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Synthetic Pyrethroids Pesticide Residues in Soils and Drinking Water Sources from Cocoa Farms in Ghana

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Abstract

The contamination of pesticides in 32 soils and 64 drinking water samples was investigated from cocoa farms in the Dormaa West District of Ghana to assess pollution status. A total of nine synthetic pyrethroids pesticides were measured with a high resolution Varian CP-3800 Gas Chromatograph equipped with ^{63}Ni electron capture detector (ECD). Eight synthetic pyrethroid residues namely fenvalerate, deltamethrin, cypermethrin, bifenthrin, permethrin, lambda-cyhalothrin, allethrin and cyfluthrin were detected with lambda-cyhalothrin and allethrin occurring most frequently in soil and water respectively. The concentrations of synthetic pyrethroids residues in the soil samples were in the ranges of; 0.02-0.03 mg/kg for lambda-cyhalothrin, 0.010-0.02 mg/kg for allethrin, 0.010-0.04 mg/kg for cyfluthrin, <0.01-0.04 mg/kg for cypermethrin, 0.02-0.06 mg/kg for deltamethrin, and <0.01-0.03 mg/kg for bifenthrin. Similarly, the synthetic pyrethroids residues in the water samples were in the ranges of; 0.01-0.05 μg/L for allethrin, 0.01-0.04 μg/L for fenvalerate, 0.01-0.04 μg/L for cypermethrin and 0.01-0.05 μg/L for deltamethrin. The concentrations of synthetic pyrethroids pesticide residues recorded in the soil samples analysed were generally below and within their respective US MRLs for agricultural soils, except the mean concentration values recorded for pesticides such as lambda-cyhalothrin at Diabaa (S2) and Krakrom (S3), allethrin at Diabaa (S2) and deltamethrin at Kwakuanya (S4), which were above their respective US MRLs for agricultural soils. The trends of synthetic pyrethroids pesticide residues in the water samples analysed from the various distances to cocoa farms decreased with an increase of water source to cocoa farm (ranking; 0-15m>16-30m>above 30m). All synthetic pyrethroids pesticide residues recorded in the water samples were below and within their respective WHO MRLs for drinking water except for deltamethrin, which exceeded the WHO MRL at Kwakuanya (S4) at distance 0-15m from a cocoa farm. The presence of synthetic pyrethroids residues in the soil and water samples analysed is an indication of the use of the pesticide by cocoa farmers in the study area. The routine monitoring of pesticide residues in the study area is necessary for the control and reduction of environmental pollution.

Keywords: environment, health, monitoring, pollution, toxicity, water quality

1. Introduction

Cocoa (*Theobroma cacao*) is the most dominant tree crop and the major agricultural export commodity in Ghana. The cocoa sector employs over 800,000 smallholder farmers and contributes about 70-100% of their annual household incomes (Appiah, 2004; Anim-Kwapong & Frimpong, 2004; Danso-Abbeam, Setsoafia, Gershon, & Ansah, 2014). The export of cocoa beans has been one of the major foreign exchange earners for the Ghanaian economy. In 2013, cocoa contributed 16.48% (US$ 2,267.3 million) of the total agricultural foreign exchange earnings for the country (Institute of Statistical Social and Economic Research [ISSER], 2014). Unfortunately, cocoa production in Ghana has over the years faced major challenges including the incidence of insect pests and diseases, which have been recognized as a major cause of declining yields in cocoa (Ayenor, Huis, Obeng-Ofori, Padi, & Röling, 2007; Ntiamoah & Afrane, 2008). In order to increase cocoa production, there has been an increase in the use of pesticides in general, particularly synthetic pyrethroids.

Despite the positive roles pesticides play in keeping pest and disease below their economic injury levels, their use have been associated with unintended environmental and human health consequences (Dankyi, Gordon, Carboo, & Fomsgaard, 2014; Owombo, Idumah, & Afolayan, 2014). In most developing countries, studies show that these consequences have often been severe due to misuse and overuse of pesticides with disregards to safety measures and effective regulations on chemical use (Fianko, Donkor, Lowor, & Yeboah, 2011; Dankyi et al., 2014). Soil contamination with pesticide residues is consequently a critical environmental problem as it pose threat to soil microflora and microfauna as well as their ecosystem (Bentum, Essumang, & Dodoo, 2006; Braschi, Gessa, & Blasioli, 2011). The chemical residues can pollute surface water as well as groundwater making them unsafe for drinking. These pesticides in the soil can enter the food web through plants and pose health risk to humans as they bio-accumulate and can be transferred along the food chain (Ortiz-Hernández, Sánchez-Salinas, Olvera-Velona, & Folch-Mallol, 2011). In recent years, several human acute and chronic illnesses have been associated with low doses of pesticide exposure (Mostafalou & Abdollahi, 2012; Gill & Garg, 2014). For instance, exposure to pesticide residues through food and drinking water were reported to, cause low sperm count in males, birth defects, increase testicular cancer and reproductive malfunction/deformities (Sosan, Akingbohungbe, Ojo, & Durosinmi, 2008; Tanner et al., 2011; Leena, Choudhary, & Singh, 2012), endocrine disruptions, immunotoxicity, and neurobehavioral and developmental disorders (Mesnage, Clair, de Vendômois, & Seralini, 2010; Cocco et al., 2013; Gill & Garg, 2014). In addition, studies in Ghana have also shown that significant amount of applied pesticides ends up in cocoa growing soils and cocoa beans (Botchway, 2000; Bentum et al., 2006; Apau & Dodoo, 2010; Agyen, 2011; Daanu, 2011; Boakye, 2012; Frimpong, Yeboah, Fletcher, Pwamang, & Adomako, 2012a; Frimpong, Yeboah, Fletcher, Adomako, & Pwamang, 2012b; Frimponget al., 2012c;Frimpong, Yeboah, Fletcher, Pwamang, & Adomako, 2012d; Dankyi et al., 2014), sediments and water (Ntow, 2001; Darko, Akoto, & Oppong, 2008; Kuranchie-Mensah et al., 2012), fruits and vegetables (Bempah & Donkor, 2011; Bempah, Donkor, Yeboah, Dubey, & Osei-Fosu, 2011), meat (Darko & Acquaah, 2007), fish (Darko et al., 2008) and human fluids (Ntow, 2001; Tutu, Yeboah, Golow, Denutsui, & Blankson-Arthur, 2011). Unfortunately, the few available reports on the levels of pesticides contamination of soils and water sources in and around cocoa farms in the cocoa industry in some parts of Ghana, have concentrated much more on organochlorines which are banned with little information on the residual levels of synthetic pyrethroids in soils and particularly in drinking water sources although these are approved for agricultural use (Environmental Protection Agency [EPA], 2009).

The Brong Ahafo region is one of the major cocoa producing regions in Ghana. Farmers in this region use pesticides extensively, particularly synthetic pyrethroids to control pests and diseases in order to increase cocoa yields. However, there is little information on synthetic pyrethroids residue levels in soils and drinking water sources (which are hand dug well on farms) from cocoa farms. This paper aims at assessing the levels of synthetic pyrethroids pesticide residues in soils and drinking water sources from selected cocoa farms in the Dormaa West District of the Brong Ahafo Region in Ghana.

2. Materials and Methods

2.1 Study Area

The study was carried out in the Dormaa West District located at the western part of the Brong-Ahafo region of Ghana (Figure 1). The area is a forest ecological zone, which is conducive for agrarian activities. It is one of the major cocoa producing districts in the country. It has a tropical climate with distinct wet and dry season. The dry season is from November to February and the rainy season from May to September. Mean annual rainfall is between 125 and 175 cm with a mean annual temperature of about 28.1 ºC (Ghana Statistical Service, 2014). The major economic activities in the district include the cultivation of food and cash crops (including cocoa), poultry and livestock farming, oil palm extraction, cassava processing and sand winning.

2.2 Sampling Design

Four cocoa growing communities namely Nkrankwanta (S1), Diabaa (S2), Krakrom (S3) and Kwakuanya (S4) were randomly selected from the district (Figure 1). Sixteen (16) cocoa farms (four farms for each community) were identified purposively with (i) distance of farm to the nearest drinking water source (hand dug wells) and (ii) age of cocoa farm (not less than 8 years and not more than 20 years with a history of at least five years of pesticides application) used as determining factors. Soils and water sampling were carried out from December 2014 to February 2015.

2.3 Soil Sampling

Two quadrats of 80 x 80 m were marked in each selected cocoa farm. In each quadrat, five (5) core soil samples were collected randomly at 0-20 cm depth with a soil auger and put together to form a composite sample, which were well mixed and a sub-sample taken. Two soil replicates were collected from each cocoa farm. These gave a total of 32 soil samples from the study area. All soil samples were kept in well-labelled sampling bags and transported to the Ecological Laboratory of the University of Ghana for analysis. The soil samples were air dried at room temperature for 7 days and then oven–dried at 105 ºC to constant weight, and sieved using 2mm mesh. Sub-soil samples were taken and analysed for pesticide residues.

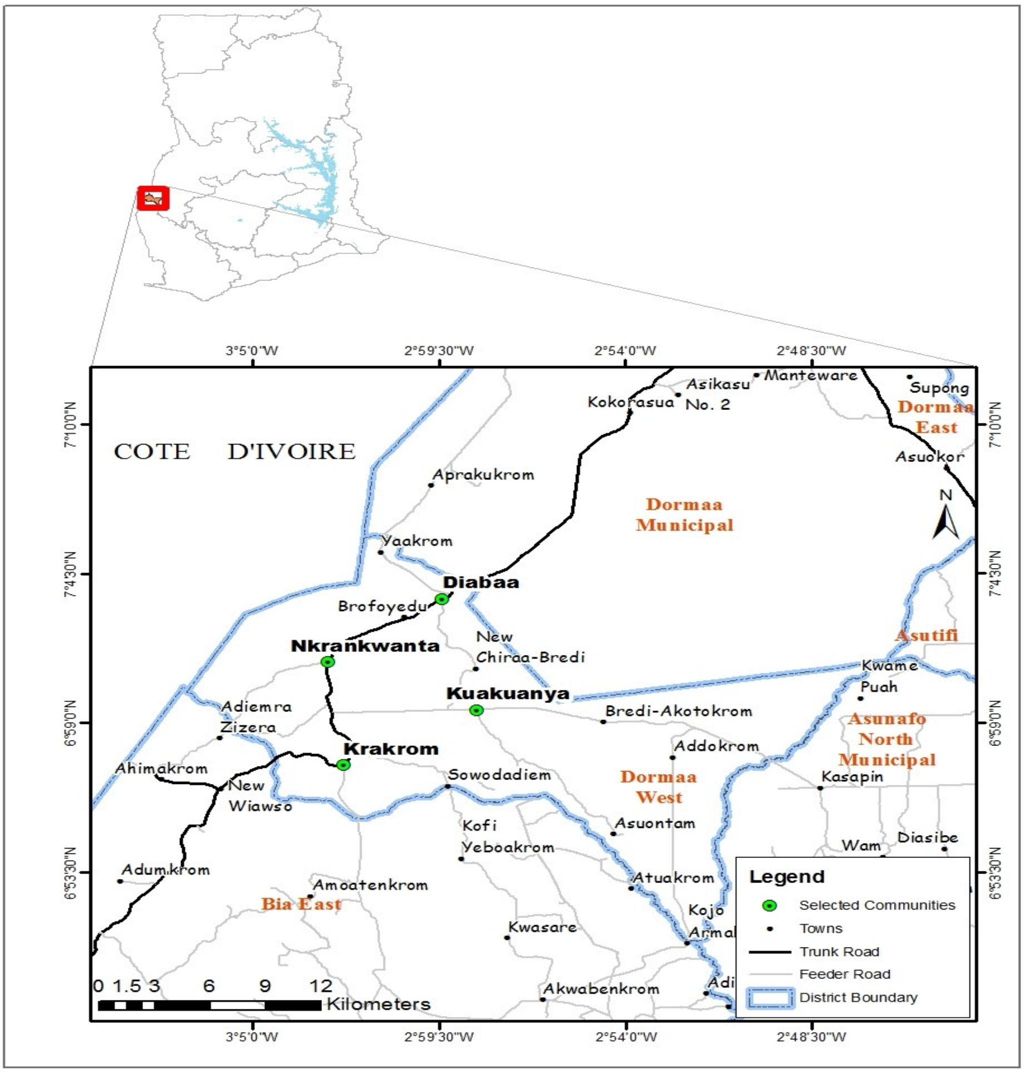

Figure 1. Map of Dormaa West District showing study communities

2.4 Water Sampling and Physico-Chemical Analysis

Water samples were collected from hand dug wells located within and around the selected cocoa farms in the study area. Wells were selected based on distance to cocoa farms in the order; 0-15 m and 16-30 m whiles wells above 30 m were used as the control. These distances were chosen based on the World Health Organization (WHO) recommendations that hand dug wells should be at least 100ft (30 m) away from an agricultural field (Harris, Hoffman, & Mazac, 1996; World Health Organization [WHO], 2004). Water samples were collected from two hand-dug wells for each of the distance categories (between 0-15 m and 16-30 m) within each selected community. Water samples were also collected from three (3) open wells (above 30 m from farms) as control (S5). Three replicates were collected from each well making a total of sixty-four (64) water samples for the study. A water sampler was used to collect water samples into 1.5 L and 500 mL polyethylene sample bottles with stoppers that had been pre-cleaned with tap water and rinsed with distilled water for pesticide residues and physico-chemical analysis respectively. Samples were labelled, stored in ice chest and transported to the Ecological Laboratory of the University of Ghana and Ghana Standards Authority Pesticides Residue Laboratory Accra, for water physico-chemical and pesticide residues analysis respectively. Samples were pre-filtered through 0.45 mL fiber glass filters (WHATMAN) to remove debris and suspended material (Darko et al., 2008).

Water physico-chemical parameters such as pH and total dissolved solids were measured in-situ using a Multi pH parameter probe meter (Model YSI 63) manufactured by YSI incorporated, USA. Turbidity was measured using a turbid meter (Model HACH 2100P) manufactured by HACH company, USA. Total suspended solid (TSS) was determined using HACH direct reading spectrophotometer (Model DR. 2010) manufactured by HACH Company, USA.

2.5Determination of Pesticide Residues in Water and Soils Samples

2.5.1Chemicals and Reagents Used for Analysis

Soil and water samples were analysed for 9 synthetic pyrethroids pesticides (allethrin, bifenthrin, fenpropathrin, λ-cyhalothrin, permethrin, cyfluthrin, cypermethrin, fenvalerate and deltamethrin). The individual certified pesticide reference standards used for the identification and quantification were obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany) with certified purity of 99% and stored in the freezer to minimize degradation. Pesticides grade acetonitrile and dichloromethane were purchased from BDH, England. Silica gel adsorbents were obtained from Phenomenex, USA. In addition, all other reagents and solvents used were of analytical grade purchased from BDH, England and included: anhydrous magnesium sulphate, anhydrous sodium sulphate, acetone, ethyl acetate, and saturated sodium chloride.

2.5.2 Standard Solutions

Stock solutions (1000 µg/mL) of the nine synthetic pyrethroids pesticides standards were prepared by pipetting the appropriate aliquot or weight of the synthetic pyrethroids pesticides into 25 mL volumetric flasks, and then dissolving and diluting to the marks with ethyl acetate with the aid of a vortex mixer (Thermolyne Maxi Mix-Plus). All solutions prepared for Gas Chromatography (GC) were filtered through a 0.45 µm nylon filter.

2.5.3 Extraction of Soil Samples

The extraction of the soil samples was carried out by the method described by Frimpong et al. (2013), with slight modification from the Ghana Standard Authority's (GSA) pesticides residue laboratory protocol. Ten grams (10g) of the soil samples were weighed into 250 mL separating flasks. A 10 mL of acetonitrile was added to each of the samples in the flasks and ultra-sonicated using Becon FS400b for 5 minutes. An additional 10 mL of acetonitrile was added, and the flasks closed tightly. The contents were placed on horizontal mechanical shaker (Ika-Werke HS 501 Digital) and set to shake continuously for 30 minutes at 300 mot/min. The contents were then allowed to stand for 10 minutes to sufficiently separate the phases or layers. A 10 mL of the supernatants were carefully taken by pipette and dried over 2 g anhydrous magnesium sulphate through filter papers into 50 mL round bottom flasks. The concentrates were then adjusted to 2 mL using the rotary film evaporator (Buchi Ratovapor R-210, USA) at 35 ºC, and made ready for silica clean up step.

2.5.4 Extraction of Water Samples

The extraction technique employed for water samples in this work was as described by Afful, Awudza, Osae, & Twumasi (2013), with slight modification from the Ghana Standard Authority's (GSA) pesticides residue laboratory protocol. A 1000 mL portion of the filtered water samples were transferred into 2 L capacity separating flasks. Then 30 mL of saturated sodium chloride solution (NaCl) was added to each sample. The samples were thoroughly mixed by inverting the flask three to four times. A 100 mL of dichloromethane as extraction solvent was then added to each sample and vigorously shaken manually for 2-3 minutes, while releasing the pressure intermittently. The phases were allowed to separate for 5 minutes and the dichloromethane extracts (organic layers) were separated from the aqueous layers. The extraction for each water sample was repeated twice with 100 mL of dichloromethane and the organic layers put together and dried over anhydrous sodium sulphate through filter paper into 50 mL round bottom flasks. The extracts from the water samples were then concentrated on rotary vacuum evaporator (Buchi Ratovapor R-210, USA) to about 1 mL and subjected to silica clean up.

2.5.5 Clean-up of Soil and Water Extracts

The silica gel clean-up processes for the soil and water extracts were carried out using themethods described by Frimpong et al. (2013) and Afful et al. (2013) respectively, with slight modifications from the Ghana Standard Authority (GSA) pesticides residue laboratory protocols. Extracts clean up were done, using polypropylene cartridge columns, packed with one-gram silica gel previously activated for 10 hours in an oven at 130 ºC. For the soil extracts, 1 cm thickness layer of anhydrous magnesium sulphate on top of the silica gel were conditioned with 6 mL acetonitrile while for the water extracts, 2 g layer of anhydrous sodium sulphate on top of the silica gel were conditioned with 6 mL dichloromethane. The concentrated extracts (from soil and water) were then loaded onto the columns/cartridges, and 100 mL round bottom flasks placed under the columns to collect the elutes. A 10 mL acetonitrile and 20 mL dichloromethane were then used to elute the clean-up columns/cartridges for the soil and water clean ups processes respectively. The total filtrates collected (elutes from soils and water) were concentrated to dryness using the rotary evaporator (Buchi Ratovapor R-210, USA) set at 40 ºC. The residues (from the soil and water extracts) were re-dissolved in 1 mL ethyl acetate by pipetting and transferred into 2 mL standard opening vials prior to quantitation by Gas Chromatography (GC) (Varian Association Inc. USA) equipped with Electron Capture Detector (GC-ECD). All extracts were kept frozen until quantification was achieved.

2.5.6 Gas Chromatography Determination

The final extracts were analysed using a Gas Chromatography (GC) - Varian CP-3800 (Varian Association Inc. USA) equipped with ^{63}Ni Electron Capture Detector (ECD) and CombiPAL Auto sampler at the Ghana Standards Authority (GSA) Pesticide Residues Laboratory, Accra. The Gas Chromatography (GC) conditions and the detector response were adjusted to match the relative retention times and response as described by Japanese analytical methods for agricultural chemicals (Syoku-An, 2006). The GC conditions used for the analysis were capillary column (fused silica capillary) coated with VF-5 ms (30 m + 10 m EZ guard column x 0.25 mm internal diameter, 0.25 µm film thickness). The injector and detector-ECD temperatures were set at 270 ºC and 300 ºC respectively. The oven temperature was programmed as follows: 70 ºC held for 2 min, ramp at 25 ºC min^{-1} to 180 ºC, held for 1 min, and finally ramp at 5 ºC min^{-1} to 300 ºC. Nitrogen was used as carrier gas at a constant flow rate of 1.0 mL min^{-1} and detector make-up gas of 29 mL $min^{-1.}$ The injection volume of the GC was 1.0 µL. The total run time for a sample was 31.4 min.

2.5.7 Quantification and Limit of Detection of Pesticides

The residue levels of synthetic pyrethroids pesticides were quantitatively determined by the external standard method using peak area. Measurement was carried out within the linear range of the detector following the procedures described by Frimponget al. (2012c), Frimpong et al. (2012d), Afful et al. (2013) and Frimpong et al. (2013). The peak areas whose retention times coincided with the standards were extrapolated on their corresponding calibration curves to obtain the concentration. The limit of detection of the pesticides determined was based on the extract of the fortified samples that were serially diluted by factor of two to give different concentrations. One out of each concentration that gave a response three times the standard deviation of the least fortified sample was noted. And this was used to estimate the statistical significance of differences between low level analyte responses and the combined uncertainties in both the analyte and the background measurement. The limits of detection for the synthetic pyrethroids pesticides in soils and water samples were 0.010 mg/kg and 0.01 µg/L respectively.

2.5.8 Quality Assurance and Quality Control

All glassware used for analysis were rigorously washed with detergent, rinsed with distilled water and thoroughly rinsed with analytical grade acetone and dried overnight in an oven at 150 ºC. The glassware were then removed from the oven and allowed to cool down and stored in dust-free cabinets. The quality of synthetic pyrethroids pesticide residues was assured through the analysis of solvent blanks, procedural matrix blanks and duplicate samples. All reagents used during the analysis were exposed to same extraction procedures and subsequently run to check for interfering substances. In the blank for each extraction procedure, no pesticide was detected. Samples of each series were analysed in duplicates. Recalibration curves were run with each batch of samples to ensure that the correlation coefficient was kept around $r^2 = 0.99$. The method used was an international method, optimized and validated using various agricultural products (Frimponget al., 2012c). A fortification level at the limit of determination of 0.010 mg/kg and at levels of 0.10 mg/kg, and 0.05 mg/kg for soil samples and fortification level at the limit of determination of 0.01 µg/L and at levels of 0.005 µg/L and 0.05 µg/L for water samples, were chosen before the analysis to evaluate the recovery of the compounds in the soil and water samples respectively. The efficiency of the analytical methods (the extraction and clean-up methods) was determined by recoveries of an internal standard. The recoveries of internal standards ranged between 70% and 100% for all the synthetic pyrethroids pesticides analysed. These recovery values show that the method used was reproducible.

2.6 Data Analysis

Statistical Package for Social Sciences (SPSS) software version 20.0 was used to generate the means for the physico-chemical parameters of water as well as the pesticide residues in soil and water. One-way Analysis of variance (ANOVA) was used to test for the significant differences and similarities between the water physico-chemical properties and pesticide residues in soil and water from the various sampled sites. A Pearson correlation analysis was also carried out to establish the degree of relationship between the physico-chemical parameters of water and the various pesticide residues detected in water samples. The statistical significance tests were carried at 5% confidence level *(p <0.05)*.

3. Results and Discussion

3.1 Physico-chemical Properties of Water Samples from the Study Area

The results of the physico-chemical parameters of water samples from the distance categories to cocoa farms are summarized in Table 1. The mean pH of the water samples ranged from 5.18 to 5.82 with a mean value of 5.51±0.23. There were no significant differences ($p > 0.05$) in pH with respect to distances of water sources to cocoa farms. However, with the exception of water samples from Kwakuanya (S4) (6.93) at a distance 0-15m, all the other sites within the various distances recorded mean pH values below the WHO acceptable limits of 6.5-8.5 recommended for drinking water (WHO, 2004). This indicates slightly acidic condition of water for the study period. The low pH values recorded could be due to the amount of cations present in the soil due to the leaching of basic cations or the presence of high level of organic matter within the soil zones whose oxidation releases carbon dioxide that reacts with water to produce a weak carbonic acid (Langmuir, 1997; Kortatsi, 2007). According to Ansa-Asare, Darko, & Asante (2009), the nature of the geology in an area could also account for low pH values. Additionally, Nkansah, Boadi, & Badu (2010), reported that pH values lower than 6.5 are considered acidic for human consumption and can cause health problems such as acidosis and adverse effects on digestive and lymphatic system. Acidity increases the capacity of the water to also attack geological materials and leach toxic trace metals into the water making it potentially unsafe for human consumption (Kortatsi, 2007).

Total dissolved solids (TDS) are a measure of the total organic and inorganic substances dissolved in water. According to Tay (2007), total dissolved solids can be used as a common indicator for water pollution. The mean total dissolved solids (TDS) for the various distances ranged from 46.3 mg/L to 65.5 mg/L with a mean of 58.7±7.61 mg/L. There were no significant differences ($p > 0.05$) in mean values of TDS among the various distances of water sources to cocoa farms. The TDS measured in the water samples were all below the (WHO, 2004) guideline limit of 1000 mg/L for drinking water, indicating that the groundwater are generally fresh and have no particulate content.

Turbidity in water is the reduction of transparency due to the presence of suspended particles (Fianko et al., 2013). The measured turbidity values ranged from 2.29 NTU to 63.6 NTU with a mean of 35.8±21.9 NTU. There were no significant differences *($p > 0.05$)* in turbidity in relation to distances of water sources to cocoa farms. With the exception of water samples from Diabaa (S2) (3.87 NTU) at a distance 0-15m, Kwakuanya (S4) (1.77 NTU) at a distance 16-30 m and the Control (S5) (2.29 NTU) at a distance above 30m, all the other sites at the various distances recorded turbidity values that far exceeded the WHO permissible limit of 5 NTU for drinking water. The high turbidity values may be due to erosion and run-off from cocoa farms that carried soil particles and deposited them in the wells as most of the wells were not protected. This could also be due to the shallow nature of the wells causing disturbance and re-suspension during water withdrawal from the well. Elevated turbidity is often associated with the possibility of microbiological contamination, as turbidity makes it difficult to disinfect water properly (Fatoki, Muyima, & Lujiza, 2001). Gyamfi et al. (2012) reported that water samples with high turbidity presents colloidal materials, which provides adsorption sites for chemicals and organisms that may be harmful or cause undesirable tastes and odors in water.

Furthermore, total suspended solids (TSS) are solids in water that can be trapped by a filter (Fianko et al., 2013). The mean TSS measured in the water samples within the various distances ranged from 4.00 mg/L to 70.0 mg/L with a mean of 38.0±23.4 mg/L. There were no significant differences *($p > 0.05$)* in TSS in relation to distances of water sources to cocoa farms. Although, TSS in drinking water does not have a WHO health based guideline limit, it is recommended that it should not exceed 500 mg/L in water for drinking (Ewusi, Obiri-Yeboah, Voigt, Asabere, & Bempah, 2013). The mean values of TSS recorded for the entire study period were far below this recommended limit/guideline. The result is line with the findings of Amoako, Karikari & Ansa-Asare (2011) who reported mean TSS values of 10.0 mg/L to 45.0 mg/L. The low values recorded in this study may be as a result of the filtering capacity of the soil.

Table 1. Summary of water physico-chemical characteristics at Dormaa West, Ghana

Distance (m)	0 – 15m		16– 30m		Above 30m		Total means	WHO MRLs
Parameters	Range	Mean± SD	Range	Mean± SD	Range	Mean± SD	Mean ± SD	
pH	5.05-6.93	5.82±0.62	5.34-5.86	5.54±0.17	4.80-5.42	5.18±0.33	5.51±0.23	6.5-8.5
TDS (mg/L)	36.0-110.0	65.5±25.0	37.0-98.0	64.3±20.5	30.0-73.0	46.3±23.3	58.7±7.61	1000
Turbidity (NTU)	3.87-205.0	63.6±23.3	1.77-121.0	41.2±6.21	1.25-2.87	2.29±0.91	35.8±21.9	5
TSS (mg/L)	6.00-277.0	70.0±26.8	3.00-126.0	40.0±5.45	2.00-6.00	4.00±2.08	38.0±23.4	

SD=Standard deviation, TDS=Total dissolved solids, TSS=Total suspended solids

Table 2. Range, mean and standard deviation of synthetic pyrethroids concentrations in (mg/kg) in soil samples from the study area

Sites	Nkrankwanta (S1)		Diabaa (S2)		Krakrom (S3)		Kwakuanya (S4)		Total means	US MRLs
Pesticides	Range	Mean± SD	Range	Mean± SD	Range	Mean± SD	Range	Mean± SD	Mean± SD	
Lambda-cyhalothrin	ND-0.02	0.02±0.00	ND-0.04	0.03±0.01	ND-0.03	0.03	ND-0.04	0.03±0.01	0.03±0.00	0.02
Allethrin	ND	ND	ND-0.02	0.02	ND-0.03	0.02±0.01	ND	ND	0.02±0.00	0.01
Cyfluthrin	ND	ND	ND-0.06	0.04±0.02	ND-0.05	0.04±0.01	ND-0.05	0.03±0.01	0.04±0.00	0.15
Cypermethrin	ND-<0.01	<0.01	ND-0.04	0.04±0.00	ND-0.05	0.03±0.02	ND-0.06	0.04±0.02	0.03±0.01	0.05
Deltamethrin	ND-0.03	0.03±0.00	ND-0.03	0.03	ND-0.04	0.04	ND-0.06	0.06	0.04±0.01	0.05
Bifenthrin	ND-<0.01	<0.01	ND-0.02	0.02	ND-0.03	0.03	ND-0.03	0.03	0.02±0.01	0.04

SD=Standard deviation, ND=non-detected/below detection level, Limit of Detection=0.010 mg/kg

3.2 Pesticide Residue Concentrations in Soil Samples from the Study Area

Table 2 presents the concentrations of synthetic pyrethroids pesticides measured in the soil samples. Recovery ranged between 73% and 100%. The limit of detection (LOD) for synthetic pyrethroids in the soil samples was 0.010 mg/kg. Soil samples analysed showed the presence of six synthetic pyrethroids pesticides namely lambda-cyhalothrin, allethrin, cyfluthrin, deltamethrin, cypermethrin and bifenthrin.

Lambda-cyhalothrin occurred in 50% of the soil samples analysed and ranged from 0.02 mg/kg at Nkrankwanta (S1) to 0.03 mg/kg at Diabaa (S2), Krakrom (S3) and Kwakuanya (S4), with a mean value of 0.03±0.00 mg/kg, which was above the United States (US) Maximum Residue Limit (MRL) of 0.02 mg/kg for agricultural soils.There were no statistically significant site differences ($p > 0.05$) in mean lambda-cyhalothrin recorded in the soil samples analysed. However, with the exception of soil samples from Nkrankwanta (S1) which recorded a mean lambda-cyhalothrin concentration of 0.02 mg/kg, all the other sites recorded mean values above the US MRL of 0.02 for agricultural soils. The concentrations of lambda-cyhalothrin in the soil samples may be attributed to the use of pesticides containing lambda-cyhalothrin as its active ingredient in the study area. Lambda-cyhalothrin has high affinity for soil as reported by Tarus Nyambati, Kituyi and Segor Chebii (2007).

Additionally, allethrin was detected in 25% of the soil samples analysed and ranged from below detection at Nkrankwanta (S1) and Kwakuanya (S4) to 0.02 mg/kg at Diabaa (S2) and Krakrom (S3). The mean allethrin concentration recorded for the entire study was 0.02±0.00 mg/kg, which was higher than the US MRL of 0.01 mg/kg set for agricultural soil. In addition, sites S2 and S3 with detectable residues recorded values that were above the US MRL of 0.01 mg/kg for agricultural soils. There were however, no significant differences ($p > 0.05$) in mean allethrin recorded in the soil samples among the various sampled sites. The occurrence of allethrin suggests the use of the pesticide in cocoa production in the study area. The use of this pesticide, however, could be attributed to the fact that allethrin is registered for use in Ghana for agricultural purposes (EPA 2009).

Cyfluthrin occurred in 43.8% of the soil samples and ranged from below detection at S1 to 0.04 mg/kg at S2 and S3, with a mean value of 0.04±0.00 mg/kg. There were no significant differences ($p > 0.05$) in mean cyfluthrin recorded in the soil samples among the various sampled sites. However, the measured means recorded were all far below the US MRL of 0.15 mg/kg for agricultural soils. The detection of cyfluthrin in the soil samples suggests its use in cocoa production in the study area.

Cypermethrin was also detected in 50% of the soil samples analysed and ranged from <0.01 mg/kg at S1 to 0.04 mg/kg at S2 and S4 with a mean value of 0.03±0.01 mg/kg. However, the measured mean concentrations of cypermethrin observed at all the sampled sites were below the US MRL of 0.05 mg/kg for agricultural soils. Similarly, no significant differences ($p > 0.05$) in mean values of cypermethrin were recorded in the soil samples. The use of cypermethrin in the control of cocoa pests and diseases by farmers in the study area, might have accounted for its detection levels in the soil samples analysed.

In addition, deltamethrin occurred in 37.5% of the soil samples analysed with a mean value of 0.04±0.01 mg/kg, which was below the US MRL of 0.05 mg/kg for agricultural soils. The measured mean concentrations of deltamethrin ranged from 0.03 mg/kg at Nkrankwanta (S1) and Diabaa (S2) to 0.06 mg/kg at Kwakuanya (S4). There were no significant differences ($p > 0.05$) in mean values of deltamethrin recorded in the soil samples among the various sampled sites. With the exception of S4 which recorded a mean deltamethrin value above the US MRL of 0.05 mg/kg for agricultural soils, all the other sampled sites recorded mean values below the US MRL for agricultural soils. The use of pesticides with deltamethrin as its active ingredient by cocoa farmers in the study area might have accounted for its detection in sampled soils.

Bifenthrin which is registered for use in cocoa production by Ghana COCOBOD was detected in 31.3% of the soil samples. The measured bifenthrin ranged from <0.01 mg/kg at S1 to 0.03 mg/kg at S3 and S4 with a mean value of 0.02±0.01 mg/kg. There were no significant differences ($p > 0.05$) in mean values of bifenthrin recorded in the soil samples among the various sites. However, the measured mean concentrations of bifenthrin recorded at all the sampled sites were below the US MRL of 0.04 mg/kg for agricultural soils. The detection of bifenthrin suggests the use of pesticides with bifenthrin as its active ingredient by cocoa farmers in the study area.

3.3 Pesticide Residues in Water Samples from the Various Distance Categories

Table 3 presents results of synthetic pyrethroids pesticides detected in the water samples from the various distance from cocoa farms. The different synthetic pyrethroids pesticides detected at significantly varying concentrations were allethrin, deltamethrin, cypermethrin and fenvalerate. Recovery ranged between 73% and 100%. The limit of detection (LOD) for synthetic pyrethroids in the water samples was 0.01 µg/L.

Allethrin was detected in 26.3% of the water samples analysed. In general, the mean allethrin concentration recorded at distances 0-15m was relatively higher than at distances 16-30m, with distances above 30m (control) recording no concentration. There were no significant differences ($p > 0.05$) in mean allethrin recorded in water with respect to distances of water sources to cocoa farms. The mean concentrations of allethrin observed at the various distances ranged from below detection to 0.05 µg/L with a mean value of 0.05±0.00 µg/L. However, the mean allethrin values recorded at all the sampled sites within the various distances were below the WHO guideline value of 0.05 µg/L for drinking water except sample from Diabaa (S2) at distance 0-15m, which recorded a mean concentration of 0.05 µg/L, which was within the WHO guideline value. On the other hand, the mean concentration of allethrin (0.05±0.00 µg/L) recorded for all the sampled sites was found to be within the WHO guideline value of 0.05 µg/L for drinking water, but higher than the mean value of 0.00069 µg/L reported by Afful et al. (2013) in water samples from the Weija Lake in Ghana. The detection of allethrin in the water samples confirmedthe use of the pesticide by cocoa farmers in cocoa production in the study area, as it was also detected in the soil samples analysed.

Similarly, fenvalerate occurred in 26.3% of the water samples analysed with a mean concentration of 0.05±0.00 µg/L, which was within WHO MRL of 0.05 µg/L set for portable water. The fenvalerate average residue observed at distances 16-30m to the nearest cocoa farms was higher than the mean value recorded at distances 0-15m, with the control sites (S5) (distances above 30m) recording no concentration of fenvalerate. There were no significant differences ($p > 0.05$) in mean values of fenvalerate among the various distances of water sources to cocoa farms. The measured mean concentrations of fenvalerate observed at all sites within the various distances were below the WHO MRL of 0.05 µg/L set for portable water except water sampled from Kwakuanya (S4) at distance 16-30m which recorded a mean concentration of 0.05 µg/L, which was comparable to the WHO MRL for drinking water. The mean concentration of fenvalerate recorded in this work was however, higher than the mean fenvalerate of 0.0086 µg/L reported in a similar study by Afful et al. (2013) in water samples analysed from the Weija Lake in Ghana. The use of pesticides with fenvalerate as its active ingredient by cocoa farmers in the study area might have accounted for their detection in water samples although it was not detected in the soil samples analysed.

Cypermethrin a very popular synthetic pyrethroids in Ghana and also registered for cocoa production (EPA, 2009) was detected in 26.3% of the water samples analysed. Cypermethrin mean concentration recorded at distances 0-15m was higher than the average value recorded at 16-30m to the nearest cocoa farms, with S5 (control) recording no concentration of cypermethrin. There were no significant differences ($p > 0.05$) in cypermethrin mean values recorded in the water samples in relation to distances of water sources to cocoa farms. Cypermethrin concentrations ranged from below detection to 0.04 µg/L with a mean value of 0.03±0.01 µg/L, which was below the WHO MRL of 0.05 µg/L for portable water. On the other hand, the mean concentrations of cypermethrin recorded at all sites with detectable residues, were below the WHO MRLs of 0.05 µg/L for portable water. However, the mean value of cypermethrin (0.03±0.0 µg/L) recorded in this study was higher than the mean concentration of 0.00025 µg/L reported by Afful et al. (2013) in water samples analysed from the Weija Lake in Ghana. The detection of cypermethrin in the water samples confirmed the use of pesticides with cypermethrin as its active ingredient in the control of cocoa pests and diseases by farmers as it was also detected in the soil samples from the study area.

Deltamethrin occurred in 26.6% of the water samples analysed with a mean concentration of 0.05±0.01 µg/L, and ranged from below detection to 0.05 µg/L. The mean concentration of deltamethrin recorded at distances 0-15m was higher than the mean concentration recorded at distances 16-30m. However, samples from the control sites recorded no value for deltamethrin. There were no significant differences ($p > 0.05$) in deltamethrin mean values recorded in relation to distances of water sources to cocoa farms. The mean concentrations of deltamethrin recorded at sampled sites with detectable residues within the various distances were found to be below the WHO MRL of 0.05 µg/L for drinking water except water sampled from Kwakuanya (S4) at distance 0-15m that recorded a mean deltamethrin concentration of 0.07 µg/L, which was above the WHO MRL for drinking water. The mean value of deltamethrin (0.05±0.01 µg/L) recorded in this study was however higher than the mean value of 0.0004 µg/L reported by Afful et al. (2013) in water samples analysed from the Weija Lake in Ghana. The presence of deltamethrin suggests it use in the study area.

Table 3. Range, mean and standard deviation of synthetic pyrethroids in (µg/L) in water samples from the study area with respect to the various distance categories.

Distance (m)	0 – 15m		16– 30m		Above 30m		Total means	WHO MRLs
Pesticides	Range	Mean± SD	Range	Mean± SD	Range	Mean± SD	Mean± SD	
Allethrin	0.04-0.05	0.05±0.01	<0.01-0.04	0.04	ND	ND	0.05±0.00	0.05
Fenvalerate	0.02-0.04	0.03±0.01	0.02-0.05	0.04±0.02	ND	ND	0.04±0.00	0.05
Cypermethrin	0.04-0.04	0.04±0.00	ND-0.02	0.02	ND	ND	0.03±0.01	0.05
Deltamethrin	0.03-0.07	0.05±0.02	ND-0.04	0.04	ND	ND	0.04±0.01	0.05

SD=Standard deviation and ND=non-detected/below detection level, Limit of detection=0.01 µg/L

Table 4. Pearson's correlation between water physico-chemical properties and pesticide residues detected

Pesticides	pH	TDS	TSS	Turbidity
Cypermethrin	0.269	-0.202	0.378	-0.380
Allethrin	-0.321	-0.361	-0.545*	-0.880**
Fenvaerate	-0.614*	0.427	-0.117	-0.388
Deltamethrin	0.980**	0.969**	0.968**	-0.382

* = Correlation is significant at 0.05 level (2-tailed), ** = Correlation is significant at 0.01 level (2-tailed), TDS = Total dissolved solids, TSS = Total suspended solids

3.4 Relationship between Water Physico-Chemical Properties and Pesticides Residue in Water

Relationship between water properties and pesticide residues detected in water samples were analysed using the correlation of Pearson (Table 4). A positive significant correlation was observed between pH and deltamethrin (at $p < 0.01$), TDS and deltamethrin (at $p < 0.01$), and TSS and deltamethrin (at $p < 0.01$). On the other hand, a negative correlation was found between pH and fenvalerate (at $p < 0.05$), turbidity and allethrin (at $p < 0.01$), and TSS and allethrin (at $p < 0.05$). The strong positive correlation between pH, TSS and TDS with deltamethrin indicates that pH, TSS and TDS of water could have enhanced the adsorption of these pesticide compound. Thus, an increase in pH, TSS and TDS resulted in a corresponding increase in the concentrations of deltamethrin. On the other hand, the negative correlation between pH and fenvalerate, turbidity and allethrin, and TSS and allethrin suggests that an increase in pH, turbidity and TSS of water results in a corresponding decrease in fenvalerate and allethrin respectively and vice versa.

4. Conclusion and Recommendations

Eight synthetic pyrethroids namely, fenvalerate, deltamethrin, cypermethrin, bifenthrin, permethrin, lambda-cyhalothrin, allethrin and cyfluthrin were detected in soil and water samples analysed from the study area. Generally, there was high frequency of occurrence for all eight synthetic pyrethroids pesticides detected in the samples (soil and water). This indicates a higher preference for synthetic pyrethroids pesticides among cocoa farmers in the study area and could be attributed to the fact that most of these pesticides are registered for use in Ghana for agricultural purposes (EPA, 2009). The concentrations of synthetic pyrethroids pesticide residues in the soil samples analysed were generally below and within their respective US MRLs for agricultural soils. However, mean concentrations of pesticide such as lambda-cyhalothrin at Diabaa (S2) and Krakrom (S3), allethrin at Diabaa (S2) and deltamethrin at Kwakuanya (S4) were above their respective US MRLs for agricultural soils. Synthetic pyrethroids pesticides may have found their way into the soils via spray drift during cocoa tree treatment, wash-off from treated cocoa tree, improper disposal of left over spray solution, sprayer wash water, pesticide containers and misuse or overuse of pesticides. The presence of synthetic pyrethroids pesticides in the soil samples analysed pose potential hazards to soil organisms and can contaminate surrounding water bodies through surface runoff and leaching. In addition, there is the possibility of translocation of these residues from the contaminated soils into the cocoa fruits (cocoa beans) through the root system, and into other crops like vegetables that are commonly intercropped in cocoa farms thereby posing health risks to consumers. In addition, the trends of synthetic pyrethroids pesticides residues in the water samples analysed from the various distances to cocoa farms followed the decreasing order of ranking; 0-15m>16-30m>above 30m (control). All the pesticide residues detected in the water samples analysed from the various distances were below and within their respective WHO MRLs for drinking water except deltamethrin which exceeded the WHO MRL at Kwakuanya (S4) at distance 0-15m. The presence of synthetic pyrethroids pesticides in the water samples could be traced to drift during pesticides spraying, runoff from treated area and leaching via the soil. The results therefore suggest that synthetic pyrethroids pesticide residue concentrations in the hand dug wells within the various distances for this study may not pose health hazards to cocoa farmers' households and communities who use water from those sources except for water from Kwakuanya (S4) at distance 0-15m.

The routine monitoring of pesticide residues in the study area is necessary for the prevention, control and reduction of environmental pollution, to minimize health risks. Also, it is recommended that domestic water wells should be sited above 30m in relation to distances from cocoa farms to reduce potential pesticide contamination as most of the water samples analysed within distances 0-30m were found to be contaminated with pesticides residues. Farmer education on safe pesticide use should be intensified to limit the levels of pesticides residues in soils and in drinking water sources as poor practices were observed from the study area.

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