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# Effect of location and grilling process on organochlorine pesticides residues in muscles of different fish species, Egypt

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**ABSTRACT**

This study aimed to investigate concentrations of organochlorine pesticides (OCPs) residues in muscles of different fish species; mullet, Nile tilapia, and catfish obtained from different locations (Nile River, Wadi El-Rayan Lake, Edku and Burullus lagoons) during July 2019. Also, it aimed to study the effect of the grilling process on these concentrations in Nile tilapia as abundant fish. Results showed that 15 compounds of OCPs were identified in raw fish samples and grilled tilapia. Concerning fish species, mullet from Edku, tilapia from Burullus, and catfish from Wadi El-Rayan were the lowest polluted than other ones. With regard to the location of catch, the Nile River was mostly polluted with OCPs and Wadi El-Rayan followed by Burullus and Edku lagoons. On the other side, grilling process reduced all compounds of OCPs; the loss rate in grilled tilapia was taken the following order; ∑HCH (72.59- 97.08%) >∑DDT (61.52- 94.94%) > ∑Heptachlor (58.37- 87.53%) > ∑Endosulfan (50.98-79.17%) > Endrin (32.06-83.95%) >∑Aldrin (21.15-100%). In conclusion, this study proved that concentrations of OCPs compounds in raw fish muscles were lower than the maximum permissible limits (MPLs), and grilling decreased sharply all OCPs compounds. So, the muscles of raw and grilled investigated fish samples were considered safe for human consumption.

## INTRODUCTION

Chemical pesticides are well recognized as an economic approach to control pests. Rivers and Lakes are excessively contaminated by heavy metals, organic compounds and hydrocarbon release from domestic, industrial and agricultural effluents. Drainage water from the lands treated with pesticides is drained into several major drains that finally discharged their waters into the Nile River or Lakes. Contamination of the Nile River water with various pesticides poses a hazardous risk to human health through contamination of the water and food supply, and also to aquatic organisms in general

(**Gaafar *et al.,* 2010; Gad, 2010; Dahshan *et al.,* 2016** and **Clasen *et al.,* 2018**). However, **Eissa *et al.* (2020)** confirmed that consumption of Nile tilapia from the Rosetta branch did not cause a significant potential health risk and can thus be considered to be safe for ordinary consumers. With regard to the Lakes and Lagoons, Wadi El-Rayan Lakes receive the agricultural waste water drainage from El-Wadi drain and vary in their physical and chemical characters (**Ali *et al.,* 2007**). The environmental pollution in Wadi El-Rayan Lakes induced physiological, biochemical and histopathological alterations in *Tilapia spp.* **(Mohamed and Sabae, 2015)**. However, the flesh of fish samples obtained from Wadi El-Rayan Lake has good safe and better quality for human consumption (**El-Sherif *et al.,* 2016** and **El-Sherif, 2017)**.

In general, the pesticide residues in closed Lakes mostly come from agricultural runoff and domestic discharge (**Abbassy *et al.,* 2019).** Concerning the Edku Lake pollution, **Haredi *et al.* (2020)** reported that the contaminated environment of fish in Edku Lake could cause harmful effects detrimental effects on the fish's health status, quality and marketability causing great economic losses for the natural resources; besides, it may affect the health of consumers. Also, Burullus Lake represents one of the most subjected Lakes to pollution at the delta's coastline and it serves as reservoirs for drainage waters, which are contaminated, with anthropogenic materials (**El-Zeiny and El-Kafrawy, 2017)**. However, the level of contamination of farmed tilapia, in the Nile delta, by pesticide residues and heavy metals was within MPLs (**Eltholth *et al.,* 2018**). In general, this variation of concentration and distribution of pesticides residues in fish may be attributed to the agricultural cycle, fertilizers, fish species, season, location of catch, processing methods and consumption behavior (**El-Sherif *et al.,* 2016; El-Sherif, 2017** and **Eltholth *et al.,* 2018).**

With regard to effect of heat treatment on contaminants, heat is applied for food in different ways (Boiling, baking, roasting, frying and grilling) to enhance their flavor, taste and increase shelf life **(Garcia-Arias *et al.,* 2003)** and to reduce the burden of contaminants in fish where lipophilic OCPs are associated with the fat portion of foods **(Domingo, 2011)**. **Abd-Allah (2013)** found that the loss rates of OCPs ranged from 18.40 to 53.24% in fried, 12.8 to 40.15% in grilled fish products, respectively. Also, **Hassanen *et al.* (2016)** reported that the grilling of *Oreochromus nilotica* collected from Manzala Lake reduced OCPs by ratio of 11% to 100% depending upon the type of pesticide residue.

Therefore, this study aims to study of OCPs in muscles of three raw fish species; mullet (*Mugil cephalus*), Nile tilapia (*Oreochromis niloticus*) and catfish (*Clarias gariepinus*) obtained from four different locations; Nile River and Lakes of Wadi El-Rayan, Edku and Burullus during July, 2019, and to compare the obtained levels with the international maximum residue limits (MRL). Also to study the effect of common grilling technique on their concentration in muscles of Nile tilapia samples as abundant fish.

## MATERIALS AND METHODS

### Study locations

The Nile River at Al-Qanater Al-Khairya City, Wadi El-Rayan Lake and Lagoons of Burullus and Edku were selected to obtain three similar fish species at the same time.

**Fish species**

Three fish species; grey mullet (*Mugil cephalus*), Nile tilapia (*Oreochromis niloticus*), catfish (*Clarias gariepinus*) samples were obtained from the four different locations during July, 2019. They were transferred to Fish processing technology Lab., National Institute of Oceanography and fisheries (NIOF), Alex., Egypt using ice box. All samples (first batch) were carefully washed with tap water and drained. Second batch; Nile tilapia samples obtained from the pre-mentioned locations were rolled with wheat bran and common grilled on hot plate at 170ºC for 20 min.

**Analytical methods**

Fish species samples; raw and grilled tilapia muscles were manually separated, minced, 15 organochlorine pesticides residues (OCPs) were determined (**Koc and Karakus, 2011**) using TSQ 8000 GC/MS at Central Lab., Alexandria, National Institute of Oceanography and Fisheries (NIOF). The obtained results of OCPs residues were expressed as mean values (ppb, ww.).

## RESULTS

**1. OCPs concentrations in fish species from different locations**

15 compounds of OCPs were identified by using TSQ 8000 GC/MS in raw and grilled edible parts of fish samples investigated. **Table (1)** shows the concentrations (ppb, w.w.) of OCPs in muscles of raw mullet fish (*Mugil cephalus*) samples obtained from different fishing locations during July, 2019. The values of sum DDT compounds (DDD, DDE and DDT) were 1.429, 0.632, 0.848 and 0.648 ppb; Endosulfan compounds (Endosulfan-I, Endosulfan-II and Endosulfan sulfate) were 6.969, 6.355, 2.687 and 4.929 ppb; Heptachlor compounds (Heptachlor and Heptachlor epoxide) were 0.921, 1.833, 1.063 and 0.524 ppb; Aldrin compounds (Aldrin and Dialdrin) were 0.582, 0.700, 0.431 and 0.076 ppb; HCH compounds (α, β, γ and δ- HCH) were 0.582, 1.329, 1.233 and 0.565 ppb; and Endrin were 1.077, 0.102, 0.719 and 0.528 ppb, in muscles of raw mullet samples from the Nile River, El-Rayan, Burullus and Edku, respectively.

Values of OCPs (ppb, w.w.) in muscles of raw Nile tilapia (*Oreochromis niloticus*) fish samples obtained from different fishing locations are presented in **Table 2**. Concentrations of sum DDT compounds (DDD, DDE and DDT) were 1.003, 5.766, 0.689 and 2.077ppb; Endosulfan compounds (Endosulfan-I, II and Endosulfan sulfate) recorded 6.638, 5.237, 3.826 and 3.109 ppb; Heptachlor compounds (Heptachlor and Heptachlor epoxide) were 1.235, 1.24, 0.478 and 0.802 ppb; Aldrin compounds (Aldrin and Dialdrin) were 0.406, 0.364, 0.124 and 0.563 ppb; HCH compounds (α, β, γ and δ-HCH) were 3.112, 1.729, 1.097 and 1.375ppb; and values of Endrin compound were 1.566, 0.810, 0.734 and 0.351ppb in muscles of raw Nile tilapia samples from the Nile River, El-Rayan, Burullus and Edku, respectively.

In general, tilapia muscles were highly polluted with sum Endosulfan, Heptachlor, HCH and Endrin compounds for the Nile River followed by DDT and Heptachlor for Wadi El-Rayan and Aldrin for Edku samples; however tilapia samples from Burullus were the lowest polluted than other ones.

**Table 1.** Concentrations (ppb, w.w.) of OCPs in muscles of raw mullet fish (*Mugil cephalus*) samples obtained from different fishing locations during July, 2019

| OCPs residues | Concentrations of OCPs residues (ppb, w.w.) | | | | *MPLs |
|---|---|---|---|---|---|
| | River Nile | El-Rayan Lake | Burullus Lagoon | Edku Lagoon | (ppb) |
| **4,4'-DDD** | 0.258 | 0.068 | 0.467 | 0.200 | |
| **4,4'-DDE** | 0.439 | 0.192 | 0.152 | 0.049 | |
| **4,4'-DDT** | 0.732 | 0.372 | 0.229 | 0.399 | |
| **∑ DDT** | **1.429** | **0.632** | **0.848** | **0.648** | **5000** |
| **Endosulfan-I** | 4.796 | 1.836 | 1.773 | 3.516 | |
| **Endosulfan-II** | 0.798 | 0.551 | 0.411 | 0.691 | |
| **Endosulfan-sulfate** | 1.375 | 3.968 | 0.503 | 0.722 | **300** |
| **∑ Endosulfan** | **6.969** | **6.355** | **2.687** | **4.929** | |
| **Heptachlor** | 0.269 | 0.991 | 0.493 | 0.196 | |
| **Heptachlor epoxide** | 0.652 | 0.842 | 0.570 | 0.328 | **300** |
| **∑ Heptachlor** | **0.921** | **1.833** | **1.063** | **0.524** | |
| **Aldrin** | 0.196 | 0.194 | 0.133 | 0.019 | |
| **Dieldrin** | 0.386 | 0.506 | 0.298 | 0.057 | |
| **∑ Aldrin** | **0.582** | **0.700** | **0.431** | **0.076** | **300** |
| **α-HCH** | 0.488 | 0.478 | 0.279 | 0.086 | |
| **β-HCH** | 0.012 | 0.057 | 0.008 | 0.030 | |
| **γ-HCH** | 0.056 | 0.168 | 0.698 | 0.165 | |
| **δ-HCH** | 0.026 | 0.626 | 0.248 | 0.284 | |
| **∑ HCH** | **0.582** | **1.329** | **1.233** | **0.565** | **300** |
| **Endrin** | **1.077** | **0.102** | **0.719** | **0.528** | **300** |

*MPLs: Maximum permissible limits as reported by EPA (2007), FAO/WHO (2007) and FDA (2020).

**Table (3)** exhibits the concentrations (ppb, w.w.) of OCPs residues in the muscles of raw catfish (*Clarias gariepinus*) samples obtained from different fishing locations. The values of sum DDT compounds (DDD, DDE and DDT) were 0.271, 0.039, 0.415 and 1.984 ppb, Endosulfan compounds (Endosulfan-I, II and Endosulfan sulfate) were 1.091, 1.331, 4.585 and 9.558 ppb; Heptachlor compounds (Heptachlor and Heptachlor epoxide) were 0.11, 0.228, 0.749 and 0.576 ppb; Aldrin compounds (Aldrin and Dialdrin) were 0.105, 0.166, 0.39 and 0.19 ppb; HCH compounds (α, β, γ and δ- HCH) were 0.172, 0.303, 0.485 and 0.687 ppb; and Endrin were 0.305, 0.031, 0.690 and 0.523 ppb in muscles of raw catfish samples from the Nile River, El-Rayan, Burullus and Edku, respectively.

In general, catfish samples were highly polluted with sum Heptachlor, and Aldrin and Endrin compounds for Burullus and DDT, Endsulfan and HCH for Edku followed by HCH compounds for the Nile River samples; however catfish samples from Wadi El-Rayan were the lowest polluted than other ones.

**Table 2.** Concentrations (ppb, w.w.) of OCPs in muscles of raw Nile tilapia (*Oreochromis niloticus*) samples obtained from different fishing locations during July, 2019

| OCPs residues | Concentrations of OCPs residues (ppb, w.w.) | | | | *MPLs |
|---|---|---|---|---|---|
| | River Nile | El-Rayan Lake | Burullus Lagoon | Edku Lagoon | (ppb) |
| **4,4'-DDD** | 0.314 | 0.614 | 0.326 | 1.116 | |
| **4,4'-DDE** | 0.255 | 0.497 | 0.041 | 0.103 | |
| **4,4'-DDT** | 0.434 | 4.655 | 0.322 | 0.858 | |
| **∑ DDT** | **1.003** | **5.766** | **0.689** | **2.077** | **5000** |
| **Endosulfan-I** | 3.780 | 1.992 | 1.730 | 1.236 | |
| **Endosulfan-II** | 0.640 | 1.060 | 1.203 | 0.498 | |
| **Endosulfan-sulfate** | 2.218 | 2.185 | 0.893 | 1.375 | **300** |
| **∑ Endosulfan** | **6.638** | **5.237** | **3.826** | **3.109** | |
| **Heptachlor** | 0.238 | 0.169 | 0.260 | 0.246 | |
| **Heptachlor epoxide** | 0.997 | 1.071 | 0.218 | 0.556 | **300** |
| **∑ Heptachlor** | **1.235** | **1.24** | **0.478** | **0.802** | |
| **Aldrin** | 0.165 | 0.165 | 0.075 | 0.290 | |
| **Dieldrin** | 0.241 | 0.199 | 0.049 | 0.273 | |
| **∑ Aldrin** | **0.406** | **0.364** | **0.124** | **0.563** | **300** |
| **α-HCH** | 0.677 | 0.100 | 0.172 | 0.254 | |
| **β-HCH** | 0.178 | 0.133 | 0.008 | 0.054 | |
| **γ-HCH** | 1.385 | 1.053 | 0.904 | 0.966 | |
| **δ-HCH** | 0.872 | 0.443 | 0.013 | 0.101 | |
| **∑ HCH** | **3.112** | **1.729** | **1.097** | **1.375** | **300** |
| **Endrin** | **1.566** | **0.810** | **0.734** | **0.351** | **300** |

*MPLs: Maximum permissible limits as reported by EPA (2007), FAO/WHO (2007) and FDA (2020).

## 2. Effect of grilling process on OCPs concentrations

**Table (4)** demonstrated the values (ppb, w.w.) of OCPs residues in the muscles of grilled tilapia fish samples which were pre-obtained from different fishing locations during July, 2019. It could be found that all compounds of OCs decreased sharply as affected by thermal process throughout grilling process. The values of sum DDT compounds (DDD, DDE and DDT) were 0.386, 0.926, 0.234 and 0.444 ppb, Endosulfan compounds (Endosulfan – I, II and Endosulfan sulfate) were 2.37, 2.576, 0.797 and 0.687 ppb, Heptachlor compounds (Heptachlor and Heptachlor epoxide) were 0.286, 0.508, 0.199and 0.10 ppb, Aldrin compounds (Aldrin and Dialdrin) were 0.118, 0.287, Nd and 0.063 ppb, HCH compounds (α, β, γ and δ- HCH) were 0.291, 0.474, 0.032 and 0.334 ppb and Endrin compound were 1.064, 0.130, 0.203 and 0.099 ppb in grilled tilapia samples obtained from the Nile River, El-Rayan, Burullus and Edku, respectively.

**Table 3.** Concentrations (ppb, w.w.) of OCPs in muscles of raw catfish (*Clarias gariepinus*) samples obtained from different fishing locations during July, 2019

| OCPs residues | Concentrations of OCPs residues (ppb, w.w.) | | | | *MPLs |
|---|---|---|---|---|---|
| | River Nile | El-Rayan Lake | Burullus Lagoon | Edku Lagoon | (ppb) |
| **4,4'-DDD** | 0.029 | Nd | 0.040 | 1.837 | |
| **4,4'-DDE** | 0.104 | 0.008 | 0.182 | 0.070 | |
| **4,4'-DDT** | 0.138 | 0.031 | 0.193 | 0.077 | |
| **∑ DDT** | **0.271** | **0.039** | **0.415** | **1.984** | **5000** |
| **Endosulfan-I** | 0.512 | 0.094 | 3.564 | 4.542 | |
| **Endosulfan-II** | 0.030 | 0.071 | 0.632 | 0.880 | |
| **Endosulfan-sulfate** | 0.549 | 1.166 | 0.389 | 4.136 | |
| **∑ Endosulfan** | **1.091** | **1.331** | **4.585** | **9.558** | **300** |
| **Heptachlor** | 0.086 | 0.216 | 0.068 | 0.027 | |
| **Heptachlor epoxide** | 0.024 | 0.012 | 0.681 | 0.549 | |
| **∑ Heptachlor** | **0.11** | **0.228** | **0.749** | **0.576** | **300** |
| **Aldrin** | 0.043 | 0.086 | 0.149 | 0.048 | |
| **Dieldrin** | 0.062 | 0.080 | 0.241 | 0.142 | |
| **∑ Aldrin** | **0.105** | **0.166** | **0.390** | **0.190** | **300** |
| **α-HCH** | 0.120 | 0.165 | 0.250 | 0.279 | |
| **β-HCH** | 0.001 | 0.010 | 0.117 | 0.019 | |
| **γ-HCH** | Nd | Nd | 0.076 | 0.297 | |
| **δ-HCH** | 0.051 | 0.128 | 0.042 | 0.092 | |
| **∑ HCH** | **0.172** | **0.303** | **0.485** | **0.687** | **300** |
| **Endrin** | **0.305** | **0.031** | **0.690** | **0.523** | **300** |

*MPLs: Maximum permissible limits as reported by EPA (2007), FAO/WHO (2007) and FDA (2020). Nd: not detected

**Table (5)** showed the reduction rate (%) of OCs in grilled tilapia samples. Grilling reduced in all OCs investigated in edible portion of tilapia samples. The reduction rate of sum DDT was ranged between 61.52 and 94.94% of tilapia from Nile River and Wadi El-Rayan, respectively. Sum Endosulfan was ranged between 50.98% and 79.17% for tilapia from El-Rayan and Burullus, respectively. Sum Heptachlor compounds reduced at range of 58.37% and 87.53% of tilapia from Wadi El-Rayan and Edku, respectively. Sum Aldrin compounds were 21.15% and 100%, HCH compounds were 72.59% to 97.08% of tilapia from Wadi El-Rayan and Burullus, respectively. In addition, Endrin ranged 32.06% and 83.95% of tilapia from the Nile River Wadi El-Rayan.

**Table 4.** Concentrations (ppb, w.w.) of OCPs in muscles of grilled Nile tilapia (*Oreochromis niloticus*) samples obtained from different fishing locations during July, 2019

| OCPs residues | Concentrations of OCPs residues (ppb, w.w.) | | | | *MPLs |
|---|---|---|---|---|---|
| | River Nile | El-Rayan Lake | Burullus Lagoon | Edku Lagoon | (ppb) |
| **4,4'-DDD** | 0.272 | 0.472 | 0.109 | 0.323 | |
| **4,4'-DDE** | 0.081 | 0.199 | 0.061 | Nd | |
| **4,4'-DDT** | 0.033 | 0.255 | 0.064 | 0.117 | |
| **∑ DDT** | **0.386** | **0.926** | **0.234** | **0.44** | **5000** |
| **Endosulfan-I** | 1.800 | 1.762 | 0.188 | 0.380 | |
| **Endosulfan-II** | 0.288 | 0.614 | 0.467 | 0.120 | |
| **Endosulfan-sulfate** | 0.282 | 0.200 | 0.142 | 0.187 | **300** |
| **∑ Endosulfan** | **2.37** | **2.576** | **0.797** | **0.687** | |
| **Heptachlor** | 0.015 | 0.087 | 0.198 | 0.067 | |
| **Heptachlor epoxide** | 0.271 | 0.421 | 0.001 | 0.033 | **300** |
| **∑ Heptachlor** | **0.286** | **0.508** | **0.199** | **0.10** | |
| **Aldrin** | 0.021 | 0.090 | Nd | 0.006 | |
| **Dieldrin** | 0.097 | 0.197 | Nd | 0.057 | |
| **∑ Aldrin** | **0.118** | **0.287** | Nd | **0.063** | **300** |
| **α-HCH** | 0.104 | 0.069 | 0.013 | 0.010 | |
| **β-HCH** | 0.051 | 0.036 | 0.001 | 0.025 | |
| **γ-HCH** | 0.101 | 0.356 | 0.018 | 0.296 | |
| **δ-HCH** | 0.035 | 0.013 | 0.000 | 0.003 | |
| **∑ HCH** | **0.291** | **0.474** | **0.032** | **0.334** | **300** |
| **Endrin** | **1.064** | **0.130** | **0.203** | **0.099** | **300** |

*MPLs: Maximum permissible limits as reported by EPA (2007), FAO/WHO (2007) and FDA (2020); Nd: not detected.

**Table 5.** Reduction rate (%) of OCPs in edible portion of Nile tilapia samples obtained from different fishing locations during July, 2019

| Location of catch | Reduction loss (%) in OCPs compounds of grilled tilapia; | | | | | |
|---|---|---|---|---|---|---|
| | ∑ DDTs | ∑Endosulfan | ∑Heptachlor | ∑ Aldrin | ∑ HCHs | Endrin |
| **The River Nile** | 61.52 | 64.30 | 76.84 | 70.94 | 90.65 | 32.06 |
| **El-Rayan Lake** | 94.94 | 50.98 | 59.03 | 21.15 | 72.59 | 83.95 |
| **Burullus Lagoon** | 66.04 | 79.17 | 58.37 | 100 | 97.08 | 72.34 |
| **Edku Lagoon** | 78.82 | 77.90 | 87.53 | 88.81 | 75.71 | 71.79 |

## DISCUSSION

Generally, the present study proved that, through the differences between the total concentrations of OCPs residues for different fish species and fishing locations, it is cleared that tilapia fish was highest in OCPs residue concentrations followed by mullet fish, but catfish was the lowest. Also, Edku Lagoon fish were highest in OCPs residue followed by Wadi El-Rayan and the River Nile but Burullus Lagoon fish was the lowest pollution.

These results are lower than that reported by **Dogheim *et al.* (1990**) who found that DDT was the predominant pesticide detected in fish samples. The concentration of lindane, β-HCH and chlordane were 0.59, 0.435 and 0.059 μg $kg^{-1}$. The levels of other OCPs (as average) were heptachlor (0.56 μg $kg^{-1}$), heptachlor epoxide (0.14 μg $kg^{-1}$), aldrin (0.59 μg $kg^{-1}$), dieldrin (0.61 μg $kg^{-1}$) and endrin (3.34 μg $kg^{-1}$) in fish samples collected from the River Nile (Beni-Suef and Fayom governorate);.

The obtained results are lower than that found by **Talab and Ghannam (2015)**; concentrations of p,p'-DDE, p,p'-DDD, p,p'-DDT, Heptachlor, α-HCH, β-HCH, γ-HCH and δ-HCH compounds were ranged 20-30, 33-68, 13-51, 15-27, 14-75, 28-67, 47-97 and 150-82 (ng/g w.w.) in raw fillets of Nile tilapia, blue tilapia, mango tilapia and bayad catfish, respectively, while endrin, endosulfan-I, endosulfan-II, endosulfan sulfate in the same fish species were below than our recorded results. Also, our results are in agreement with those findings by **El-Sherif *et al*. (2016)** in some detected compounds and less in another compounds for raw Nile tilapia and mullet fish fillets from Wadi El-Rayan 1st Lake where a ranges of detected components were 0.017-0.028 4,4'-DDD, UD-0.032 4,4'-DDE, 0.065-0.126 endosulfan-I, 0.188-0.244 endosulfan-II, 0.003-0.021 endosulfan-sulfate,UD-0.043 endrin, 0.021-0.052 β-HCH, 0.037- 0.067 γ HCH and 0.014-0.0.043 δ-HCH (ppm, ww). On the other hand, our results are higher than all organochlorine pesticides residues levels (<0.002-0.01μg/kg) that detected by **Mohamed and Sabae (2015)** in flesh of *Tilapia spp*. from Wadi El-Rayan Lakes.

The results obtained by **Abbassy *et al.* (2019)**; sum of DDTs was 916.53 ppb in tilapia fish from Edku Lake were very higher than our results about tilapia fish (2.077 ppb). Also, **Said *et al.,* (2008)** reported that the concentrations of DDTs in *Oreochromus niloticus* and *Clarries spp.* tissues from Brullus lagoon were ranged from 2.76 to 24.23 and from 14.16 to 45.13 ng g-1 ww, respectively and higher than our results. In addition, our results are lower than those findings by **Eissa *et al.,* (2020)** who showed that the concentrations of p,p′-DDE (< LOQ-40 μg/kg ww) in muscles of Nile tilapia (*Oreochromis niloticus*) samples were collected from 3 sampling sites (El-Rahawy, Sabal and Tala) situated along the Rosetta branch of the Nile samples. Also, **Hassan *et al.,* (2020)** found that Nile Tilapia fish had the highest concentrations of DDE, endrin, methoxychlor, DDD and endosulfan sulfate with 6.27, 25.02, 6.98, 33.35 and 47.82 ppb, respectively. While, long fin catfish had the highest concentrations of D-chlordane, α-HCH and aldrin with 82.27, 11.89 and 11.73ppb, respectively.

Also, The present study revealed that all OCPs concentrations in muscles of different investigated fish species; mullet, tilapia and catfish samples from different fishing locations; River Nile, Wadi El-Rayan Lake, Burullus and Edku Lagoons were

very lower than the maximum permissible limits (MPLs) as set in Tables (1,2,3 & 4) by Environmental Protection Agency **(EPA, 2007),** Food and Agricultural Organization/World Health Organization **(FAO/WHO, 2007)** and Food and Drug Administration **(FDA, 2020).**

Concerning the effect of grilling process on concentrations of organochlorine pesticides residues in raw muscles of Nile tilapia fish obtained from different fishing locations; the River Nile, Wadi El-Rayan Lake, Burullus and Edku Lagoons, it was found that the concentrations of all detected OCPs were sharply lost at different rates during grilling operation. The different rates of loss OCPs during grilling was depending upon the type, nature and solubility of pesticide residue itself and grilling conditions. This decrease of OCPs was due to high heat treatment used during grilling and caused the loss of OCPs with dripped fats and oils, where OCPs a high solubility in fats or oils **(Domingo, 2011)**.

These results are in accordance with that reported by **Abd-Allah (2013)** whom found that the loss rates of OCPs ranged from 18.40 - 53.24% in fried, 12.8 - 40.15% in grilled tilapia muscles from Nile River. **Hassanen *et al.* (2016)** decided that the OCPs of *Oreochromus niloticas* collected from Manzala Lake were reduced by ratio of 11% - 100% by grilling process. Similar results were reported by **Eid and Salem (2012)** who found that OCPs concentrations (α-HCH, γ-HCH and Endrin) were decreased in *Tilapia nilotica* samples after frying by 85, 79 and 80% respectively and by grilling 87, 52 and 100%, respectively, while, in *Clarias lazera* samples the levels decreased by 100, 54 and 83%, respectively by frying and 100, 50 and 100, respectively by boiling. However, **El-Sherif (2017)** reported that the highest loss of OCPs was observed for samples that cooked by boiling followed by frying and grilling, the average of total loss were 80.2 and 78.3% in boiled silver carp and catfish samples, respectively and total loss were 60.6 and 54.3% of fried samples and 39.0 and 32.1% of grilled fish samples, respectively. Also, our results are in agreement with many authors **(Trotter *et al.*, 1989; Zabik *et al.*, 1995; Witczak, 2009; El-Sherif *et al.*, 2016; Hassanen *et al.*, 2016; El-Sherif, 2017** and **Salah Eldin *et al.*, 2019).**

## CONCLUSION

This study proved that the concentrations of detected pesticides varied according to fish species and fishing locations. Total concentrations of OCPs residues appeared abundances in fish muscles and fishing locations in the following order: tilapia fish > mullet fish > catfish, about fishing locations; Edku Lagoon > Wadi El-Rayan > the Nile River > Burullus Lagoon. The concentrations of OCPs in muscles of raw mullet, tilapia and catfish samples were lower than the maximum permissible limits (MPLs). Grilling process caused a sharply decrease in all OCPs of tilapia samples. Finally, variation in concentration levels of OCs are affected by location of catch, fish species, season, feeding habitats, age, sex, lipid content and characteristics of OCPs.

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## REFERENCES

**Abbassy, M. A.; Abdel-Halim, K. Y. and El-Meseiry, M. A.** (2019). Bioaccumulation of pesticides in aquatic system of Edku Lake, Egypt: An Approach for risk associated with fish consumption. Academic Journal of Life Sciences, 5 (9): 61-69.

**Abd-Allah, Sh. S.** (2013). Studies on chemical and microbiological contaminants in some fish species and the influence of some cooking and processing methods on these contaminants. M. Sc. Thesis, Fac. of Agric. AL-Azher Univ., Cairo, Egypt.

**Ali, M. H.; Abdel-Tawab, A. A. and Soliman, A. M.** (2007). Monitoring of water quality and some pollutants of Man-Made Lake (Wadi El-Rayan First Lake, Egypt). Egypt. J. Aquat. Biol. Fish., 11(3):1235–1251.

**Clasen, B.; Loro, V. L.; Murussi, C. R.; Tiecher, T. L.; Moraes, B. and Zanella, R.** (2018). Bioaccumulation and oxidative stress caused by pesticides in Cyprinus carpio reared in a rice-fish system. Sci. Total Environ., 626:737–743, https://doi.org/10.1016/ j.scitotenv. 2018.01.154.

**Dahshan, H.; Megahed, A. M.; Abd-Elall, A. M. M.; Abd-El-Kader, M. A.; Nabawy, E. and Elbana, M. H.** (2016). Monitoring of pesticides water pollution-the Egyptian River Nile. Journal of Environmental Health Science & Engineering, 14:15, 2-9.

**Dogheim, S. M.; Nasr, E. N.; Almaz, M. M. and El-Tohamy, M. M.** (1990). Pesticide residues in milk and fish samples collected from two Egyptian governorates. J. Assoc. off. Anal. Chem., 73: 19-21.

**Domingo, J. L.** (2011). Influence of cooking processes on the concentrations of toxic metals and various organic environmental pollutants in food. Crit. Rev. Food Sci. Nutr., 51(1): 29-37.

**Eid, A. M. and Salem, Amani, M.** (2012). Effect of different traditional cooking styles on pesticides residues in fresh Tilapia nilotica and Clarias lazera fish in Gharbia markets. Alexandria Journal of Veterinary Sciences, 35 (1): 197-209.

**Eissa, F.; Ghanema, K.** and **Al-Sisi, M.** (2020). Occurrence and human health risks of pesticides and antibiotics in Nile tilapia along the Rosetta Nile branch, Egypt. Reports, https://doi.org/10.1016/j.toxrep.2020.03.004

**El-Sherif, S. A.; Ibrahim S. M. and Abd El-Gahfour S. A.** (2016). The validity of some dominant fish obtained from Wadi El-Rayan Lake for human consumption. Int. J. Adv. Res. **4**(10): 1278-1285.

**El-Sherif, S. A.** (2017). Determination of some contaminants in silver carp and catfish flesh from Wadi El-Rayan Lake and the effect of traditional cooking methods on their concentrations. Int. J. of Chem. Tech. Research, 10 (12): 143-154.

**Eltholth, M.; Fornace, K.; Graced, D.; Rushton, J. and Häslerb, B.** (2018). Assessing the chemical and microbiological quality of farmed tilapia in Egyptian fresh fish markets. Global Food Security, 17: 14–20.

**El-Zeiny, A. & El-Kafrawy, H.** (2017). Assessment of water pollution induced by human activities in Burullus Lake using Landsat 8 operational land imager and GIS. The Egyptian Journal of Remote Sensing and Space Science, 20: 49-56.

**EPA** (2007). U.S. Environmental Protection Agency (March 27, 2007), Pesticides and food: What the pesticide residue limits are on food. EPA Gov. Retrieved on September 15, 2008.

**FAO/WHO** (2007). Safety evolution of certain food additives and contaminants. International program on chemical safety. WHO Geneva.

**FDA** (2020). Food and Drug Administration, Fish and Fishery Products Hazards and Controls Guidance Fourth Edition – March 2020, 157pp.

**Gaafar, A. Y.; El-Manakhly, E. M.; Soliman, M. K.; Soufy, H.; Zaki, M. S.; Mohamed, Safinaz G. and Hassan, Sh. M.** (2010). Some pathological, biochemical and hematological investigations on Nile tilapia (*Oreochromis niloticus*) following chronic exposure to edifenphos pesticide. J. of American Sci., 6 (10):542-551.

**Gad, N. S.** (2010). Organochlorine pesticides and trace metals contamination in some marketable fish in Egypt. Egypt. J. of Aquatic Res., 36 (4): 633-642.

**Garcia-Arias, M. T.; Pontes, E. A.; Garcia-Linares, M. C.; Garcia-Fernandaz, M. C. and Sanchez-Muniz, F. J.** (2003). Cooking-Freezing Reheating (CFR) of sardine (Sardina pilchardus) fillets. Effects of different cooking and reheating procedures on the proximate and fatty-acid compositions. Food Chem., 83:349-356.

**Haredi, A. M.; Mourad, M.; Tanekhy, M.; Wassif, E. and Abdel -Tawab, H. S.** (2020). Lake Edku pollutants induced biochemical and histopathological alterations in muscle tissues of Nile Tilapia (*Oreochromis niloticus*). Toxicology and Environmental Health Sciences. *https://doi.org/10.1007/s13530-020-00042-w*.

**Hassan, M. A; Ahmed, A. M.; Marzouk, N. M. and Maky, M.A.** (2020). Detection of organochlorine pesticides residues in Nile fish and its risks in Qena City. International Journal of Veterinary Sciences, 3 (1): 51-65.

**Hassanen, F. S.; Kamel, E. A.; Gaafar, R. A. M. and Shaheen, A. A.** (2016). Effect of grilling on pesticides residues in *O. niloticus* muscles. Benha Vet. Med. J., 30 (1):348-357.

**Koc, F. & Karakus, E.** (2011). Determination of organochlorinated pesticide residues by gas chromatography-mass spectrometry after elution in a florisil column. Kafkas Univ. Vet. Fak Dreg. 17 (1):65 -70.

**Mohamed, F. S. & Sabae, S. Z.** (2015). Monitoring of pollution in Wadi El-Rayan lakes and its impact on fish. Int. J. of Develop., 4 (1): 1-28.

**Said, T. O.; El Moselhy, K. M.; Rashad, A. M. and Shreadah, M. A.** (2008). Organochlorine contaminants in water, sediment and fish of Lake Burullus, Egyptian Mediterranean Sea. Bull. Environ. Contam. Toxicol., 81:136-146.

**Salah Eldin, Doha A.; Ahmed, S. A.; Mohamed, Laila A.; Soliman, W. S. and Younes, A. M.** (2019). Impact of frying and grilling on pesticide residues in

Egyptian freshwater fishes. Report and Opinion, **11**(6). http://www.sciencepub .net/report.

**Talab, A. S. and Ghannam, Hala E.** (2015). Organochlorine pesticide residues in raw and cooked fish fillets. J. of Biodiversity and Environ. Sci., 7(3): 292-303.

**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.

**Witczak, A.** (2009). Effect of heat treatment on organochlorine pesticide residues in selected fish species. Pol. J. Food Nutr. Sci., 59 (3):231-235.

**Zabik, M. E.; Zabik, M.; Booren, A.; Nettles, M.; Song, J.; Welch, R. and Humphrey, H.** (1995). Pesticide and total polychlorinated biphenyls in Chinook salmon and carp harvested from the Great Lakes: effects of skin-on and skin-off processing and selected cooking methods. J. Agric. Food Chem., 43: 993-1001.