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SOME CHEMICAL CONTAMINANTS OF PUBLIC HEALTH HAZARD IN DRIED EGG

(With 2 Tables)

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(Received at 31/10/1999)

بعض الملوثات الكيميائية ذات الخطورة على الصحة العامة في البيض المجفف

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تم جمع أربعون (٤٠) عينة من صفار البيض المجفف و عشر عينات من بياض البيض المجفف من أماكن مختلفة بمحافظة الإسكندرية و قد تم تجميع هذه العينات خلال ستة أشهر و ذلك بهدف بيان مدى تواجد متبقيات الملوثات الكيميائية التي إشتملت على المركبات الهيدروكلورينية و الفسفورية العضوية و كذلك المعادن الثقيلة و متبقيات المضادات الحيوية و قد تم تحليل هذه العينات باستخدام جهاز كروماتوجرافيا الغاز لبيان مدى تواجد المركبات الهيدروكلورونية و المركبات الفسفورية العضوية و قد أظهرت النتائج تواجد المركبات الهيدروكلورونية في صفار البيض بنسب ١٧,٥ و ٢٢,٥ و ١٠ و ١٢,٥ و ١٠% لكل من ال د.د.ت. المركب و مركبات الهكساميكلوهكسان و الهبتاكلور و الهبتاكلور ايبوكسيد و الديلدرين و الإندرين على التوالي و قد كانت متوسطاتها كالتالي ٥,١٦ ± ٠,٠٣ ٠,٤٣ ± ٠,٠٣ ٠,٦٥ ± ٠,٠٢ ١,١٦ ± ٠,٠١ ٠,٧٥ ± ٠,٠١ ٠,٤٨ ± ٠,٠٣ جزء في المليون على التوالي. و قد إتضح أيضاً من الدراسة خلو عينات بياض البيض المجفف من التلوث بالمركبات الهيدروكلورونية بالإضافة إلى ذلك فقد أشارت النتائج إلى عدم تلوث جميع عينات البيض المجفف التي تم فحصها بمركبات المبيدات الفسفورية العضوية و قد قيمت النتائج طبقاً للمعايير الدولية التي أقرتها منظمة الصحة العالمي و الأغذية و الزراعة و قد إتضح من النتائج أن كل العينات التي إحتوت على المركبات الهيدروكلورونية كانت أقل من الحد المسموح به. و قد أجرى هذا البحث أيضاً لتحديد و قياس بقايا بعض المعادن الثقيلة و هي الزئبق و الكاديوم و الرصاص في عينات البيض المجفف و ذلك باستخدام جهاز الإمتصاص الذرى الطيفي و قد أظهر تحليل هذه العينات تواجد الزئبق و الكاديوم و الرصاص في صفار البيض بمتوسطات قدرها ٠,٠٦١ ± ٠,٠١٧ ٠,٠٤٩ ± ٠,٠٣١ ٠,٠٧١ ± ٠,٠١٣ جزء في المليون على التوالي. أما بياض البيض فقد إحتوى فقط على الرصاص بمتوسط

قدره 0.0089 ± 0.0029 جزء في المليون. وقد أشارت النتائج إلى تواجد هذه العناصر تحت الحد المسموح به كما إتضح أيضاً من النتائج عدم تواجد بقايا المضادات الحيوية و قد نوقت الأهمية الصحية لمتقيات هذه الملوثات الكيميائية كل على حدة.

SUMMARY

Forty samples of dried egg yolk and ten samples of dried egg albumen were collected from different localities in Alexandria Governorate. The samples were analysed for determination of some organochlorine and organophosphorus insecticides residues and heavy metals as well as detection of residual levels of antibiotics. Organochlorine and organophosphorus insecticide residues were determined using gas chromatography with electron capture and flame photometric detectors. The obtained results revealed the presence of organochlorine insecticides residues in analysed dried egg yolk samples at percentages of 17.5, 22.5, 12.5, 10, 12.5 and 10 for total DDT, total Hexachlorocyclo hexane, Heptachlor, Heptachlorepoide, Dieldrin and Endrin, with mean values of 5.16 ± 0.03 , 0.43 ± 0.03 , 0.65 ± 0.02 , 1.16 ± 0.01 , 0.75 ± 0.01 and 0.48 ± 0.03 ppb, respectively. On the other hand, non of organochlorine insecticide residues could be detected in dried egg albumen. Moreover, all analysed dried egg samples were free from detectable amounts of organophosphorus insecticide residues. The results were evaluated according to International Standards of FAO/WHO. The obtained data indicated that all analyzed dried egg yolk samples contain organochlorine insecticide residues below the permissible limit. The samples were analysed for determination of cadmium, lead and mercury by using Atomic Absorption Spectrophotometer. The results indicated these metals were present in dried egg yolk samples at percentages of 25, 27.5 and 100 for mercury, cadmium and lead with mean values of 0.061 ± 0.017 , 0.049 ± 0.031 and 0.071 ± 0.013 ppm, respectively. While lead was the only detectable metal in dried egg albumen samples at a percentage of 100 with a mean value of 0.089 ± 0.029 ppm. The results were evaluated according to International Standards. The obtained results indicated the presence of metal contaminants in dried egg samples with values below the recommended limits. The results of antibiotic residues indicated that all examined samples did not contain any inhibitory substances. The public health importance of existed toxic chemical residues was discussed.

Key words: Dried egg.

INTRODUCTION

Egg products which include liquid, dried or frozen whole eggs, yolk and/or white are more likely to be polluted with chemical contaminants of public health hazard including pesticides, heavy metals and antibiotic residues (Hubbert *et al.*, 1996).

Pesticides have increased steadily to the extent that the annual production of synthetic pesticide chemicals has exceeded one billion kilograms. This may be largely contributed to prevent insect from making undue inroads into man's food supply and to break the vector chain for diseases carried by insects to man and domestic animals (Green *et al.*, 1987 and Maybury, 1989). However, despite this high productivity, public awareness is currently focused on the wide spread use of pesticides in the environment and their relationship to environmental health (Carson, 1990).

The problem of pesticide residues in food has been considered the main side effect of environmental pollution by pesticides leading to injury of non-target organisms concerns the health of the workers and consumers, as renal failure and hepatic cancer.

Another form of chemical pollutants are heavy metals which have recently come to forefront dangerous substances causing serious health hazards in human. Mercury poisoning results in neurological damage, loss of vision and paralysis. It also passes human placenta causing chromosomal disorders and teratogenicity (Sorensen, 1991). Cadmium is a highly cumulative poison with a biologic half-life of about 20-30 years in human (Manahan, 1992). Kidney is the most sensitive organ to chronic cadmium exposure. It produces nephrotoxicity, proteinuria, osteomalacia and pathological fracture (Friberg and Elinder, 1985).

Lead poisoning leads to anaemia, liver dysfunction, muscle pain, nephropathy and neuropathy of both central and peripheral nervous system (Gold Frank *et al.*, 1990). Its biological half-life in bone is about 27 years (Shibamoto and Bjeldanes, 1993).

Antibiotic therapy has been also widely employed in treatment of diseases in poultry farms. In some instances, the antibiotics are incorporated into foods as a dietary supplement and used as preservatives. Consequently, whatever the mode of antibiotic administration, antibiotic residues may be found in eggs produced by hens. The presence of antibiotics in food represents a potential hazard to consumers due to their allergic properties.

Dried eggs provide a unique well-balanced source of nutrients for human and are used in a wide variety of foods specially all types of bakery products (Bennion, 1980).

There is a lack of information about the degree of dried egg contamination with some chemical pollutants in Egypt. So, the present investigation aimed to throw a light on the limit of their residues in dried eggs sold in Alexandria Governorate.

MATERIAL and METHODS

Sampling. Forty dried egg yolk and ten dried egg albumen samples were collected over six months from different localities in Alexandria Governorate. The samples were packed in polyethylene bags and kept dry till the analysis was carried out.

I. Determination of pesticide residues:

Gas chromatography. Column (9 ft x 1/4 I.D.) packed with 1.5% OV-17/1.95, OV-210 on Gas Chromatography-Q-80-100 mesh for qualitative and quantitative determination of organochlorine residues and column (9 ft x 1/4 I.D.) packed with 10% DC-200 on gas chromatography 80-100 mesh for confirmation of organochlorine residues while column (7 ft x 1.8 I.D.) packed with 4% SE-30/6% OV-210 on gas chromatography 80-100 mesh for qualitative determination of organophosphorus pesticide residues.

Acetonitrile, petroleum ether, ethyl acetate, N-hexane and all solvents were distilled from all-glass apparatus and subjected to general purity tests according to FDA Manual (1992).

Pesticide reference standard solutions were used in determining the retention times of each of the investigated pesticides.

Chlorinated hydrocarbon pesticides used were: 1) D.D.T complex. 2) HCH (Hexachlorocyclo-hexane), isomers (α , and γ). 3) Heptachlor and Heptachlorepoide. 4) Aldrin, Dieldrin and Endrin.

Organophosphorus pesticides used were: 1) Diazinon. 2) Malathion. 3) Dimethoate. 4) Dursban.

Preparation, extraction and clean up procedure: The method applied was according to the steps described by FDA Manual (1992).

The residues of organochlorine and organophosphorus pesticides were determined by using G.C.PYE Unicam Gas Chromatography, with 63 NI electron capture detector, with flame photometric detector.

II. Determination of heavy metal residues:

Analytical procedures for metal concentrations:

Samples examined for cadmium, lead and mercury analysis were ashed according to AOAC (1980). The trace metals content of ashed residues were measured using Atomic Absorption Spectrophotometry (Perkin Elmer, model 2380, USA) with alteration of standard burner head of A.A.S. in relation to the light beam of examined metals.

III. Detection of antibiotic residues:

Detection of antibiotic residues in examined samples were carried out using agar gel diffusion test described by Bogaerts and Wolf (1980) and a well technique according to Coretti (1961).

RESULTS and DISCUSSION

Environmental pollution represents a major problem in the world especially in developing countries. Egypt suffers from biosphere pollution (air/soil/water). Pollutants in water including heavy metals and pesticides that come from agricultural and industrial wastes may accumulate in food chains causing serious health problems to consumers.

I. Pesticide residues:

The obtained results in this investigation revealed the occurrence of organochlorine insecticide residues in dried egg yolk samples (Table 1).

1. Dichloro-Diphenyl Trichloroethane (DDT):

DDT and its derivatives were present in dried egg yolk samples at percentages of 10, 15, 15 and 7.5 with mean values of 0.99 ± 0.05 , 2.21 ± 0.02 , 2.1 ± 0.02 and 0.24 ± 0.01 for DDT-O'P, DDT-P'P, DDE-P'P and DDD-P'P, respectively. The frequent detection of DDT-P'P and DDE-P'P gave an indication for the degradation of DDT to its metabolites.

2. Total Hexachlorocyclohexane (, β and γ HCH):

Residues of alpha HCH were detected in 15% of examined dried egg yolk samples. Its concentration varied from 0.11 to 0.32 with a mean value of 0.19 ± 0.02 , while beta HCH was detected in 20% of dried yolk samples with a mean value of 0.33 ± 0.02 and its residues ranged from 0.13 to 0.65 ppb. On the other hand, gamma HCH failed to be detected in all examined samples. It can be noticed that β isomer was the highest detectable insecticide in dried egg yolk samples and this

may be attributed to its strong persistency and stability than other isomers (Dogheim *et al.*, 1989).

3. Heptachlor and Heptachlorepoide:

Heptachlor residues were detected in 12.5% of dried egg yolk samples with values ranged from 0.19-1.1 ppb with an average of 0.65 ± 0.02 , while Heptachlorepoide residues were present at a percentage of 10 with values varied from 0.29 - 2.02 ppb with a mean value of 1.16 ± 0.01 ppb. It was noticed that Heptachlorepoide is more detectable than Heptachlor isomers. This may be attributed to its strong persistency and stability than other isomers (Venant *et al.*, 1991).

4. Aldrin, Dieldrin and Endrin:

Aldrin residues were completely absent in all examined samples while Dieldrin residues were detected in 12.5% of examined dried egg yolk samples with values ranged from 0.31-1.12 ppb with an average of 0.75 ± 0.01 ppb.

The absence of Aldrin in all examined samples may be explained on the basis of its continuous degradation into Dieldrin within the living tissues through the different metabolic processes (Dogheim *et al.*, 1989).

Endrin residues were detected in dried egg yolk samples at a percentage of 10 with a mean value of 0.48 ± 0.03 and its quantities varied from 0.23 - 0.82 ppb.

The presence of organochlorine insecticide residues in dried egg yolk samples may be due to their presence in eggs (Frank *et al.*, 1985 and Abou-Zeid, 1997) which can be derived from either topical treatment of birds and spraying of poultry houses. However, the main source of contamination is likely to be chicken feed. Other sources may include water, soil and air (WHO, 1982).

In addition to the long persistence of organochlorine pesticides and their chemical stability in the soil, water and air lead to the contamination of crops and ultimate accumulation in animal derived foods (UNEP, FAO, WHO\GEMS, 1981; 1988 and 1992).

The obtained results in Table (1) proved that none of the determined insecticide residues in examined dried egg yolk samples exceeded the permissible limits permitted by FAO/WHO (1992) which are 100, 100, 50, 200 and 100 ppb for Lindane, Aldrin and Dieldrin, Heptachlorepoide, Endrin and total DDT, respectively. This may be attributed, in a part, to their presence in egg under the permissible limits (Kwon *et al.*, 1992 and El-Mekkawi *et al.*, 1994) and in another part due to continuous degradation of organochlorine pesticides in the

environment and the successful trials to control the misuse and application of pesticides.

Although the determined levels of residues in all examined dried egg yolk samples were below the permissible limit established by FAO/WHO (1992), the dangerous effect of such residues arises from their cumulative effects which appears later leading to hazards in human beings as carcinogenesis, renal failure, liver cirrhosis and optic nerve manifestation are of special concern (Dixon, 1980 and Amr, 1992).

The recorded results in this study show the contamination of dried egg yolk samples by organochlorine insecticide residues while it could not be detectable in dried egg albumen. This may be due to organochlorine pesticides are lipophilic in nature, so they are easily partitioned into the fat portion of eggs. Moreover, the residues of organochlorines are not removed easily even during thermal treatment or other processing due to its high stability and persistency (Harper, 1980 and Perez *et al.*, 1982).

The obtained results show the absence of organophosphorus insecticide residues in all examined dried egg samples which may be attributed to the fact that they are water soluble, showed continuous degradation within the living tissue through the metabolic pathway and easily excreted from the body few days after consumption, so it could not be detected in dried egg samples.

II. Heavy metal residues:

It is clear from Table (2) that the heavy metal residues were detected in the examined dried egg yolk samples at percentages of 25, 27.5 and 100 for mercury, cadmium and lead, respectively.

The concentrations of mercury ranged from 0.041 - 0.081 ppm with a mean value of 0.061 ± 0.017 ppm and cadmium varied from 0.045 - 0.065 ppm with a mean value of 0.049 ± 0.031 ppm. Also the concentrations of lead varied from 0.061 to 0.083 ppm with an average value of 0.071 ± 0.013 ppm.

In contrast, lead was the only detectable heavy metal in dried egg albumen samples at a percentage of 100 and its concentrations ranged from 0.071 - 0.093 ppm with a mean value of 0.089 ± 0.029 ppm.

The presence of heavy metal residues in dried egg samples may be attributed to their presence in eggs which derived from hens which ingest contaminated water, feed of plant or animal origin (Leonzio and Massai, 1989; El-Hoshy and Ashoub, 1998).

A permissible limit of 2 ppm was established for lead in food (Pearson, 1976). Also, the German guideline of cadmium in animal tissues is 0.5 ppm (Kluge-Berge *et al.*, 1992). Moreover, the Food and Drug Administration (FDA, 1992) established 0.5 ppm as a guideline for mercury in food (Gomez and Markakis, 1974).

Accordingly, the levels of mercury, cadmium and lead in examined dried egg samples are below these limits. Although the determined levels of mercury, cadmium and lead residues in all examined dried egg samples were below the permissible limits. The dangerous effects of such residues arise from their cumulative effect (Miller, 1971 and Timbrell, 1982).

III. Antibiotic residues:

Our results revealed absence of inhibitory substances in all dried egg samples.

In conclusion, the presence of organochlorine and heavy metal residues at low levels and low percentages below the permissible limits and absence of both organophosphorus and antibiotic residues in dried egg samples may be attributed to strict hygienic measures in consideration regarding the raw material used in manufacture of such products particularly eggs. The cumulative properties of such residues may constitute a public health hazard.

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Table 1: Analytical results of organochlorine insecticide residues (ppb) in dried egg yolk samples.

Pesticide	Min.	Max.	Mean	S.E.	Positive samples		Permissible Limit
					No.	%	
1. DDT complex							
- OP-DDT	0.39	1.31	0.99	0.05	4	10	100 ppb
- PP-DDT	1.16	3.92	2.21	0.02	6	15	
- PP-DDE	1.27	4.95	2.10	0.02	6	15	
- PP-DDD	0.21	0.28	0.24	0.01	3	7.5	
- Total DDT	2.13	7.89	5.16	0.03	7	17.5	
2. Hexachlorocyclohexane:							
- HCH	0.11	0.32	0.19	0.02	6	15	100 ppb
- JCH	0.13	0.65	0.33	0.02	8	20	
- γ-HCH	--	--	--	--	--	--	
- Total HCH (Lindane)	0.23	0.94	0.43	0.03	9	22.5	
3. Heptachlor	0.19	1.10	0.65	0.02	5	12.5	50 ppb
- Heptachlorepoixide	0.29	2.02	1.16	0.01	4	10	
4. Aldrin	--	--	--	--	--	--	
- Dieldren	0.31	1.12	0.75	0.01	5	12.5	100 ppb
- Aldrin & Dieldrin	0.31	1.12	0.75	0.01	5	12.5	
- Endrin	0.23	0.82	0.48	0.03	4	10	

No. of examined dried egg yolk samples = 40.

Table 2: Statistical analytical results of heavy metal residues (ppm) in examined dried eggs.

Type of Sample	Type of Heavy metal	Min.	Max.	Mean	S.E.	Positive samples No.	Permissible Limit %
1. Dried egg yolk	Mercury	0.041	0.081	0.061	0.017	10	25
	Cadmium	0.045	0.065	0.049	0.031	11	27.5
	Lead	0.061	0.083	0.071	0.013	40	100
2. Dried egg albumen	Mercury	--	--	--	--	--	--
	Cadmium	--	--	--	--	--	--
	Lead	0.071	0.093	0.089	0.029	10	100

No. of examined dried egg yolk sample = 40; No. of examined dried egg albumen = 10