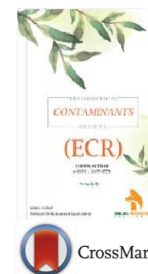


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RESEARCH ARTICLE

HEALTH RISK ASSESSMENT OF ORGANOCHLORINE PESTICIDE RESIDUE IN CRUDE PALM OILS IN SELECTED PALM PLANTATIONS IN CROSS RIVER STATE, NIGERIA

Nwuyi O. Sam-Uket^a, Bassey S. Okori^{b*}^a Department of Animal and Environmental Biology, Faculty of Biological Sciences, University of Cross River State, Nigeria^b Department of Chemical sciences, Clifford University, Owerrinta, Abia State, Nigeria*Corresponding author's e-mail: sambassey28@gmail.com

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ABSTRACT

Crude palm oil (CPO) is thought to be the most produced and consumed vegetable oil globally. However, when oil palm is grown using pesticides, it may contain OCP residues, which could be hazardous to humans due to their bioaccumulation in the body. For this reason, this study was conducted. Over the course of six months, the processed CPO (n = 18) was gathered once a month from the oil palm plantations owned by JP, Calaro, and Mrs. P. (control). The samples underwent analysis using gas chromatography mass spectrometry (GC-MS/MS) to detect the presence of organochlorinated pesticides (OCPs). Dieldrin and endosulphan II residues were found in the CPO from oil palm plantations (Calaro and JP) that were sprayed with pesticides. The CPO from the Calaro plantation had the highest total OCPs (0.23 ng/mL), followed by the CPO from the JP oil palm plantation (0.14 ng/mL). Dieldrin residues from Calaro and JP oil palm plantations were found in CPO, which is unsafe for ingestion by humans, because they exceeded FAO/WHO acceptable limits for pesticides in food. Since dieldrin was the main source of non-carcinogenic contamination for consumers, it had the highest EDI values in CPO from Calaro (1.23×10^{-4} ng/kg/day) and JP oil palm (7.03×10^{-5} ng/kg/day). This indicates that dieldrin has the highest potential to cause serious health problems. Dieldrin's EDI values in CPO from Calaro and JP oil palm plantations were higher than the RfD value of 0.00005, suggesting a possible risk to health. CPO from Calaro had the highest HR after ingesting dieldrin, suggesting a higher risk of developing cancerous conditions. The government must therefore educate farmers on the best practices for applying pesticides and create laws and policies to regulate the proper use of pesticides on agricultural land in light of the possibility that CPO processed from oil palm that is cultivated with pesticides may be contaminated and that there may be health risks involved.

KEYWORDS

Organochlorine pesticide, Crude palm oil

1. INTRODUCTION

The usage and application of pesticides, particularly organochlorine pesticides (OCPs), in agriculture and public health has led to higher concentrations of this class of chemicals. According to some study, this has contaminated a number of environmental matrices worldwide, including drinking water, food crops, oil palm, crude palm oil (CPO), and food fish species from various agrarian ecosystems in Nigeria (Nemr et al., 2012; Bazrafshan et al., 2013; Buah-Kwofie et al., 2018). OCPs are widely used in forestry, public health, and the home sphere in addition to their advantageous role in pest and vector control in agriculture (Aliyeva et al., 2012). Due to this situation, OCPs are now a vital tool in agricultural production, with the result that pesticides of all kinds are used to produce one-third of agricultural products (Liu et al., 2002). Certain pesticides, like organochlorine pesticides (OCPs), have the ability to prevent and control weeds, insects, and disease vectors, but they can also be hazardous to wildlife, humans, and the environment (Behrooz et al., 2009; Aliyeva et al., 2012). Strong evidence has been found linking pesticide exposure to a variety of diseases (Mostafalou and Abdollahi, 2013). The study's aim is to determine the degree of contamination in CPO from Elaise guineensis grown on pesticide-contaminated soil in Southern Cross River, South Nigeria. The objectives are:

- To ascertain the OCP content in the CPO of oil palm grown on some Southern Cross River plantations' pesticide-contaminated soils.
- To look into the health risk of consuming OCPs from CPO contaminated with pesticides.

2. METHODS

2.1 Study Area

Cross River is located between longitudes 07° 15' and 09° 28' East and latitudes 05° 32' and 04° 27' North. The state is bordered to the north by Benue State and to the east by the states of Akwa Ibom, Anambra, and Imo. Its total area is approximately 21,000 km². The international border with Cameroon forms the eastern boundary, while the southern boundary is traced along the Cross River into the Bight of Bonny. The climate of Cross River is tropical rainforest with distinct wet and dry seasons. In general, the months and seasons have a significant impact on the climate. The rainy season has two peaks in June/July and September, and it lasts from mid-March to October. The total yearly precipitation is between 1500 and 1900 mm.

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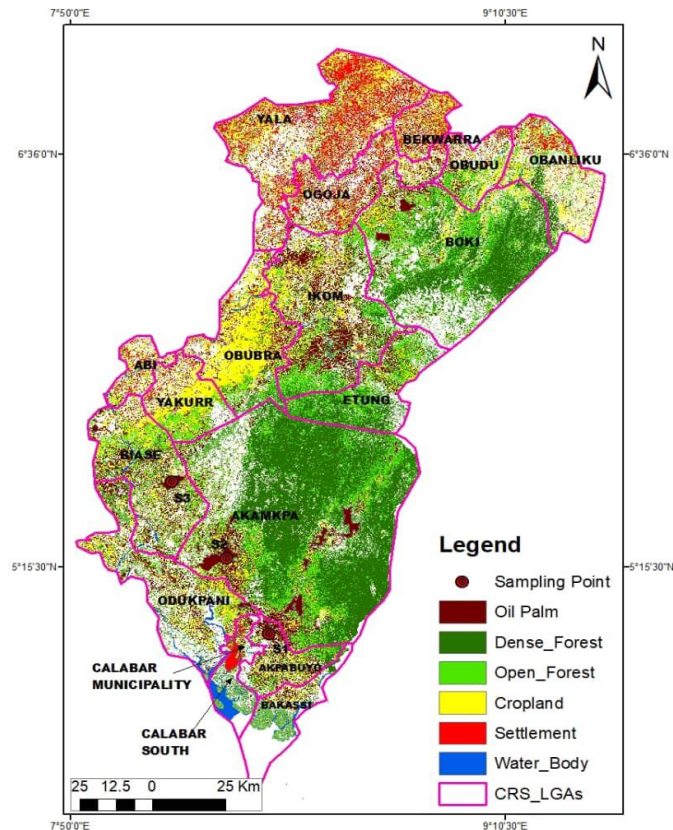
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Table 1a: Geocoordinate of the sampling points

S/N	Points	Location	Latitude	Longitude
1	Mrs. P Oil Palm	Akpabuyo	05° 02'31.281"N	008° 26'41.757"E
2	Calaro Oil Palm	Akamkpa	05° 17'20.438"N	008° 18'48.911"E
3	JP Oil Palm Estate	Biase	05°32'14.735"N	008° 08'57.853"E

**Figure 1:** Map of Cross River showing the oil palm plantations

2.2 Harvesting of oil palm fruit

Using a long-armed sickle, matured oil palm fruit was harvested from the following plantations: Mrs. P. (Plantation 1: control), Calaro (Plantation 2), and JP (Plantation 3). The fruits were individually transported to their respective oil palm mill for milling as soon as they were harvested. Following the milling procedure, the CPO were gathered.

2.3 Collection of CPO samples

Over the course of six months (January – June), CPO samples ($n = 18$) were collected monthly, separately, from Mrs. P. oil palm plantation, Calaro oil palm plantation, and JP oil palm plantation into a well-treated one-liter sampling container. Before the actual sampling, the sampling container was treated with 10% nitric acid, cleaned with detergent, rinsed with distilled water, and dried in a hood. The samples were collected and promptly labeled appropriately. They were then stored in an ice chest and transported to the environmental laboratory of Mifor Laboratory Calabar for analysis of organic compounds.

2.4 Chemical analysis of OCP

A 50 mL polypropylene centrifuge tube containing 10 μ L of internal standard (0.1 ng/ μ L) and 1 g of CPO sample was weighed, and the sample was diluted with 5 mL of water. Next, a ceramic homogenizer-containing centrifuge tube was filled with 10 mL of acetonitrile. The tube was hand-shook vigorously for one minute after it was sealed. To achieve good phase separation, 4 g of NaCl was added to the tube and shaken vigorously for a few seconds. Next, the tube was centrifuged at 5000 rpm for 5 minutes. Five milliliters of the supernatant were added to a fifteen milliliter EMR (enhanced matrix removal)-lipid d-SPE tube that held the EMR adsorbent. Five milliliters of water were added to the tube to activate the adsorbent prior to transferring the supernatant. After one minute of vortexing, the tube was centrifuged for five minutes at 5,000 rpm. Then, after transferring the 5 mL of supernatant to an EMR-lipid polish tube with 2 g of salts (NaCl/MgSO₄ = 1:4), the tube was vortexed for a minute. Using the

extraction cartridges, 2 milliliters of the supernatant were moved to a 10-milliliter glass tube. Under a mild nitrogen stream, the supernatant was evaporated until it was completely dry. Before GC-MS/MS (gas chromatography-triple quadrupole mass spectrometry) analysis, the residue was filtered through a 0.22 μ m PTFE filter and redissolved in 100 μ L of ethyl acetate.

A triple quadrupole mass spectrometer was connected to a SHIMADZU GCMS-TQ8050 system for the GC-MS/MS analysis. GC separation was performed using the Rxi-5Sil MS capillary column (30 m \times 0.25 mm i.d., film thickness = 0.25 μ m). Sample injection was done in splitless mode while the injector temperature was maintained at 250 $^{\circ}$ C. The carrier gas was helium (99.999%), with a flow rate of 1.69 milliliters per minute. Utilized was the subsequent oven temperature program: 50 $^{\circ}$ C for one minute, then increase to 125 $^{\circ}$ C at a rate of 25 $^{\circ}$ C/min, then increase to 300 $^{\circ}$ C at a rate of 10 $^{\circ}$ C/min and hold for another two minutes. At 230 $^{\circ}$ C, mass spectrometry was carried out using electron impact ionization (70 eV) in multiple reaction monitoring (MRM) mode. A temperature of 250 $^{\circ}$ C was used for the transfer line. The collision gas used was argon (99.999%). The injection volume was 1 μ L, and the solvent delay was set for 4 minutes. Two MRM mass transitions per target compound were tracked, providing the retention time (tR) and useful information for both qualitative and quantitative analysis. Groupings were adjusted to achieve low background noise and sufficient sensitivity.

2.5 Estimated daily intake (EDI)

The EDI (ng per kilogram per day) was calculated according to a study as shown in equation 1 (Sana et al., 2021).

$$EDI = \frac{ADI \times CM}{BW} \quad (1)$$

Where ADI = average daily intake of palm oil per person per day (kg/person/day)

CM = Concentration of OCP in palm oil (ng mL⁻¹)

BW = Average body weight

The ADI of palm oil per person per day was taken as 0.0527 kg/person/day, and the average body weight will be taken as 60 kg (EPA, 2002). When the EDI is above the reference oral dosage (RfD) of each OCPs, then there is a potential health danger. RfD is the oral reference dose for a particular compound intake (mg/kg/day) α -HCH (0.8), β -HCH (0.8), γ -HCH (0.3), pp'-DDT (0.5), op'-DDT (0.5), pp'-DDD (0.5), op'-DDD (0.5), pp'-DDE (0.5), aldrin (0.03), dieldrin (0.00005), endrin (0.3), heptachlor epoxide isomer B (0.013), endosulphan 1 (0.006), Endrin aldehyde (0.3), endosulphan II (0.006), endosulphan sulphate (0.006), endrin ketone (0.3), and heptachlor (0.5) (Witczak, 2014).

2.6 Target hazard quotient (THQ)

It is of paramount importance to investigate the health risk associated with the consumption of the palm oil. This was achieved by the use of the Target Hazard Quotient. The THQ describes the ratio of the evaluated dose to the reference dose (Chien et al., 2002). When the THQ is above 1, then there is a potential health risk. It was calculated as shown in equation 2 below:

$$THQ = \frac{EF \times ED \times ADI \times CM \times 0.001}{RfD \times BW \times AT} \quad (2)$$

Where EF = exposure frequency (365 days)

ED = exposure duration (60 years)

CM = concentration of OCPs in palm oil (ng mL⁻¹)

0.001 = conversion factor

RfD = reference oral dosage

BW = average body weight

AT = average time of exposure for 365 days in 60 years for adults when considering non-carcinogenic effects (But AT = 365 days \times 60 years = 21900 days).

2.7 Hazard ratio (HR)

The HR reveals the carcinogenic potential of ingesting OCPs. The HR was calculated according to a study as shown in equation (3). Oral Slope Factor (OSF) values were obtained from the Integrated Risk Information System (IRIS), United States Environmental Protection Agency (USEPA) for each contaminant (Dougherty et al., 2000; USEPA, 2007). HR values above 1 represent a likelihood of causing health issues, particularly cancer within a life span of 60 years.

$$HR = \frac{EDI}{CBC} \quad (3)$$

Where:

HR = health ratio via ingestion route

EDI (ng kg⁻¹ d⁻¹) = estimated daily intake

CBC (ng kg⁻¹ d⁻¹) = cancer bench mark concentration

The CBC was calculated according to equation 4 as shown below:

$$CBC = \frac{RL \times OSF \times BW}{ADI} \quad (4)$$

Where RL = risk level (10⁻⁶)

OSF = oral slope factor (mg/kg/day)

BW = body weight (60 kg)

ADI = average daily intake of palm oil (0.0527 kg/person/day)

N/B: Oral slope factor of dieldrin = 16 mg/kg/day

2.8 Statistical analysis

Data obtained was subjected to descriptive statistics (ranges, mean, and standard deviation). Analysis of variance (ANOVA) was used to test for the significant difference in the concentration of OCPs between the CPO from the different oil palm plantation compared to the control at 0.05 level of significance and at their relevant degree of freedom. All analysis were carried out using predictive analytical software (PASW) version 20.

3. RESULTS

Table 1: Concentration of OCP in CPO from oil palm cultivated using pesticides

OCP concentration (ng/mL)	Mrs P plantation (control)	Calaro oil palm plantation	JP oil palm plantation	FAO/WHO (2020)
Hexachlorocyclohexanes (HCHs)	ND	ND	ND	0.01
Endosulphan 1	ND	ND	ND	-
Dieldrin	ND ^a	0.14 ± 0.07 ^b (0.07 - 0.25)	0.08 ± 0.03 ^b (0.05 - 0.12)	0.02
Endrin aldehyde	ND	ND	ND	0.05
Endosulphan II	ND ^a	0.09 ± 0.07 ^b (0.01 - 0.26)	0.06 ± 0.05 ^a (0.01 - 0.13)	-
DDD	ND	ND	ND	0.1
Endosulphan sulphate	ND	ND	ND	1.0
Heptachlor epoxide (Isomer B)	ND	ND	ND	-
Endrin Ketone	ND	ND	ND	0.05
Total OCP	ND^a	0.23 ± 0.07^b(ng/mL)	0.14 ± 0.04^b (ng/mL)	

Values are in Mean ± standard deviation; ranges in parenthesis (); ND - not detected

Values with different superscript compared to the control is significantly different (p<0.05)

Table 2: Estimated daily intake of ingesting OCPs from CPO processed from contaminated oil palm

Organo-chlorinated Pesticides (ng/kg/day)	Mrs P plantation (control)	Calaro oil palm plantation	JP oil palm plantation	RfD values
Hexachlorocyclohexanes (HCHs)	-	-	-	0.8
Endosulphan 1	-	-	-	0.006
Dieldrin	-	1.23 x 10 ⁻⁴	7.03 x 10 ⁻⁵	0.00005
Endrin aldehyde	-	-	-	0.3
Endosulphan II	-	7.91 x 10 ⁻⁵	5.27 x 10 ⁻⁵	0.006
DDD	-	-	-	0.5
Endosulphan sulphate	-	-	-	0.006
Heptachlor epoxide (Isomer B)	-	-	-	0.013
Endrin Ketone	-	-	-	0.3

Table 3: Target hazard quotient of ingesting OCPs from CPO processed from contaminated oil palm

Organo-chlorinated Pesticides	Mrs P plantation (control)	Calaro oil palm plantation	JP oil palm plantation
Dieldrin	-	2.46 x 10 ⁻³	1.41 x 10 ⁻³
Endosulphan II	-	1.32 x 10 ⁻⁵	8.78 x 10 ⁻⁶

Table 4: Health ratio (HR) of ingesting OCPs from CPO processed from contaminated oil palm

Organo-chlorinated pesticides	Mrs P plantation (control)	Calaro oil palm plantation	JP oil palm plantation
Dieldrin	-	6.76 x 10 ⁻³	3.86 x 10 ⁻³
Endosulphan II	-	-	-

Note: HR of Endosulphan II was not calculated because its carcinogenic slope factor is unavailable

4. DISCUSSION

The CPO from the control plantation (Mrs. P. plantation) did not record any OCP residue, but the current study found differences in the concentration of OCPs in the CPO from Calaro and JP oil palm plantations. This is consistent with research by who also noted contamination during oil palm tree cultivation and fruit harvesting as a result of pre- and post-planting farming techniques like the use of fungicides and fertilizer residues (Ayodele and Oluyomi, 2011). Since no pesticides were used during the cultivation of the oil palm, the CPO from the control plantation (Mrs. P. plantation) did not record any OCP residue, confirming the theory that pesticide use contaminates food crops. In addition, the Calaro oil palm plantation's CPO had the highest dieldrin and endosulphan II concentrations when compared to the control group. This indicates that in Calaro's CPO, where more pesticides are applied, the oil palm absorbed these residues into their fruit at higher concentrations. The pesticides used for oil palm cultivation also increased the concentration of the previously mentioned OCPs in the soil (Amasha et al., 2020). The differences in the quantity of pesticides applied, the timing of application, photo-degradation, absorption by plant parts, sorption at the soil level, chemical decomposition (pH, humidity, and temperature), and biological degradation by microorganisms could all be contributing factors to the variations in OCP concentrations in the CPO from Calaro and JP oil palm plantations (Sharma et al., 2019). The results of the current investigation indicate that the total OCP in the CPO from Calaro and JP oil palm plantation was less than the total OCP reported by Sana et al. (2021) for a study on the OCPs in Pakistani buffalo and cow milk, and for a study on the OCP residues in Chinese vegetable oils (Cui et al., 2020). Moreover, dieldrin was the most common OCP in the current investigation. A group researcher found that hexachlorocyclohexanes (HCHs) were the most common OCP (Sana et al., 2021). A studied OCPs from food items in the Cambodian market and found that dichloro diphenyl trichloroethanes (DDT) was the most common OCP (Wang et al., 2011). A studied OCP residues in Ghanaian cocoa beans and found that DDT was the most common OCP (Okoffo et al., 2016). Dieldrin levels in JP and Calaro oil palm plantations were higher than FAO/WHO acceptable limits for pesticides in food, meaning the oil is safe to eat. There is a need to reduce pesticide use because, despite current indications of minimal danger, residues will continue to accumulate over time to the point where they could pose serious health risks to consumers, especially given the well-documented potential for OCPs to become toxic to human health, wildlife, and the environment (Behrooz et al., 2009; Aliyeva et al., 2012). OCP poisoning may cause convulsions, headaches, dizziness, nausea, vomiting, tremors, confusion, muscle weakness, slurred speech, salivation, and sweating (FAO/WHO 2020).

4.1 Health risk assessment

We also effectively used the health risk model to predict the potential carcinogenic and non-carcinogenic health implications of ingesting OCP from CPO from oil palm cultivated with pesticides in order to further investigate the possible health risks of doing so. The results of this study indicate that in CPO from Calaro and JP oil palm, dieldrin had the highest EDI and THQ values. As a result, dieldrin was the most consumed OCP from CPO from Calaro and JP oil palm plantations. It also had the greatest potential to cause major health problems, making it the main non-carcinogenic contamination source for consumers. Further evidence that dieldrin may eventually result in major health problems comes from the fact that the EDI values of dieldrin in CPO from Calaro and JP oil palm plantations were higher than the RfD value of 0.00005. Consumers of CPO from Calaro are at a higher risk of developing carcinogenic conditions, as indicated by the current study, which found that the HR from dieldrin ingestion was highest in our sample. The THQ of ingesting endosulphan and dieldrin, as well as the HR of ingesting dieldrin in CPO from oil palm plantations (Calaro and JP oil palm plantations) grown with organochlorine pesticides, were less than 1, indicating the need to reduce pesticide use, particularly since dieldrin's earlier EDI indicated that there was. This is due to the possibility that these residues could accumulate over time to the point where they endanger customers' health. Since pesticide poisoning can result in convulsions, headaches, dizziness, nausea, vomiting, tremors, confusion, muscle weakness, slurred speech, salivation, sweating, and cancer, it is best to prevent it as much as possible (FAO/WHO, 2020).

5. CONCLUSION

The study's findings indicate that the use of pesticides during oil palm plantation cultivation contaminates processed CPO with OCP residues. Because more pesticides were applied there, Calaro had the highest dieldrin and endosulphan II residues. Dieldrin levels in the CPO from Calaro and JP oil palm plantations were too high to be consumed. Dieldrin

was the most consumed OCP, according to EDI and THQ, and it also posed the greatest risk of major health problems. It was also the main cause of non-carcinogenic contamination for CPO consumers from Calaro and JP oil palm plantations. Dieldrin's HR in Calaro's CPO showed the greatest potential for long-term carcinogenic effects.

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ETHICAL ISSUE

Authors are aware of, and comply with, best practice in publication ethics specifically with regard to authorship (avoidance of guest authorship), dual submission, manipulation of figures, competing interests and compliance with policies on research ethics. Authors adhere to publication requirements that submitted work is original and has not been published elsewhere in any language.

COMPETING INTERESTS

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

AUTHORS' CONTRIBUTION

Bassey S. Okori perform the experimental design, prepared the manuscript text and compiled the data. Nwuyi O.Sam-Uket performed the literature review and manuscript edition.

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