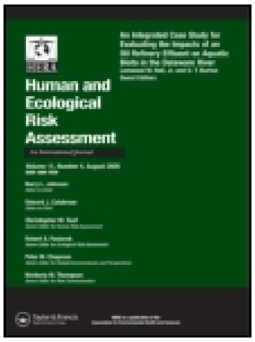

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Analysis of Some Pesticide Residues in Tomatoes in Ghana

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ABSTRACT

Pesticide residues, both natural and synthetic, can be found in most of the things we eat, for example, fruits, vegetables, bread, meat, poultry, fish, and the processed foods made from them. Some of this pesticide contamination is legal, but does this mean it is safe? Much of it is illegal, with residues found in excess of regulatory safe levels. Identifying and determining the level of trace contaminants in our food and environment is critical in protecting and improving human health and the environment. This study evaluates the residue levels of select pesticides used on tomato crops in Ghana that are likely to have accumulated in the tomatoes during application. The results obtained confirm that pesticide residues were indeed present in the tomatoes and further analysis quantified the amount present. Analysis of some organochlorine and organophosphorus residue levels in the fruits indicated that chlorpyrifos, which is an active ingredient of pesticides registered in Ghana under the trade name dursban 4E or terminus 480 EC for use on vegetables, has the greatest residue level of 10.76 mg/kg. The lowest residue level observed was that of pirimiphos-methyl with 0.03 mg/kg. Human health risk assessment was performed on the results obtained from the analysis using Human Health Evaluation computerized software-RISC 4.02. The risk assessment showed cancer risk for adults and children due to the presence of endosulfan and chlopyrifos. Endosulfan is not registered in Ghana as a pesticide for use on vegetables, therefore the detection of endosulfan in several samples indicates misuse of agrochemicals among Ghanaian farmers.

Key Words: pirimiphos-methyl, fenitrothion, lambda-cyhalothrin, chlorpyrifos, folpet, cypermethrin, endosulfan.

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INTRODUCTION

Food, fresh water, and salt water are subject to extensive, and often illegal, contamination by a cocktail of toxic synthetic and natural pesticides. As pesticides break down in the environment at different rates, the more persistent ones (which resist breakdown) can contaminate food and water for years. Even though we only consume these residues in small amounts, little is known about their long-term effects on human health in Ghanaian residents.

The use of pesticides during agricultural production has increasingly been causing concerns due to their adverse effects on human health. A pesticide is any substance or mixture of substances intended for preventing or controlling any unwanted species of plants and animals and also includes any substances or mixture of substances intended for use as a plant-growth regulator, defoliant, and desiccant (Hicks 2000). The term "pesticide" includes any substance used for the control of pests during the production, storage, transport, marketing, or processing of food for man or animals or which may be administered to animals for the control of insects or arachnids in or on their bodies. Pesticides are toxic chemicals used in preventing, destroying, repelling, or mitigating pests (NRC 1993; USEPA 2005)

Pesticide residues in or on plants may be unavoidable even when pesticides are used in accordance with Good Agricultural Practice (PS 1997; Uysal-Pala and Bilisli 2006). Pesticide residues are reduced by processing or household preparation stages such as washing, peeling and cooking, and so on (Petersen *et al.* 1996; Dikshit *et al.* 2003). Processing studies allow a better estimate of the consumer exposure to the residues (PS 1997). Endosulfan is a broad-spectrum organochlorine insecticide and acaricide used to kill mites. Technically, endosulfan contains a mixture of α- and β-isomers in the approximate ratio of 70:30. In soil and on plant surfaces, endosulfan sulfate is the primary degradation product of endosulfan. Endosulfan is a highly toxic pesticide in U.S. Environmental Protection Agency (USEPA) toxicity class I and its suspected endocrine disrupter (EXTOXNET 1996; IPCS 1998; EC 2000; FOE 2001). Some endosulfans have been listed as potential endocrine disrupters by the German Federal Environment Agency (EXTOXNET 1995; Warhurst 2004).

Pesticide residues, both natural and synthetic, can be found in all the things we eat (*e.g.*, fruits, vegetables, bread, meat, poultry, fish, and the processed foods made from them). Some of this contamination is legal, but does this mean it is safe? Much of the contamination is illegal, with residues detected in excess of legal safety levels (Hurst *et al.* 1991).

Identifying and determining the level of trace contaminants in our food and environment is critical to protecting and improving human health and the environment. The accurate measurement of residues helps to better protect our community and develop superior production practices.

Good agricultural practices in the use of pesticides is the officially recommended or authorized usage of pesticides under practical conditions at any stage of production, storage, transport, distribution, and processing of food and other agricultural commodities, although there are variations in requirements within and between regions and minimum quantities necessary to achieve adequate control. Pesticides being applied in such a manner leave residues in the smallest amount, so as not to cause harm to humans or animals during their life time (FAO/WHO 1975).

Some pesticides registered for use on vegetables in Ghana include the following: cypermethrin, chlorpyrifos, fenitrothion, folpet, lambda-cyhalothrin, and pirimiphos-methyl. Endosulfan is not registered to be used on vegetables in Ghana.

Over the years pesticide use has become a common agricultural practice in Ghana. However, lack of knowledge of the types, uses, and the effects of these pesticides among small-and large-scale farmers has resulted in their misuse and consequently their accumulation in various foods and feed items.

Ghana is mainly an agricultural nation with most of its food crops produced by local farmers. Levels of chemicals in these food crops are of great concern. Over time, these pesticides can accumulate in the bodies of humans, causing various health-related problems, such as disrupting the endocrine system, which can influence development, growth, reproduction, and behavior. Therefore, it is important to know the type of pesticides that are used locally on food crops and their health effects and thus the need for pesticide residue analyses.

MATERIALS AND METHOD

Equipment

The Gas Chromatograph (GC) used for the pesticide residue analysis was a Varian CP-3800 series equipped with the ^{63}Ni selective Electron Capture Detector and Phosphorus Detector. The GC column employed was capillary column, GS-Q (30 m $\times$ 0.53 mm i.d), J&W Scientific, CA, USA, for organochlorine and organophosphorous pesticides. Also used were a rotary vacuum evaporator (B'U'CHI type), Hamilton Beach Commercial Blender, a Libror EB-3200s weighing balance with capacity of 3200 g and a B'U'CHI water bath, B-480/B-485.

Sample Collection

Tomato fruits were collected (different market places) from Kumasi in the Ashanti region and Cape Coast in the central region of Ghana. The samples from Kumasi, which were cultivated at Bolgatanga and Akumadan in the Upper East and Brong Ahafo regions of Ghana, respectively, were labelled 1 and 4. The other samples collected from Cape Coast were cultivated in Burkina Faso, which shares a boundary with Ghana to the north were labelled 2 and 3. In all, about 200 tomato fruits were sampled for the study.

Sample Preparation

Approximately 650 g of the fresh samples were weighed and each sample was blended into a paste before being extracted. The entire reagent used was of the analytical grade supplied by BDH Chemical Ltd., UK.

Process of Extraction

The method of extraction used was USEPA Method 3510 (Edgell and Wesselman 1989) for extracting multi-residue pesticides in non-fatty crops. The extraction was done with ethyl acetate as the solvent. Sodium hydrogen carbonate ($NaHCO_3$) was used to neutralize any acid that may be present and anhydrous sodium sulphate (Na_2SO_4) was used to remove water from the sample matrix.

A 100 g portion of the paste was weighed into a flat bottom flask and 40 mL ethyl acetate was added and the mixture was shaken. A 5 g portion of sodium hydrogen carbonate was added to the mixture followed by 20 g of sodium sulphate, and the entire mixture was shaken vigorously. The mixture was centrifuged at a speed of 1800 rmp for 5 min, and then the organic layer was decanted into a round bottom flask. This procedure was repeated for four other 100.0 g portions of the same sample. This was repeated for the other samples (*i.e*., 2, 3, and 4).

The extracts for each sample were concentrated using a rotary evaporator and each concentrate was dissolved in 2.0 mL *n*-hexane for clean-up. The clean-up was done to remove extraneous materials from the extract before analysis.

Procedure for Clean-up (USEPA Method 3620B)

A 10 mm chromatographic column was filled with 3 g activated silica gel and topped up with 2 to 3 g of anhydrous sulphate. Next, 10 mL of *n*-hexane was added to the column by opening the tap and allowed to run through it to wet and rinse the sodium sulphate and the silica gel. The residue in 2 mL *n*-hexane was then transferred onto the column and the extract vial rinsed thrice with 2 mL hexane and added to the column. The column was then eluted with 80 mL *n*-hexane at a rate of 5 mL/min into a round bottom flask as fraction one. The column was eluted again with 50 mL *n*-hexane at the same rate into a round bottom flask and the eluent as fraction two. A third elution was done using 15 mL dichloromethane and collected at the same flow rate into a round bottom flask as fraction three (USEPA 1989a).

This procedure was repeated for the concentrates of the other three samples. All three fractions of each sample were concentrated to dryness using a rotary evaporator. Each residue was then dissolved in 2 mL ethyl acetate for GC (gas chromatograph) analysis. The Electron Capture Detector (ECD) and Flame Photometric Detector (FPD) were used for organochlorine and organophosphorous species, respectively.

Quality Control

The efficiency of the method or recovery percentage from the tomatoes was evaluated using reference solutions. Spike levels of reference solutions to the samples had recovery percentages that ranged between 63–95% and relative standard deviations (RSD) ranging between 3–26%.

RESULTS AND DISCUSSION

The concentration of the organochlorine and organophosphorus pesticides found in the tomato fruits sampled from Kumasi and Cape Coast markets are presented in Tables 1 and 2. Samples were analyzed for organochlorines including folpet, α-endosulfan, β-endosulfan, lambda-cyhalothrin, cypermethrin, and organophosphorus pesticides that included pirimiphos-methyl, chlorpyrifos, and

Table 1. Organochlorine pesticides.

Samples	Folpet (mg/kg)	α-Endosulfan (mg/kg)	β-Endosulfan (mg/kg)	Lambda-Cyhalothrin (mg/kg)	Cypermethrin (mg/kg)
1	0.52	0.08	0.09	0.05	1.26
2	0.22	0.30	0.43	0.14	3.46
3	0.35	0.22	0.04	1.45	0.35
4	0.35	0.09	0.03	0.35	0.96
MRL	**2.00**	**0.50**	**0.50**	**0.10**	**0.50**

fenitrothion. These chemicals are the active ingredients that are sold under various trade names as pesticides for vegetables.

The great demand for quality and disease-free agricultural product has resulted in the use of sophisticated chemicals to meet such needs. The frequent use of these chemicals can result in their accumulation in food crops and consequently in the people who consume them.

The aim of this project was to ascertain whether pesticide residues in tomato fruits are significant enough to merit the maximum attention from the public. To do this, it was necessary to sample tomato fruits that were readily available to the public/consumers and then analyze them to determine whether there are residues present and then quantify them. The results confirmed that pesticide residues were indeed present in the fruits of tomatoes and further analysis indicated the amount that was present. Analysis of some organochlorine and organophosphorus residue levels in the fruits indicated that chlorpyrifos, which is an active ingredient of pesticides registered in Ghana under the trade name dursban 4E or terminus 480 EC for use on vegetables, has the greatest residue level of 10.76 mg/kg. The lowest residue level observed was that of pirimiphos-methyl at 0.03 mg/kg.

The greatest concentrations recorded for each pesticide were as follows: folpet (0.52 mg/kg), α-endosulfan (0.30 mg/kg), β-endosulfan (0.43 mg/kg), lambda-cyhalothrin (1.45 mg/kg), cypermethrin (3.46 mg/kg), pirimiphos-methyl (0.15 mg/kg), fenitrothion (0.09 mg/kg), and chlorpyrifos (10.76 mg/kg). Each of these chemicals has their allowed maximum residue levels in various vegetables that have been indicated in Tables 1 and 2.

The minimum concentrations recorded for each pesticide were as follows: folpet (0.22 mg/kg), α-endosulfan (0.08 mg/kg), β- endosulfan (0.03 mg/kg), lambda-cyhalothrin (0.05 mg/kg), cypermethrin (0.35 mg/kg), pirimiphos-methyl (0.03 mg/kg), fenitrothion (0.00 mg/kg), and chlorpyrifos (0.18 mg/kg).

Table 2. Organophosphorus pesticides.

Sample	Pirimiphos-methyl (mg/kg)	Chlorpyrifos (mg/kg)	Fenitrothron (mg/kg)
1	0.03	0.18	ND
2	0.07	0.70	ND
3	0.11	10.76	0.09
4	0.15	0.20	0.04
MRL	**1.00**	**0.50**	**0.01**

For tomatoes, the allowed Maximum Residue Limits (MRL) for cypermethrin, chlorpyrifos, and α- and β-endosulfan are 0.5 mg/kg each. For folpet its MRL is 2.0 mg/kg, which is valid from 2001 to 2011. The MRL for fenitrothion is 0.010 mg/kg, which has been valid since 2006. That of lambda-cyhalothrin is 0.1 mg/kg, which has been valid since 2006. Pirimiphos-methyl has a MRL of 1.0 mg/kg, valid since 2005 (EC 2002, 2003; Codex 2006a,b).

The concentrations of α- and β-endosulfan in each of the samples were less than their MRLs, indicating that consumers are safe from any effects that might result from ingestion of contaminated foods. However, endosulfan is not registered in Ghana as a pesticide for use on vegetables, thus, their presence in tomatoes indicates misuse of agrochemicals among Ghanaian farmers. The central nervous system is the primary target affected by exposure to β-endosulfan. Inhaling, eating, or drinking high doses of endosulfan can cause convulsions and death (ATSDR 1995).

Tomato fruits from Burkina Faso have greater concentrations of endosulfan as compared to those grown inland. The Ghana EPA allows the use of chlorpyrifos and cypermethrin on vegetables. The results indicated that the residue levels of cypermethrin in samples 1, 2, and 4 are greater than the MRL for cypermethrin, whereas the concentration of chlorpyrifos in sample 2 and 3 is greater than the MRL for chlorpyrifos. Chlorpyrifos in sample 3 was the greatest concentration recorded in the entire sampling; however, this concentration is within the range between 0.05 mg/kg to 15 mg/kg (ATSDR 1997), the value set by the U.S. Food and Drug Administration (FDA) for agricultural products. The concentrations of folpet α-endosulfan and β-endosulfan in all four samples were less than the MRLs. Fenitrothion was detected in samples but was detected in samples 3 and 4. The concentration of pirimiphos-methyl in all four samples was less than the MRL.

Concentrations of lambda-cyhalothrin were detected at levels less than the MRL for all the samples except sample 1.

CANCER–HUMAN HEALTH RISK ASSESSMENT

Human health risk assessment was performed on the results obtained from the analysis using Human Health Evaluation computerised software-RISC 4.02 (USEPA 1989b). The estimated lifetime cancer risk for adult and child consumers of tomatoes from the study area are as follows: for CTE and RME: 4.5E-04 and 3.7E-03 for chronic, 3.7E-03 and1.9E-03 for subchronic, and 1.5E-04 and 3.7E-04 for acute for chlorpyrifos (Table 3). This implies that 5 and 4 (for chronic) out of 1000, 4 and 2 (for subchronic) out of 1000 adult consumers are at increased risk to suffer from cancer-related cases in the case of chlorpyrifos, whereas for acute 2 and 4 out of 10,000 consumers are at increased risk to suffer from cancer-related cases.

In the case of children, CTE and RME estimated cancer risks are as follows: 7.7E-04 and 1.9E-03 for chronic (*i.e.*, 8 and 2 out of 10,000 and 1000 consumers are at increased risk to suffer from cancer, respectively), 3.9E-04 and 9.7E-04 for subchronic (*i.e.*, 4 and 10 out of 10,000 consumers are at increased risk to suffer from cancer, respectively) and 7.7E-05 and 1.9E-04 for acute (*i.e.*, 8 and 2 out of 100,000 and 10,000 consumers are at increased risk to suffer from cancer).

Table 3. Carcinogenic risk data.

			Chronic		Subchronic		Acute	
Sample code	Exposure media	Exposure route	CTE	RME	CTE	RME	CTE	RME
Cypermethrin	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Chlorpyrifos	Vegetable	Oral-adult-child	4.5E − 04	3.7E − 03	7.5E − 04	1.9E − 03	1.5E − 04	3.7E − 04
			7.7E − 04	1.9E − 03	3.9E − 04	9.7E − 04	7.7E − 05	1.9E − 04
Fenitrothion	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Folpet	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Lambda-cyhalothrin	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Pirimiphos-methyl	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Endosulfan	Vegetable	Oral-adult-child	4.7E − 08	3.9E − 07	7.8E − 08	2.0E − 07	1.6E − 08	3.9E − 08
			8.0E − 08	2.0E − 07	4.0E − 08	1.0E − 07	8.0E − 09	2.0E − 08

Table 4. Hazard index data.

			Chronic		Subchronic		Acute	
Sample code	Exposure media	Exposure route	CTE	RME	CTE	RME	CTE	RME
Cypermethrin	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Chlorpyrifos	Vegetable	Oral-adult-child	3.3E + 00	8.3E + 00	3.3E + 00	8.3E + 00	3.3E + 00	8.3E + 00
			8.6E + 00	2.1E + 01	8.6E + 00	2.1E + 01	8.6E + 00	2.1E + 01
Fenitrothion	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Folpet	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Lambda-cyhalothrin	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Pirimiphos-methyl	Vegetable	Oral-adult-child	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
			0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00	0.0E + 00
Endosulfan	Vegetable	Oral-adult-child	1.0E − 02	2.5E − 02	1.0E − 02	2.5E − 02	1.0E − 02	2.5E − 02
			2.6E − 02	6.5E − 02	2.6E − 02	6.5E − 02	2.6E − 02	6.5E − 02

The estimated lifetime cancer risk for adult and child consumers of tomatoes from the study area are as follows: for CTE and RME: 4.7E-08 and 3.9E-07 for chronic, 7.8E-08 and 2.0E-07 for subchronic, and 1.6E-08 and 3.9E-08 for acute for Endosulfan (Table 3). This implies that 5 and 4 (for chronic) out of 100,000,000, and 10,000,000, respectively, whereas 4 and 2 (for subchronic) out of 100,000,000, and 10,000,000 (respectively) adult consumers are at increased risk to suffer from cancer-related cases in the case of endosulfan. For acute 2 and 4 out of 100,000,000 consumers are at increased risk to suffer from cancer-related cases.

In the case of children, CTE and RME estimated cancer risks are as follows: 8.0E-08 and 2.0E-07 for chronic (*i.e.*, 8 and 2 out of 100,000,000 and 10,000,000 consumers are at increased risk to suffer from cancer, respectively), 4.0E-08 and 1.0E-07 for subchronic (*i.e.*, 4 and 1 out of 100,000,000 and 10,000,000 consumers are at increased risk to suffer from cancer, respectively), and 8.0E-09 and 2.0E-08 for acute (*i.e.*, 8 and 2 out of 1,000,000,000 and 100,000,000 consumers are at increased risk to suffer from cancer).

The other pesticides, cypermethrin, fenitrothion, folpet, lambda-cyhalothrin, and pirimiphos-methyl from the risk assessment studies did not pose any cancer risk (USEPA 1999).

NON-CANCER–HUMAN HEALTH RISK ASSESSMENT

The respective hazard indices for chronic, subchronic, and acute oral exposure of adult and children consumers for CTE and RME are as follows: 3.3E + 00 and 8.3E + 00 for chlorpyrifos for adults, respectively, while 8.6E + 00 and 2.1E + 01 for children, respectively (Table 4). The respective hazard indices for chronic, subchronic, and acute oral exposure of adults and children for CTE and RME are as follows: 1.0E-02 and 2.5E-02 for endosulfan for adults, respectively, while 2.6E-02 and 6.5E-02 for children, respectively.

The hazard index for chlorpyrifos was greater than 1, which is an indication of contamination by chlorpyrifos. However, for all the other pesticides, the hazard indices were less than 1, which means there is no hazard risk for consumers of the tomatoes (USEPA 1995).

CONCLUSION

Results indicate that consumers of tomatoes produced in Ghana, as well as those imported from Burkina Faso, are exposed to pesticide residues. Although these residues were detected in very small amounts does not mean that their presence in the fruits should just be ignored. These pesticides have the potential to affect human health and therefore we should be concerned and address the issue appropriately.

The detection of both α- and β-endosulfan in several samples shows the misuse of pesticides in Ghana. The information of misuse in Ghana is scarce, indicating a need for further monitoring to indicate exactly what pesticides are being used and in what quantities by Ghana's farmers.

The human health risk assessment indicates that chlorpyrifos and endosulfan are a real challenge for tomato production in Ghana and effort should be made to educate both farmers and consumers on their health implications.

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