# Monitoring of some Organochlorines and Organophosphorus residues in imported and locally raised chicken and bovine muscles in Egypt.

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Abstract: Organochlorine and Organophosphorus Pesticides are found in many parts of the environment in small concentrations, but they have cumulative characteristics thus become a threat to human health and life. This study is focused on detection and determination of some of these pesticides in the muscles of chicken and bovine in the Egyptian market whether it was imported or raised in Egypt. Samples of locally raised chicken and bovine were collected from 5 areas: Al Giza, El Saf, El Ayat, El Hawamdeya, El Badrasheen (5 samples of chicken and 5 samples of meat from each area). Imported chicken were collected from the local market, all of the samples were imported from Brazil (25 samples), meanwhile, bovine samples were imported from: Netherlands, India, Sudan and Brazil (5 samples from each country except Brazil, 10 samples). Samples were prepared, then extraction, partitioning and clean up processes has been conducted and the aliquots were analyzed by Gas Chromatography (GC) using ECD (electron capture detector) and FPD (Flame Photometric Detector). The study showed that a lot of samples were contaminated with organochlorine pesticides while fewer samples were contaminated with organophosphorus pesticides.

Key words: Organochlorin, Organophosphorus, meat, chicken, residues.

### INTRODUCTION

Pesticides have been widely used throughout the world since the middle of the last century; however, most of them have been banned since the 1970s (many of the organochlorine compounds). Pesticides were mainly used in agriculture. Organochlorine and Organophosphorus have a high toxic effects and persistence in the environment, posing considerable hazards. The problem becomes more serious when bioaccumulation of these lipophilic compounds (such as lindane and heptachlor) is taken into consideration<sup>[1]</sup>. The necessity for continual monitoring and surveillance of these substances in natural surroundings has been recognized<sup>[2]</sup>.

Due to the general prevalence of pesticides, it is important to detect and determine the concentration levels of these pesticides residue, in environmental samples, especially in food<sup>[3]</sup>. Since these analyses do not readily degrade in the environment and are lipophilic, with a tendency to bioaccumulate, they can be found at high concentrations in fatty foods, especially chicken, meats and fish and this may result in deleterious effects in man who eats these products<sup>[4]</sup>.

A lot of researches had been conducted to detect and evaluate the concentration of many different pesticides, for example, fifteen random chicken muscle samples were analyzed in Portugal<sup>[5]</sup> for detection of Organophosphorus pesticides (chlorpyrifos, ethion, fenitrothion and methidathion). They reported that the fenitrothion and chlorpyrifos were not detected in all examined samples. On the other hand, ethion was found in four samples in concentrations of 40.55, 51.37, 31.315 and 46.575 ppb while methidathion were detected in another two samples at levels of 3.25 and 4.41 ppb. Also, they mentioned that the maximum residue limit (MRL) established by FAO/WHO for both ethion and methidathion residues was 200 ppb. Yess et al, estimated insecticide residues in poultry meat, which was used as infant food in U.S.A<sup>[6]</sup>. They reported that the concentrations of DDT, dieldrin, heptachlor and heptachlorobenzen in different examined poultry meat samples were 0.003, 0.0009, 0.0003 and 0.0003 ppm, respectively.

Garrido et al.<sup>[7]</sup> analyzed the pesticides in 30 samples, 10 from chicken, 10 from pork and 10 from lamb. Only three organochlorines, Endosulfan II alpha, Endosulfan II sulfate and dichloran were detected in three different lamb samples, while Endosulfan II alpha was detected in only one pork sample, at concentration

levels lower than the LOQ values. However, pesticide residues were not found in any of the other 10 chicken samples analyzed.

In Egypt, Mansour stated that many of the banned or severely restricted pesticides (shown in Table I) have been used in Egypt and a number of "probable human carcinogenic (group B)" and "possible human carcinogenic (group C)" pesticides were banned in an official way by Ministerial Act No. 874 for the year 1996[8]. During the season of 2001/2002, the number of pesticides registered in Egypt reached 330 formulations belonging to 175 active ingredients. Each of the insecticides and fungicides represent 34% of the total active ingredients number. Herbicides, acaricides, rodenticides, and nematicides represent 21, 6, 3, and 2%, respectively. Included in these classes of pesticides are compounds belonging to Class IA of WHO "extremely hazardous" (e.g., aldicarb, chlorophacinone, difenacoum, diphacinone), as well as Class IB "highly hazardous" (e.g., carbosulfan, fenamiphos, methomyl, oxamyl, triazophos). Moreover, pesticides such as propargite, mancozeb, maneb, folpet, procymidone, captan, and cyproconazole of "group B", as well as dimethoate, carbaryl, ethofenprox, dicofol, iprodione, benomyl, triadimefon, atrazine, oxyfluorfen, oxadiazon, linuron, simazine, and pendimethalin of "group C" may be still available for use and local formulation in Egypt.

Sallam et al, [9] determined Organochlorine pesticide in a total of 270 meat samples; comprising the muscle, liver, and kidney collected from 90 carcasses (30 each camel, cattle and sheep) slaughtered in Sharkia Province<sup>[9]</sup>. All samples were analyzed for their residual contents of DDT compounds (DDTs), hexachlorocyclohexane isomers (HCHs), lindane (c-HCH), aldrin, dieldrin, endrin, hexachlorobenzene (HCB), toxaphene, and chlordane compounds. The results indicated that 54.4% (49/90), 51.1% (46/90), 47.8% (43/90), 44.4% (40/90), 33.3% (30/90) and 15.6% (14/90) of the examined carcasses were contaminated with DDTs, HCHs, lindane, aldrin, dieldrin and endrin, respectively. The other contaminants (HCB, toxaphene, and chlordane) were only present in less than 10% of the analyzed carcasses.

Amongst the three meat animal species examined, the incidence of contamination as well as the residual concentrations of all the pesticides detected in camel carcasses were lower than those detected for cattle and sheep. The contamination level of the studied organochlorines followed the order: DDTs > HCHs > lindane > dieldrin > aldrin > endrin > toxaphene > HCB > chlordane; while the order for the contamination in the analyzed organs was liver > kidney > muscle. Heat treatment of some selected

samples (boiling for 1.5 h) produced overall reductions of 40.4%, 55.0%, 32.4%, 33.5%, 29.2%, 42.7% and 38.2% in DDTs, lindane, dieldrin, aldrin, endrin, toxaphene and HCB contents, respectively. The residual contents of the organochlorines detected in all of the contaminated samples analyzed from the three different species were well below the respective maximal permissible limits set by local or international organizations.

Therefore, the aim of this study is to analyze different types of meat samples (chicken and bovine) obtained from the local market for pesticide residues.

#### MATERIALS AND METHODS

Collection of Samples: Twenty five samples of chicken muscles and twenty five meat muscles of the locally raised animals were collected from 5 different areas of Giza governorate: Al Giza, El Saf, El Ayat, El Hawamdeya, El Badrasheen (5 chicken samples and 5 meat samples from each area).

For the imported chicken and meat, 25 samples of imported chicken were collected from different super markets and all of the samples were a Brazilian production. For the imported meat, 25 samples were collected from the super markets, and 5 samples were collected from each country of origin: Netherlands, India, and Sudan except for Brazil (10 samples).

Samples were packed in a transparent polyethylene page, stored in an ice box (contains ice during collecting the samples) and then all of the samples were stored in a deep-freezer at -20 °C until the time of analysis.

Chemicals and Reagents: All of the individual pesticide reference standards purity (>98.0%) were obtained from Dr. Ehrenstorfer, Augsburg in Germany, Organochlorines were prepared in n-Hexane, while Organophosphorus pesticides were prepared in Ethyl Acetate (HPLC grade). Organochlorine pesticides examined were:  $\alpha$ ,  $\gamma$  and  $\Delta$  HCH, Heptachlor, Aldrin, Heptachlor - epoxide, γ- Chlordan, Endosulfan II, Dieldrin, p,p DDE, Endrin, p,p DDD and p,p, DDT. Organophosphorus pesticides examined were: Ethoprophos, Cadusafos, Propetamphos, Diazinon, Chlorpyrifos-Me, Promopos-Me, Chlorpyrifos, Quinalphos, prothiophos, Ethion, Azinophos-Me. nhexane), acetone, Methylene chloride and Sodium sulphate anhydrous were purchased from Al Nasr Company for chemicals, Egypt. Ethyl acetate (HPLC grade) was purchased from SDS and acetonitril (HPLC grade) was purchased from Alliance (U.S.A), while Florisil used as a packed column (PR Grade, 60-100 mesh) was purchased from Silica (Silica Co., USA).

## **Analytical Procedures:**

Extraction: Extraction of tissue samples was conducted as described by Abd El-Kader<sup>[10]</sup>. 20 g of sample (chicken or meat) were weighed and placed in a blender with 20 g of sodium sulphate anhydrous and blended in a high speed then 120 ml of n-hexan and 120 ml of acetone were added to the blended sample, then the sample and the solvents were blended for 15 minutes. The extract was washed 3 times with distilled water in a separatory funnel. The sample moisture was dried by passing on anhydrous sodium sulphate and the solvents were evaporated at 40 C in a rotary evaporator till dryness.

Partitioning: Partitioning of the extracted muscle samples was carried out according to Leon et al.[11]. 500 ml petroleum ether were partitioned with 500 ml acetonitrile. Both two solvents were mixed in separatory funnel then separated from each other for use in sample partitioning. Sample was transferred by 25 ml petroleum ether and 10 ml acetonitrile to a 100 ml separatory funnel, then 15 ml n-hexan were added. Separatory funnel was shaken vigorously for one minute. After separation of the two solvents layers, acetonitrile was collected in a flask after passing through anhydrous sodium sulphate to remove any moisture. Another 20 ml of acetonitrile were added to petroleum ether in the separator funnel and the previously mentioned steps were repeated for two times. The petroleum ether layer was discarded and acetonitrile was evaporated by rotary evaporator.

Clean up Procedure: The clean up of extracted muscle samples was conducted according to Mills et  $al.^{[12]}$ . A 30 cm glass stoppered column was filled with 6 g of activated florisil (60 – 100 mesh) and topped with 2 g of anhydrous sodium sulphate. The sample extract was transferred to the florisil column which was already saturated with n-hexan. The column was eluated with 200 ml eluant (50% methylene chloride + 1.5% acetonitrile + 48.5% n-hexan) at the rate of 5 ml/ min. The collected eluant was concentrated on rotary evaporator at 40 C and dissolved in 2 ml of ethyl acetate for residue analysis.

Preparation of Blank Solution: The same volume of solvents (petroleum ether - acetone) and sodium sulphate anhydrous which was used in sample extraction were subjected to the same extraction, partitioning and clean up procedures as the examined samples to detect any possible traces of the studied pesticides.

## Chromatography Analysis:

Organophosphorus Pesticides: Agilent 6890 GC with

FPD detector (P-mode) was used in the study. Conditions: initial temperature was 160°C and held for 2 min., then raised 3°C/min to 240°C held for 10 min., run time was 32 min., pressure 25 PSI, flow 3.4 ml/min., column PAS-1701 (30 m x 0.32 mm (i.d.) x 0.25 μm film thickness). Detector temperature was 250°C; injector temperature was 240°C. Sample identification was confirmed on another column (CP-Ci113 CB) under a temperature program (150 - 250°C), detector temperature; 250°C, injector temperature; 240°C and flow rate of 3.4 mL/min.)

Organochlorines Pesticides: Hewlett Packard GC Model 6890 equipped with Ni63 - electron capture detector was used for analysis. GC Conditions:HP-5MS capillary column (30 m length X 0.32 mm internal diameter (i.d..), X 0.25 µm film thickness), carrier gas N2 at a flow rate of 4 ml/min., injector and detector temperatures were 300 °C and 320 °C respectively: The column temperature was initial oven temperature, 180°C for 2 min., raised at 3°C/min. and then held at 220°C for 1 min., then raised at 9°C/min. to 280°C and then held for 2 min., until a total time of 30 minutes had elapsed, DB-17 (J & scientific) capillary column (30m length X 0.32 mm internal diameter (i.d.) X 0.25 um film thickness). Operating temperature were: column temperature was programmed 160°C to 230°C at a rate of 3°C/min. and detector temperature 320°C with nitrogen carrier gas flow at 4 ml/min. was used to confirm the detected pesticides.

**Determination of Percentage Rate of Recovery:** The reliability of analytical method was examined by fortifying the tested pesticides with known quantities of tested pesticides following the same procedures of extraction, partitioning, clean up and analysis. The percent range of recovery of Organochlorine pesticides ranged from 75% to 93%, while in Organophosphorus pesticides, the percent range of recovery varied from 80% to 99%.

#### RESULTS AND DISCUSSION

The present study focus on detecting and measuring the concentration of some selected organochlorines and organophosphorus in bovine and chicken muscle samples whether was imported or was locally raised in Egypt.

As background, Mansour stated that the authorities in Egypt considered alto of pesticides as a severely restricted compounds from usage (listed in Table (1-A)) long time ago, however, most of these pesticides are still being used in Egypt. Therefore, some residues of these pesticides were detected in this study<sup>[8]</sup>.

Table 1-A: List of some banned or severely restricted pesticides:

| Pesticide           | Pesticide                      |
|---------------------|--------------------------------|
| Aldicarb            | Captafol                       |
| Aldrin              | Chlorobenzilate                |
| HCH (mixed isomers) | Ethylene dichloride            |
| Chlordane           | Pentachlorophenol              |
| Dieldrin            | Toxaphene                      |
| Heptachlor          | Binapacryl                     |
| Lindane             | Bromacil                       |
| DDT                 | Ethylene oxide                 |
| Mercury compounds   | Hexachlorobenzene              |
| Cyhexatine          | Dinoseb and dinoseb salts      |
| Endrin              | Maleic hydrazide               |
| Ethylene dibromide  | Parathiona <sup>a</sup>        |
| Fluoroacetamide     | Parathion-methyla <sup>a</sup> |
| 2,4,5-T             | Phosphamidona <sup>a</sup>     |
| Chlorodimeform      | Monocrotophosa <sup>a</sup>    |
| Paraquat            | Methamidophosaa                |

<sup>&</sup>lt;sup>a</sup> Added due to their formulations may cause problems under use conditions in DCs, not according to FAO/UNEP criteria for PIC list.

**6.1. Organochlorine Residues:** For Organochlorine pesticides, the study shows that a lot of meat samples were contaminated with Organochlorines especially that obtained from locally raised animals (Table 1).

In Table (1), all of the imported chicken samples contained residues of Organochlorines. Aldrin, Endosulfan II and Dieldrin were the most frequently found pesticides in these samples and p,p DDE was the least frequent one, but none of these concentrations exceeded the maximum residue limits given by EC and FAO/WHO (Codex alimentarius)<sup>[13]</sup>.

All of imported bovine samples contained residues of Organochlorines (Table 2). Endosulfan II, Dieldrin, Aldrin, p,p DDE and p,p, DDT were the most frequently found pesticides in these samples respectively, while the least frequent one was  $\gamma$  – HCH. In these samples, 4 samples out of 7 samples contained  $\gamma$  – Chlordan and 5 samples out of 18 samples contained Endosulfan II and exceeded the maximum residue limits.

Data in Table (3) show that all of locally raised chicken samples contained residues of Organochlorines. Endosulfan II, Dieldrin, Aldrin and p,p, DDT was the most frequently found pesticide, meanwhile Endrin and p,p DDD were the least frequent ones. There were 3 samples contained residues of Endosulfan II and had exceeded the maximum residue limits (2 samples of them were from Hwamdeya and 1 sample from Badrashen).

Results also show that all of locally raised bovine samples contained residues of Organochlorines. Aldrin, Endosulfan II, Dieldrin and p,p, DDT was the most frequently found pesticides, while Endrin was the least frequent one. These findings are similar to chicken samples. 5 samples out of 25 samples contained Endosulfan II had exceeded the maximum residue limits, 2 from Giza, 2 from Hwamdeya and one from El Ayaat. (Table 4)

**Organophosphorus Residues:** For Organophosphorus pesticides, the study show that fewer samples were contaminated with pesticides of this group compared to Organochlorines previously mentioned.

In Table (5) Propetamphos, Pirimiphos –Me and Ethion were not detected in the 25 samples of locally raised chicken, while Ethoprophos, Chlorpyrifos-Me, Chlorpyrifos and Quinalphos were the most frequently found pesticides.

Regarding Diazinon, 2 samples (obtained from El Ayaat) had exceeded the maximum residue limits for this pesticide. Also for Ethoprophos only one sample (obtained from El Ayaat) had exceeded the maximum residue limits. For Chlorpyrifos only one sample (obtained from El Ayaat) had exceeded the maximum residue limits, while only one sample contained Azinphos-Me (obtained from Giza) had exceeded the maximum residue limits.

Table Results given in (6) show Propetamphos, Diazinon and Ethion were not detected in the 25 samples of locally raised bovine. On the other hand, Ethoprophos, Cadusafos, Chlorpyrifos and Chlorpyrifos-Me were the most frequently found pesticides. For Ethoprophos, 2 samples ( obtained from Badrashen) out of 8 contained residues of this pesticide and had exceeded the maximum residue limits. For Azinphos-Methyl, only one sample contained residues of this pesticide (obtained from Giza) and had exceeded the maximum residue limits.

Propetamphos, Ethion and Azinphos-Methyl were not detected in the 25 samples of imported bovine. Meanwhile, Ethoprophos and Chlorpyrifos were the most frequently found pesticides (Table 7). For Ethoprophos, 3 samples (2 samples from India and 1 from Sudan) out of 32 samples were found to contain residues of this pesticide and had exceeded the maximum residue limits.

Finally, data in Table (8) show that only Ethoprophos, Pirimiphos –Methyl and Chlorpyrifos were only detected in some samples of imported chicken while the other pesticides were not. None of the detected residues exceeded the maximum residue limits.

These results more or less were near to the data that obtained by Rafat *et al.*<sup>[14]</sup> who found that 23 out of 115 chicken samples and 131 out of 270 meat samples analyzed for organochlorine pesticide residues were contaminated with these pesticide residues.

In the chicken samples,  $\alpha$ -HCH residues were found in 11 samples and the concentration ranged from 0.004 to 0.100 ppb, while  $\beta$ -HCH range was 0.008–0.080 ppb and it was found in 5 samples, and  $\gamma$ -HCH was detected in one sample.For Heptachlor and

Table 1: Residues of some Organochlorine pesticides on 25 samples of imported chicken (ppb)

| Pesticide           | Min.  | Max.   | Mean ± SE         | MRL* | (+ ) Inc | dence | Violation |    |  |
|---------------------|-------|--------|-------------------|------|----------|-------|-----------|----|--|
|                     |       |        |                   |      | No.      | %     | No.       | %  |  |
| α - НСН             | 0.919 | 6.398  | $1.820 \pm 0.063$ | 200  | 9        | 36    | 0         | 0% |  |
| γ - HCH             | 0.151 | 1.035  | 0.442 ± 0.013     | 20   | 8        | 32    | 0         | 0% |  |
| Δ - HCH             | 3.493 | 44.344 | 11 ± 0.460        | 200  | 14       | 56    | 0         | 0% |  |
| Heptachlor          | 0.932 | 3.493  | 1.600 ± 0.035     | 200  | 6        | 24    | 0         | 0% |  |
| Aldrin              | 0.059 | 0.662  | $0.259 \pm 0.006$ | 200  | 24       | 96    | 0         | 0% |  |
| Heptachlor- epoxide | 0.053 | 1.907  | 0.406 ± 0.0009    | 200  | 15       | 60    | 0         | 0% |  |
| γ - Chlordan        | 0.383 | 2.090  | 1.280 ± 0.019     | 50   | 15       | 60    | 0         | 0% |  |
| Endosulfan II II II | 0.760 | 6.324  | 3.460 ± 0.054     | 50   | 24       | 96    | 0         | 0% |  |
| Dieldrin            | 0.548 | 12.660 | 3.440 ± 0.104     | 200  | 24       | 96    | 0         | 0% |  |
| p,p DDE             | 0.281 | 2.727  | $1.260 \pm 0.036$ | 300  | 5        | 20    | 0         | 0% |  |
| Endrin              | 0.498 | 3.110  | 1.210 ± 0.031     | 50   | 16       | 64    | 0         | 0% |  |
| p,p DDD             | 1.296 | 18.396 | 3.020 ± 0.140     | 300  | 22       | 88    | 0         | 0% |  |
| p,p, DDT            | 0.706 | 9.405  | $6.330 \pm 0.140$ | 300  | 15       | 60    | 0         | 0% |  |

MRL\* all of MRLs were obtained from:

Table 2: Residues of some Organochlorines pesticides on 25 samples of imported bovine (ppb)

| Pesticide           | Min.  | Origin    | Max.    | Origin    | Mean ± SE           | MRL | (+ ) In | cidence | Violatio | on |
|---------------------|-------|-----------|---------|-----------|---------------------|-----|---------|---------|----------|----|
|                     |       |           |         |           |                     |     | No.     | %       | No.      | %  |
| α - НСН             | 0.919 | )Brazil)  | 8.901   | (India)   | $4.824\ \pm\ 0.129$ | 200 | 6       | 24      | 0        | 0  |
| γ - НСН             | 0.157 | (Brazil)  | 0.350   | (Brazil)  | $0.023 \pm 0.003$   | 20  | 5       | 20      | 0        | 0  |
| Δ - НСН             | 2.797 | (Holland) | 17.003  | (Sudan)   | 8.400 ± 0.218       | 200 | 10      | 40      | 0        | 0  |
| Heptachlor          | 0.830 | (Brazil)  | 3.709   | (Brazil)  | $1.870 \pm 0.045$   | 200 | 6       | 24      | 0        | 0  |
| Aldrin              | 0.061 | (India)   | 0.421   | (Brazil)  | $0.174 \pm 0.004$   | 200 | 12      | 48      | 0        | 0  |
| Heptachlor- epoxide | 0.055 | (India)   | 1.257   | (Brazil)  | $0.329 \pm 0.015$   | 200 | 8       | 32      | 0        | 0  |
| γ - Chlordan        | 0.364 | (Holland) | 340.744 | (Sudan)   | 176.900 ± 6.644     | 50  | 7       | 28      | 4        | 57 |
| Endosulfan II II    | 0.837 | (Brazil)  | 154.167 | (Indian)  | 19.600 ± 1.170      | 50  | 18      | 72      | 5        | 28 |
| Dieldrin            | 0.473 | (Brazil)  | 26.260  | (India)   | $8.540 \pm 0.360$   | 200 | 15      | 60      | 0        | 0  |
| p,p DDE             | 0.186 | (Brazil)  | 21.290  | (Brazil)  | $5.050 \pm 0.272$   | 300 | 11      | 44      | 0        | 0  |
| Endrin              | 0.669 | (India)   | 1.220   | (Holland) | 1 ± 0.010           | 50  | 4       | 16      | 0        | 0  |
| p,p DDD             | 0.686 | (India)   | 1.340   | (Sudan)   | 1 ± 0.009           | 300 | 6       | 24      | 0        | 0  |
| p,p, DDT            | 0.652 | (Brazil)  | 15.324  | (India)   | 4.800 ± 0.184       | 300 | 11      | 44      | 0        | 0  |

A) FAO/WHO Food Standards, Codex alimentarius, Maximum Residue Limits of Pesticide in Food (13).

B) Pesticide EU-MRLs, Regulation (EC) NO 396/2005, updated on 02/12/2009 (16).

Table 3: Residues of some Organochlorines pesticides on 25 samples of locally raised chicken (ppb)

| Pesticide           | Min.  | Origin      | Max.    | Origin      | Mean ± SE        | MRL | (+ ) Incidence |    | Violation |      |
|---------------------|-------|-------------|---------|-------------|------------------|-----|----------------|----|-----------|------|
|                     |       |             |         |             |                  |     | No.            | %  | No.       | %    |
| α - НСН             | 0.180 | (Badrashen) | 95.682  | (Hwamdeya)  | $8.37 \pm 0.968$ | 200 | 15             | 60 | 0         | 0    |
| γ - HCH             | 2.249 | (Hwamdeya)  | 0.027   | (Badrashen) | 0.33 ± 0.021     | 20  | 17             | 68 | 0         | 0    |
| Δ - НСН             | 0.473 | (Badrashen) | 143.767 | (Hwamdeya)  | 31.80 ± 1.870    | 200 | 18             | 72 | 0         | 0    |
| Heptachlor          | 0.360 | (El Saf )   | 37.357  | (Badrashen) | 6.95 ± 0.364     | 200 | 17             | 68 | 0         | 0    |
| Aldrin              | 0.015 | (Badrashen) | 7.770   | (Giza)      | 0.67 ± 0.064     | 200 | 24             | 96 | 0         | 0    |
| Heptachlor- epoxide | 0.007 | (Badrashen) | 4.987   | (Hwamdeya)  | 0.44 ± 0.0432    | 200 | 21             | 84 | 0         | 0    |
| γ - Chlordan        | 0.070 | (Badrashen) | 8.438   | (Hwamdeya)  | 2.07 ± 0.088     | 50  | 15             | 60 | 0         | 0    |
| Endosulfan II       | 2.536 | (Giza)      | 79.394  | (Hwamdeya)  | 26.40 ± 0.880    | 50  | 24             | 96 | 3         | 12.5 |
| Dieldrin            | 0.330 | (El Ayaat ) | 72.797  | (Hwamdeya)  | 15 ± 0.692       | 200 | 24             | 96 | 0         | 0    |
| p,p DDE             | 0.030 | (Badrashen) | 2.626   | (Hwamdeya)  | 0.88 ± 0.036     | 300 | 14             | 56 | 0         | 0    |
| Endrin              | 0.069 | (Badrashen) | 1.041   | (Giza)      | 0.47 ± 0.010     | 50  | 14             | 56 | 0         | 0    |
| p,p DDD             | 0.090 | (Badrashen) | 38.906  | (Giza)      | 6.15 ± 0.512     | 300 | 13             | 52 | 0         | 0    |
| p,p, DDT            | 0.035 | (Badrashen) | 93.850  | (El Saf)    | 1.48 ± 0.040     | 300 | 23             | 92 | 0         | 0    |

| Pesticide           | Mın.  | Origin      | Max.    | Origin      | Mean ± SE         | MRL | (+ ) In | cidence | Violation |    |
|---------------------|-------|-------------|---------|-------------|-------------------|-----|---------|---------|-----------|----|
|                     |       |             |         |             |                   |     | No.     | %       | No.       | %  |
| α - НСН             | 0.213 | (Badrashen) | 27.712  | (Hwamdeya)  | $5.47 \pm 0.306$  | 200 | 16      | 4       | 0         | 0  |
| γ - HCH             | 0.064 | (Giza )     | 5.199   | (Hwamdeya)  | 1.40 ± 0.081      | 20  | 13      | 52      | 0         | 0  |
| Δ - HCH             | 12.73 | (El Saf)    | 34.155  | (Giza )     | $10.97 \pm 0.367$ | 200 | 19      | 76      | 0         | 0  |
| Heptachlor          | 0.668 | (Giza )     | 9.786   | (Badrashen) | 7.57 ± 0.102      | 200 | 14      | 56      | 0         | 0  |
| Aldrin              | 0.049 | (El Ayaat)  | 45.905  | (El Saf)    | 2.43 ± 0.021      | 200 | 25      | 100     | 0         | 0  |
| Heptachlor- epoxide | 0.036 | (Giza )     | 0.546   | (Giza )     | 0.28 ± 0.007      | 200 | 15      | 60      | 0         | 0  |
| γ - Chlordan        | 0.340 | (El Ayaat)  | 10.034  | (Hwamdeya)  | 1.99 ± 0.101      | 50  | 15      | 60      | 0         | 0  |
| Endosulfan II       | 1.388 | (Badrashen) | 0126.28 | (El Ayaat)  | 30.80 ± 1.270     | 50  | 25      | 100     | 5         | 20 |
| Dieldrin            | 0.889 | (Giza )     | 63.464  | (El Ayaat)  | 11.40 ± 0.504     | 200 | 25      | 100     | 0         | 0  |
| p,p DDE             | 0.077 | (Badrashen) | 27.695  | (El Ayaat)  | 3.85 ± 0.295      | 300 | 14      | 56      | 0         | 0  |
| Endrin              | 0.695 | (Giza )     | 7.756   | (El Ayaat)  | 2.61 ± 0.116      | 50  | 9       | 36      | 0         | 0  |
| p,p DDD             | 0.325 | (El Ayaat)  | 1.846   | (Giza )     | 3.03 ± 0.174      | 300 | 13      | 12      | 0         | 0  |
| p,p, DDT            | 0.548 | (El Ayaat)  | 218.291 | (Hwamdeya)  | 13.80 ± 1.720     | 300 | 25      | 100     | 0         | 0  |

| Table 5. | Daciduae | of como | Organophosphorus | posticidos s | 25    | comple  | of locally  | roised objeken (nn | h)  |
|----------|----------|---------|------------------|--------------|-------|---------|-------------|--------------------|-----|
| Table 5: | Residues | or some | Organophosphorus | pesticides c | on 25 | samples | oi iocany i | aisea chicken (bb  | ומי |

| Pesticide       | Min.         | Origin      | Max.  | Origin     | Mean ± SE         | MRL                 | (+ ) Inci | dence | Violati | on  |
|-----------------|--------------|-------------|-------|------------|-------------------|---------------------|-----------|-------|---------|-----|
|                 |              |             |       |            |                   |                     | No        | %     | No      | %   |
| Ethoprophos     | 1.37         | (Badrashen) | 44.46 | (El Ayaat) | $15.70 \pm 0.780$ | 10                  | 4         | 16    | 1       | 16  |
| Cadusafos       | 3.45         | (Badrashen) | 8.13  | (El Saf)   | 5.79 ± 0.132      | No MRLs established | 2         | 8     | ?       | ?   |
| Propetamphos    | ND           |             | ND    |            | _                 | No MRLs established | _         | _     | ?       | ?   |
| Diazinon        | 41           | (El Ayaat)  | 66.37 | (El Ayaat) | 53.70 ± 0.716     | 20                  | 2         | 8     | 2       | 100 |
| Chlorpyrifos-Me | 6.27         | (El Saf)    | 26.06 | (Giza)     | 14.90 ± 0.404     | 50                  | 3         | 12    | 0       | 0   |
| Pirimiphos -Me  | ND           |             | ND    |            | ND                | 50                  | _         | _     | _       | _   |
| Chlorpyrifos    | 10.75        | (Badrashen) | 67.04 | (El Ayaat) | 3.32 ± 1.190      | 50                  | 3         | 12    | 1       | 33  |
| Quinalphos      | 3.34         | (El Saf)    | 21.08 | (El Saf)   | 12.40 ± 0.355     | No MRLs established | 3         | 12    | ?       | ?   |
| Prothiophos     | 55.77 (Giza) |             |       |            | _                 | No MRLs established | 1         | 4     | ?       | ?   |
| Ethion          | ND           |             | ND    |            | _                 | No MRLs established |           |       | ?       |     |
| Azinphos-Me     | 17.50 (Giza) |             |       |            |                   | 10                  | 1         | 4     | 1       | 100 |

Table 6: Residues of some Organophosphorus pesticides on 25 samples of locally raised bovine (ppb)

| Pesticide       | Min.            | Origin     | Max.  | Origin      | Mean ± SE        | MRL                 | (+ ) Inci | dence | Violation |    |
|-----------------|-----------------|------------|-------|-------------|------------------|---------------------|-----------|-------|-----------|----|
|                 |                 |            |       |             |                  |                     | No.       | %     | No.       | %  |
| Ethoprophos     | 2.37            | (Giza)     | 16.18 | (Badrashen) | $8.06 \pm 0.223$ | 10                  | 8         | 32    | 2         | 25 |
| Cadusafos       | 0.07            | (El Saf)   | 66.06 | (Hwamdeya)  | 17.40 ± 1        | No MRLs established | 6         | 24    | ?         | ?  |
| Propetamphos    | ND              |            | ND    |             | ND               | No MRLs established | _         | _     | ?         | ?  |
| Diazinon        | ND              |            | ND    |             | ND               | 50                  | _         |       | _         | _  |
| Chlorpyrifos-Me | 11.87           | (Hwamdeya) | 37.30 | (Hwamdeya)  | 22.10 ± 0.435    | 50                  | 4         | 16    | 0         | 0  |
| Pirimiphos -Me  | 8.88 (Hwamdeya) |            |       |             | _                | 50                  | 1         | 0     | 0         | 0  |
| Chlorpyrifos    | 8.21            | (Giza)     | 73.23 | (Badrashen) | 26.50 ± 1.270    | 1000                | 5         | 20    | 0         | 0  |
| Quinalphos      | 10.82           | (El Saf)   | 13.22 | (Giza)      | 12 ± 0.068       | No MRLs established | 2         | 8     | ?         | ?  |
| Prothiophos     | 12.99           | (Giza)     | 18.44 | (El Saf)    | 16 ± 0.111       | No MRLs established | 3         | 12    | ?         | ?  |
| Ethion          | ND              |            | ND    | -           | _                | No MRLs established |           | _     | ?         | ?  |
| Azinphos-Me     | 3.10            | (Giza)     | 26.96 | (Badrashen) | 16.30 ± 0.485    | 10                  | 3         | 12    | 1         | 33 |

Table 7: Residues of some Organophosphorus pesticides on 25 samples of imported bovine (ppb)

| Pesticide       | Min.    | Origin  | Max.  | Origin  | Mean ± SE         | MRL                 | (+ ) In | cidence     | Viola | tion |
|-----------------|---------|---------|-------|---------|-------------------|---------------------|---------|-------------|-------|------|
|                 |         |         |       |         |                   |                     | No.     | %           | No.   | %    |
| Ethoprophos     | 0.26    | (Sudan) | 21.25 | (India) | $6.25 \pm 0.278$  | 10                  | 8       | 32          | 3     | 37.5 |
| Cadusafos       | 6.62    | (India) | 19.61 | (India) | 13.10 ± 0.368     | No MRLs established | 2       | 8           | ?     | ?    |
| Propetamphos    | ND      |         | ND    |         | <del></del>       | No MRLs established | _       | <del></del> | ?     | ?    |
| Diazinon        | 4.28    | (Sudan) | 15.26 | (Sudan) | 8.89 ± 0.228      | 50                  | 3       | 12          | 0     | 0    |
| Chlorpyrifos-Me | (India) | 11.96   |       |         | <del>-</del>      | 50                  | 1       | 4           | 0     | 0    |
| Pirimiphos -Me  | 6.74    | (Sudan) | 13.43 | (Sudan) | 10.10 ± 0.189     | 50                  | 2       | 8           | 0     | 0    |
| Chlorpyrifos    | 9.74    | (Sudan) | 29.02 | (Sudan) | $16.60 \pm 0.309$ | 1000                | 7       | 28          | 0     | 0    |
| Quinalphos      | 11.84   | (India) | 23.43 | (India) | $17.60 \pm 0.328$ | No MRLs established | 2       | 8           | ?     | ?    |
| Prothiophos     | 6.17    | (Sudan) | 10.8  | (Sudan) | 8.49 ± 0.131      | No MRLs established | 2       | 8           | ?     | ?    |
| Ethion          | ND      |         | ND    |         | <del></del>       | No MRLs established |         |             | ?     | ?    |
| Azinphos-Me     | ND      |         | ND    |         | <del></del>       | 10                  |         |             |       |      |

Table 8: Residues of some Organophosphorus pesticides on 25 samples of imported chicken (ppb)

| Pesticide       | Min.  | Max.  | Mean ± SE           | MRL                 | (+ ) Inci    | dence       | Violation   |             |  |
|-----------------|-------|-------|---------------------|---------------------|--------------|-------------|-------------|-------------|--|
|                 |       |       |                     |                     | No.          | %           | No.         | %           |  |
| Ethoprophos     | 1.521 | 3.86  | $2.60\ \pm\ 0.0472$ | 10                  | 3            | 12          | 0           | 0           |  |
| Cadusafos       | ND    | ND    | <del></del>         | No MRLs established | <del></del>  | <del></del> | ?           | ?           |  |
| Propetamphos    | ND    | ND    | <del>_</del>        | No MRLs established | <del></del>  | _           | ?           | ?           |  |
| Diazinon        | ND    | ND    | <del>_</del>        | 20                  | <del></del>  | <del></del> | <del></del> | <del></del> |  |
| Chlorpyrifos-Me | ND    | ND    | <del></del>         | 50                  | <del></del>  | <del></del> | <del></del> |             |  |
| Pirimiphos -Me  | 29.14 |       |                     | 50                  | 1            | 4           | 0           | 0           |  |
| 1.5             | 5.080 | 43.93 | 19.70 ± 0.844       | 50                  | 3            | 12          | 0           | 0           |  |
| ` '             | ND    | ND    | <del>_</del>        | No MRLs established | _            | _           | ?           | ?           |  |
| Prothiophos     | ND    | ND    | <del>_</del>        | No MRLs established | _            | _           | ?           | ?           |  |
| Ethion          | ND    | ND    | <del>_</del>        | No MRLs established | <del>-</del> | _           | ?           | ?           |  |
| Azinphos-Me     | ND    | ND    |                     | 10                  | _            | _           | _           | _           |  |

Heptachlor epoxide, the mean concentration was 0.052 and 0.030 ppb respectively. Heptachlor residues were found in 3 samples while Heptachlor epoxide residues were found in 1 sample.

In the same study, 270 meat samples were analyzed for the same pesticides; the mean concentration of Aldrin residues was 0.470 and found in 1 sample. pp- DDD residues were found in 6 samples with concentration range 0.010-0.150 ppb, while pp- DDE residues were found in 74 samples with concentration range of 0.005-0.150 ppb, and finally pp- DDT residues were found in 16 samples with concentration range of 0.010-0.500 ppb.

Also in analyzed the meat samples for the organochlorine pesticides,  $\alpha$ -HCH residues were found in 21 samples with concentration range of 0.006–0.500 ppb, while  $\beta$ -HCH residues were found in 14 samples with concentration range of (0.009–0.050) and  $\gamma$ -HCH residues were found in 37 samples with concentration of 0.005–2.890 ppb. For Heptachlor and Heptachlor epoxide, concentration range was 0.010–0.220 ppb and 0.010–0.050 ppb respectively. Heptachlor were found in 10 samples while Heptachlor epoxide was found in 7 samples.

In another study conducted by Tao *et al.*<sup>[15]</sup>, 113 chicken muscle samples were collected from 12 provinces in China and were tested for organochlorines residues, and the concentrations of the residues were (in ng/g):  $\alpha$  –HCH (0.009  $\pm$  0.008),  $\beta$  -HCH (0.026  $\pm$  0.022),  $\gamma$  –HCH (0.008  $\pm$  0.008),  $\Delta$  –HCH (0.011  $\pm$  0.011), p,p-DDT (0.035  $\pm$  0.028), p,p-DDD (0.026  $\pm$  0.016), p,p-DDE (0.034  $\pm$  0.021), o,p-DDT (0.008  $\pm$  0.006), o,p-DDD (0.010  $\pm$  0.012) and o,p-DDE (0.009  $\pm$  0.012).

## 6.1. Organochlorine Pesticide Residues:

- **6.2.** Organophosphorus Pesticide Residues: In conclusion, many samples were found to be contaminated with either organochlorines or organophosphorus. These pesticides might have reached the animals via different ways:
- 1- Waste water of irrigation after spraying the crops,
- 2- Animal feeds that were contaminated during the pesticides treatments,
- 3- Air that was polluted with pesticides during spraying

These residues will accumulate in the human body by eating the meat of these animals which were contaminated with pesticides and this will result in diseases and other hazards for human. Therefore, we recommend that there should be a continuous program designed for analyzing and monitoring meat and meat products in the Egyptian local market before using to make sure that the levels of pesticide residues don't exceed the MLR. Also the usage of the banned pesticide should be stopped. In addition, Government should apply a training program to the farmers and show them the hazardous effects of using such banned pesticides or even using pesticides higher than the recommended doses.

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