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HUMAN EXPOSURE TO ORGANOCHLORINE PESTICIDES IN VEGETABLES FROM MAJOR CITIES IN SOUTH-SOUTH NIGERIA

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HIGHLIGHTS

- The vegetables were contaminated with OCPs
- The OCPs in the vegetables were lower than their respective MRLs
- The daily intakes were below their respective FAO/WHO acceptable limits
- The TCR values indicated low to moderate carcinogenic risks
- OCPs in the vegetables originated from both historical and recent usage

RESPONSES TO REVIEWERS COMMENTS

Reviewer #1: Overview and general recommendation:

This work contains useful information on the human exposure of OCPs in vegetables from major cities in Nigeria. The paper needs some revisions as there are a few things to be checked and/or corrected. They are given below:

Specific comments:

- Comment: In the introduction, please more elaborate the reason why the authors only selected four vegetables as the samples in the study, e.g. if most consumed, how many consumption per year. Why these vegetables are so important to be analyzed?
 Response: More elaborate reasons why the authors only selected four vegetables as the samples in the study have been given.
- Comment: Why the samples only collected from four major cities in Nigeria, are they
 most populated or vegetables producer? Please provide the profiles of sampling area.
 Response: Samples were collected from four major cities in the south-south region of
 Nigeria and not Nigeria in general. They are the most populated cities in the region.
 The profiles of sampling area have been included.
- 3. **Comment:** Are the vegetables locally produced? Or are cultivated from other cities. **Responses:** The vegetables are locally produced.
- 4. **Comment:** Page 3. Please provide the sampling period in the method. **Response:** The sampling period has been added in the method section
- 5. **Comment:** Page 10. The authors have tried to identify potential sources of contamination and suggested that several OCPs are from recent usage. Please provide data or reference of OCPs usage in Nigeria, e.g. from other study or exports and imports of OCPs

Response: Data or reference of OCPs usage in Nigeria, e.g. from other study or exports and imports of OCPs has been included in the introduction and source identification section of discussion.

Comment: In the results section, a PCA was performed. Please provide the method of PCA in the method section.
 Personal This has been included

Response: This has been included

Reviewer #3:

The paper presents results on vegetables as a source of persistent pollutant exposure to the consumers in the study area. Although similar studies have been carried out on leafy vegetables in the southwest and Northern Nigeria, the study adds to the information available on presence of pesticide residues in leafy vegetables sampled from Nigeria.

Abstract

Comment: The abstract was well written
Response: Thank you
Comment: In the first few mentions of ng g-1, the Author should indicate that concentrations are fresh weights; ng g-1fresh wt or (fw) or dry weight (dwt).
Response: This has been included
Comment: Other editorial corrections are indicated in the annotated manuscript
Responses: These has been effected

Introduction

Comment: Syntax errors are indicated in the annotated manuscript **Response:** Theses has been effected

Materials & Methods

Sampling and sample pre-treatment

Comment: How significant are the markets that the samples were obtained from? There is no information on this in the study.

Response: The significance of the markets has been added

Comment: In Figure 1, is it possible to add a sample collection map showing the 4 markets used as sample locations in the 4 cities?

Response: This has been added

Sample Preparation and Extraction.

Comment: Line 99-100, The type of extractor used and volume of solvent was not stated. I am surprised that the extraction was carried out for 10 hours.

Response: These were not stated because the detailed procedure have been previously reported.

Comment: QUECHERS (Quick Easy Cheap Effective Rugged Safe) method are highly beneficial analytical approach which simplifies the analysis of multiple pesticide residues in fruit, vegetables and cereals within a shorter time. This is will be good for future work of this nature.

Response: This has been noted by authors. Thank you

Comment: What was the percentage purity of the stream of Nitrogen used for extraction? **Response:** This has been included

Quantification of the extract samples

Comment: The GC-MS mode adopted for the qualitative and quantitative analysis was not stated.

Response: This has been added

Quality Assurance

Comment: Recovery results look good and appear to be well carried out. However, were any certified plant-based reference materials extracted to guarantee the analytical precision. If not, the addition of a certified/standard reference material in a plant tissue would be a good addition to future reports of this nature.

Response: This has been noted by authors. Thank you

Risk assessment

Comment: Page 5, Line 134-135, "In this study, the body weight of 80 Kg for adults and 15 Kg for children were used and ingestion rate = 165 g day-1 was adopted (Tesi et al., 2021)." The reference cited in SM2 is different from what was cited in the manuscript.

Response: This section has been rephrase and revised accordingly

Comment: For the ingestion rate, the author needs to rely on/use sub-national or national survey data or world health organization/FAO data such as cluster diet as used by numerous authors across the world. In reality, children and adults do not consume the same quantity of vegetables daily!

Response: WHO/FAO value has been used to recompute the health risk.

Comment: Bodyweight of 80kg is on the high side for adults, the author needs to check up on other works carried out in your country not necessary from your research group. The average weight for adults in Africa should be around 60-70

(https://stats.areppim.com/stats/stats_weightboysng.htm; https://doi.org/10.1186/1471-2458-12-439)

Response: Body weight of 60 kg has been used to recomputed the health risk

Comment: In the risk assessment, what were the health endpoints for which the authors computed the hazard risk index? Where are these for Cardiovascular? Respiratory? Etc.... The specific health endpoint is highly important in risk assessment and characterization. The authors should search for health endpoints and the target organs of the oral Reference Dose and cancer slope factor used for the risk estimation.

Response: The health endpoints has been included in the manuscript while the target organs of the oral Reference Dose and cancer slope factor used for the risk estimation has been included in the supplemental material SM3.

Comment: The limitation or shortcoming of the assumed values used for the study needs to be stated. For example; the absorption and bioavailability rates of pesticide residues in the children and adults, the age of adults and children, and the concentration used for the analysis (mean concentration, 10 percentile, 90th percentile.....)

Response: The limitation or shortcoming has been added

Data Analysis

Comment: For analysis of variance, were the data tested for normality and equal variance? Were any transformations necessary? Almost all contaminant or toxicology data is left-censored with a skewed distribution best treated by log-transformation (Log (n+1)). **Response:** Data were tested for normality using the Shapiro Wilks and Kolmogorov Smirnov tests and no transformation was done.

Comment: ANOVA can only be carried out when the assumptions are satisfied and if failed, a non-parametric test can be carried out. None of the Table is showing the result from the ANOVA or the means separation. Can the author utilise statistics (either parametric or non-parametric test) to check if differences exist among/between concentrations across the 4 locations as well as the 4 vegetables?

Response: The non-parametric Kruskal Wallis H test was applied to determine significant variation in the concentrations of OCPs in the vegetables since the data were not normal based on the Shapiro Wilks and Kolmogorov Smirnov tests. The results of the Kruskal Wallis H test are shown in supplemental material.

Results Presentation

Comment: In Table 1, DDT, DDD, and DDE should be written as p,p'-DDT, p,p'-DDD, and p,p'-DDE respectively. This correction should be made on all mentions (Figures and Tables inclusive).

Response: This has been effected.

Comment: The classification of the OCP as shown in Tables 1-2 is wrong. Methoxychlor also known as Methoxy-DDT or Dimethoxy-DDT or p,p'-Dimethoxydiphenyltrichloroethane should belong to the DDT group.

Response: This has been effected

Comment: Line 163, the range presented for green leaves is different from the range 7.63 - 54.2 ng g-1 reported in Table 2.

Response: This has been corrected

Comment: Line 168, maximum residue limits (MRLs) stipulated by the European Union for the respective congeners were not presented in any of the Tables.

Response: This has been corrected

Comment: 178-180, the statement "The occurrence pattern of OCPs in the vegetables followed the order Σ HCHs > Σ Chlordanes > Σ Drins> Σ DDTs > Σ Endosulfans for all the sampled cities in southern Nigeria" is not true for all cases. The author should cross-check. For example; in Table 1, the statement is not true for all the cities, Chlordane was highest for Warri (Bitter leaves), Yenagoa (Bitter leaves) and so on.

Response: This has been revised

Comment: In Figure 1, is it possible to add a sample collection map showing the 4 markets used as sample locations in the 4 cities?

Response: This has been added

Comment: In Fig 2, italicise the scientific names and use another design such as a pie chart where the percentage contribution of each class/group can be shown

Response: Pie chart has been used here

Comment: In Fig 3 C-E, the pie chart should be bigger or use other designs to make it easier for the reader to grasp or maintain the current size, remove the caption and include a colour identification code for each of the congeners.

Response: This has been done

Comment: In Fig 4, The authors should reference the benchmark values on the graphs with a horizontal delimitation which will show clearly what is "above" and "below" those thresholds. It would be much nicer to read and easier to see. For example, a line showing the benchmark values (HRI=1) behind the bars.

Response: Benchmark values has been referenced on the graphs with a horizontal delimitation in yellow colour

Comment: There is a need for authors to cross-check the data presented in Tables and Figures with data presented in the result section.

Response: This has been done

DISCUSSION

Comment: Re-writing presenting this study in a longitudinal format, reflecting on other research on vegetables and food products across Nigeria and other countries of the world might be good and show clearly that it's an ongoing problem.

Response: This has been done (See Table 3).

Comment: Page 6, Line 170-176, there is a need to state the mean concentration or range of the OCP reported in the studies. Were their concentrations in wet weight or dry weight? **Response:** This has been added. However, no information was available whether the concentrations were in wet or dry weight

Comment: Page 9, Line 235-247, The possible health effects (both non-carcinogenic or systemic and carcinogenic) due to exposure to the OCP congeners were not stated. How does the risk estimate in this study differs or in similarity to other studies in your country? **Response:** This has been added

Comment: Can the author present a longitudinal review to show current trends or levels of the pesticides in the country's vegetables in comparison with detected levels in this study? **Response:** This has been done (See Table 3)

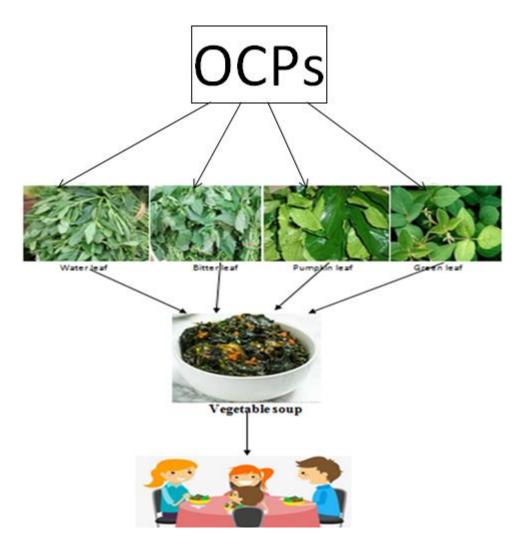
Conclusion

Comment: See annotated manuscript

Response: This has been corrected

References Comment: The references cited in supplementary materials were not listed in the supplementary file. **Response:** The references has been listed

GRAPHICAL ABSTRACT



HUMAN EXPOSURE TO ORGANOCHLORINE PESTICIDES IN VEGETABLES FROM MAJOR CITIES IN SOUTH-SOUTH NIGERIA

3

4 5 **ABSTRACT**

Contamination of vegetables with organochlorine pesticides (OCPs) during cultivation could 6 affect their nutritional value and also results in adverse health effects to consumers. Thus, 7 8 this study evaluates human exposure to OCPs in vegetables from major cities in south-south 9 Nigeria. A total of eighty vegetables consisting of Vernonia amygdalina, Telfairia occidentalis, Desmodium intortum cv and Talinum triangulare obtained from four major 10 cities (Warri, Benin, Yenagoa and Port-Harcourt) in south-south Nigeria were analysed for 11 OCPs using a gas chromatograph coupled with a mass selective detector. The results showed 12 that the mean concentration of $\Sigma 20$ OCPs in the vegetables ranged from 11.6 to 37.7 ng g⁻¹ 13 fresh wt for Vernonia amygdalina, 2.48 to 37.0 ng g⁻¹ fresh wt for Telfairia occidentalis, 14 7.63-54.2 ng g^{-1} fresh wt for *Desmodium intortum cv* and 27.3 to 57.3 ng g^{-1} fresh wt for 15 *Talinum triangulare*. The concentrations of OCPs were generally lower than their respective 16 Maximum Residue Limits (MRLs). The EDI values of the $\Sigma 20$ OCPs in the vegetables 17 ranged from 141 to 464 ng kg⁻¹ bw day⁻¹ for children and 26.5 to 87.0 ng kg⁻¹ bw day⁻¹ for 18 adults. The EDI of Σ Drins, Σ Chlordanes and Σ DDTs for the vegetables were below their 19 respective acceptable EDI values set by FAO/WHO Joint Meeting on Pesticide Residue. The 20 21 hazard index values for children and adults were generally < 1 suggesting that there is no potential non-carcinogenic risk for children and adults consuming the vegetables. The total 22 cancer risk values were above 1×10^{-6} and indicated that children and adults have moderate 23 and low carcinogenic risks respectively from ingestion of these vegetables. The isomeric 24 ratios and principal component analysis result showed that OCPs in these vegetables 25 originated from both historical and recent usage in vegetable cultivation. 26 27

28

31

29 Keywords: Vegetables, OCPs, Daily intake, Risks, Nigeria

30 1.1 Introduction

Vegetables constituted the second most consumed food category in West Africa after cereals 32 (Stadlmayr et al., 2013). Vegetables are bought from farmers or traders and consumed raw or 33 cooked. They provide extra vitamins and minerals to the body and play essential nutritional, 34 35 social and economic roles for consumers (Bolor et al., 2018; Adeleye et al., 2019; Tesi et al., 2021). During cultivation, several pests are attracted to vegetables. To control and manage 36 these pests as well as increase yield, farmers applied pesticides. Approximately, between 37 125,000 and 130,000 metric tonnes of pesticides is used annually in Nigeria (Asogwa and 38 Dongo, 2009). The importation of pesticides rose progressively from \$13 million in 2001 to 39

40 \$28 million in 2003 (FAO, 2005) and this trend has continue till date (PAN, 2007; Oyekunle et al., 2011). For instance, a total of one hundred and forty-seven thousand, four hundred and 41 forty-six (147, 446) tonnes of pesticides comprising five hundred and eighty-four (584) 42 43 tonnes of toxic pesticides were imported into Nigeria for crops production in 2018 (FAO Statistics, 2020). In 2019, Nigeria import \$306 million worth of pesticides while in 2020, 44 Nigeria import \$321 million worth of pesticides resulting in 4.87% import growth 45 46 (https://oec.world/en/profile/hs/pesticides). Organochlorine pesticides are classic persistent organic pollutants extensively applied to prevent and eradicate fungi, insects, weeds, and 47 48 bacterial effects in agriculture (Gereslassie et al., 2019; Tesi et al., 2020). OCPs are of global concern because of their toxicity, persistency, bioaccumulation tendency, ecological and 49 human health effects via the food chain (Yang et al., 2012; Bai et al., 2015). Some health 50 51 effects of OCPs include endocrine disruption, development of cancer cells, immunological 52 and neurological disorders, foetal and reproductive, neuro-toxicological and immunotoxicological disorders; reproductive and foetal defects, enzyme inhibition; 53 54 cryptorchidism, and low sperm concentration (Botella et al., 2004; Barlow, 2005; Yucra et al., 2006; Qin et al., 2019; Tesi et al., 2020; Emoyan et al., 2021). 55

There is currently an increase in the rate of vegetables consumption worldwide (Thompson 56 and Agbugba, 2013; Adetunde et al., 2018; Tesi et al., 2021). Thus, studies assessing the 57 58 healthy state of vegetables with reference to their OCPs contents are needed as residues of 59 OCPs may contaminate and accumulate in vegetables during cultivation. In Nigeria, some studies on OCPs in vegetables have been documented (Benson and Arowajoye, 2011; Akan 60 et al., 2014; Njoku et al., 2017; Adefemi et al., 2018; Ibrahim et al., 2018; Adeleye et al., 61 62 2019). However, none of these studies have evaluated OCPs content in Vernonia amygdalina, Desmodium intortum cv and Talinum triangulare and only two studies have assessed OCPs in 63 64 Telfairia occidentalis. Also, these previous studies were done in northern and south western

65 Nigeria. Vernonia amygdalina, Telfairia occidentalis, Desmodium intortum cv and Talinum triangulare are very popular, readily available, cheap and mostly consumed leafy vegetables 66 by majority of the populace in south-south Nigeria. They are generally cultivated in any open 67 68 land space near homes, in roadsides, farmlands and gardens. Data on OCPs in the above mentioned vegetables are however lacking in south-south region of Nigeria. Thus, the aim of 69 the present study is to assess OCPs concentrations in vegetables, health risks of OCPs 70 associated with consumption of vegetables and sources of OCPs in the vegetables from four 71 major cities in south-south Nigeria. Such data is essential for quality control of food and 72 73 moreover helps consumers to make an informed decision about the choice of foods in order to reduce risk. 74

75

76 **1.2 Materials and Methods**

77 **1.2.1** Study area

The study area comprised four major cities of Warri, Benin, Yenagoa and Port-Harcourt in 78 79 south-south Nigeria (Figure 1). These four major cities are the commercial capitals of Delta, Edo, Bayelsa and Rivers States respectively. These cities are densely populated with 80 population of 943,000 for Warri, 1,841,000 for Benin, 352,285 for Yenagoa and 3,325,000 81 for Port Harcourt in 2022 based on the United Nations - World Population Prospect 82 83 (https://www.macrotrends.net/cities). Four markets from each of the cities were used (Figure 84 1). These markets include Main market (WM1), Okere market (WM2), Igbudu market (WM3) and Polokor market (WM4) from Warri City; Oba market (BM1), Ekiosa market 85 (BM2), New Benin market (BM3) and Ikpoba Hill market BM4) from Benin City; Opolo 86 87 market (YM1), Swali market (YM2), Tombia market (YM3) and Kpasia market (YM4) from Yenagoa City; Oil Mill market (PM1), Mile 1 market (PM2), Abuloma market (PM3) and 88 89 Rumuigbo market (PM4) from Port Harcourt City. These markets are significant because

90 they are the biggest, most visited and located at the centre of these cities. Thus, they were91 selected based on their sizes, location and their popularity.

92

93 1.2.2 Sample collection

Four vegetables were sampled and they include; Vernonia amygdalina, Telfairia occidentalis, 94 Desmodium intortum cv and Talinum triangulare popularly called bitter leaves, pumpkin 95 leaves, green leaves and water leaves respectively. Sample collection was done between 96 October and November, 2021. Five composites of each vegetable were purchased from four 97 98 markets in each of the four major cities giving eighty samples in all. Vegetables were locally cultivated in these cities and also from neighbouring towns. Vegetable samples were wrapped 99 100 with aluminium foil, labeled and transported to the laboratory. Thereafter, the vegetables 101 were rinsed with distilled water, sliced into pieces and kept in refrigerator at 4 °C prior to 102 analysis.

103

104

105

106 **1.2.3 Reagents**

107 The reagents used in this study were of spectra grade and include OCPs standards from Accu
108 Standard, USA, dichloromethane from GFS Chemicals, Columbus, n-hexane from Ultrafine
109 Limited, London, Florisil from Labtech Chemicals, Italy and anhydrous sodium sulfate from
110 Merck, Germany.

111 **1.2.4** Sample extraction and clean up

112 The USEPA method 3550C as earlier described by Tesi *et al.* (2021) was followed for OCPs 113 extraction and cleans up in the vegetables. Briefly, 10 g of the homogenous vegetable sample 114 was weighed into an extraction thimble, and extraction was carried out for 10 hours using dichloromethane/hexane mixture. The extract was concentrated using a rotary evaporator,
cleaned up was carried out on a column packed with acidified silica gel, Florisil, anhydrous
Na₂SO₄ and copper powder. Elution was carried out with 40 mL of DCM/hexane. The eluent
was collected and evaporated to near dryness with under a stream of nitrogen gas (99.99 %
pure) and kept in a vial for analysis.

120 1.2.5 Instrumental analysis

121 The determination of the OCPs in the vegetables was performed using a gas chromatograph 122 (Agilent 6890N) coupled with a mass selective detector (GC–MSD). The injection 123 temperature of 250 °C, detector temperature was 290 °C and column was DB-17 ($30m \times 250$ 124 $\mu m \times 0.25 \mu m$). High purity helium gas with a steady flow rate of 0.8 mL/min was used as 125 the carrier gas. A 1 μ L sample was injected into the GC–MS in splitless mode. Initial oven 126 temperature was 150 °C, increase to 280 °C at 6 °C/min and final temperature was 300 °C. 127 The GC-MSD mode adopted for the analysis was the selective ion mode (SIM).

128 **1.2.6** Quality control and assurance

Spiked recovery technique and blank analysis were used for quality control in this study. 129 Already analysed sample was spiked with standard solution of the OCPs and the spiked 130 sample was analysed. Then the percent recovery was computed. The percent recovery of the 131 132 OCPs ranged from 88.6%–102 %. The relative standard deviation for replicate analyses (n =3) was < 6 %. Analysis of blank samples was done to obtain the limits of detection (LOD) 133 and quantification (LOQ). The LOD was obtained from the concentration of the OCPs that 134 formed a signal/noise ratio of 3, while the LOQ is the concentration of the OCPs that formed 135 a signal/noise ratio of 10. The values of LODs, LOQs, R² and % recoveries of OCPs are 136 shown in Supplemental Material SM1. 137

138

139 1.2.7 Statistical Analysis

Data analyses were completed using the IBM Statistical Product and Service Solutions (IBM SPSS version 25). Descriptive statistics such as mean, standard deviation and range were used. Also, inferential statistics such as the non-parametric Kruskal Wallis H test was applied to determine significant variation in the concentrations of OCPs in the vegetables after the Shapiro Wilks and Kolmogorov Smirnov tests were used to determine the normality of the data. Principal component analysis and isomeric ratios were used to determine the sources of OCPs in the vegetables.

147

148 **1.3** Risk assessment of OCPs in the vegetables

149 **1.3.1 Estimation Daily Intake (EDI)**

150 The EDI of OCPs from consumption of these vegetables was computed from the equation:

151 EDI (ng kg⁻¹ bwday⁻¹) =
$$\frac{OCPs Concentration \times Ingestion Rate}{Body Weight}$$
 (1)

152 In this study, the body weight of 60 Kg for adults and 15 Kg for children were used (Tesi et

153 *al.*, 2021) and ingestion rate = 46.4 g day^{-1} was adopted (WHO, 2012a, b).

154

155 1.3.3 Non-carcinogenic and carcinogenic risks

The OCPs non-carcinogenic risk from ingestion of the vegetables was assessed as hazard index (HI) while the carcinogenic risk was assessed as total cancer risk (TCR) using the equations below (USEPA, 2009):

159
$$HI = \left[\frac{C \times \ln gR \times EF \times ED}{BW \times AT_{nc}} \times 10^{-6}\right] / RfD$$
(2)

160

161
$$TCR = \frac{C \times IngR \times EF \times ED \times SFO}{BW \times ATca} \times 10^{-6}$$
(3)

163 The definition of terms and values for each of the variables in the equations (1) and (2) are given in supplemental material SM2 and SM3 and Tesi *et al.* (2021). The HI value > 1164 suggests that non-cancer risk is present and vice versa while TCR value above 1×10^{-6} 165 suggests that presence of cancer risk is present and vice versa (USEPA, 2010). However, 166 New York State Department of Health (2007) classified TCR values into: very low risk (TCR 167 value $< 10^{-6}$), low risk (TCR value $< 10^{-6}$ but $< 10^{-4}$), moderate risk (TCR value $> 10^{-4}$ but $< 10^{-4}$ bu 168 10⁻³), high risk (TCR value >10⁻³ but 10⁻¹) and very high risk (TCR value $\ge 10^{-1}$). In this 169 study, the health endpoints for which the risks were computed include neurotoxicity, systemic 170 171 toxicity, immunotoxicity, reproductive toxicity and developmental toxicity.

172

173 **1.4 Results and Discussions**

174 1.4.1 Concentration of OCPs in vegetables

The concentrations of OCPs in the vegetables are shown in Tables 1 and 2. There was no 175 significant difference (p > 0.05) in the OCPs concentrations in each vegetable type across the 176 four locations (Tables SM4-SM7) as well as among the different vegetables (Table SM8). 177 However, the mean concentrations of $\Sigma 20$ OCPs in *Vernonia amygdalina* followed the order: 178 Benin > Port-Harcourt > Warri > Yenagoa while the mean concentrations of $\Sigma 20$ OCPs in 179 Telfairia occidentalis followed the order: Port-Harcourt > Benin > Yenagoa > Warri. For 180 Desmodium intortum cv and Talinum triangulare, the order of $\Sigma 20$ OCPs concentrations 181 were: Port-Harcourt > Benin > Yenagoa > Warri and Benin > Yenagoa > Port-Harcourt > 182 Warri respectively (Table 1). The $\sum 20$ OCPs concentrations found in the vegetables varied 183 from 11.6 to 37.7 ng g⁻¹ for Vernonia amygdalina 2.48 to 37.0 ng g⁻¹ for Telfairia 184 *occidentalis*, 7.63-54.2 ng g^{-1} for *Desmodium intortum cv* and 27.3 to 57.3 ng g^{-1} for *Talinum* 185 triangulare. On the average, the concentrations of OCPs in the vegetables obtained in this 186 study followed the trend: Talinum triangulare > Desmodium intortum cv > Vernonia 187

188 amygdalina > Telfairia occidentalis (Table 2). The concentrations of OCPs obtained in this study were generally lower than their respective maximum residue limits (MRLs) stipulated 189 by the European Union (2005; 2012) and World Health Organization (2010). The 190 191 concentrations of OCPs obtained in our study with others reported in literature are shown in Table 3. The OCPs concentrations in vegetables from south-south, Nigeria were comparable 192 to OCPs concentrations reported in some vegetables from Lagos, Nigeria (Oyeyiola et al., 193 194 2017), Ghana (Bolor et al., 2018), Togo (Kolani et al., 2016) and Pakistan (Majeed et al., 2020). However, the concentrations of OCPs obtained in this study were higher than those 195 196 reported by Shoiful et al. (2013) for vegetables in Indonesia but lower than others (Adefemi et al., 2018; Agnandji et al., 2018; Adeleye et al., 2019; Odewale et al., 2021; Olutona et al., 197 2021; Omeje et al., 2021; Suleiman et al., 2021). 198

199 **1.4.2** Profiles of OCPs homologues in vegetables

200 The profiles of OCPs homologues in the vegetables are given in Figures 2 and 3 (a-e). On the average, the occurrence pattern of OCPs in the vegetables followed the order Σ HCHs > 201 Σ Chlordanes > Σ DDTs > Σ Drins > Σ Endosulfans for Vernonia amygdalina; Σ Drins > 202 Σ HCHs > Σ Chlordanes > Σ Endosulfans > Σ DDTs for *Telfairia occidentalis*, Σ Drins > 203 Σ Chlordanes > Σ HCHs > Σ Endosulfans > Σ DDTs for *Desmodium intortum cv* and Σ HCHs 204 $> \sum DDTs > \sum Chlordanes > \sum Drins > \sum Endosulfans for$ *Talinum triangulare*(Figure 2). The205 mean ∑HCHs concentrations ranged from 1.18 to 30.5 ng g⁻¹. T. triangulare from Port-206 207 Harcourt and T. occidentalis from Warri have the maximum and minimum Σ HCHs concentrations respectively. The concentrations of Σ HCHs in these vegetables followed the 208 order Talinum triangulare > Desmodium intortum cv > Vernonia amygdalina > Telfairia 209 210 occidentalis. Among the HCHs γ -HCH was the dominant congener in V. amygdalina and D. *intortum cv* accounting for 41 % and 42 % of the total Σ HCHs respectively while α -HCH 211 212 was the dominant HCH congener in T. occidentalis and T. triangulare constituting 65 % and

32 % respectively of the Σ HCHs (Figure 2a). The mean concentrations of Σ DDTs ranged 213 from not detected to 19.5 ng g⁻¹. The maximum Σ DDTs concentration was found in T. 214 triangulare from Benin whereas Σ DDTs was not found in *T. occidentalis* and T. triangulare 215 from Yenagoa and D. intortum cv from Warri. The concentrations of Σ DDTs in these 216 vegetables followed the order: Talinum triangulare > Vernonia amygdalina > Desmodium 217 *intortum cv* > *Telfairia occidentalis*. Among the DDTs, DDE is the dominant congener. DDE 218 constituted 86 %, 64 %, 100% and 63 % of the ∑DDTs for *V. amygdalina*, *T. occidentalis*, *D*. 219 intortum cv and T. triangulare respectively (Figure 2b). Only DDE was detected in D. 220 221 intortum cv. However, DDD was not detected in V. amygdalina and DDT was not detected in T. occidentalis. The mean concentrations of Σ Chlordanes in the vegetables from southern 222 Nigeria ranged from 0.08 ng g⁻¹ for *Telfairia occidentalis* from Warri to 16.2 ng g⁻¹ for 223 Desmodium intortum cv from Port-Harcourt. The concentrations of Σ Chlordanes in these 224 vegetables followed the order: *Desmodium intortum cv > Talinum triangulare > Vernonia* 225 amygdalina > Telfairia occidentalis. γ -chlordane (45 %), heptachlor epoxide (69 %), 226 heptachlor (52 %) and α -chlordane were the predominant chlordane congeners in Vernonia 227 amygdalina, Telfairia occidentalis, Desmodium intortum cv and Talinum triangulare 228 respectively (Figure 2c). The mean Σ Endosulfan concentrations ranged from not detected to 229 6.77 ng g⁻¹. The ∑endosulfan was not detected in Vernonia amygdalina from Warri and Port-230 Harcourt and *Desmodium intortum cv* from Yenagoa. The concentrations of Σ Endosulfans in 231 these vegetables followed the order: Talinum triangulare > Vernonia amygdalina > 232 Desmodium intortum > Telfairia occidentalis. Endosulfan II was not detected in any of the 233 vegetables in this study while endosulfan sulphate was found only in Telfairia occidentalis 234 from Warri and Desmodium intortum cv from Port-Harcourt. Endosulfan I was the 235 predominant congener among the endosulfans constituting 71 to 100 % of the Σ endosulfans 236 in these vegetables. The mean concentrations of Σ Drins varied between not detected in 237

Vernonia amygdalina from Yenagoa to 19.5 ng g⁻¹ in Desmodium intortum cv from Port-238 Harcourt. The concentrations of Σ Drins in these vegetables followed the order: *Desmodium* 239 intortum cv > Talinum triangulare > Telfairia occidentalis > Vernonia amygdalina. Aldrin 240 was the dominant congener among the Σ Drins in Vernonia amygdalina, Telfairia 241 *occidentalis* constituting 94 % and 49 % respectively of the Σ Drins. Dieldrin was dominant 242 in Desmodium intortum cv accounting for 49 % of the SDrins while endrin constituted 44 % 243 of the \sum Drins in *Talinum triangulare* to be the dominant congener. Endrin aldehyde was 244 found only in Telfairia occidentalis from Benin, Talinum triangulare from Benin and Port-245 246 Harcourt while endrin ketone was found only in Telfairia occidentalis from Warri.

247

248 1.4.3 Estimated Dietary Intake

249 The estimated EDI values of OCPs are displayed in supplemental material SM9. The EDI (in ng kg⁻¹ bw day⁻¹) from ingestion of the vegetables varied between 42.4 and 179, 4.6 to 107, 250 35.9 to 91.4, 8.2 to 45.7, 30.8 to 95.9 and 141 to 464 for Σ HCHs, Σ DDTs, Σ Chlordanes, 251 Σ Endosulfans Σ Drins and Σ 20 OCPs respectively for children ingestion and 8.0 to 33.5, 0.9 252 to 20.0, 6.7 to 17.1, 1.5 to 8.6, 5.8 to 18.0 and 26.5 to 87.0 for SHCHs, SDDTs, 253 Σ Chlordanes, Σ Endosulfans, Σ Drins and Σ 20 OCPs respectively for adults ingestion. The 254 estimated daily intake obtained in this study were lower than the acceptable daily intake of 255 100, 500 and 10,000 ng kg⁻¹ bw day⁻¹ for Σ Drins, Σ Chlordanes and Σ DDTs respectively 256 established by FAO/WHO Joint Meeting on Pesticide Residue (WHO, 2010). 257

258

259 1.4.4 Estimated non-carcinogenic and carcinogenic risks

The HI and TCR values of OCPs in the vegetables are displayed in Figure 4 and supplemental materials SM10 and SM11. The HI values for children and adults consuming the vegetables varied between 0.06 and 2.75 and from 0.01 to 0.69 respectively. The HI

values for children and adults were generally < 1 suggesting that non-cancer risk is absent from consuming these vegetables except for *Vernonia amygdalina*, *Telfairia occidentalis*, and *Desmodium intortum cv* from Port Harcourt for child exposure. The HI values obtained in this study were far lower than the HI values previously reported for OCPs in vegetables (Adefemi *et al.*, 2018; Adeleye *et al.*, 2019). Adefemi *et al.* (2018) and Adeleye *et al.* (2019) reported HI values of 15.8 to 54.1 and 68.916 to 116 respectively for children; and 7.91 to 27.2 and 19.182 to 32.35 for adults respectively.

The TCR values from children and adults consuming the vegetables varied between $5.96 \times$ 270 $10^{\text{-5}}$ and $1.08\times10^{\text{-3}}$ and from $5.42\times10^{\text{-6}}$ to $9.77\times10^{\text{-5}}$ respectively. The TCR values were >271 1.0×10^{-6} indicating that there is cancer risk for children and adults consuming the 272 vegetables. Based on the NYSDOH classification, the TCR values for children fall into the 273 274 moderate risk while those of adults fall into the low risk category. Heptachlor epoxide and aldrin contributed significantly to the HI and TCR values. The possible long term 275 carcinogenic effects of OCPs exposure from consumption of the vegetables include skin, 276 lung, liver, pancreas, prostate, breast, kidney and brain cancers. In this study, the health risk 277 assessment was done following the assumptions that the concentrations of the OCPs remain 278 unchanged during the whole exposure duration. It was also assumed that absorption and 279 bioavailability rates are 100%. Thus, the results of the health risk assessment of OCPs in the 280 281 vegetables should be considered as indicative and not to be interpreted as accurate prediction 282 of observed health outcomes.

283

1.5 Source Identification of OCPs in the vegetables

285 1.5.1 Isomeric ratios

The isomeric ratios of OCPs used in source identification in the vegetables are shown in supplemental material SM12. For *Vernonia amygdalina*, the ratios of heptachlor

288 epoxide/heptachlor and DDT/SDDTs indicated that the OCPs in the vegetables were from historical usage whereas other isomeric ratios indicated that the OCPs were from recent 289 usage. For *Telfairia occidentalis*, the ratios of γ -HCH/ Σ HCHs and β -HCH/ γ -HCH indicated 290 291 that the OCPs are from recent usage whereas the other isomeric ratios indicated that the OCPs were from historical usage. For Desmodium intortum cv, all the isomeric ratios 292 suggested that the OCPs were from recent usage. For *Talinum triangulare*, ratios of α -293 HCH/ γ -HCH and γ -HCH/ Σ HCHs suggested that the OCPs were from recent usage whereas 294 the other isomeric ratios suggested that the OCPs were from historical usage. This result 295 296 implies and also confirmed that OCPs are still being imported and used in farming in Nigeria despite their banned. 297

298

299 1.5.2 Principal Component Analysis (PCA)

300 The PCA result of the OCPs in these vegetables is displayed in supplemental material SM13 and Figure SM1. Two factors were extracted and make up for 73.439 % of the OCPs data set. 301 302 Factor 1 explained 39.743 % of the total variance with positive high loading on HCHs (.847) and endosulfans (.886) and moderate loading on DDTs. The association of these OCP 303 homologues with component 1 indicates that they are from similar source and have related 304 physicochemical properties (Kim and Smith, 2001; Devi et al., 2013). Factor 2 explained 305 306 33.697 % of total variance in the OCPs dataset with high positive loading on chlordanes 307 (.920) and drins (.887). The association of the chlordanes and drins in this component suggested that they originated from similar source(s) in these vegetables. 308

309 **1.6 Conclusion**

The results of this study have shown that the vegetables from south-south Nigeria were contaminated with OCPs but the concentrations of OCPs in the vegetables were below their respective MRLs. The HI values suggested a potential non-carcinogenic health risk for

| 313 | children consumers and no potential risks for adult consumers of the vegetables. The TCR |
|---------------------------------|---|
| 314 | values indicated that children have moderate carcinogenic risk while adults have low |
| 315 | carcinogenic risk from consuming the vegetables. The isomeric ratios and PCA results |
| 316 | showed that OCPs in these vegetables from southern Nigeria originated from both historical |
| 317 | and recent usage in the vegetable cultivation. |
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| 322 | Conflict of Interest |
| 323 | There is no conflicting interest among authors. |
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| | Vernonia amygdalina | | | | | Telfairia occidentalis | | | | Desmodium intortum cv | | | | Talinum triangulare | | | |
|--------------------|---------------------|------------|------------|------------|-----------|------------------------|-----------|------------|-----------|-----------------------|------------|------------|------------|---------------------|------------|------------|----|
| | WR | BN | YG | PH | WR | BN | YG | PH | WR | BN | YG | PH | WR | BN | YG | PH | |
| a-HCH | ND | 0.89± 0.5 | 0.94±0.5 | ND± 0.04 | 0.07±0.05 | 0.80±0.08 | 0.73±0.03 | 8.37±0.05 | 0.59±0.07 | ND | 3.14±0.07 | 2.02±0.05 | 3.57±0.02 | 8.85±0.03 | 8.25±0.04 | ND | 10 |
| 3-HCH | 1.70±0.5 | 7.12± 0.09 | ND | ND± 0.01 | 0.56±0.08 | 0.24±0.38 | ND | ND | 0.69±0.05 | 2.39±0.01 | 1.94±0.03 | 6.27±0.06 | 1.94±0.06 | 2.41±0.09 | 6.37±0.03 | 8.25±1.05 | 10 |
| ү-НСН | ND | 9.67± 1.00 | 2.24± 0.09 | ND± 0.09 | ND | 1.36±0.80 | 0.15±0.01 | ND | 1.76±0.02 | 6.68±0.06 | 2.68±0.08 | 1.96±0.02 | ND | 4.44±0.06 | 8.45±0.09 | 0.03±1.01 | 10 |
| 5-HCH | 1.60±0.09 | ND | ND | 6.19± 0.5 | 0.55±0.6 | 1.07±0.09 | 0.95±0.07 | 0.57±0.5 | 0.26±0.09 | ND | ND | 0.71±0.04 | 2.37±0.09 | 1.50±0.08 | 7.45±0.06 | 0.03±0.06 | 10 |
| ∑HCHs | 2.30±1.20 | 17.68±1.00 | 3.18± 0.90 | 6.19± 1.00 | 1.18±0.80 | 3.47±1.05 | 1.83±0.80 | 8.94±0.40 | 3.30±0.50 | 9.07±0.66 | 7.76±0.80 | 10.96±0.60 | 8.06±0.90 | 17.20±0.93 | 30.52±1.50 | 9.15±0.78 | |
| p,p'-DDE | 2.30±0.6 | 7.09± 0.05 | ND | 0.18± 0.05 | ND | 0.08±0.03 | ND | 0.98±0.60 | ND | 1.00±0.50 | 0.27±0.03 | 0.90±0.04 | 8.34±0.06 | 8.02±0.06 | ND | 8.14±0.08 | 50 |
| o,p'-DDD | ND | ND | ND | ND | 0.07±0.01 | ND | ND | 0.53±0.01 | ND | ND | ND | ND | 2.72±0.09 | 8.25±0.07 | ND | ND | 50 |
| p,p ' -DDT | ND | ND | 1.70± 0.50 | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | 3.27±0.08 | ND | ND | 50 |
| Vethoxychlor | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | |
| ∑DDTś | 2.80±0.55 | 7.09± 0.9 | 1.70± 1.01 | 0.18±0.70 | 0.07±0.90 | 0.08±0.60 | ND | 1.51±0.05 | ND | 1.00±0.11 | 0.27±1.05 | 0.90±1.35 | 11.06±1.04 | 19.25±1.05 | ND | 8.14±1.05 | |
| a-Chlordane | ND | ND | 1.54± 0.02 | 5.24±0.08 | 0.08±0.05 | ND | ND | 2.08±0.04 | ND | 3.06±0.04 | ND | ND | 2.00±1.05 | ND | 1.15±0.06 | 10.24±0.08 | |
| -Chlordane | 4.37± 1.10 | 1.66± 0.9 | ND | 6.16±0.05 | ND | ND | ND | 0.96±0.02 | 0.65±0.2 | 4.22±0.1 | ND | ND | 3.71±0.03 | 0.75±0.06 | ND | 0.61±0.01 | |
| Heptachlor | ND | 0.12± 0.5 | 2.70± 0.90 | 0.93±0.02 | ND | 0.16±0.90 | 0.78±0.06 | ND | 0.72±0.05 | 7.60±0.06 | ND | 8.94±0.07 | 1.56±0.08 | ND | 1.93±0.09 | 0.53±0.05 | 10 |
| Heptachlor epoxide | ND | 2.50± 1.00 | ND | 1.88±0.04 | ND | 1.55±0.03 | 0.54±0.08 | 6.91±0.05 | 0.26±0.06 | ND | 0.51±0.50 | 7.27±1.05 | 0.22±0.04 | 1.71±0.05 | 2.72±0.04 | 2.18±0.04 | 10 |
| ∑Chordanes | 4.37±0.9 | 4.28± 1.50 | 4.24± 0.70 | 14.21±0.50 | 0.08±0.25 | 1.71±1.02 | 1.32±0.55 | 9.95±0.55 | 1.63±0.90 | 14.88±0.89 | 0.51±0.68 | 16.21±0.94 | 7.49±0.88 | 2.46±1.45 | 5.80±0.83 | 13.56±0.92 | |
| Endosulfan I | ND | 6.77± 0.77 | 2.48± 0.6 | ND | ND | 0.91±0.04 | 1.54±0.04 | 0.51±0.08 | 0.87±0.09 | 0.25±0.05 | ND | 1.13±0.07 | 0.68±0.06 | 5.78±0.03 | 6.02±0.08 | 4.15±0.07 | 50 |
| Endosulfan II | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | 50 |
| Endosulfan sulfate | ND | ND | ND | ND | 0.02±0.60 | ND | ND | ND | ND | ND | ND | 5.49±0.08 | ND | ND | ND | ND | 50 |
| ∑Endosulfans | 0± 0.5 | 6.77± 0.9 | 2.48± 0.2 | 0±0.5 | 0.02±1.05 | 0.91±0.5 | 1.54±0.70 | 0.51±.60 | 0.87±1.05 | 0.25±1.05 | 0±0.50 | 6.62±0.99 | 0.68±1.05 | 5.78±1.50 | 6.02±1.04 | 4.15±1.00 | |
| Aldrin | 3.47±1.50 | 1.92± 0.3 | ND | 5.17±0.07 | ND | ND | ND | 8.00±0.03 | 1.83±0.50 | ND | ND | 4.07±0.08 | 0.01±0.02 | ND | ND | 3.43±0.06 | 10 |
| Dieldrin | ND | ND | ND | ND | 1.10±1.00 | ND | ND | 1.50±0.08 | ND | 7.85±0.04 | ND | 9.32±0.50 | ND | ND | ND | ND | 10 |
| Endrin | ND | ND | ND | 0.65±0.4 | ND | ND | ND | 6.60±0.01 | ND | 4.07±0.01 | 1.61±0.09 | 6.13±0.02 | ND | 7.49±0.01 | 0.94±1.05 | ND | 10 |
| Endrin aldehyde | ND | ND | ND | ND | ND | 0.20±0.07 | ND | ND | ND | ND | ND | ND | ND | 4.77±0.08 | ND | 2.50±0.08 | 10 |
| Endrin Ketone | ND | ND | ND | ND | 0.03±0.01 | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | ND | |
| ∑Drins | 3.47±1.40 | 1.92± 0.80 | 0± 0.3 | 5.82±1.10 | 1.13±0.55 | 0.2±1.40 | 0.82±1.03 | 16.1±1.50 | 1.83±1.22 | 11.92±1.30 | 1.61±1.06 | 19.52±1.50 | 0.01±0.03 | 12.26±0.63 | 0.94±1.50 | 5.93±1.36 | |
| ∑20 OCPs | 12.94±1.05 | 37.7± 1.50 | 11.6± 0.80 | 26.40±0.90 | 2.48±1.05 | 6.37±1.11 | 5.51±0.90 | 37.01±0.99 | 7.63±1.05 | 37.12±1.00 | 10.15±1.50 | 54.21±0.99 | 27.30±0.68 | 57.24±1.22 | 43.28±1.04 | 40.93±1.50 | |

 Table 1: Mean OCPs concentrations (ng g⁻¹) in vegetables with respect to locations

WR = Warri; BN = Benin; YG = Yenagoa; PH = Port-Harcourt

| | Vernonia | | Telfairia | | Desmodiu | т | Talinum | | | |
|-----------------------------|-----------------|-----------------|-----------------|-----------|-----------------|------------|-----------------|---------------------------|--|--|
| | amygdalin | <i>a</i> (n=20) | occidental | is (n=20) | intortum c | ev (n=20) | triangular | <i>triangulare</i> (n=20) | | |
| | Mean± SD | RANGE | Mean± SD | RANGE | Mean± SD | RANGE | Mean± SD | RANGE | | |
| α-HCH | 0.46±0.53 | ND-0.94 | 2.49 ± 3.93 | 0.07-8.37 | 1.44 ± 1.42 | ND-3.14 | 5.21 ± 4.15 | ND-8.85 | | |
| β-НСН | 1.96 ± 3.46 | ND-7.12 | 0.2 ± 0.27 | ND-0.56 | 2.82 ± 2.41 | 0.69-6.27 | 4.74 ± 3.07 | 1.94-8.25 | | |
| γ-HCH | 2.98 ± 4.58 | ND-9.67 | 0.38 ± 0.66 | ND-1.36 | 3.27 ± 2.31 | 1.76-6.68 | 3.23 ± 3.05 | ND-8.45 | | |
| σ-НСН | 1.95 ± 2.93 | ND-6.19 | 0.79 ± 0.26 | 0.55-1.07 | 0.24 ± 0.33 | ND-0.71 | 3.05 ± 3.00 | 0.87-7.45 | | |
| ΣHCHs | 7.34±7.09 | 2.3-17.68 | 3.86±3.52 | 1.18-8.94 | 7.77 ± 3.26 | 3.3-11.0 | 16.2 ± 10.4 | 8.06-30.5 | | |
| p,p'-DDE | 2.52±3.31 | ND-7.09 | 0.27 ± 0.48 | ND-0.98 | 0.54 ± 0.49 | ND-1.00 | 6.13 ± 4.09 | ND-8.34 | | |
| p,p'-DDD | ND | ND | 0.15±0.26 | ND-0.53 | ND | ND | 2.74 ± 3.89 | ND-8.25 | | |
| p,p'-DDT | 0.43 ± 0.85 | ND-1.70 | ND | ND | ND | ND | 0.82 ± 1.64 | ND-8.27 | | |
| Methoxychlor | ND | ND | ND | ND | ND | ND | ND | ND | | |
| ΣDDTs | 2.94±2.97 | 0.18-7.08 | 0.42±0.73 | ND-1.51 | 0.54 ± 0.49 | ND-1.00 | 9.69± 8.07 | ND-3.27 | | |
| α -Chlordane | 1.7 ± 2.47 | ND-5.24 | 0.54±1.03 | ND-2.08 | 0.77 ± 1.53 | ND-3.06 | 3.35 ± 4.67 | ND-19.5 | | |
| γ-Chlordane | 3.05 ± 2.75 | ND-6.16 | 0.24 ± 0.48 | ND-0.96 | 1.22 ± 2.02 | ND-4.22 | 1.27 ± 1.66 | ND-10.2 | | |
| Heptachlor | 0.94±1.25 | ND-2.70 | 0.24±0.37 | ND-0.78 | 4.32 ± 4.61 | ND-8.94 | 1.01 ± 0.89 | ND-3.71 | | |
| Heptachlor epoxide | 1.1±1.29 | ND-2.50 | 2.25±3.17 | ND-6.91 | 2.01 ± 3.51 | ND-7.27 | 1.71 ± 1.07 | 0.22-2.72 | | |
| Σ Chordanes | 6.78±4.96 | 4.24-14.2 | 3.27±4.51 | 0.08-9.95 | 8.31± 8.39 | 0.51-16.2 | 7.33 ± 4.65 | 2.46-13.6 | | |
| Endosulfan I | 2.31±3.19 | ND-6.77 | 0.74±0.65 | ND-1.54 | 0.56 ± 0.53 | ND-1.13 | 4.16 ± 2.46 | 0.68-6.02 | | |
| Endosulfan II | ND | ND | ND | ND | ND | ND | ND | ND | | |
| Endosulfan sulfate | ND | ND | 0.01±0.01 | ND-0.02 | 1.37 ± 2.75 | ND-5.49 | ND | ND | | |
| ∑Endosulfans | 2.31±3.19 | ND-6.77 | 0.75±0.64 | 0.02-1.54 | 1.94± 3.14 | ND-6.62 | 4.16 ± 2.46 | 0.68-6.02 | | |
| Aldrin | 2.64 ± 2.20 | ND-5.17 | 2.21±3.88 | ND-8.00 | 1.48 ± 1.93 | ND-4.07 | 0.86 ± 1.71 | ND-3.43 | | |
| Dieldrin | ND | ND | 0.65 ± 0.77 | ND-1.50 | 4.29 ± 4.99 | ND-9.32 | ND | ND | | |
| Endrin | 0.16±0.33 | ND-0.65 | 1.65 ± 3.30 | ND-6.60 | 2.95 ± 2.70 | ND-6.13 | 2.11 ± 3.62 | ND-7.49 | | |
| Endrin aldehyde | ND | ND | 0.05 ± 0.10 | ND-0.20 | ND | ND | 1.82 ± 2.29 | ND-4.77 | | |
| Endrin Ketone | ND | ND | 0.01 ± 0.02 | ND-0.03 | ND | ND | ND | ND | | |
| ∑Drins | 2.8±2.46 | ND-5.82 | 4.56±7.70 | 0.2-16.10 | 8.72 ± 8.66 | 1.61-19.52 | 4.79 ± 5.62 | 0.01-12.3 | | |
| $\overline{\Sigma}$ 20 OCPs | 22.2±12.3 | 11.6-37.7 | 12.8±16.2 | 2.48-37.0 | 27.3 ± 22.4 | 7.63-54.2 | 42.2 ± 12.3 | 27.3-57.2 | | |

 Table 2: Summary statistics of OCPs concentrations (ng g⁻¹) in the vegetables

| Location | Vegetable types | No. of samples | No. of OCPs compounds analysed | ∑HCHs | ∑DDTs | ∑Chlordanes | ∑Endosulfans | ∑Drins | ∑OCPs | References |
|--|--|-------------------|--------------------------------------|--|--|--|--|--|--|-------------------------------|
| South-south, Nigeria | Vernonia amygdalina, Telfairia occidentalis, Desmodium intortum cv and Talinum triangulare | 80 | 20 | 1.18-30.5 | ND-7.08 | 0.08-16.2 | ND-6.77 | ND-19.5 | 2.48-57.3 | This Study |
| Gombe, Northern Nigeria | Tomato, onion, pepper, and chili pepper | 36 | 10 | 242-329 | | 98.0-124 | 214-286 | 257-373 | 257-492 | Suleiman <i>et al.</i> (2021) |
| Nsukka and Enugu, South-east, Nigeria | Fluted pumpkin, <i>Amaranthus</i> leaf, waterleaf, and scent leaf | NA | 9 | <dl-504< td=""><td><dl-221< td=""><td><dl-733< td=""><td><dl-54.8< td=""><td><dl-771< td=""><td>461-1255</td><td>Omeje <i>et al.</i> (2021)</td></dl-771<></td></dl-54.8<></td></dl-733<></td></dl-221<></td></dl-504<> | <dl-221< td=""><td><dl-733< td=""><td><dl-54.8< td=""><td><dl-771< td=""><td>461-1255</td><td>Omeje <i>et al.</i> (2021)</td></dl-771<></td></dl-54.8<></td></dl-733<></td></dl-221<> | <dl-733< td=""><td><dl-54.8< td=""><td><dl-771< td=""><td>461-1255</td><td>Omeje <i>et al.</i> (2021)</td></dl-771<></td></dl-54.8<></td></dl-733<> | <dl-54.8< td=""><td><dl-771< td=""><td>461-1255</td><td>Omeje <i>et al.</i> (2021)</td></dl-771<></td></dl-54.8<> | <dl-771< td=""><td>461-1255</td><td>Omeje <i>et al.</i> (2021)</td></dl-771<> | 461-1255 | Omeje <i>et al.</i> (2021) |
| Nsukka and Enugu, South-east, Nigeria | Okro, cucumber, carrot, and watermelon | NA | 9 | <dl-517< td=""><td><dl-80.8< td=""><td><dl-733< td=""><td><dl-73.3< td=""><td><dl-242< td=""><td>144-1255</td><td>Omeje et al. (2021)</td></dl-242<></td></dl-73.3<></td></dl-733<></td></dl-80.8<></td></dl-517<> | <dl-80.8< td=""><td><dl-733< td=""><td><dl-73.3< td=""><td><dl-242< td=""><td>144-1255</td><td>Omeje et al. (2021)</td></dl-242<></td></dl-73.3<></td></dl-733<></td></dl-80.8<> | <dl-733< td=""><td><dl-73.3< td=""><td><dl-242< td=""><td>144-1255</td><td>Omeje et al. (2021)</td></dl-242<></td></dl-73.3<></td></dl-733<> | <dl-73.3< td=""><td><dl-242< td=""><td>144-1255</td><td>Omeje et al. (2021)</td></dl-242<></td></dl-73.3<> | <dl-242< td=""><td>144-1255</td><td>Omeje et al. (2021)</td></dl-242<> | 144-1255 | Omeje et al. (2021) |
| Iwo, Southwest, Nigeria | Carrot, Onion, Cabbage, Garlic and Ginger | 50 | 17 | <dl-950< td=""><td><dl-4898< td=""><td><dl-2700< td=""><td><dl-4320< td=""><td><dl-44200< td=""><td><dl-14150< td=""><td>Olutona <i>et al.</i> (2021)</td></dl-14150<></td></dl-44200<></td></dl-4320<></td></dl-2700<></td></dl-4898<></td></dl-950<> | <dl-4898< td=""><td><dl-2700< td=""><td><dl-4320< td=""><td><dl-44200< td=""><td><dl-14150< td=""><td>Olutona <i>et al.</i> (2021)</td></dl-14150<></td></dl-44200<></td></dl-4320<></td></dl-2700<></td></dl-4898<> | <dl-2700< td=""><td><dl-4320< td=""><td><dl-44200< td=""><td><dl-14150< td=""><td>Olutona <i>et al.</i> (2021)</td></dl-14150<></td></dl-44200<></td></dl-4320<></td></dl-2700<> | <dl-4320< td=""><td><dl-44200< td=""><td><dl-14150< td=""><td>Olutona <i>et al.</i> (2021)</td></dl-14150<></td></dl-44200<></td></dl-4320<> | <dl-44200< td=""><td><dl-14150< td=""><td>Olutona <i>et al.</i> (2021)</td></dl-14150<></td></dl-44200<> | <dl-14150< td=""><td>Olutona <i>et al.</i> (2021)</td></dl-14150<> | Olutona <i>et al.</i> (2021) |
| Southwest, Nigeria | Carrot, Cucumber, Tomato and watermelon | 144 | 9 | - | - | 903-1368 | 1438-4256 | 1986-4182 | 4329-9508 | Odewale et al. (2021) |
| Lagos, Southwest, Nigeria | Cabbage, cameroun, green and chilli peppers, carrot, lettuce, tomato and scotch honnet | NA | 13 | 1.03-2.88 | 10.75 | 0.73-1.57 | 0.09-0.29 | 0.4-2.06 | 14.8-131 | Oyeyiola et al. (2017) |
| Southwest, Nigeria | Fluted pumpkin and Amaranthus | 32 | 9 | - | - | 103-349 | 788-3449 | 3999-5698 | 5366-9496 | Adeleye et al. (2019) |
| Ekiti Southwest, Nigeria | <i>Senecio biafrae</i> (Wet season) | 8 | 17 | ND-4.0 | 41.0-166 | ND-68.0 | ND-245 | ND-299 | 136-932 | Adefemi et al. (2018) |
| Ekiti Southwest, Nigeria | <i>Senecio biafrae</i> (Dry season) | 8 | 17 | ND-91.0 | 63.0-379 | 29.0-299 | ND-286 | 20.0-246 | 189-908 | Adefemi <i>et al.</i> (2018) |
| Cotonou and Seme-Kpodji, Benin Republic | Lactuca sativa L and Solanum macrocarpum L | 31 | 10 | 66-415 | 44-127 | - | 310-2313 | 5.0-66.0 | 569-2780 | Agnandji <i>et al.</i> (2018) |
| Kumasi, | Cabbage, Lettuce and | 15 | 8 | 0.78-9.54 | 175-190 | 2.29 | 0.60-28.47 | 25.8 | - | Bolor et al. (2018) |

Table 3: Comparison of OCPs concentrations (ng g⁻¹) in vegetables from south-south Nigeria with others reported in literature

| Ghana | Onion | | | | | | | | | |
|-----------|---|-----|----|--------------|--------------|--------------|-----------|--------------|--------------|-------------------------------|
| Togo | Tomato, cabbage and | 130 | 19 | < 0.001-93.8 | < 0.001-1.52 | < 0.001-1.59 | - | < 0.001-0.70 | < 0.001-97.9 | Kolani et al. (2016) |
| | lettuce | | | | | | | | | |
| Indonesia | Carrot, potato, cucumber, corn and | 21 | 21 | < 0.05 | <0.03 | < 0.03 | - | < 0.03 | <0.03 | Shoiful <i>et al</i> . (2013) |
| Pakistan | onions Tomato, brinjal, gourd, okra and spinash | 45 | - | 0.06-0.72 | 0.09-2.53 | - | 0.04-1.17 | - | - | Majeed et al. (2020) |

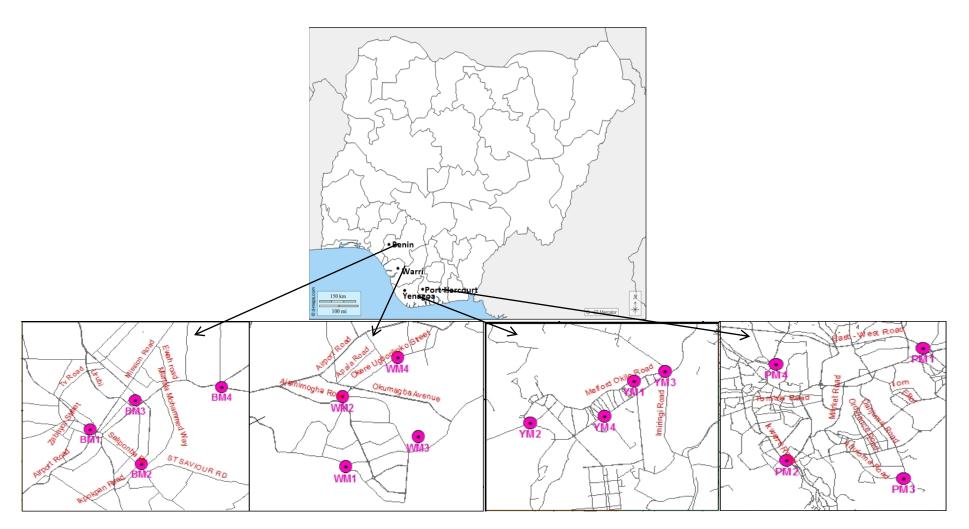


Figure 1: Map of Study area showing the locations of the markets

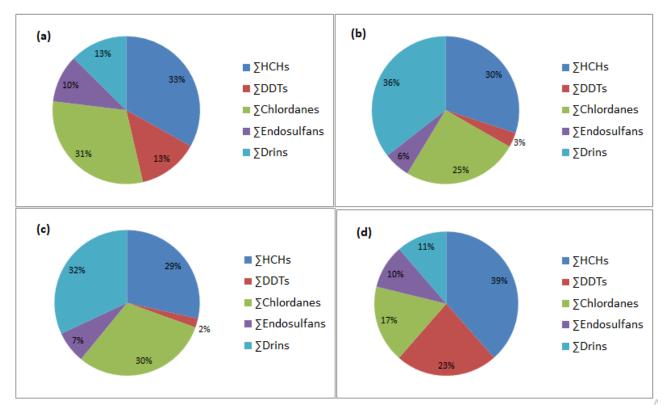


Figure 2: Occurrence pattern of OCPs homologues in the vegetables (a) *Vernonia amygdalina* (b) *Telfairia occidentalis* (c) *Desmodium intortum cv* (d) *Talinum triangulare*

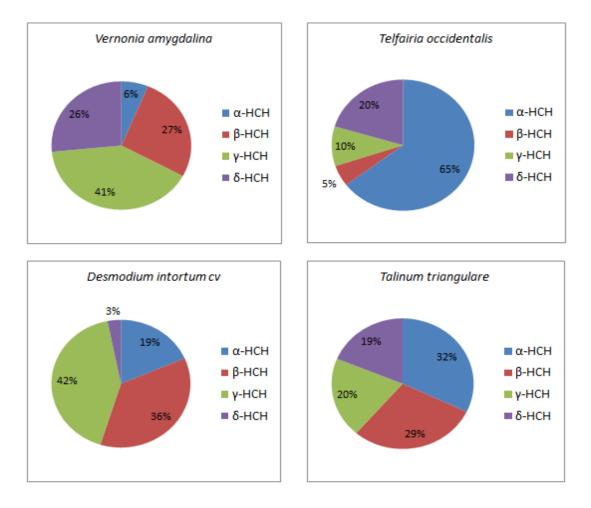


Figure 3a: Percentage composition of HCHs congeners in the vegetables

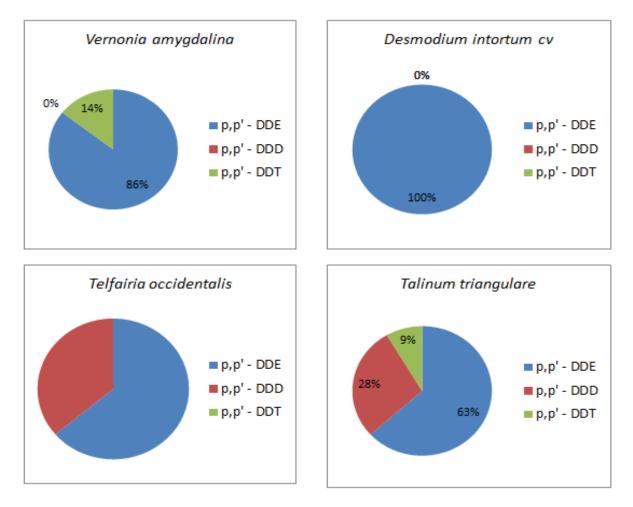
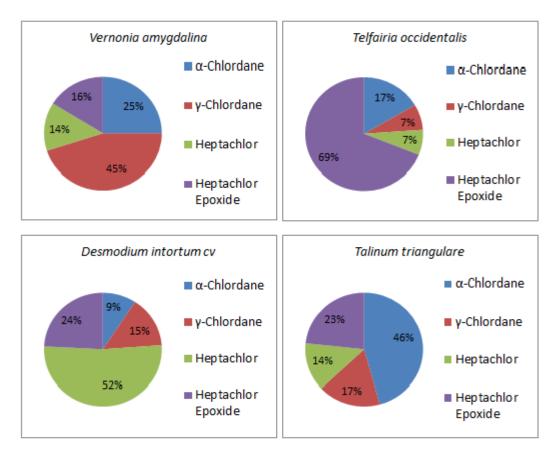


Figure 3b: Percentage composition of DDTs congeners in the vegetables





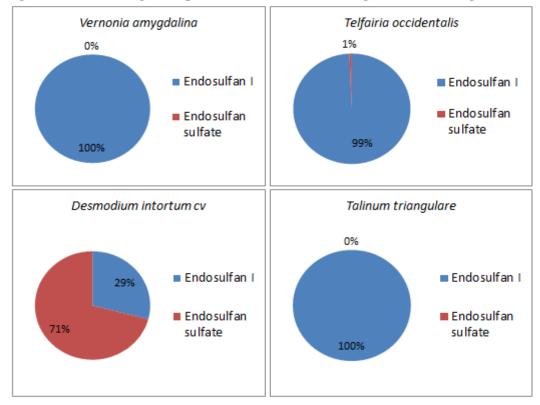


Figure 3d: Percentage composition of Endosulfans congeners in the vegetables

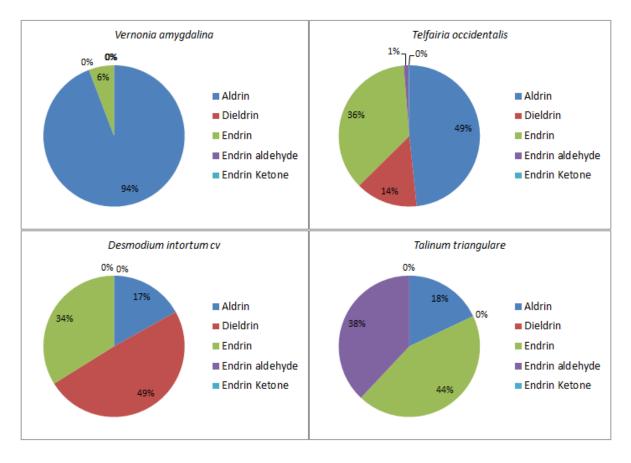


Figure 3e: Percentage composition of Drins congeners in the vegetables

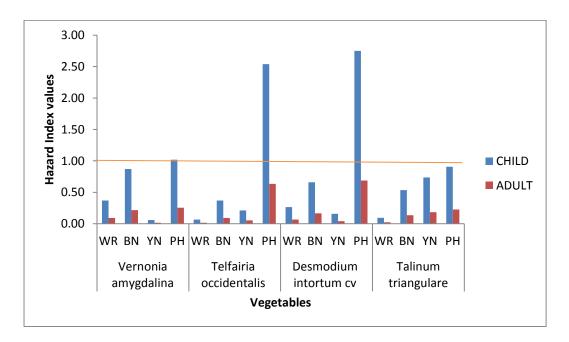


Figure 4a: Hazard index values of OCPs in the vegetables (The yellow line behind the bars indicates the benchmark value)

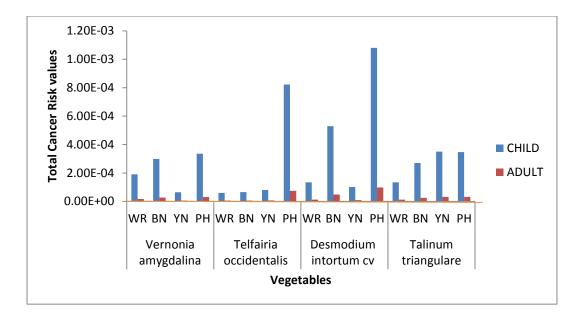


Figure 4b: Total cancer risk values of OCPs in the vegetables (The yellow line behind the bars indicates the acceptable risk value of 1×10^{-6})