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ORGANOCHLORINE RESIDUES IN BUFFALOE AND CATTLE TISSUES IN ASSIUT GOVERNORATE

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ABSTRACT :

Pesticide residues were studied in 164 tissue samples (120 fat, 20 liver and 24 muscle samples) collected from Assiut Governorate during March-April 1992 by GC-ECD.

Liver and muscle samples were found to contain negligible amounts of organochlorine pesticides that never exceeded Extraneous Residue Limit (ERL) ERL's, while fat samples contained relatively higher amounts that rarely exceeded the permissible limits.

DDT derivatives (p,p'-DDT, o,p'-DDT, p,p'-DDD and p,p'-DDE), total HCH isomers (α-, β-, γ- and δ-isomers) and hexachlorobenzene (HCB) were the most frequent pesticides, followed by aldrin and dieldrin, endrin and heptachlor and heptachlor epoxide. Only one buffaloe fat sample (1.3%) exceeded endrin released by the Codex Committee on Pesticide Residue (CCPR) of FAO/WHO in 1990.

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INTRODUCTION :

There is a growing awareness and alarm regarding the possible hazards and economic losses to ever increasing number of toxic substances that enter the food chain. Many of the contaminants enter food directly or indirectly as a result of human activities.

Pesticides occupy a rather unique position among the many chemicals that man encounters daily, in that they are deliberately added to the environment for many purposes. Over the years, vast quantities of chlorinated pesticides have been used in Egypt for crop protection and control of disease transmitting insects.

Public concern about the hazards of health and the environment from the use of agricultural chemicals had been increased dramatically in recent years. Much of the public concern is focused on the residues of the organochlorine compounds. Some of the most prominent pesticides of this group includes DDT, aldrin, dieldrin, lindane (gamma-HCH) and heptachlor. Such chemicals are characterized by their great tendency to accumulate and persist in animal tissues as they are all fat soluble pesticides. They reach man through the food chain and accumulate in various organs, but mainly in fatty tissues. However, pesticides from other groups, such as organophosphates and carbamates that are not free of problems associated with their use, do not pose serious long term residue problems (Fries, 1970).

In Egypt, although the use of chlorinated hydrocarbon pesticides had been curtailed since the early 1970s, high amounts of these chemicals were found in meat, milk and fish samples collected from some Egyptian Governorates (Saleh, 1986; El-Shafei, 1988; Dogheim et al., 1988 and 1990 and salem et al., 1995).

The present work reports the levels of organochlorine pesticide residues in buffaloe and cattle tissues from Assiut Governorate, Egypt.

MATERIALS AND METHODS :

A total number of 120 perinephric fat samples (80 buffaloes and 40 cattle), 20 liver samples (10 of each) and 24 muscle samples (14 of buffaloes and 10 of cattle) from buffaloe and cattle carcasses generally consumed by man were collected randomly during March-April 1992 from the slaughter houses in Assiut Governorate. The samples were transferred into the laboratory, minced and frozen until analysis.

Extraction of tissue samples:

Liver and muscle samples were extracted according to Food and Drug Administration, PAM1 (1988).

Adipose tissue samples were extracted according to the method established by the Federal Institute for the Control of Infectious Diseases in Livestock in Austria (Anonymous,

1988) and related to Stijve and Cardinale, (1974).

Clean-up was carried out according to the method that followed in fat samples (Anonymous, 1988).

Pesticide reference standards:

Alpha-HCH (Supelco Nr. 4-8493), Beta-HCH (Supelco Nr. 4-8494), Gamma-HCH (Supelco Nr. 4-9049) and delta-HCH (Supelco Nr. 4-8495), p,p'-DDE (Supelco Nr. 4-9017), p,p'-DDD (Supelco Nr. 4-9009), o,p'-DDT (Ehrenstorfer P1111), p,p'-DDT (Supelco Nr. 4-9019), Heptachlor and heptachlor epoxide (Supelco Nr. 4-9041 and 4-9042), Aldrin and dieldrin (Supelco Nr. 4-9000 and 4-9024), Hexachlorobenzene (Supelco Nr. 4-8508) and endrin (Supelco Nr. 4-9032). Standard solution of reference materials were prepared in petroleum ether.

Gas chromatograph- Carlo Erba MEGA HRGC 5330 with ^{63}Ni ECD and split-splitless injector and column, HP ULTRA1 50m X 0.2mmX 0.33um was used, according to the method of Suzuki, et al (1979).

RESULTS AND DISCUSSION :

The results of organochlorine pesticide residues are presented in table 1 which show that liver, muscle and fat samples of buffaloes and cattle were found to contain DDT and its metabolites, HCH isomers, heptachlor , heptachlor epoxide, aldrin and dieldrin, HCB and endrin residues with different frequencies and levels (table 1)

According to the previous studies carried out in many countries, i.e., Egypt (Saleh, 1986; El-Shafei, 1988 and Dogheim et al., 1988, 1990 and 1991), Denmark (Bro-Rasmussen, 1968); France (De Lavaur and Hascoet, 1974); Italy (Pastore and Vecchia, 1974), the pesticides most commonly found in the various animal foodstuffs are lindane, the isomers of BHC (HCH), DDT and its metabolites (DDE and DDD), dieldrin and heptachlor epoxide. HCB and HCH gamma and alpha isomers were the most frequently detected pesticides in Germany (Knoeppler, 1976) and in Austria (Jarc, 1980).

DDT derivatives o,p´ and p,p´, DDE and DDD contributing the DDT complex were detected in all tissue samples of liver, muscle and adipose tissues with 100% frequency. Highest amounts of the DDT complex were determined in adipose tissue samples. This could be attributed to the high solubility and tendency of DDT and its metabolites to accumulate and store in fatty tissues (WHO, 1979 and 1989).

Residual amounts detected in buffaloe tissue samples were almost higher than that in cattle.

DDE constituted more than 80% of the DDT complex residues in both animal tissues, but p,p´-DDT was represented by 8% (table 2) . DDE was the most frequently presented

TABLE (1) : Mean, range values (ppm)[a] and percentage's frequency (%F) of organochlorine pesticide residues detected in buffaloe and cattle tissue samples collected from Assiut Governorate in 1992.

		Buffaloes			Cattle		
Pesticides	Values	Liver	Muscle	Fat	Liver	Muscle	Fat
DDT complex	Mean	0.041	0.023	0.213	0.023	0.018	0.108
	Range	0.011-0.080	0.010-0.053	0.025-0.869	0.015-0.029	0.004-0.051	0.009-0.758
	Frequency	100	100	100	100	100	100
Total HCH isomers	Mean	0.032	0.010	0.030	0.024	0.012	0.017
	Range	0.002-0.083	0.004-0.022	0.002-0.138	0.001-0.032	0.001-0.019	0.001-0.048
	Frequency	100	100	96.25	100	100	87.5
γ-HCH (Lindane)	Mean	0.002	0.002	0.004	0.001	0.001	0.002
	Range	0.001-0.005	0.001-0.005	0.001-0.039	0.0004-0.003	0.001-0.002	0.001-0.005
	Frequency	100	85.7	30	90	60	22.5
Heptachlor & Hep. epoxide	Mean	0.006	0.009	0.011	0.001	0.0015	0.008
	Range	0.0003-0.009	0.009-0.009	0.0003-0.061	0.001-0.001	0.001-0.002	0.008-0.008
	Frequency	50	7.1	12.5	20	20	2.5
Aldrin & dieldrin	Mean	0.003	0.001	0.004	0.0007	0.002	0.003
	Range	0.001-0.009	0.001-0.002	0.001-0.009	0.0006-0.001	0.001-0.003	0.0003-0.008
	Frequency	40	21.4	35	40	20	15
Endrin	Mean	0.008	0.001	0.013	0.001	0.002	0.006
	Range	0.001-0.016	0.0002-0.001	0.001-0.121	0.001-0.002	0.0007-0.003	0.002-0.011
	Frequency	50	50	30	40	50	12.5
HCB	Mean	0.001	0.0006	0.004	0.001	0.001	0.004
	Range	0.0004-0.004	0.0002-0.001	0.0003-0.017	0.0006-0.002	0.0006-0.003	0.0002-0.022
	Frequency	100	71.4	98.75	100	70	92.5

a = on fat basis

derivative (100%) with the highest amounts, more than 80% of the DDT complex in all tissue samples indicating the continuous degradation of DDT to the less toxic and more persistent derivative as reported or studied by Fries et al., (1972) and Hayes, (1975); who reported that DDE is more resistant to metabolic degradation than DDT in animals and man. Also, DDE is found in almost all the living organisms because of its strong affinity with body fat (Jensen and Jasson, 1976).

In spite of the known information about the prohibition use of DDT in Egypt along the last fifteen years according to the authorities report, the continuous use of the acaricide dicofol (Kelthane®) replaced DDT as the primary source of environmental DDE and contains as much as 0.6 % p,p´ and o,p´-DDT (Camoni et al., 1983); this indicates the continuous contamination of the environment by DDT and DDE.

Edwards, (1973) reported that DDT has an average half life in the soil of about 3 years, and 5-10% of the amounts applied still remains 10 years after the application, while Nash and Woolson, (1967) stated that as much as 40% of DDT were still present 17 years in a Maryland

Table 2. Mean, maximum values and frequency (f) of individual organochlorine pesticide residues detected in buffaloes and cattle fat samples collected from Assiut Governorate.

Pesticides	Animal species					
	Buffaloes			Cattle		
	mean	max.	f	mean	max.	f
p,p'-DDT	0.010	0.037	81.3	0.006	0.013	25
o,p'-DDT	0.017	0.120	8.8	0.021	0.035	5
p,p'-DDE	0.183	0.727	100	0.105	0.758	100
p,p'-DDD	0.020	0.068	93.8	0.006	0.018	27.5
DDT complex	0.213	0.869	100	0.108	0.758	100
α-HCH	0.004	0.026	50	0.004	0.026	72.5
β-HCH	0.028	0.126	96.3	0.015	0.048	77.5
γ-HCH	0.004	0.039	30	0.002	0.005	22.5
δ-HCH	0.002	0.003	10	ND	ND	0
HCH isomers	0.030	0.138	96.3	0.017	0.048	87.5
Heptachlor	ND	ND	0	0.004	0.004	2.5
Hept. epoxide	0.011	0.061	12.5	0.004	0.004	2.5
Total heptachlors	0.011	0.061	12.5	0.008	0.008	2.5
Aldrin	ND	ND	0	ND	ND	0
Dieldrin	0.004	0.009	35	0.003	0.008	15
Aldrin & Dieldrin	0.004	0.009	35	0.003	0.008	15
Endrin	0.013	0.121	30	0.006	0.011	12.5
HCB	0.004	0.017	98.8	0.004	0.022	92.5

ND = not detected.

soil after the application; which means a permanent source of pollution with DDT. On the basis of ERL of 5 mg/kg total DDT complex residues in animal tissues, all the analyzed samples were prooved to he below the limit.

Total hexachlorocyclohexane isomers (alpha, beta, gamma and delta) were detected at the same frequency as DDT in liver and muscle samples and with less frequency in fat samples. Detected residues of total HCH isomers were almost high in buffaloe than in cattle tissues especially in adipose tissue and liver samples. This may be attributed to the body fat condition, which was noticed during sample's collection and almost seemed that cattle contain more fat in its carcasses. Spence et al., (1990) recorded that the reduction in BHC residue levels occurred by redistribution and dilution throughout the increased body fat.

α- and β-HCH were the most frequent isomers detected in all tissues of both animals, but β-HCH constituted more than 80% of the total HCH residues in all the analyzed samples (table 2). All the investigated tissues were below the ERL's (2 ppm) for total HCH isomers and lindane.

Beta isomer was the most pronounced HCH isomer detected in all tissue samples of buffaloes and cattle, which agreed with Posgay

et al., (1980) and constituted more than 80% of the total HCH residues in all tissues. This isomer is the most persistent and slowly eliminated from the body (Pfeilsticker, 1973) and has the ability to accumulate in fat tissues 10-30 times than lindane (Heeschen, 1980), but it is not the most hazardous one (Scholz et al., 1985). Its predominant occurrence in the tissue samples indicate the continuous degradation of the more toxic alpha and gamma isomers (Dogheim et al., 1988).

Lindane (gamma isomer of HCH) which is used alone as a pesticide and also is the most toxic isomer of this group was always detected in insignificant amounts. Almost no differences in the residual amounts of lindane were observed between the different tissues of the two animal sorts.

α-HCH was also more frequent in tissue samples as β-HCH but with very low values. This may be attributed to the possible use of technical HCH that contained a large proportion of α-HCH or the external treatment of animals with HCH based veterinary preparations, which could be ingested or absorbed through skin (Harper, 1980). Gamma-HCH transforms to the alpha and delta isomers, which are of 4 and 50 times less insecticidal activity, respectively (Newland et al., 1969).

Taking into consideration that the increase in HCH total isomers was mainly due to the beta isomer which is only a minor component in the technical HCH pesticide (Pfeilsticker, 1973). It might be concluded that these exceeding amounts could be attributed to the degradation process of the other isomers giving elevated amounts to the most persistent and relatively least hazardous beta isomer. Jensen, (1983) stated that a- and g-HCH isomers may isomerize into the b-isomer in the living organisms .

A sum of heptachlor and heptachlor epoxide defined as total heptachlors were detected in liver, muscle and adipose tissue samples at low frequencies and levels but they could not be detected (table 2). Higher residual amounts of heptachlors were almost detected in buffaloe tissue samples. Adipose tissue samples expressed the highest levels of total heptachlors' residues in contrast to other tissue samples due to the storage of these compounds in the animal fat (Rusoff et al., 1963 and Bruce et al., 1965).

Wilkinson et al., (1964) indicated that toxic residues mostly heptachlor epoxide, will persist in the soil for as long as 9 years, the possibility thus exists for contamination of crops from these fields. However, it was translocated from the soil into certain crops (Bruce and Decker, 1966), subsequently stored in the fat of dairy cows (Rusoff et al., 1963).

Referring to the ERL´s of the (CCPR, 1990) presence of 0.2 mg/kg of heptachlor and its epoxide in animal tissue, it could be revealed that all detected residues in buffaloes and cattle tissues were below the permissible limit.

Aldrin residue definition includes dieldrin residues as well. Inspite of dieldrin being an insecticide by itself it is also resulting as a degradation product from aldrin application. A statement that was mentioned by Bann et al., (1956) that dieldrin is the form of which aldrin usually being stored in fats.

The magnitude of aldrin and dieldrin residues revealed low levels and few frequencies in liver, muscle and adipose tissue samples of both animals.

Aldrin residues were absent almost from all samples as the total residues resulted mainly from dieldrin (table 2). All analyzed buffaloes and cattle tissue samples contained aldrin and dieldrin residues below the extraneous residue limit.

Concerning solubility and tendency to accumulate in fatty tissues (Kiigemgi et al., 1958 and Humphrays, 1988), endrin was detected at higher amounts in adipose tissues of buffaloes and cattle but still at low frequencies.

As far as human safety is concerned, endrin is the most hazardous pesticide from the cyclodiene group. The extraneous residue limit of endrin in animal tissues is 0.1 mg/kg (CCPR, 1990). All liver and muscle samples from buffaloes and cattle were safe for human consumption where endrin residues were absent or below the permissible limits. Only one fat sample from (1.3%) buffaloe out of 80 exceeded the ERL. Cattle adipose tissue never exceeded the limit.

The fungicide, industrial product and by-product HCB, has since the early 1970s been recognized as an environmental contaminant comparable to DDT and PCBs in industrialized countries (Acker and Schulte, 1970 and Stijve, 1971). On the other hand, HCB contamination does not seem to be a serious problem in Egypt, since only very low residue levels were found which never exceeded the permissible limits. The increase in its frequency in this study may indicate a recent exposure of our environment .

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متبقيات المبيدات العضوية الكلورينية في أنسجة الجاموس والأبقار في محافظة أسيوط

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*قسم الطب الشرعى والسموم – كلية الطب البيطرى ـ جامعة أسيوط ** الهيئة القومية للسيطرة على الأمراض المعدية – وزارة الصحة – مودلنج – النمسا ، *** المعمل المركزى للمبيدات – معهد البحوث الزراعية – وزارة الزراعة – الدقى – القاهرة

إن تزايد المخاطر البيئية من جراء استخدام المبيدات الحشرية فى الأغراض الزراعية وفي الطب البيطرى أدى إلى تلوث العديد من المزروعات والأغذية الحيوانية مما ساعد على زيادة هذه المركبات فى أنسجة الحيوانات ومنتجاتها . وهذا دفعنا إلى إجراء هذه الدراسة لاستبيان مستوى تلوث أنسجة الجاموس والأبقار فى مدينة أسيوط ببعض المبيدات الحشرية العضوية الكلورينية .

وقد تم فى هذه الدراسة قياس مستويات مركبات ال د.د.ت.ومركبات سادس كلوريد الهكسان الحلقية بالإضافة إلى مركبات الهبتاكلور والهبتاكلورأبوكسيد ومركبات الالدرين والديلدرين والاندرين ومركبات سادس كلوريد البنزين وذلك فى أنسجة عدد ١٦٤ عينة . وقد استخدم جهاز الفصل الغازى المزود بكاشف إليكترونى (GC-ECD) لهذا الغرض .

أوضحت النتائج وجود كميات قليلة جداً من المركبات الكلورينية فى كبد وعضلات العينات المفحوصة والتى لاتزيد عن الكميات المسموح بها دولياً بواسطة المنظمات العالمية (ERL) بينما أظهرت عينات الدهن وجود كميات عالية نسبياً ولكن نادراً ما تكون أعلى من الكميات المسموح بها عالمياً .

كما أظهرت النتائج أن مشتقات ال د.د.ت.وسادس كلوريد الهكسان الحلقية مجتمعة وسادس كلوريد البنزين كانت أعلى ظهوراً يليها مركبات الالدرين والديلدرين والاندرين والهبتاكلور أبوكسيد كما أظهرت عينة واحدة فقط من دهن الجاموس (١,٣٪) زيادة فى كمية الاندرين عن الكميات المسموحة بواسطة منظمة الصحة العالمية ومنظمة الأغذية والزراعة عام ١٩٩٠ ومن ثم فإن الفحص الدورى والمستمر لتواجد المبيدات الحشرية فى الأغذية والمنتجات الحيوانية يعد من الامور الهامة التى يجب أن تؤخذ فى الاعتبار لتدارك أخطارها.