type of soil for cultivation, fertilizers and source of water used for irrigation (Abdullahi et al., 2008). In addition, colourants which are added to some of these flavour enhancers may contain some trace metals such as lead (Ekpo and Jimmy, 2005).

Interaction with sellers of powdered ginger and pepper (red and green) as spices in the local markets indicated that, these fruits are processed by the individual seller whereas most of the seasonings are imported. These spices may easily be contaminated by heavy metals from the soil or aerial depositions as these spices are dried on the ground or on roof tops. Moreover, commercial mills used may also introduce some amount of metals into the

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seasonings due to wear and tear of the machinery.

Heavy metals content of different spices, bouillon cubes, food condiments and aromatic herbs have been investigated in many countries (Garcia et al., 2000; Lopez et al., 2000; Divrikli et al., 2006; Ozkutlu et al., 2006; Nnorom et al., 2007). However, there is limited information on the levels of heavy metals in these seasonings on the Ghanaian markets. This work therefore seeks to bridge that gap by providing information especially to the Ghanaian populace on the levels of heavy metals of these most consumed seasonings. Information will further be provided on the sources of these seasonings and the extent to which they are contaminated with these heavy metals for future studies and effective comparative analysis. The objective of this study therefore was to evaluate human exposure to some heavy metals through consumption of mixed and unmixed seasonings in some markets in Ghana.

MATERIALS AND METHODS

Sample collection and preparation

Twenty two different powdered samples were purchased at random from local shops and hawkers from the Asafo (A), Railway (R) and Central markets (C) in Kumasi, Ashanti Region of Ghana located in the transitional forest zone about 270 km north of the national capital, Accra. The samples were purchased after preliminary investigations had indicated that they were the most consumed seasonings in the metropolis. The samples were grouped into unmixed seasonings (M) and mixed seasonings (UM). The unmixed seasonings were rosemary (*Rosmarinus officinalis)*, anise (*Pimpinella anisum*), garlic (*Allium sativum*), nutmeg (*Myristica fragrans*), prekese (*Tetrapleura tetraptera*), Senegal pepper (*Xylopia aethiopica*), ashanti pepper(*Piper guineense*), ginger (*Zingiber officinale*), red pepper (*Capsicum annuum*), green pepper (*Capsicum frutescens*). The mixed seasonings were adobo red, adobo yellow and curry. The samples were then dried using the Astell scientific dryer at 45°C for 24 h, cooled and stored in plastic bags prior to analysis.

Apparatus

All glassware and plastic containers used were washed with detergent solution followed by soaking in 10% (v/v) nitric acid overnight. They were rinsed with distilled water followed by 5% potassium permanganate, rinsed with distilled water and dried before use (Voegborlo and Akagi, 2007). Blanks and standard were prepared alongside. Analytical reagents (AnalaR) grade chemicals (BDH Chemicals Ltd., Poole. Emgland) were used throughout the study.

Digestion of samples

Digestion of samples was carried out by the method of Akagi and Nishimura (1991). About 1 g of each sample was weighed into a 50 ml digestion tube and 1 ml H_2O, followed by 2 ml HCl, 5 ml HNO_3 : $HClO_4$ (1:1) and 2 ml H_2SO4 were added. After heating at 200°C till solution was clear, samples were cooled and filtered into standard 50 ml volumetric flask and made to the volumetric mark. The digest were analyzed for Fe, Zn, Cu, Cd and Pb by air-acetylene flame atomic absorption spectrometer (Spectr AA 220, Australia). The automatic mercury analyzer model HG-5000 was for the determination of Hg.

Statistical analysis

Means and standard deviations were computed using Statsgraphics Centurion XV, 2005 Version 15 statistical software (Statpoint. Inc, USA). Where necessary, one way analysis of variance (ANOVA) was used to test if any significant differences existed between concentrations of a particular metal in a particular seasoning group using the same statistical software.

RESULTS AND DISCUSSION

Heavy metals in seasonings

Iron (Fe), zinc (Zn), copper (Cu), mercury (Hg) and lead (Hg) levels were determined in all seasonings. The mean (±standard deviation) and range of the concentration of metals in the groups of seasonings are presented in Tables 1 and 2, respectively. The results are means of three replicates. Cadmium was mostly below the detection limit (0.01 mg/kg) in unmixed seasonings but was present in most of the mixed seasonings. The levels of iron in both groups of seasonings were mostly high.

Iron is the most needed micronutrient in plants (Divrikli et al., 2006). The iron contents in the seasonings were in the range 19.4 in rosemary (UM-1) to 971.40 mg/kg in nutmeg (UM-4) for unmixed seasonings. Red pepper (UM- 14) from central market (C) was almost three times the concentration of red pepper (UM-16) from Railway market whereas red pepper (UM-15) from Asafo was almost two times the concentration for UM-16. The concentration of Fe in the various red peppers differed significantly ($p < 0.05$).

Mixed seasonings had iron levels in the range of 83.36 to 480.82 mg/kg with Adobo (yellow) (M-2) recording the least while curry (M-4) having the highest (Table 2). Four different brands of curry were analyzed. Curry (M-4) from Ghana had the highest value of Fe (480.82 mg/kg) while curry (M-5) from Nigeria had the lowest value of 149.13 mg/kg. This suggests that curry powder produced in Ghana may be a rich source of Fe. No significant difference ($p > 0.05$) was found in the concentration of Fe between curry (M-3) and (M-4).

Rosemary (UM-1) had lower level of iron whereas in ginger, levels were higher than values reported in literature (Divrikli et al., 2006; Ozkutlu et al., 2006; Koc and Sari, 2009). Levels of iron in ginger (UM-8, UM- 9 and UM-10) and garlic (UM-3) were also higher than levels reported by Hashmi et al. (2007). Generally, the levels of iron in unmixed seasonings were higher than mixed seasonings. The higher Fe content in unmixed seasonings could be due to combination of the same plants parts (Nnorom et al., 2007) such as fruits whereas the Fe content in the mixed seasoning could be due to low levels of Fe in the individual spice. This is because the sellers

Table 1. Mean levels (± SD) (mg/kg) of iron, zinc, copper, cadmium and lead in unmixed (UM) seasoning.

Sample	Source	Sample code	Fe	Zn	Cu	Cd	Pb
Rosemary	China (CH)	UM-1	19.4±1.6	2.6±0.4	0.9±0.2	ND	1.5±0.2
Anise	*	UM-2	500.4±5.9	34.6±1.9	9.3±0.1	ND	1.2±0.6
Garlic	India	UM-3	58.6±8.6	14.9±1.5	3.2±0.2	ND	1.0±0.1
Nutmeg	France	UM-4	109.0±1.6	10.8±2.0	10.1±2.6	0.9±1.6	1.1±0.6
Prekese	Ghana	UM-5	24.8±0.8	2.4±0.1	2.9±0.1	ND	1.0±0.8
Senegal pepper	*	UM-6	45.5±1.0	5.1±0.9	5.5±3.9	ND	0.7±0.1
Ashanti pepper	Mali	UM-7	100.6±0.8	17.8±0.4	4.5±0.2	ND	0.6±0.1
Ginger	Ghana (GH-C)	UM-8	698.3±1.4	17.6±1.8	4.7±1.3	ND	1.8±0.6
Ginger	Ghana (GH-A)	UM-9	408.4±10.5	14.1±0.7	3.7±0.2	ND	0.9±0.3
Ginger	Ghana (GH-R)	UM-10	590.1±4.0	11.8±1.8	3.7±0.7	ND	1.6±0.5
Green pepper	Ghana (GH-C)	UM-11	94.3±8.4	19.4±0.8	1.5±0.2	ND	1.4±0.1
Green pepper	Ghana (GH-A)	UM-12	226.2±0.0	16.9±1.1	1.3±0.0	ND	1.0±0.1
Green pepper	Ghana (GH-R)	UM-13	105.9±1.1	15.4±1.1	1.4±0.0	ND	0.9±0.2
Red pepper	Ghana (GH-C)	UM-14	971.4±1.8	14.9±1.6	6.7±0.2	ND	1.4±0.6
Red pepper	Ghana (GH-A)	UM-15	614.6±1.0	9.5±0.4	7.2±0.2	ND	1.0±0.0
Red pepper	Ghana (GH-R)	UM-16	345.9±12.7	7.5±0.7	7.7±0.3	ND	1.4±0.5

*Source not known; (C), Central market; (A), Asafo market; (R), Railway market; SD, standard deviation.

Table 2. Mean level (± SD) (mg/kg) of iron, zinc, copper, cadmium and lead in a mixed (M) seasonings.

Sample	Source	Sample code	Fe	Zn	Cu	Cd	Pb
Adobo (red)	United states of America (U.S.A)	M-1	91.56±5.51	1.72±0.32	1.81±0.04	0.06±0.00	1.39±0.64
Adobo (yellow)	United states of America (U.S.A)	M-2	83.36±3.49	2.76±0.93	1.73±0.03	0.05±0.00	1.08±0.03
Curry	France (FR)	M-3	204.32±4.37	17.68±0.46	5.77±0.14	0.01±0.01	0.91±0.10
Curry	Ghana (GH)	M-4	480.82±0.95	13.67±0.28	7.70±1.72	0.02±0.02	1.05±0.16
Curry	Nigeria (NIG)	M-5	149.13±4.15	13.87±0.80	6.40±0.13	ND	0.84±0.15
Curry	South Africa (SA)	M-6	214.93±5.40	26.78±0.61	6.39±0.11	ND	0.63±0.10

purchase the same plants parts such as fruits or leaves from different sources, mix them and mill them into a single homogeneous product. The high levels of iron in all the seasonings could also be due to contamination during milling. Research indicates that grinding of spices in commercial mills contaminates them to about between 3 and 5 folds, due to wear and tear of the machine parts (Janitha et al., 1988).

Even though Fe is an essential element needed by the body, consumption of an excessive amount can lead to health effects such as enlarged liver and joint diseases (Hoffman, 2009). To safeguard the health of the public, organizations such as Nutrigold Technical (2007) and FAO/WHO have established tolerable levels known as the Recommended Daily Allowance (RDA) and Provisional Tolerable Weekly Intake (PTWI) for most elements.

In order not to exceed the RDA of 14.8 mg/day of Fe for a 60 kg body weight, a person must not consume more than 762.89 g of rosemary daily. Iron (Fe) toxicity is therefore insignificant.

Zinc plays important roles in growth and development in humans (Colak et al., 2005). Zinc deficiency is of growing concern in the developing world because consumption of plants food has inhibitory effect on zinc absorption (Divrikli et al., 2006). It is reported by Hotz and Brown (2004) that an estimated 20% of the world population was at risk of inadequate zinc intake. Delayed neurological and behavioural development in children occurs as a result of zinc deficiency (Caulfield et al., 1998). Unmixed seasonings had zinc content between 2.40 and 34.60 mg/kg (Table 1). Anise (UM-2) recorded the highest amount of Zn whereas *Prekese* (UM-5) which differed significantly ($p<0.05$) from Anise (UM-2), recorded the lowest. Ginger (UM-8) from Central market had the highest level of Zn (17.6 mg/kg) whereas ginger (UM-10) from Railway market had the lowest Zn concentration of 11.8 mg/kg. Green pepper from the various markets, showed variation in Zn concentration though no significant difference (p>0.05) was observed (Table 1). Zinc level in red pepper (UM-14), purchased from Central market was two times the concentration in red pepper (UM-16) from Railway market. Concentration of zinc in red pepper from the various markets differed significantly (p<0.05).

Zn levels in mixed seasonings were between 1.72 and 26.7 8 mg/kg with Adobo (red) from U.S.A having the least while curry seasonings from South Africa (S.A) have the highest. The four different brands of curry analyzed showed that curry (M-6) from S.A had the highest Zn content whereas curry (M-4) from Ghana had the least. This suggests that curry from S.A may be a rich source of Zn. Though there was no significant difference (p>0.05) in concentration of Zn between any type of Adobo, curry (M-3) and curry (M-6) differed significantly (Table 2).

Ginger (UM-8, UM-9 and UM-10) and garlic (UM-5) had concentrations of Zn higher than that reported previously (Ozkutlu et al., 2006, Hashmi et al., 2007). Rosemary (UM-1) had lower Zn level as compared to other studies (Divrikli et al., 2006; Koc and Sari, 2009). The mean zinc content of red pepper and curry seasonings in this study ranged from 7.40 to 14.9 mg/kg and 13.67 to 26.78 mg/kg as compared to 10.40 to 35.0 and 13.65 to 29.90 mg/kg, respectively, in Nigeria (Nnorom et al., 2007; Awode et al., 2008).

Zinc levels in all samples were lower than the standard level (100 mg/kg) set by FAO/WHO (2003). The differences in Zn content from same plants parts purchased from the various markets could be due to factors such as type of soil for cultivation and drying environment. The high levels of zinc in the seasonings reflect the normal composition expected in plant derived products (Onianwa et al., 1999). Research indicates that microntrients are generally higher in leaves than in other above ground parts in plants (Basgel and Erdemoglu, 2005). However, Zn in rosemary, a leafy spice, was quite low. This may be due to low zinc content in the soil used for cultivation.

Copper helps in iron metabolism by helping in oxygen transport as well as utilization and absorption of iron in humans (Özçelik et al., 2002). The concentration of copper in unmixed seasonings ranged from 0.90 to 10.10 mg/kg (Table 1). Rosemary (UM-1) recorded the lowest Cu concentration while nutmeg (UM-4) recorded the highest. Ginger (UM-9 and UM-10) from Asafo and Railway markets recorded similar Cu values. The concentrations of Cu in green pepper from all three markets were very close. This suggests that though samples differ in localities, their copper content may be influenced by the same factors. There was no significant difference (p>0.05) in Cu content between any type of ginger, red pepper or green pepper (Table 1).

The copper level in mixed seasonings was between 1.73 and 7.70 mg/kg (Table 2). The lowest level was found in Adobo (yellow) (M-2) from U.S.A and the highest in curry (M-4) from Ghana. Copper in the four brands of curry was highest in curry from Ghana followed by Nigeria, South Africa and France in that order. No significant difference was found in Cu content between any type of Adobo or curry seasonings.

Rosemary (UM-1) had lower levels of copper as compared to those reported previously in other studies (Ozcan, 2004; Divrikli et al., 2006; Koc and Sari, 2009). Ginger (UM-8, UM-9 and UM-10) garlic and nutmeg (UM-4) had higher levels of copper as compared to values reported in other literatures (Ozkutlu et al., 2006; Hashmi et. al., 2007; Krejpcio et al., 2007). The level of Cu in all groups of seasonings was below 10 mg/kg (FAO/WHO, 2003) permissible in plants.

It has been reported that presence of cadmium in food is mostly derived from various sources of environmental contamination and has no biological importance in higher organisms such as humans and plants (Adriano, 1984). Cadmium is extremely toxic even at low levels (WHO, 1989). Cadmium toxicity is characterized by chest pain, cough with foamy and bloody sputum, and death of the lining of the lung tissues due to excessive accumulation of watery fluids (Duruibe et al., 2007).

Cadmium levels in unmixed seasonings ranged below 0.01 and 0.90 mg/kg (Table 1). Nutmeg (UM-4) was found to contain the highest Cd content whereas Cd in the rest of the seasonings was below detection limit.

In Table 2, the cadmium levels in mixed seasonings ranged from below detection limit to 0.06 mg/kg. Adobo (red) (M-1) recorded the highest Cd content. The highest Cd level in the curry samples was found in M-4 from Ghana though below the permissible limit (0.20 mg/kg). There was no significant difference between any type of Adobo or curry seasoning investigated.

Cadmium was not detectable in Rosemary by Divrikli et al. (2006), while Koc and Sari (2009) reported 3.1 µg/g of cadmium in rosemary. The concentration of cadmium in pepper and ginger was reported to range from 0.25 to 1.07 mg/kg and 0.02 to 0.04 mg/kg, respectively, by Awode et al. (2008) and Krejpcio et al. (2007) but was not detected in any type of pepper or ginger in this study. Concentration in nutmeg (UM-4) was higher than reported levels (0.05 mg/kg) by Krejpcio et al. (2007). The level of cadmium in curry seasonings were lower than levels reported in literature (Nnorom et al. 2007). According to Chizzola et al. (2003), cadmium levels could be considered normally low in plants. Generally, all the seasoning samples with the exception of nutmeg were below the permissible level (0.20 mg/kg) set by the FAO/WHO (2001) for spices. Though cadmium content in the seasonings were low, its presence could be due to contamination of raw materials, technological processes or colouring agents used as they contain cadmium salts (Cabrera et al., 1995).

Spices are suspected to obtain lead during growth in lead contaminated soils or in the course of milling or other processing procedures (Woolf and Woolf, 2005).

The use of pesticides contaminated with heavy metals during the growing of herbs and spices may also be a source of lead contamination in the final product (Galal-Gorchev, 1991).

In unmixed seasonings, levels of Pb ranged from 0.6 to 1.8 mg/kg. Ashanti pepper (UM-7) from Mali recorded the least Pb level while ginger (UM-8) from GH (Railway) recorded the highest (Table 1). Ginger, red pepper and green pepper from the various markets showed differences in the concentrations of Pb. However, no significant difference (p>0.05) was found in Pb content between either group of seasoning. It was observed that out of the three markets, seasonings from Central had the highest Pb content. This is also evident in seasonings from Railway market which is located between two busy roads. Wheeler and Rolfe (1979) found that Pb levels in vegetation increased linearly with traffic density. Pb concentration in mixed seasonings (Table 2) was between 0.63 and 1.39 mg/kg. The highest Pb level was found in Adobo (red) (M-1) from the U.S.A and curry seasonings (M-6) from S.A had the lowest. Within the different brands of curry, Pb levels in curry (M-4) from Ghana had the highest Pb level whereas Curry (M-6) from S.A had the lowest.

Pb has been reported as non detectable in some spices and herbs (Divrikli et al., 2006; Nnorom et al., 2007). However, in other studies (Chizzola et al., 2003; Gupta et al., 2003; Koc and Sari, 2009) where Pb content was determined, the highest Pb level ranged from 2.00 to 200 mg/kg as compared to 0.37 and 2.8 mg/kg in this present study. Concentration in nutmeg (UM-4) was higher than reported levels (0.36 mg/kg) by Krejpcio et al. (2007).

The level of lead in all seasoning samples were above the permissible limit required in spices (0.3 mg/kg) by the FAO/WHO (2003). The lead content in seasonings could be attributed to the addition of lead during processing to impart colour, sweet taste or increase the weight of these products (Kakosy et al. 1996). Some studies reported that traffic density also increases the lead burden in the environment thereby increasing the lead content in vegetation (Rodriguez-Flores and Rodriguez-Castellon, 1982; Buszewski et al., 2000; Nabulo, 2004).

It must be said that though data on the rate of consumption of seasoning in Ghana is unavailable, it is unlikely that a person will consume high amounts (> 20g) of seasonings in a day. This suggest that intake of the seasonings will have negligible effects on the health of consumers. Thus, the high levels of toxic metals (Cd and Pb) suggest the need for consumption advisory.

Conclusion

The study shows that iron, zinc, copper and lead were present in all seasonings whereas only 23% of the sampled seasonings contained cadmium. Mercury was not detected in any of seasonings analyzed. The toxic metals, cadmium and lead were mostly above the FAO/WHO permissible level.

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