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Effects of Organochlorine pesticide residues in Some Beans and Tomatoes Sold at Amai Market

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Abstract

Pesticides are plant-defensive products vastly used in agriculture to augment the yield, improve the quality, and extend the storage life of food crops. However, its indiscriminate use, inner properties, and the likelihood of having effects on non-target organisms have made it a pollutant of concern in the environment. This study examined the effects of pesticide residues in local beans and tomato samples. Samples of beans and tomatoes were purchased randomly from the Amai market in the Ukwani Local government area of Delta State. A total of twenty-five organochlorine pesticide (OCP) residues in these samples were analyzed with a gas chromatography-electron captured detector (ECD). The estimated acceptable daily intake (EADI) and estimated chronic daily intake (ECDI) of aldrin and Dichlorodiphenytetraethene (DDT) concentration in Beans and tomatoes was determined based on European Union (EU) and United States Agency of Toxic Substances and Disease Registry (ATSDR) standards. The study results revealed levels of organochlorine pesticide residues among beans and tomato samples. Dichlorodiphenytetraethene (DDT) had the highest value of 0.0004mg/kg among the OCP residues detected in beans followed by aldrin, with a value of 0.0002mg/kg in tomatoes. The study, thus suggests that OCPs concentrations in beans and tomatoes are unsafe for human consumption. The study therefore calls for incessant monitoring of agricultural farmlands because continuous exposure to pesticide-contaminated food products sold in Amai market (beans, tomatoes) could affect the health of consumers.

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Introduction

Pesticides are plant-defensive products vastly used in agriculture to improve the quality and extend the storage life of food crops (Fernndez-Alba and Garca-Reyes 2008). They provide society with a vast range of benefits, especially in agricultural productivity through the control of diseases (Bowles and Webster, 1995; FDA, 1999). The world market for pesticides has increased substantially from $2.7 billion in the 1970s, to $18.5 billion in the 1990s and $32.7 billion in 2001. The USA pesticide production is 34 percent, followed by China and the European Union countries (France, Germany, UK) (Pimentel, 2009; Lewis *et al.*, 1997; Rosen *et al.*, 1997; Francis *et al.*, 2003; Van Emden and Peakall, 1999, Chapman, 2002).

There has been an increasing concern about dietary ingestion of pesticide residues by adults and children who feed on beans and tomatoes (UNDP 2000). Consumption of beans and tomatoes has increased substantially over time because of their health benefit (Lehotay, 2000; Holland *et al*., 2004; Hamilton *et al*., 2004; Gilden *et al*., 2010). In the United States of America, for example, the consumption of beans and tomatoes doubled from 1990 to 2000 (UNDP, 2000). Xenobiotics are a major cause for concern all over the world, because of the ability of their metabolite residues to remain in the soil after degradation by artificial or natural means and adverse effects on humans and the eco-biota (Chapman, 2002).

In 2001, about 74 percent of agricultural products (beans and tomatoes) in the United States of America (USA) were reported to contain at least one pesticide residue higher than the acceptable level(Kouradsen, 2003). This increase in the level of pesticide residues has risen in developing countries such as Africa, Asia, South and Central America, Eastern Mediterranean (FDA, 1999). The maximum residue levels (MRLs) or tolerances in the United States limit the types and amounts of residues that can be legally present in beans and tomatoes sensitivity of modern GC- and LC-MS(/MS) instruments(Lehotay *et al*., 2007).

In Nigeria, the Federal Environmental Protection Agency FEPA (1991) has established criteria, guidelines, specifications, and standards for pesticide usage. It is, therefore, necessary to conduct regular monitoring of pesticide residues and their stable metabolites to ascertain

if their concentrations meet the prescribed limits as established by the Nigerian government (Atuanya and Onuoha, 2018)
Hence, this research investigated and identified pesticide residues in beans and tomatoes sold at Amai market

Materials and Methods

Study area

The study was carried out in Amai. Amai market is located in the hub of Amai village close to Novena University. Beans and tomatoes are sold for consumption daily. The beans and tomatoes are cultivated and harvested within Amai.

Samples Collection

A random sampling of beans and tomatoes were purchased from the market. Samples were enclosed in a clean blotting paper and wrapped inside a clean, paper envelope. The addition of a small sachet of silica gel to the envelope helped to reduce the moisture content in the enclosed samples. Samples were provided to the analytical laboratory for analysis.

Organochlorine Pesticide Residue Extraction

The persistent organochlorine pesticide residues from beans and tomato samples were analyzed chromatographically with gas chromatography equipped with an electron capture detector(GC/ECD) as described by Papadakis *et al*., (2015).
Thirty grams (30 g) aliquot of well-mixed samples were placed separately into a solvent-rinsed beaker.50:50 mixed with acetone were prepared. One milliliter (1 ml) of decachlorobiphenyl was added and mixed thoroughly using a glass stir rod. 1.5 g of anhydrous sodium sulfate (Na_2SO4) was weighed and added to the sample and mixed thoroughly to form a free-flowing powder. 50ml of the solvent was mixed and added to the samples. Samples were placed in the Sonicator and Sonicated for about 10 – 15 minutes at about 60^0C. The extract was decanted into a round bottom flask. The procedure was repeated once more an additional 50 ml of solvent was mixed, Sonicated, and allowed to settle in the beaker before it was decanted into the round bottom flask. The samples were extracted with a 2 ml concentrated rotary evaporator. 5 ml of hexane was added to the extract. It was then allowed to evaporate to reduce the volume to 2 ml. The final hexane volume was 2 ml, giving final sample weight of 15g/ml.

Sample cleanup

A syringe was used to transfer 2 ml of hexane into a 10 ml vial extract in a fume cupboard. 5ml of the 1:1 Sulfuric acid was carefully added, It was ensured there was no exothermic reaction. The cap of the flask was capped tightly and vortexed for 1 min. The phases were allowed to separate for at least 3 minutes, the upper hexane layer was observed and ensured that it was not highly colored. After the cleanup procedure, each fraction was evaporated with a rotary evaporator and concentrated in a nitrogen stream to 1 ml. The fractions were analyzed for pesticide residues.
The organochlorine residue concentrations were determined using a Hewlett-Packard (H_P) 5890 series II equipped with an electron capture detector with an autosampler. The chromatographic separation was achieved by using an HP-1 of 30 x 0.25 mm internal diameter (ID). The chromatographic temperature program was kept at 100 ^{O}C for 1 min; and increased to 200^{O}C for 2 min. The injection volume was 1 ml. The detector temperature was maintained at 300^{O}C.

Calculation for sample analysis

The concentration of each analyte range in a sample was calculated directly from the instrument using the data analysis software. The final sample weight and the dilution factor was used in the batch file and the final results were generated by the software.

The response factor method was also employed

$$Cf = \frac{Area\ (p) \times Rf \times Vf \times DF \times 1000}{Wi}$$

Where:
Cf = Final Sample concentration (mg/L)
Area (p) = Measured area of peak (peaks)
Wi = Initial weight extracted (g dry weight)
Vf = Final extract volume (mL).
Df = Dilution factor of sample or extract if diluted.
RF= Response factor from the calibration standard calculation

$$RF = \frac{Concentration\ (P)}{Area\ (P)}$$

Concentration (p) = Concentration of peak or Total concentration of range
Area = Area of peak or total across range.

Human health risk assessment of pesticide residues in beans and tomatoes (dietary)

To estimate the carcinogenic and non-carcinogenic risk of detected pesticides to humans, using two population groups (young children and adults) the estimated acceptable daily intake (EADI) was used. EADI was

obtained by multiplying the residual pesticide concentration (ug/kg) in each ready-to-eat vegetable by the consumption rate in Nigeria (L/day or kg/day) and dividing the product by the body weight (kg)(Fianko *et al*., 2011). The hazard quotient (HQ) was then obtained from the ratio of EADI and reference dose. The reference dose (RFD) of each pesticide is the exposure that is likely to be without an appreciable risk or deleterious effects and was provided by the USFDA (1999). The Food and Agricultural Organization (FDA,1999) quotes the per capita consumption of ready-to-eat vegetables in Nigeria as 9 kg. The following formula was used to estimate the dietary intake.

EADI =C x CR/BW(Fianko *et al*. 2011).

EADI is the estimated average daily intake, C is the concentration of pesticide residues, CR represents the consumption rate of beans and tomatoes and BW represents the body weight of the age group. The Food and Agricultural Organization (FDA,1999) quotes the per capita consumption of beans and tomatoes in Nigeria as 9 kg., while body weight was set at 70 kg for the adult population group.

Hazard quotient (HQ): Hazards quotients were obtained by dividing the EADI by their corresponding reference dose (RfD).

Hazard quotient (HQ) =EADI/RfD(Fianko *et al*., 2011).

Hazard index (HI): using the hazard quotient equation above, the hazard index (HI) was obtained. The hazard index is used to assess the risk involved in exposure to mixtures of the detected pesticides belonging to the same chemical group (organochlorines).

Hazard Index(HI)=$\sum_i^n HQl$.(Fianko *et al*., 2011).

Statistical analysis

The data were collated and results were presented in figures, tables, and graphs. The results were expressed as mean ± standard error(S.E). The recorded data were subjected to statistical analysis using Statistical Package for Social Sciences (I BMSPS) Microsoft Excel 2010 (ogbeibu, 2005).

COMPONENT	**Beans**	**Tomatoes**
ALDRIN	0.00±0.00	0.0002±0.00
a – BHC	0.00±0.00	0.00±0.00
b – BHC	0.00±0.00	0.00±0.00
d – BHC	0.00±0.00	0.00±0.00
Gamma - BHC (LINDANE)	0.000±0.00	0.00±0.00
ALPHA – CHLORDANE	0.00±0.00	0.00±0.00
GAMMA – CHLORDANE	0.00±0.00	0.00±0.00
ATRAZINE	0.00±0.00	0.00±0.00
p,p DDD	0.00±0.00	0.00±0.00
p,p DDE	0.00±0.00	0.00±0.00
p,p DDT 4, 4 DDT	0.004±0.00	0.00±0.00
DIEDRIN	0.00±0.00	0.00±0.00
ENDOSULFAN 1	0.00±0.00	0.00±0.00
ENDOSULFAN 11	0.00±0.00	0.00±0.00
ENDOSULFAN SULFATE	0.00±0.00	0.00±0.00
ENDRIN	0.00±0.00	0.00±0.00
ENDRIN ALDEHYDE	0.00±0.00	0.00±0.00
HEPTACLOR	0.00±0.00	0.00±0.00
HEPTACHOR EPOXIDE	0.00±0.00	0.000±0.00
METHOXYCHLOR	0.00±0.00	0.00±0.00
DIAZINON	0.00±0.00	0.00±0.00
PHOSPHORO METHYL GLYCINE	0.00±0.00	0.00±0.00
TCMX	0.00±0.00	0.00±0.00
CARBAMATE	0.00±0.00	0.00±0.00

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DECACHLOROBIPHENYL	0.00±0.00	0.00±0.00
TOTAL OCP (mg/Kg)	0.0004±0.00	0.0002±0.00

Results

Table 1 **: Mean concentration of pesticide residues in beans and Tomatoes**

KEY :OCP; Organochlorine pesticide residue, BDL;Below detectable limit

Table I above shows a total of 25 OCP residues were investigated. They are; aldrin, a-BHC, b-BHC, d-BHC, gamma lindane, alpha-chlordane, gamma chlordane, atrazine, p,p DDD, p,p DDE, p,p DDT 4, 4 DDT, dieldrin, endosulfan 1, endosulfan 11, endosulfan sulfate, endrin, endrin aldehyde, heptachlor, heptachlor epoxide, methoxychlor, diazinon, phosphoro methyl glycine, TCMX, carbamate and decachlorobiphenyl.

In the beans and tomatoes in Amai; out of the 25 OCP residues that were investigated only one (1) OCP residue (DDT) in beans was detected; the remaining OCP residues (aldrin, a-BHC, b-BHC, d-BHC, alpha- chlordane, gamma chlordane, atrazine, p,p DDD, p,p DDE, p,p DDT 4, 4 DDT, dieldrin, endosulfan 1, endosulfan 11, endosulfan sulfate, endrin, endrin aldehyde, heptachlor, heptachlor epoxide, methoxychlor, diazinon, phosphorus methyl glycine, TCMX, carbamate and decachlorobiphenyl) in tomatoes below the detectable limit (BDL) (table 1).

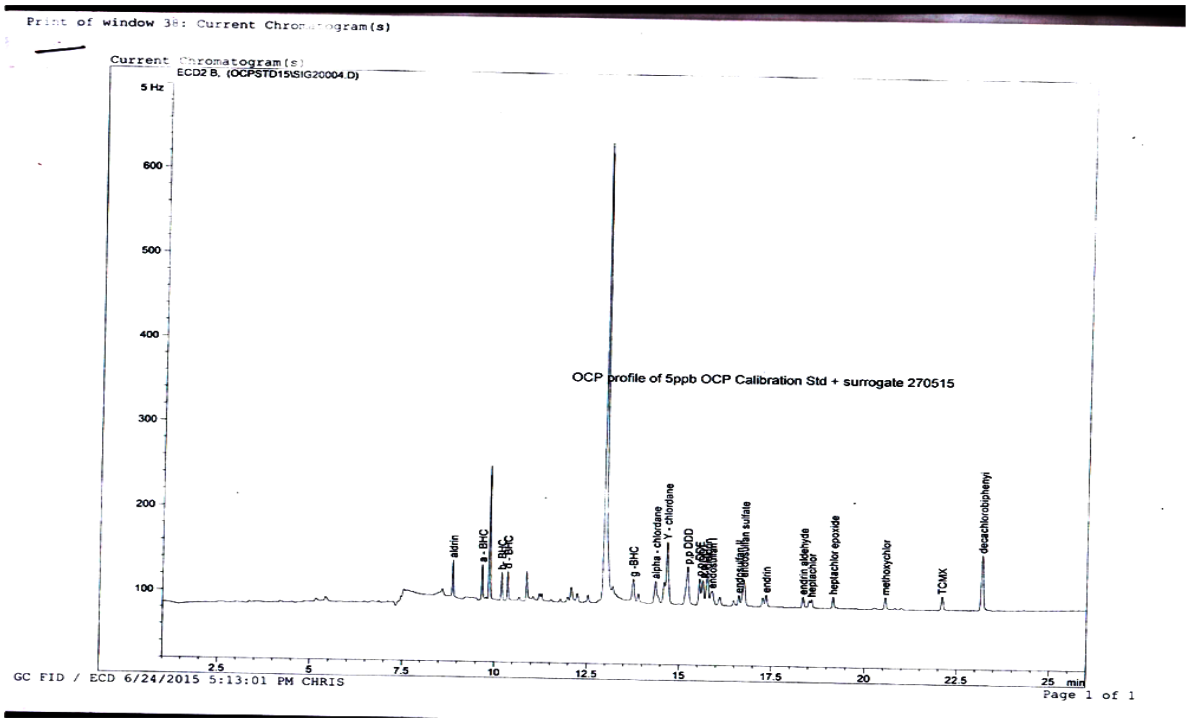

Figure 1: Organochlorine pesticide (OCP) residue profiles in beans and tomatoes

The beans and tomatoes samples in Amai that were investigated, revealed that there were no interference peaks obtained for the blank sample chromatogram at the same retention time as the target compounds in the chromatogram in (Figure I).

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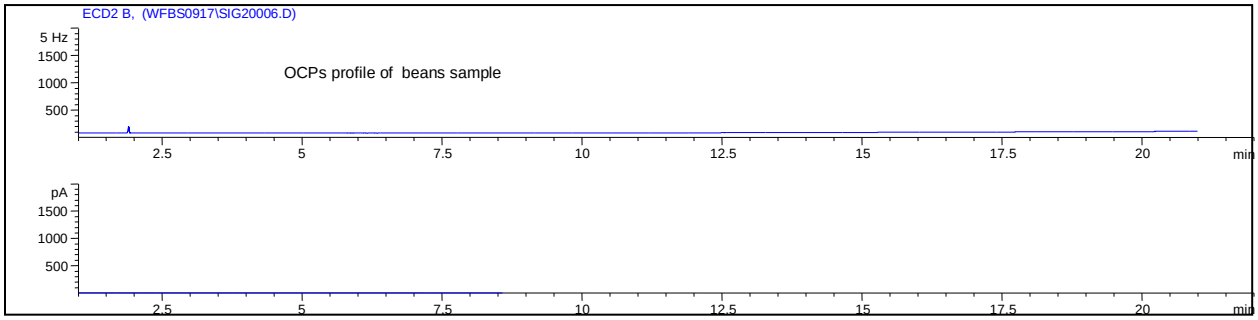

Figure 2: Chromatogram of beans and tomatoes samples

The concentration of pesticide residues in beans and tomatoes sold in Amai market, Delta state has been demonstrated. Twenty-five (25) pesticides were assessed in these consumable products (Table.1) with dichlorodiphenytetraethane having the highest mean concentration (0.0004mg/kg) in the beans sample, tomatoes (0.002±0.00) as shown in Table I.

Table 2: Estimated acceptable daily intake(EADI) and hazard quotient (HQ) for OCPs in **consumption of beans**

`Pesticide	Concentration	CR	child		Adult		RQ
DDT	0.0004	9	EADI	HQ	EADI	HQ	
			1.8E-04	0.6	2.57E-05	0.08	
Hazard index			0.6		0.08		5.0E-07

Discussion

All over the world, pesticide application has been an integral part of successful agricultural practices. However, indiscriminate use, coupled with the inherent properties of pesticide compounds and their possibility of having effects on nontarget organisms has made them the pollutant of concern in the environment. Beans and tomatoes are of great importance in Nigeria because of their health benefits(Osibanjo and Bamgbose, 1999). The concentration of pesticide residues in beans and tomatoes sold in Amai , Delta state has been demonstrated. Figures 1 and 2 show the representative chromatogram standard and sample interference peaks were obtained for the blank sample chromatogram at the same retention time as the target compounds. The mean recovery values for the spiked samples are shown in Table I. The procedure employed in this study is reproducible, efficient, and reliable for the analysis of OCPs as stipulated by EU guidelines for evaluating accuracy and precision methods (European Union,2005). The chromatogram result in this study aligns with that of the European Union, (2005); (Osibanjo and Bamgbose, 1999)and Keikotlhaile*et al.* (2010) assessed beans and tomatoes in Addo Ekiti, Ogun, and Ghana. Reports from this investigation revealed that organochlorine pesticide residues in beans ranged from 0.0002mg/kg to 0.0004mg/kg (Table I). Twenty-five (25) pesticides were assessed in these consumable products (Table I) with dichlorodiphenyltrichloroethane (DDT) having the highest mean concentration (0.0004mg/kg) in the beans sample.

The presence of these organochlorine pesticide residues in beans showed that farmers still use them for the control of pests. This is because of their potency, efficiency, and low cost compared with alternative pesticides (banned or not)(Akinnifesi *et al*., 2006; Idowu *et al*., 2013).

Several activities (volatilization, photolysis, penetration through the plant surface, inadequate training of personnel, inappropriate use of pesticides, waste from industrial chemical production, pesticides runoff from agricultural areas, sewage and refuse dump) could be attributed to the levels of chlorinated hydrocarbon compounds in agricultural produce sold in Benin City markets (Celik *et al*, 1995; Adeyemi *et al*., 2011; Ezemonye *et al*., 2008). The findings in this study agree with the findings of the FDA,1999; FEPA,1991; Ogunleye *et al*., 2010; Osibanjo *et al*., 1999; Keikotlhaile *et al*., 2010). Ezemonye *et al*.(2008) observed that organochlorine pesticide residue in local beans ranged from 0.0004 to 0.76mg/kg in Benin City, Addo Ekiti, Ogun, Lagos, and Ghana metropolises. Previous studies have demonstrated that organochlorine pesticides like gamma lindane and aldrin are toxic and can affect non-

target organisms other than the organisms of interest, thereby causing a great menace to the ecosystem and consumers(Papadakis *et al.* 2015; Celik *et al*, 1995). The use of organochlorine pesticides for the control of pests by farmers is a global issue (Cook *et al*., 1999). These compounds are characterized by high persistence, low polarity, low aqueous solubility, and high lipid solubility (lipophilicity). They are ecotoxic, non-biodegradable, and able to bioaccumulate and biomagnify in living organisms (Lars, 2000; Afful *et al*., 2010). The major concerns are their toxic effects such as interfering with the reproductive systems and fetal development as well as their capacity to cause cancer, cardiovascular disease, asthma, and other health-related diseases (Atuanya and Onuoha, 2018).

The ecological and human health risk assessment of dietary intake of organochlorine pesticide concentration(gamma lindane and aldrin) in beans in this study is shown in (Table 2). The risk quotient, estimated acceptable daily intake (EADI), hazard quotient (HQ), and hazard index(HI) for gamma lindane in beans sample were calculated using two population groups(child and adult) with varying body weights(35and 70kg)(Ezemonye *et al*., 2008). The study revealed that the risk quotient(RQ)of gamma lindane was (5.0E-07), estimated acceptable daily intake for both population groups was (EADI)(1.8E-04) and (2.57E-05). The hazard quotient (HQ) estimated were(0.6)and (0.08). The hazard indexes (HI)were (5.0E-07),(0.6 and 0.08). Aldrin,a-BHC, b-BHC, Alpha-chlordane, Gamma chlordane, Atrazine, P'p DDD, P'P DDE, P'p DDT 4,4DDT, Dieldrin, Endosulfan 1, Endosulfan11, Endosulfan sulfate, Endrin, Endrin aldehyde, Heptachlor, Heptachlor epoxide, Methoxychlor, Diazinon, Phosphoromethylglycine, TCMX, Carbamate and Decachlorobiphenyl were below detection limit(BL). Similar results were observed by Okoya*et al*., (2013) in Ado Ekiti.

Organochlorine pesticide residues at any given concentration are highly toxic, bioaccumulated, and not readily biodegraded(Osibanjo, 1999; UNDP, 2000; FDA,1999; Ezemonye *et al.*, 2008). Though the estimated concentrations in this study were minute, ecological risk assessment showed that there is a potential for toxic effects on beans and tomatoes upon exposure to organochlorine pesticides. Risk projections for humans from dietary also showed that there is potential for cancer effects. Projections showed that both children and adults were at high health risk. The result in this study is in line with the United States Agency for Toxic Substances and Disease Registry (ATSDR, 2002) and the European Union(EU, 2005) standard for estimation of OCPs(0.00001 to 1mg/kg) in beans that could be considered as unsafe to human

Conclusion

The study showed the presence of OCPs with varying concentrations in beans and tomatoes. The study therefore calls for continuous sensitization to farmers because continuous exposure to pesticide-contaminated food products sold in Amai markets (beans and tomatoes) could affect the health of consumers. Thus, more efforts should be geared toward reducing the indiscriminate and illegal use of pesticides (banned or approved). Orientation exercises for farmers to ensure proper application of pesticides and procurement of appropriate instruments of application should be conducted regularly.

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