



## Seasonal variations on the levels of some pesticide residues in dairy farms

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### ABSTRACT

The present work was carried out to study the effect of seasonal variations on the levels of pesticide residues in water, feeds and milk. Our results illustrated that the significantly highest concentrations of OCs in water and feed were detected during summer [(1.05±0.35, 0.72±0.24, 0.77±0.25, 2.5±0.70 and 0.81±0.27 µg/L, for p.p.DDT, aldrin, β-BHC, lindane and γ.chlordane respectively in water samples) and (30.0±10.20, 45.00±13.02, 65.33±21.90, 35.33±13.20, 43.67±13.30, 22.00±7.33 and 19.13±8.6 µg/kg, for p.p.DDD, p.p.DDE, p.p.DDT, aldrin, lindane, γ.chlordane and methoxychlor, respectively in feed samples)], while the lowest were detected during spring, on the other hand, none of pesticides detected in winter which may be attributed to environmental factors. The significantly highest concentration of diazinon in water and feed was recorded during autumn (78.75±26.30 µg/L and 140.00±64.60 µg/kg, respectively), while the lowest levels were detected during spring. Also, the highest concentrations of deltamethrin was detected during spring in water (55.30±18.50 µg/L) and during summer in feed samples (78.40±26.70 µg/kg), while the lowest levels were detected during autumn in water and during spring in feed samples. The significantly highest concentrations of residues in milk were detected during summer (p.p.DDD, p.p. DDE, p.p.DDT, heptachlor, heptachlor epoxide, γ.chlordane, methoxychlor, diazinon and deltamethrin were 20.10±6.01, 52.30±17.20, 33.20±20.06, 1.16±0.38, 6.72±0.90, 2.570±2.23, 0.70±0.20, 58.50±19.20 and 45.13±15.10 µg/kg, respectively) and the lowest were detected in winter. Our results concluded that the seasonal variations greatly influence the levels of all examined pesticide residues with variable degree specially diazinon and deltamethrin.

**Keywords:** seasonal variations, OCs, Diazinon, Deltamethrin, Milk, Water, Feed.

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### 1. INTRODUCTION

Pesticide residues in milk, milk products, animal feed and water emphasizing the need of monitoring of pesticide residues regularly to prevent human hazards. Human and animal cannot avoid exposure to pesticides directly especially during its application as well as through consumption of contaminated cereals, vegetables and fruits harvested in same country or imported products from other countries such as egg, milk and meats. Presence of persistence and hazardous agrochemicals in human diet is an emerging problem in developing countries which encounter poor institutional progress and slow financial development (Shahzadi et al., 2013). Many factors affect the environmental fate of

pesticides as the chemical behavior, solubility in water and adsorption to the soil and sediments as well as microbial, chemical and physical degradation (Ian, 2004). The high level of pesticide residues in feedstuffs may be due to post-harvest treatment, commercial aerial application and feedstuffs manufactured from plant material, grass and hay treated during the growing season with insecticides (Fernandez et al., 2008). About 75 % pesticides are being in use by the developed countries for saving farmer money by killing the unwanted insects or pests during summer in the crops by increasing production yield (Zahoor et al., 2013). Pesticides are widely used in agriculture and can be transferred to animal's milk in a

number of ways through contaminated feeds and water as well as during application of pesticides. The feed to milk transference of pesticides is influenced by the quantity ingested, absorption, pesticide metabolism and excretion by animals in production (LeDoux, 2011). Pesticide residues ruminants milk represent a public health hazard, since milk and dairy products are widely consumed by infants, children and adults throughout the world. In view of this, many countries have enacted regulations to set the permissible limits of such pesticide residues in milk and dairy product to protect the consumers (Tsiplakou *et al.*, 2010). This work aimed to study the effect of seasonal variations on levels of pesticide residues in water, feeds and milk.

## 2. MATERIAL AND METHODS

### 2.1. Sampling

A total of 144 samples were collected from two dairy farms located at Kaliobia governorate during winter, spring, summer and autumn 2014. Two samples were collected per visit from water, feed and milk in three visits with month interval per every season. Water samples were collected from drinkers and the main source from each farm per visit and stored in darkness at 4°C and extracted within 14 days according to Vanderford *et al.*, (2011). While feed samples included all components of the animal diet (roughage, silage, soybean concentrates and minerals), from storage room and feed troughs. All the samples were placed in plastic bags and transported in ice boxes and stored at -20°C until processing (Fagnani *et al.*, 2011). Raw milk (1L) were collected directly from the cooling tanks of the farms in air tight well labeled polyethylene bags. Every 300 ml of chilled milk was placed in bags and stored at -20 °C and were analyzed within 48 hours (Fagnani *et al.*, 2011).

### 2.2. Standards

Analytical standards of organochlorine (93-99%) procured from Sigma-Aldrich, standards of diazinon were from obtained from GmbH (Augsburg, Germany) and

deltamethrin (97% to 99%) was granted by Pak China.

### 2.3. Determination of organochlorine

Determination OCs in water samples was carried out by using gas chromatography method using electron capture detector (ECD) according to Erkmen *et al.*, (2013), while determination in feeds was according to Panseri *et al.*, (2013). OCs in milk was detected was carried out according to Heck *et al.*, (2007) by using gas chromatography analysis.

### 2.4. Determination of diazinon

The procedure of water samples was carried out by using gas chromatography method using flame photometric detector (FPD) according to Martinez Vedail *et al.* (2002), while in feeds the protocol of QuEChERS Mini-residue method used for analysis of diazinon concerning commodities with high fat content was followed (Anastassiades *et al.*, 2003). Because there are a lot of modifications of the QuEChERS method published, the procedure followed in this study is described in detail (QuEChERS, 2009). Diazinon detection in milk was carried out by GC equipped with an electron capture detector according to Walorczyk (2008).

### 2.5. Determination of deltamethrin

The determination of deltamethrin in water samples was done with HPLC-UV by chromatograph equipped with UV detector according to Flavio *et al.*, (1999), while in feeds was used HPLC-UV detector according to Boussahel *et al.*, (2006). Deltamethrin detection in milk: was conducted on HPLC system, Photodiode array (PDA) detector according to Darko and Acquah (2008).

### 2.6. Statistical analysis

SPSS Microsoft version 16.0.1. One-way ANOVA tests were applied to check the significant difference (SPSS, 2004).

## 3. RESULTS

It is clear from Table (1) that the highest mean values of p.p. DDT, aldrin,  $\beta$ -BHC, lindane and  $\gamma$ .chlordaneresidues detected in water samples was observed during summer were  $1.05\pm0.35$ ,  $0.72\pm0.24$ ,  $0.77\pm0.25$ ,  $2.5\pm0.70$  and  $0.81\pm0.27$   $\mu\text{g/L}$ , respectively, while the lowest concentration was observed during spring. None of them were detected during winter. Heptachlor epoxide was detected in water samples during autumn ( $0.17\pm0.05$   $\mu\text{g/L}$ ), followed by summer ( $0.21\pm0.07$   $\mu\text{g/L}$ ) and lastly during spring ( $0.03\pm0.01$   $\mu\text{g/L}$ ). Alpha-HCH was detected only in water samples collected during spring at a level of  $1.93\pm0.64\mu\text{g/L}$ , while its level during summer was  $0.99\pm0.33\mu\text{g/L}$ .

The results demonstrated in Table (1) revealed that the concentrations of diazinon was significantly highest during autumn ( $78.75\pm26.30$   $\mu\text{g/L}$ ), followed by summer ( $63.9\pm21.3\mu\text{g/L}$ ) finally spring ( $60.8\pm20.26$   $\mu\text{g/L}$ ), while not detected during winter. The highest mean value of deltamethrin was recorded during spring ( $55.30\pm18.50$   $\mu\text{g/L}$ ) followed by summer ( $26.99\pm8.19\mu\text{g/L}$ ) and finally autumn ( $17.2\pm5.40$   $\mu\text{g/L}$ ), while during winter all the examined samples were deltamethrin free.

The obtained results illustrated in Table (2) showed that the highest significant mean values of p.p.DDD, p.p.DDE, p.p.DDT, aldrin, lindane,  $\gamma$ .chlordane and methoxychlor were recorded in feed samples collected during summer ( $30.0\pm10.20$ ,  $45.00\pm13.02$ ,  $65.33\pm21.90$ ,  $35.33\pm13.20$ ,  $43.67\pm13.30$ ,  $22.00\pm7.33$  and  $19.13\pm8.6$   $\mu\text{g/kg}$ , respectively), while the lowest was recorded during spring. The highest concentration of heptachlor was detected in feed samples collected during autumn ( $40.60\pm13.03$   $\mu\text{g/kg}$ ) followed by spring ( $32.33\pm10.50\mu\text{g/kg}$ ) and summer ( $20.00\pm6.20$   $\mu\text{g/kg}$ ). The data demonstrated in Table (2) revealed that the highest mean values of diazinon were detected in feed samples collected during summer ( $140.00\pm64.60$   $\mu\text{g/kg}$ ) followed by autumn ( $57.00\pm19.90\mu\text{g/kg}$ ) and finally during

spring ( $14.00\pm4.06$   $\mu\text{g/kg}$ ). Deltamethrin at a high level was detected in feed samples collected during summer ( $79.97\pm27.2\mu\text{g/kg}$ ) followed by autumn ( $78.40\pm26.70$   $\mu\text{g/kg}$ ) then during spring ( $15.48\pm5.20\mu\text{g/kg}$ ).

The results illustrated in Table (3) clarified that the highest mean concentration of p.p. DDD, p.p. DDE, p.p.DDT, heptachlor, heptachlor epoxide,  $\gamma$ .chlordane and methoxychlor were detected in milk samples collected during summer ( $20.10\pm6.01$ ,  $52.30\pm17.20$ ,  $33.20\pm20.06$ ,  $1.16\pm0.38$ ,  $6.72\pm0.90$ ,  $2.570\pm2.23$  and  $0.70\pm0.20$   $\mu\text{g/kg}$ , respectively), the lowest were detected in spring. The highest mean values of aldrin and lindane were detected in milk samples collected during summer ( $5.33\pm1.77$  and  $26.70\pm8.20$   $\mu\text{g/kg}$ , respectively), while the lowest were detected in winter ( $2.45\pm 0.81$  and  $0.65\pm0.21$   $\mu\text{g/kg}$ , respectively). The obtained results showed that  $\alpha$ -BHC and  $\beta$ -BHC were detected only in milk samples collected during autumn and spring with at mean concentrations of  $9.30\pm3.09$ ,  $8.30\pm2.76$ ,  $11.20\pm3.73$  and  $10.20\pm0.34$   $\mu\text{g/kg}$ , respectively. The results demonstrated in Table (3) showed that the highest mean value of diazinon was detected in milk samples during summer ( $58.50\pm19.20$   $\mu\text{g/kg}$ ) followed by spring ( $42.50\pm14.20$   $\mu\text{g/kg}$ ) then autumn ( $16.18\pm5.60\mu\text{g/kg}$ ) and finally winter ( $7.00\pm2.30$   $\mu\text{g/kg}$ ). The highest mean value of deltamethrin was detected in milk samples during summer ( $45.13 \pm 15.10$   $\mu\text{g/kg}$ ) followed by autumn ( $30.93 \pm 10.80$   $\mu\text{g/kg}$ ) then spring ( $26.80\pm8.90\mu\text{g/kg}$ ), while it was not detected in winter.

#### 4. DISCUSSION

The findings about p.p.DDT, aldrin,  $\beta$ -BHC, lindane and  $\gamma$ .chlordane residues were nearly similar to that obtained by Abdel Razik (2006), Badach et al., (2007) and Enbaia et al., (2014) who concluded that the incidences of organochlorines were relatively high in aquatic environment from

Table (1): Effect of seasonal variations on the levels of pesticide residues in water samples (n=48) ( $\mu\text{g/L}$ ).

Pesticides	Summer			Autumn			winter			spring		
	Min	Max	Mean $\pm$ SE	Min	Max	Mean $\pm$ SE	Min	Max	Mean $\pm$ SE	Min	Max	Mean $\pm$ SE
p.p.DDD	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
p.p.DDE	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
p.p.DDT	0.40	1.70	1.05 $\pm$ 0.35 <sup>a</sup>	0.65	1.87	0.76 $\pm$ 0.26 <sup>b</sup>	ND	ND	ND	0.30	1.90	0.60 $\pm$ 0.20 <sup>b</sup>
Dieldrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Aldrin	0.45	0.99	0.72 $\pm$ 0.24 <sup>a</sup>	0.06	0.39	0.25 $\pm$ 0.17 <sup>b</sup>	ND	ND	ND	0.30	0.138	0.03 $\pm$ 0.01 <sup>c</sup>
Heptachlor	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Heptachlor epoxide	0.09	0.311	0.21 $\pm$ 0.07 <sup>b</sup>	0.50	0.29	0.17 $\pm$ 0.05 <sup>a</sup>	ND	ND	ND	0.09	0.046	0.03 $\pm$ 0.01 <sup>c</sup>
$\alpha$ -BHC	0.11	1.10	0.99 $\pm$ 0.33 <sup>b</sup>	ND	ND	ND	ND	ND	ND	0.81	3.86	1.93 $\pm$ 0.46 <sup>a</sup>
B-BHC	0.11	1.15	0.77 $\pm$ 0.25 <sup>a</sup>	ND	ND	ND	ND	ND	ND	0.14	2.01	1.02 $\pm$ 0.34 <sup>a</sup>
Lindane	0.43	3.7	2.50 $\pm$ 0.70 <sup>a</sup>	0.29	2.33	1.31 $\pm$ 0.43 <sup>b</sup>	ND	ND	ND	0.11	2.23	1.19 $\pm$ 0.39 <sup>b</sup>
Endrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
$\gamma$ .chlordan	0.61	1.00	0.84 $\pm$ 0.06 <sup>a</sup>	ND	ND	ND	ND	ND	ND	0.32	0.78	0.62 $\pm$ 0.20 <sup>a</sup>
HCB	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Methoxychlor	0.13	1.80	0.81 $\pm$ 0.27 <sup>a</sup>	0.14	0.98	0.71 $\pm$ 0.23 <sup>a</sup>	ND	ND	ND	ND	ND	ND
Diazinon	33.00	94.40	63.90 $\pm$ 21.3 <sup>b</sup>	25.00	94.15	78.75 $\pm$ 26.30 <sup>a</sup>	ND	ND	ND	50.00	91.00	60.80 $\pm$ 20.26 <sup>b</sup>
Deltamethrin	11.00	32.80	26.99 $\pm$ 8.19 <sup>b</sup>	13.00	22.00	17.20 $\pm$ 5.40 <sup>b</sup>	ND	ND	ND	23.00	87.00	55.30 $\pm$ 18.50 <sup>a</sup>

The different litter in the same row indicates significant difference according to one way ANOVA ( $p \leq 0.05$ ).

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Table (2): Effect of seasonal variations on the levels of pesticides residues in feeds samples (n=48).

Pesticides	Summer			Autumn			winter			Spring		
	Min	Max	Mean±SE	Min	Max	Mean± SE	Min	Max	Mean±SE	Min	Max	Mean±SE
p.p.DDD	12.80	47.00	30.00±10.20 <sup>a</sup>	18.00	22.00	20.00±6.20 <sup>b</sup>	ND	ND	ND	14.20	23.70	17.09±5.20 <sup>b</sup>
p.p.DDE	15.30	60.44	45.00±13.02 <sup>a</sup>	17.50	56.00	41.00±11.50 <sup>a</sup>	ND	ND	ND	20.32	30.11	24.00±8.03 <sup>b</sup>
p.p.DDT	15.00	115.66	65.33±21.90 <sup>a</sup>	31.00	50.00	36.00±12.50 <sup>c</sup>	ND	ND	ND	30.70	50.09	40.00±13.06 <sup>b</sup>
Dieldrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Aldrin	7.00	41.00	35.33±13.20 <sup>a</sup>	11.00	45.00	32.00±12.70 <sup>b</sup>	ND	ND	ND	ND	ND	ND
Heptachlor	18.00	22.00	20.00±6.20 <sup>c</sup>	38.00	42.00	40.60±13.03 <sup>a</sup>	ND	ND	ND	13.00	38.00	32.33±10.50 <sup>c</sup>
Heptachlor epoxide	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
α-BHC	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
B-BHC	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Lindane	13.00	58.00	43.67±13.30 <sup>a</sup>	16.00	40.00	37.67±13.90 <sup>b</sup>	ND	ND	ND	28.00	41.00	39.67±12.90 <sup>b</sup>
Endrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
γ.chlordane	11.00	30.00	22.00±7.33 <sup>a</sup>	ND	ND	ND	ND	ND	ND	14.80	32.40	23.50±7.30 <sup>a</sup>
HCB	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Methoxychlor	3.14	33.00	19.13±6.60 <sup>a</sup>	1.01	22.00	18.60±5.20 <sup>b</sup>	ND	ND	ND	ND	ND	ND
Diazinon	11.90	266.00	140.00±64.60 <sup>a</sup>	13.40	75.00	57.00±19.90 <sup>a</sup>	ND	ND	ND	1.80	18.00	14.00±4.06 <sup>c</sup>
Deltamethrin	67.10	92.00	79.97±27.20 <sup>a</sup>	63.30	93.50	78.40±26.70 <sup>a</sup>	ND	ND	ND	6.30	20.10	15.48±5.20 <sup>b</sup>

The different litter in the same row indicates significant difference according to one way ANOVA ( $p \leq 0.05$ )

Table (3): Effect of seasonal variations on the levels of pesticides residues in milk samples (n= 48).

Pesticide	Summer			Autumn			winter			spring		
	Min	Max	Mean±SE	Min	Max	Mean±SE	Min	Max	Mean±SE	Min	Max	Mean±SE
p.p.DDD	0.18	42.20	20.10±6.01 <sup>a</sup>	0.19	32.50	18.17±6.07 <sup>b</sup>	ND	ND	ND	0.19	26.00	13.00±4.33 <sup>c</sup>
p.p.DDE	1.10	68.00	52.30±17.20 <sup>a</sup>	1.07	33.00	21.60±7.07 <sup>b</sup>	ND	ND	ND	2.09	31.20	21.03±7.01 <sup>b</sup>
p.p.DDT	0.22	44.40	33.20±11.06 <sup>a</sup>	0.22	38.80	25.30±8.90 <sup>b</sup>	0.17	3.00	2.00±0.66 <sup>c</sup>	0.33	32.70	23.00±7.67 <sup>b</sup>
Dieldrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Aldrin	0.70	6.10	5.33±1.77 <sup>a</sup>	0.11	5.50	3.00±1.00 <sup>b</sup>	0.06	3.91	2.45±0.81 <sup>c</sup>	ND	ND	ND
Heptachlor	0.50	1.90	1.16±0.38 <sup>a</sup>	0.70	0.90	0.80±0.26 <sup>b</sup>	ND	ND	ND	0.70	1.00	0.83±0.27 <sup>b</sup>
Heptachlor epoxide	0.15	13.27	6.71±2.23 <sup>a</sup>	0.15	4.10	3.72±1.24 <sup>b</sup>	ND	ND	ND	0.90	1.50	1.17±0.57 <sup>c</sup>
α-BHC	ND	ND	ND	0.61	12.70	8.30±2.76 <sup>a</sup>	ND	ND	ND	0.81	17.97	9.30±3.09 <sup>a</sup>
B-BHC	ND	ND	ND	0.19	23.10	11.20±3.73 <sup>a</sup>	ND	ND	ND	0.14	20.10	10.20±3.40 <sup>a</sup>
Lindane	0.30	33.10	26.70±8.20 <sup>a</sup>	0.70	12.90	9.80±3.26 <sup>b</sup>	0.20	1.04	0.64±0.21 <sup>d</sup>	0.95	4.00	2.12±0.70 <sup>c</sup>
Endrin	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
γ.chlordane	0.10	3.70	2.57±0.85 <sup>a</sup>	ND	ND	ND	ND	ND	ND	0.17	0.20	0.16±0.05 <sup>c</sup>
HCB	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Methoxychlor	0.40	1.10	0.70±0.23 <sup>a</sup>	ND	ND	ND	ND	ND	ND	0.50	2.10	0.80±0.26 <sup>a</sup>
Diazinon	26.60	96.40	58.50±19.20 <sup>a</sup>	2.20	30.16	16.18±5.60 <sup>c</sup>	8.40	18.80	7.00±2.30 <sup>d</sup>	26.60	66.40	42.50±14.20 <sup>b</sup>
Deltamethrin	3.14	54.50	45.13±15.10 <sup>a</sup>	13.00	48.56	30.93±10.80 <sup>b</sup>	ND	ND	ND	2.09	31.00	26.80±8.90 <sup>c</sup>

The different litter in the same row indicates significant difference according to one way ANOVA ( $p \leq 0.05$ ).

different locations in Egypt during summer and autumn. Variation in levels of pesticide residues during different season may be due to physical and chemical factors of environment, so the environmental variations can affect the concentration of pesticide residues (Ian, 2004).

On the other hand, these findings were disagreed with those obtained by Samia El-Hoshy (1989) and Abou-Arab et al., (1995) who found that the total DDT were predominant in water samples collected during winter. Also, pesticides applied directly to the soil may be washed off by rain into nearby bodies of surface water or percolate through the soil to lower soil layers and ground water (Kamrin, 1997), or may be due to illegal uses of pesticide at certain season (Aydin et al., 2013).

Results concerning heptachlor epoxide were nearly similar to those obtained by Ahmed (1997) and Abdel Razik (2006) who found that the concentration of heptachlor-epoxide was ranged from 0.23-58.17  $\mu\text{g/L}$  in surface water collected in Kafr El-Sheikh governorate. On the other hand, Aydin et al., (2013) detected heptachlor epoxide in water samples collected during spring, while samples collected during summer were free. It is clear that  $\alpha$ -HCH was detected only in water samples collected during spring. The current results agreed to some extent with those reported by Abdel Razik et al., (1988). On the other hand the findings were disagreed with Abdel Razik (2006) who detected  $\alpha$ -BHC in water samples collected during winter in Kafr El-Sheikh governorate. The results illustrated in Table (1) clarified that there were significance differences among different seasons for the mean values of p.p.DDT, aldrin, heptachlor epoxide,  $\alpha$ -BHC and lindane, this may be attributed to the degradation of pesticides during different season, which depend on the chemistry of pesticides, environmental factors, temperature, rainfall, and pH, while for  $\beta$ -BHC,  $\gamma$ -chlordane and methoxychlor showed no significance differences

between each two seasons (summer and spring), this may be due to exposure to nearly similar environmental factors.

Our results indicated that, there were significant differences in the mean values of diazinon among samples collected during different seasons, this may be attributed to the frequent application of pesticides in the agricultural sector during summer and autumn, while limited application occurred during spring, also may be due to its washed off by rains besides to different environmental factors. OPs pesticides are strongly adsorbed to soil and not very mobile in water that infiltrates toward groundwater, or run off into surface water, water streams, rivers, lakes and underground aquifers and become contaminated from direct application, drift, run-off from treated areas, or waste materials (Malhat and Nasr, 2011). Our result agreed with Ahmed (1997) who detected diazinon in water samples collected during winter from three farms, Hela et al., (2005) and Bagheri (2007) who detected highest diazinon concentration in water samples collected during summer and with Badach et al., (2007) who detected diazinon in ground water sources during spring and autumn.

High significant differences were recorded in the mean values of deltamethrin among summer, autumn and spring, this may be attributed to repeat application of deltamethrin those seasons. On the other hand, Badach et al., (2007) didn't detect deltamethrin in water samples collected during spring and autumn from different water sources. The results indicated that contamination of drinking water inside dairy farms may be attributed to spraying of animal by deltamethrin and diazinon (applicable pesticides in chosen dairy farms) during certain seasons without any precautions to avoid contamination of drinkers as covering or emptying to drinkers and filling or washing sprayer near them, as well as absence of special area for pesticides application. Our results concluded that seasonal variations

significantly affect the level of pesticide residues pollution of water samples.

Concerning to feed samples, the obtained results in table (2) came in accordance with those Beaton *et al.*, (2010) who found that heavy rains may lead to runoff water carrying pesticide residues into streams, ponds and wells, so the level of pesticides after application in winter can reach to non-detectable level. The high level of pesticide residues in feedstuffs may be due to post-harvest treatment, commercial aerial application and feedstuffs manufactured from plant materials, grass and hay treated during the growing season with insecticides (Fernandez *et al.*, 2008).

The highest concentration of heptachlor was detected in feed samples collected during autumn. These findings were nearly similar those obtained by Prasad and Chhabra (2001) who reported that heptachlor was detected in cereal grains, oil-cakes, bran, legume and non-legume forages and crop used for animals feed.

Results revealed that the highest mean values of diazinon was in feed samples collected during summer followed by autumn and finally during spring that may be attributed to application during growing seasons (summer and spring) of cereals which used as feed for dairy farms or contaminated from spraying in near areas. Also, Morgan and Ashraf (2008) mentioned that small amounts of diazinon applied outdoors can be carried indoor by air, dust, soil and pets creating the potential for exposure.

It is clear that deltamethrin was detected at a high level in feed samples collected during summer followed by autumn then spring, while not detected in winter, which may be attributed to contamination of feed due to animal spray inside dairy farm in summer and autumn to eliminate ectoparasites, while not sprayed in winter. As well as deltamethrin is a rapidly biodegradable pyrethroid. Moreover, Bedi, (2012) mentioned that deltamethrin has

been registered for vast applications for control of insect on cotton, rice, wheat, vegetables, fruits, grain storage and for public health programs in mosquito control to prevent transmission of vector borne diseases.

It is clear from Table (2) that the statistical analysis showed significant differences among summer, autumn and spring for the mean values of p.p.DDD, p.p.DDE, p.p.DDT, aldrin, heptachlor, lindane,  $\gamma$ .chlordane, methoxychlor, diazinon and deltamethrin, while they were not detected in winter, this may be attributed to exposure to different environmental factors as temperature, humidity, rain, soil pH, different types of crops which grow in different seasons and need different treatment of pesticides.

Our results indicated that contamination of feedstuff inside dairy farms may be attributed to spraying of animal by deltamethrin and diazinon (applicable pesticides in chosen dairy farms) during certain seasons without taking any precautions to avoid contamination of feed troughs and storage rooms as covering or emptying of feeders.

Our results concluded that seasonal variations have great effect on the level of pesticide residues in feed samples.

Concerning to milk samples, the results illustrated in table (3) clarified that the highest mean concentration of p.p.DDD, p.p. DDE, p.p.DDT, heptachlor, heptachlor epoxide,  $\gamma$ .chlordane and methoxychlor were detected during summer, while the lowest were detected in spring. Furthermore none of them detected in winter, this may be due to the lipophilic nature of these insecticides which make it descend in milk reflecting body contamination through feed, water and application in certain season (summer, autumn and may be spring), so collected milk samples in this season are being found always contaminated. The current findings of agree, quite well, with



those recorded by Enb (1987), John (2001) and Malhat and Hagag (2012).

The findings about aldrin and lindane were nearly similar those obtained by Enb (1987), Doghiem et al., (1990) and John (2001) reported that aldrin was detected in milk samples collected from Jaipur city, Rajasthan, India during summer, autumn and winter.

The findings about  $\alpha$ -BHC and  $\beta$ -BHC agreed, quite well, with those recorded by Enb (1987) and Doghiem et al., (1990). On the other hand, Malhat and Hagag (2012) reported that  $\alpha$ -HCH not detected in any of cow's milk samples collected from five districts of El-Qalubiya Governorate, Egypt during spring.

Concerning the milk samples, the highest mean value of diazinon was detected during summer followed by spring then autumn and finally winter. The current results agreed to some extent, with Malhat and Hagag (2012) who found that diazinon at the range of 0.005-0.586 mg/kg in 100 milk samples collected from five districts located in Qalubiya Governorate during spring at limit of detection 0.001mg/kg.

The data demonstrated in Table (3) showed that the highest mean value of deltamethrin was detected in milk samples during summer followed by autumn then spring, while not detected in winter. The current results disagreed, to some extent, with those reported by Nasr et al., (2007) who failed to detect deltamethrin in fresh milk collected from different locations at Gharbia Governorate in analyzed samples during all seasons.

The statistical analysis showed significant differences among all seasons for mean values of p.p.DDD, p.p.DDE, p.p.DDT, aldrin, heptachlor, heptachlor epoxide, lindane,  $\gamma$ .chlordane, diazinon and deltamethrin, this variation in results may be attributed to illegal application of some organochlorine till now in some agriculture areas, while diazinon and deltamethrin may be attributed to the frequency of spraying

each farm according to data that have been collected and documented for each farm before. Moreover,  $\alpha$ -BHC,  $\beta$ -BHC and methoxychlor showed no significant differences.

Our results concluded that the levels of pesticide residues especially diazinon and deltamethrin in feed, water and milk were greatly affected by season.

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