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Original article

Determination of pesticides residue content in watermelon fruit from Ghana

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Summary

Introduction – **Though watermelon has several benefits, pesticide residues resulting from pesticide application in recent times, appear to have overshadowed the good side of watermelon. The study looked at pesticide residue levels of watermelon grown in Ghana.** *Materials and methods* – **Pesticides in watermelon from seven locations in Ghana (Nsadwir, Ayensudo, Cape Coast, Sekondi-Takoradi, Kumasi, Accra and Bolgatanga) were analyzed. A total of 200 watermelon fruits were collected. Pesticides were extracted from 10.0 g of the fruit using a multi-residue extracting method for non-fatty food crops (Essumang *et al.*, 2013). Clean-up was done using a 10 mm chromatographic column with a 1:1 solvent mixture of cyclohexane and dichloromethane, and the samples were analyzed using Varian CP – 3800 series equipped with the ^{63}Ni selective Electron capture detector (ECD) and a Pulsed Flame-Photometric Detector (PFPD).** *Results and discussion* – **Fifty-six percent of the samples were found to contain pesticide residue contents above the MRLs. Of these, 39% were organochlorine (OC) and 17% being organophosphate (OP). The levels of the organophosphate (OP) pesticides in the watermelon fruit ranged from 0.9 to 4,383.2 µg kg^{-1} whereas that of the organochlorine (OC) ranged between 0.7–34.5 µg kg^{-1} as well as the synthetic pyrethroids pesticides in the range of 0.1–6.4 µg kg^{-1}.** *Conclusion* – **In general, some of the pesticides residues in the watermelon fruit had mean levels higher than the maximum residue limits (MRLs) set by the WHO/FAO. With these high levels, it is likely that consumers may also suffer some accumulation with its attendant health effects due to some of these pesticides.**

In view of the high pesticides (OC and OP) levels determined, the Ghana Standards Authority and the Ministry of Food and Agriculture must collaborate to probe for other pesticide options that would boost profits of watermelon growers and as well reduce the health and environmental risks frequently associated with pesticides application. The Ghana Standards Authority must again ensure strict application of laws that regulate pesticides influx into the country.

Keywords
Citrullus lanatus, limited maximum residues – LMR, organophosphorus, organochlorine, pyrethroid, human health

Significance of this study

What is already known on this subject?

- Pesticide applications have resulted in environmental and food contaminations. Pesticides are absorbed or adsorbed by the crops and transferred to the human body when consumed. Pesticides are linked to numbers of negative health issues such as cancer.

What are the new findings?

- In most parts of Ghana where watermelon is produced or consumed, the mean levels of organophosphate and organochlorine pesticides exceeded the maximum residue limits set by the WHO/FAO.

What is the expected impact on horticulture?

- Our results offer the opportunity for all types of stakeholders, producers, consumers, extensionists, and policy makers to be aware of the pesticide misuse among watermelon farmers in Ghana, and to change the practices so that the fruits produced in the country are of the highest quality and safe for human consumption.

Résumé

Détermination de la teneur en résidus de pesticides dans les pastèques du Ghana.

Introduction – **Les applications de pesticides sur les cultures de pastèque semblent avoir éclipsé les bénéfices nutritionnels de ce fruit. Cette étude a porté sur l'analyse des concentrations de résidus de pesticides trouvés dans les pastèques produites en différents sites au Ghana.** *Matériel et méthodes* – **Les pesticides dans les fruits de la pastèque ont été analysés sur 200 échantillons provenant de sept sites au Ghana (Nsadwir, Ayensudo, Côte du Cap, Sekondi-Takoradi, Kumasi, Accra et Bolgatanga). Les pesticides ont été extraits à partir de 10,0 g de fruit et les échantillons traités ont été analysés par chromatographie en phase gazeuse couplé au détecteur à capture d'électrons (ECD) et au détecteur photométrique à flamme pulsée (PFPD).** *Résultats et discussion* – **Cinquante-six pour cent des échantillons présentaient des résidus de pesticides au-dessus des limites maximales de résidus (LMR). Parmi ceux-ci, 39% étaient des pesticides organochlorés (OC) et 17% des organophosphates (OP). Les niveaux de pesticides OP dans les pastèques variaient entre 0,9 à 4383,2 µg kg^{-1} alors que ceux des pesticides**

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OC variaient entre 0,7 et 34,5 µg kg^{-1} et ceux des pyréthroïdes synthétiques se situaient entre 0,1 et 6,4 µg kg^{-1}. *Conclusion* **– Certains résidus de ces pesticides dans les pastèques présentaient des niveaux moyens supérieurs à la LMR fixée par l'OMS/FAO. Avec des niveaux aussi élevés, il est probable que les consommateurs subissent également une certaine accumulation de ces pesticides avec des risques sanitaires associés. La Ghana Standards Authority devrait utiliser ces résultats pour assurer une application stricte des lois qui réglementent les pesticides dans le pays.**

Mots-clés
Citrullus lanatus, limites maximales de résidus – LMR, organophosphoré, organochloré, pyréthrinoïdes, sécurité sanitaire des aliments

Introduction

Watermelon is a vegetable plant cultivated for its fruit and falls within the same family of cucumbers, pumpkins, squash and gourds. It is a member of the *Cucurbitaceae* family, genus *Citrullus* name *Citrullus lanatus* (Dane and Liu, 2007). They are cultivated in rows of approximately twelve feet apart and its cultivation necessitates appropriate combination of water, weather conditions and care. Approximately 60 days following planting, the fruits nearest the root, the "crown set" are produced. Inside the next 30 days crown set can be harvested with other fruits down the vine, which mature after the crown set (Asare, 2011).

Watermelons have been found to contain no fat, no cholesterol or saturated fat and provide about 80 calories per serve (Sundia, 2007). Even though watermelons are approximately 95% water, they have loads of vitamins. For instance vitamin A, B6, C, and supplementary nutrients critical for excellent health are present in watermelon (Inuwa and Aina, 2011). The vitamin A increases numbers of lymphocytes that help fight infections, improve the immune system, and support good health (Zold, 2008). The Vitamin B6 assists in the improvement of serotonin, dopamine and melatonin neurotransmitters that help the body deal with anxiety (Inuwa and Aina, 2011). The vitamin C prevents infections and helps retard the aging process and cataracts development. The vitamin C helps in reinforcing blood vessels, bones and restoring injured tissues (Zold, 2008). Potassium which alleviates muscle pains occur in small amounts in watermelon (Asare, 2011). Small quantities of calcium and iron are also contained in watermelons (Asare, 2011).

Watermelons again contain lycopene, a potent antioxidant that has been found to fight and prevent diseases (Adorini, 2005). The lycopene was thought to be present only in tomatoes but recently it has been found to occur in watermelons at higher levels than any other vegetables or fruits (Adorini, 2005). Watermelons have 9.09 mg lycopene compared with 4 mg found in one cup of tomato juice (Asare, 2011). Pills and capsules forms of vitamins contain 5–10 mg lycopene which is the prescribed daily amount. Thus, consuming a plateful of watermelon daily would provide about the same health benefits as ingesting over-the-counter vitamins (Asare, 2011).

Lycopene also helps fight cardiovascular diseases by preventing the hardening of the arteries (Adorini, 2005). Watermelon diet helps control acid-base balance, lowers cholesterol levels, increases the amounts of water in urine and helps eliminate excess water from the body, helps clean the kidneys and prevents bladder and kidney stones development (Sundia, 2007).

Thus, watermelon is undoubtedly a fruit with enormous health benefits. However, this essential fruit is prone to insects such as aphids, cabbage loppers, cucumber beetles, cutworms, thrips, leaf miners and spider mites infestations during cultivation. However, the infestations can be managed with pesticides or by biological means. Lady beetles and lacewings, food bran and molasses are some of the biological methods that can be utilized to deal with pest infestations (Bell *et al*., 2001).

Diseases as such Alternaria, leaf blight, anthracnose, bacterial rind necrosis, bacterial wilt, gummy stem blight, downy, mildew, cercospora leaf spot, fusarium wilt, powdery mildew, pythium, southern blight and verticillium wilt often attack watermelon crops during cultivation (Bell *et al*., 2001). Viruses such as watermelon mosaic virus-2, tobacco ring spot virus, papaya ring spot virus, squash mosaic virus, cucumber mosaic virus and zucchini yellow mosaic virus also affect watermelon production (Bell *et al*., 2001). Weed management is vital to successful watermelon production. During production, annual grasses, perennial grasses and broadleaf weeds emerge throughout the cultivation season and must be controlled to ensure maximum yield (Bell *et al*., 2001).

Efforts to manage diseases, control pests and weeds, and to maximize crop yield have resulted in the application of harmful substances which impinge on human health, animals, and the environment. Thus, humans are continually exposed to varieties of lethal chemicals (Eskenazi and Marks, 2006). Humans are exposed to these lethal substances even while in the womb of the mother. Studies have shown occurrence of pesticides in human milk, water, cow's milk and other dairy products (Aguilar and McLachlan, 2006). Contamination by lethal substances is a widespread problem and depends on the environmental conditions, weather conditions, educational and socio-economic disparities in individual situation (Aguilar and McLachlan, 2006).

Pesticides are not a problem of just the developing world. Industrialized countries still utilize large amounts of pesticides and these form the basis of many health and environmental problems. Every country requires reforms to eradicate the impairments originating from pesticide exposure. Exposure to pesticides is a serious quandary in the developing world, where Africa and for that matter, Ghana is no exception. Loads of active components in pesticide are known or suspected carcinogens. Individual pesticides have been found by laboratory investigations or epidemiological studies to cancers such as multiple myeloma, soft tissue sarcoma, Ewing's sarcoma, lymphoma, non-Hodgkin's lymphoma, leukaemia, melanoma, neuroblastoma, germ-cell tumours, retinoblastoma (eye tumour), cancer of the oesophagus, stomach, prostate, testis, breast, ovary, cervix, bladder, thyroid, lung, brain, kidney, pancreas, liver, colon and rectum (Rahbar *et al*., 2002).

Pesticide exposure has also been associated with impaired development of the nervous system which can cause reduced intelligence and behavioural abnormalities (Rahbar *et al*., 2002). There are data that link pesticides to a range of effects on the central nervous system, the peripheral nervous system and the pre-birth developing brain. These effects include increased aggressive behaviour in children, intense depression effects that may lead to suicides, postponed neu-

ropathy which involves corrosion of the peripheral nerves in the limbs with muscular pains and influenza-like symptoms, behavioural change, impaired attentiveness and memory disorder, worsened sense of smell, neuromuscular deficits, Parkinson's disease and parkinsonism, a disorder with symptoms similar to Parkinson's disease, but may be reversible among other effects (Rahbar *et al*., 2002). In view of these effects, it is clear that the population of Ghana where there is no apposite monitoring of the kind(s) of pesticides employed for diverse functions are at risk from these chemicals.

One worrying effect of the problem has to do with the fact that the people who handle the pesticides lack knowledge of their nature, composition and proper application of such pesticides. This has led to scenarios with pesticide misapplication. The problem occurs because most developing countries have very weak or non-existing pesticide control (Essumang *et al*., 2013).

This study investigated the contents of various pesticide residues in watermelon fruits sampled in monitored farms and on city markets in Ghana. The data were compared with the safety levels recommended by the WHO/FAO (Zakia, 2015) and noted as the maximum residue levels (MRLs). Fruit consumption pattern in Ghana has recently shifted towards watermelon due to intense education on its numerous health benefits such as the ability to lower blood cholesterol levels, combat cardiovascular diseases and prevent cancers (Kpodo *et al*., 2015). It is thus essential to show that fruits promoted for health benefits are healthy for customers and the environment.

Materials and methods

Equipment

The gas chromatographic (GC) instrument employed for the pesticide residue analysis was a Varian CP–3800 series equipped with the ^{63}Ni selective Electron capture detector (ECD) for the analysis of organochlorines (OCs) and synthetic pyrethroids pesticides and a Pulsed Flame-Photometric Detector (PFPD) for organophosphorus (OPs) pesticides and capable of temperature programming. The GC column employed was a GS-Q (30 m × 0.53 mm i.d.) supplied by J&W Scientific Inc., Folsom, California, USA. Also employed were a high speed Binatone domestic blender (Model BGL-401), Stuart Scientific Flask Shaker (Model SF1), a Sartorius weighing balance (Model LE632P) with a weighing capacity of 620 g. Other equipment employed include: a rotary vacuum evaporator, Buchi type (Model R-114), a Buchner funnel fitted with a Buchi B-169 vacuum system, a water bath (Model B-840) and 1 L separating funnel (Silber Brand).

Reagents

Chemicals/reagents used were of analytical quality and were supplied by BDH Chemical Ltd. in the United Kingdom. The chemicals/reagents used include: anhydrous sodium sulphate, dichloromethane, acetone, hexane, cyclohexane and florisil.

Sampling and sample preparation

Watermelon fruit samples were randomly collected from seven different locations (Figure 1). These locations were Nsadwir and Ayensudo in the Komenda-Edina-Eguafo-Abrem (KEEA) municipality in the Central Region of Ghana where 60–70% of Ghana's watermelon production is grown (from monitored farms), Takoradi (Western), Kumasi (Ashanti), Cape Coast (Central), Bolgatanga (Upper East), and Accra (Greater Accra) where watermelon harvests have the largest market (several market places). Each sampling site was divided into five sections and watermelon fruits were collected from each site daily for a period of 90 days.

A total of 200 watermelon fruits were randomly collected, packed in a brown paper cardboard box and were transported to the laboratory. Watermelon fruits were sampled from various farms 3–4 weeks after pesticide application. For randomizing the fruit sampling an imaginary W was designed across both the fields and the market places. The fruits were collected along the four arms of the W at 2 m apart (Essumang *et al*., 2013). Thirty percent (n = 60) of the watermelon fruits were collected from the monitored farms whilst 70% (n = 140) were collected from the monitored markets.

Extraction of pesticides and determination

The watermelon fruit samples were divided into flesh and rind (2 cm from rind) (Essumang *et al*., 2013). Each was homogenized separately employing a high speed Binatone domestic blender (Model BLG-401) preceding sample extraction. The extraction protocol for multi-residue pesticides in non-fatty food crops was utilized with acetone as the extracting solvent (Essumang *et al*., 2013).

A 10-g portion of the homogenized watermelon fruit sample was weighed into a 500-mL flat bottom flask and 100 mL acetone was added. The mixture was then fitted onto a Stuart Scientific Flask Shaker (Model SF1) and shaken for 72 h. After the 72 h, the mixture was filtered using a Buchner funnel fitted with a BUHCI B-169 vacuum system through Whatman No. 1 filter paper. The filtrate was quantitatively transferred into an acetone washed 1-L separating funnel for partitioning. In partitioning the filtrate, 40-mL portion of 1:1 mixture of n-hexane and Dichloromethane was added to the filtrate. The funnel was corked and shaken vigorously for 3 min. Occasionally, the tap of the separating funnel was opened to expel pressure. The funnel was fitted onto a retort stand and its content allowed separate for 1 h (Essumang *et al*., 2013). The partitioning was repeated five times using 40 mL each of the 1:1 n-hexane-dichloromethane mixture. The organic layer was drained into an acetone washed 500 mL flat bottom flask. A 30-g portion of anhydrous sodium sulphate dried at 105 °C for 24 h (cooled in a dessicator) was added to the partitioned extract and set aside to stand for 48 h to eliminate traces of water. The partitioned and dried extract was filtered again using a Buchner funnel fitted with a Buchi B-169 vacuum system. The filtrate was transferred quantitatively into another acetone washed 500 mL Buchi round bottom flask, fitted onto a vacuum rotary evaporator and evaporated to dryness. The procedure was repeated for all other watermelon fruit samples (Essumang *et al*., 2013). Each extract concentrate was dissolved in 5 mL of 1:1 mixture of n-hexane and dichloromethane for a clean-up.

Procedure for a clean-up

Preceding the gas chromatographic (GC) analysis, the sample clean-up was done to eliminate co-extractives or unrelated materials from the extracts (Essumang *et al*., 2013). A 10-mm chromatographic column was filled with 3 g Florisil material activated at 130 °C for 24 h. This was topped up with 3 g anhydrous sodium sulphate. A 20-mL portion of 1:1 solvent mixture of cyclohexane and dichloromethane mixture was added onto the column. The tap of the column was then opened to permit the solvent mixture through to wet and rinse the column.

The pesticide residue in 5 mL of 1:1 solvent mixture of n-hexane and dichloromethane was quantitatively transferred onto the column and the extract vial was rinsed thrice with the 1:1 n-hexane-dichloromethane mixture and added onto the column. The column was then eluted with a 100-mL portion of 1:1 cyclohexane and dichloromethane mixture at a rate of about 1 mL min^{-1} into a round bottom flask. The eluent was then evaporated to almost dryness using the vacuum rotary evaporator. The residue was then dissolved in 1 mL solvent mixture of 1:1 acetone and cyclohexane for gas chromatograph (GC) analysis.

Spiked sample preparation

Spiked samples for the watermelon fruits were prepared from 40% (n=80) of the 200 fresh watermelon fruits sampled by adding standard pesticide spiking solution to 10.0 g of the fruit. The samples were spiked with a stock solution of the pesticides containing 1 µgm L^{-1} of the pesticides in ethyl acetate. The spiked samples were prepared at the time of extracting the pesticide residues and were subjected to the same extraction and clean-up procedure as described for the fruit. Samples were spiked with 0.1 mL of the standard as a way to establish the effectiveness of the extraction protocol. The proportion recovered was then evaluated by dividing the spike sample result by the expected result as follows:
Expected spike result = sample result + standard concentration

$$\text{Percent recovery} = \frac{\text{spike result}}{(\text{expected result} \times \text{spike volume})} \times 100$$

Results and discussion

Quality control

Recuperation of different pesticides at 0.10 µg kg^{-1} fortification from watermelon fruit samples with florisil clean-up column is shown in Tables 1–3. The achieved recuperations were within the range of 79.0–86.0% already reported (Essumang *et al.*, 2013). Watermelon fruits from Nsadwir, Ayensudo, Cape Coast, Sekondi-Takoradi, Kumasi, Accra and Bolgatanga were analyzed for organophosphorus, organochlorine and synthetic pyrethroids pesticides. The mean concentration of each pesticide was compared to the WHO/FAO maximum residue levels (MRLs) (Essumang *et al.*, 2013).

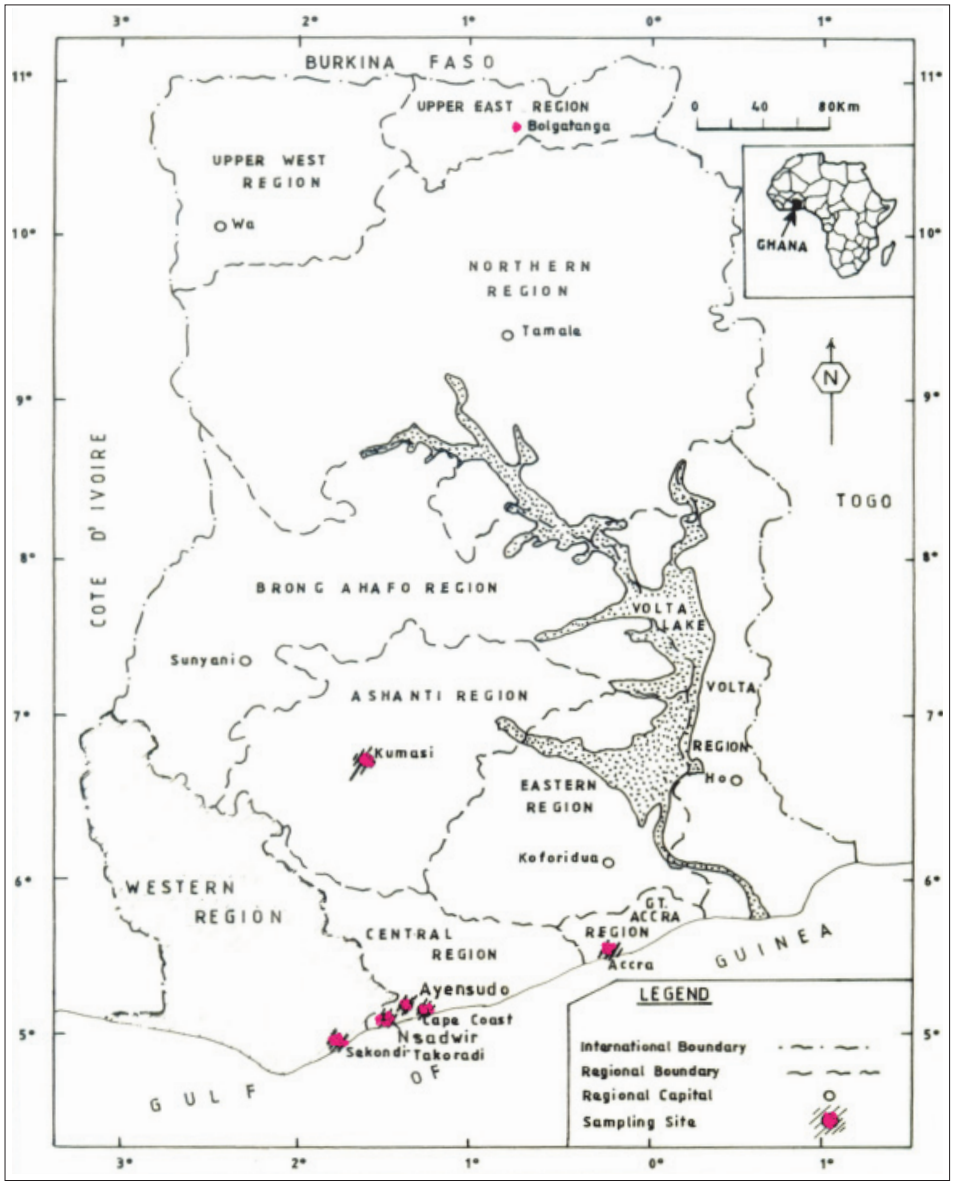

FIGURE 1. Sampling sites of watermelon fruit in Ghana.

Organophosphate pesticide residues

The mean levels of organophosphate pesticides residues in the watermelon fruits are shown in (Table 4). Pirimipphos mean residue levels across all sampling sites ranged between 3.4–19.1 µg kg^{-1} and were below the WHO/FAO acceptable level of 30.0 µg kg^{-1} (Zakia, 2015). The mean levels were also below the 83.6 µg kg^{-1} and 34.3 µg kg^{-1} detected in tomato and pepper fruits respectively (Mohammed, 2006). The mean residue levels of Fonofos, Chlorfenvinp and Profenofos ranged between 5.5–17.5 µg kg^{-1}, 5.5–10.4 µg kg^{-1} and 6.5–26.5 µg kg^{-1} respectively. Samples from Kumasi market had mean residue levels of 17.5 µg kg^{-1} and 26.5 µg kg^{-1} for Fonofos and Profenofos respectively. These mean residue levels were above the 10.0 µg kg^{-1} maximum residue levels recommended by WHO/FAO. Except Pirimipphos, Fonofos, Chlorfenvinp and Profenofos, the mean residue levels of Methamedophos, Enthoprophos, Diazinon, Phorate and Dimethoate were above the WHO/FAO recommended maximum residue levels of 4.0 µg kg^{-1}, 0.4 µg kg^{-1}, 2.0 µg kg^{-1}, 0.7 µg kg^{-1} and 2.0 µg kg^{-1}, respectively. Their mean residual levels ranged from 7.5–32.2 µg kg^{-1} for Methamedophos, 3.9–21.8 µg kg^{-1} for Enthoprophos, 3.5–16.0 µg kg^{-1} for Diazinon, 3.5–9.9 µg kg^{-1} for Phorate and 21.2–89.9 µg kg^{-1} for Dimethoate.

The Enthoprophos mean residue levels across all sample sites exceeded the 0.22 µg kg^{-1} in garden eggs (Raihanah *et al*., 2015). Phorate mean residue levels as well exceeded the 0.32 µg kg^{-1} detected in garden eggs for analogous study (Asiedu, 2013). The mean residue levels of Pirimipphos which ranged from 3.4–19.1 µg kg^{-1} were above the 8.3 µg kg^{-1} and 34.3 µg kg^{-1} detected in pepper and tomato fruits respectively (Mohammed, 2006; Crentsil *et al*., 2011; Drouillet *et al*., 2011). The mean levels of Chlorpyrifos were exceedingly high in all samples.

The mean levels of Chlorpyrifos ranged from 176–3,049 µg kg^{-1}. These mean residue levels were generally above the 678.3 µg kg^{-1} and 878.8 µg kg^{-1} detected in pepper and tomatoes respectively (Mohammed, 2006). The mean levels of Chlorpyrifos were 160–30, 490% above the WHO/FAO recommended maximum residue levels (Zakia, 2015). The mean levels of Chlorpyrifos also exceeded the 0.01 µg kg^{-1} detected in some fruits and vegetables (Thai Agricultural Standard, 2008) and the 0.25 µg kg^{-1} in garden eggs (Raihanah *et al*., 2015). Fenitrothion, Malathion and Chlorfenvinp mean residue levels ranged from 5.4–81.6 µg kg^{-1}, 8.8–40.1 µg kg^{-1} and 5.5–10.4 µg kg^{-1} respectively. These exceeded the maximum residue levels of 10.0 µg kg^{-1} for Fenitrothion, 3.0 µg kg^{-1} for Malathion and 10.0 µg kg^{-1} for Chlorfenvinp recommended by WHO/FAO.

Generally, organophosphate pesticides in the watermelon fruits followed the order Nsadwir > Ayensudo > Cape Coast > Accra > Sekondi-Takoradi > Kumasi > Bolgatanga. The elevated mean residue levels of the organophosphate pesticides

Table 1. Recovery study of various organophosphorus pesticides at 0.1 µg kg^{-1} fortification at 95% confidence interval. [Sample replication=3, replication number 40% (n=80)].

Pesticides	Mean (%)	Standard deviation	Standard error	Standard error %
Methamedophos	83.50	0.008	8.94E-04	1.07E-03
Enthoprophos	81.50	0.184	2.05E-02	2.50E-02
Phorate	84.00	0.090	1.00E-02	1.19E-02
Diazinon	84.50	0.006	6.70E-04	7.92E--04
Dimethoate	80.50	0.010	1.11E-03	1.37E-03
Pirimipphos	86.50	0.010	1.11E-03	1.28E-03
Chlorpyrifos	83.50	0.011	1.22E-03	1.46E-03
Fenitrothion	80.50	0.007	7.82E-02	9.71E-02
Parathion	84.00	0.007	7.82E-02	9.30E-02
Fonofos	79.50	0.007	7.82E-02	9.83E-02
Profenofos	79.00	0.023	2.57E-03	3.25E-03
Malathion	79.00	0.010	1.11E-03	1.40E-03
Chlorfenvinp	81.00	0.006	6.70E-04	8.27E-04

Table 2. Recovery study of various organochlorine pesticides at 0.1 µg kg^{-1} fortification at 95% confidence interval [Sample replication = 3, replication number 40% (n = 80)].

Pesticides	Mean (%)	Standard deviation	Standard error	Standard error %
Lindane	82.00	0.009	1.00E-03	1.21E-03
Heptachlor	78.00	0.009	1.00E-03	1.28E-03
Aldrin	79.00	0.006	6.70E-04	8.48E-04
Endosulfan	77.00	0.010	1.11 E-03	1.44E-03
DDE	82.00	0.009	1.00E-03	1.21E-03
Dieldrin	76.50	0.009	1.00E-03	1.30E-03
DDD	87.50	0.008	8.94E-04	1.02E-03
DDT	82.00	0.008	8.94E-04	1.09E-03
Methoxychlor	78.00	0.010	1.11E-04	1.42E-04
Endrin	80.50	0.006	6.70E-04	8.32E-04

Table 3. Recovery study of various synthetic pyrethroid pesticides at 0.10 µg kg^{-1} fortification at 95% confidence interval. [Sample replication = 3, replication number 40% (n = 80)].

Pesticides	Mean (%)	Standard deviation	Standard error	Standard error %
Bifenthrin	83.50	0.009	1.00E-03	1.19E-03
Lambda-Cyhalothrin	86.50	0.007	7.82E-04	9.04E-04
Permethrin	83.00	0.006	6.70E-04	8.07E-04
Cyfluthrin	83.50	0.009	1.00E-03	1.19E-03
Cypermethrin	82.50	0.004	4.47E-04	5.41E-04
Fenvalerate	82.50	0.008	8.94E-04	1.08E-03
Deltamethrin	86.50	0.007	7.82E-04	9.04E-04

in majority of the fruits may have originated from pesticide applications few weeks or months before fruits were harvested (Essumang *et al*., 2013) and at the time of harvest, no significant pesticide losses due to washing and degradation might have occurred (Essumang *et al*., 2013). The high mean residue levels of the pesticides could also be attributed to the frequency application (Essumang *et al*., 2013) since it could contribute to the high pesticide levels in the fruits through accumulation.

Organochlorine pesticide residues

The mean residue levels of organochlorine pesticides in the watermelon fruits ranged from 3.7–8.2 µg kg^{-1} in Nsadwir (Table 5). The mean residue levels of Heptachlor, Aldrin, Dieldrin, Endosulfan and Endrin were above the WHO/FAO maximum residue levels. The mean residue levels of Heptachlor, Aldrin, Dieldrin, Endosulfan and Endrin ranged from 7.7–9.4 µg kg^{-1}, 1.1–9.1 µg kg^{-1}, 3.0–7.1 µg kg^{-1}, 8.0–28.6 µg kg^{-1} and 2.0–13.9 µg kg^{-1} respectively. These high mean levels could be attributed to the failure of crops to breakdown the pesticides fast enough, thus resulting in an accumulation in the fruit (Essumang *et al*., 2013). Lindane, DDT, DDE, DDD and Methachlor had mean residue levels which ranged from 4.7–9.9 µg kg^{-1}, 2.9–11.1 µg kg^{-1}, 3.7–6.1 µg kg^{-1}, 2.6–7.3 µg kg^{-1} and 5.6–15.4 µg kg^{-1} respectively. Except Lindane, DDD and Methoxychlor, the other had mean residue levels above the WHO/FAO permitted MRLs (Table 5). The mean residue levels of Lindane were below the 100.0 µg kg^{-1}, 10.0 µg kg^{-1}, 133.0 µg kg^{-1}, 129.0 µg kg^{-1}, 40.0 µg kg^{-1}, 100.0 µg kg^{-1} and 19.0 µg kg^{-1} in papaya, mango, pineapple, tomato, lettuce, cabbage and onion respectively (Mohammed, 2006). However, the mean residue levels of Lindane were comparable with the 4.0 µg kg^{-1}, 9.0 µg kg^{-1} and 8.0 µg kg^{-1} in watermelon, pear and okro respectively (Mohammed, 2006; Crentsil *et al*., 2011; Drouillet *et al*., 2011; Asiedu, 2013; Tuan, 2014). The mean residue levels of Methoxychlor in the watermelon fruit samples compared favourably with the 6.0 µg kg^{-1}, 8.0 µg kg^{-1}, 4.0 µg kg^{-1}, 3.0 µg kg^{-1} and 6.0 µg kg^{-1} in papaya, banana, pineapple, mango and lettuce respectively (Mohammed, 2006; Crentsil *et al*., 2011; Drouillet *et al*., 2011; Asiedu, 2013; Tuan, 2014).

The Ayensudo samples had mean residue levels of Lindane, Endosulfan, DDE, DDD, DDT and Methoxychlor below the WHO/FAO permitted MRLs. These low mean values could be due to the fact that these pesticides are no longer used since they are banned and the residues could have originated from previous application of these pesticides on the cropping sites and due to their persistent nature some levels still remain in the soil for the fruit crop to uptake (Essumang *et al*., 2013).

The mean residue levels of Heptachlor, Aldrin, Dieldrin and Endrin in the Ayensudo samples exceeded the WHO/FAO MRLs (Table 5). These high mean levels could have originated due to the frequency of pesticide applications as farmers reported they apply pesticides to the crops every 10–14 days from germination till fruits were harvested. The frequency of the pesticides application could have resulted in elevated

Table 4. Concentrations of various organophosphorus pesticides (in µg kg^{-1}) in watermelon fruits sampled in several cropping sites (A = Nsadwir; E&F = Ayensudo; K = Cape Coast; G = Sekondi-Takoradi; I = Kumasi; H = Accra; J = Bolgatanga). Mean values ± SD [Sample replication = 3, replication number 70% (n = 140)].

Pesticides*	A	E&F	G	H	I	J	K	WHO levels
Meth	4.9±22.0	13.8±3.0	7.5±0.9	16.5±1.0	13.3±3.0	15.3±1.0	32.2±27.0	4.0
Enth	3.9±1.9	6.0±2.3	6.0±1.4	8.2±0.9	21.8±10.0	7.9±1.7	12.0±2.0	0.4
Phor	9.3±5.0	5.8±4.2	8.5±4.7	3.5±0.9	8.6±5.5	6.8±6.4	9.9±3.6	0.7
Diaz	5.7±3.7	7.3±2.3	7.2±0.2	3.8±2.7	13.4±13.0	6.8±2.6	16.6±6.7	2.0
Dim	21.9±19.0	26.4±13.0	89.9±74.0	84.4±71.0	61.6±13.0	37.5±10.0	34.8±7.0	2.0
Pir	8.7±7.8	3.4±3.0	6.3±6.2	9.3±1.9	19.1±15.4	16.4±2.1	16.8±5.4	30.0
Chp	3,049.0±243.7	1,936.0±55.0	1,405.0±26.0	314.0±267.0	207.0±126.0	249.0±26.0	176.0±94.0	10.0
Feni	81.6±45.6	8.0±3.5	13.4±12.0	5.5±0.8	11.2±4.3	5.4±0.3	14.8±3.0	10.0
Para	8.0±5.0	7.6±4.4	7.8±2.0	5.2±1.4	7.8±4.1	5.9±0.2	9.3±2.6	3.0
Fono	5.6±1.9	6.0±3.3	9.2±5.1	5.5±0.8	17.5±10.0	7.7±0.4	9.9±0.3	10.0
Prof	6.5±4.1	9.6±5.6	8.9±8.0	6.7±0.5	26.5±19.0	8.5±5.9	7.1±1.1	10.0
Mala	12.1±10.7	10.2±6.7	8.8±0.7	30.1±16.0	40.1±15.0	15.7±3.0	13.1±5.0	3.0
Chlf	7.7±5.5	6.9±3.8	5.6±2.5	6.2±0.1	5.5±0.3	10.4±2.0	5.9±0.4	10.0

* Meth: Methamedophos, Enth: Enthoprophos, Phor: Phorate, Diaz: Diazinon, Dim: Dimethoate, Pir: Pirimipphos, Chp: Chlorpyrifos, Feni: Fenitrothion, Para: Parathion, Fono: Fonofos, Prof: Profenofos, Mala: Malathion, Chlf: Chlorfenvinp.

Table 5. Concentrations of various organochlorine pesticides (in μg kg^{-1}) in watermelon fruits sampled in several cropping sites (A = Nsadwir; E&F = Ayensudo; K = Cape Coast; G = Sekondi-Takoradi; I = Kumasi; H = Accra; J = Bolgatanga). Mean values ± SD [Sample replication = 3, replication number 70% (n = 140)].

Pesticides	A	E& F	G	H	I	J	K	WHO levels
Lindane	7.1±1.4	6.8±2.9	4.7±0.8	4.9±0.9	4.8±0.8	8.0±0.5	9.9±0.3	10.0
Heptachlor	8.2±2.4	9.3±6.4	8.7±6.3	9.3±2.9	8.2±5.1	9.4±3.8	7.7±2.5	0.10
Aldrin	5.6±1.4	3.8±3.6	1.1±0.0	7.1±2.5	4.9±3.5	9.1±2.7	1.8±0.9	0.10
Endosulfan	8.0±3.2	9.8±3.5	9.7±2.4	12.3±1.8	11.0±3.5	16.5±4.7	28.6±8.2	10.0
DDE	3.8±2.6	4.4±4.1	4.8±3.3	3.7±0.7	6.1±4.6	3.9±1.6	3.8±1.8	0.10
Dieldrin	4.9±1.6	3.5±3.4	3.5±0.7	4.1±0.2	3.0±0.2	7.1±3.3	5.7±2.1	10.0
DDD	3.7±1.2	5.4±2.8	3.4±3.1	2.6±2.1	7.3±7.0	3.2±1.1	2.6±2.0	10.0
DDT	6.8±2.0	4.7±2.8	2.9±0.8	3.2±2.7	6.7±3.6	11.1±4.5	4.1±3.3	6.0
Methoxychlor	6.4±3.2	11.7±9.0	5.6±4.9	15.4±8.6	8.5±7.4	14.3±1.3	14.6±2.9	50.0
Endrin	7.6±6.0	4.9±2.1	2.1±2.0	2.0±0.1	8.4±6.0	13.9±6.5	5.7±3.7	0.2

residue mean levels in the soil and those levels might have become accessible to the fruit crops for uptake (Essumang *et al*., 2013).

The Sekondi-Takoradi watermelon fruits had mean residue levels of 4.7 μg kg^{-1}, 9.7 μg kg^{-1}, 4.8 μg kg^{-1}, 3.4 μg kg^{-1}, 2.9 μg kg^{-1}, 5.6 μg kg^{-1}, 8.7 μg kg^{-1}, 1.1 μg kg^{-1}, 3.5 μg kg^{-1} and 2.1 μg kg^{-1} for Lindane, Endosulfan, DDE, DDD, DDT, Methoxychlor, Heptachlor, Aldrin, Dieldrin and Endrin respectively. Except DDT, Methoxychlor and Dieldrin, the mean residue levels of the rest of the organochlorine pesticides which were present in the Sekondi-Takoradi watermelon fruits exceeded the WHO/FAO MRLs indicated in Table 5 (Zakia, 2015).

The mean residue levels of Heptachlor, Aldrin, DDE, DDT, DDD, Methoxychlor and Endrin in Ayensudo watermelon fruits were higher than those in watermelon fruits sampled from the Sekondi-Takoradi metropolis (Table 5). Accra watermelon fruits samples had residue mean levels of Lindane, Endosulfan, Dieldrin, Heptachlor, Aldrin to be 4.9 μg kg^{-1}, 12.3 μg kg^{-1}, 4.1 μg kg^{-1}, 9.3 μg kg^{-1} and 7.1 μg kg^{-1} (Table 5) respectively. Methoxychlor, DDD, DDE, DDT and Endrin residue mean residue levels were 15.4 μg kg^{-1}, 2.6 μg kg^{-1}, 3.7 μg kg^{-1}, 3.2 μg kg^{-1} and 2.0 μg kg^{-1} (Table 5) respectively. Of these, mean residue levels of Heptachlor, Aldrin, DDE and Endrin were higher than the WHO/FAO MRLs (Zakia, 2015) (Table 5). These high mean levels might be explained by the ability of the pesticide compounds to resist photolysis, thereby resulting in slower degradation rates, long persistence periods in fruits and soils, and consequent accumulation in the fruit crop (Essumang *et al*., 2013).

The lower mean residue levels of DDT and its metabolite DDD might have been due to their persistence in the soil years after DDT has been banned (Essumang *et al*., 2013). Although mean residue levels of organochlorine pesticides were generally high, some of the fruits had mean residue levels lower than the maximum residue levels permitted by some countries. For example, Lindane had mean residue level lower than the 20.0 μg kg^{-1} permitted by Australia and 10.0 μg kg^{-1} permitted by Russia, Japan and the European Union (Chiu *et al*., 2015). Except the Bolgatanga samples, the mean residue levels of Endrin were below the 10.0 μg kg^{-1} maximum residue levels permitted by Russia, the European Union, Japan and Australia (Chiu *et al*., 2015). The mean residue levels of Aldrin were also below the 20.0 μg kg^{-1} and 30.0 μg kg^{-1} maximum residue levels permitted by Japan and Russia respectively (Chiu *et al*., 2015). The mean residue levels of Aldrin were again below the 10.0 μg kg^{-1} maximum residue levels permitted by the European Union and Australia (Chiu *et al*., 2015). Dieldrin had mean residue levels below the 20.0 μg kg^{-1} maximum residue levels permitted by Russia and the 10.0 μg kg^{-1} maximum residue levels permitted by the European Union, Japan and Australia (Chiu *et al*., 2015).

The Kumasi samples had mean residue levels of Lindane, DDE, Methoxychlor, DDD and DDT below the WHO/FAO MRLs (Table 5). Though these mean residue levels were low, the presence of these molecules in the fruit indicates that pesticides are a contamination problem. However, the mean residue levels of Heptachlor, Aldrin, Endosulfan, Dieldrin and Endrin were above the WHO/FAO maximum residue levels (Table 5). Generally, the high mean residue levels of the organochlorine pesticides indicate possible human and environmental health risk (Essumang *et al*., 2013).

Synthetic pyrethroid residues

The mean residue levels of the synthetic pyrethroid residues in watermelon fruits from the different sample sites were below the WHO/FAO (Zakia, 2015) recommended maximum residue limits (MRLs) (Table 6). These results could be due to rapid metabolization of the pesticide compounds in the crop and/or, by the ability of the watermelon crop to extensively dilute the residues through considerable uptake of water by its roots (Thai Agricultural Standard, 2008). The application of the pesticides at lower concentrations than recommended by manufacturers might have as well contributed to the lower levels of the pyrethroid (Essumang *et al*., 2013). The mean residue levels of the synthetic pyrethroid were also below the MRLs permitted by some countries. Bifenthrin mean residue levels in samples were below the 10.0 μg kg^{-1} maximum residue levels permitted by Russian Hygiene Authority (RHA) and the Thailand Agricultural Standard (TAS) (Lam *et al*., 2003; Chiu *et al*., 2015). Deltamethrin, Fenvalerate and Cypermethrin levels were below the 10.0 μg kg^{-1} maximum residue level permitted by the RHA (Chiu *et al*., 2015). The mean residue levels of the synthetic pyrethroid residues in watermelon fruits from the sample sites were also below the mean levels detected in similar studies. For example, the mean residue levels of permethrin across the sample sites in the range of 1.7–2.8 μg kg^{-1} were below the 6.0 μg kg^{-1} and 90.0 μg kg^{-1} in Pear and Lettuce respectively (Mohammed, 2006). Cypermethrin mean residue levels fell in the range of 0.8–3.5 μg kg^{-1} and were below the 44.0 μg kg^{-1} and 60.0 μg kg^{-1} detected in pineapple and watermelon respectively in a similar study (Mohammed, 2006).

The results from the sampling sites showed that watermelon farmers in Ghana apply similar or same types of pesticides on their crops. For example, Methamedophos occurred in all samples at mean residue levels of 3.2–10.1 µg kg^{-1} for Nsadwir, 6.8–17.2 µg kg^{-1} for Ayensudo, 6.9–8.2 µg kg^{-1} for Sekondi-Takoradi, 15.2–17.8 µg kg^{-1} for Accra, 10.9–15.7 µg kg^{-1} for Kumasi, 14.2–16.5 µg kg^{-1} for Bolgatanga and 12.5–51.9 µg kg^{-1} for Cape Coast. A comparison of the mean residue levels of pesticides in the fruits showed that generally, the mean residue levels of organophosphate and organochlorine pesticides were above the WHO/FAO MRLs. This observation gives course for concern as watermelon fruits appeared to have been contaminated with organophosphate (OP) and organochlorine (OC) pesticides at levels that are expected to be perilous to humans and the environment (Zakia, 2015).

The presence of pesticide residues in the watermelon fruits across all the sampling sites confirms an obvious case of contamination of the fruits by pesticides. The use of uncalibrated and faulty spraying apparatus with enlarged nozzles, the application of pesticides too close to the harvesting periods, and the application of pesticides at high levels could all have contributed to the contaminations monitored in watermelon. A worrying scenario of gross misapplication of some pesticides was also observed since the pesticides determined in this study are registered for use on cotton plants and for cocoa sacks, and as pet swabs (Zakia, 2015). A check from the Ghana Standards Authority (GSA) revealed that some of the pesticides which were detected in this study are not approved for use on watermelon crop, but have been registered for use on cocoa trees instead. The GSA recommends Ridomil (metaloxyl), Maxim (fludioxonil), Busan (1-methyl-3, 5, 7-triaza-1-azoniatricyclodecane chloride), Alanap (naptalam) and Gavel (ozamide) as the approved pesticides for watermelon crop. This indicates a large-scale misuse of pesticides among the farmers in Ghana with particular reference to application on watermelon. Since pesticides are manufactured according to the morphology of the crops for which they are developed, it is a failure of the farmers not to request for expert recommendation, and from the extension services not assisting the farmers accordingly.

Conclusion

This study was to investigate the mean levels of pesticide residues in watermelon fruits from monitored farms and markets in Ghana. The results showed that three classes of pesticides, *viz*. the organophosphate, organochlorine and the synthetic pyrethroid pesticides occurred in the watermelon fruits. Seven different synthetic pyrethroid were present in the watermelon fruits across the sampling site (Table 6). However, their mean residue levels were below the WHO/FAO (Zakia, 2015) recommended maximum residue levels and they represented 44% (n = 88) of the total samples. Though the synthetic pyrethroid pesticides do not appear to possess any significant human and environmental health risks, their occurrence in fresh fruit is of grave concern, and efforts must be made to ensure consumer's health is not compromised. Fifty six percent of the samples (n = 112) were found to contain pesticide residue contents above the WHO/FAO MRLs. Of these, 39% (n = 78) were organochlorine (OC) and 17% (n = 34) being organophosphate (OP). The mean residue levels of the organophosphate pesticides were very high as compared to the organochlorine pesticides. Thus Ghana's watermelons are contaminated with significant levels of pesticides and the order of contamination was organophosphate pesticides > organochlorine pesticides > synthetic pyrethroid pesticides. The high mean levels of the pesticides coupled with their synergic characters suggest that humans and the environment are at risk of the pesticides. Fruit consumption pattern in Ghana has recently shifted towards watermelon due to intense education on its numerous health benefits such as the ability to lower blood cholesterol levels, combat cardiovascular diseases and prevent cancers (Kpodo *et al*., 2015). Therefore, it is essential that fruits promoted for health benefits are healthy for customers. The Ghana Standards Authority and the Ministry of Food and Agriculture must ensure strict implementation of laws that regulate pesticides influx into the country whilst ensuring that requisite education is given to farmers on the recommended pesticides for the watermelon crops, the period that pesticides must be applied, and the need to seek expert advice in order to reduce the effects of the pesticides on the watermelon crops, consumers and the environment.

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Table 6. Concentrations of various synthetic pyrethroids (in µg kg^{-1}) in watermelon fruits sampled in several cropping sites (A = Nsadwir; E&F = Ayensudo; G = Sekondi-Takoradi; H = Accra; I = Kumasi; J = Bolgatanga; K = Cape Coast). Mean values ± SD [Sample replication = 3, replication number 70% (n = 140)].

Pesticides*	A	E&F	G	H	I	J	K	WHO levels
Bifen	3.8±0.1	0.9±0.1	1.5±0.1	2.1±0.1	1.7±1.0	2.4±0.0	1.2±0.1	20.0
Lamb	5.3±1.0	1.4±0.1	2.2±1.0	2.4±1.0	2.4±0.1	1.5±0.0	1.6±0.1	20.0
Perm	2.8±1.0	2.0±0.1	2.0±0.1	2.3±0.1	3.3±1.0	2.3±1.0	1.7±0.0	50.0
Cyflu	4.5±10	2.3±0.1	1.2±0.1	2.0±1.0	2.5±0.1	2.6±0.0	1.8±0.0	20.0
Cyper	3.5±0.1	1.6±0.1	1.2±1.0	1.3±1.0	0.8±0.1	1.7±0.0	1.7±0.0	50.0
Fenv	3.2±1.0	1.7±0.1	2.1±0.1	1.8±0.1	1.5±0.1	3.0±0.0	2.0±0.0	20.0
Deltam	3.3±0.1	1.4±0.1	1.4±0.1	2.1±0.1	1.7±0.1	0.9±0.1	1.0±0.0	10.0

* Bifen: bifenthrin, Lamb: Lambda-Cyhalothrin, Perm: Permethrin, Cyflu: Cyfluthrin, Cyper: Cypermethrin, Fenv: Fenvalerate, Deltam: Deltamethrin.

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