1 **Original Research Article**

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3 Effects of Processing on the Levels of Pesticides in 4 some Commonly Consumed Meats from Sagamu, 5 South-western Nigeria

6

7 **ABSTRACT**

8 Technological and kitchen processes can partially or fully remove or degrade pesticide residues to other
9 compounds often less toxic, which makes safer products for human consumption. This study was
10 conducted to determine the effects of processing on levels of pesticides in some commonly consumed
11 meat in Nigeria.

12 Cow, goat and pork muscles were purchased from three abattoirs in Sagamu, South-western Nigeria.
13 Each meat sample was separately packaged in a polyethylene bag and transported to the laboratory
14 immediately, where they were processed (boiled and fried) on the same day. Samples of raw, boiled and
15 fried meat were extracted and cleaned up before being quantitatively analyzed using gas chromatograph
16 with pulsed flame photometric detector.

17 The results showed that 35 pesticide residues were detectable in all the meat samples. However, only 10
18 of them were significantly affected by the processing methods. The levels of some organochlorine
19 pesticide (OCPs) residues and an organophosphorus pesticide (OPP) residue were relatively higher in all
20 the meat samples but were significantly (P<0.05) decreased by the processing methods; with frying being
21 better. The levels of carbamate, triazine, chlorophenoxy, dinitroanilin, chloroalkylthio, benzoylurea and
22 phenylurea were somewhat low in all the samples and were not significantly (P>0.05) affected by the
23 boiling and frying methods.

24 The mean levels of all the detected pesticide residues were far below the various internationally set
25 maximum residue limits for meat samples; making the meats analyzed safe for consumption.
26 Furthermore, processing methods greatly depleted pesticide residues, especially OCPs and OPP, in the
27 meats analyzed.

28 *Keywords: Pesticide residues; cow; goat; pork; red meat; boiling; frying; maximum residue limit.*

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1. INTRODUCTION

Pesticides are substances intended for destroying, preventing, repelling, attracting, or controlling any pest and in some cases species of plants or animals that are not desirable at the period of production, storage, transport, distribution and processing of food, agricultural commodities, or animal feeds or those that are given to animals for the control of ectoparasites [1].

Generally, pesticides have been classified relative to target organism and chemical structure. Classes of pesticides according to target organism mainly include herbicides, insecticides, disinfectants, rodenticides and fungicides. In lieu of chemical structure, they have been classified into 6 major groups which are organochlorine (OC), organophosphorus (OP), carbamate, triazine, chlorophenoxy and pyrethroid [2].

Globally, the use of pesticides is increasing due to the need to feed the world's ever-growing population, while the amount of land available for food production is continually limited [1]. The increase in the use of pesticides has simultaneously contributed to the hazards on human health and to environmental pollution. Pesticides and their metabolites find their ways into the human body through the water cycle and especially the food chain. Breeding animals can accumulate persistent organic pollutants from contaminated feed and water, and/or from pesticide application in animal production areas (treatment of cowsheds, pigsties, sheepfolds, warrens and/or treatment of animals themselves) [3]. While pesticide compounds are mostly stored in the fat and muscle of animals, they can also reach other compartments such as the brain, liver and lungs to cause unique damages [4].

Regular surveys on the levels of pesticides in meats have been reported in the developed world. Driss and Bouguerra [5], Garcia-Regueiro *et al*. [6] and Krauthacker *et al*. [7] reported the levels of organochlorine pesticides (OCPs) in some foodstuffs of animal origin from Tunisia, Spain and Yugoslavia respectively. Suzuki *et al*. [8] surveyed pesticide residues in imported Australian meat, while Cantoni *et al*. [9] also reported the OCPs in pork collected from Italy. Furthermore, in a surveillance of marketed foods (including meat products, fish, egg, milk and dairy) in Barcelona, Spain from 2001-2006, Fontcuberta *et al*. [10] discovered a gradual disappearance of regulated OCPs as a consequence of the growing worldwide implementation of current regulatory agreement.

In addition, efforts have been made in some developing countries to evaluate pesticide residues in meats. The contents of OCPs in camel, cattle and sheep carcasses slaughtered in Sharkia province of Egypt

were determined and reported to be well below the respective minimal permissible limits set by local or international organizations [11]. Blankson-Arthur *et al*. [12] reported the levels of OCP residues in grasscutter tissues from Ghana as being below the acceptable maximum residue limits (MRL) of WHO/FAO Codex Alimentarius Commission.

However, limited reports are available in Nigeria on this same subject. The concentrations of OCP residues in cow, pig and goat were stated to be higher than some values reported in similar animals at some other parts of the world but below FAO's maximum residue limit (MRL) [13]. This makes it needful to provide recent information on pesticide residue in meat and thereby protect meat consumers in Nigeria. It has been observed that technological and kitchen processes can partially or fully remove or degrade pesticide residues to other compounds often less toxic, which makes products safer for human consumption [14,15]. A reduction rate of 44% has been detected for DDT after thermal processing of lamb meat cuts [16]. A reduction rate of 60% in lindane contents has been reported for cooked beef meat after heating at 115 ^{o}C for 2 hours [17]. Also, a high reduction rate of 65% in lindane contents has been reported in rabbit meat after boiling for 1.5 hours [18]. However, there is dearth of information on the extent of reduction of specific pesticide residue in cow, pork and goat meats after conventional processing of boiling and frying. More so, limited information also exist on the most effective processing method for the depletion of pesticide residues in meat as consumed in Nigeria. Therefore, this study was designed to investigate the effect of kitchen processing (boiling and frying) on the levels of pesticide residues in some commonly consumed meats (cow, pork and goat) in Nigeria.

2. MATERIALS AND METHODS

2.1 Sampling

Cow, goat and pork meat samples were purchased from three abattoirs in Sagamu, Ogun State, South-western Nigeria. Sampling was done within February 2012. The meat samples were separately packaged in a polyethylene bag and transported to the laboratory immediately. Each pulled raw meat sample of about 5 Kg was sub-divided into three subsamples. A raw subsample was retained that way for analysis. The remaining two subsamples were boiled (without additional condiment) and the broths discarded. One of these boiled subsamples was then further fried. The samples - raw, boiled and fried for each of cow,

pork and goat meat - were then analyzed for various pesticide residues within 72 hours after processing. All samples were stored at -18 ^{o}C before the analysis.

2.2 Chemicals

All reagents used were of analytical grade from Merck group, Darmstadt, Germany. They include, surrogate standard solution, TFE-fluorocarbon, sodium dihydrogen phosphate, disodium hydrogen phosphate, sulphuric acid, sodium hydroxide, sodium chloride, redistilled analytical grade methylene dichloride (dichloromethane), anhydrous sodium sulphate, nitrogen gas, florisil, and hexane.

2.3 Meat Processing

On delivery of the meat samples to the laboratory, they were thoroughly washed, cut into about 100 g pieces and washed again with tap water as available in the laboratory. Both processing methods – boiling and frying followed the conventional procedures used in preparing meat for table consumption in Nigeria. The boiling was done with water at 120 ^{o}C until the pieces were well cooked and tender. The deep frying was done in different oil portion, for each of the meat type at 120 ^{o}C. Frying was achieved within 15 minutes. All the processing methods were carried out without the addition of any ingredient or additive.

2.4 Pesticide Residue Extraction and Clean-up

The pesticide residue extraction and analysis was carried out by following the modified standard test methods of Manual of Analytical Methods for the Analysis of Pesticides Residues in Human Environmental Samples, EPA-600/8-80-038 [19]. The method is as described subsequently:

The sub-sample materials were pulverized using the laboratory milling machine Janke & Kunkel (IKA Labortechnik, Breisgau, Germany). The pulverized samples were stored at -18 ^{o}C until analysis. 10.0 g of the sample was weighed and extracted after the addition of the surrogate standard solution to the sample and later transferred to the extracting bottle that was cocked with TEF-fluorocarbon.

Fifty milliliters of the phosphate buffer was added, followed by pH measurement with the addition of drops of 9 M sulfuric acid or 10 M sodium hydroxide solution for pH adjustment to 7, if necessary. One gram of the sodium chloride salt was added to the sample, sealed and shaken to dissolve the salt. Twenty milliliters of the redistilled analytical grade methylene dichloride (dichloromethane) was measured and added to the sample. The sample was extracted for about 30 minutes. The extract was filtered into the

Erlenmeyer flask. Then, the extraction was repeated two more times with fresh solvent and the filtrates were combined. The combined extract was dried by pouring through a drying column containing a 10 cm column of anhydrous sodium sulphate (previously rinsed with methylene dichloride), and the filtrate was concentrated in the concentrator flask with a stream of nitrogen. The wall of the concentrator flask was rinsed with extracting solvent so as to bring the final volume of the extract to 5 mL. The clean-up of the concentrated extract was carried out using florisil packed into column. The concentrated extract was eluted with hexane and later concentrated to the required final volume of 5 mL.

2.5 Determination of Pesticide Residue Concentrations

The Agilent 6890 gas chromatography (GC) with Pulsed Flame Photometric Detector (PFPD) (Agilent Technologies Inc., Wilmington, Delaware) was used for the quantitative analysis. The conditions of the GC are as presented in Table 1. Fortified samples were used for the determination of percentage recoveries of all the pesticide residues under study. This revealed recoveries above 85% for all residues. The detection limit was 0.1 µg/kg for hexachlorobenzene (HCB), lindane, chloropyrifos, dichlorodiphenyltrichloroethane (DDT), aldrin, heptachlor, endrin, heptachlor epoxide, endosulfan and chlordane, whereas all other residues determined were detectable at 0.01 pg/Kg.

Table 1. Gas chromatography conditions for the pesticide analysis

Parameters	**Specification**
GC:	HP 5890/6890 Powered with HP ChemStationrev.A09.01[1206] Software
Injection temperature:	Split injection
Split Ratio:	20:1
Carrier Gas	Hydrogen
Flow Rate:	1.0mL/min
Inlet Temperature:	250 ^{o}C
Column Type:	HP 5MS
Parameters	Specification
Column Dimensions:	30 m x 0.25 mm x 0.25 µm
Oven Program:	Initial at 60 ^{o}C for 2 minutes First Ramp at 10 ^{o}C/min to 200 ^{o}C Second Ramp at 8 ^{o}C/min to 300 ^{o}C constant at 5 minutes
Detector:	PFPD
Detector Temperature:	300^{o}C
Hydrogen Pressure:	22psi
Compressed Air:	28psi

2.6 Statistical Analysis

All values were reported as means ± standard deviation (SD), and all analysis were carried out in triplicates. Data were subjected to one-way analysis of variance (ANOVA) to determine the differences in and the effects of processing on the multi-residual pesticide contents of the meat samples. Significant differences among the mean were determined by least significant difference (LSD). All data analyses were performed using the statistical package for social sciences (SPSS) version 22.

3. RESULTS

3.1 Effect of Processing on the Levels of Organochlorine and Organophosphorus Pesticide Residues in Cow, Pork and Goat Muscle Samples from Sagamu, Ogun State

The levels of organochlorine pesticides (OCP) and organophosphorus pesticides (OPP) in various meat (muscle) samples as affected by processing methods are presented in Table 2. The table shows the presence of four OPPs and twelve OCPs in the various samples. The levels of OCPs in the raw cow muscle range from 0.76 ng/Kg of alachlor to 221.14 µg/Kg of dichlorodiphenyl trichloroethane (DDT); while the levels of OPPs in the same sample range from 0.73 ng/Kg of phosmet to 143.75 µg/Kg of chlorpyrifos. Comparing the concentration of the various OCPs and OPPs in the three muscle samples, the levels of mollinate, dimethoate, chlorothaloniol, alachlor, metachlor and phosmet are not significantly different ($p>0.05$) from one another. All other OCPs and OPPs are significantly different ($p<0.05$) in concentration in the three muscle samples.

The levels of mollinate, dimethoate, chlorothaloniol, alachlor, metachlor and phosmet are somewhat low in all the samples and are not significantly ($p>0.05$) affected by the processing (boiling and frying) methods. All other OCPs and OPPs are higher in concentrations and were significantly decreased ($p<0.05$) by the processing methods. The result of this study shows a significant reduction in the levels of 9 out of 12 OCP residues determined in the cow, pork and goat muscle samples. In all, the frying method diminished the pesticide residues better (Table 2).

Table 2. Effect of processing on the levels (ng/Kg edible portion on dry weight basis) of organochlorine and organophosphorus pesticides in commonly consumed meat (muscle) samples in Nigeria

S/N	Pesticide	Cow			Pork			Goat		
		Raw	Boiled	Fried	Raw	Boiled	Fried	Raw	Boiled	Fried
1.	Mollinate[#]	1.48±0.05[a]	1.48±0.07[a] (0.0)	1.48±0.05[a] (0.0)	1.50±0.04[a]	1.50±0.10[a] (0.0)	1.50±0.08[a] (0.0)	1.46±0.06[a]	1.46±0.03[a] (0.0)	1.46±0.09[a] (0.0)
2.	Dimethoate[#]	1.25±0.07[a]	1.25±0.03[a] (0.0)	1.25±0.05[a] (0.0)	1.30±0.05[a]	1.30±0.03[a] (0.0)	1.30±0.09[a] (0.0)	1.22±0.02[a]	1.22±0.03[a] (0.0)	1.22±0.02[a] (0.0)
3.	Chlorothaloniol[*]	2.71±0.04[a]	2.71±0.04[a] (0.0)	2.71±0.03[a] (0.0)	2.65±0.05[a]	2.65±0.04[a] (0.0)	2.65±0.04[a] (0.0)	2.61±0.07[a]	2.61±0.05[a] (0.0)	2.61±0.09[a] (0.0)
4.	Alachlor[*]	0.76±0.01[a]	0.76±0.02[a] (0.0)	0.76±0.02[a] (0.0)	0.79±0.02[a]	0.79±0.03[a] (0.0)	0.79±0.01[a] (0.0)	0.74±0.01[a]	0.74±0.02[a] (0.0)	0.74±0.01[a] (0.0)
5.	Metachlor[*]	0.98±0.04[a]	0.98±0.02[a] (0.0)	0.98±0.05[a] (0.0)	0.95±0.04[a]	0.95±0.02[a] (0.0)	0.95±0.04[a] (0.0)	0.96±0.04[a]	0.96±0.04[a] (0.0)	0.96±0.06[a] (0.0)
6.	Phosmet[#]	0.73±0.02[a]	0.73±0.04[a] (0.0)	0.73±0.04[a] (0.0)	0.75±0.06[a]	0.74±0.02[a] (1.4)	0.74±0.04[a] (1.4)	0.74±0.03[a]	0.73±0.04[a] (1.4)	0.73±0.06[a] (1.4)

Values represent mean ± standard deviation (SD), n=3

Values (of specific pesticide residue) with different alphabets for each animal are significantly different (at $p<0.05$) from one another as affected by processing method

[*] - Organochlorine pesticide; [#] - Organophosphorus pesticides

Values in ( ) represent percentage reduction in pesticide levels, relative to the raw samples

Table 2. Effect of processing on the levels (µg/Kg edible portion on dry weight basis) of organochlorine and organophosphorus pesticides in commonly consumed meat (muscle) samples in Nigeria (continued)

S/N	Pesticide	Cow			Pork			Goat		
		Raw	Boiled	Fried	Raw	Boiled	Fried	Raw	Boiled	Fried
7.	HCB[*]	4.51±0.10[a]	3.96±0.05[b] (12.2)	3.66±0.22[c] (18.8)	4.30±0.03[a]	3.91±0.01[b] (9.1)	3.33 ±0.02[c] (22.6)	2.80±0.17[a]	2.49±0.02[b] (11.1)	1.59±0.07[c] (43.2)
8.	Lindane[*]	3.99± 0.05[a]	3.63± 0.06[b] (9.0)	1.50± 0.01[c] (62.4)	1.70± 0.02[a]	1.49± 0.03[b] (12.4)	1.06± 0.01[c] (37.6)	1.48±0.03[a]	1.30±0.03[b] (12.2)	1.02±0.00[c] (31.1)
9.	Chlorpyrifos[#]	143.75±0.30[a]	138.34± 0.40[b] (3.8)	101.58±0.63[c] (29.3)	148.10±0.34[a]	133.34±0.44[b] (6.7)	102.35±0.04[c] (30.9)	115.10±0.10[a]	99.22±0.68[b] (13.8)	72.01±0.16[c] (37.4)
10.	DDT[*]	221.14±0.35[a]	197.17± 0.50[b] (10.8)	138.04±0.48[c] (37.6)	166.63±0.23[a]	150.94±0.36[b] (9.4)	112.30±0.06[c] (32.6)	98.21± 0.27[a]	63.95±0.35[b] (34.9)	56.54±0.50[c] (42.4)
11.	Aldrin[*]	2.28± 0.06[a]	1.88± 0.06[b] (17.5)	1.49±0.03[c] (34.6)	5.13±0.05[a]	4.71± 0.10[b] (8.2)	3.70±0.06[c] (27.9)	4.67±0.05[a]	3.73±0.04[b] (20.1)	2.30± 0.04[c] (50.7)

12.	Heptachlor*	0.91± 0.01[a]	0.82± 0.01[b] (9.9)	0.64±0.05[c] (29.7)	1.80± 0.03[a]	1.62±0.03[b] (10.0)	1.12± 0.02[c] (37.8)	1.55±0.02[a]	1.45±0.03[b] (6.5)	1.22±0.01[c] (21.3)
13.	Endrin*	3.76± 0.02[a]	3.16± 0.12[b] (16.0)	2.55±0.12[c] (32.2)	2.35±0.04[a]	2.20±0.02[b] (6.4)	1.52±0.01[c] (35.3)	2.05±0.07[a]	1.79±0.04[b] (12.7)	1.15±0.03[c] (43.9)
14.	Hept.Epoxide*	2.54±1.89[a]	2.21±0.09[b] (13.0)	1.69±0.01[c] (33.5)	2.20±0.06[a]	2.0±0.04[b] (9.1)	1.71±0.03[c] (22.3)	2.19±0.03[a]	1.61±0.02[b] (26.5)	1.35±0.02[c] (38.4)
15.	Endosulfan*	17.26±0.09[a]	16.46±0.06[b] (4.6)	14.92±0.06[c] (13.6)	15.73± 0.10[a]	12.76±0.12[b] (18.9)	11.15±0.12[c] (29.1)	7.86±0.04[a]	5.06±0.08[b] (34.9)	4.18±0.11[c] (46.8)
16.	Chlordane*	7.90±0.40[a]	6.24±0.01[b] (21.0)	4.03±0.00[c] (49.0)	6.81±0.07[a]	5.95±0.10[b] (12.6)	2.56±0.00[c] (62.4)	5.86±0.07[a]	5.14±0.04[b] (11.9)	2.31± 0.03[c] (60.6)

Values represent mean ± standard deviation (SD), n=3.

Values (of specific pesticide residue) with different alphabets for each animal are significantly different (at $p<0.05$) from one another as affected by processing method

* - Organochlorine pesticide; # - Organophosphorus pesticides

Values in ( ) represent percentage reduction in pesticide levels, relative to the raw samples

HCB = Hexachlorobenzene

DDT = Dichlorodiphenyltrichloroethane

178 The result further indicated a reduction of 4.6% for endosulfan to 34.9% for DDT in the boiled muscle
179 samples relative to the raw. The frying yielded a reduction of 13.6% for endosulfan to 62.4% for lindane
180 and chlordane in the fried muscle samples compared with the raw samples. From another perspective,
181 boiling reduced the residues by 4.6-21.6%, 6.4-18.9%, and 6.5-34.9% relative to the raw cow, pork, and
182 goat muscle samples respectively, whereas frying reduced the residues by 13.6-62.4%, 22.3-62.4% and
183 21.6-60.6% relative to the raw cow, pork, and goat muscle samples respectively (Table 2).

184 All the meat (muscle) varieties show detectable levels of four OPP residues (0.73-148.10µg/kg) of which
185 chlorpyrifos was the most prevalent (Table 2). Only chlorpyrifos was significantly depleted by the cooking
186 (boiling and frying) methods. Other residues seem to be too minute or with characteristics that made them
187 not readily depleted by the cooking methods. Boiling reduced chlorpyrifos residue by 3.8 – 13.8%
188 whereas frying reduced it by 29.3 – 37.4%.

189 **3.2 Effect of Processing on the Levels of Carbamate and Triazine Pesticides in Cow, Pork**
190 **and Goat Muscle Samples from Sagamu, Ogun State**

191 The levels of carbamate and triazine pesticides in various muscle samples as affected by conventional
192 processing methods are as presented in Table 3. The table shows the presence of one carbamate and
193 five triazine pesticides in the various samples. The levels of aldicarb, simazine, atrazine, carbofuran,
194 terbuthylazine, and cyanazine are very low in all the samples and are not significantly (p>0.05) depleted
195 by the processing methods. Comparing the concentration of the various carbamate and triazine
196 pesticides in the three muscle samples, the levels of aldicarb, simazine, atrazine, carbofuran,
197 terbuthylazine, and cyanazine are not significantly (p>0.05) different from one another. The result
198 indicated no significant (p>0.05) percentage reduction in all of carbamate and triazine pesticide residues
199 after treatment with conventional processing methods.

200

Table 3. Effect of processing on the levels (ng/Kg edible portion on dry weight basis) of carbamate and triazine pesticides in commonly consumed meat (muscle) samples in Nigeria

S/N	Pesticide	Cow			Pork			Goat		
		Raw	Boiled	Fried	Raw	Boiled	Fried	Raw	Boiled	Fried
1.	Aldicarb*	0.23±0.01 [a]	0.23±0.02 [a] (0.0)	0.23±0.01 [a] (0.0)	0.25±0.02 [a]	0.25±0.02 [a] (0.0)	0.25±0.03 [a] (0.0)	0.27±0.01 [a]	0.27±0.02 [a] (0.0)	0.27±0.02 [a] (0.0)
2.	Simazine#	2.44±0.06 [a]	2.44±0.04 [a] (0.0)	2.44±0.05 [a] (0.0)	2.34±0.02 [a]	2.34±0.02 [a] (0.0)	2.34±0.04 [a] (0.0)	2.40±0.03 [a]	2.40±0.05 [a] (0.0)	2.40±0.03 [a] (0.0)
3.	Atrazine#	2.22±0.03 [a]	2.22±0.05 [a] (0.0)	2.22±0.02 [a] (0.0)	2.28±0.05 [a]	2.28±0.03 [a] (0.0)	2.28±0.06 [a] (0.0)	2.25±0.04 [a]	2.25±0.04 [a] (0.0)	2.25±0.03 (0.0)[a]
4.	Carbofuran#	1.11±0.04 [a]	1.11±0.03 [a] (0.0)	1.11±0.02 [a] (0.0)	1.19±0.03 [a]	1.19±0.05 [a] (0.0)	1.19±0.05 [a] (0.0)	1.14±0.03 [a]	1.14±0.04 [a] (0.0)	1.14±0.04 [a] (0.0)
5.	Terbuthylazine#	1.01±0.03 [a]	1.01±0.02 [a] (0.0)	1.01±0.04 [a] (0.0)	1.07±0.04 [a]	1.07±0.05 [a] (0.0)	1.07±0.03 [a] (0.0)	1.00±0.04 [a]	1.00±0.03 [a] (0.0)	1.00±0.02 [a] (0.0)
6.	Cyanazine#	3.06±0.02 [a]	3.06±0.04 [a] (0.0)	3.06±0.05 [a] (0.0)	3.14±0.04 [a]	3.14±0.03 [a] (0.0)	3.14±0.05 [a] (0.0)	3.02±0.03 [a]	3.02±0.04 [a] (0.0)	3.02±0.04 [a] (0.0)

Values represent mean ± standard deviation (SD), n=3.

Values (of specific pesticide residue) with different alphabets for each animal are significantly different (at $p<0.05$) from one another as affected by processing method

* - Carbamate pesticide, # - Triazine pesticides.

Values in ( ) represent percentage reduction in pesticide levels, relative to the raw samples

3.3 Effect of Processing on the Levels of Chlorophenoxy, Dinitroanilin and Chloroalkylthio Pesticides in Cow, Pork and Goat Muscle Samples from Sagamu, Ogun State

The levels of chlorophenoxy, ditroanilin and chloroalkylthio pesticides in various muscle samples as affected by processing methods are as presented in Table 4. The table shows the presence of eight chlorophenoxy, one dinitroanilin and one chloroalkylthio in the various samples. The levels of 2,4,6-trichlorophenol (2,4,6-TCP), 4-(2-methyl-4-chlorophenoxy) acetic acid (MCPA), mecoprop, 2, 4-dichlorophenoxy (2,4-D), 1, 2-dibromo-3-chloropropane (DBCP), dichloroprop, fenoprop, 4-(2, 4-dichlorophenoxy) butyric acid (2,4-DB), pendimethalin and captan are very low in all the samples and are not significantly ($p>0.05$) affected by the processing methods.

Comparing the concentration of the various chlorophenoxy, ditroanilin and chloroakylthio pesticide residues in the three muscle samples, their levels are not significantly ($p>0.05$) different from one another. The result also indicated no percentage reduction in all the chlorophenoxy, dinitroanilin and chloroalkylthio pesticide residues (Table 4).

Table 4. Effect of processing on the levels (ng/Kg edible portion on dry weight basis) of chlorophenoxy, dinitroanilin and chloroalkylthio pesticides in commonly consumed meat (muscle) samples in Nigeria

S/N	Pesticide	Cow			Pork			Goat		
		Raw	Boiled	Fried	Raw	Boiled	Fried	Raw	Boiled	Fried
1.	2,4,6-TCP*	0.02±0.01[a]	0.02±0.00 [a] (0.0)	0.02±0.00 [a] (0.0)	0.03±0.00 [a]	0.03±0.01 [a] (0.0)	0.03±0.00 [a] (0.0)	0.02±0.00 [a]	0.02±0.01 [a] (0.0)	0.02±0.00 [a] (0.0)
2.	MCPA*	1.14±0.04[a]	1.14±0.02 [a] (0.0)	1.14±0.02 [a] (0.0)	1.10±0.02 [a]	1.10±0.05 [a] (0.0)	1.10±0.04 [a] (0.0)	1.06±0.02 [a]	1.06±0.04 [a] (0.0)	1.06±0.03 [a] (0.0)
3.	Mecoprop*	0.73±0.03[a]	0.73±0.02 [a] (0.0)	0.73±0.02 [a] (0.0)	0.78±0.03 [a]	0.78±0.02 [a] (0.0)	0.78±0.02 [a] (0.0)	0.70±0.02 [a]	0.70±0.01 [a] (0.0)	0.70±0.03 [a] (0.0)
4.	2,4-D*	2.05±0.04[a]	2.05±0.04 [a] (0.0)	2.05±0.02 [a] (0.0)	2.10±0.03 [a]	2.10±0.04 [a] (0.0)	2.10±0.04 [a] (0.0)	2.01±0.03 [a]	2.01±0.02 [a] (0.0)	2.01±0.02 [a] (0.0)
5.	DBCP*	2.43±0.04[a]	2.43±0.07 [a] (0.0)	2.43±0.03 [a] (0.0)	2.47±0.03 [a]	2.47±0.05 [a] (0.0)	2.47±0.05 [a] (0.0)	2.40±0.03 [a]	2.40±0.04 [a] (0.0)	2.40±0.04 [a] (0.0)
6.	Dichloroprop*	2.44±0.02[a]	2.44±0.04 [a] (0.0)	2.44±0.03 [a] (0.0)	2.47±0.04 [a]	2.47±0.03 [a] (0.0)	2.47±0.04 [a] (0.0)	2.41±0.02 [a]	2.41±0.03 [a] (0.0)	2.41±0.04 [a] (0.0)
7.	Fenoprop*	1.81±0.04[a]	1.81±0.02 [a] (0.0)	1.81±0.03 [a] (0.0)	1.86±0.02 [a]	1.86±0.03 [a] (0.0)	1.86±0.03 [a] (0.0)	1.78±0.04 [a]	1.78±0.02 [a] (0.0)	1.78±0.02 [a] (0.0)
8.	2,4-DB*	2.35±0.02[a]	2.35±0.03 [a] (0.0)	2.35±0.02 [a] (0.0)	2.33±0.02 [a]	2.33±0.01 [a] (0.0)	2.33±0.02 [a] (0.0)	2.30±0.03 [a]	2.30±0.02 [a] (0.0)	2.30±0.03 [a] (0.0)
9.	Pendimethalin#	2.08±0.02[a]	2.08±0.04 [a] (0.0)	2.08±0.03 [a] (0.0)	2.05±0.02 [a]	2.05±0.01 [a] (0.0)	2.05±0.02 [a] (0.0)	2.01±0.02 [a]	2.01±0.03 [a] (0.0)	2.01±0.03 [a] (0.0)
10.	Captan^	1.22±0.02[a]	1.22±0.02 [a] (0.0)	1.22±0.03 [a] (0.0)	1.20±0.02 [a]	1.20±0.01 [a] (0.0)	1.20±0.02 [a] (0.0)	1.12±0.01 [a]	1.12±0.02 [a] (0.0)	1.12±0.01 [a] (0.0)

Values represent mean ± standard deviation (SD), n=3.

Values (of specific pesticide residue) with different alphabets for each animal are significantly different (at $p<0.05$) from one another as affected by processing method

*- Chlorophenoxy pesticide, # - Dinitroanilin pesticide and ^ - Chloroalkylthio

Values in ( ) represent percentage reduction in pesticide levels, relative to the raw samples

2, 4, 6-TCP = 2, 4, 6-trichlorophenol

MCPA = 4-(2-methyl-4-chlorophenoxy) acetic acid

2, 4-D = 2, 4-dichlorophenoxy

DBCP = 1, 2-dibromo-3-chloropropane

2, 4-DB = 4-(2, 4-dichlorophenoxy) butyric acid

3.4 Effect of Processing on the Levels of Benzoylurea and Phenylurea in Cow, Pork and Goat Muscle Samples from Sagamu, Ogun State

The levels of benzoylurea and phenylurea pesticide residues in the various muscle samples as affected by processing methods are presented in Table 5. The table shows one phenylurea and two benzoylurea pesticides in the various samples. The levels of isoproturon, chlorotoluron and fenprothrin are very low in all the samples and are not significantly (p>0.05) affected by processing methods. Comparing the concentration of the benzoylurea and phenylurea pesticide residues in the three muscle samples, their levels are not significantly different (p>0.05) from one another. The result indicated no percentage reduction in all the benozoylurea and phenylurea pesticide residues after boiling and or frying.

Table 5. Effect of processing on the levels (ng/Kg edible portion on dry weight basis) of benzoylurea and phenylurea pesticides in commonly consumed meat (muscle) samples in Nigeria

S/N	Pesticide	Cow			Pork			Goat		
		Raw	Boiled	Fried	Raw	Boiled	Fried	Raw	Boiled	Fried
1.	Isoproturon*	2.19±0.12 [a]	2.19±0.07 [a] (0.0)	2.19±0.09 [a] (0.0)	2.29±0.15 [a]	2.29±0.21 [a] (0.0)	2.29±0.17 [a] (0.0)	2.14±0.06 [a]	2.14±0.12 [a] (0.0)	2.14±0.20 [a] (0.0)
2.	Chlorotoluron*	1.68±0.09[a]	1.68±0.13[a] (0.0)	1.68±0.16[a] (0.0)	1.65±0.11[a]	1.65±0.07[a] (0.0)	1.65±0.13[a] (0.0)	1.60±0.08[a]	1.60±0.07[a] (0.0)	1.60±0.10[a] (0.0)
3.	Fenprothrin#	1.87±0.17[a]	1.87±0.11[a] (0.0)	1.87±0.09[a] (0.0)	1.90±0.12[a]	1.90±0.21[a] (0.0)	1.90±0.14[a] (0.0)	1.81±0.09 [a]	1.81±0.08 [a] (0.0)	1.81±0.13 [a] (0.0)

Values represent mean ± standard deviation (SD), n=3.

Values (of specific pesticide residue) with different alphabets for each animal are significantly different (at $p<0.05$) from one another as affected by processing method

* - Benzoylurea pesticide, # - Phenylurea pesticides

Values in ( ) represent percentage reduction in pesticide levels, relative to the raw samples

3.5 The Levels of Pesticide Residues in Cow, Pork and Goat Muscle Samples from Sagamu, Ogun State Compared with Available International Maximum Residue Limits (MRLs)

The highest residue levels of pesticides in the various muscle samples were compared with international standards from some developed countries and organization as presented in Table 6. The highest levels of the pesticides in all samples analyzed (as found in raw samples) were relatively low and none was above the least maximum residue limit (MRL) set by the various international regulatory agents. Maximum residue limit for alachlor, endrin, cyanazine, 2,4,6-trichlorophenol (2,4,6-TCP), mecoprop, fenoprop, isoproturon, chlorotoluron and DBCP are not available.

Table 6. The levels of pesticide residues in cow, pork and goat muscle samples from Sagamu, Ogun state compared with available international Maximum Residue Limits (MRLs)

	Pesticide	Concentration (µg/Kg)			MRLs (µg/Kg)			
		Cow Raw	Pork Raw	Goat Raw	Australia[a]	FAO/WHO[b]	EU[c]	Japan[d]
1.	Mollinate	0.00148	0.00150	0.00146	50	NS	NS	NS
2.	Dimethoate	0.00125	0.00130	0.00122	50	50	NS	50
3.	Chlorothalonil	0.00271	0.00265	0.00261	20	20	NS	20
4	Alachlor	0.000760	0.000790	0.000740	NS	NS	NS	20
5.	Metalachlor	0.000980	0.000950	0.000960	50	NS	NS	NS
6.	Phosmet	0.000730	0.000750	0.000740	50-1000	1000	100	200
7.	HCB	4.51	4.30	2.80	1000	NS	200	200
8.	Lindane	3.99	1.70	1.48	2000	10	20	20
9.	Chlorpyrifos	144	148	115	500	1000	NS	50-500
10.	DDT	221	167	98.2	5000	5000	1000	1000
11.	Aldrin	2.28	5.13	4.67	50	200	200	200
12.	Heptachlor	0.91	1.80	1.55	200	NS	NS	200
13.	Endrin	3.76	2.35	2.05	NS	NS	NS	50

HCB = Hexachlorobenzene; DDT = Dichlorodiphenyltrichloroethane; NS – Not set
a – Australian Pesticides and Veterinary Medicines Authority (APVMA) [20]
b – FAO/WHO [21]
c – Commission Regulation [22]
d – The Japan Food Chemical Research Foundation [23]

Table 6. The levels of pesticide residues in cow, pork and goat muscle samples from Sagamu, Ogun state compared with available international Maximum Residue Limits (MRLs) (continued)

	Pesticide	Concentration (µg/Kg)			MRLs (µg/Kg)			
		Cow Raw	Pork Raw	Goat Raw	Australia[a]	FAO/WHO[b]	EU[c]	Japan[d]
14.	Hept.Epoxide	2.54	2.20	2.19	200	200	200	200
15.	Endosulfan	17.3	15.7	7.86	200	200	50	100
16.	Chlordane	7.90	6.81	5.86	200	50	50	80
17.	Aldicarb	0.000230	0.000250	0.000270	10	10	NS	NS
18.	Simazine	0.00244	0.00234	0.00240	50	NS	NS	20
19.	Atrazine	0.00222	0.00228	0.00225	10	NS	NS	20
20.	Carbofuran	0.00111	0.00119	0.00114	50	50	NS	50
21.	Terbuthylazine	0.00101	0.00107	0.00100	10	NS	NS	NS
22.	Cyanazine	0.00306	0.00314	0.00302	NS	NS	NS	NS
23.	2,4,6-TCP	0.0000200	0.0000300	0.0000100	NS	NS	NS	NS
24.	MCPA	0.00114	0.00110	0.00106	50	100	NS	80
25.	Mecoprop	0.000730	0.000780	0.000700	NS	NS	NS	50
26.	2,4-D	0.00205	0.00210	0.00201	200	NS	NS	200

2,4,6- TCP = 2, 4, 6-trichlorophenol; MCPA = 4-(2-methyl-4-chlorophenoxy) acetic acid; 2, 4-D = 2, 4-dichlorophenoxy; NS – Not set
a – Australian Pesticides and Veterinary Medicines Authority (APVMA) [20]
b – FAO/WHO [21]
c – Commission Regulation [22]
d – The Japan Food Chemical Research Foundation [23]

Table 6. The levels of Pesticide residues in cow, pork and goat muscle samples from Sagamu, Ogun state compared with available international Maximum Residue Limits (MRLs) (continued)

	Pesticide	Concentration (µg/Kg)			MRLs (µg/Kg)			
		Cow Raw	Pork Raw	Goat Raw	Australia[a]	FAO/WHO[b]	EU[c]	Japan[d]
27.	DBCP	0.00243	0.00247	0.00240	NS	NS	NS	NS
28.	Dichlorprop	0.00244	0.00247	0.00241	20	NS	NS	NS
29.	Fenoprop	0.00181	0.00186	0.00176	NS	NS	NS	NS
30.	2,4-DB	0.00235	0.00233	0.000230	200	NS	NS	200
31.	Pendimethalin	0.00208	0.00205	0.00201	10	NS	NS	NS

32.	Captan	0.00122	0.00120	0.00112	50	NS	NS	50
33.	Isoproturon	0.00219	0.00229	0.00214	NS	NS	NS	NS
34.	Chlorotoluron	0.00168	0.00165	0.00160	NS	NS	NS	NS
35.	Fenpropathrin	0.00187	0.00190	0.00181	NS	10	NS	100

DBCP = 1,2-dibromo-3-chloropropane; 2,4-DB = 4-(2, 4-dichlorophenoxy) butyric acid; NS= Not set

a – Australian Pesticides and Veterinary Medicines Authority (APVMA) [20]

b – FAO/WHO [21]

c – Commission Regulation [22]

d – The Japan Food Chemical Research Foundation [23]

4. DISCUSSION

It has been proven that the technological and kitchen processes can partially or fully remove or degrade organochlorine pesticide residues to other compound often less toxic, which makes products safer for human consumption [14,15,24]. The result patterns show that the frying method is a better method to minimize the OCP residues in the various meats (Table 2). This makes it necessary to reach a compromise around pesticide residues (toxicants), palatability and healthy nature of these foodstuffs for human consumption. The significant loss in OCP residues of the cow, pork and goat meats after boiling and frying might be due to the polarity of the compounds which made them leach with the fat into the broth as affected by heat treatment applied.

The percentage reduction of lindane residues (37.6% in pork and 31.1% in goat) in fried samples compared with raw samples is similar to reduction rates (17-35%) in lindane content of ovine meat after grilling, roasting and cooking as reported by Conchello *et al*. [25]. However, these figures are lower than 55% reduction reported by Sallam and Mohammed Ali Morshedy [11] for camel, cattle and sheep boiled for 1.5hrs, 60% reduction reported by Jan & Malnersic [17] in cooked beef meat after heating at 115^{o}C for 2hrs and 65% reduction reported by Mirna and Coretti [18] in rabbit meat after boiling for 1.5hrs. Nonetheless, these reports [11,17,18] are similar to the 62.4% reduction in lindane content noted in the fried cow meat. Variation in these percentage reductions seem to mainly reflect the long heating time of 90-120 minutes applied in various studies compared with 20-30 minutes cooking done in this study. The biological and physiological characteristics of the various meat samples might have also contributed.

The meat samples analyzed in this study showed 33-42% reduction of DDT in the fried samples (Table 2). This is comparable with 40% and 44% reduction in DDT residue reported by Sallam and Mohammed Ali Morshedy [11] and Bayarri *et al*. [16], respectively in various heat treated meats. The varied lengths of time allowed for processing seem not to count here. The percentage reduction of aldrin (35-51%) and endrin (32-44%) residues are somewhat higher than 33.5% and 29.2% reported by Sallam and Mohammed Ali Morshedy [11], respectively for aldrin and endrin residues.

Similar to the trend with the OCPs, frying is quite more favorable to minimize the level of chlorpyrifos (an OPP residue) in the three kinds of meat. There is paucity of data on the effect of processing on the levels of OPP residues in meat samples so very little comparison can be done.

The results on Table 3 show that the cow, pork and goat meats have detectable amount of carbamate and triazine residues in them, though at infinitesimal quantities (0.23-3.14 ng/Kg). The carbamate (aldicarb) and all the triazine residues being not significantly reduced by the cooking methods might be due to the very limited amount of the residues available in the meat samples. As far as our search covered, no data is available on the effect of processing on the levels of carbamate and triazine in meat. Thus, useful comparison is no quite feasible. Though there were detectable amount of chlorophenoxy, dinitroanilin, chloroalkylthio, benzoylurea, and phenylurea residues in the raw cow, pork and goat meats (Tables 4 and 5), the processing methods (boiling and frying) did not significantly (p>0.05) affect the levels of these residues in the various meats. The minute levels of residues might also be the main contributing factor to this outcome along with their physico-chemical characteristics.

A comparison of the detected pesticide residues with international standards [20-23] of Maximum Residue Limits (MRLs) shows that they are totally below the MRLs (Table 6). This affirms the safety of these foodstuffs to their consumers in Sagamu, Ogun state and beyond. However, it is still very necessary to regularly monitor the levels of these residues in the meats to provide safety information to consumers and ensure that good agricultural practices are maintained by meat farmers.

5. CONCLUSION

This study indicates some levels of contamination of cow, pork and goat meats by the various pesticide residues determined. The organohlorine pesticides (OCP) and organophosphorus (OPP) residues were predominantly found in the samples analyzed. Processing methods – boiling and frying – significantly (p<0.05) reduced the levels of OCPs (lindane, HCB, DDT, aldrin, heptachlor, endrin, heptachlor epoxide, endosulfan, and chlordane) and OPP (chloropyrifos) in the cow, pork and goat meats. Though not universally promoted as healthy food processing method, frying is shown to be exceptionally advantageous in reducing these residues in meats. All the meat varieties are safe for consumption, since

the levels of pesticide residues in them are completely below the MRLs set by the various international organizations.

REFERENCES

1. Dudje Y, Kamara Y, Omoigui L, Tegbaru A, Teli A, Onyibe E. Guide to safe and effective use of pesticides for crop production in Borno state, Nigeria: Introduction. IITA 2008; 23:1-15.
2. LeDoux M. Analytical methods applied to the determination of pesticide residues in foods of animal origin. A review of the past two decades. J. Chromatogr. A. 2011; 1218:1021-36.
3. Pagliuca G, Gazotti T, Zironi E, Sticca P. Determination of organophosphorus pesticide residues in wastewater samples using binary-solvent liquid-phase microextraction and solid-phase microextraction: A comparative study. J. Chromatogr. A. 2005; 1071:67-70.
4. Covaci A, Gheorghe A, Schepens P. Organochlorine pesticides (OCPs). Chemosphere. 2004; 56:757-66.
5. Driss MR, Bouguerra ML. Organochlorine residues in the Tunisian market basket. Acta Biol. Hung. 1987; 38:101-6.
6. Garcia-Regueiro JA, Diaz I, Monfort JM. Determination of organochlorine pesticides by capillary column gas chromatography (CGC) in meat and meat products from markets in Gerona (Spain). Rev. Agroquim Technol. Aliment. 1987; 27:405-17.
7. Krauthacker B, Reiner E, Simeon V, Skrinjaric-Spoljar M. Residues of organochlorine pesticides in some foodstuffs of animal origin collected in Croatia, Yugoslavia. Arh. Hig. Rada Toksikol. 1988; 39:27-31.
8. Suzuki T, Ishizaka T, Sasaki K, Saito Y, Fukuda Y. Pesticide residues in imported Australian meats. J. Food Hyg. Soc. Japan. 1989; 30:48-53.
9. Cantoni C, Fabbris F, Rogledi S. A survey of organochlorine pesticides in pork in 1988. Ind. Aliment (Pinerolo Italy). 1989; 28(268):117-18.
10. Fontcuberta M, Arques JF, Villalbi JR, Martinez M, Centrich F, Serrahima E, et al. Chlorinated organic pesticides in marketed food: Barcelona, 2001–2006. Sci. Total Environ. 2008; 389:52-57.

11. Sallam KI, Mohammed Ali Morshedy AE. Organochlorine pesticide residues in camel, cattle and sheep carcasses slaughtered in Sharkia Province, Egypt. Food Chem. 2008; 108:154-64.
12. Blankson-Arthur S, Yeboah O, Golow A, Osei Tutu A, Denutsui D. Levels of Organochlorine pesticide in residues in Grasscutter (*Thryononyms swinderianus*) tissues. Res. J. Environ. Earth Sci. 2011; 3:350-57.
13. Osibanjo O, Adeyeye A. Organochlorine Pesticide Residues in Foodstuffs of Animal Origin in Nigeria. Bull. Environ. Contam. Toxicol. 1997; 58:206-12.
14. Zabik ME, Zabik MJ, Booren AM, Nettles M, Song J-H, Welch R, et al. Pesticdes and total polychlorinated biphenyls in Chinook salmon and carp harvested from the Great Lakes: Effects of skin-off processing and selected cooking methods. J. Agric. Food Chem. 1995; 43:993-1001.
15. Abou-Arab AA. Degradation of organochlorine pesticides by meat starter in liquid media and fermented sausage. Food Chem. Toxicol. 2002; 40:33-41.
16. Bayarri S, Conchello P, Arino A, Lazaro R, Herrera A. DDT, DDT metabolites, and other organochlorines as affected by thermal processing in three commercial cuts of lamb. Bull. Environ. Contam. Toxicol. 1994; 52:554-59.
17. Jan J, Malnersic S. Effects of some food additives and fat content in meat on lindane transformation to non-polar compounds during cooking. J. Agric. Food Chem. 1982; 30:1253-56.
18. Mirna A, Coretti K. Influence of processing on the degradation of pesticides in meat products. Meat Sci. 1979; 3:97-107.
19. Sherma J, Beroza M. Analysis of pesticide residues in human and environmental samples: a compilation of methods selected for use in pesticide monitoring programs. U.S. Environmental Protection Agency, Washington, D.C., EPA/600/8-80/038 (NTIS PB82208752). 1980. Accessed 5 June 2018. Available: https://nepis.epa.gov/Exe/ZyPDF.cgi?Dockey=20007QPD.PDF.
20. Australian Pesticides and Veterinary Medicines Authority (APVMA). Agricultural and Veterinary Chemicals Code Instrument No. 4 (*MRL Standard*) Amendment Instrument 2018. Accessed 1 June 2018. Available: https://www.legislation.gov.au/Details/F2018C00434/Download.
21. FAO/WHO. Codex Alimentarius pesticide database. 2018. Accessed 1 June 2018. Available: http://www.fao.org/fao-who-codexalimentarius/codex-texts/dbs/pestres/pesticides/en/.

22. Commission Regulation (EC) No 1881/2006. Setting maximum levels for certain contaminants in foodstuffs. 2006. Accessed 3 June 2018. Available: http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2006:364:0005:0024:EN:PDF.
23. The Japan Food Chemical Research Foundation. Maximum Residue Limits (MRLs) List of Agricultural Chemicals in Foods. 2018. Retrieved June 15, 2018 from http://db.ffcr.or.jp/front/
24. Garcar J, Hrusovsky J, Smirjak M. The effect of processing on the contents of organochlorine pesticide residues in canned meat. Vet. Med. (Praha) 1987; 32:619-26.
25. Conchello MP, Herrera A, Arino A, Lazaro R, Bayarri S. Effect of several kitchen treatments on hexachlorocyclohexane residues in ovine meat. Bull. Environ. Contam. Toxicol. 1993; 51:612-18.