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Full Length Research Paper

Heavy Metals and Organochlorine Pesticides Residue Levels in Natural Spices Sold in Kumasi, Ghana

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Abstract

Heavy metals and organochlorine pesticides residue levels were assessed in some natural spices sold in Kumasi. A total of twenty samples comprising of ten different spices were purchased from vendors from Asafo and Central markets in Kumasi, Ghana, homogenized and digested with acid mixture (1:1 HNO_3: $HClO_4$). Sample solutions were analyzed for THg using an Automatic Mercury Analyzer Model HG-5000 and for the other metals using SHIMADZU Flame Atomic Absorption Spectrometer (AA 240 FS). The organochlorine pesticides were determined with Shimadzu gas chromatograph GC-2010 with ECD. THg concentrations ranged from 1 ppb to 35 ppb for all spices, and were below EU maximum limit in food (500 ppb), that for Cd also ranged from below detectable limit to 0.47 ppm, and 25% of the samples were above the EU maximum limit in food (0.05 ppm). Lindane concentration (µg/kg) for all the spices ranged from 10 to 180, HCB range from below detectable limit (1 ppb) to 166 ppb. Although, most of the organochlorine residues detected were below the maximum limits set by the Codex Alimentarius Commission (CAC), bioaccumulation of these residues is likely to pose health problems in higher organisms like human beings.

Keywords: Heavy metals, Organochlorine, Pesticides, Spice, Ghana.

INTRODUCTION

Spice, a major group of the food additives is a vegetable substance of indigenous or exotic origin which is or has a hot, pigment taste, used to enhance taste of foods or to add to them the stimulant ingredients contained in them. It can also be defined as a dried seed, fruit, root, bark or vegetative substance derived from the non- leafy parts of plants. It is used as a food additive for the purpose of flavouring or food flavourings, and sometimes as a preservative by killing or preventing the growth of harmful bacteria (Adamson, 2004).

There are many reasons for which people use spices, though, taste probably tops the list. There are several spices that simply smell good and those smells can be alternately soothing or exciting. Some of the spices used in Ghana include Onion, Ginger, Garlic, Cloves, Dawadawa, Cinnamon and Rosemary. There is no doubt that spices do have value far beyond enhancing the taste of food. However, besides adding to the taste, spices have multifarious functions that include combating foodborne microorganisms, reducing food poisoning (Sherman and Billing, 1999), antioxidant function (Pokorney, 1991), and antimicrobial activity (Shelef, 1983).

Heavy metals may be present in agricultural soils at low levels but can be accumulated by plant over a period. This has led researchers all over the world to study the pollution with heavy metals of air, water, and foods to avoid their harmful effects and to determine their suitability for human consumption(Naeem et al., 2009; Nkansah and Amoako, 2010; Sarojam and Chen, 2010). A research conducted in Saudi Arabia on 18 local spices (cumin, coriander, nigella, cinnamon, fenugreek, basibic, nasturitium, mahalib, turmeric, thyme, black pepper, ginger, safflower, cardamon, garden sage, nutmeg, anise and mahaleb cherry) revealed high levels of some trace metals (Al-Eed et al., 2002). The concentration of lead (Pb) ranged from trace to 14.30

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mg/kg on dry weight basis, cadmium (Cd) ranged from 1.25 mg/kg to 3.05 mg/kg, cobalt (Co) from 0 to 0.64 mg/kg and that of selenium (Se) from 0 to 13.3 mg/kg. Some of these concentrations are above the limit approved by WHO/FAO and this makes it dangerous due to the health implications associated with them. Other countries have also established daily intake metals in foodstuff depending on the levels found in foodstuff in the country. It is therefore necessary to determine the levels of heavy metals in the spices sold in the Ghanaian market to help policy makers.

Persistent organic pollutants (POPs) such as organochlorines have been of great concern due to their occurrence in high concentration even in remote ecosystems, despite bans on production and usage (Iwata et al., 1994; Guruge and Tanabe, 2001). Organochlorine pesticides (OCPs) such as hexachlorocyclohexane (HCH) and 1,1,1- trichloro-2,2′-bis(p-chlorophenylethane (DDT) are ubiquitous anthropogenic environmental contaminant (Willet et al., 1998; Nakata et al., 1998).

In Ghana organochlorine pesticides residue work has been done on meat (Darko and Acquaah, 2007), dairy products (Darko and Acquaah, 2008) and vegetables (Botwe *et al*., 2011). No work has been done on spices which plays a major role in Ghanaian dishes.

This work, thus, seeks to provide information on levels of pesticide residues and heavy metals in spices that will assist in a scientific assessment of the impact of pesticides and heavy metals on public health, agriculture and the environment in Ghana.

MATERIALS AND METHODS

A total of twenty (20) samples comprising ten different spices were purchased from different traders in the selected markets. Some of the spices were obtained in the powdered form and others in the raw form as their powdered form were not available.

Digestion and analysis of heavy metal

Samples were washed thoroughly to remove soil and dirt that might be deposited on spices with distilled water and dried in an oven at a temperature of 30^{o}C overnight. The samples were ground in a laboratory motar.

Total Hg (THg) was analysed following Akagi and Nishimura (1991) procedure, where 0.5 g of each dried homogenized spice samples were weighed into 50 ml digestion tubes. De-ionized water (1 ml) was added, the tubes were swirled to ensure complete wetting of samples, 2 ml (1 + 1 HNO_3 and $HClO_4$) was then added followed by 5 ml of H_2SO_4. The samples were heated at 200 ± 5°C until clear digest was obtained, without covering the flasks and then cooled to room temperature and diluted to 50 ml with de-ionized water. Total Hg in samples was analysed with a semi - automatic Mercury analyzer (HG 5000, Sanso Seisakusho Co. Ltd). The rest of the digest was used to determine five other heavy metals: zinc (Zn), manganese (Mn), iron (Fe), lead (Pb), cadmium (Cd) using SHIMADZU Flame Atomic Absorption Spectrometer (AA 240 FS).

Extraction and analysis of pesticides

Analytical standards of lambda-cyhalothrin (97.5%), DDTs (97.0-99.5%), dimethoate (98.0%), endosulfan (98.0-99.5%), HCHs (97.7-98.5%), and methoxychlor (95%) were supplied by Dr Ehrenstorfer, D-86199 Augsburg, Germany. All organic solvents used were of GC grade (Sigma, Munich, Germany or BDH, VWR International, UK).

All glassware and other apparatus such as spatula were washed thoroughly with detergent and rinsed with water and then with acetone.

All samples were transported to pesticide residues laboratory, Ghana Atomic Energy Commission, and were refrigerated (at 5 ^{o}C).The samples were blended with electronic blender and were transferred into a “zip-lock” rubber bags.

The extraction procedure was according to methods described by EPA methods 8081A and 3510 with some modification as used by Darko and Acquaah, (2007). 4 g of each homogenised sample was weighed into 50 ml centrifuge tube. 8 g of Na_2SO_4 was added, stirred and allowed to stand for 20 min. 20 ml of hexane was added to each samples and shaken for 2 min using an electronic shaker. The samples were then centrifuged at 2000 rpm for 2 min and the supernatant was filtered through a cotton wool overlaid with 2 g of Na_2SO_4 into 50 ml volumetric flask. The extraction was repeated with 20 ml hexane and 10 ml hexane. The filtrate was evaporated to less than 5 ml with a rotary evaporator at 40 ˚C, and transferred into a 5 ml volumetric flask where it was topped up with hexane to the mark.

Clean up

The florisil column (500 mg/8 ml) cartridge was conditioned with 5 ml of hexane. A receiving flask was placed under the column to collect the eluate. 5 ml of sample extract (filtrate) was loaded into the column and elute the column with 10 ml (3, 3, 4 ml) of hexane from above. The sample extract was concentrated to dryness using the rotary evaporator with water bath aided with the water chiller. The residue was dissolved with 1 ml of hexane and then transferred quantitatively into 2 ml vial for quantification on to the gas chromatograph.

The prepared sample solutions were tested by GC

General operation conditions were as follows: injector temperature 250^{0}C; detector temperature, 300^{0}C; column temperature 90^{0}C; the carrier gas was nitrogen, the sample injection volume was 1µl; and the injection mode was splitless. Optimal conditions were investigated for determination of organochlorine pesticides. Each pesticide was detected within 63minutes.

RESULTS AND DISCUSSION

Heavy metals analysis

A total of twenty samples, comprising of ten different natural spices from two different markets at Asafo and Central market, were collected for analysis. The samples were analysed for six heavy metals. Three of the metals namely zinc (Zn), manganese (Mn), and iron (Fe) are essential micronutrients and lead (Pb), cadmium (Cd) and mercury (Hg) are toxic metals.

Total Mercury (THg), one of the toxic elements with serious health implications determined range from below detectable limit to 0.035 µg/g. The PWTI for THg is 4 µg/kg body weight (bw) (JECFA, 2011). The maximum level of 1.0 mg/kg Hg has been set by EU, (2006). The levels of THg in the samples were below the maximum level set by EU, and it is unlikely to exceed PWTI set by JECFA. The maximum concentration of THg was slightly higher in this study than 0.025 mg/kg reported by Nkansah and Amoako, (2010). The cadmium levels for all the spices ranged from below detectable limit to 0.47µg/g. The PWTI for Cd is 7 µg/kg body weight (bw) (JECFA, 1989). The maximum level of 0.05 mg/kg Hg has been set by EU, (2006). A study by Nnorom *et al.* (2007) on trace metals in bouillon cubes and some natural spices such as thyme reported levels for Cd ranging from below detection to 5.05 mg/kg which is far above levels recorded for this study. Another research work done in Saudi Arabia on heavy metal levels of 18 common spices also showed high levels of 1.25 to 3.05 mg/kg for Cd (Al-Eed et al., 2002). The Zn concentrations of the samples ranged from 0.78 -11.47µg/g. A study by Nnorom *et al.* (2007) recorded a range from 1.00 to 48.10 µg/g for zinc. The Fe concentration ranged from 21.24 - 360.17 µg/g. A study by Nnorom *et al.,* (2007) reported results of some of spices recording iron levels as high as 419.05 µg/g. The Mn concentration of this study ranged from 1.77-70.0 µg/g (Table 1 below) with Grain of paradise sample from central market having the highest Mn concentration.

Pesticides Analysis

A total of twenty (20) samples comprising ten different spices were purchased. Nine (9) organochlorine pesticides; Lindane, HCB, Haptachlor epoxide, Trans-nanochlor, Dieldrin, o, p' DDT, p, p' DDT, o, p' DDE and p, p' DDE, residues were determined on the spices.

Work done in some farming communities in the Ashanti region of Ghana has indicated the presence of organochlorine pesticide residues in fish (Osafo-Acquaah, 1997) vegetables, water, sediments, mother's milk and blood samples (Ntow, 2001).

Results in Table 2 show that the mean concentration of lindane in the spices fluctuated between 2.0 and 180 µg/kg. Lindane was detected in all the 20 samples of the spices from Kumasi. The average concentration of lindane in the spices from Kumasi was 43.30 µg/kg. This level is below the WHO limit of 6.0µg/kg. The concentration of HCB in the spices ranged from below detectable limit to 166 µg/kg. The percentage detection rate in the spices was 30.0 %. Heptachlor was detected in 85.0 % of the spices collected from Kumasi. The concentration of Heptachlor in the samples ranged from below detectable limit to 920 µg/kg. The concentration of Trans – nanochlor in the spices ranged from below detectable limit to 277 µg/kg. The percentage detection of Trans –nanochlor in the spices collected was 90.0%.

The concentration of Dieldrin in the spices ranged from below detectable limit to 52 µg/kg. Out of 20 spices collected from the markets in Kumasi, 25.0% were found to contain Dieldrin. From the result, out of 20 spices collected, 80 % contained p, p' DDT and 35% contained o, p' DDT. The concentration of p, p' DDT ranged from below detectable limit to 90.0 µg/kg. The WHO recommended maximum residue limit of p, p' DDT is 500 µg/kg. The concentration of o, p' DDT ranged from below detectable limit to 96.0 µg/kg.

The concentration of o, p' DDE ranged from below detectable limit to 90 µg/kg with a percentage detection rate of 25% for the samples collected from markets at Kumasi. The concentration of p, p' DDE ranged from below detectable limit to 122 µg/kg with 75 % of the samples being detected to contain p, p' DDE. The results from the study show that residues of organochlorine pesticides are present at concentrations close to the WHO Maximum Residue Levels (MRLs) in the spices samples. These pesticides might have originated from pesticides used.

CONCLUSION

The results from this study show that heavy metals and residue of organochlorine pesticides are present in spices with concentrations lower than their maximum recommended residue levels. Although these residues occurred at very low concentrations in the samples, they may accumulate to higher levels in human beings who consume these products. The results will help in a scientific assessment of the implications of pesticide residues with regards to human risks in Ghana.

Table 1. Average metal concentrations (µg/g) in spices

Spice	Location	Hg (µg/g)	Pb µg/g	Cd (µg/g)	Mn (µg/g)	Fe (µg/g)	Zn (µg/g)
Rosemary (RO)	Asafo Market	0.001	<dl	<dl	5.08	162.94	3.5
	Central Market	0.003	<dl	0.39	6.13	202.16	2.52
Nutmeg (NU)	Asafo Market	0.008	<dl	<dl	7.84	27.66	3.03
	Central Market	0.005	0.88	<dl	10.04	22.06	3.16
Ashanti pepper (AS)	Asafo Market	<dl	<dl	0.23	11.53	36.49	4.23
	Central Market	0.001	<dl	0.19	16.94	32.83	5.04
Garlic (GA)	Asafo Market	0.005	<dl	<dl	1.77	47.63	2.8
	Central Market	0.001	<dl	<dl	2.76	21.71	2.98
Anise (AN)	Asafo Market	0.002	<dl	<dl	11.2	105.6	8.64
	Central Market	0.002	<dl	0.47	25.78	360.17	11.47
Red pepper (RE)	Asafo Market	0.001	<dl	0.17	6.82	108.09	4.32
	Central Market	0.002	<dl	<dl	5.08	236.65	5.27
Ginger (GN)	Asafo Market	0.005	0.99	<dl	26.9	130.04	3.41
	Central Market	0.001	1.36	<dl	57.41	119.39	1.45
Senegal pepper (SE)	Asafo Market	0.001	<dl	<dl	7.08	31.11	1.67
	Central Market	0.002	<dl	<dl	20.83	27.47	1.74
Aidan (AI)	Asafo Market	0.035	<dl	<dl	5.05	22.43	0.78
	Central Market	0.002	<dl	<dl	9.34	21.24	1.59
Grain of paradise (GR)	Asafo Market	0.005	<dl	<dl	55	65.06	2.3
	Central Market	0.003	1.81	<dl	70	71.5	1.64

<dl means below detectable limit

Table 2. Average Organochlorine pesticides concentrations (µg/kg) in spices

Spice	Location	Lindane	HCB	Hepatchlor epoxide)	Trans-nanochlor	Dieldrin	o,p-DDT	p,p-DDT	o,p-DDE	p,p-DDE
Rosemary (RO)	Asafo Market	9.0	<dl	22.0	19.0	<dl	<dl	61.0	<dl	3.0
	Central Market	70.0	70.0	142.0	30.0	<dl	<dl	90.0	<dl	20.0
Nutmeg (NU)	Asafo Market	69.0	22.0	128.0	36.0	<dl	1.0	52.0	1.0	44.0
	Central Market	70.0	19.0	130.0	40.0	<dl	<dl	60.0	<dl	40.0
Ashanti pepper (AS)	Asafo Market	59.0	73.0	50.0	149.0	13.0	45.0	47.4	9.0	105.0
	Central Market	83.0	166.0	920.0	277.0	27.0	96.0	25.0	90.0	122.0
Garlic (GA)	Asafo Market	22.0	<dl	28.0	9.0	<dl	<dl	1.0	<dl	41.0
	Central Market	20.0	<dl	220.0	40.0	<dl	<dl	<dl	<dl	50.0

Table 2. Continue

Anise (AN)	Asafo Market	180.0	<dl	220.0	10.0	<dl	<dl	<dl	<dl	10.0
	Central Market	25.0	<dl	303.0	40.0	<dl	<dl	68.0	<dl	30.0
Red pepper (RE)	Asafo Market	2.0	<dl	28.0	21.0	41.0	22.0	<dl	14.0	13.0
	Central Market	38.0	<dl	54.0	40.0	52.0	23.0	<dl	13.0	17.0
Ginger (GN)	Asafo Market	7.0	<dl	2.0	3.0	<dl	<dl	6.0	<dl	<dl
	Central Market	20.0	<dl	14.0	30.0	<dl	42.0	71.0	<dl	<dl
Senegal pepper (SE)	Asafo Market	16.0	<dl	<dl	5.0	<dl	<dl	5.0	<dl	40.0
	Central Market	24.0	59.0	28.0	26.0	27.0	57.0	22.0	<dl	50.0
Aidan (AI)	Asafo Market	7.0	<dl	<dl	<dl	<dl	<dl	8.0	<dl	<dl
	Central Market	10.0	<dl	<dl	<dl	<dl	<dl	10.0	<dl	<dl
Grain of paradise (GR)	Asafo Market	49.0	<dl	22.0	2.0	<dl	<dl	4.0	<dl	13.0
	Central Market	86.0	<dl	18.0	10.0	<dl	<dl	21.0	<dl	<dl

<dl means below detection limit

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