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Assessment of Type and Concentration of Pesticide Residues in Fluted Pumpkin (Telfairia occidentalis) and Green Amaranth (Amaranthus hybridus) Sold in Akure, Ondo State, Nigeria

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ABSTRACT: Pesticide residues have been reported in edible agricultural products, posing a huge public health risk. This study investigated the type and concentration of pesticide residues present in common vegetables, Fluted Pumpkin (*Telfairia occidentalis*) and Green Amaranth (*Amaranthus hybridus*), sold in four major markets in a capital city, Akure, Ondo State, Southwest Nigeria. Using a series of Gas Chromatography techniques, twenty-three (23) pesticide residues including herbicides, organophosphates, organochlorines and pyrethroids were identified. The mean pesticide residues with the highest concentration (in ppm) in each group include Herbicide: Atrazine (Fluted pumpkin – 0.35, Amaranth – 0.18, Maximum Residue Limit (MRL) – 0.05); Organophosphate: Mevinphos (Fluted Pumpkin – 0.047, Amaranth – 0.043, MRL – 0.01); Organochlorine: Lindane (Fluted Pumpkin – 0.67, Amaranth – not detected (ND), MRL – 0.01) and Aldrin (Fluted Pumpkin – ND, Amaranth – 0.19, MRL – 0.01); Pyrethroid: Permethrin (Fluted Pumpkin – 1.13, Amaranth – 0.23, MRL – 0.05). The frequency of pesticide residues contamination was higher in Fluted Pumpkin (12) than in Green Amaranth (9). The presence of multi-pesticide residues in the vegetables at levels above the WHO MRL and the attendant toxic effect makes pesticide contamination of leafy vegetables a priority public health concern in Akure. This study has brought to the fore the need for more attention to be paid to pesticide contamination of locally-consumed agricultural products against the high incidence of health challenges (such as cancer, birth defect, impaired immune function, and neurobehavioral disorder) linked to pesticide contamination.

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The use of chemicals to combat weeds and pests in Nigeria to improve crop yield has led to the indiscriminate use of pesticides. Erhunmwunse *et al.* (2012) reported that about 15,000 metric tonnes of pesticides comprising 135 different chemical types are imported annually into the country to meet the market demands. Pesticides are generally composed of herbicides and insecticides, and other chemical types such as nematicides and fungicides (Liebman and Dyck, 1993). Despite the benefits from pesticides in controlling weed and insect pests, evidence in the last few decades have shown that they could also be

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detrimental to human health and the ecosystem (Tadesse and Asferachew, 2008). For example, the accidental ingestion of pesticides by humans could potentially cause a wide range of diseases and health complications such as cancer, cardiovascular diseases, birth defect, dermatitis, impaired immune function, neurobehavioral disorder, and fatal allergy (Ogar *et al.,* 2011). Vegetables are a major component of Nigerian diet as in other parts of the world. The Food and Agricultural Organization (FAO) and the World Health Organization (WHO) recommended a daily vegetable intake of 200 g per person. However, the Nigerian national average is well below the recommended standard (FAO, 1991). Vegetables are of good nutritional value because they contain micronutrients, vitamins, minerals, and dietary fibre; therefore, serve as sources of supplements for diets (Chadha and Oluoch, 2003). Fluted pumpkin (*Telfairia occidentalis*) and Green Amaranth (*Amaranthus hybridus)* are commonly grown vegetables, commercially available and widely consumed in Nigeria for its nutritional value, especially in the southern part of the country, including Akure (Okunlola and Ofuya, 2010; Okunlola and Akinrinola, 2014). Vegetables are also economically viable as they are commonly grown by farmers to improve their income. For instance, vegetable production contributed between 33 to 37% of family income among peri-urban vegetable farmers in 2008 production season (Okunlola and Akinrinola, 2014). They also serve as a source of income for women (Owombo *et al.,* 2012) who have gardens/small farming units behind their homes. However, the attack of these vegetables by various insect pests is a major concern to farmers for not only reducing the quality and yield of produce, but also for causing economic loss (Sithanatham *et al.,* 2003). Pesticides are important to achieve effective pest control; the longer they persist, the greater the chances of exposure to targeted pests. However, this prolonged exposure eventually becomes directly or indirectly harmful to non-target organisms (e.g., microfauna and microflora). The use of pesticides inadvertently leads to residues in food crops and the environment. It is reported that only 1% of pesticides used are effective with the remaining 99% becoming unwanted residues settling in water bodies, soil and atmosphere or even absorbed by organisms (Zhang *et al.,* 2011). Pesticide residues in produce (e.g. vegetables or food crops) are a direct result of the application of pesticides on the field (Businelli *et al.,* 1992), and their toxic effects manifest in humans when bioaccumulation occurs along the food chain after initial uptake by edible plants. Maximum Residue Limits (MRLs) are set to reflect the highest amount of pesticide residue expected in foods when pesticides are applied correctly in accordance with approved conditions of use. MRLs are primarily trading standards but also serve to ensure that residue levels do not pose negative risks to consumers. It has been severally documented that pesticide residues above recommended MRLs have been found in fruits and vegetables in Nigeria. For instance, vegetables such as *Telfairia occidentalis* and *Cetosia argentea* purchased from six markets in Lagos State, Nigeria, were found to contain as high as 2.35 mg/kg of aldrin which was far above the WHO limit (0.02 mg/kg) (Njoku *et al.,* 2017).

Despite the various studies on the presence and fate of pesticide residues in food, there are a few gaps in information regarding pesticide residue levels in leafy vegetables sold in Akure Metropolis, the capital city of Ondo State, Nigeria. Therefore, the objective of the paper was to assess the type and concentration of Pesticide Residues in Fluted Pumpkin (*Telfairia occidentalis*) and Green Amaranth (*Amaranthus hybridus*) Sold in Akure, Ondo State, Nigeria.

MATERIALS AND METHODS

Study location: The study was carried out in Akure metropolis, the capital city of Ondo state, located in Southwest Nigeria at latitude 7.2571˚ N and longitude 5.2058˚ E of the equator. It is situated in the rainforest ecological zone.

Sample collection and preparation: The two vegetables (*T. occidentalis* and *A. hybridus*) were purchased from four major markets in Akure, namely, Nepa, Isikan, Odopetu and Oba. The locations were coded randomly to avoid prejudice towards any market. For each market, replicate samples of vegetable size of at least 2 kg were purchased randomly from vendors, following the methods described by the Codex Alimentarius Commission (2000). The samples were collected in clean polythene bags and taken to the Environmental Biology Research Laboratory of the Federal University of Technology Akure, Nigeria for further studies. At the laboratory, edible portions of the sampled vegetables were picked, chopped, and prepared for subsequent analyses.

Extraction and clean-up of vegetable samples: The method of extraction used for the vegetables is the United States Environmental Protection Agency (USEPA) method 3510 for extracting pesticide residues in non-fatty crops using ethyl-acetate, demonstrated by Akan *et al*. (2013). Triplicate subsamples of 100 g of each of the sample was weighed using an Electronic Weighing Balance, JA 3003 (S. Mettler HZ-300g/0.01g), washed thoroughly with distilled water and blended into a paste. The paste was transferred into a conical flask and 40 ml of ethyl-acetate was added and shaken vigorously. A 5 g portion of sodium hydrogen carbonate ($NaHCO_3$) was added to the mixture. After shaking for a minute, 20 g of anhydrous sodium sulphate (Na_2SO_4) was also added and the entire mixture shaken vigorously for 1 h. To neutralize any acid present, $NaHCO_3$ was added while anhydrous sodium sulphate (Na_2SO_4) was added to the homogenate to remove water from the sample matrix. The mixture was filtered into a labeled container and thereafter, centrifuged at a speed of 2000 rpm for 10 mins using 80-2 Electronic Centrifuge. The organic layer was decanted into a container and a 1:1 mixture of 5 ml ethyl-acetate and cyclo-hexane was added prior to clean-up.

The vegetable extracts were cleaned up using a 10 mm chromatographic column filled with 3 g activated silica gel and topped up with 3 g of anhydrous sodium sulphate. N-hexane (20 ml) was used to rinse out the column. The residue in 2 ml N-hexane was transferred into the column and the extract was rinsed thrice with 2 ml N-hexane. The procedure was repeated for all the samples. The samples were collected in 2 ml vials, sealed, and preserved in a refrigerator at a temperature of 4^{o}C.

Pesticide residue determination using Gas Chromatography: Determination of pesticide residues were conducted at the National Agency for Food and Drugs Administration and Control (NAFDAC) Laboratory, following the Netherlands Analytical Methods of Pesticide Residues and Food Stuff (2006).

Determination of herbicide residues: The Shimadzu Japan, GC/MS-QP2010 equipped with fluorescence detector was used to analyse the herbicide extracts. The column was held at 60^{o}C in injection volume of 1 µl programmed to 250^{o}C. The mass spectrometer detector was held at 250^{o}C above the maximum column temperature. The sample size of 1 µl was split in 100-1 unto the column so that the total charge on the column was 1. Helium was used as the carrier gas at a linear velocity of 46 cmsec^{-1} and pressure of 100.2 kPa. The instrument conditions were capillary column coated with Hp5 MS (30 m x 0.25 µm, 0.25 mm id). The oven temperature was programmed at an initial temperature of 80^{o}C for one minute, to 200^{o}C for two minutes and 280.0^{o}C for final hold time of one minute. The column flow rate was 1.18 mL/minute. The detection of pesticides was performed using GC-ion trap MS with optional MSN mode. The scanning mode offer enhanced selectivity over either full scan or selected ion monitoring (SIM). At the elution time of each pesticide using selected ion monitoring, the ratio of the intensity of the matrix ions increases exponentially versus that of the pesticide ions as the concentration of the pesticide approach the detection limit, decrease the accuracy at lower levels. The GC-ion trap MS was operated in MSN mode and perform tandem MS function by injecting ions into the ion trap and destabilizing matrix ions, isolating only the pesticide ions. The retention time, peak area and peak height of the sample was compared with those of the standards for quantitation.

Determination of insecticides residues: The instrument used was Agilent (7890A) Gas Chromatograph (GC) equipped with Agilent 7693 Autosampler with Flame Photometric Detector (FPD). The instrument condition was capillary column coated with HP-5 (30 m x 0.25 µm x 0.32 mm id). The Carrier Gas was Nitrogen, Column flow rate of 2.7 mL/min and injection temperature set at 250^{o}C. For the organophosphate residues, the oven was set at an initial temperature of 60^{o}C for one minute, 200^{o}C for 2 min and 280^{o}C for 3 min with a total runtime of 28 min; while for organochlorines and pyrethroids, the oven was set at an initial temperature of 80^{o}C for 1 min, 180^{o}C for 3 min and 300^{o}C for 2 min with a total runtime of 28 min. 1 ml of the sample was injected into the GC. The splitless mode was used for the sample injection. The identities of the organophosphates in sample extracts were confirmed by spiking and comparing their retention times with those of standards. Concentrations were determined by computer calculation making use of both the response factors of the residues and the internal standard.

Data analysis: The data were compared with the maximum residues limits (MRLs) set by the European Union (European Union Database, 2014) to identify samples with pesticide residues above the standard limits. The data from the study were subjected to Analysis of variance (ANOVA) and the means separated by Tukey's Test at $p<0.05$; SPSS (Version 17.0) was used for all analyses.

RESULTS AND DISCUSSIONS

An herbicide, atrazine, was detected in all the fluted pumpkin samples in all the markets except MK 3 and the lowest residue level detected (7.03 x 10^{-2} ppm) exceeded atrazine's maximum residual limit (MRL; 5.00 x 10^2 ppm) (Table 1). Though the highest mean concentration of atrazine observed in fluted pumpkin was 34.5 x 10^{-2} ppm from MK 2, the upper limit of the range was 91 x 10^{-2} ppm. The atrazine levels in the green amaranth leave samples collected from the markets were lower than the MRL except in MK 4 where concentrations were as high as 18.03 x 10^2 ppm (range: 0.00 – 32.20 x 10^{-2} ppm) exceeding the MRL (Table 2). Alachlor, on the other hand, was undetected in all samples collected from all the markets (Tables 1 and 2). Houjayfa *et al*. (2020) similarly reported high concentrations of atrazine in green amaranth (treated with the herbicide) collected in Cameroon, which exceeded the MRL. The occurrence of atrazine in the two leafy vegetables sampled in this study is consistent with the findings of Gushit *et al.* (2013) who detected atrazine residues in some root crops and leafy vegetables in Plateau State, Nigeria, though in low level in the leafy vegetables.

Table 1: Mean concentrations of herbicide residues in Fluted Pumpkin leaves from markets in Akure, Nigeria.

Samples	**Herbicides (ppm x 10^{-2})**	
	Atrazine	**Alachlor**
MK 1	7.033 ± 1.33[a]	ND
MK 2	34.500 ± 3.43[c]	ND
MK 3	ND	ND
MK 4	13.367 ± 1.97[b]	ND
MRL (EU)	5	1

Means followed by the same letter along the column are not significantly different (p>0.05) using Tukey's Test. MK = Market; MRL = Maximum Residue Limit set by the European Union; ND = Not Detected.

Farmers make use of herbicides in land preparation and 62% of the farmers in Akure were reported to control weeds with herbicides (Adeola, 2012). This explains how contamination may occur if such farmlands are not given enough time for the herbicides to breakdown before planting vegetables on them. Atrazine is a choice herbicide in land preparation because it is cheap and affordable to the relatively poor vegetable farmers.

Table 2: Mean concentrations of herbicide residues in Green Amaranth leaves from markets in Akure, Nigeria.

Samples	**Herbicides (ppm x 10^{-2})**	
	Atrazine	**Alachlor**
MK 1	ND	ND
MK 2	3.067 ± 0.21[a]	ND
MK 3	ND	ND
MK 4	18.033 ± 1.43[b]	ND
MRL (EU)	5	1

Means followed by the same letter along the column are not significantly different (p> 0.05) using Tukey's Test. MK = Market; MRL = Maximum Residue Limit set by the European Union; ND = Not Detected.

Atrazine is readily available in soil to plants because it is loosely bound to the soil thus making its release to plants easy. It is highly volatile and can also be found in the particulate and vapor phases of air (ATSDR, 2003) and will be broken down in the soil over a growing season. The high levels of atrazine can be attributed to the fact that it accumulates in the growing tips and the leaves. Atrazine contamination is of importance as it is classified as priority pollutant because it is harmful to organisms even at microgram (µg) levels (Agdi *et al.,* 2000*)*. It is linked to incidences of premature deliveries, birth defects and acts on the brain to disrupt the hormone that triggers ovulation (Heather, 2009).

Dichlorvos and mevinphos were detected in all the market samples at levels exceeding their MRLs (1 x 10^{-2} ppm) in both fluted pumpkin and green amaranth (except mevinphos in MK 4) (Tables 3 and 4). Of all the organophosphates residues detected, mevinphos had the highest mean concentration in both vegetables (*T. occidentalis*- 4.694 x 10^{-2} ppm; *A. hybridus*- 4.298 x 10^{-2} ppm), these were observed in MK 2 samples. In all the markets, dichlorvos was detected in both fluted pumpkin and green amaranth in concentrations slightly above the MRL (*A. hybridus* range: 1.000 x 10^2 ppm – 2.100 x 10^{-2} ppm; *T. occidentalis* range: 1.300 x 10^2 ppm – 3.700 x 10^{-2} ppm). No other organophosphate residue was detected in green amaranth in all the markets. However in fluted pumpkin leaves, diazinon and pirimiphos-methyl were detected in two markets each below the MRL (though a replicate each from Mk 1 and MK 3 had concentrations slightly above the MRL) (Table 3). In a similar trend to the detection of dichlorvos in this study, Akan *et al.* (2013) reported high levels of dichlorvos in vegetables from agricultural areas of Borno State, Nigeria. Dichlorvos, just like all other organophosphates insecticides, are toxic at very low concentrations but the effect is short-lived such that repeated applications are required at short time intervals. Dichlorvos is classified as highly hazardous by WHO because of its high oral and dermal toxicity (WHO, 2019).

It is an organophosphate that is commonly used in developing countries. Expectedly, the levels of organophosphates were low because of their short half-life; however, the need for repeated application must have accounted for the concentrations above the MRLs in the samples collected. Mevinphos, on the other hand, is classified as highly toxic, and the use is restricted internationally. Its effectiveness as an insecticide over a wide range of insect pests may be responsible for making it a choice in the management of insect pests and a probable disregard for Good Agricultural Practice (GAP) by farmers may account for its high levels in the samples collected. Since most organophosphates share common toxicity mechanism, an exposure to several of them and their breakdown products as observed in this study could intensify their toxic effects.

The concern over detection of organophosphates in edible vegetables is due to their status as toxic insecticide group commonly used, they affect the human nervous system even at very low levels of exposure. They are also suspected to have adverse effect on male fertility, increase the risk of Parkinson's disease (Bloomquist *et al.,* 2002), cause attention deficit hyperactivity disorder (ADHD) in children, and behavioral problems and lower cognitive function in infants (PAN, 2013).

Table 3: Mean concentrations of organophosphate residues in Fluted Pumpkin leaves from markets in Akure, Nigeria.

Samples	Organophosphate pesticides (ppm x 10^{-2})				
	Dichlorvos	**Mevinphos**	**Diazinon**	**Dimethoate**	**Pirimiphos-methyl**
MK 1	1.721 ± 0.91^a	3.504 ± 0.93^b	0.203 ± 0.97^a	3.277 ± 0.88^a	1.700 ± 0.07^a
MK 2	1.864 ± 0.43^a	4.694 ± 1.03^{bc}	ND	ND	ND
MK 3	2.850 ± 0.83^b	1.379 ± 0.03^a	0.366 ± 0.03^a	ND	ND
MK 4	2.040 ± 0.21^a	1.310 ± 0.05^a	ND	ND	2.518 ± 1.52^b
MRL (EU)	1	1	1	2	5

Mean followed by the same letter along the column are not significantly different (p>0.05) using Tukey's Test. MK = Market; MRL = Maximum Residue Limit set by the European Union; ND = Not Detected.

Table 4: Mean concentrations of organophosphate residues in Green Amaranth leaves from markets in Akure, Nigeria.

Samples	Organophosphate pesticides (ppm x 10^{-2})				
	Dichlorvos	**Mevinphos**	**Diazinon**	**Dimethoate**	**Pirimiphos-methyl**
MK 1	1.769 ± 0.43^a	3.420 ± 1.23^a	ND	ND	ND
MK 2	1.452 ± 0.99^a	4.298 ± 0.53^a	ND	ND	ND
MK 3	1.603 ± 0.91^a	1.219 ± 0.03^b	ND	ND	ND
MK 4	1.803 ± 0.23^a	ND	ND	ND	ND
MRL (EU)	1	1	1	2	5

Mean followed by the same letter along the column are not significantly different (p>0.05) using Tukey's Test. MK = Market; MRL = Maximum Residue Limit set by the European Union; ND = Not Detected.

Organochlorines such as lindane, chlorothalonil and aldrin exceeded their MRLs in the fluted pumpkin samples collected from all the markets (Figure 1). Other organochlorines such as α-BHC, pp'-DDD, pp'-DDT, and dieldrin were detected in fluted pumpkin samples collected from at least one market. The highest organochlorine residue level detected from all four markets was lindane at 67.06 x 10^{-2} ppm, which exceeded its MRL (1.00 x 10^{-2} ppm).

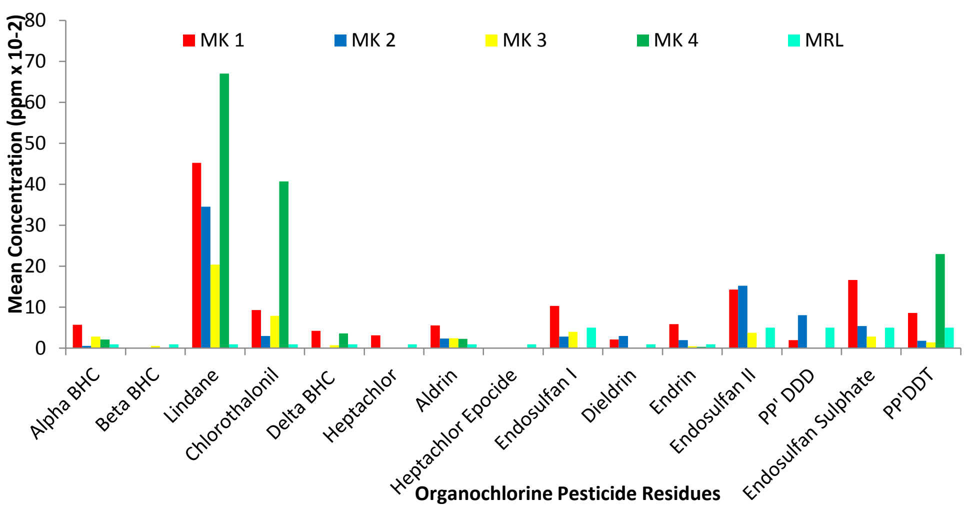

Fig 1: Mean concentrations of organochlorine residues in samples of fluted pumpkin leaf from markets in Akure. MK = market; MRL = Maximum Residue Limit set by the European Union.

Aldrin, lindane, pp'-DDT, beta-BHC, delta-BHC, chlorothalonil, heptachlor, heptachlor epoxide, dieldrin, and endrin, were detected in green amaranth samples in concentrations higher than their MRLs (Figure 2). The top three organochlorine residues with the highest concentrations were aldrin in MK 1 (20.00 x 10^2 ppm), lindane in MK 3 (13.00 x 10^2 ppm) and pp'-DDT in MK 4 (13.00 x 10^2 ppm), which exceeded their MRLs, 1.00 x 10^2 ppm, 1.00 x 10^2 ppm and 5.00 x 10^2 ppm, respectively. Furthermore, alpha-BHC, endosulfan I and II, endosulfan sulphate, and pp'-DDD, were detected below their MRLs. These findings are consistent with the study by Abah *et al.* (2021) who assessed six organochlorine residues (DDT, DDD, aldrin, heptachlor, dieldrin and endosulfan) in Green Amaranth collected in Taraba State, Nigeria, and found DDT to have the highest concentration while endosulfan had the least concentration. DDT concentrations in Green Amaranth collected in Southwestern Nigeria, also exceeded the MRL in a study by Adeleye *et al.* (2019).

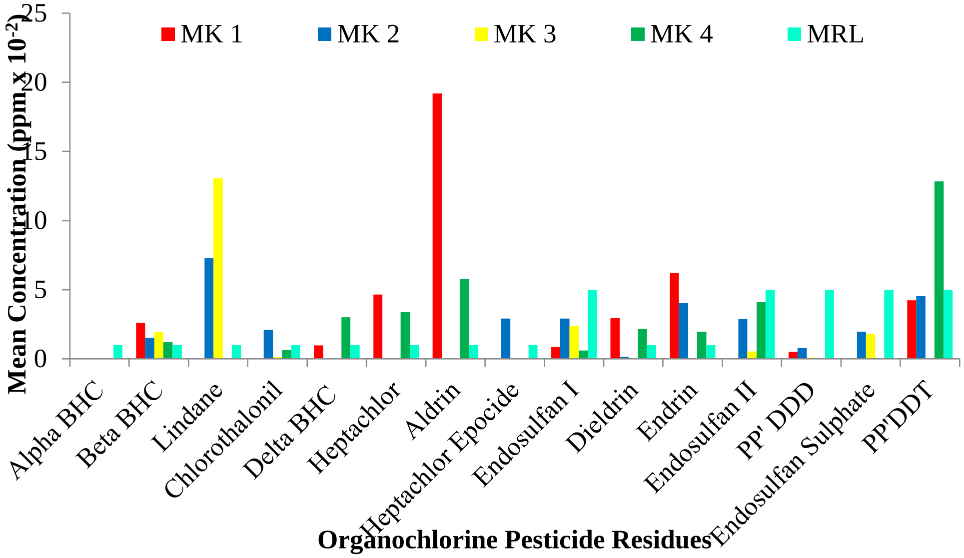

Fig 2: Mean concentrations of organochlorine residues in green amaranth in samples from markets in Akure. MK = Market; MRL = Maximum Residue Limit set by the European Union.

Generally, the levels of organochlorines in the fluted pumpkin can be attributed to the perennial nature of the vegetable; hence, uptake and bioaccumulation can occur over long-term. This becomes worrisome considering the level of consumption of the vegetable across the West and Central Africa (Alegbejo, 2012) and it is even taken fresh to cure some ailments such as convulsion (Gbile, 1986). The leaf alone is useful in the management of liver problems, and impaired immune system (Eseyin *et al.,* 2005; Adaramoye, 2007). Fresh leaves are ground and the juice used as tonic for the treatment of anaemia, chronic fatigue, and diabetes (Aderibigbe *et al.,* 1999; Dina *et al.,* 2006; Alada, 2010). The high concentration of organochlorines detected in the fluted pumpkin sampled, is consistent with the findings of Bempah *et al.* (2011) where aldrin, dieldrin, endrin and pp'-DDT residues were detected in edible vegetables in Ghana. Also, lindane, endosulfan, aldrin and endrin were detected in various vegetable species from market gardens across Togo (Madjouma *et al.,* 2012). In Nasarawa State, fluted pumpkins, spinach and sorrel leaves were assessed for organochlorine residues, and it was found that fluted pumpkins contained high levels of pp'-DDT and endrin (Ibrahim *et al.,* 2018). Organochlorine residues were also reported in vegetables other than leafy ones by Nsikak and Aruwajoye (2011), though at low levels but with the concern of public safety in view of possible bioaccumulation in the long-term. Detection of organochlorine in the vegetables can be adduced to past (and possible present) use on cash crops such as cocoa, kolanut and palm trees, that were previously planted on the farms now used for vegetable farming and even in some vegetable farms the cash crops are still planted along with the vegetables. The persistent ability of organochlorine insecticides makes the soil a source of continuous crop contamination until the total absorption or breakdown. There is also the possibility of illegal use of these organochlorines on the farms. The high concentrations of lindane in the fluted pumpkin is of particular concern, considering the health impact. They are associated with risk of liver and breast cancer, endocrine destruction especially in males; they also impact on human immune system (ASTDR, 2005). Endosulfan either as isomers (Endosulfan I and Endosulfan II) or metabolite (Endosulfan Sulphate) was detected in levels higher than the MRL across the four markets. It is known to cause birth defects in male reproductive systems, and it is an endocrine disruptor, linked to Autism in children (PAN, 2013). The higher levels of pp'-DDT over its metabolites (DDD) in fluted pumpkin suggest recent usage of DDT on the farms. DDT persists in the environment with gradual degradation to DDD and DDE (dichloro diphenyl dichoroethylene) under aerobic condition (Klumpp *et al.,* 2002). DDT has been highlighted as a suspected human carcinogen by USEPA, European Union and International Agency for Research on

Cancer (IARC). The pyrethroid levels were remarkably high, particularly for permethrin, as it was detected in fluted pumpkin samples collected from all the markets (Table 5). The concentration of the permethrin detected ranged from 38.61 x 10^2 ppm to 113.02 x 10^2 ppm which was far above the MRL (5.00 x 10^2 ppm). The concentration of Lambda-cyhalothrin, on the other hand, was below the MRL (100 x 10^2 ppm) in all the markets except in MK 1 with concentrations doubling the MRL; 211.900 x 10^2 ppm (Table 5).

Table 5: Mean concentrations of pyrethroid residues in Fluted Pumpkin leaves from markets in Akure, Nigeria.

Samples	**Pyrethroid (ppm x 10^{-2})**	
	Lambda-Cyhalothrin	**Permethrin**
MK 1	211.900 ± 2.04^{a}	113.020 ± 2.33^{a}
MK 2	7.220 ± 1.12^{b}	67.820 ± 1.92^{b}
MK 3	3.840 ± 0.37^{bc}	93.200 ± 2.33^{ab}
MK 4	1.880 ± 0.23^{c}	38.610 ± 1.13^{c}
MRL (EU)	100	5

Mean followed by the same letter along the column are not significantly different (p>0.05) using Tukey's Test. MK = Market; MRL = Maximum Residue Limit set by the European Union.

Similar to the fluted pumpkin leaves, the concentration of permethrin detected in green amaranth collected from MK 1, 2 and 4 ranged from 19.30 x 10^2 ppm to 23.07 x 10^2 ppm, which were higher than the MRL (Table 6). However, lambda-cyhalothrin was detected below the MRL in all four markets Table 6), which is in contrast with the findings of Fan *et al.* (2013). They reported concentrations of lambda-cyhalothrin higher than the MRL in green amaranth in China, this may be as a result of choice of pesticides in use by vegetable farmers in China.

Table 6: Mean concentrations of pyrethroid residues in Green Amaranth leaves from markets in Akure, Nigeria.

Samples	**Pyrethroid (ppm x 10^{-2})**	
	Lambda-Cyhalothrin	**Permethrin**
MK 1	27.280 ± 1.22^{a}	19.390 ± 1.72^{a}
MK 2	14.140 ± 1.63^{b}	20.180 ± 0.44^{a}
MK 3	6.70 ± 0.98^{ab}	3.590 ± 0.11^{b}
MK 4	74.750 ± 1.66^{c}	23.070 ± 1.78
MRL (EU)	100	5

Mean followed by the same letter along the column are not significantly different (p>0.05) using Tukey's Test. MK = Market; MRL = Maximum Residue Limit set by the European Union

Pyrethroids, characteristically have a wide spectrum of activity, with high biodegradability, low mammalian and avian toxicity while also demonstrating strong selectivity for insects and invertebrates (Ecobichon, 1996; Fishel, 2005). The effectiveness of pyrethroid may be due to its peculiar sorption to particulates which reduces bioavailability to non-target organisms while ensuring its concentration in the area of direct application. Pyrethroids are linked to disruption of the endocrine system which can affect reproduction and sexual development.

Permethrin levels were very high (far above the MRL) in the fluted pumpkin leaf samples studied. In contrast, lower levels were reported in vegetables collected in Ghana (Bempah *et al.*, 2011) and in other edible parts of plants (Stone *et al.*, 2009). The high concentration of permethrin in the vegetable sampled could be due to its indiscriminate use by Akure farmers. From the study, organochlorine compounds were the most detected pesticide residues while herbicides were the least detected in the Fluted Pumpkins and Green Amaranth, i.e., organochlorines > organophosphates > pyrethroids > herbicides (see Figures 3 and 4).

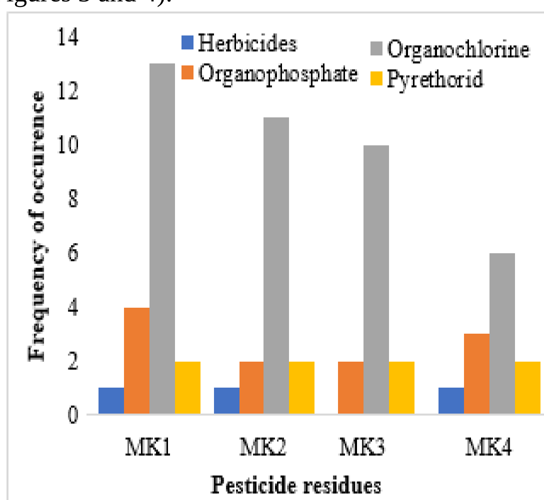

Fig 3: Incidence of Pesticide (herbicides and insecticides) residues in fluted pumpkin leaves from selected markets in Akure. MK = Market

Of all the four markets sampled, MK 1, 2 and 4 appeared to have more vegetables contaminated with organochlorines while MK 3 have more vegetables contaminated with organophosphates and pyrethroids. This implies that most of the vegetables sold in MK 1, 2 and 4 might be from vegetable farms which were previously used for cash crops where organochlorines has been used to control insect pests.

On the other hand, vegetables in MK 3 might be from more recent lands used for farms treated with organophosphates and pyrethroids to manage pests. The wide range of level of detected pesticides observed in this study is due to the diverse source of the vegetables sold in the Akure markets. Some are from peri-urban farms while some are from far distanced villages.

In a similar pesticide residue study in Southeast Nigeria on vegetables by Omeje *et al.* (2021) which included fluted pumpkin and green amaranth, they observed that although the residues detected fell below the MRLs, the vegetables contained diverse pesticides such as aldrin, carbofuran, g-chlordane, chlorpyrifos, dichlorobiphenyl, dichlorvos, endosulfan, heptachlor, hexachlorobenzene (HCB), isopropylamine, lidane, t-nonachlor and profenofos. Likewise, in another recent study by Omeje *et al.* (2022), they observed the presence of violating residues of organochlorines and organophosphates in some fresh fruits (apple, carrot and cucumber) and vegetables (cabbage and green beans) sold in a market in Lagos and two markets in a Southeastern state in Nigeria.

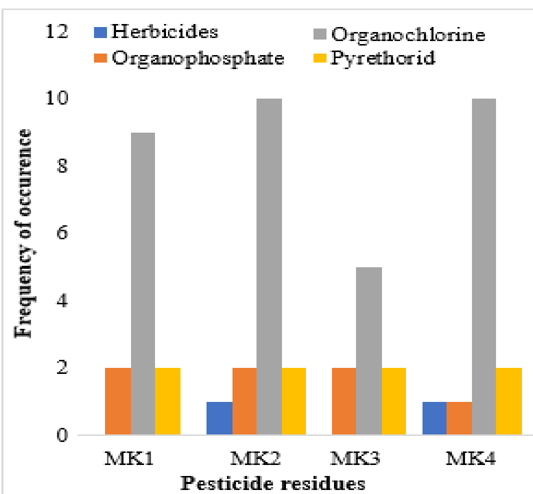

Fig 4: Incidence of Pesticide (herbicides and insecticides) residues in green amaranth leaves from selected markets in Akure. MK = Market

Conclusions: The assessment of the type and concentration of pesticide residues in fluted pumpkins and green amaranth leaves sold in Akure markets, Ondo State, Nigeria, revealed the presence of various chemical groups such as herbicides, organophosphates, organochlorines and pyrethroids at levels that exceed the maximum residue limits set by the European Union. The widespread usage of pesticides, their stability and tendency to bioaccumulate make them particularly dangerous when consumed by humans. Of greater concern is the exposure of the vegetables to multiple pesticide residues as research reveals that ingesting multiple pesticide residues even below the arbitrary 'safe' limit still portends grave health consequences due to synergistic toxicity. Therefore, there should be regular monitoring of the leafy vegetables sold across Nigerian markets by designated authorities. Government policies on the controlled use of pesticides in agricultural products need to be strictly enforced while offenders are adequately punished to serve as deterrent to others.

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Data Availability Statement: Data are available upon request from the first author or corresponding author or any of the other authors.

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