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Assessment of Organochlorine Pesticide Residue Levels in Fruits and Vegetables Sold in Federal Capital Territory (FCT), Nigeria

K.C. Igwegbe [a*], V.I. Onwukeme [b*] and A.P. Okeke [b]

[a] Food Safety and Applied Nutrition (FSAN), National Agency for Food and Drug Administration and Control (NAFDAC), FCT, Abuja, Nigeria.
[b] Department of Pure and Industrial Chemistry, Faculty of Physical Sciences, Nnamdi Azikiwe University, Awka, Anambra State, Nigeria.

Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

The study evaluated the organochlorine pesticide residue levels in fruits and vegetables sold in Federal Capital Territory, Abuja. Six samples were bought from eleven major markets across the six Area Councils of FCT, Abuja. The samples were mixed to form composite groups of the fruits and vegetables, and prepared using QuEChERS method. It was analysed using Agilent 7890 Gas Chromatography equipped with a micro-cell Electron Capture Detector (μECD). The analysis revealed that the hazard index (HI) for all the fruits and vegetables studied were well below the levels of adverse health effect for chronic exposure except for Onion in children which was 1.2287.

**Corresponding author: Email: valentineifenna@yahoo.com, kelvin.igwegbe@nafdac.gov.ng;*

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The HI for the various fruits and vegetables in children were higher than those for the adult. For example, the HI for Tomato, Green Amaranth Leaves and Pepper all in children were 0.4485, 0.4411 and 0.3981 respectively while the HI for Tomato, Green Amaranth Leaves and Pepper all in adult were 0.1121, 0.1103 and 0.0995 respectively. The calculated cancer risk associated with the consumption of these fruits and vegetable showed values well below the upper bound of $1.0x10^{-6}$ except for Aldrin, Dieldrin, Heptachlor, Heptachlor Epoxide. a-BHC, b-BHC, and d-BHC especially in children. From the hazard index and cancer risk analysis, Aldrin and Dieldrin have been implicated as the pesticides of concern since they were present in reasonable concentrations for almost all the samples and age groups studied. The highest potential of cancer risk was found in Onions for children. Also, the total cancer risk implicated Onions (with a value of $1.2x10^{-4}$ for adult and $4.7x10^{-4}$ for children) and Pepper (with a value of $5.5x10^{-5}$ for adult and $2.2x10^{-4}$ for children) as the most contaminated foodstuffs with pesticide residues in the studied area. The order of the total cancer risk from the study was Onions > Pepper > Tomato > Green Amaranth Leaves > Fluted Pumpkin Leaves > Okra for both adult and children. In conclusion it was observed that pesticide residues of Endrin, o,p'-DDE and Heptachlor in Onions, Green Amaranth Leaves and Pepper were above the Codex maximum residue level (MRL). Endrin in Onions had 858.0%, o, p'-DDE in Onions had 276.0%, o, p'-DDE in Green Amaranth Leaves had 100.8%, Heptachlor in Pepper had 168.4% and Heptachlor in Onions had 119.2%. Also, the hazard index of chronic exposure and cancer risk of children for Onions requires urgent attention as their values were well above the acceptable limit. Therefore, periodic monitoring of pesticides residues in these fruits and vegetables cannot be over emphasized, but will go a long way to prevent, control and reduce environmental pollution and health risks. Also, taking precautionary measures like proper cooking before consumption of these foodstuffs is advised.

Keywords: Pesticide; pesticide residue; organochlorine; QuEChERS method; maximum residue level; hazard quotient/index/indices; cancer risk.

1. INTRODUCTION

Fruits and vegetables are commonly grown with pesticides to prevent and control pest infestation. Preventing and controlling pest infestation in the farm leads to higher yield [1]. Pesticides are also used to preserve or transport harvested crops during storage and handling. Therefore, contamination with pesticides may occur at any point from farm to fork [2,3]. The remains or residues of these pesticides in food become an analytical concern and an important threat to life because some are highly persistent to environmental degradation. Pesticide exposure may also lead to wide array of health problems such as cancer, birth defect, neural and kidney damage, immune suppression, diminished intelligence, hormone disruption, reproductive abnormalities, congenital disabilities, etc. [4]. Therefore, this necessitates the identification and quantification of pesticide residues in agricultural products such as fruits and vegetables. It is a very important concern to both developed and developing countries. For example, in 2015 European Union (EU) issued a ban on the importation of beans from Nigeria into Europe due to their high residue level (0.03mg/kg to 4.6mg/kg) of Dichlorvos [5]. This study will provide experimental data, dietary survey and statistical analysis on some pesticides residues in selected fruits and vegetables which will help enhance better regulation of pesticide use in Nigeria.

2. METERIALS AND METHODS

2.1 Study Area

The experimental samples were bought from eleven major markets in FCT, *viz.* Abaji, Bwari, Dutse-Alhaji, Garki, Gwagwalada, Kuje, Kwali, Utako, Nyanya, Wuse, and Zuba, while the control samples which were grown without pesticides spray were collected from a personal farm in Garki. The various markets were drawn from all the six area councils of the FCT. Each market was divided into four quadrants and the same cost of samples were bought from each quadrant. The samples were sorted and mixed to form composite samples of each type *i.e* tomatoes from one market were mixed with those from another market. Thus, six composite samples were formed representing the markets in FCT, *viz.* Tomato (*Solanum lycopersicum*), Pepper (*Capsicum spp*), Okra (*Abelmoschus esculentus*), Onion (*Allium cepa*), Fluted Pumpkin Leaves (*Telfairia occidentalis*) and Green Amaranth Leaves (*Amaranthus spp*).

FCT is the capital city of Nigeria, which is located between latitude of 8.25^{o} N - 9.20^{o} N, and

longitude of 6.45°E - 7.39° E. It is located north of the confluence of the Niger River and Benue River. It was created in 1976 and made the Federal Capital Territory in 1991. FCT is bordered by Niger, Kaduna, Nasarawa and Kogi states.

2.2 Sample Pre-Treatment and Preparation

The samples were processed and prepared using QuEChERS method [6]. Dirt and particles were removed, and they were washed and rinsed with running water. The composite samples were grinded separately using dry ice in Stephan Electronic 2010 Blender to control temperature and spill of the liquid content (cryogenic milling). This process is used to increase homogeneity of the samples or to reduce the sub-sampling variation and to enhance the extraction of analytes. The grinded samples were packed separately in a clean disposable closed plastic plates and stored in the fridge at about 4°C until required for extraction.

2.3 Sample Extraction

Sample extraction were carried out using QuEChERS method [6]. 10 g of each sample was weighed into different 50mL centrifuge tubes and labelled accordingly. 10 mL of acetonitrile was added to each, followed by 0.1 mL of 50 ppm PF-38 and the mixture homogenized with a vortex mixer. Extraction salt were added to each and the mixture homogenized again with a vortex mixer. They were then centrifuged at 3000 rpm at 0°C for 5 minutes and 6 mL of the upper layers were decanted into different 14 mL centrifuge tubes and labelled accordingly. Clean up salt were added to each decanted upper layer and the mixture homogenized with a vortex mixer. They were then centrifuged again at 3000 rpm at 0°C for 5 minutes and the supernatants decanted into different test tubes and labelled accordingly. 50% Formic Acid was added for each as buffer solution at the rate of 10 µL per mL followed by 0.125 mL of 20 ppm Triphenylphosphate (TPP) as an internal standard and the mixture homogenized. 1 mL of each solution was transferred into different test tubes and labelled accordingly. It was then placed into the Turbo Vap LV for drying. After drying, each test tube was reconstituted or made up to the volume with n-hexane:acetone (4:1, v/v). The mixture was then homogenized and transferred to vials for instrumentation.

2.4 Instrumentation

The instrumentation was done with an Agilent 7890 Gas Chromatography equipped with a micro-cell Electron Capture Detector (µECD) and a 7683B auto sampler in accordance with Agilent Advanced User Guide [7]. The samples were injected on splitless mode (60 mL/min, 0.75min) using an injector equipped with a 10 µL syringe and a 4 mm i.d. tap GW liner. The separation of pesticides was carried out with a HP-5 capillary column (30 m length, 0.32mm i.d., 0.25 µm

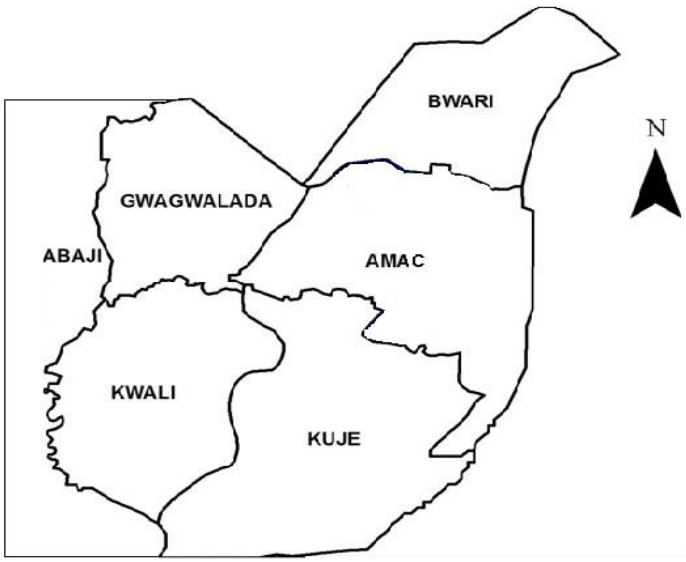

Fig. 1. Map of FCT showing the area councils

film thickness) containing a 5% phenyl methyl siloxane as stationary phase. The GC-ECD operating conditions were as follows: injector temperature was 250°C; detector temperature was 310°C; initial oven temperature was 100°C for 1 min, then raised at 10°C/min to 200°C, held at 200°C for 2 min, then raised at 10°C/min to 300°C, and held at 300°C for 1 min. The run time was 24 min per sample. Nitrogen was used as a carrier gas at a flow rate of 4 mL/min.

In the GC-ECD, the gas chromatography column separates the sample into its individual components or analytes, which are then passed through the electron capture detector. The detector uses a radioactive beta particle emitter and a makeup gas like Nitrogen or Methane. The radioactive isotope releases electrons that collide with the makeup gas, causing more electrons to be released. This creates a current, which is measured by the detector. Different chemical species will absorb the free electrons in different amounts, causing a drop in the current. The observed drop can be used to determine which compounds are present, and in what concentrations. By this the different types of pesticides present in the fruits and vegetables can be identified and quantified. The data produced by the instrument for each sample were in "relative %"/10g of the sample analyzed and thereafter converted to mg/kg.

2.5 Calibration and Method Development

The instrument calibration and method development was done using analytical protocol based on application of QuEChERS technique [8]. The pesticide standards were run singly to get their retention time using the highest concentration (*i.e.* 0.5ppm). Thereafter, the calibration curve of each pesticide of interest was derived by running serially diluted or calibrated standard solutions of 0.01, 0.05, 0.10, 0.25, 0.50ppm. The linearity of calibration curve (R^2) was determined by plotting the concentration versus the peak area. The limit of detection (LOD) and limit of quantification (LOQ) were evaluated.

2.6 Quality Assurance and Control

All analytical procedures were monitored using strict quality assurance and control measures in accordance with QuEChERS method [6]. Blender and all materials used for preparation of samples were well-washed and rinsed with acetone before reuse. Chemicals used in the sample preparation and analyses were of spectra grade. The use of internal standard (*i.e.* PF-38 and Triphenylphosphate), blank determination, percent recovery determination, limit of detection and limit of quantification were also carried out for quality control and assurance. Blank analyses were carried out in order to check for any interference or background value of pesticides contracted during the bench work. Recovery analysis was performed to evaluate the precision and efficiency of the analytical procedures using standard addition method. The recoveries were determined by comparing the peak areas of the pesticides after spiking with those un-spiked and percentage recoveries were calculated based on proportion of concentrations of the analytes detected from the spiked samples.

2.7 Health Risk Assessment

This involves non-carcinogenic risk and carcinogenic risk assessment. The non-carcinogenic risk assessment involved the calculation of the estimated daily dose, hazard quotient and hazard indices of the different fruits and vegetables for adult and children. In this study, health risk assessment model was derived from USEPA IRIS [9] guideline and applied to estimate the carcinogenic and non-carcinogenic risks for adults and children [10]. The estimated daily doses of the fruits and vegetables for adult and children were calculated using Equation 3 while the hazard quotients (HQ) was calculated using Equation 4. The hazard index (HI) of the fruits and vegetables were calculated as the sum of the hazard quotients of the various pesticides while the cancer risks were calculated using Equation 5.

$$EDI = \frac{C_i \,.\, FCR \,.\, F \,.\, ED}{B_W \,.\, AT} \quad (1)$$

For daily exposure to these pesticides the Exposure Factor (EF) is equal to 1

$$EF = \frac{(F \,.\, ED)}{AT} = 1 \quad (2)$$

Thus,

$$Adjusted\ EDI = \frac{C_p . FCR}{B_W} \quad (3)$$

Where;

EDI = Estimated Daily Intake (in mg/kg-day)
C_i = Concentration of Pesticide Residues (in mg/kg, from the experimental result)

FCR = Food Consumption Rate (in kg/day) (see Appendix 1)
F = Exposure Frequency (in days/year)
ED = Exposure Duration (in years)
B_w = Body Weight (60 kg for adult and 15 kg for children)
AT = Average Time (in days)

$$HQ = \frac{EDI_i}{RfD_i} \quad (4)$$

Where;

RfD_i = Reference Dose (in mg/kg-day)

$$Cancer\ Risk = C_i.\frac{FCR_i.F_i.ED_i.CF}{B_W.AT}.\mathrm{SF}_i \quad (5)$$

Where;

CF = Conversion Factor as 1.0 x 10^{-6} (in kg/mg)
SF_i = Oral Slope Factor (in mg^{-1}kg-day)

Food Consumption Rate (FCR) is calculated from a survey data recovered from the study area while Estimated Daily Intake (EDI), Hazard Quotient (HQ) and Cancer Risk were calculated with reference to the standard assumption of adults and children having an average body weights of 60 kg and 15 kg respectively. Cancer risk of 1.0 x 10^{-6} implies that 1 in 1,000,000 persons has the lifetime risk of developing cancer through the pathway (of ingestion only). The maximum risk value or threshold is 1.0 x 10^{-6} but values greater than that is unacceptable.

3. RESULTS AND DISCUSSION

3.1 Results

Table 1. Organochlorine Pesticide Residues found in the fruits and vegetables under review

S/N	Pesticides	Tomato (mg/kg)	Pepper (mg/kg)	Okra (mg/kg)	Onion (mg/kg)	Fluted Pumpkin (mg/kg)	Green Amaranthus (mg/kg)
1	Aldrin	0.0140	0.0310	0.0095	0.0439	0.0019	0.0140
2	a-BHC	0.0064	0.0161	0.0025	0.0368	0.0802	0.0227
3	b-BHC	0.0164	0.0475	0.0000	0.0147	0.0003	0.0017
4	d-BHC	0.0157	0.0084	0.0070	0.0660	-	0.0075
5	Chlorothalonil	0.0097	0.0000	0.0000	0.0334	0.0000	0.0000
6	o, p' - DDE	-	0.0105	0.0091	0.0276	-	0.0504
7	p, p' - DDE	0.0014	0.0440	-	0.0096	0.0087	-
8	Dieldrin	0.0043	0.0401	-	0.0292	0.0000	0.0025
9	Endosulfan I	0.0002	0.0799	0.0002	0.0292	0.0043	0.0026
10	Endosulfan II	-	0.0332	-	0.0129	-	0.0082
11	Endrin	-	0.0072	-	0.0858	-	-
12	Heptachlor	0.0164	0.0842	0.0254	0.0596	0.0221	0.0430
13	Heptachlor Epoxide	0.0009	0.0058	-	0.0346	-	0.0311
14	Lambda Cyhalothrin	-	0.0823	-	0.0287	-	-
15	Lindane	0.0018	0.0066	0.0037	0.0256	0.0008	0.0037
16	PCB-153	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

3.2 Codex Maximum Residue Level (MRL)

Table 2. Codex MRLs of Pesticide Residues under review

S/N	Pesticides	Tomato (mg/kg)	Pepper (mg/kg)	Okra (mg/kg)	Onion (mg/kg)	Fluted Pumpkin (mg/kg)	Green Amaranthus (mg/kg)
1	Aldrin	0.10	0.10	0.10	0.05	0.05	0.05
2	a-BHC	2.00	1.00	1.00	1.00	2.00	2.00
3	b-BHC	2.00	1.00	1.00	1.00	2.00	2.00
4	d-BHC	2.00	1.00	1.00	1.00	2.00	2.00
5	Chlorothalonil	5.00	7.00	-	1.50	-	-

S/N	Pesticides	Tomato (mg/kg)	Pepper (mg/kg)	Okra (mg/kg)	Onion (mg/kg)	Fluted Pumpkin (mg/kg)	Green Amaranthus (mg/kg)
6	o, p' - DDE	0.05	0.05	0.05	0.01	0.05	0.05
7	p, p' - DDE	0.05	0.05	0.05	0.01	0.05	0.05
8	Dieldrin	0.10	0.10	0.10	0.05	0.05	0.05
9	Endosulfan I	0.50	0.50	0.50	0.20	2.00	2.00
10	Endosulfan II	0.50	0.50	0.50	0.20	2.00	2.00
11	Endrin	0.05	0.05	0.05	0.01	0.01	0.01
12	Heptachlor	0.05	0.05	0.05	0.05	0.05	0.05
13	Heptachlor Epoxide	0.05	0.05	0.05	0.05	0.05	0.05
14	Lambda Cyhalothrin	0.30	0.30	0.20	0.20	0.30	0.30
15	Lindane	2.00	1.00	1.00	1.00	2.00	2.00
16	PCB-153	0.05	0.05	-	-	-	-

Table 3. Reference Dose (RfD) (mg/kg/d) and Oral Slope Factor (SF) (mg-1kgd)

S/N	Pesticides	RfD	SOURCE	SF	SOURCE
1	Aldrin	3.00E-05	USEPA, [13]	1.70E+01	USEPA, [14]
2	a-BHC	5.00E-04	USEPA, [13]	6.30E+00	USEPA, [14]
3	b-BHC	2.00E-04	USEPA, [13]	1.80E+00	USEPA, [14]
4	d-BHC	2.00E-04	USEPA, [13]	1.10E+00	USEPA, [14]
5	Chlorothalonil	1.50E-02	USEPA, [13]	1.70E-02	USEPA, [13]
6	o, p' - DDE	3.00E-03	USEPA, [14]	3.40E-01	USEPA, [10]
7	p, p' - DDE	3.00E-03	USEPA, [14]	3.40E-01	USEPA, [10]
8	Dieldrin	5.00E-05	USEPA, [10]	1.60E+01	USEPA, [10]
9	Endosulfan I	6.00E-03	USEPA, [13]	-	-
10	Endosulfan II	6.00E-03	USEPA, [13]	-	-
11	Endrin	3.00E-04	USEPA, [15]	-	-
12	Heptachlor	5.00E-04	USEPA, [13]	4.50E+00	USEPA, [13]
13	Heptachlor Epoxide	1.30E-05	USEPA, [13]	9.10E+00	USEPA, [13]
14	Lambda Cyhalothrin	5.00E-03	USEPA, [15]	-	-
15	Lindane	3.00E-04	USEPA, [13]	1.30E+00	USEPA, [13]
16	PCB-153	2.00E-05	USEPA, [12]	2.00E+00	USEPA, [12]

3.3 Discussion

Sixteen organochlorine pesticides were the only analytes covered. The samples were collected from only eleven major markets and mixed to form composite samples for the studied area. Thus, the results/findings are average values of the true data expected in the studied area. The health risk assessment considered ingestion pathway as the only means of exposure to these analytes. The primary source of the samples investigated and the points of contamination were not considered. Further studies to assess the source and points of contamination may be considered in other research. The survey instrument (or questionnaire) distributed used a proportional allocation of sample size based on the projected population size from the last population census in 2006. The calibration curves of all the pesticide standards were found to exhibit good linearity, with correlation coefficients (R^2) of more than 0.9979. The limit of detection (LOD) was 0.000001-0.000006 while the limit of quantification (LOQ) was 0.000002-0.000017. The Residual Standard Deviation (RSD) was 0.0001-0.0009%. The percentage recovery (R%) values of the analytes in the spiked samples were in the range of 83-118% for Tomato and 89-117% for Fluted Pumpkin Leaves. The blank determination recorded no peak at all.

From Table 1 and 2, a total of twelve organochlorine pesticides were detected in tomatoes while o, p' – DDE, Endosulfan II, Endrin, and Lambda Cyhalothrin were below detection limit and none of them exceeded the Codex MRL. These represents the organochlorine pesticides the farmers used in cultivation of these crops. For pepper, almost all the sixteen organochlorine pesticides studied were detected and all the analytes were below the Codex MRL except for Heptachlor (168.4%). For Okra, seven organochlorine pesticides were

Table 4. Hazard Quotients (HQ) and Hazard Indices (HI) for samples under review

S/N	Pesticides	Tomato		Pepper		Okra		Onion		F. Leaves	Pumpkin	G. Leaves	Amaranth
		Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children	Adult	Children
1	Aldrin	0.0226	0.0902	0.0310	0.1239	0.0087	0.0350	0.0788	0.3151	0.0016	0.0066	0.0085	0.0338
2	a-BHC	0.0006	0.0025	0.0010	0.0039	0.0001	0.0005	0.0040	0.0159	0.0041	0.0165	0.0008	0.0033
3	b-BHC	0.0040	0.0159	0.0071	0.0285	0.0000	0.0000	0.0040	0.0158	0.0000	0.0002	0.0002	0.0006
4	d-BHC	0.0038	0.0152	0.0013	0.0051	0.0010	0.0039	0.0178	0.0711	-	-	0.0007	0.0027
5	Chlorothalonil	0.0000	0.0001	0.0000	0.0000	0.0000	0.0000	0.0001	0.0005	0.0000	0.0000	0.0000	0.0000
6	o, p' – DDE	-	-	0.0001	0.0004	0.0001	0.0003	0.0005	0.0020	-	-	0.0003	0.0012
7	p, p' – DDE	0.0000	0.0001	0.0004	0.0018	-	-	0.0002	0.0007	0.0001	0.0003	-	-
8	Dieldrin	0.0042	0.0167	0.0241	0.0964	-	-	0.0314	0.1257	0.0000	0.0000	0.0009	0.0037
9	Endosulfan I	0.0000	0.0000	0.0004	0.0016	0.0000	0.0000	0.0003	0.0010	0.0000	0.0001	0.0000	0.0000
10	Endosulfan II	-	-	0.0002	0.0007	-	-	0.0001	0.0005	-	-	0.0000	0.0001
11	Endrin	-	-	0.0007	0.0029	-	-	0.0154	0.0616	-	-	-	-
12	Heptachlor	0.0016	0.0064	0.0051	0.0202	0.0014	0.0056	0.0064	0.0257	0.0011	0.0045	0.0016	0.0063
13	Heptachlor Epoxide	0.0035	0.0138	0.0134	0.0537	-	-	0.1434	0.5735	-	-	0.0434	0.1736
14	Lambda Cyhalothrin	-	-	0.0005	0.0020	-	-	0.0003	0.0012	-	-	-	-
15	Lindane	0.0003	0.0011	0.0007	0.0026	0.0003	0.0014	0.0046	0.0184	0.0001	0.0003	0.0002	0.0009
16	PCB-153	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Hazard Indices		**0.1121**	**0.4485**	**0.0995**	**0.3981**	**0.0120**	**0.0480**	**0.3085**	**1.2341**	**0.0135**	**0.0539**	**0.1103**	**0.4411**

Table 5. Cancer Risk for samples under review

S/N	Pesticides	Tomato		Pepper		Okra		Onion		F. Pumpkin Leaves		G. Amaranth Leaves	
		Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child
1	Aldrin	1.2E-05	4.6E-05	1.6E-05	6.3E-05	4.5E-06	1.8E-05	4.0E-05	1.6E-04	8.4E-07	3.4E-06	4.3E-06	1.7E-05
2	a-BHC	2.0E-06	7.8E-06	3.0E-06	1.2E-05	4.3E-07	1.7E-06	1.2E-05	5.0E-05	1.3E-05	5.2E-05	2.6E-06	1.0E-05
3	b-BHC	1.4E-06	5.7E-06	2.6E-06	1.0E-05	0.E+00	0.E+00	1.4E-06	5.7E-06	1.4E-08	5.6E-08	5.6E-08	2.2E-07
4	d-BHC	8.4E-07	3.3E-06	2.8E-07	1.1E-06	2.1E-07	8.5E-07	3.9E-06	1.6E-05	-	-	1.5E-07	6.0E-07
5	Chlorothalonil	8.0E-09	3.2E-08	0.E+00	0.E+00	0.E+00	0.E+00	3.1E-08	1.2E-07	0.E+00	0.E+00	0.E+00	0.E+00
6	o, p' – DDE	-	-	1.1E-07	4.3E-07	8.6E-08	3.4E-07	5.0E-07	2.0E-06	-	-	3.1E-07	1.2E-06
7	p, p' – DDE	2.3E-08	9.2E-08	4.5E-07	1.8E-06	-	-	1.8E-07	7.0E-07	7.6E-08	3.0E-07	-	-
8	Dieldrin	3.3E-06	1.3E-05	1.9E-05	7.7E-05	-	-	2.5E-05	1.0E-04	5.6E-09	2.3E-08	7.3E-07	2.9E-06
9	Endosulfan I	-	-	-	-	-	-	-	-	-	-	-	-
10	Endosulfan II	-	-	-	-	-	-	-	-	-	-	-	-
11	Endrin	-	-	-	-	-	-	-	-	-	-	-	-
12	Heptachlor	3.6E-06	1.4E-05	1.1E-05	4.5E-05	3.2E-06	1.3E-05	1.4E-05	5.8E-05	2.6E-06	1.0E-05	3.5E-06	1.4E-05
13	Heptachlor Epoxide	4.1E-07	1.6E-06	1.6E-06	6.4E-06	-	-	1.7E-05	6.8E-05	-	-	5.1E-06	2.1E-05
14	Lambda Cyhalothrin	-	-	-	-	-	-	-	-	-	-	-	-
15	Lindane	1.1E-07	4.4E-07	2.6E-07	1.0E-06	1.3E-07	5.3E-07	1.8E-06	7.2E-06	2.5E-08	1.0E-07	8.7E-08	3.5E-07
16	PCB-153	0.E+00	0.E+00	0.E+00	0.E+00	0.E+00	0.E+00	0.E+00	0.E+00	0.E+00	0.E+00	0.E+00	0.E+00
Total Cancer Risk		**2.3E-05**	**9.3E-05**	**5.5E-05**	**2.2E-04**	**8.5E-06**	**3.4E-05**	**1.2E-04**	**4.7E-04**	**1.6E-05**	**6.6E-05**	**1.7E-05**	**6.8E-05**

detected and all were below the Codex MRL. For Onions, all the sixteen organochlorine pesticides were detected and three were above the Codex MRL, *viz.* o, p'–DDE (276.0%), Endrin (858.0%) and Heptachlor (119.2%) and required urgent attention and control. For Fruited Pumpkin Leaves, a total of seven organochlorine pesticides were detected while eleven were detected in Green Amaranth Leaves. None of the analytes were above the Codex MRL except for o, p'-DDE in Green Amaranth Leaves which had 100.8% and calls for concern and further monitoring.

The result of this study revealed that of all the fruits and vegetables investigated, only Pepper, Onions, and Green Amaranth Leaves reported the presence of pesticide residues above Codex MRL. It is pertinent to note that this result agrees with Njoku et al., [11] that reported the presence of Aldrin in all the vegetables purchased from six markets in Lagos state. He also reported that some of the pesticide residues were above the maximum residue levels and can pose carcinogenic and non-carcinogenic health effects.

3.4 Health Risk Assessment

US Environmental Protection Agency (USEPA) Guidelines [12] were used to calculate the non-carcinogenic and carcinogenic hazards. Non-carcinogenic hazards are characterized by hazard index which is the ratio of the estimated daily intake and the reference dose (RfD) of the analyte while carcinogenic hazards are characterized by estimated daily intake, oral slope factor (SF) and conversion factor (CF).

From Table 4, it is observed that the Hazard Quotients for all pesticides residues detected were all satisfactory and the Hazard Index for all the fruits and vegetables studied were well below the level at which adverse health effect for chronic exposure was observed except for Onion in children, which was 1.2287. This calls for concern over prolonged use of these Onion from the area by children. The percentages of chronic reference dose for Heptachlor Epoxide and Aldrin in Onions for children were 57.35 and 31.51 respectively. It was observed that the percentage of chronic reference dose were higher for children than for adults. The result is consistent with that of Adeleye et al., [16] that reported HI > 1 for Aldrin, Dieldrin and Heptachlor Epoxide in Fruited Pumpkin Leaves and Green Amaranth Leaves for adult and children. This result showed that Heptachlor Epoxide, Aldrin and Dieldrin occurred more in the fruits and vegetables studied and as such, their use required strict monitoring.

Table 5 shows that eleven organochlorine pesticides have cancer effect, namely; Aldrin, a-BHC, b-BHC, d-BHC, Chlorothalonil, o,p'–DDE, p,p'–DDE, Dieldrin, Heptachlor, Heptachlor Epoxide and Lindane. It is worthy of note that only twelve out of the sixteen organochlorine pesticides studied had oral slope factors (Table 3). The calculated cancer risk associated with consumption of these fruits and vegetables showed values well below the upper bound of $1.0x10^{-6}$ except for few pesticides. The highest potential of cancer risk was found in Onions for children. Also, the total cancer risk implicated Onions and Pepper as the most contaminated foodstuffs with pesticide residues in the studied area. The result is consistent with that of Adeleye et al., [16] that reported Aldrin and Dieldrin to pose carcinogenic health risks to adult, while Aldrin, Dieldrin, Heptachlor and Heptachlor Epoxide to pose carcinogenic health risks to children. It is pertinent to note that Aldrin and Dieldrin are already banned due to concern about their impact on human health such as increased risk of breast cancer. Aldrin is readily converted to Dieldrin, which is one of the most persistent of all organochlorine pesticides [17]. Also, Heptachlor is readily converted to Heptachlor Epoxide and other products in the environment. Heptachlor Epoxide degrades more slowly and, as a result, it is more persistent than Heptachlor [18].

4. CONCLUSION

From the hazard index and cancer risk analysis, Aldrin and Dieldrin have been implicated as the pesticides of concern since they were present in reasonable concentrations for almost all the samples and groups studied. Other pesticides of interest is Heptachlor and Heptachlor Epoxide detected in Onions and Pepper for adult and children.

Generally, the study showed that children in the studied area has more cancer risk than adult. The highest potential of cancer risk was found in Onions for children. Also, the total cancer risk implicated Onions and Pepper as the most contaminated foodstuffs with pesticide residues in the studied area. The order of the total cancer risk from the study was Onions > Pepper > Tomato > Green Amaranth Leaves > Fluted

Pumpkin Leaves > Okra for both adult and children. Therefore, periodic monitoring of pesticides residues in these fruits and vegetables cannot be over emphasized, but will go a long way to prevent, control and reduce environmental pollution and health risks. Also, taking precautionary measures before consumption of these foodstuffs is advised.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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Appendix 1. Food Consumption Rate (FCR)

The Food Consumption Rate (FCR) for each family (in Naira/day) was calculated from the survey and summed together before being divided by the number of families (N) studied (Equation 6).

$$\text{FCR}\ (Naira/day) = \frac{\text{FCR}_1 + \text{FCR}_2 + \cdots + \text{FCR}_\text{N}}{\text{N}} \quad (6)$$

To convert the Average FCR (in Naira/day) to FCR (in g/day), we used the dry weight (g) of the same cost value (1000 Naira) of the various fruits and vegetables in Equation 7.

$$\text{FCR}\ (\text{g}/day) = \frac{\text{FCR}\ (\text{Naira}/day)\ \text{x Dry weight of 500 Naira value (g)}}{500\ (\text{Naira})} \quad (7)$$

Table 6. Food Consumption Rate (FCR)

S/N	Fruits/Vegetables	FCR (Naira/day)	FCR (g/day)
1	Tomato	63.81	2.91
2	Pepper	36.59	1.80
3	Okra	30.23	1.67
4	Onion	47.34	3.22
5	F. Pumpkin Leaves	40.10	1.54
6	G. Amaranth Leaves	33.67	1.09

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