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Pesticide Residue Contamination of Some Cereals and their Consequential Health Implication in the Food Chain of Taraba Northern Geo-political Region, Taraba State

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Authors' contributions

This work was carried out in collaboration among all authors. This work was carried out in collaboration among all authors. Author BWB designed the concept of the research and participated in sampling and data collection, and wrote report of the study. Author NWY helped in designing the concept and proof read the proposal and the final report of the manuscript. Author AA was involved in sampling, data collection and data analysis. Author SAF was actively involved in the processes of sampling and data collection of the manuscript. All authors read and approved the final manuscript. All authors read and approved the final manuscript.

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ABSTRACT

Aim: To investigate the Pesticide Residues (PRs) contamination of some cereals and determine their health risks in the food chain of Taraba North Geo-political region.
Study Design: Maize, Millet, Rice and Sorghum, and their soils were randomly collected from farmers' field in three (3) LGAs of Taraba North.

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Place and Duration of Study: The samples were collected from Ardo-kola, Karim-lamido and Zing LGAs of Taraba State. They are processed at Laboratories of Biological Sciences Department of Taraba State University and Analyzed at the Department of Chemistry, Yobe State University, Damaturu, Yobe State, between September, 2021 – March, 2022.

Methodology: Soils and ripe cereals stalks were collected applying the principles of randomization. These samples were processed, pulverized into powdered form, and analyzed for pesticide residues (PR) using the Gas Chromatograph - Mass Spectrometer (GC-MS).

Results: Ten (10) pesticide residues in different concentrations were recorded, with some of them occurring above the permissible limits. Isopropylamine accounted for 21% of total contaminants' concentrations, followed by carbofuran (14%), dichlorvos (12%), t-nonachlor (11%), heptachlor (10%), HCB and g-chlordane all at 9%, DDT (7%), Endosulfan (6%), and Aldrin at 1%. Health implications showed that some of the cereals are unsafe for consumption. In Ardo-Kola and Zing, only millet had a Hazard Index (HI) of less than 1, while in Karim-Lamido, millet and sorghum recorded HI ≤ 1. In Ardo-Kola trend in HI values in adullts was rice (53.32) > sorghum (24.35), > maize (10.99), > millet (0.82), while in children, the trend was rice (64.50), > sorghum (24.71), > maize (10.65), > millet (0.90). In Karim-Lamido, HI values in adults were in the following order: rice (54.64) > maize (38.44), > millet (0.62), > sorghum (0.35). At the same time, HI in children followed the order; rice (66.10) > maize (46.50), > sorghum (0.74), > millet (0.73). In Zing, HI values were in descending order of rice (32.20) > sorghum (12.27) > maize (8.27) > millet (0.47), in adults, and rice (38.97) > sorghum (14.86) > maize (9.99) > millet (0.55), in children. Overall, children are at a higher risk of toxicity than adults.

Conclusion: It was deduced at the end of the study that pesticides residue are bioaccumulating in the food chain due to excessive use in many agricultural phases. Hence we recommended intensified efforts in check-mating these products in our agricultural systems.

Keywords: Safety; pesticide; residue; toxicity; contaminants.

1. INTRODUCTION

Agricultural soils have suffered massive contamination in the past decades due to the indiscriminate use of agrochemicals [1,2]. Although it has massively contributed to the increase in global food production [3], through improved crop yield and reduced crop loss [4], indiscriminate agrochemical use has resulted in the loss of soil biodiversity while simultaneously causing various health-related problems due to the complex nature of these chemicals [5]. Plants absorb nutrients from the soil, and when the soil is contaminated, plants can also absorb the contaminants, bioaccumulating them in the food chain [6,7,8,9]. Human exposure to pesticides mainly occurs via inhalation, dermal absorption, and oral ingestion [10]. Oral ingestion is the primary exposure route, with food and water being the major drivers [11]. The long-term effects associated with environmental pollution by agrochemicals, even at low concentrations, are of great concern worldwide [12]. Agrochemicals contain impurities that, when transferred to humans through the food chain, may affect food quality and safety [13,14]. Further, prolonged intake may lead to chronic accumulation in humans, causing disruptions of numerous biochemical processes and leading to numerous diseases [15,16]. Therefore, it becomes essential to monitor food quality and safety given that plant uptake is one of the major pathways through which pesticide residues (PRs) enter the food chain [17,18].

By target organism, pesticides can be divided into many groups, and two of the most well-known pesticide families are the organochlorine and the organophosphate families. Chlorinated hydrocarbons, known as organochlorine pesticides (OCPs), were widely used in agriculture and insect control from the 1940s to the 1960s [19]. Although organochlorine pesticides have been used for a very long time, their substantial toxicity to humans, plants, and other organisms, as well as their long-lasting natural persistence and potential for bioaccumulation, make them unsuitable for usage [19,20]. These findings necessitated the bans and restrictions placed by several nations, by national and international bodies, on OCPs but due to porous borders and weak regulations, these chemicals find their way into the country, where they are still used by some farmers ignorantly [21,22]. Although these restrictions have resulted in an increase in the use of synthetic pesticides that are alternatives, such as organophosphates (OPPs) in the 1960s, lower persistence carbamates [20], and pyrethroids (PYRs) in the 1980s [19], in several developed nations, OCPs are still in use in some developing nations. Zhang [23] reported that 90% of pesticide-related chemicals, such as toxophene, DDT, aldrin, and dieldrin are both carcinogenic and mutagenic, while carbaryl, ethylene dibromide, and parathion are mutagenic. Hence, it is crucial to determine the extent of contamination in areas known for high farming activities, as they are significant food suppliers across the country. This has the potential to rapidly spread contaminated food products across the country.

Taraba North is an active farming zone where pesticides are used on a large scale to cultivate different crops, especially cereals like maize, millet, rice, and sorghum. Cereals remain one of the most significant sources of food and animal feed, so assessing their safety or compliance with the law is crucial [24]. Knowing PRs can accumulate in the soil, there is also the possibility of them bioaccumulating in the tissues of farm products and, hence, entering the food chain, where they can cause many life-threatening ailments. There is no literature on this effect in the study area, given its background as a robust agricultural zone. Hence, the need to carry out this study to serve as a piece of baseline information for policymakers, farmers, and the public at large. Hence, this study aims at ascertaining the level of contamination by OCPs, OPPs, and PYRs in 200 samples of cereals in the study area and evaluating the risk associated with the consumption of the identified PRs. This study is vital since it will aid the government in creating regulatory guidelines for pesticides in food because it will identify the risk linked with the PRs levels found as well as their health implications in the food chain of Taraba's northern geopolitical zones.

2. MATERIALS AND METHODS

2.1 Study Area

The study was conducted in the Taraba Northern geo-political district of Taraba State, with an area of about 14,227 km^2 and a population estimate of about 981,900 inhabitants with a growth rate of 2.91% according to the National Population Commission [25]. It is made up of six (6) LGAs; Ardo-Kola, Jalingo, Karim-Lamido, Lau, Yorro, and Zing. The region falls within latitude 8.88° North and longitude 11.37° East and is 351 meters above sea level. The principal occupation of people in this area is agriculture, producing crops such as maize, rice, sorghum, millet, cassava, and yam. Different varieties of vegetables are also produced in commercial quantities. Taraba North is characterized by dry and wet seasons spanning a period of eight months from April to October. The mean annual rainfall is estimated at 1058 mm, with the wetting months being August and September. The dry season lasts five months, from December to March, with December and January being the driest months. The humidity drops to about 15% while the mean annual temperature is estimated at 28°C. Maximum temperatures range between 30°C – 39.4°C while minimum temperatures range from 15°C – 23°C. These characteristics favor agricultural activities.

2.2 Sampling and Sample Collection

Three local government areas (LGAs) were selected randomly. In each LGA, wards notable for producing grains of maize, Guinea corn, millet, and rice were purposely identified and randomly selected for sample collection. In each sampling unit, soils at a depth of 0–30 cm were randomly collected as described by [26,27]. The soils were pooled into a composite sample and analyzed for possible pesticide residue contaminants. From the selected farms, harvested grains were randomly collected with the farmers' permission and processed in the lab for analysis.

2.3 Processing of Samples

In the laboratory, all samples (soil and grains) were air-dried, milled, pulverized, and sieved through a mesh size of 2 mm. The fine powder was placed in a clearly labeled plastic container and analyzed for pesticide residue contaminants.

2.4 Methods of Analysis

Pesticide residues were analyzed using Gas Chromatography–Mass Spectrometry (GC–MS) (Agilent Tech. GC7890B, MSD 5977A). During the GC-MS analysis, 5g of air-dried samples were weighed into a 50 mL Flask, to which 30 mL aliquots of an extraction solvent mixture of n-hexane and dichloromethane (1/1, v/v) were added and then ultrasonically extracted at room temperature for 30 min. At the end of the extraction, each mixture was centrifuged at 5000 r/min for 10 min. The extraction procedure was repeated twice, and the extracts from each time were combined into a pear-shaped bottle and dissolved in 20 mL of n-hexane twice and then subjected to clean-up. The dissolved solution was added to five grams of copper and shocked for 5 min, then centrifuged at 8000 r/min for 5 min. The supernatant was added to 3 mL of concentrated sulfuric acid, shaken for 2 min, and then centrifuged at 2000 r/min for 5 min. The supernatant was added to 1 mL of concentrated sulfuric acid and shaken again for 2 min, then centrifuged at 2000 r/min for 5 min. The supernatant solvent was dried by a rotary evaporator. The residue was reconstituted with 1 mL of n-hexane and mixed in a vortex stirrer [28-31]. The contamination rate was determined using the original rate equation with minor modifications of terms, as in [27].

The rate of contamination and the risk to health were also determined using the following equations:

Rate of Contamination by Individual Contaminant

$$(RC_{ic}) = \frac{VC_{if}}{TVC_f} \quad (1)$$

Where; $\boldsymbol{VC_{if}}$ is the value of individual contaminant in the farm, and $\boldsymbol{TVC_f}$ is the total value of contaminants in the farm.

Rate of LGAs Contamination by Individual Contaminant

$$(\boldsymbol{RDC_{ic}}) = \frac{VC_{id}}{TVC_d} \quad (2)$$

Where; $\boldsymbol{VC_{id}}$ is the value of individual contaminants in the LGA, and $\boldsymbol{TVC_d}$ is the total value of contaminants in the whole studied area.

Rate of Contamination in the whole Geo-political Region

$$(\boldsymbol{RSC_{ic}}) = \frac{VC_{is}}{TVC_s} \quad (3)$$

Where; $\boldsymbol{VC_{is}}$is the value of individual contaminant in the LGAs, and $\boldsymbol{TVC_s}$ is the total value of contaminants in the district.

Risk to health due to intake of metal-contaminated grains was calculated using the hazard quotient (HQ) as described by [33] and given as;

evaporated in a rotary evaporator to dry at a low temperature and weak vacuum. The residue was

$$\boldsymbol{HQ} = \frac{Div\ x\ C_{contaminant}}{RfD\ x\ B_o} \quad (4)$$

Where, (Div) is the daily intake of vegetables (kgd^{-1}), ($C_{contaminant}$) is the concentration of PRs ($mgkg^{-1}$), RfD is the oral reference dose for the contaminant, and Bo is the human body mass (kg), whereas potential risk to human health due to more than one contaminant known as the Hazard Index (HI) was calculated as described by [33], which is the total sum of all the Hazard Quotients as revealed in the equation below;

$$\boldsymbol{HI} = \sum \boldsymbol{HQ} \quad (5)$$

The hazard index assumes that the magnitude of the adverse effect will be proportional to the sum of the multiple metal exposures.

2.5 Statistical Analysis

The mean standard deviation was used to express all experimental data in this study (SD). The one-way ANOVA was used to calculate statistical comparisons, and ***p 0.001 was regarded to be of extreme significance.

2.6 International Standard for Permissible Limits

The maximum permissible limits established by reputable international regulators, specific to the detected heavy metals in this current study, are summarized in Table 1.

3. RESULTS AND DISCUSSION

3.1 Mean Concentration of Pesticide Residues in Cereals

The mean concentration of pesticide residues (PRs) in cereals obtained from Ardo-kola is presented in Table 2. The table shows that all PRs tested were found in different concentrations in one cereal or the other. The contamination with PRs showed 80% in maize and 50% in millet. Rice had all queried PRs recorded at 100%, while in sorghum, 70% of the queried PRs were recorded.

In maize, isopropylamine had the highest mean concentration of 0.1109 + 0.001 mg kg^{-1}, while aldrin recorded the least mean concentration of 0.0100 + 0.003 mg kg^{-1}, but carbofuran and g-chlordane were not recorded at all. In millet, isopropylamine still recorded the highest concentration (0.1303 + 0.006 mg kg^{-1}), while heptachlor was the least at 0.0013 + 0.000 mg kg1. In the same vein, endosulfan, aldrin, carbofuran, dichlorvos, and g-chlordane were absent. In rice, carbofuran (0.1892 + 0.000 mg kg^{-1}) had the highest PR recorded, while the least was recorded in aldrin (0.0158 + 0.000 mg kg^{-1}). In sorghum, aldrin, DDT, and heptachlor were not recorded, as carbofuran appeared the highest with a mean concentration of 0.1961 + 0.000 mg kg^{-1} while isopropylamine (0.0010 + 0.000 mg kg^{-1}) was the least recorded.

Rice had the highest cumulative contaminant concentration (CCC) at 1.1704 mg kg^{-1} (58.87%), followed by maize, sorghum, and millet, which had mean concentrations of 0.4043 mg kg^{-1} (20.33%), 0.2329 mg kg^{-1} (11.71%), and 0.1806 mg kg^{-1} (9.08%), respectively. Isopropylamine (0.3983 $mgkg^{-1}$) > carbofuran (0.2953 $mgkg^{-1}$) > HCB (0.2219 $mgkg^{-1}$) > dichlorvos (0.2210 $mgkg^{-1}$) > t-nonachlor (0.2177 $mgkg^{-1}$) > heptachlor (0.1980 $mgkg^{-1}$) > g-chlordane (0.1812 $mgkg^{-1}$) > DDT (0.1285 $mgkg^{-1}$) >endosulfan (0.1005 $mgkg^{-1}$) > Aldrin (0.0258 $mgkg^{-1}$) was the order of occurrence of the Total Contaminant Concentration (TCC).

During the study, some PRs concentrations were found above the permissible limits. These include isopropylamine in maize, millet, and rice; HCB in rice; endosulfan in rice; DDT in maize and rice; t-nonachlor in maize and rice; dichlorvos in rice; heptachlor in maize and rice; and g-chlordane in rice. In contrast, the rest of the PRs' concentrations were recorded below the international permissible limits.

The mean concentration of pesticide residues in karim-lamido as recorded in Table 3, showed that all the studied cereals contain one or more pesticide residues, with some occurring even above the standard permissible limits. Among those that exceeded the permissible limits is isopropylamine in maize (0.1716 + 0.003 mg kg^{-1}), millet (0.1499 + 0.002 mg kg^{-1}), and rice (0.1613 + 0.004 mg kg^{-1}). Endosulfan concentrations in maize (0.0747 + 0.004 mg kg-1) and rice (0.0558 + 0.003 mg kg-1); DDT concentrations in maize (0.0743 + 0.004 mg kg-1) and rice (0.0807 + 0.001 mg kg-1). t-nonachlor in maize (0.0997 + 0.001 mg kg^{-1}) and in rice (0.1160 + 0.000 mg kg^{-1}). Dichlorvos (0.1187 + 0.003 mg kg^{-1}) in maize and rice (0.1498 + 0.000 mg kg1). Heptachlor in maize (0.0755 + 0.004 mg kg^{-1}) and rice (0.1514 + 0.001 mg kg^{-1}), and g-chlordane in rice (0.1100 + 0.000 mg kg^{-1}). The remaining pesticide residue concentration was recorded below the permissible limits.

Moreover, cumulative contaminant concentration (CCC) showed the additive concentration of a contaminant in a food item (Table 3). CCC followed this order: rice>maize>sorghum>millet, with CCC and PCC values of 1.2057 mg kg^{-1} (52.61%), 0.6488 mg kg^{-1} (28.31%), 0.2398 mg kg^{-1} (10.46%), and 0.1972 mg kg^{-1} (8.60%), respectively. TCC occurred in the following order: isopropylamine (0.4841 mg kg^{-1}) > carbofuran (0.2983 mg kg^{-1}) > dichlorvos (0.2774 mg kg^{-1}) > t-nonachlor (0.2628 mg kg^{-1}) > HCB (0.2375 mg kg^{-1}) > heptachlor (0.2269 mg kg^{-1}) > g-chlordane (0.1871 mg kg^{-1}) > DDT (0.1550 mg kg^{-1}) > endosulfan (0.1305 mg kg^{-1}) > aldrin (0.0319 mg kg^{-1}).

Furthermore, TCC recorded for each pesticide residue showed that isopropylamine had the highest concentration at 0.4841 mg kg^{-1} (21.12%), followed by carbofuran at 0.2983 mg kg^{-1} (13.01%), and dichlorvos at 0.2774 mg kg^{-1} (12.10%). The next were t-nonachlor 0.2628 mg kg^{-1} (11.46%), HCB 0.2374 mg kg^{-1} (10.36%), Heptachlor 0.2269 mg kg^{-1} (9.90%), g-chlordane 0.1879 mg kg^{-1} (8.16%), DDT 0.1550 mg kg^{-1} (6.76%), and endosulfan 0.1305 mg kg^{-1} (5.69%). The least TCC was recorded in aldrin, at 0.0319 mg kg^{-1} (1.39%). During the study, 100% of queried PRs were reported in both maize and rice, 40% in sorghum, and 30% in millet.

In Zing (Table 4), all queried PRs were recorded in different concentrations across different food items. 100% representation was recorded in rice, 80% in maize, 70% in sorghum, and 50% in millet. In maize, isopropylamine (0.0696 + 0.000 mg kg-1) had the highest PR concentration, while HCB (0.0069 + 0.000 mg kg-1) recorded the least, as carbofuran and g-chlordane were undetectable. In millet, isopropylamine had the highest concentration recorded at 0.0742 + 0.001 mg kg^{-1}, while heptachlor (0.0012 + 0.000 mg kg^{-1}) had the lowest. However, endosulfan, aldrin, carbofuran, dichlorvos, and g-chlordane were not detected. In rice, carbofuran had the highest PRs concentration recorded (0.1187 + 0.001 mg kg^{-1}) while aldrin had the lowest at a concentration of 0.0098 + 0.000 mg kg^{-1}. In sorghum, carbofuran was also the highest with a concentration of 0.0629 + 0.005 mg kg^{-1}, while endosulfan (0.0009 + 0.000 mg kg^{-1}) was the least, as aldrin, DDT, and heptachlor were not recorded.

Table 1. Permissible limits and oral reference dose of pesticide residues as approved by reputable international regulators

Pesticide Residues (PRs)	PL ($mgkg^{-1}$)	Reputable International Regulators	Ref	Reference Dose ($mgkg^{-1}d^{-1}$)	Ref
Aldrin	0.2	Food and Agricultural Organization (FAO) and World Health Organization (WHO)	[32]	3.0×10^{-5}	[33]
Carbofuran	NA	NA	[32]	5.0×10^{-3}	[34]
DDT	0.1	Food and Agricultural Organization (FAO) and World Health Organization (WHO)	[32]	5.0×10^{-4}	[33]
Dichlorvos (DDVP)	0.01	International Food Standards/Codex Alimentaruis, FAO/WHO	[32]	5.0×10^{-4}	[35]
Endosulfan	0.5 – 5	Food and Agricultural Organization (FAO) and World Health Organization (WHO)	[32]	6.0×10^{-3}	[35]
g-chlordane	0.01	International Program on Chemical Safety (IPCS)	[32]	6.0×10^{-5}	[36]
HCB (Hexachlorobenzene)	0.5	United States Environmental Protection (USEP)	[32]	8.0×10^{-4}	[35]
Heptachlor	0.01	International Food Standards/Codex Alimentarius FAO/WHO	[32]	5.0×10^{-5}	[34]
Isopropylamine	0.1	United States Environmental Protection (USEP)	[32]	1.75	[37]
t-nonachlor	0.04	Canadian Health Measures Survey (CHMS)	[32]	NA	[38]

Table 2. Mean Concentrations ($mg\ kg^{-1}$) of Pesticides Residues in Cereals of Ardo-Kola LGA

Pesticide Residues	Maize	Millet	Rice	Sorghum	TCC	PCC	PL
Isopropylamine	0.1109 + 0.001	0.1303 + 0.006	0.1561 + 0.002	0.0010 + 0.000	0.3983	20.03%	0.1
HCB	0.0122 + 0.001	0.0288 + 0.001	0.1653 + 0.004	0.0156 + 0.000	0.2219	11.16%	0.5
Endosulfan	0.0461 + 0.003	0.0000	0.0534 + 0.003	0.0010 + 0.000	0.1005	5.05%	0.5
Aldrin	0.0100 + 0.000	0.0000	0.0158 + 0.000	0.0000	0.0258	1.29%	0.2
DDT	0.0470 + 0.002	0.0038 + 0.000	0.0777 + 0.001	0.0000	0.1285	6.46%	0.01
t-nonachlor	0.0626 + 0.002	0.0164 + 0.000	0.1095 + 0.001	0.0292 + 0.000	0.2177	10.94%	0.4
Carbofuran	0.0000	0.0000	0.1892 + 0.000	0.1061 + 0.000	0.2953	14.85%	NA
Dichlorvos (DDVP)	0.0680 + 0.000	0.0000	0.1463 + 0.002	0.0067 + 0.001	0.2210	11.11%	0.1
Heptachlor	0.0475 + 0.002	0.0013 + 0.000	0.1492 + 0.001	0.0000	0.1980	9.95%	0.01
g-chlordane	0.0000	0.0000	0.1079 + 0.001	0.0733 + 0.003	0.1812	9.11%	0.1
CCC	0.4043	0.1806	1.1704	0.2329	1.9882	99.95%	
PCC	20.33%	9.08%	58.87%	11.71%		99.99%	

Where: TCC = Total Contaminant Concentration, CCC = Cumulative Contaminant Concentration, PCC = Percentage Contaminant Concentration, and PL = Permissible Limit.

Table 3. Mean Concentrations (mg kg^{-1}) of Pesticides Residues in Cereals of Karim-Lamido LGA

Pesticide Residues	Maize	Millet	Rice	Sorghum	TCC	PCC	PL
Isopropylamine	0.1716 + 0.003	0.1499 + 0.002	0.1613 + 0.004	0.0013 + 0.000	0.4841	21.12%	0.1
HCB	0.0188 + 0.002	0.0301 + 0.000	0.1725 + 0.002	0.0161 + 0.000	0.2375	10.36%	0.5
Endosulfan	0.0747 + 0.004	0.0000	0.0558 + 0.003	0.0000	0.1305	5.69%	0.5
Aldrin	0.0155 + 0.001	0.0000	0.0164 + 0.000	0.0000	0.0319	1.39%	0.2
DDT	0.0743 + 0.004	0.0000	0.0807 + 0.001	0.0000	0.1550	6.76%	0.01
t-nonachlor	0.0997 + 0.001	0.0172 + 0.000	0.1160 + 0.000	0.0299 + 0.000	0.2628	11.46%	0.4
Carbofuran	0.1065 + 0.000	0.0000	0.1918 + 0.002	0.0000	0.2983	13.01%	NA
Dichlorvos (DDVP)	0.1187 + 0.003	0.0000	0.1498 + 0.000	0.0089 + 0.000	0.2774	12.10%	0.1
Heptachlor	0.0755 + 0.004	0.0000	0.1514 + 0.001	0.0000	0.2269	9.90%	0.01
g-chlordane	0.0771 + 0.002	0.0000	0.1100 + 0.000	0.0000	0.1871	8.16%	0.1
CCC	0.8324	0.1972	1.2057	0.0562	2.2915	99.95%	
PCC	36.32%	8.60%	52.61%	2.45%		99.80%	

Where: *TCC = Total Contaminant Concentration, CCC = Cumulative Contaminant Concentration, PCC = Percentage Contaminant Concentration, and PL = Permissible Limit.*

Table 4. Mean Concentrations (mg kg^{-1}) of Pesticides Residues in Cereals of Zing LGA

Pesticide Residues	Maize	Millet	Rice	Sorghum	TCC	PCC	PL
Isopropylamine	0.0696 + 0.000	0.0724 + 0.001	0.0941 + 0.003	0.0080 + 0.000	0.2441	20.10%	0.1
HCB	0.0069 + 0.000	0.0171 + 0.000	0.1004 + 0.000	0.0097 + 0.000	0.1341	11.04%	0.5
Endosulfan	0.0289 + 0.001	0.0000	0.0305 + 0.000	0.0009 + 0.000	0.0603	4.96%	0.5
Aldrin	0.0091 + 0.000	0.0000	0.0098 + 0.000	0.0000	0.0189	1.55%	0.2
DDT	0.0291 + 0.000	0.0024 + 0.000	0.0470 + 0.001	0.0000	0.0785	6.46%	0.01
t-nonachlor	0.0373 + 0.001	0.0108 + 0.000	0.0642 + 0.006	0.0170 + 0.001	0.1293	10.64%	0.4
Carbofuran	0.0000	0.0000	0.1187 + 0.001	0.0629 + 0.005	0.1816	14.95%	NA
Dichlorvos (DDVP)	0.0417 + 0.000	0.0000	0.0906 + 0.000	0.0057 + 0.000	0.1380	11.36%	0.1
Heptachlor	0.0282 + 0.000	0.0012 + 0.000	0.0920 + 0.001	0.0000	0.1214	9.99%	0.01
g-chlordane	0.0000	0.0000	0.0641 + 0.004	0.0438 + 0.000	0.1079	8.88%	0.1
CCC	0.2508	0.1039	0.7114	0.1480	1.2141	99.93%	
PCC	20.65%	8.55%	58.59%	12.19%		99.98%	

Where: *TCC = Total Contaminant Concentration, CCC = Cumulative Contaminant Concentration, PCC = Percentage Contaminant Concentration, and PL = Permissible Limit.*

HCB was found above the permissible limit in rice; DDT in maize and rice; t-nonachlor in rice; and Heptachlor in maize and rice, while the rest of the PR concentrations were below the permissible limits in all food items. Cumulative Contaminant Concentration (CCC) was highest in rice (0.7114 mg kg-1), followed by maize (0.2508 mg kg-1), sorghum (0.1480 mg kg-1), and millet (0.1039 mg kg-1). TCC was as follows: Isopropylamine 0.2441 mg kg-1 (20.10%) > carbofuran 0.1816 mg kg-1 (14.95%) > dichlorvos 0.1380 mg kg-1 (11.36%) > HCB 0.1341 mg kg-1 (11.04%) > t-nonachlor 0.1293 mg kg-1 (10.64%) > heptachlor 0.1214 mg kg-1 (9.99%) > g-chlordane 0.1079 mg kg^{-1} (8.88%) > DDT 0.0785 mg kg^{-1} (6.46%) > endosulfan 0.0603 mg kg^{-1} (4.96%) > aldrin 0.0189 mg kg^{-1} (1.55%).

The concentrations of pesticide residue recorded in this study might be portraying a trend in the differential bioaccumulation potentials of these food items for the contaminants. This could also be a result of the different formulations of the pesticide residues in the chemical pesticides used in the farms prior to this study, which is an important function of the availability of such pesticide residues in the soils of the study area. Omeje [32] believed that these trends in pesticide residues, especially when taking into account their degrees of contamination found in their study, may be depicting certain of the examined food crops with greater concentrations as more sensitive to the bioaccumulation process compared to others. The result obtained could also mean that the quest for more staple foods, especially the yearning of the government to make rice more available to meet the rising demands, has allowed farmers to probably apply more pesticides to rice fields to realize mega harvests and, in the process, make more contaminants available to the soil medium for plant uptake and subsequent bioaccumulation. Hence, there are more contaminants in rice than in maize, sorghum, and millet. Consumer preference for food types could also have been a contributing factor, with demands for rice and maize being higher than those for sorghum and millet; hence, the need to apply more pesticides to rice and maize and realize a larger harvest than for sorghum and millet. Susceptibility to diseases and pest attacks may also have contributed to the presence of pesticide residues in food, indicating that rice and maize are more susceptible to pests and diseases than sorghum and millet, resulting in the application of more pesticides to rice and maize, resulting in a higher concentration than in sorghum and millet. Treatments of crops with different pesticides usually result in multiple residues in the samples [39,40]. In this study, most of the pesticides detected are above the permissible limits, which is in contrast to [40], whose findings reported that even at their highest concentrations, the PRs were below the permissible limits. The presence of pesticide residues in cereal food is a major concern for consumers due to toxic adverse health effects, especially for children who consume a high quantity of cereal meal and its related products [41]. [42] Reported pesticide residue contamination of food items in Osun State, Nigeria, at various concentration levels. [43] In Ondo State, Nigeria, investigated PRs in *Phaseolus vulgaris* and reported higher levels of contamination, some above the permissible limits.

3.2 Pesticide Residues (PRs) in Soils of the Study Area

3.2.1 Pesticide residues (PRs) percentage concentrations in soils of Ardo-kola: In Fig. 1, the concentration of isopropylamine is highest (72.52%) in the millet farm and lowest (0.47%) in the sorghum farm. HCB was detected in all farms. The millet farm had the highest concentration (15.79%), while the maize farm had the lowest (3.26%). Endosulfan was recorded in maize, rice, and sorghum farms, with concentrations of (11.07%) and (0.49%) as the highest and lowest in maize and sorghum farms, respectively, while none was detected in millet farms. Aldrin was only recorded in maize farms as the highest (2.72%) and rice farms as the lowest (1.49%), while it was undetected in millet and sorghum farms. DDT was not recorded in sorghum farms but was found in maize, millet, and rice farms, with maize farms having the highest (12.29%) and millet farms having the lowest (2.21%) concentrations. t-nonachlor was detected in all cereal farms of Ardo-kola, with the maize farm recording the highest (15.48%) while the millet farm recorded the lowest (8.73%). Carbofuran was only detected in rice and sorghum farms, with sorghum farms being the highest (44.49%), while rice farms recorded the lowest (17.12%). Dichlorvos was found in three (3) farms, namely, maize, rice, and sorghum, with the concentration on the maize farm being the highest (16.62%) and that of the sorghum farm being the lowest (3.63%). Heptachlor was undetected in sorghum farms but had a high concentration (13.83%) in rice farms and the lowest (0.73%) concentration in maize farms. g-chlordane was only present in Sorghum and Rice farms at the highest (31.33%) and lowest (10.07%) levels, respectively, but was not detected in Maize and Millet farms. It can be seen also from the Fig. 1 that there is a significant difference in mean concentration of pesticides residue from both soils and cereals at different p values.

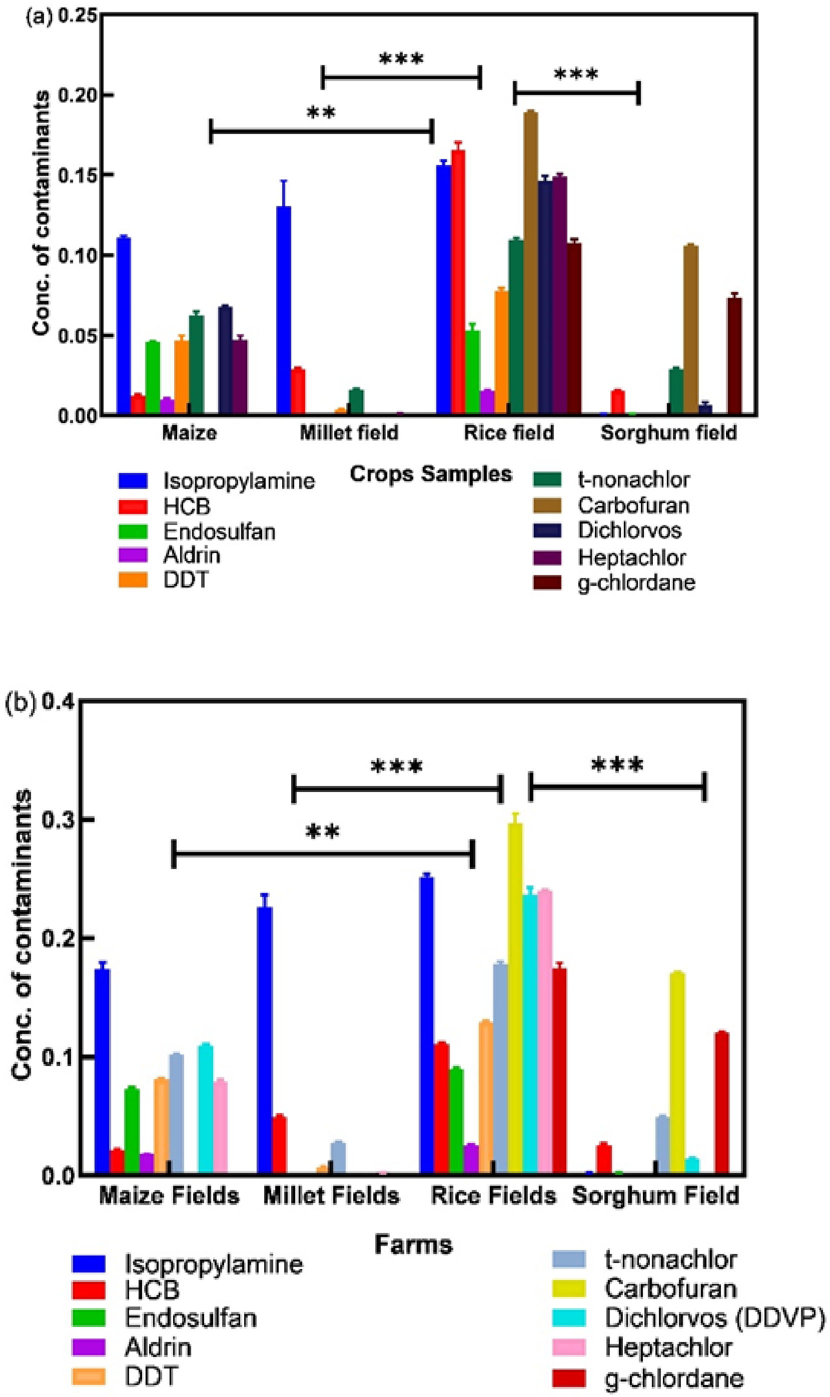

Fig. 1. Rate of Contamination of Individual Contaminant (mg kg^{-1}) in (a) Cereals and (b) Soils of Ardo-kola LGA

3.2.2 Pesticide residues (PRs) percentage concentrations in soils of Karim-Lamido:

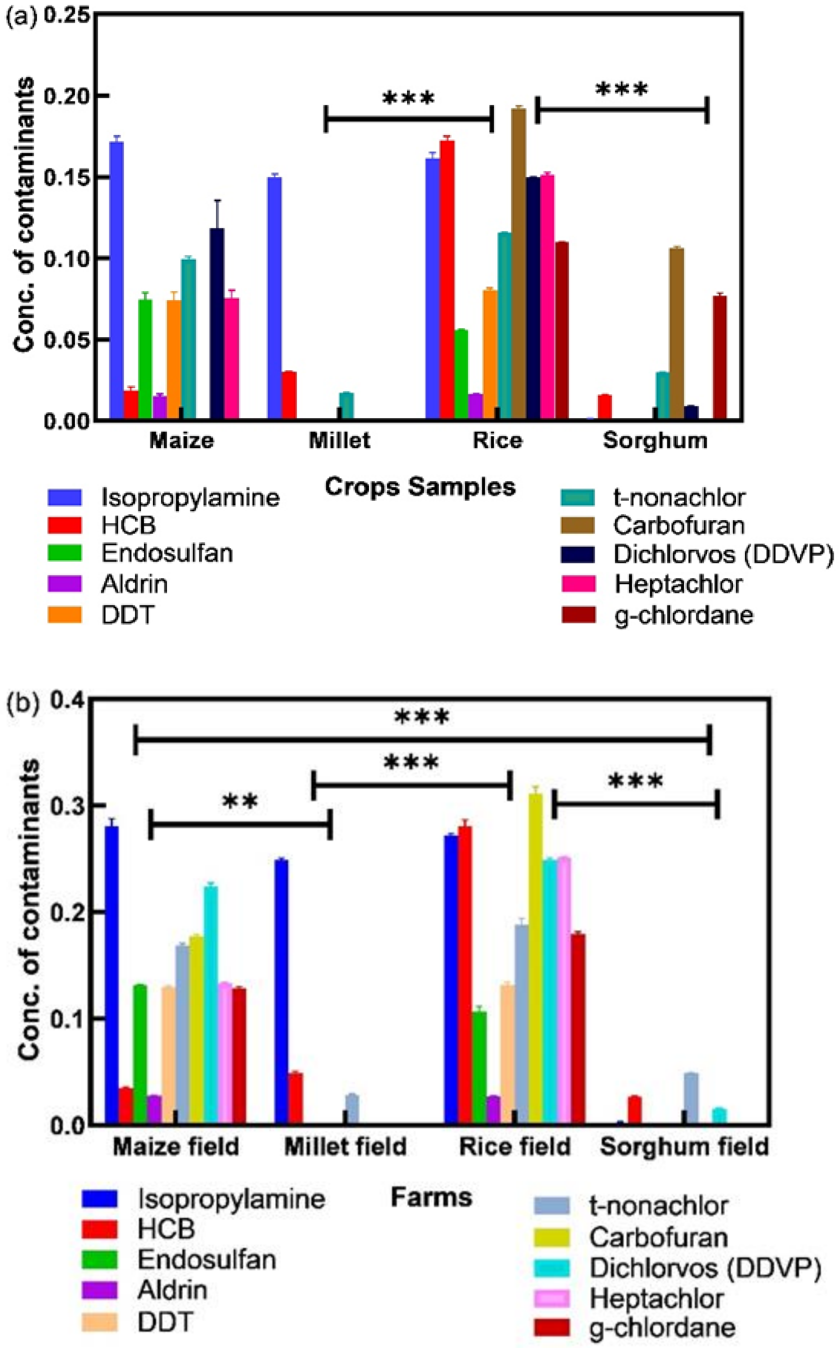

Fig. 2. Rate of Contamination of Individual Contaminant (mg kg^{-1}) in (a) Cereals of Karim-Lamido LGA and (b) Soils of Karim-Lamido LGA

Fig. 2 presents the PRs percentage concentration in the soils of Karim-Lamido LGA. And it can be deduced that isopropylamine was detected in all farms, with the highest concentration (76.3%) in the millet farm and the least concentration (2.97%) in the sorghum farm. HCB was also present in all four (4) farms, with the highest concentration (28.48%) in the sorghum farm and the least concentration (2.45%) in the maize farm. Endosulfan was detected in only maize and rice farms, the highest (9.14%) and lowest (5.35%), respectively; none was detected in millet and sorghum farms. Aldrin was seen in maize farms (1.94%) and rice farms (1.35%) as the highest and lowest, respectively, while it was undetected in millet and sorghum farms. DDT followed the pattern observed with Aldrin, as it is present in only maize (8.99%) and rice (6.58%), the highest and lowest concentrations, respectively, while remaining undetected in both millet and sorghum farms. t-nonachlor was detected in all farms, with the highest concentration (51.85%) in the sorghum farm and the lowest concentration (8.60%) in the millet farm. Carbofuran, Heptachlor, and G-chlordane were detected only in rice and maize farms at concentrations of 15.57% and 12.32%, 12.58% and 9.28%, and 8.99% and 8.97%, respectively. While Dichlorvos was detected in three (3) farms, which include maize, rice, and sorghum, with the sorghum farm being the highest with 16.68% and the rice farm the lowest with 12.45%. Fig. 2 also showed significant values at different level of probability.

3.2.3 Pesticide residues (PRs) percentage concentrations in soils of Zing:

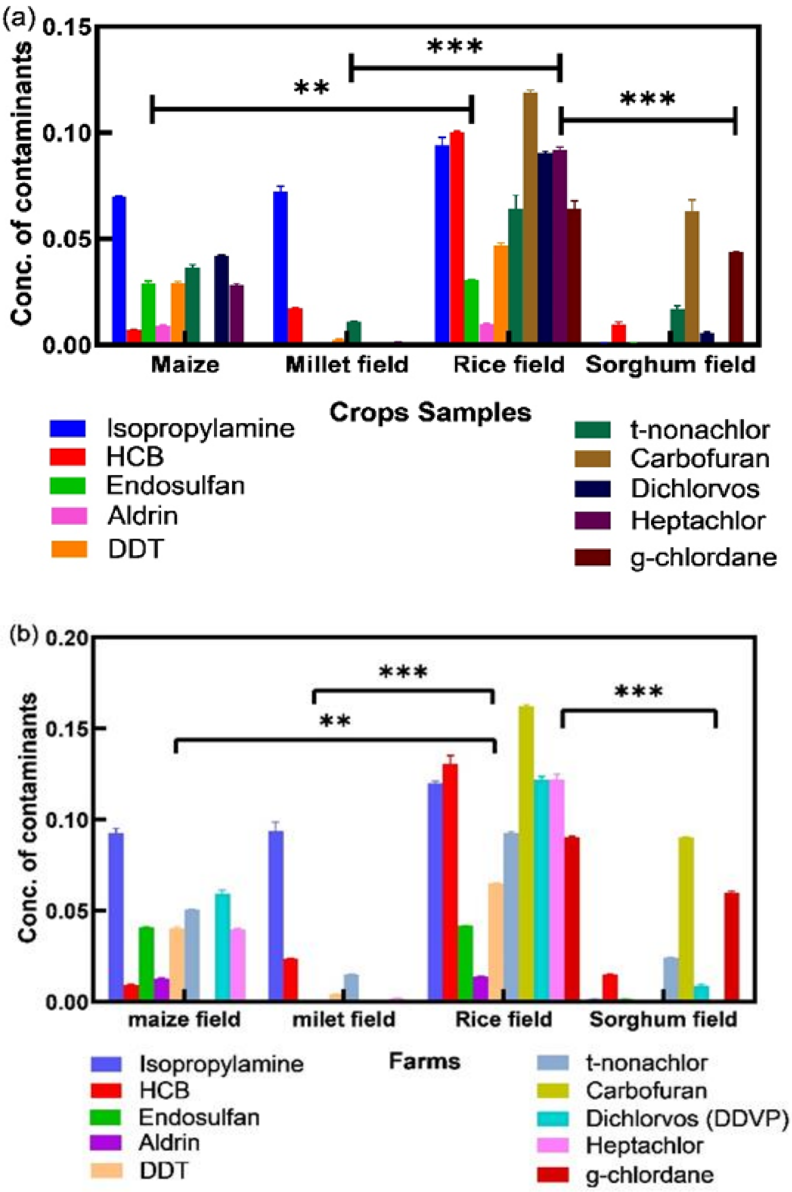

Fig. 3. Rate of Contamination of Individual Contaminant (mg kg^{-1}) in (a) Cereals and (b) Soils of Zing LGA

The PRs percentage concentrations of Zing are as seen in Fig. 3, and it can be deduced that isopropylamine, HCB, and t-nonachlor were detected in all of the study farms. Isopropylamine was highest (67.72%) in millet farms and lowest (0.79%) in sorghum farms. HCB was found to be highest (17.07%) in millet farms and lowest (2.71%) in maize farms, while t-nonachlor was highest (14.68%) in maize farms and lowest (9.65%) in rice farms. Endosulfan, DDT, dichlorvos, and heptachlor were detected in three (3) farms, with peak and low concentrations of 11.82% and 0.74% in maize and rice farms, 11.71% and 2.88% in maize and millet farms, 17.13% and 4.46% in maize and rice farms, and 12.71% and 1.44% in rice and maize farms, respectively. Aldrin, carbofuran, and g-chlordane were detected in only two farms. Aldrin was highest (3.75%) in maize farms and lowest (1.46%) in rice farms; carbofuran was highest in sorghum farms (44.71%) and lowest in rice farms (16.83%); and g-chlordane was also found to be highest (29.79%) and lowest (9.42%) in sorghum and rice farms, respectively, with the Fig .3 showing statistical difference at various level of probability.

Isopropylamine, DDT, t-nonachlor, heptachlor, and g-chlordane were the most prevalent across all LGAs (Fig. 4), with 21.18%, 7.0%, 11.50%, 10.41%, and 9.71%, respectively, in Ardo-Kola. Zing recorded the highest percentage concentration in HCB (10.86%), aldrin (1.64%), and carbofuran (15.28%), while endosulfan (6.18%) and dichlorvos (12.67%) were the highest in Karim-Lamido. Fig. 4 below showed that there is a significant difference between cereals of Zing and Karim-Lamido (**), cereals of Zing and Ardo-Kola (*) but no significant difference between cereals of Karim-Lamido and Ardo-Kola.

Generally, during the study, total contaminant concentrations in the study area were compiled and presented in Fig. 5. This revealed that the most abundant contaminant during this study was isopropylamine (21%), followed by carbofuran (14%), dichlorvos (12%), t-nonachlor (11%), heptachlor (10%), HCB and g-chlordane (both recording 9%), DDT (7%), endosulfan (6%), and aldrin (1%).

The differential concentration of PRs recorded in this study might not be unconnected to the indiscriminate use of these pesticides by farmers in resolving pest challenges on their farms. This has led to the use of whatever pesticides were available on the market without actually paying attention to the contents of the pesticides. In agricultural areas of Borno State, Nigeria, Chlorpyrifos, Diazinon, Dichlorvos, and Fenitrothionin residues in several plants were found, according to research by [44]. It could also be the result of multiple pesticide applications to address pest problems without settling on a single one. It might also be that due to poor legislation, product control, and monitoring, some manufacturers of these products have developed ways of creating multiple formulations that make these residues available in their products. Many research studies have documented residual pesticides in different soil types. p,p'-DDT levels in soil samples were found to be lower than those of its metabolites (p,p'-DDD, p,p'-DDE, and p,p'-methoxychlor), indicating that p,p'-DDT was

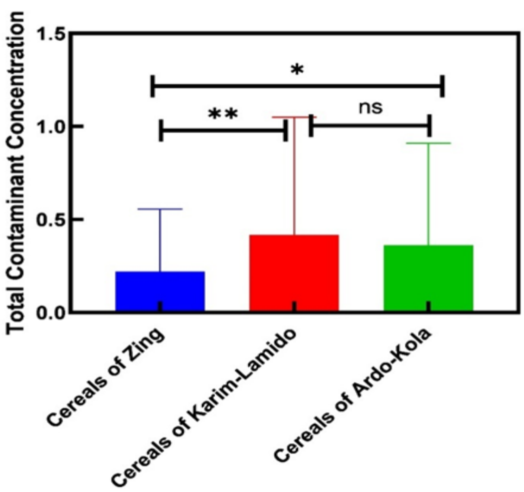

Fig. 4. The comparison of total Contaminant Concentration (TCC) in Cereals of all three LGA

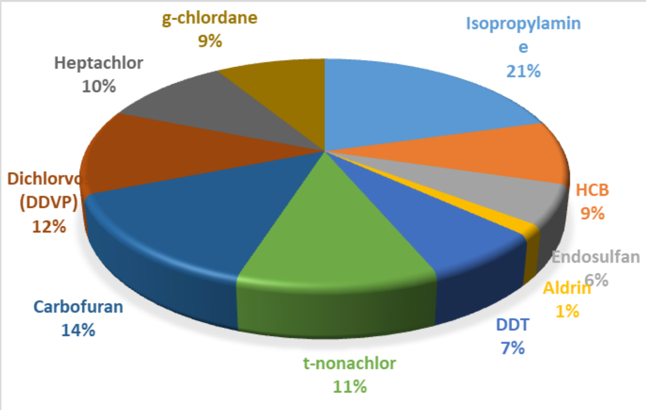

Fig. 5. Rate of Contamination of Individual Contaminant (mg kg^{-1}) in Soils of Taraba North

a pesticide component used in the farm over time [44,45]. [45] Reported higher residues of heptachlor epoxide than heptachlor, which is an indication that heptachlor had probably been transformed into heptachlor epoxide, signifying the higher stability of these derivatives. This could also be a confirmation that heptachlor epoxide could be resistant to environmental degradation, and because of its persistent nature, heptachlor should have been phased out of agrochemical stores. However, its presence today is an indication that it is being marketed under false names or labels or that it is being added to other pesticides currently in use by Nigerian farmers as part of the many active ingredients. [46] Reported seventeen (17) pesticide residues and derivatives that include; 2,4 dichloro, hexachlorobenzene (HCB), endosulfan, aldrin, *p,p'*-DDD, g- chlordane, profenofos, carbofuran (a carbamate), DDVP, dichlorvos, heptachlor, t-nonachlor, isopropylamine, glyphosate, biphenyl, dichlorobiphenyl and lindane. These PRs were found to be within the maximum residue limit hence they suggested indiscriminate uses as a contributing factor.

3.3 Health Implications

This shows the potential health consequences of consuming cereals from each of the study areas during the period of the study. This was determined as explained by equations 1 and 2, using the daily intake for cereals as 1,120 kg and 0.3733 kg [40], the oral reference dose (RfD) as in Table 1, the body weight (Bo) as 70 kg for adults and 19.25 kg for children [41], and the contaminant concentrations as in Tables 2, 3, and 4. Hazard values greater than or equal to one (1) indicate a higher risk to one's health. During the period of study, the Hazard Quotient (HQ) revealed that children are at greater risk of toxicity than adults.

3.3.1 Health implications of consuming cereals from Ardo-Kola LGA: The health implications due to the consumption of cereals from Ardo-Kola are in the following order: rice > maize > sorghum > millet (Table 5). In adults, rice recorded the highest HQ due to g-chlordane (28.7733) and the least due to isopropylamine (0.0014). In maize HQ, the concentration was highest as a result of the PR aldrin (3.200) and was least due to isopropylamine (0.0010). The highest and lowest HQs in sorghum were due to g-chlordane (23.4560) and endosulfan (0.0027), respectively while the highest and lowest HQs in millet were due to HCB (0.5760) and isopropylamine (0.0012). In children, HQ was highest and lowest in rice, maize, sorghum, and millet due to g-chlordane (34.8737) and isopropylamine (0.0018); aldrin (3.8784) and isopropylamine (0.0012); g-chlordane (23.6908) and t-nonachlor (0.0109); and HCB (0.6981) and isopropylamine (0.0014), respectively.

3.3.2 Health implications of consuming cereals from Karim-Lamido LGA: The health implication of consuming cereals from Karim-Lamido (Table 6) is presented below. It can be deduced from the table that in adults, HQ in rice was highest due to g-chlordane (29.3333) and was lowest as a result of isopropylamine (0.0015). Maize recorded the highest and lowest HQs due to g-chlordane (20.5600) and isopropylamine (0.0016). In millet and sorghum, the highest and lowest HQs were due to HCB (0.6020) and isopropylamine (0.0014); and dichlorvos (0.2848) and HCB (0.0322), respectively. While in children, the highest and lowest HQs in maize, millet, rice, and sorghum were due to g-chlordane (24.9190) and isopropylamine (0.0019); HCB (0.7296) and isopropylamine (0.0017); g-chlordane (35.5523) and isopropylamine (0.0018); and HCB (0.3903) and t-nonachlor (0.0112).

3.3.3 The health implications of consuming cereals from Zing LGA: Table 7 shows the implications of consuming cereals from Zing. The table shows that in adults, maize had the highest and lowest HQs due to aldrin (4.8533) and isopropylamine (0.0006); in millet, HCB (0.3420) and isopropylamine (0.0007); in rice, HQs were highest and lowest in g-chlordane (17.0933) and isopropylamine (0.0009); and in sorghum, g-chlordane and endosulfan recorded highest (11.6800) and lowest (0.0024) HQs respectively. In children, the highest and lowest HQs in maize were due to aldrin (5.8823) and isopropylamine (0.0008). HCB (0.4145) and isopropylamine (0.0008) are the highest and lowest HQs in millet. In rice, g-chlordane (20.7173) was responsible for the highest HQ, while isopropylamine (0.0010) had the lowest. In sorghum, the highest and lowest HQs were due to g-chlordane (14.1563) and t-nonachlor (0.0063).

To safeguard consumers' health from pesticide toxicity through food consumption, it is significant to evaluate the hazards associated with such pesticide exposure through consumption. During the period of this study, Hazard Quotient and Hazard Index were evaluated as described by the [33], and the results obtained showed that the food items investigated are indeed contaminated with HI values greater than 1 in most cases. This could not be far from a long-term accumulation of these pesticides in both the soils and seeds of the food items. DDTs, endosulfans, and chlordane in soil from Thailand and China mostly came from past application, according to the source evaluation of OCP residues by [47]. This could be the case found in soil and cereals of Taraba north as it is a tradition that some of the harvest from the previous year is stored and used as seed for planting in the coming planting season. And when such seeds are already contaminated from the previous planting season, they tend to bioaccumulate more contaminants when planted the next season, increasing the chances of exceeding the permissible levels of these contaminants in food for human consumption. The assessment of the health risks associated with pesticide residues in several food crops in Ejura, Ghana, and discovered that maize was polluted with pyrethroid, organochlorine, and organophosphate pesticides [48]. These were represented by Aldrin, Dieldrin, Heptachlor, Endrin, y-chlordane, and Chlorfenvinphos in maize, as well as Heptachlor and p,p-DDD in cowpea, which were all viewed as potentially systemically harmful candidates for consumers. The dangers that pesticide residues bring to human health should not be ignored, as stressed by [49]. Aldrin, for instance, may result in higher metabolism of d-glutaric acid. Skin sensitization, enzyme induction, allergic reactions/rash, and contact dermatitis are all possible side effects of lindane. Moreover, HCB can result in skin atrophy, bulb development, irreversible hair loss, and photosensitivity [34].

The result obtained in this study could also be that, due to the farmers' eagerness to realize and achieve quicker solutions, they have doubled the concentrations of the pesticide in their spraying tanks and, hence, made more contaminants available to the crops for bioaccumulation than usual, which will invariably increase the concentration of the contaminants in the food products. [50] Reported a HI 1, indicating that pesticide residues in the crops they studied caused long-term harm to human health. They explained that consumption of pesticide-tainted food crops has the potential to cause cancer in children and adults, with risks exceeding one in a million people. They also reported that aldrin, dieldrin, chlordane, p,p'-DDT, p,p'-DDD, p,p'-DDE, heptachlor, and heptachlor epoxide could have a cancerous influence on the health of children who consume food crops. Due to their lifetime exposure to these residues in food crops, people are at risk of acquiring various ailments. Thus, looking at the significance of the studied food crops, applications of recalcitrant and highly toxic pesticides on farmlands where these crops are planted should be done with the utmost caution [51].

Children might be more prone or susceptible to toxicity because they are more exposed to toxic substances in the environment than adults. They breathe, drink, and eat more than adults, and their behavioral patterns, that include playing in the fields, usually increases their chances of exposure to potentially toxic substances. Furthermore, children are likely to be more vulnerable to environmentally acquired hazards or toxins because, their systems are continually developing, which most often are incapable of metabolizing, detoxifying, and excreting toxic substances. It is also worth noting that for developmental toxicants, a safe dose for an adult could have lethargic effects in a child [52].

Table 5. Health Hazard of Consuming Cereals from Ardo-Kola LGA

	ADULTS				CHILDREN			
Pesticide Residue	**Maize**	**Millet**	**Rice**	**Sorghum**	**Maize**	**Millet**	**Rice**	**Sorghum**
Isopropylamine	0.0010	0.0012	0.0014	0.0000	0.0012	0.0014	0.0018	0.0000
HCB	0.2440	0.5760	3.3060	0.3120	0.2957	0.6981	4.0069	0.3781
Endosulfan	0.1480	0.0000	0.1448	0.0027	0.1490	0.0000	0.1587	0.0323
Aldrin	3.2000	0.0000	8.4267	0.0000	3.8784	0.0000	10.2132	0.0000
DDT	1.5040	0.1216	2.4864	0.0000	1.8229	0.1474	3.0135	0.0000
t-nonachlor	0.0701	0.0814	0.1223	0.0327	0.0234	0.0061	0.0409	0.0109
Carbofuran	0.0000	0.0000	0.6054	0.3395	0.0000	0.0000	0.7333	0.3429
Dichlorvos (DDVP)	2.1760	0.0000	4.6816	0.2144	2.6373	0.0000	5.6742	0.2599
Heptachlor	1.5200	0.0416	4.7744	0.0000	1.8423	0.0504	5.7866	0.0000
g-chlordane	0.0000	0.0000	28.7733	23.4560	0.0000	0.0000	34.8737	23.6908
Hazard Index	**10.9964**	**0.8218**	**53.3223**	**24.3573**	**10.6502**	**0.9034**	**64.5028**	**24.7149**

Hazard Index (HI) was in the order rice (53.3223) > sorghum (24.3573) > maize (10.9964) > millet (0.8218) in adults and children the order is rice (64.5028) > sorghum (24.7149) > maize (10.6502) > millet (0.9034)

Table 6. Health Hazard of Consuming Cereals from Karim-Lamido

	ADULTS				CHILDREN			
Pesticide Residue	**Maize**	**Millet**	**Rice**	**Sorghum**	**Maize**	**Millet**	**Rice**	**Sorghum**
Isopropylamine	0.0016	0.0014	0.0015	0.0000	0.0019	0.0017	0.0018	0.0000
HCB	0.3760	0.6020	3.4500	0.0322	0.4557	0.7296	4.1814	0.3903
Endosulfan	0.1992	0.0000	0.1488	0.0000	0.2414	0.0000	0.1803	0.0000
Aldrin	8.2667	0.0000	8.7467	0.0000	10.0193	0.0000	10.6011	0.0000
DDT	2.3776	0.0000	2.5824	0.0000	2.8817	0.0000	3.1299	0.0000
t-nonachlor	0.1117	0.0193	0.1299	0.0335	0.0372	0.0064	0.0433	0.0112
Carbofuran	0.3408	0.0000	0.6138	0.0000	0.4131	0.0000	0.7349	0.0000
Dichlorvos (DDVP)	3.7984	0.0000	4.7936	0.2848	4.6037	0.0000	5.8099	0.3452
Heptachlor	2.4160	0.0000	4.8448	0.0000	2.9282	0.0000	5.8720	0.0000
g-chlordane	20.5600	0.0000	29.3333	0.0000	24.9190	0.0000	35.5523	0.0000
Hazard Index	**38.448**	**0.6227**	**54.6448**	**0.3505**	**46.5012**	**0.7377**	**66.1069**	**0.7467**

Hazard Index (HI) for both adults and children in maize, millet, rice, and sorghum were 38.4480, 0.6227, 54.6448, and 0.3505, 46.5012, 0.7277, 66.1069, and 0.7467

Table 7. Health Hazard of Consuming Cereals from Zing

	ADULTS				CHILDREN			
Pesticide Residue	**Maize**	**Millet**	**Rice**	**Sorghum**	**Maize**	**Millet**	**Rice**	**Sorghum**
Isopropylamine	0.0006	0.0007	0.0009	0.0000	0.0008	0.0008	0.0010	0.0000
HCB	0.1380	0.3420	2.0080	0.1940	0.1673	0.4145	2.4337	0.2351
Endosulfan	0.0771	0.0000	0.0813	0.0024	0.0934	0.0000	0.0986	0.0029
Aldrin	4.8533	0.0000	5.2267	0.0000	5.8823	0.0000	6.3347	0.0000
DDT	0.9312	0.0768	1.5040	0.0000	1.1286	0.0931	1.8229	0.0000
t-nonachlor	0.0418	0.0121	0.0719	0.0190	0.0139	0.0040	0.0240	0.0063
Carbofuran	0.0000	0.0000	0.3798	0.2013	0.000	0.0000	0.4604	0.2440
Dichlorvos (DDVP)	1.3344	0.0000	2.8992	0.1824	1.6173	0.0000	3.5139	0.2211
Heptachlor	0.9024	0.0384	2.9440	0.0000	1.0937	0.0465	3.5682	0.0000
g-chlordane	0.000	0.0000	17.0933	11.6800	0.0000	0.0000	20.7173	14.1563
Hazard Index	**8.2788**	**0.47**	**32.2091**	**12.2791**	**9.9973**	**0.5589**	**38.9747**	**14.8657**

Hazard Index (HI) was in the order rice (32.2091) > sorghum (12.2791) > maize (8.2788) > millet (0.4700) in adults and the same order of rice (38.9747) > sorghum (14.8657) > maize (9.9973) > millet (0.5589) in children

4. CONCLUSION

Although the use of OCP pesticides was abolished globally several decades ago, OCP residues were still identified in cereals and soil from Taraba north. This investigation of the pesticide residues in cereals from Taraba north, revealed a lack of compliance to the OCPs ban and the weakness of policy implementation by the regulating body as OCPs, OPPs, and PYRs were detected in varying concentrations. This also presented samples with above permissible limits of the PRs. As population increases, more demand will be place on food supply and more pesticides introduced to insure yield. This extremely significant information can be used to identify a group of food contaminants for additional toxicological research and improved consumer risk assessment.

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CONSENT FOR PUBLICATION

All Authors have consented to this publishing this article in the Beni Suef Journal of Basic and Applied Sciences

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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