PESTICIDE RESIDUES CONTAMINATION OF VEGETABLES AND THEIR PUBLIC HEALTH IMPLICATIONS IN GHANA

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ABSTRACT

This study aimed at determining the residues concentrations of pesticides in vegetables, and assessing the health risk due to the daily consumption of contaminated vegetables for adults in the prominent farming community of Akumadan in Ghana. Fifteen pooled samples, belonging to five vegetable types (tomato, cabbage, pepper, onion and eggplants) purchased from local markets across Ghana were analysed for DDTs, endosulfan, HCHs, methoxychlor, dimethoate and lambda-cyhalothrin using GC-MS. The concentrations of the pesticides ranged from 0.01 to 46.95 µg/kg wet weight. Dietary data were collected from 130 residents. By combining the dietary and contaminant data, dietary exposure to the identified pesticides was calculated. Dietary exposure to pesticide residues at Akumadan is low and there is no associated health risk. However, the results of persistent contaminants are of particular health concern because of this persistence. The estimated exposure distributions were shown to be insensitive to valuation of the non-detect residue samples

Keywords: *contamination, environment, health risk, pesticide residues and vegetables*

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INTRODUCTION

Pesticides applied to food crops in the field can leave potentially harmful residues. Organochlorine pesticides in particular can persist in foodstuffs for a considerable period. If crops are sprayed shortly prior to harvest without an appropriate waiting period, even organophosphate residues can persist up until the food is in the hands of the consumer (Bull, 1992). In Ghana, the increase in urban population and food demand has catalysed the use of chemical pesticides for food production (Amoah, Drechsel, Abaidoo and Ntow, 2006).

Vegetables are grown extensively in Ghana, and constitute large portion of the diet of the average Ghanaians. Vegetables are essential for a healthy and balanced diet, as well as adding variety, interest and flavour to the menu. But vegetables also attract a wide range of pests and diseases, and require intensive pest management. About 87% of the farmers who grow vegetables in Ghana use pesticides (Dinham, 2003). Many of these farmers spray the same wide range of pesticides on all vegetables and ignore pre-harvest intervals (Ntow, Gijzen, Kelderman and Drechsel, 2006). Sometimes farmers spray pesticides one day before harvest to sell 'good-looking' vegetables. This practice, in particular, exposes consumers to pesticides.

Though it is sometimes thought that residues are destroyed if food is properly washed and cooked, this is not always the case. Washing and cooking may reduce pesticide residues in food; boiling may remove only 35-60% of organophosphate residues and 20-25% of organochlorines (Bull, 1992). Residues above tolerance limits do occur in cooked food. Consumption of contaminated food is an important route of human exposure to pesticide residues, and may pose a public health risk (MacIntosh et al., 1996). The objective of this study, therefore, was to determine the levels of pesticide residues in vegetables from Ghanaian markets and estimate the cancer and non-cancer health risk due to dietary exposure to vegetable for the adult population of Akumadan, a prominent vegetable farming community in Ghana.

MATERIALS AND METHODS

Sampling of vegetables: Identical vegetable items were purchased from local designated markets across all the 10 regions in Ghana. The vegetable items, which were tomato, pepper, onion, cabbage, and eggplants were considered representative of the diet of the Ghanaian population according to a survey in 2005 (Ntow et al., 2006). At each market, vegetable samples were collected from three randomly selected sellers. Three composite samples of identical vegetable items (each containing 3 bulbs of tomato, 3 cabbages, 10 pepper fruits, 3 bulbs of onion, and 3 eggplants) were drawn from each market. The samples were wrapped in aluminium foil according to type, packed in polythene bags, and transported to the CSIR Water Research Institute Laboratory in Accra where they were given a cold-water wash with a soft brush to remove adhering soil particles. Samples were kept in a freezer at - 4^{o}C until required for extraction, which was carried out within 24hrs of arrival at the laboratory in Accra.

Sample preparation and extraction: 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).

In preliminary studies, the various sample types (homogenised vegetable samples placed in 250ml standard joint borosilicate bottles) were spiked with appropriate volumes of previously prepared stock solutions of pesticide compounds. Each fortification level was prepared in three replicates. The bottles were capped, manually shaken to ensure thorough mixing, and stored in a deep freezer at -4^{o}C for 24hrs to simulate sample storage conditions. The average recoveries for all compounds varied from 83-91%. Triplicate analysis also gave a standard error of about 10%. Residue data were not corrected for efficiency of recovery. The limit of quantification (LOQ) was set to 0.01g/kg fresh weight for all compounds (calculated from real samples as being 10 times the signal to noise ratio). During the sample extraction, blanks were regularly processed (one in ten).

The frozen samples were thawed and each cut into four equal segments (quartered). Opposite segments were discarded in order to reduce the bulk of the material needing to be processed. Following this step, the vegetables were cut into small pieces. For the analysis, individuals of identical vegetable item were pooled as one composite. From this composite, three replicate samples were prepared for extraction and subsequent instrumental analysis. This design yielded three residue values for each vegetable item. Sample preparation and extraction followed the procedures described in Ferrer *et al.* (2005). About (10g) of each vegetable was weighed into a porcelain mortar, and ground with 50g of anhydrous sodium sulphate. The powdered sample was extracted in ethyl acetate. The extract was rotary evaporated at 40^{o}C, and the residue re-dissolved in hexane. Sample clean up followed the procedure of Hsu, Biggs and Saini (1991).

Instrumental analysis: The analysis of vegetable samples for pesticide compounds was performed on a GC-MS (Agilent 6890 Series GC System) coupled with an Agilent 5973N mass selective detector-electron impact ionization. The capillary column was HP-5MS (length 30m; I.D 0.25mm and film thickness 0.25µm) and packed with 5% phenyl methyl siloxane. The GC-MS was operated in the selected ion-monitoring mode at the following conditions: injection port 250^{o}C (splitless, pressure 22.62psi; purge flow 50ml/min; purge time 2.0min; total flow 55.4ml/min). Column oven: initial 70^{o}C held 2min, programming rate 25^{o}C/min (70 to 150^{o}C); 10oC/min (150 to 200^{o}C); 8^{o}C/min (200 to 280^{o}C) and held 10min at 280^{o}C. The carrier gas was nitrogen at 15psi; detector make-up, 30 ml/min. The injection volume was 1µl (Agilent 7683 Series injector).

Risk assessment: Health risk assessment was performed based on vegetable consumption data obtained for Akumadan, a prominent farming village located at about 95km northwest of Kumasi (1°57'W, 7°24'N) (Ntow, 2001) in the Ashanti Region. The inhabitants of Akumadan have a long history of relying upon vegetables as major component of their diet, of relying on vegetables for commerce, and of valuing vegetables for their cultural and social importance.One hundred and thirty (130) healthy adults were randomly sampled from the general population. Data collected for each vegetable included frequency of consumption over the past 12 months (number of times per day, week, month or year) and the quantity consumed on each occasion. (Ntow *et al.*, 2006). The total daily dietary exposure, E_t (mg/kg body weight), that results from eating a combination of contaminated vegetables was calculated as follows (USEPA, 1989):

$$E_t = \sum_{i=I}^{n} \left(C_f\right)_i \left(L\right)_i \qquad \text{..(1)}$$

where

C_f = the concentration (mg/kg) of the contaminant in the vegetable,

L = the amount (kg/day body weight) of contaminated vegetable consumed

I = the number of different vegetable types consumed.

For each contaminant, the average daily dietary exposure, E (mg/kg body weight), level for the population was calculated by:

$$E = C_f \times L \qquad \text{..(2)}$$

Vegetable consumption was expressed as daily consumption divided by body weight, which was set at 50kg for an adult (Tejada *et al.*, 1995). To account for residues concentrations that were below the limit of quantification, two alternative assumptions were used. The first assumption that actual concentrations for non-detect (ND) samples were equal to the limit of quantification. However, this would overestimate the concentrations if there would be no contamination of the sampled vegetables. Therefore, exposures with the alternate assumption that actual concentrations for all non-detect samples were equal to zero were also calculated. These two assumptions provide a range of exposure estimates for each contaminant with non-detects.

To assess potential public health risks, exposure concentrations were compared to the minimal risk levels (MRLs) for the individual pesticide components. These were derived from the United States Agency for Toxic Substances and Disease Registry (ATSDR) in a manner similar to the way in which the reference dose (RfD) and reference concentration are determined by the United States Environmental Protection Agency (USEPA) (Jiang et al., 2006). The oral MRLs are for: total DDTs 0.0005 (listed as p,p'-DDT); Endosulfan 0.002; HCHs 0.00001 (listed as γ-HCH); cyhalothrin 0.01, and methoxychlor 0.005mg/kg/day (ATSDR, 2007). For dimethoate an RfD value of 0.0002 mg/kg/day was applied (EXTOXNET, 2007). The non-cancer health risks from the consumption of vegetables by Akumadan residents were assessed by estimating the relevant hazard ratios (HRs) (Equ. 3). These hazard ratios were calculated by dividing the average daily exposure by the MRL. A HR > 1 indicates that the average exposure level exceeded the benchmark concentration (Dougherty et al., 2000).

Hazard Ratio (HR) = Average daily exposure/Minimal risk level ....(3)

RESULTS AND DISCUSSION

The results for pesticide residues that were detected in the five types of vegetable crops are shown on Table 1. Residues concentration of DDTs ranged from 0.01 to 0.45µg/kg wet weight, with the highest concentration in cabbage. Among the pesticides detected in cabbage, endosulfan showed the highest mean concentration. Lambda cyhalothrin was the only pesticide that did not show up in quantifiable amounts in cabbage. While all the measured pesticides were detected in quantifiable amounts in tomato and onion, pepper and eggplants exhibited quantifiable amounts of only three of the pesticides. Relatively high residue concentrations of methoxychlor were found in all vegetables except in cabbage. Mean concentrations of methoxychlor varied markedly among the vegetables, and were highest in onion, and lowest in cabbage. According to the dietary survey, a healthy adult ate 565 ± 108 g vegetables each day (Table 1). There was a gender-specific difference in the rate of vegetable consumption: women consumed more vegetables than men. In this survey, five vegetable crops were examined for individual dietary consumption (Table 2). Tomato and eggplants, together accounted for about 70% of total vegetable consumption in the survey population, and the order of consumption was tomato > eggplants > onion > pepper > cabbage (Table 2).

The results of an evaluation of the non-cancer risks to human health associated with the consumption of vegetables containing DDTs, endosulfan, HCHs, methoxychlor, dimethoate, and lambda cyhalothrin is shown in Fig. 3. The hazard ratios (HRs) of non-cancer risk (based on ND = 0 and ND = limit of quantification) at Akumadan were all < 1 for vegetables. The estimated pesticides dietary intakes and their associated HRs were insensitive to the treatment of non-detect residue samples (Table 3 & Fig. 3). For instance, changing the non-detect value from zero to the limit of quantification did not markedly change the exposure levels for all of the contaminants. The value assigned to non-detects also did not have an effect on the calculated HRs. The potential health risk of a daily intake of HCHs-contaminated vegetable had a HR of over 0.10 which was due to consumption of tomato and onion, followed by methoxychlor and dimethoate, with HRs of over 0.05 and 0.03 for vegetables. Onions were found to be the largest contributor to lambda cyhalothrin exposures, accounting for 89% of the estimated distribution. Among the vegetable items, tomato and onion were the principal contributors to the average pesticide exposure among individuals (Table 3).

Endosulfan, dimethoate, lambda cyhalothrin, DDTs, HCHs and methoxychlor were detected in vegetable samples. Endosulfan, dimethoate, and lambda cyhalothrin are among the most commonly used pesticides in vegetable production in some parts of Ghana. For instance, vegetable farmers in a survey by Ntow *et al.* (2006) admitted to spraying lambda cyhalothrin (Karate 2.5 EC/ULV), dimethoate (Perferkthion 400 EC) and endosulfan (Thionex 35 EC/ULV, Thiodan 50 EC) on tomato, pepper, okra, eggplant, cabbage and lettuce. Although DDTs, HCHs and methoxychlor did not appear to be used in vegetable farming (they are banned from agricultural use), their occurrence in the samples are not surprising. This is mainly because these chemicals (organochlorines), relative to other classes of pesticides are resistant to environmental degradation, which allows them to accumulate in plant and animal tissue. Besides, vegetable farmers in Ghana use pesticides which are not registered and which may have found their way into the country through unapproved routes.

Generally, vegetable farming in Ghana is fraught with abuse, misuse and overuse of pesticides. Pests and diseases pose big problems in vegetable production and these have led many farmers to use chemical pesticides, even if they have received no training in the choice of chemicals or application technique. The concentrations of DDTs, endosulfan, and lambda-cyhalothrin measured in vegetables in this study were lower than those measured for lettuce in Amoah et al. (2006). The results regarding pesticide residues also indicate that several pesticides are used within a crop-growing season. As also described by Danso, Drechsel and Fialor (2002) and Ntow *et al.* (2006), vegetable farmers mix cocktails of various pesticides to increase the potency of the compounds.

Since vegetables are one of the most important food sources in Ghana, intake of especially toxic pesticides from vegetables is of great concern for human health risk. This study provided insight into the magnitude of potential exposures from vegetable contamination. Initial screening of the data presented showed that estimated exposure to a number of contaminants in the average vegetable diet of adults did not exceed benchmark concentration for non-cancer effects. This indicates that exposure to pesticides was unlikely to result in any adverse health effects. However, it is important to note that some of the identified pesticides have been banned for agricultural uses in Ghana. DDT, for example, has been banned for over a decade. Lindane (γ-HCH) is among the list of restricted pesticides in Ghana. However, because many of these compounds, such as DDT, are highly persistent, they continue to show up at significant levels in the food supply, indicating the importance of considering persistence when assessing potential risks.

Health risk assessment is very subjective (Raschke and Burger, 1997) and is only as accurate as the available information. There are a number of important limitations in this study. A first source of uncertainty in this analysis arises from the vegetable consumption data. Data on vegetable consumption were collected from a sub-population of farmers, which may not be representative of dietary patterns of members of certain age, ethnic, socio-economic, and other subpopulations. Better characterisation of the consumption of vegetables across a wider sample size and occupational diversity may be needed to reduce the uncertainty associated with the consumption data. Other equally important uncertainties, which this study did not consider are cancer risks, possible interactions among various toxic chemicals, and different vegetable consumption patterns within and between populations. In spite of the limitations associated with the analysis, the results point to potential exposures to contaminants in vegetables and represent an important step toward better characterisation of these exposures.

CONCLUSION

Average daily dietary exposures to 6 vegetable contaminants for 130 Ghanaian adult males and females at Akumadan were assessed. For all of the chemicals included in the analysis, estimated exposures were due to consumption of vegetable items that contained measurable levels of the contaminant, and were insensitive to valuation of the non-detect residue samples (because of the low detection rate observed in the residues data for these chemicals). This analysis has focussed on exposure to contaminants that result from their presence in one type of food source - vegetables. It is commendable to extend the scope of exposures to include food items such as fruits, cereals and other exposure media such as outdoor air and surface water. The range of contaminants should also be expanded to include carcinogenic agents such as PCBs, dioxins, and heavy metals. Strategically, agricultural extension service as well as public education should be carried out to educate vegetable farmers in Ghana, the public health implication in the use certain pesticides in vegetable farming.

Table 1: Concentrations [mean (SE) in µg/kg wet weight] of pesticide residues in vegetables from Ghanaian markets (n = 3)

	Tomato	Cabbage	Pepper	Onion	Eggplants
DDTs[a]	0.03 (0.01)	0.45 (0.16)	< 0.01	0.02 (0.01)	0.01 (0.01)
Endosulfan[b]	0.21 (0.09)	3.03 (0.75)	< 0.01	1.85 (0.84)	< 0.01
HCHs[c]	0.22 (0.06)	0.62 (0.12)	0.34 (0.14)	0.08 (0.02)	< 0.01
Methoxychlor	28.21 (0.38)	0.85 (0.14)	7.43 (2.15)	46.95 (0.34)	1.00 (0.03)
Dimethoate	0.01 (0.01)	0.43 (0.26)	< 0.01	1.76 (0.13)	0.22 (0.07)
Lambda cyhalothrin	0.05(0.01)	< 0.01	0.01 (0.01)	0.11 (0.03)	< 0.01

[a] *p,p*'-DDE + *p,p,*-DDT + *p,p,*-DDD

[b] -endosulfan + -endosulfan + endosulfan sulphate

[c] -HCH + -HCH + -HCH + -HCH

Table 2: Daily consumption (mean ± SE) of various vegetables in the Akumadan population

Vegetables	Daily consumption (g/person)	Percentage of total consumption (%)
Tomato	200 ± 47	35
Cabbage	5 ± 2	1
Pepper	10 ± 6	2
Onion	150 ± 92	27
Egg plants	200 ± 38	35
Total	565 ± 108	100

Table 3: Calculated dietary intake of pesticides from consumption of vegetables

Pesticide components	Daily intake (pg/kg body weight)					
	Tomato	Cabbage	Pepper	Onion	Eggplants	Total intake
ND [a] = 0						
DDTs [b]	120	40	-	60	40	260
Endosulfan [c]	840	300	-	5550	-	6690
HCHs [d]	880	60	70	240	-	1250
Methoxychlor	113000	90	1490	140000	4000	258580
Dimethoate	40	40	-	5280	880	6240
Lambda cyhalothrin	40	-	2	330	-	372
ND = LOQ [e]						
DDTs	120	45	2	60	40	267
Endosulfan	840	303	2	5550	40	6735
HCHs	880	62	68	240	40	1290
Methoxychlor	1128	85	1486	140850	4000	259261
Dimethoate	40	43	2	5280	880	6245
Lambda cyhalothrin	40	1	2	330	40	413

[a] Non-detect (that is, below the limit of quantification)

[b] *p,p*'-DDE + *p,p,*-DDT + *p,p,*-DDD

[c] -endosulfan + -endosulfan + endosulfan sulphate

[d] -HCH + -HCH + -HCH + -HCH

[e] Limit of quantification

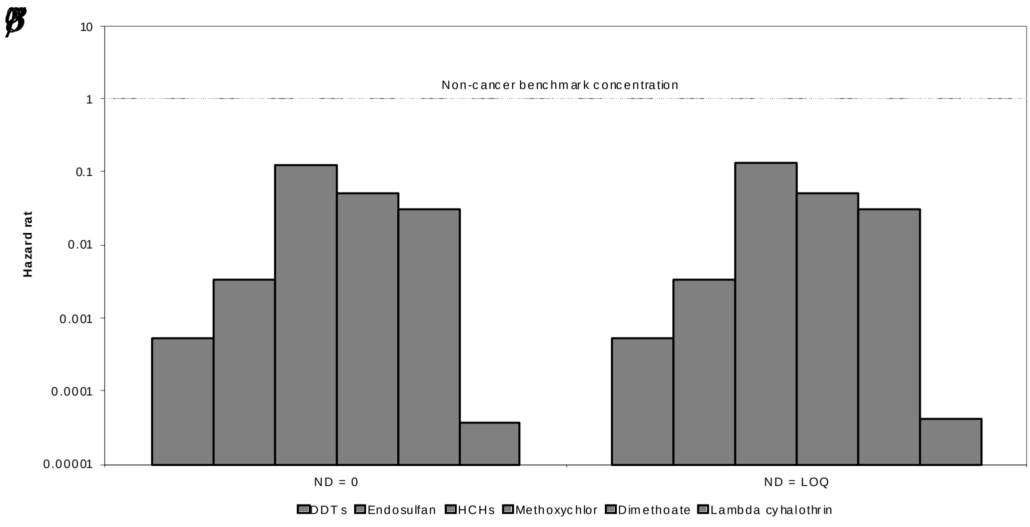

Fig.3: Non-Cancer hazard ratios for daily tomato, cabbage, pepper, onion and eggplants consumption by people of Ghana

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