Impact of frying and grilling on pesticide residues in Egyptian freshwater fishes

Doha A. Salah Eldin[1], Shahat A. Ahmed[2], [1]Laila A. Mohamed[1], [1]Waleed S. Soliman[1] and Abdelgayed M. Younes[1]

[1] Department of Hydrobiology, Veterinary Division, National Research Centre, Egypt.
[2] Department of Food Hygiene and Control, Faculty of Veterinary Medicine, Cairo University, Egypt.
dr.doha61@yahoo.com

Abstract: The research amid to estimate the effect of frying and grilling on the concentration of some organochlorine, organophosphorus and pyrethroid residues in the meat of two of mostly consumed freshwater fishes in Egypt (Nile tilapia and catfish) from three locations (El-Hawamdeyya, El-Marioteya and Kafr-Elsheikh). The study revealed that the dominant organochlorine, organophosphorus and pyrethroid residues in the raw fish meat were Hexachlorobenzene (HCB), methyl parathion and cypermithrin were detected in both fish species and from the three locations. Frying caused significant losses in the concentrations of most of examined pesticides and the biggest losses was observed for heptachlor epoxide in catfish from El-Hawamdeyya and Es-fenvalerate in catfish from Kafr-Elsheikh by 100% loss while, grilling the dominant reduction was observed for methoxychlor in Nile tilapia from El-Hawamdeyya by 61%. The levels of pesticides detected in the raw fish were low and did not exceed the maximum permitted levels expect for methyl parathion, diazinon, ethyl parathion, deltamethrin for catfish samples from Kafr-Elsheikh and cypermithrin for fish samples from El-Hawamdeyya. Frying still reduced the content of pesticide residues in fish meat and improving its quality than grilling.

[Doha A. Salah Eldin, Shahat A. Ahmed, Laila A. Mohamed, Waleed S. Soliman and Abdelgayed M. Younes. **Impact of frying and grilling on pesticide residues in Egyptian freshwater fishes.** *Rep Opinion* 2019;11(6):1-12]. ISSN 1553-9873 (print); ISSN 2375-7205 (online). http://www.sciencepub.net/report. 1. doi:10.7537/marsroj110619.01.

Key words: Pesticides, Nile tilapia, Catfish, Fry, Grill, Egypt

Introduction

In Egypt the River Nile is the principle fresh water resource and life, it represents more than 97%of Egypt's water resources Ali *et al.* (2008). as a sequence of increasing industry, agriculture, Urbanization and tourism, human activities are responsible for different pollution sources for the environment and the aquatic ecosystems Bin-Dohaish *et al.* (2008).

Fish is an essential source of food and represents a main part of many natural food chains. It is highly nutritious due to its high protein content and the presence of omega-3fattyacids, fats, amino acids and vitamins; it also contains a number of minerals, including Ca, Fe, Cd, Pb, Cu, and Zn (Sofia, 2005; Aremu and Ekunode, 2008). Fish may carry many contaminants that cause many potential effects on fish itself and the organisms that consume them, including humans (Burger and Gochfeld, 2005).

Worldwide pesticide usage has increased dramatically during the past two decades, coinciding with changes in farming practices and increasingly intensive agriculture. Enviromental pollution caused by pesticides, especially the aquatic ecosystems, has become a serious problem, contamination of water by pesticides, either directly or indirectly can lead to fish kills, reduced productivity, or elevated concentrations of undesirable chemicals in edible fish tissue which can affect the health of human consuming these fish Josef *et al.* (2011).

According to FAO (2002) a pesticide is any chemical that kills controls, repel, or modifies the behavior of a pest. Pesticides can be classified according to their chemical structure to organochlorines, organophosphorus and pyrethroids.

Organochlorine pesticides (OCPs) are synthetic, non polar, toxic and can withstand in the environment for long periods. They are ubiquitous and persistent pollutants due to bioaccumulation in the food chain either as such or as their metabolites, thus causing concern on the animals at the top of the food chain. Due to their persistency and lipophilicity, OCPs from the water column can easily be accumulated by biota. Mostly, fishes are able to uptake contaminants through gills and food intake and, eventually, transfer them to humans through consumption of these organisms Zhou *et al.* (2008).

The Organophosphorus (OP) insecticides are neurotoxicants are regarded as being less persistent compared with OCs, but some reports have indicated that residues of OPs can withstand for extended periods in organic soil and surrounding drainage systems in Egypt by Abdel-Halim *et al*. (2006). Due to their cheapness and effectiveness to control pests, weeds and diseases, they had been widely used and became more and more important in agricultural production after OCs were forbidden. OP residues can concentrate and diffuse by the effect of biological enrichment and food chains; therefore, it might appear in food products and cause potential risk for human health Sun *et al.* (2011).

Pyrethroids are synthetic analogues of the natural pyrethroids, extracts of the ornamental Chry-Santemum Cinerariaefolium and its related species used to control of insects for decades, they were selective, safe and had short half lives extremely toxic to fish. Pyrethroids are heat stable and photo stable, slightly soluble in water.

Pesticides reach aquatic environment by direct application, spray drift, aerial spraying and runoff from factories, sewage and agricultural drainage which has been considered a major risk on human health, (Salah El Dien and Nasr, 2004). They cause severe toxic problems including developmental abnormalities, growth suppression, impairment of immune function and cancer promotion El Nemr *et al.* (2003).

Fish are usually cooked by different ways before consumption (boiling, roasting, baking, frying and grilling) to enhance their flavor and taste; increase shelf life. Also cooking processes are known to reduce the risk of pollutants in fish Garcia-Arias *et al* (2003). The objective of our study was to investigate the levels of pesticides from organochlorines, organophosphorus and pyrethroids in Nile tilapia and catfish from different Egyptian locations (El-Hawamdeyya, El-Mariouteya and Kafr-Elsheikh) with the effect of two cooking methods (grilling and frying) on the presence of these pesticide residues in the studied fish samples.

2. Materials and Methods

Collection of fish samples

A total of random samples of 60 Nile tilapia and 60 Cat fish samples was collected from three locations in River Nile (El-Hawamdeyya, El-Mariouteya and Kafr-Elsheikh). Samples were transferred to the laboratory in ice box under a complete aseptic condition without undue delay. Each fish species was subdivided into 3 groups (20fish samples in each group) then each group was subdivided to 4 subgroups with 5 composite fish samples in each subgroup. The soft parts of fish samples were removed and a muscle tissue sample was taken from the dorsal muscle and kept in deep freezer for the analyses of pesticide residues.

Detection of pesticides in the examined fish samples

Reagents

Extraction and cleanup

At room temperature, the muscle samples were thawed and ten gram of boneless muscles samples were processed to extract pesticides residues. In this study, QuEChERS method (Quick Easy Cheap Effective Rugged and Safe Pesticide multi-residue method) was used according to Anastassiades *et al.* (2003) and Payá *et al.* (2007) to prepare and extract samples for the pesticide residue analysis. The procedure includes initial single-phase extraction of 10 g sample with 10 ml acetonitrile, followed by liquid–liquid partitioning formed by the addition of 4 g anhydrous $MgSO_4$ plus 1 g NaCl. Removal of residual water and cleanup are completed simultaneously by using a rapid method called dispersive solid-phase extraction (dispersive-SPE), in which 150 mg anhydrous $MgSO_4$ and 25 mg primary secondary amine (PSA) sorbent are directly mixed with 1 ml acetonitrile extract. The dispersive-SPE with PSA effectively removes many polar matrix components, such as organic acids, certain polar pigments, and sugars.

Quantitative determination of organophosphorus pesticides (OPPs) and organochlorines pesticides (OCPs) and pyrethroids

Method sensitivity and recovery were determined by using samples spiked with the tested compounds. Before analysis, relevant standards were run to check column performance, peak height, resolution, and limits of detection. Peaks were identified by comparison of sample retention time values with those of the corresponding pure standard compounds. With each set of samples, to be analyzed, a solvent blank, a standard mixture and a procedural blank were run in sequence to check for contamination, peak identification and quantification. The average recovery percentages of OPPs for samples at different levels were determined and calculated for all tested compounds. Digestion, extraction and detection of pesticides concentrations in the muscle samples were carried out at the Central Agricultural Pesticides Laboratory, Agricultural Research Center, Ministry of Agriculture & Land Reclamation, Dokki, Giza, Egypt.

For cooking fish samples which having pesticide residues were taken for deboning and mincing with blender, then the fish mince was used for different cooking methods. For deep frying samples were deep fried in vegetable oil at 180°C for 15 minutes until golden brown colour appears. For grilling samples were heated at 65 °C for 10 minutes using electric grill.

Statistical analysis

The results were statistically analyzed by SPSS/20(SPSS INC., USA), using ANOVA, the presence or absence of significant difference was specified at 95% confidence interval and significance level of $P<0.05$.

3. Results

Table (1) and illustrated by figures (1) and (3) shows the mean values of different pesticides residues from organochlorines, organophosphorus and pyrethroids in Nile tilapia from the three locations. For organochlorines, methoxychlor was detected only in Nile tilapia from El-Hawamdeyya. The highest mean values of (HCB) were recorded from Kafr-Elsheikh and the lowest were from El-Hawamdeyya. For Endosulfan sulphate was detected from Kafr-Elsheikh only. The highest mean value of aldrin was detected in samples from Kafr-Elsheikh and the lowest from El-Marioteya while, it was not detected from El-Hawamdeyya, dieldrin was detected only from El-Marioteya samples. From the organophosphorus pesticide that was detected diazinon the highest mean value was recorded from Kafr-Elsheikh and the lowest was from El-Hawamdeyya.

Methyl parathion was the highest organophosphorus pesticides detected from all locations with the highest mean value from El-Marioteya and the lowest from Kafr-Elsheikh, ethyl parathion was not detected from El-Hawamdeyya and the highest mean value was detected from El-Marioteya and the lowest from Kafr-Elsheikh, Malathion was not detected from El-Marioteya but the highest mean value was recorded from Kafr-Elsheikh and the lowest from El-Hawamdeyya. Azinphos methyl was detected only from Kafr-Elsheikh samples.

From pyrethroid residues deltamethrin was only found in tilapia samples from Kafr-Elsheikh, Es-fenvalerate was another pyrethroid detected in tilapia from El-Marioteya with the highest value from El-Marioteya and the lowest was from Kafr-Elsheikh and it was not detected from El-Hawamdeyya. Cypermithrin is a synthetic widely used pyrethroid pesticide the highest average value recorded in tilapia from El-Hawamdeyya while, the lowest value was recorded from Kafr-Elsheikh.

Organochlorine, organophosphorus and pyrethroid residues in catfish samples from the three locations were presented in table (1) and illustrated by figures (2) and (3) indicated the presence of six organochlorines (methoxychlor, heptachlor epoxide, HCB, endosulfan sulphate, aldrin and dieldrin). Methoxychlor was only found in catfish samples from El-Hawamdeyya, heptachlor epoxide and HCB were recorded in all fish samples and the highest mean value were recorded from Kafr-Elsheikh and the lowest was from El-Hawamdeyya. For endosulfan sulphate was recorded in catfish samples from Kafr-Elsheikh and dieldrin from El-Marioteya only while, aldrin was not detected from El-Hawamdeyya and the highest mean value was detected from El-Marioteya and the lowest from Kafr-Elsheikh.

Six organophosphorus residues were detected in catfish samples from the three locations and methyl parathion was the highest one detected in all fish samples with the highest average value was recorded from Kafr-Elsheikh and the lowest was from El-Marioteya; ethyl parathion were also had the same pattern. For malathion which was also recorded from all catfish samples the highest mean value was from Kafr-Elsheikh and the lowest from El-Hawamdeyya. Ethion and azinphos methyl were detected in catfish samples from Kafr-Elsheikh only.

Four pyrethroid residues were detected in catfish samples from the three locations and cypermithrin was recorded in all catfish samples with the highest mean value from El-Hawamdeyya and the lowest from El-Marioteya, deltamethrin was recorded from Kafr-Elsheikh and fenopropathrin from El- Marioteya only. The highest mean value of Es-fenvalerate was recorded from El-Marioteya and the lowest from Kafr-Elsheikh.

Table (1) Mean Values of pesticide residues in Nile tilapia and catfish from different locations (ng/g)

Pesticide	**Nile tilapia**			**Catfish**			**MPL**
	El-Hawamdeyya	El-Marioteya	Kafr-Elsheikh	El-Hawamdeyya	El-Marioteya	Kafr-Elsheikh	
Methoxychlor	0.64±0.21	ND	ND	1.5±0.6	ND	ND	**-**
Heptachlor epoxide	0.24±0.18	0.35±0.1	0.4±0.2	0.05±0.04	0.6±0.15	0.7±0.25	**300 FDA (2000)**
HCB	3.2±0.95	3.4±0.9	5.2±1.05	4.4±0.8	6.1±0.5	6.3±0.8	**200 EU (2011)**
Endosulfan sulphate	ND	ND	0.3±0.2	ND	ND	0.7±0.3	**10 CREM/CBU (1998)**
Aldrin	ND	0.4±0.2	0.45±0.2	ND	0.55±0.15	0.2±0.1	**300 CAC (2009)**
Dieldrin	ND	0.45±0.2	ND	ND	0.4±0.15	ND	**300 CAC (2009)**
Diazinon	5.1±0.9	6.4±0.45	8.5±0.55	7.2±0.3	7±0.5	8.6±0.8	**2 USFDA (1991)**
Methyl parathion	10±0.85	12.3±0.6	8.5±0.3	12.75±0.55	10.25±0.9	17.45±0.5	**2 USFDA (1991)**
Ethyl parathion	ND	9.4±0.35	7.8±0.4	ND	9.7±1.2	10.2±0.13	**2 USFDA (1991)**
Malathion	2.25±0.3	ND	5.43±0.2	5±0.6	ND	6.6±0.3	**20 EU (2011)**
Ethion	ND	ND	ND	ND	ND	6.4±0.2	**10 EU (2011)**
Azinphos methyl	ND	ND	0.37±0.2	ND	ND	4.2±0.4	**-**
Deltamethrin	ND	ND	0.15±0.095	ND	ND	0.6±0.25	**0.5 FAO/WHO (2013)**
Es-Fenvalerate	ND	3.25±0.35	0.33±0.2	ND	5.3±0.9	0.2±0.1	**50 CFRUS (2006)**
Cypermithrin	2.25±0.3	0.25±0.1	0.2±0.15	2.8±0.6	0.3±0.04	0.55±0.2	**2 FAO/WHO (2013)**
Fenpropathrin	ND	1.1±0.23	ND	ND	1.4±0.7	ND	**-**

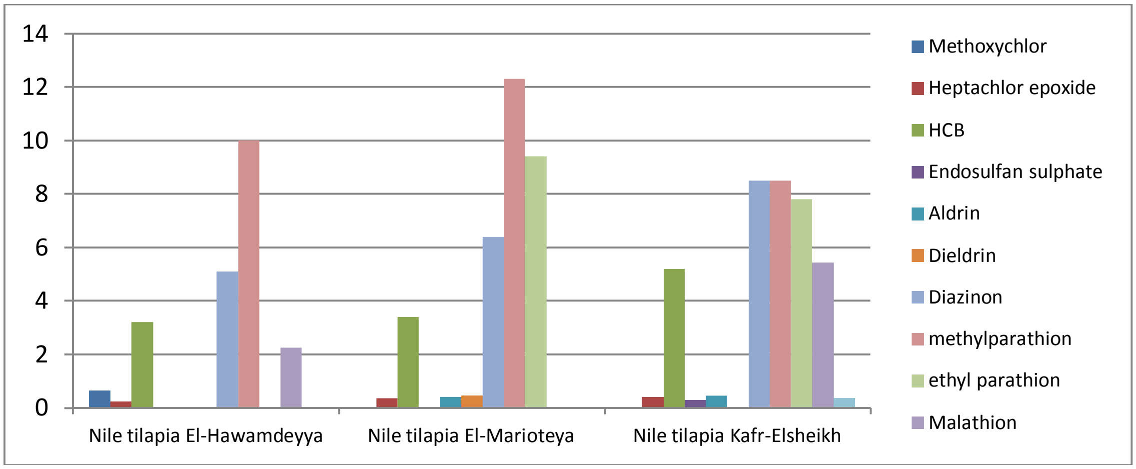


Fig (1) Mean Values of organochlorine and organophosphorus residues in Nile tilapia from different locations (ng/g).

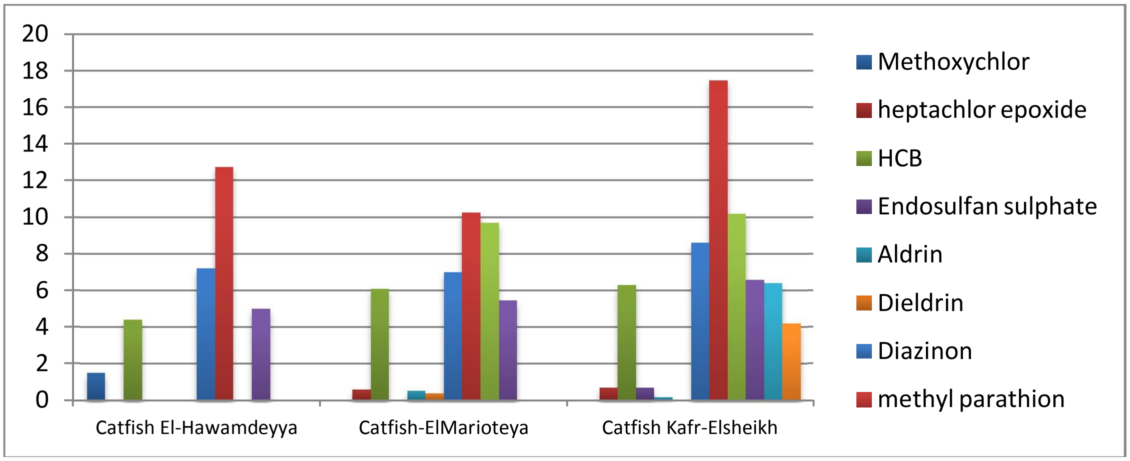


Fig (2) Mean Values of organochlorine and organophosphorus residues in catfish from different locations (ng/g).

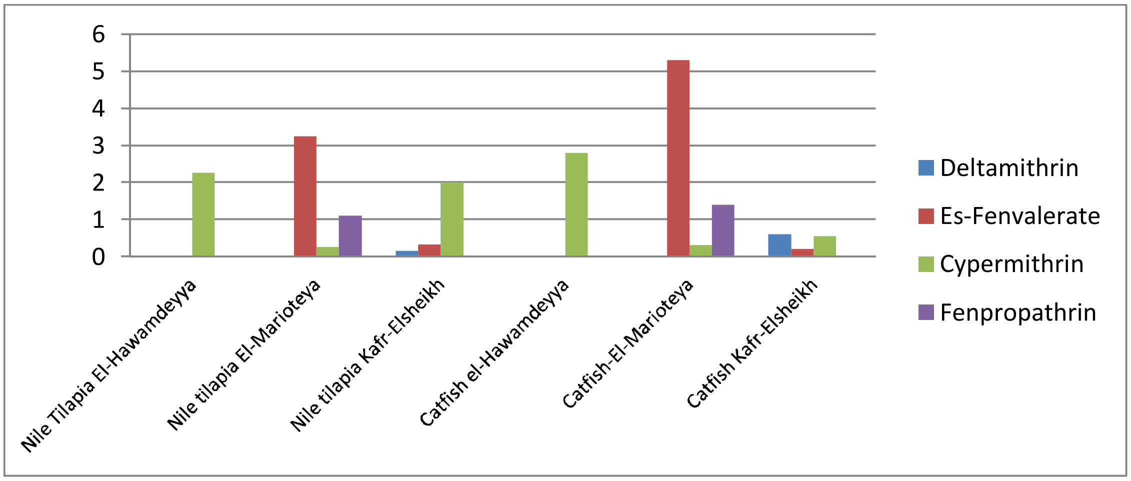
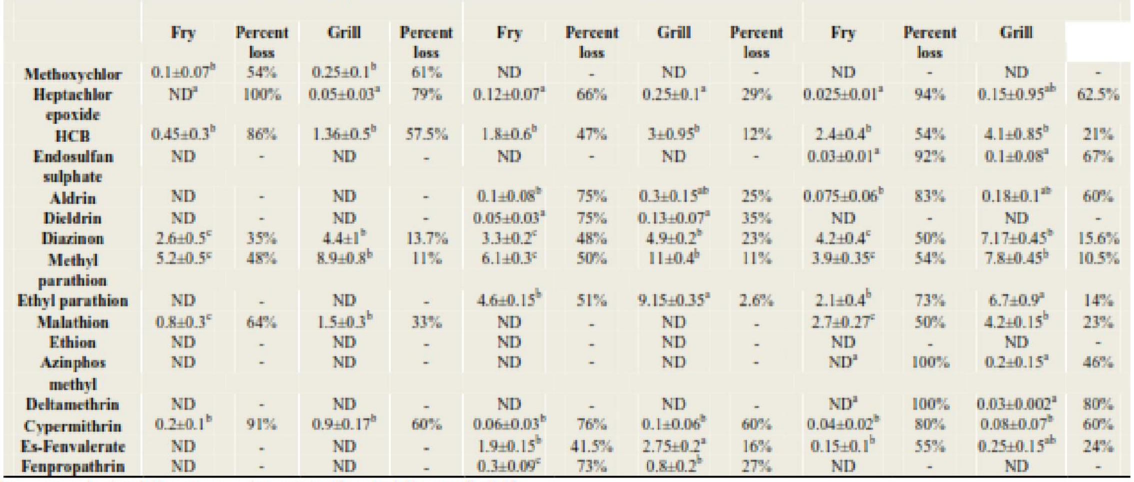


Fig (3) mean values of pyrethroids residues in Nile tilapia and Catfish from different locations

The results shown in table (2) illustrated the mean values and percent loss of the detected pesticides in Nile tilapia from the three locations after frying and grilling and the highest reduction level caused by frying of tilapia from El-Hawamdeyya was heptachlor epoxide with 100% and the lowest was 35% for diazinon while, for grilling the highest was 61% for methoxychlor and the lowest was 11% for methyl parathion. In Nile tilapia (El-Marioteya) frying caused the highest reduction level for Cypermithrin 76%and the lowest was 41.5% for Es-Fenvalerate. On the other hand the highest reduction level caused by grilling was 60% for Cypermithrin and the lowest was 11% for methyl parathion. Tilapia from Kafr-Elsheikh showed the highest reduction level by frying for cypermithrin with 80% and the lowest was for HCB and methyl parathion with 54% while, for grilling the highest was 60% for deltamithrin and the lowest was 15.6% for diazinon.

From the results recorded in table (3) showed the mean values and percent loss of pesticide residues for catfish from the three locations and the highest reduction level caused by frying for heptachlor epoxide (100%) from El-Hawamdeyya, Aldrin and Es-Fenvalerate (100%) from Kafr-Elsheikh and dieldrin (75%) from (El-Marioteya) and the lowest was recorded for malathion (47%) from El-Hawamdeyya and 36% for HCB from El-Marioteya and 44% from Kafr-Elsheikh for HCB also. While, grilling caused loss for 50% for Cypermithrin from (El-Marioteya) and the lowest was 18% for diazinon and malathion from (El-Hawamdeyya).

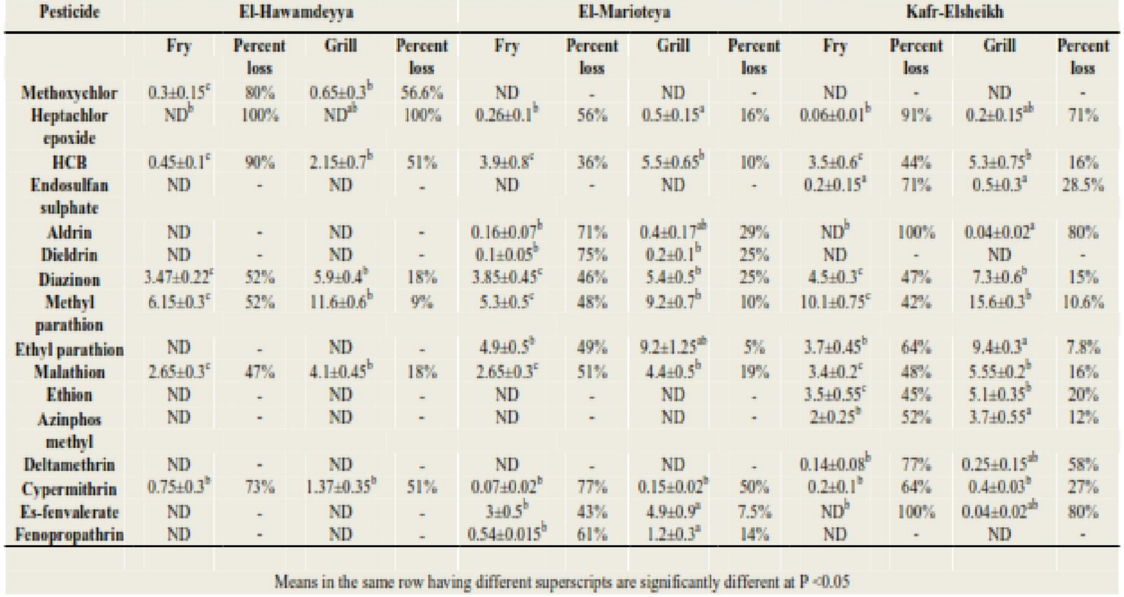
Table (2) Mean values and percent loss of detected pesticide residues in Nile tilapia from the three locations after frying and grilling (ng/g)



Pesticide	El-Hawamdeyya				El-Marioteya				Kafr-Elsheikh			
	Fry	Percent loss	Grill	Percent loss	Fry	Percent loss	Grill	Percent loss	Fry	Percent loss	Grill	
Methoxychlor	0.1 ± 0.07^{b}	54%	0.25 ± 0.1^{b}	61%	ND	-	ND	-	ND	-	ND	-
Heptachlor epoxide	ND^{a}	100%	0.05 ± 0.03^{a}	79%	0.12 ± 0.07^{a}	66%	0.25 ± 0.1^{a}	29%	0.025 ± 0.01^{a}	94%	0.15 ± 0.95^{ab}	62.5%
HCB	0.45 ± 0.3^{b}	86%	1.36 ± 0.5^{b}	57.5%	1.8 ± 0.6^{b}	47%	3 ± 0.95^{b}	12%	2.4 ± 0.4^{b}	54%	4.1 ± 0.85^{b}	21%
Endosulfan sulphate	ND	-	ND	-	ND	-	ND	-	0.03 ± 0.01^{a}	92%	0.1 ± 0.08^{a}	67%
Aldrin	ND	-	ND	-	0.1 ± 0.08^{b}	75%	0.3 ± 0.15^{ab}	25%	0.075 ± 0.06^{b}	83%	0.18 ± 0.1^{ab}	60%
Dieldrin	ND	-	ND	-	0.05 ± 0.03^{a}	75%	0.13 ± 0.07^{a}	35%	ND	-	ND	-
Diazinon	2.6 ± 0.5^{c}	35%	4.4 ± 1^{b}	13.7%	3.3 ± 0.2^{c}	48%	4.9 ± 0.2^{b}	23%	4.2 ± 0.4^{c}	50%	7.17 ± 0.45^{b}	15.6%
Methyl parathion	5.2 ± 0.5^{c}	48%	8.9 ± 0.8^{b}	11%	6.1 ± 0.3^{c}	50%	11 ± 0.4^{b}	11%	3.9 ± 0.35^{c}	54%	7.8 ± 0.45^{b}	10.5%
Ethyl parathion	ND	-	ND	-	4.6 ± 0.15^{b}	51%	9.15 ± 0.35^{a}	2.6%	2.1 ± 0.4^{b}	73%	6.7 ± 0.9^{a}	14%
Malathion	0.8 ± 0.3^{c}	64%	1.5 ± 0.3^{b}	33%	ND	-	ND	-	2.7 ± 0.27^{c}	50%	4.2 ± 0.15^{b}	23%
Ethion	ND	-	ND	-	ND	-	ND	-	ND	-	ND	-
Azinphos methyl	ND	-	ND	-	ND	-	ND	-	ND^{a}	100%	0.2 ± 0.15^{a}	46%
Deltamethrin	ND	-	ND	-	ND	-	ND	-	ND^{a}	100%	0.03 ± 0.002^{a}	80%
Cypermithrin	0.2 ± 0.1^{b}	91%	0.9 ± 0.17^{b}	60%	0.06 ± 0.03^{b}	76%	0.1 ± 0.06^{b}	60%	0.04 ± 0.02^{b}	80%	0.08 ± 0.07^{b}	60%
Es-Fenvalerate	ND	-	ND	-	1.9 ± 0.15^{b}	41.5%	2.75 ± 0.2^{a}	16%	0.15 ± 0.1^{b}	55%	0.25 ± 0.15^{ab}	24%
Fenpropathrin	ND	-	ND	-	0.3 ± 0.09^{c}	73%	0.8 ± 0.2^{b}	27%	ND	-	ND	-

Means in the same row having different superscripts are significantly different at $P < 0.05$

Table (3) Mean values and percent loss of detected pesticide residues in catfish from the three locations after frying and grilling (ng/g)



Pesticide	El-Hawamdeyya				El-Marioteya				Kafr-Elsheikh			
	Fry	Percent loss	Grill	Percent loss	Fry	Percent loss	Grill	Percent loss	Fry	Percent loss	Grill	Percent loss
Methoxychlor	0.3 ± 0.15^{c}	80%	0.65 ± 0.3^{b}	56.6%	ND	-	ND	-	ND	-	ND	-
Heptachlor epoxide	ND^{b}	100%	ND^{ab}	100%	0.26 ± 0.1^{b}	56%	0.5 ± 0.15^{a}	16%	0.06 ± 0.01^{b}	91%	0.2 ± 0.15^{ab}	71%
HCB	0.45 ± 0.1^{c}	90%	2.15 ± 0.7^{b}	51%	3.9 ± 0.8^{c}	36%	5.5 ± 0.65^{b}	10%	3.5 ± 0.6^{c}	44%	5.3 ± 0.75^{b}	16%
Endosulfan sulphate	ND	-	ND	-	ND	-	ND	-	0.2 ± 0.15^{a}	71%	0.5 ± 0.3^{a}	28.5%
Aldrin	ND	-	ND	-	0.16 ± 0.07^{b}	71%	0.4 ± 0.17^{ab}	29%	ND^{b}	100%	0.04 ± 0.02^{a}	80%
Dieldrin	ND	-	ND	-	0.1 ± 0.05^{b}	75%	0.2 ± 0.1^{b}	25%	ND	-	ND	-
Diazinon	3.47 ± 0.22^{c}	52%	5.9 ± 0.4^{b}	18%	3.85 ± 0.45^{c}	46%	5.4 ± 0.5^{b}	25%	4.5 ± 0.3^{c}	47%	7.3 ± 0.6^{b}	15%
Methyl parathion	6.15 ± 0.3^{c}	52%	11.6 ± 0.6^{b}	9%	5.3 ± 0.5^{c}	48%	9.2 ± 0.7^{b}	10%	10.1 ± 0.75^{c}	42%	15.6 ± 0.3^{b}	10.6%
Ethyl parathion	ND	-	ND	-	4.9 ± 0.5^{b}	49%	9.2 ± 1.25^{ab}	5%	3.7 ± 0.45^{b}	64%	9.4 ± 0.3^{a}	7.8%
Malathion	2.65 ± 0.3^{c}	47%	4.1 ± 0.45^{b}	18%	2.65 ± 0.3^{c}	51%	4.4 ± 0.5^{b}	19%	3.4 ± 0.2^{c}	48%	5.55 ± 0.2^{b}	16%
Ethion	ND	-	ND	-	ND	-	ND	-	3.5 ± 0.55^{c}	45%	5.1 ± 0.35^{b}	20%
Azinphos methyl	ND	-	ND	-	ND	-	ND	-	2 ± 0.25^{b}	52%	3.7 ± 0.55^{a}	12%
Deltamethrin	ND	-	ND	-	ND	-	ND	-	0.14 ± 0.08^{b}	77%	0.25 ± 0.15^{ab}	58%
Cypermithrin	0.75 ± 0.3^{b}	73%	1.37 ± 0.35^{b}	51%	0.07 ± 0.02^{b}	77%	0.15 ± 0.02^{b}	50%	0.2 ± 0.1^{b}	64%	0.4 ± 0.03^{b}	27%
Es-fenvalerate	ND	-	ND	-	3 ± 0.5^{b}	43%	4.9 ± 0.9^{a}	7.5%	ND^{b}	100%	0.04 ± 0.02^{ab}	80%
Fenopropathrin	ND	-	ND	-	0.54 ± 0.015^{b}	61%	1.2 ± 0.3^{a}	14%	ND	-	ND	-

Means in the same row having different superscripts are significantly different at $P < 0.05$

4. Disscussion

The risk of pesticides increased in the third world countries due to the lake of regulations, awareness and efficient monitoring programs dealing with the incidence of these hazardous materials in foods so that the present study was planned to evaluate the presence of organochlorines, organophosphorus and pyrethroids in fresh water fish samples (Nile tilapia and catfish) and the effect of frying and grilling on reducing these contaminants from different locations.

Organochlorines

Are a class of non polar toxic chemical compound classified as dichlorodiphenylethane, cyclodienes and chlorinated benzenes (Ademoroti, 1996).

HCB was the highest organochlorine pesticide residue detected from all fish samples from the three locations. It is still widely distributed in the environment Barber *et al.* (2005), Long term exposure to HCB in humans results in liver disease with associated skin lesions for the general population (ATSDR, 2002). In all locations catfish contained higher values than tilapia and according to (EU, 2011) which established a permissible limit for HCB that not exceed 200ng/g in fish from this point our values were far below these limits. The HCB concentrations recorded in this study was compared to those reported earlier by Yahia and Elsharkawy (2014) for tilapia species from Egypt and Adu-Kumi *et al.* (2010) for catfish and tilapia species from Ghana and the values were lower than our results.

Trials to reduce HCB in studied fish samples by frying and grilling, indicated frying had a great effect on reduction of HCB in tilapia and catfish, the highest reduction values was recorded for tilapia from El-Hawamdeyya (86%) and the lowest from El-Marioteya (47%) and for catfish the highest was recorded from El-Hawamdeyya (90%) and the lowest from El-Marioteya (36%) also. While, grilling had a moderate effect on reducing HCB with the highest reduction values detected for tilapia and catfish were from El-Hawamdeyya (57.5%) and (51%) respectively. while, the lowest reduction levels for tilapia and catfish were from El-Marioteya with (12%) and (10%) respectively. Our results were similar to Zabik *et al.* (1996) who studied the effect of grilling on HCB residues in muscle tissue of Lake trout and found that grilling had a minimal effect on reducing the pesticide content by 12-38%.

Heptachlor epoxide was anther organochorine residue was detected from all fish samples from the three studied locations which is derived as a result of the breakdown of heptachlor by bacteria that present in the environment. It has been bioconcentrated in aquatic and terrestrial organisms. It is a possible human carcinogen and impairs the immune functions. (ATSDR, 2007). Catfish was higher in heptachlor than tilapia from El-Marioteya and Kafr-Elsheikh while, the reverse from El-Hawamdeyya. Our results were below 300 ng/g set by FDA (2000) as a maximum permissible limit for heptachlor epoxide in seafood. Ogbeide *et al. (*2015) and Zhao *et al. (*2013) found heptachlor epoxide in different fish samples with nearly similar results. On the other hand Malik *et al. (*2007) detected heptachlor epoxide in *Channa punctutus* fish from Gomti River, India with mean 0.06±0.07ng/g that was lower than our results. From the previous Egyptian studies Abbassy *et al.* (2003) detected heptachlor epoxide in tilapia muscle from Lake Manzala with mean 0.059ng/g and Nasr *et al.* (2009) obtained it in tilapia from fresh water canals with mean 0.63ng/g and from drain water canals 1.78ng/g which were nearly similar to our results.

Frying showed a great effect on the reduction of heptachlor epoxide in studied catfish samples. Frying had shown a complete reduction in catfish samples from El-Hawamdeyya and the lowest reduction was recorded for catfish from El-Marioteya with a percent loss (56%). While, grilling had shown no significant decrease ($P<0.05$) for heptachlor epoxide from catfish and tilapia samples from all the studied locations. This difference may be due to variation in fish age and size that affect the fat ratio which affect the residue level after heat treatment this in agree with (Witezak, 2009) who also found a minimal effect for frying (5%) loss on heptachlor epoxide after heat treatment of Common carp with frying which was in reverse to our results.

Aldrin is a chlorinated pesticide that is used against soil-dwelling pests. It was detected in fish samples from El-Marioteya and Kafr-Elsheikh only in ranges below the maximum permissible limit set by Codex Alimentarius Commission (CAC, 2009) that aldrin in fish must not exceed 300 ng/g.

The aldrin concentration recorded in the study was compared to those reported earlier by Darko *et al.* (2008) and Zhou *et al.* (2007) found it in Nile tilapia and Red lip mullit with results agreed with our findings. On the other hand Zhao *et al.* (2013) and Malik *et al.* (2007) found aldrin in different fish species with higher values than our results. From the previous Egyptian studies Abou-Arab *et al.* (1994) detected aldrin in tilapia with mean 2.7 ng/g from Lake Manzala (Egypt), Yahia and Elsharkawy (2014) recorded aldrin in tilapia muscle from Elwasta and Mankbad, Assuit (Egypt) with mean 0.61ng ng/g and 0.22ng/g respectively, and Nasr *et al.* (2009) determined aldrin in tilapia from fresh water and drain water canals with mean 0.8 ng/g and 0.61 ng/g respectively, these findings were in agree with our study.

A complete reduction of aldrin caused by frying catfish samples from Kafr-Elsheikh while, only 71% reduction in catfish samples from El-Marioteya. In tilapia aldrin was reduced by 83%from Kafr-Elsheikh and by 75% from El-Marioteya. grilling had a no significant effect on reduction of aldrin all studied fish samples.

From previous studies Sahu (2018) detected aldrin in Catla fish from India with mean 16 ng/g which was reduced to12 ng/g (25%) by grilling and was not detected after frying this indicated the great effect of frying in depletion of aldrin from fish samples. Another study by Witezak (2009) found aldrin in Carp fish with mean 0.135±0.074 ng/g which was reduced after frying to0.119±0.046 ng/g (11.9%) which was lower than our results.

Dieldrin is a derivative from aldrin and is a highly persistent organochlorine compound that has low mobility in soil, can be lost to the atmosphere and bioaccumulate, It is considered that all the available information on aldrin and dieldrin taken together, including studies on humans, supports the view that, for practical purposes, these chemicals make very little contribution, if any, to the incidence of cancer in humans (FAO/WHO, 1993).

Dieldrin was found in fish samples from El-Marioteya only with an average below the maximum permissible limit established by Codex Alimentarius Commission (CAC, 2009) that dieldrin in fish must not exceed 300ng/g, so that our results were below this limit which considered fish from this location do not carry a hazard from dieldrin residue. The detectable levels of such residues varied in quantities dependent on the way of nutrition and the fat content of a particular species of fish, type of tissue examined and exposure of examined fish to different pesticides before caughting as well as the degree of accumulation of these compounds in examined samples as previously reported by Ademoroti *et al.* (1996).

By comparing our results and those obtained in other Egyptian locations Ahmed and El-Saad (2010) measured dieldrin in tilapia muscle from fish markets with mean 1.69 ng/g, Abou-Arab *et al.* (1995) detected dieldrin in Nile tilapia from River Nile with mean 3.85ng/g and Nasr *et al.* (2009) determined dieldrin in tilapia from fresh water and drain water canals with mean1.4 ng/g and 1.5 ng/g respectively. In another studies by Yahia and Elsharkawy (2014), Aly and Badawy (1984) and El-Mekkawi *et al.* (2009) they could not detect dieldrin in tilapia and catfish from different Egyptian water sources.

Frying caused significant reduction of dieldrin in catfish samples by 75% while, grilling caused 25% reduction in catfish samples. By comparing our findings with the other studies Sahu (2018) found dieldrin in Catla fish from India with average 870 ng/g which was reduced by grilling to 301 ng/g (65%) and to 134ng/g (84%) by frying which was slightly near to our results.

Endosulfan sulphate is a restricted organochlorine pesticide resulted from the breakdown of endosulfan in water and has the ability to accumulate in living organisms especially the aquatic one. It therefore represents a potential hazard in the aquatic environment (ATSDR, 2015). It was detected in fish samples from Kafr-Elsheikh only with average levels below the permissible limit set by CREM/CBU (1998) that not exceeds 10 ng/g in fish.

By comparing our results with the other previous studies Ogbeide *et al.* (2015) detected endosulfan sulphate in catfish from Owan River (Nigeria) with mean 0.26±0.23ng/g, Malik *et al.* (2007) recorded it in *Channa Punctutus* fish from Gomti River (India) with mean 0.22±0.24ng/g and Darko *et al.* (2008) found Endosulfan sulphate in *Nile tilapia* from Ghana with range 0.84-2.32 ng/g that were slightly near to our results, while Zhao *et al. (*2013) and Amodio-Cocchieri and Arnese (1988) did not detect endosulfan sulphate in common carp from China and Italy respectively.

Frying and grilling had no significant decrease of Endosulfan sulphate from the studied fish samples as shown in a study by Elsherif *et al.* (2016) who found endosulfan sulphate in *Nile tilapia* from Wadi El-Rayan lakes (Egypt) with mean 20 ng/g which was not affected by grilling and increased to 40 ng/ by frying due to the increase in water loss.

Methoxychlor can be found in sediments reaching the aquatic environment and its degradation may take many months. Methoxychlor is ingested and absorbed by living organisms, it accumulates in the food chain**.** It was found in fish samples from El-Hawamdeyya only with values higher than that recorded by Akan *et al.* (2013) which was 0.23±0.01ng/g for *Tilapia zilli* and 0.43±0.24ng/g for *Clarias gariepinus* obtained from Alu Dam, Nigeria.

Grilling caused a great reduction of methoxychlor in tilapia samples by 61% higher than frying which caused loss in methoxychlor residue by 54% only. While, in catfish samples frying caused higher reduction for methoxychlor (80%) than grilling (56.6%).

Organophosphorus pesticides

In an effort to substitute the persistent organochlorine pesticides, agricultural sectors have shifted towards organophosphorus pesticides. However organophosphorus pesticides are generally much more toxic to vertebrates compared to other classes of insecticides even though they rapidly degrade in the environment and have the ability to concentrate in lipid tissues of aquatic organisms Chambers *et al*. (.2001).

Methyl parathion is a nonsystemic organophosphorous insecticide and acaricide used to control pests in a wide array of crops (ATSDR 2001). It is highly toxic to aquatic organisms and has been classified as “extremely hazardous” for the environment by the WHO (2005). It was the highest pesticide detected in all fish samples from the three locations with mean values higher than the maximum permissible limit set by USFDA (1991) that it must not exceed 2ng/g in fish so that the results were higher than this limit and there is a potential great health risk from systemic toxicity to the consumers.

Methyl parathion recorded in this study was compared to those reported earlier by Yahia and Elsharkawy (2014) and Phani *et al.* (2018) from tilapia (Egypt) and Common carp (India) respectively, in levels were slightly near to our records. Enbaia *et al.* (2015) found parathion methyl in Flathead gray mullet from Tripoli markets (Libya) with a minimal level below the permissible limits about 0.0032±0.04 ng/g. while, Malhat and Nasr (2011) could not detect methyl parathion in fish tissues from many Egyptian drainage canals.

Frying had a moderate effect on reducing methyl parathion for tilapia the highest percent loss (54%) from tilapia Kafr-Elsheikh and lowest (48%) from El-Hawamdeyya. While, in catfish the highest reduction loss by frying was (52%) from El-Hawamdeyya and the lowest from Kafr-Elsheikh (42%). Grilling had a minimal effect on reducing methyl parathion with 11% loss from tilapia (El-Hawamdeyya and El-Marioteya) and 10.8% from Catfish samples (Kafr-Elsheikh). Sahu (2018) studied the effect of frying and grilling on methyl parathion from Catla fish which was reduced by 56% and 36% after frying and grilling respectively.

Diazinon is one of the most frequently used pesticides in the agricultural region worldwide. which may affect a wide range of target organisms including fish (ORUC, 2010). From our study diazinon was found in all fish samples from the studied locations with mean values exceeded the permissible limit established by USFDA (1991) that diazinon in fish does not overcome 2ng/g.

From the other previous studies Yahia and Elsharkawy (2014) detected diazinon in Nile tilapia (Egypt) with mean 6.46ng/g and 2.27 ng/g but it was not detected in catfish samples from the same locations which were lower than our results. Another study Malhat and Nasr (2011) found diazinon in fish tissues from ElMenofi drain (Egypt) and their level was 9.23 ng/g and this result was higher than our findings. However, in Ghana lagoons Essumang *et al.* (2009) obtained diazinon in fish samples in a range below to our study in a range 0.1-0.3 ng/g.

Frying and grilling had variable degrees of reduction among different species and within the same species attributed to the age, size, location and degree of pesticide contamination in the raw samples, frying for tilapia the highest reduction loss was (50%) from Kafr-Elsheikh and the lowest was (35%) from El-Hawamdeyya while, for catfish the highest percent loss was (52%) from El-Hawamdeyya and the lowest was (46%) from El-Marioteya. On the other hand grilling had a minimal effect on reducing diazinon with the highest reduction loss was recorded from catfish and tilapia (El-Marioteya) with (25%) and (23%) loss respectively, and the lowest from tilapia (El-Hawamdeyya) with (13.7%) and from catfish (Kafr-Elsheikh) with (15%).

Ethyl parathion (Parathion) is a broad spectrum organophosphate pesticide used to control many insects and mites (Worthing, 1987). It was found in fish samples from El-Marioteya and Kafr-Elsheikh with average values higher than the permissible limits established by USFDA (1991) that it does not exceed 2 ng/g in fish. By comparing the other studies Yahia and Elsharkawy (2014) detected parathion in Nile tilapia from Elwasta, Assiut (Egypt) in a concentration lower that our findings with mean 6.38±0.96 ng/g and could not detect it in catfish samples.

Frying had a good effect on reducing ethyl parathion in tilapia by (73%) and by (64%) in catfish from Kafr-Elsheikh. On the other hand grilling had no significant effect as usual on reducing ethyl parathion.

Malathion was found in all fish samples except from tilapia in El-Marioteya region and their mean values were below the permissible limit set by the EU (2011) that not exceeds 20 ng/g in fish.

From the previously mentioned studies Erewa and Ikpesu (2017), Aktar *et al.* (2009) and Yahia and Elsharkawy (2014) recorded malathion in *tilapia mariae* (Nigeria), fish samples (West Bangal), Nile tilapia and catfish from Elwasta (Egypt) respectively, with mean values lower than our results.

Loss of malathion might be caused by degradation, hydrolysis and chemical as well as physical factors such as solubility in water, voltialbilty, thermostabilty and chemical reactions as indicated by Liska *et al.* (1967) and Lee and Lee (1985). frying and grilling had a moderate and variable degrees of reduction with the highest reduction among tilapia from El-Hawamdeyya was (64%) for frying and (33%) for grilling while, among catfish the highest reduction percent was from El-Marioteya (51%) for frying and (19%) for grilling. these percents were lower than that recorded by Hassan *et al.* (1993) from Nile tilapia which was reduced after frying by 93.8% and Common carp reduced after frying by 82.9% also.

Ethion was detected only in cat fish samples from Kafr-Elsheikh in a level below the EU (2011) permissible limit that it does not exceed 10ng/g.

The mean values of ethion recorded in the study compared to those reported earlier by Yahia and Elsharkawy (2014) obtained ethion from catfish (Mankbad) in a minimal concentration and could not detect it in Nile tilapia from the same location, Zarger and Gill (2018) measured ethion in market fish sampled from different regions of Punjab in average which were lower than our findings. Another study Soltan *et al.* (2016) detected ethion in *Oreochromis niloticus* reared in earthen ponds that received agricultural drainage in Egypt with mean 5.3±3ng/g and ethion was measured by Singh *et al.* (2015) in fish muscle of *Puntius sp* from River Deomoni (West Bengal) with mean 2.95±0.4 ng/g these were slightly near to our results. Frying and grilling of catfish showed marked reduction in ethion by (45%) for frying and (20%) for grilling.

As shown in tables azinphos methyl was only recorded from Kafr-Elsheikh fish samples with average values lower than that recorded by Moradi and Goishani (2012) and Shayeghi *et al.* (2012) for mullit and Cyprinus carpio respectively.

A complete reduction in azinphos methyl was detected in tilapia samples after frying while; grilling reduced it by 46% only. Frying of catfish had a moderate effect and caused reduction by 52% on the other hand grilling showed no significant effect for its reduction in the examined fish samples.

Pyrethroids

Pyrethroids are less volatile so they are not persistent in the environment to their rapid degradation within days to several months (CDC, 2017). The natural and synthetic pyrethroids have emerged as a major class of highly active pesticides that have proved to be good substitutes for organochlorines, organophosphates due to their lower persistence and comparatively lower mammalian toxicity (Parvez and Raisuddin, 2006).

Cypermithrin is a synthetic widely used pyrethroid pesticide, it was found in all fish samples from the three locations and the only pyrethroid detected in fish samples from El-Hawamdeyya with mean values below 2 ng/g which was established as the permissible limit by FAO/WHO (2013) except fish samples from El-Marioteya that exceeded this limit.

Concerning the previous studies Riaz *et al.* (2018) detected cypermithrin in Anguilla rostrata from Qadirabad (Pakistan) also Jabeen *et al.* (2015) found cypermithrin in Channa marulius fish from Indus River (Pakistan) with and Mahboob *et al.* (2015) detected it in fish samples from River Ravi (Pakistan) in levels which were slightly near to our findings.

Frying of tilapia reduced greatly cypermithrin by 91% from El-Hawamdeyya, 81% from Kafr-Elsheikh and by 76% from El-Marioteya. On the other hand grilling of tilapia reduced cypermithrin by 60% from the three locations. Among catfish the highest reduction level was obtained from El-Marioteya was 77% caused by frying and the lowest was 51% from El-Hawamdeyya while, grilling caused a moderate effect with the highest reduction level obtained from El-Hawamdeyya by 51% and the lowest from Kafr-Elsheikh by 27% only. These results were slightly near from recorded by Sahu (2018) for Catla fish from India with mean 31 ng/g for cypermithrin and reduced by frying to 16 ng/g (48.3%) and by grilling to 21ng/g (32.25%).

Deltamethrin was recorded only in fish samples from Kafr-Elsheikh with mean values for tilapia lower than 0.5 ng/g as a permissible limit set by FAO/WHO (2013) but catfish samples exceeded this limit.

Deltamethrin recorded in this study was compared to those reported earlier by Jabeen *et al.* (2015) detected deltamethrin in Cyprinus carpio and Channa marulius from Indus River (Pakistan) with range 0.490-0.839 ng/g and 0.5-0.2ng/g respectively, which was in good agreement with our findings. While, Mahboob *et al.* (2015) measured deltamethrin in fish samples from River ravi (Pakistan) in a range ND-3.45ng/g which was higher than our results. Another study by Choudhury *et al. (*2013) found deltamethrin in Crirrhinus mrigala fish from different fish markets of Jorhat district (India) with mean 0.0013ng/g which was very low in compare with our results. Frying only significantly reduced deltamethrin by 77% from catfish samples (Kafr-Elsheikh) while, grilling had no significant changes in the examined fish samples.

Es-fenvalerate is one of the pyrethroid which is a moderately toxic pesticide and carries the signal word WARNING on the label. (Casida and Quistad, 1998). it was detected in fish samples from El-Marioteya and Kafr-Elsheikh with mean values below 50 ng/g which was set by the Code of Federal Regulations of the United States of America (2006).

Frying of tilapia had a moderate effect on reducing Es-fenvalerate by (55%) from Kafr-Elsheikh and by (41.5%) from El-Marioteya however in catfish samples frying caused a complete reduction among samples from Kafr-Elsheikh and only 43% reduction from El-Marioteya. Grilling also had no significant effect on Es-fenvalerate from the examined fish samples.

Fenopropathrin is a synthetic pyrethroid with insecticidal/acaricidal properties. It was only detected in fish samples from El-Marioteya with values higher than recorded by Choudhury *et al. (*2013) in Labeo rohita fish from different fish markets of Jorhat district (India.) Frying reduced considerably fenopropathrin by (73%) in tilapia and by (61%) in catfish on the other hand grilling reduced it by (27%) in tilapia and had no significant decrease in catfish samples.

Our results are in good agreement with Trotter *et al.* (1989) and Zabik *et al.* (1995); Witezak (2009) and Abd-Allah (2013) who showed that the reduction level in pesticide residues of processed fish tissues was depending upon the nature and the solubility of the pesticide itself, fish species, preparation treatment and the method of cooking or processing.

5. Conclusion

A general trend from the results indicates a greater increase in pesticide residue levels in Kafr-Elsheikh and El-Marioteya compared with El-Hawamdeyya in addition, higher values were realized for the detected pesticide residues in catfish as they are bottom feeders and in close contact to the sediment and their adipose tissues higher than tilapia in this study this gives clear trend of the extent of chemical contamination in these areas.

Frying is a good method for reducing many types of pesticides than grilling so it may be used to improve the quality of fish.

Finally there is a need for continued monitoring of pesticide residues in fish even when the exposure is low also monitoring is required in River Nile since the river serves as a source of drinking water, irrigation and fish for the local inhabitants in our study areas in order to ensure the safety of the population in close proximity with these contaminated river and its diverse resources and more orientation for farmers is needed to ensure the proper handling of pesticides and its application instrument. Also there is a need to intensify the enforcement of relevant laws against the importation and illegal use of banned pesticides in developing countries including Egypt.

References

1. Abdel-Halim, K.Y., Salama, A.K. and El-Khateeb, E.N. (2006): Organophosphorus pollutant (OPP) in aquatic environment at Damitta governorate, Egypt: implications for monitoring and biomarker respon.
2. Abou-Arab, A.A.K., Gomaa, M.N.E., Badawy, A. and Naguib, K. (1995): distribution of organochlorine pesticides in the Egyptian aquatic ecosystem.J.Food Chemistry.54 (2):141-146.
3. Adermoroti, C.M.A. (1996): *Enviromental chemistry and toxicology*. Floudex press LTd.Ibadan, pp: 79-208.
4. Adu-Kumi, S., Kawana, M., Shiki, Y., Yeboah, P.O., Carboo, D., Pwamang, J., Morita, M. and Suzuki, N. (2010): organochlorine pesticides (OCPs). dioxins- like polychlorinated biphenyls (DL-PCBs) polychlorinated dibenzo-P-dioxins and polychlorinated dibenzo furans (PCDD/Fs) in edible fish from lake volta, lake bosumtwii and weja lake in Ghana. J.Chemosphere.81 (6)675-684.
5. Ali, S.M., Sabae, S.Z., Fayez, M.and Hegazi, N.A. (2008): Sugar and starch industries as a potential source of water pollution of the River Nile south of Cairo: Microbiological and chemical studies. Journal of Egyptian Academic Society for Enviromental Development.9:25-45.
6. Aly O.A. and Badawy MI. (1984): Organochlorine residues in fish from the River Nile. Egypt Bull Environ Contam Toxicol.33:246–52.
7. Amodio-Cocchieri R. and Arnese A. (1988): Organochlorine pesticides residues in fish from southern Italian rivers. J. Bull. Environ. Contam. Toxicol.40:233-239.
8. Anastassiades, M., Lehotay, S.J., Štajnbaher, D. and Schenck, F.J. (2003): Fast and Easy Multiresidue Method Employing Acetonitrile Extraction/Partitioning and “Dispersive Solid-Phase Extraction” for the Determination of Pesticide Residues in Produce. Journal of AOAC International. 86(2): 412–431.
9. Aremu, M.O. and & Ekunode, O.E. (2008): Nutritional evaluation and functional properties of Clarias lazera (African catfish) from River Tammah in Nasrawa state, Nigeria. Am. J. Food. Technol.3, 264-274.
10. ATSDR "Agency for Toxic substances Portal. Heptachlor/Heptachlor epoxide. August 2007. https://www.atsdr.cdc.gov/toxprofiles/tp.asp?id=746 & tid=135
11. ATSDR "Agency for Toxic substances – toxicological profile: Endosulfan. August 2015. https://www.atsdr.cdc.gov/toxprofiles/tp.asp?id=609 & tid=113.
12. ATSDR "Agency for Toxic substances – toxicological profile: Methyl parathion. September 2001. https://www.atsdr.cdc.gov/toxprofiles/tp.asp?id=636 & tid=117
13. Barber J.L., Sweetman A.J; van Wijk D. and Jones KC. (2005): Hexachlorobenzene in the global environment: emissions, levels, distribution, trends and processes. Sci Total Environ.349:1–44.
14. Bin-Dohaish, E.A. (2008): Effects of environmental pollution with alkyl phenol (4-NONYL phenol) on reproduction of Tilapia, *Oreochromus* spillers (Teleosls). Egyptian Journal of Aquatic Research.34:336-355.
15. Burger, J. and Gochfeld, M. (2005): Heavy metals in commercial fish in New Jersey. Environ.Res.99:403-412.
16. CAC (Codex Alimentarius Commission). (2009): FAO/WHO Food STDs codex alimentarius pesticide residues in food - MRLs & Extraneus MRLs last updated March. http://www.codexalimentarius.net/mrls/pestdes/jsp/pest_q-e.jsp.
17. Casida J.E. and Quistad G.B. (1998): Golden age of insecticide research: past, present or future? Annu.Rev.Entomol. 43:1-16.
18. CDC"Centers for Disease Control and Prevention" (2017): Insecticides (Pyrethroids).

https://www.cdc.gov/nceh/dls/oatb_capacity_09.html.

19. Chambers, H.W., Boone, J.S., Carr, R.L. and Chanber, J.E. (2001): *Chemistry of organophosphorus insecticides*, In: Robert IK (ed) Handbook of pesticides toxicology.2nd ed.Acdamic Press, California pp: 913-917.
20. Code of Federal Regulations of the United States of America (2006): U.S. Government Printing Office.pp.445-446.
21. Choudhury B.H., Das B.K. and Chutia P. (2013): Evaluation of pesticide residues in fish tissue samples collected from different markets of Jorhat district of Assam, India. International Journal of Scientific and Engineering Research. (4): 2286-2299.
22. Consultancy and Research for Environmental Management/Centre of the Promotion of imports from developing on Enviromental and health issues relevant to exporters. To Eu.CREM, Rotterdam, pp: 11-42.
23. Darko G., Akoto O. and Oppong C. (2008): Persistent organochlorine pesticide residues in fish, sediments and water from Lake Bosomtwi. Ghana. J. Chemosphere.72:21–4.
24. El-Mekkawi H., Diab M., Zaki M.and Hassan A. (2009): Determination of chlorinated organic pesticide residues in water, sediments and fish from private fish farms at Abbassa and Sahl Al-Husainia, Sharkia Governorate. Aus. J. Basic Appl. Sci. 3(4): 4376-4383.
25. El Nemr, A., Said, T.O., Khaled, E.L., Sikaily, A. and Abd-Allah, A.M.A. (2003): Polychlorinated biphenyls and chlorinated pesticides in mussels collected from Egyptian Mediterranean coast. Bull. Environ. Contam. Toxicol.71:290-297.
26. Enbaia S., ElSawetta N., Abujnah Y., Greiby I., Hakam., Alzanad A., Benzitoum A., Omar A.A. and Amra H.A. (2015): Occurrence of organophosphorus pesticide residues in some fish species collected from local market in Tripoli, Libya. International Journal of Current Microbiology and Applied Sciences. 4(1): 925-937.
27. EU (European Commission Regulation). No 310/2011 of 28 March 2011 amending Annexes II and III to Regulation (EC) No 396/2005 of the European Parliament and of the Council as regards maximum residue levels for aldicarb, bromopropylate, chlorfenvinphos, endosulfan, EPTC, ethion, fenthion, fomesafen, methabenzthiazuron, methidathion, simazine, tetradifon and triforine in or on certain products. Off J EurUnion.86/1(54):1–51.
28. FAO "Food and Agriculture Organization"(2002): International Code of conduct on the distribution and use of pesticides.
29. FDA "Food and Drug Administration" (2000): Guidance for industry: Action levels for poisonous or deleterious substances in human food and animal feed.
30. Hassan I.M., Abdallah M.A., Naguib M.M. and Abou Donia M.A. (1993): Toxicity, distribution, accumulation and cooking loss of malathion in tissues of tilapia and common carp fishes. http://grasasyaceites.revistas.csic.es
31. Josef V., Alzbeta S. and Zdenka S. (2011): The effects of pyrethroid and triazine pesticides on fish physiology. Pesticides on the modern world-pests control and pesticides exposure and toxicity assessment. www.intechopen.com pp: 377-402.
32. Lee O.K.and Lee E.H. (1985): Heat stability of organochlorine pesticide residues in Loach. Bull. Nat. Fish. Univ. Pusan. 25 (1), 85-91.
33. Liska B.J., Stemp A.R. and Stadelman W.J. (1967): Effect of method of cooking on chlorinated pesticide residue content in edible chicken tissues. Food Technol. 21,435-438.
34. Mahboob S., Niazi F., AlGhanim K., Sultana S., Al-Misned F. and Ahmed Z. (2015): Health risks associated with pesticide residues in water, sediments and the muscle tissues of *Catla catla* at Head Balloki on the River Ravi. Environ. Monit. Assess.187:81-91.
35. Malhat F. and Nasr I. (2011): Organophosphorus pesticides residues in fish samples from the River Nile tributaries in Egypt. Bull Environ Contam Toxicol. 87(6):689–92.
36. Malik A., Singh K.P. and Ojha, P. (2007): Residues of Organochlorine Pesticides in Fish from the Gomti River, India. J. Bull. Environ. Contam Toxicol.78:335–340.
37. Meister R.T. ed. (1987). Farm Chemicals Handbook. Willoughby, OH: Meister publishing Co.
38. Moradi A.M. and Goishani R. (2012): Determination of concentration of organophosphate pesticides (diazinon – malathion – azinphos methyl) in mullet (Liza aurata) muscle tissue from Babolrud, Gorganrud and Tajan river estuaries. Int. J. Mar. Sci. Eng. 2(4):255-258.
39. Ogbeide O., Tongo I. and Ezemonye, L. (2015): Risk assessment of agricultural pesticides in water, sediment, and fish from Owan River, Edo State, Nigeria. Environ. Monit. Assess. 187:654-670.
40. Oruc, E. (2010): System and lipid peroxidation in the liver of Cyprinus carp. Environ. Toxicol. https://dx.doi./10.1002/Tox.571-578.
41. Parvez S. and Raisuddin S. 2006. Copper modulates non-enzymatic antioxidants in the freshwater fish, *Channa punctata* (Bloch) exposed to deltamethrin. Chemosphere, 62: 1324-1332.
42. Payá P., Anastassiades M., Mack D., Sigalova I., Tasdelen B., Oliva J. and Barba A. (2007): Analysis of pesticide residues using the Quick Easy Cheap Effective Rugged and Safe (QuEChERS) pesticide multiresidue method in combination with gas and liquid chromatography and tandem mass

spectrometric detection. Anal. Bioanal. Chem. 389: 1697–1714.

43. Phani R.S., Prasad K.R.S. and Mallu V.R. (2018): Identification of methyl parathion pesticide residues in different fish species of Mangalore (Karnataka) region. International Journal for Research in Engineering Application and Management.375-377.
44. Riaz G., Tabinda A.B., Kashif M., Yasar A., Mahmood A., Rasheed R., Khan M.I., Iqbal J., Siddique S. and Mahfooz Y. (2018): Monitoring and spatiotemporal variations of pyrethroid insecticides in surface water, sediment, and fish of the river Chenab Pakistan. Environmental Science and Pollution Research.25:22584–22597.
45. Safia S. (2005): Metal contamination in commercially important fish and shrimp species collected from aceh. (Indonesia), Penang and Perak (Malaysia) University Sciences Malaysia, Penang, Malaysia.
46. Sahu R.K. (2018): studies on pesticide and metallic residues in aquatic foods and effect of processing methods on pesticide residues. Phd. thesis. Faculty of Veterinary Science, Rajendranagar, Hyderabad-500030.
47. Salah El-Dien W.M. and Nasr I.N. (2004): Study of some organochlorine, pyrethroids and organophosphorus pesticide residues in fresh water crayfish (*procambarus clarkii*). J Egypt Vet Med Ass.64:41–51.
48. Shayeghi M., Khoobdel M., Bogheri F., Abtahi M. and Zeraati H. (2012): Organophosphorous residue in *Liza aurata* and *Cyprinus carpio*. J. Trop. Biomed. 2(7): 564–569. doi: 10.1016/S2221-1691(12)60098-7
49. Singh S., Bhutia D., Sarkar S., Rai B.K., Pal J., Bhattacharjee S. and Bahadur M. (2015): Analyses of pesticide residues in water, sediment and fish tissue from river Deomoni flowing through the tea gardens of Terai Region of West Bengal, India. International Journal of Fisheries and Aquatic Studies. 3(2): 17-23.
50. Soltan M., Hassaan M., Abaas F. and Khattaby A. (2016): Agricultural drainage water as a source of water for fish farming in Egypt. J. Ecology and Evolutionary Biology. 1(3)68-75.
51. Sun X., Zhu F., Xi J., Lu T., Liu H. and Tong Y. (2011): Hollow fiber liquid-phase microextraction as clean-up step for the determination of organophosphorus pesticides residues in fish tissue by gas chromatography coupled with mass spectrometry. Mar Poll Bull. 63:102–7.
52. Trotter W. J., Corneliussen P. E., Laski R. R. and Vanelli J. J. (1989): Levels of polychlorinated biphenyls and pesticides in blue fish before and after cooking. J. Assoc. Off. Anal. Chem. 72:501-503.
53. USFDA (Food, Drug Administration). (1991): Action levels, tolerances and guidance levels for poisonous or deleterious substances in seafood.NSSP Guidance Documents Chapter II. Growing Areas: 04 Action; 2007279–547 http://www.cfsan.fda.gov/~ear/nss4-42d.htm.
54. WHO "World Health Organization" (2004): Recommended classification of pesticides by hazard and guide line to classification. WHO, Geneva, Switzerland, Public Health Impact of Pesticides used in Agricutlure.51:86.
55. Worthing C.P. (1987): *The pesticide Manual*: A world Compendium. 8th Ed. The British Crop protection council. Croydon. England.
56. Yahia D. and Elsharkawy E.E. (2014): Multi pesticide and PCB residues in Nile tilapia and catfish in Assiut city. J. Science of the Total Environment. 466–467: 306–314.
57. Zabik M.E., Zabik M.J. Booren, A.M; Daubenmire, M.A; Welch, R. and Humphrey, H. (1995): pesticides and total polychlorinated biphenyls residues in raw and cooked walleye and white Bass harvested from the Great Lakes. Bull. Environ. *Contam. Toxicol*.54:396-402.
58. Zabik, M.E., Booren A., Zabik, M. J., Welch, R. and Humphrey, H. (1996): Pesticide residues, PCBs and PAHs in baked, charbroiled, salt boiled and smoked Great Lakes lake trout. *Food Chem*. 55, 231–237.
59. Zargar N.H. and Gill J.P.S. (2018): Studies on Levels of Pesticides Residues in Market Fish of Punjab (India). Int. J. Curr. Microbiol. App. Sci.7(8): 2899-2905. https://doi.org/10.20546/ijcmas.2018.708.307.
60. Zhao Z., Zhang L., Wu J. and Fan C. (2013): Residual levels, tissue distribution and risk assessment of organochlorine pesticides (OCPs) in edible fishes from Taihu Lake, China. J. Environ Monit Assess.185:9265–9277.
61. Zhou R., Zhou L. and Kong, Q. (2007): Persistent chlorinated pesticides in fish species from Qiantang River in East China. J. Chemosphere.68: 838-847.
62. Zhou R., Zhu L., Chen Y. and Kong Q. (2008): Concentrations and Characteristics of organochlorine pesticides in aquatic biota from Qiantang River in China. J. Environ. Pollut.151:190-199.

5/28/2019