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Eco-harmful effects of carbamates pesticide residues in tomatoes and beans samples Sold at Amai Market, Delta State

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Abstract

Pesticides are plant defensive products generally used in agriculture to augment the yield, make the quality better and extend the storage life of food crops. However, its inconsistent use, invivo properties and the ability of having effects on non-target organisms has made it a pollutant of concern in the environment. This study examined the effects of carbamates pesticide residues in tomatoes and beans samples Sold at Amai Market. Samples of tomatoes and beans were purchased randomly from Amai market in Ukwani L.G.A, Delta State. A total of eight carmates pesticides (CP) residues in these samples were analyzed with gas chromatography- electron captured detector (ECD). The estimated acceptable daily intake of Aldicarb and Carbofuran in tomatoes and oxamyl, 3-Hydroxyl carbofuran and Aldicarb present in beans was determined based on European Union (EU) and United States Agency of Toxic Substances and Disease Registry (ATSDR) standards, using two population age groups (child and adult).

The study results revealed levels of carbamates pesticide residues among the tomatoes and beans. Aldicarb had the highest value of 2.40 ng/ml followed by Carbofuran with a value of 1.51ng/ml in tomatoes. In the bean sample, Aldicarb had 5. 22 ng/ml followed by Oxamyl with 2. 22 ng/ml and 3-Hydroxylcarbofuran had 1.61ng/ml respectively. The study thus suggests that Carbamates pesticides concentration in tomatoes and beans are hazardous for human consumption. The study therefore calls for continuous inspection of agricultural farmlands because continuous exposure to carbamates pesticide contaminated tomatoes and beans products sold at Amai markets could affect the health of consumers.

Keywords: Tomatoes; Beans; Carbamate Pesticides; Aldicarb

Introduction

Pesticides are plant defensive products generally used in agriculture to augment the yield, make the quality better and extend the storage life of food crops. However, its inconsistent use, invivo properties and the ability of having effects on non-target organisms has made it a pollutant of concern in the environment [1]. They provide society with a wide range of merits, especially in agricultural productivity by the control of diseases. The world market for pesticides has increased substantially from $2.7 billion in the 1970s, to $18.5 billion in 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) [2]. There has been an increasing concern about ingestion of carbamates pesticide residues by adults and children who consume tomatoes and beans [1].

Consumption of tomatoes and beans have increased tremendously over time because of their health benefit. In the United States of America, for example, the consumption tomatoes and beans doubled from 1990 to 2000 (UNDP, 2000). Xenobiotics are core cause for worry all over the world, because of their persistence leaving behind metabolite residues after degradation on plants, eco-biota and humans [1]. In Nigeria, there have been reports of some levels of pesticide residues in ready to eat agricultural fruit vegetables, processed beverages, soft drinks, and bottle drinking [1]. The results have continually revealed contamination by organochlorine pesticides in water and agricultural produce sold in Nigeria markets.

The Carbamates pesticides (CPs) include Aldicarb, Oxamyl, 3-Hydroxylcarbofuran, Carbofuran, Methomyl, Aldicarb Sulfone

and Methiocarb. Residue analysis provides a measure of the nature and level of any chemical contamination within the environment and of its persistence (OECD, 2003). The pesticides must undergo extensive efficacy, environmental, and toxicological testing to be registered by governments for legal use in specified applications (OECD, 2010). Hence, this research investigated the levels of Cabrmates pesticides residues in tomatoes and beans sold at Amai Market.

Materials And Methods

Study area

This study was conducted in Amai, Ukwuani Local Government Area in Delta State, Nigeria. The inhabitants of Amai, like most other rural communities in Delta State, engage in farming activities. The produce is consumed by the farmers while the excess is sold to traders who come from nearby towns such as Warri and beyond.

Samples collection

Random sampling of tomatoes and beans were purchased from Amai Market. Samples were enclosed in 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.

Carbamates pesticide residue extraction

The persistent Carbamates pesticide residues from tomatoes and beans samples were analyzed chromatographically with gas chromatography equipped with electron capture detector (GC/ECD) as described by [Papadakis et al. 2015]. Thirty grams (30 g) aliquot of well mixed samples was 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 (Na2SO4) were weighed and added to the sample and mixed thoroughly to form a free-flowing powder.50ml of the solvent were mixed and added to the samples. Samples were placed in the Sonicator and Sonicated for about 10 – 15 minutes at about 600C. The extract was decanted into a round bottom flask. The procedure was repeated once more an additional 50 ml of solvent were mixed, Sonicated and allowed to settle in the beaker before it was decanted into the round bottom flask. The samples were extracted with 2ml 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

Asyringe was used to transfer 2ml 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 3mins, the upper hexane layer was observed and ensured that it was not highly colored. After the cleanup procedure, each fraction was evaporated with rotary evaporator and concentrated in a nitrogen stream to 1ml. The fractions were analyzed for pesticide residues. The Carbamates pesticides residues concentrations were determined using a Hewlett-Packard (HP) 5890 series II equipped with electron capture detector with 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^{0}C for 1 min; increased to 200^{0}C for 2 min. The injection volume was 1ml. The detector temperature was maintained at 300^{0}C [2-7].

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.

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

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

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

Concentration (p) = Concentration of peak or Total concentration of range

Area = Area of peak or total across range.

Risk assessment of pesticide residues

Ecological risk assessment

The Risk quotient (RQ) method was used to determine the risk of Carbamates pesticide exposure to non-target tomatoes and beans. RQ is the ratio of the measured environmental concentration (MEC)to the predicted no effect concentration (PNEC). The predicted no effect concentration (PNEC) was obtained by multiplying the LC50 with an assessment factor (AF) of 100. The assessment factor takes into account the uncertainty in

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extrapolation from laboratory toxicity tests for a limited number of dietary and non dietary products to the real environment. The LC50 was obtained from Mineau (2005).

Risk Quotient (RQ)=MEC" / "PNEC [Papadakis et al. 2015].

Health risk estimations

To assess the health risk associated with exposures to pesticideresidues through consumption of tomatoes and beans (dietary intake). The guidelines for potential risk assessment drawn up by the USFDA were used.

Human health risk assessment of Carbamates pesticide residues in Tomatoes and Beans (dietary)

To estimate the carcinogenic and non-carcinogenic risk of detected Carbamates residues 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 tomatoes and beans by the consumption rate in Nigeria (L/day or kg/day) and dividing the product by the body weight (kg) [1]. 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 (2021). The food and agricultural organization (FDA,1999) quotes the per capita consumption of tomatoes and beans in Nigeria as 9kg -20.5kg. The following formula was used to estimate the dietary intake.

$$EADI = \frac{C \times CR}{BW}$$ [Fianko et al. 2011].

EADI is the estimated average daily intake, C is the concentration of Carbamates pesticide residues, CR represents consumption rate of tomatoes and beans while BW represents the body weight of age group. The food and agricultural organization (FDA, 2021) quotes the per capita consumption of tomatoes and beans in Nigeria as 9kg -20.5kg, while body weight was set at 70 kg for adult population group.

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

$$\textbf{Hazard quotient (HQ)} = \frac{EADI}{RfD}$$ [Fianko et al. 2011].

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

$$\textbf{Hazard Index (HI)} = \sum_i^n HQI.$$ [Fianko et al. 2011].

Results

Figure 1 above revealed the Chromatogram of eight Carbamates pesticide residues from beans sample analyzed. It shows that Methiocarb had the highest retention time of 20.538S followed by Carbofuran with 19.679s, adicarb with 17.688, Oxamyl was 11.914 and Aldicarb Sulfone and Aldicarb Sulfoxide had the lowest values, respectively [8-12].

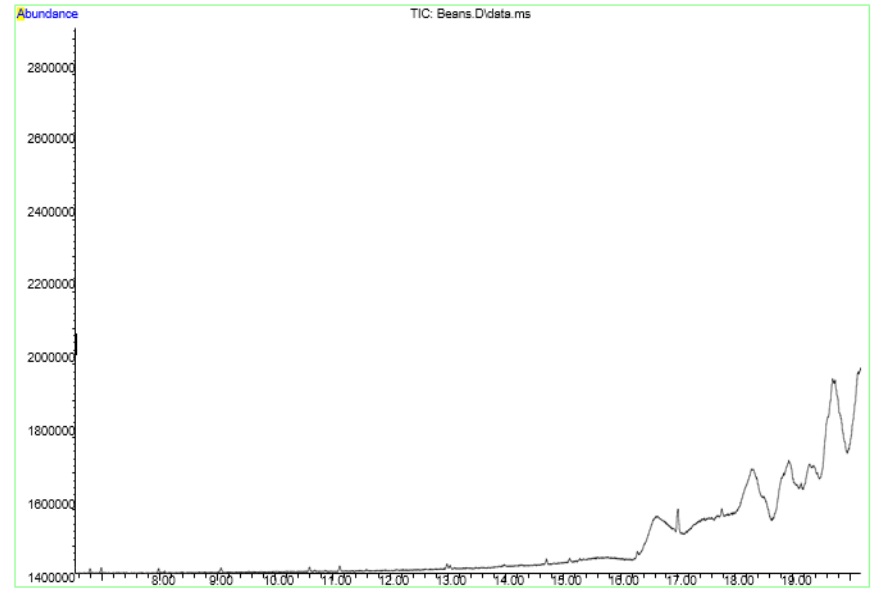

Figure 1: Chromatogram of Beans Sample.

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Table 1 above revealed the mean concentration of eight (8) carbamates pesticide residues in beans sample analyzed. The analysis revealed that Aldicarb had the highest mean concentration of 5.22ng/ml followed by Oxamyl with 2.06ng/ml and 3-Hydroxycarbofuran had 1.11ng/ml while Aldicarb Sulfoxide, Aldicarb Sulfone, Methomyl and Methiocarb were below 1ng/ml, respectively. The results revealed that such a level of concentration could pose a threat to consumers according to USFDA, (2021).

Table 1: Mean concentration of Carbamates pesticide Residue in Beans.

Target Compounds	R.T. QIon	Response	Conc Units	Dev(Min)	Qvalue
1) Aldicarb Sulfoxide	7.68	TIC	6966	0.07	ng/ml# 37
2) Aldicarb Sulfone	9.345	TIC	5683	0.77	ng/ml# 17
3) Oxamyl	11.754	TIC	4778	2.06	ng/ml# 16
4) Methomyl	14.718	TIC	49560	0.76	ng/ml# 9
5) 3-Hydroxycarbofuran	15.531	TIC	6293	1.11	ng/ml# 15
6) Aldicarb	17.534	TIC	8799	5.22	ng/ml# 10
7) CarboFuran	19.508	TIC	7189	0.31	ng/ml# 35
8) Methiocarb	20.538	TIC	9673	0.63	ng/ml# 23

The Figure 2 above revealed the Chromatogram of eight Carbamates pesticide residues from tomatoes sample analyzed. It shows that Carbofuran had the highest rention time with 19.679s followed by adicarb with 17.688, Oxamyl was 11.914 and Aldicarb Sulfone and Aldicarb Sulfoxide had the lowest values, respectively. Table 2 above revealed the mean concentration of eight carbamates pesticide residues in tomatoes. The results showed that Aldicarb had the highest mean concentration of 2.40ng/ml, followed by Carbofuran with 1.51ng/ml while Aldicarb Sulfoxide, Aldicarb Sulfone, oxamyl had values below 1ng/ml. Methiocarb, Methomyl and 3-Hydroxycarbofuran were not detected, respectively. The results revealed that such a level of concentration could pose a threat to consumers according to USFDA, (2021) [13-18].

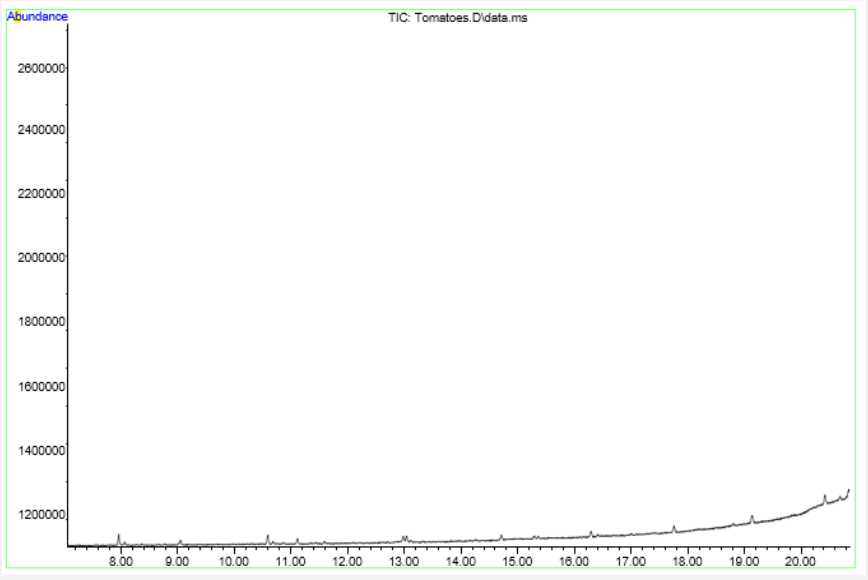

Figure 2 : Chromatogram of Tomatoes Sample.

Table 3 above revealed the estimated acceptable daily intake of beans as 20.5kg going by United States Food and Drug Administration recommendations. The results above show that children consume beans more with a value of 4.60 while adults had 2.5. The hazard index also revealed that it was more on the children 1. 5 than the adults with 1. 4.

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Table 2: Mean concentration of Tomatoes.

Compound	R.T. QIon	Response	Conc Units	Dev (min)
Target Compounds				**Qvalue**
1) Aldicarb Sulfoxide	7.743	TIC	5803	0.06 ng/ml# 34
2) Aldicarb Sulfone	8.973	TIC	2253	0.03 ng/ml# 23
3) Oxamyl	11.914	TIC	4800	0.06 ng/ml# 25
4) Methomyl	0		0	N.D.
5) 3-Hydroxycarbofuran	0		0	N.D.
6) Aldicarb	17.688	TIC	95418	2.40 ng/ml# 10
7) CarboFuran	19.679	TIC	4373214	1.51 ng/ml# 21
8) Methiocarb	0		0	N.D.

Table 3: Above revealed the estimated acceptable daily intake.

`Pesticide	Concentration	CR	child		Adult		RQ
Aldicarb	5.22ng/ml	20.5	EADI	HQ	EADI	HQ	
			4. 60	2. 0	2.5	1. 0	
Hazard index			1. 5		1. 4	1.2	

Discussion

The use of pesticide has played a core part in successful agricultural practices worldwide. However, misuse, followed with the invivo properties of carbamates pesticide compounds and their possibility of having effects on organisms that are not of interest has made them the pollutant of concern in the environment. Beans and Tomatoes are of great importance in Nigeria because of their nutritional value [1].

The Figure 1 above revealed the Chromatogram of eight Carbamates pesticide residues from beans sample analyzed. It shows that Methiocarb had the highest retention time of 20.538S followed by Carbofuran with 19.679s, Adicarb with 17.688, Oxamyl was 11.914 and Aldicarb Sulfone and Aldicarb Sulfoxide had least values respectively. Table 1 above revealed the mean concentration of eight (8) carbamates pesticide residues in beans sample analyzed. The analysis revealed that Aldicarb had the highest mean concentration of 5.22ng/ml followed by Oxamyl with 2.06ng/ml and 3-Hydroxycarbofuran had 1.11ng/ml while Aldicarb Sulfoxide, Aldicarb Sulfone, Methomyl and Methiocarb were below 1ng/ml, respectively [19-23].

The results revealed that such a level of concentration could pose a threat to consumers according to USFDA, (2021). The results in Figure 1 and Table 1 representative chromatogram standard and samples. No interference peaks were obtained for the blank sample chromatogram at the same retention time as the targets compounds. The mean recovery values for the spiked samples are shown in Table 1. The procedure employed in this study is reproducible, efficient and reliable for the analysis of CPs as stipulated by EU guidelines for evaluating accuracy and precison method (European Union,2005).

The chromatogram result in this study is in consonance with that of European Union, (2005 [1], [Keikotlhaile et al.] assessed Beans and Tomatoes in Addo Ekitti, Benin City, Ogun and Ghana. Reports from this investigation revealed that Carbamates pesticide residues in ranged from Aldicarb 5.22ng/ml followed by Oxamyl with 2.06ng/ml and 3-Hydroxycarbofuran had 1.11ng/ml while Aldicarb Sulfoxide. Figure 2 above revealed the Chromatogram of eight Carbamates pesticide residues from tomatoes sample analyzed. It shows that Carbofuran had the highest rention time with 19.679s followed by adicarb with 17.688, Oxamyl was 11.914 and Aldicarb Sulfone and Aldicarb Sulfoxide had least values, respectively. Many activities (volatilization, photolysis, penetration through the plant surface, inadequate training of personnel, inappropriate use of Carbamate 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 Amai market [3,1]. The findings in this study are in consonance with the findings of USDA, 2021 [Keikotlhaile et al.] [1] observed that Carbamates pesticide residue in Beans and Tomatoes ranged from 0.0001 to 0.75mg/kg in Benin City.

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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 great menace to ecosystem and to consumers [Papadakis et al. 2015], [4]. The use of organochlorine pesticides for the control of pest by farmers is a global issue [5]. These compounds are characterized by high persistent, low polarity, low aqueous solubility and high lipid solubility (lipophilicity). They are ecotoxic, non-biodegrable and able to bioaccumulate and biomagnify in living organisms [Lars, 2000], [6]. The major concerns are their toxic effects such as interfering with the reproductive systems and foetal development as well as their capacity to cause cancer, cardiovascular disease, asthma and other health related diseases [4].

Table 2 above revealed the mean concentration of eight carbamates pesticide residues in tomatoes. The results showed that Aldicarb had the highest mean concentration of 2.40ng/ml, followed by Carbofuran with 1.51ng/ml while Aldicarb Sulfoxide, Aldicarb Sulfone, oxamyl had values below 1ng/ml. Methiocarb, Methomyl and 3-Hydroxycarbofuran were not detected respectively. The results revealed that such level of concentration could pose threat to consumers according to USFDA, (2021). Table 3 above revealed the estimated acceptable daily intake of beans as 20.5kg going by United States Food and Drug Administration recommendations. The results above shows that children consume beans more with a value of 4.60 while adults had 2.5.

The hazard index also revealed that it was more on the children 1. 5 than the adults with 1. 4. The results above is in consonance with previous findings of UNDP, 2000 who states that carbamtes pesticide residues at any given concentration are highly toxic, bioaccumulate 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 of toxic effects to tomatoes and beans upon exposure to carbamates pesticides. Risk projections for human's daily intake also revealed that there is potential for cancer effects. Projections showed that both children and adult were at high health risk. The result in this study is in line with the United States Agency of Toxic Substances and Disease Registry (ATSDR, 2002) and European union (EU, 2005) standard for estimation of CPs(1ng/ml to 5ng/ml) in tomatoes and beans could be considered as unsafe to human.

Conclusion

The study calls for continuous inspection of agricultural farmlands because continuous exposure to carbamates pesticide contaminated tomatoes and beans products sold at Amai markets could affect the health of consumers.

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