# MONITORING OF ORGANOCHLORINE PESTICIDE RESIDUES IN BOTH FRESH AND PACKED MILK

#### BY

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# ABSTRACT

A total of 525 samples were collected from greater Cairo (Cairo, Giza and Qualiobia Governorates) during the autumn season as it follows the summer period during which the maximum use of pesticides takes place for controlling cotton pests.

Samples were extracted and cleaned up and pesticide residues were detected by using GLC apparatus equiped with an electron capture detector.

Results indicated that aldrin residues were detected in 28.57% and 86.67% of packed and bulk milk respectively. Residue levels ranged from 0 to 0.1024 and from 0 to 0.1049 mg/L in both packed and bulk milk.

Dieldrin residues were detected only in packed milk. The residue levels ranged from 0 to 0.0484 mg/L. Only 14.29% of packed milk samples contained dieldrin.

Chlordane was detected in 85.71% and 93.33% of both packed and bulk milk samples respectively. The residue levels ranged from 0 to 0.1155 and 0 to 1.271 mg/L respectively.

Only 14,29% and 26.67% of the packed and bulk milk samples respectively contained  $\Sigma DDT$ . Residues levels ranged from 0 to 11,9950 and 0 to 3,786 mg/L were detected respectively.

Packed milk contained endrin in 14.29% of samples at levels ranged from 0.0 to 0.3644 mg/L, while no residues were detected in bulk milk samples.

Heptachlor residues levels ranged from 0.0308 to 2.1312 and from 0 to 1.0143 mg/L in both packed and bulk milk samples. Hundred percent of packed milk and 73.33% of bulk milk were contaminated with heptachlor residues.

Lindane residues were detected in 85.77% and 80% of both packed and bulk milk respectively. The residues varied between 0.0 to 0.0098 mg/L and 0.0 to 0.0111 mg/L respectively.

# INTRODUCTION

Now measurable amounts of pesticide residues find their way to pollute our food. Organochlorine pesticides were detected in human milk samples in Finland (Russalo. - Rauhaman 1988); in Italy (Dommarco et al., 1987)' in Norway (Skaare et al., 1988)' in Sweden (Noren and Sjovall, 1987)' in Kenya (Kanja et al., 1992)' in Zimbabwe (Chikuni et al., 1991); in Germany (Ehrenstorfer et al., 1991); in Yugoslavia (Krauthacker, 1991); in Egypt (Dogheim et al., 1990) and 1991) and in Canada (Frank et al., 1985).

Data also indicate that cow milk is also polluted by organochlorine pesticide residues as reported before by Jahn et al., 1991, Heeschen and Bluthgen, 1985 and Bluthgen et al., 1984 (in Germany); Salch, 1991 (in USA); Lucisano et al., 1982 (in Italy); Juszkiewicz, 1982 and Juszkiewicz and Niewiadowska, 1984 (in Poland). El-Alfy, 1981 reported that all samples collected from Qualiobia province in Egypt contained lindane and endrin with a range between 0.01 to 0.03 ppm. Abdel-Gawaad and Shams El Din, 1989 and 1990 detected endrin, dieldrin, lindane and DDT in fresh and powder milk.

### MATERIALS AND METHODS

# Milk Samples:

Total of 135 packed ultra - hightreated milk samples and

390 samples of bulk milk comprising of fresh cow and buffalo milk were collected from three governorates (Cairo, Giza and Qualiobia).

The samples were kept in deep-freezer under -18°C until analysis.

#### Standard used:

The following primary standards were obtained from the repository of the U.S. Environment Protection Agency at Research Triangle Park, N.C. These standards included: Aldrin, Dieldrin, BHC, Chlordane, DDE, DDD, DDA, DDT, Endrin, Heptachlor and Lindane.

The working standard solutions of three concentrations were prepared daily 1,2 and 4 nanogram per microliter with pesticide quality hexane solvent.

#### Extraction and Clean up:

The same method used before by Abdel-Fatah et al., 1992 was used for extraction and clean up.

# GC Detection:

Hewlett Packard, Model HP 5890 A programable gas chromatography equipped with electron capture detector was used under the operating conditions, initial temperature 150°C, temperature rate 5°C per minute; final oven

mean in packed milk samples was 0.0521  $\pm$  0.1377 ppm while the range was from 0.0 to 0.3644 ppm.

Heptachlor residues were detected in 100% of packed milk samples. The mean of detected residues were  $0.565\pm0.3553$  ppm in packed and bulk milk samples respectively. The maximum range of residues was 2.1312 ppm in packed milk while in bulk milk samples respectively. The maximum range of residues was 2.1312 ppm in packed milk while in bulk milk samples was

temperature 240°C; oven maximum temperature 250°C; injector temperature 250°C; Detector temperature 300°C, flow rate of carrier gas 40 ml N2/min.

# RESULTS

Results in Table I show that aldrin residues were detected in 78.57% of packed milk samples. The residue level ranged from 0.0 to 0.01024 mg/L. The mean value was 0.0309 mg/L with a standard deviation of 0.0383. Aldrin

Table 1: Percentage of incidence of different chlorinated hydrocarbon inscontaminated the packed and bulk milk samples within specific concentration r

neaM ageravA mqq ni U.S ±	Percent	egnar sənbisəsi mqq ni	Rind of Milk	Defected residues
(), ()3()9 ± (0,0383) (), ()247 ± (), ()293	72.82 73.68	42()1.() - ().() (4()1.() - ().()	Packed	niublA
$(0.0259 \pm 0.0403$ $7450.0 \pm 4120.0$	17.28	SSII.() - ().() ()172.1 - ().()	Packed bulk	Chlordane
£810.0 ± 0000.0 *CLN	14.29	#840.0- ().()	Packed	niabləid
7812.4 ± 1707.1 8079.0 ± 7888.0	14,29	0209.11 - 0.0 887.£ - 0.0	Packed	Taaz
7781.() ± 128().() *(LN	14.29	*(1,V) - (),3644	Packed	niabnə
882() ± ().7288 ().4276 ± ().3553	100.001	S181.S - 8060.0 6410.1 - 0.0	Packed	Teptachlor
$2 \xi(0)(0, 0) \pm \xi(0)(0, 0)$ $2 \xi(0)(0, 0) \pm 1 \xi(0)(0, 0)$	07.28 00.08	89()(),() - (),() 111(),() - (),()	Packed	anabai.

Lindsine or y-BHC, residues were detected in 85,70% and 80,00% of the sinalysed packed and bulk milk samples respectively. The mean of residues was 0,0033 ppm in packed samples and 0,0051 in bulk milk samples.

Table 2: Average detected residues (ADR) of it cide in tested milk samples.

ADR/MRI ratio	*J/Sm	MIN	Insecticide
24.4 04.11 75.0 14.21 27.02 22.87	0800.0 0200.0 0800.0 0020.0 8000.0 0800.0 0010.0	7320.0 8220.0 2200.0 7077.0 6310.0 8174.0 8400.0	Aldrin Chlordane Dieldrin EDDT Heptachlor Heptachlor

MRL = maximum residue level

residues were also detected in 86.67% of bulk milk samples. The residue levels ranged from 0.0 to 0.1049 mg/L. The mean value was 0.0247 mg/L, with a standard deviation of 0.0293.

Chlordane residues were detected in 85.71% of packed in 0.0.0 from 0.0.0 or 0.0.0 or

Dieldrin residues in packed milk ranged from (),() to 0.0484 with mean equal (),(0)69 ± (),(0)183. The percent of incidence of dieldrin was 14.29% in packed samples and 0.0% in bulk samples.

Sum DDT residues included DDE, DDD, DDT sind DDA. The DDA was the only analogue detected in the packed milk while in the bulk milk DDE was detected. The DDT residues were detected in 14.29% of packed samples and in 26.67% of bulk milk. The residue levels ranged from 0.0 to 11.9950 ppm at mean of  $1.7071 \pm 4.5167$  ppm in packed milk samples. The residue levels of ZDDT were in bulk milk than in packed milk samples.

Endrin residues were detected in 14,29% of packed samples but not detected in bulk samples. The average

Results in table 2, showed the average detected residues (ADR) as calculated from all samples and the maximum residue level (MRL).

These results indicated that aldrin, chlordane, DDT and heptachlor residues were exceeded the MRL. The hightest ratio was shown in heptachlor it was 78.55 folds more than the MRL.

#### Discussion

For economic reasons, organochlorine pesticides, have been, and are still used extensively in Egypt (Abdel Gawaad 1985 and CAPMAS 1992). The presence of high levels of organochlorine pesticide residues in milk in Egypt was reported by El Alfy 1981, Abdel Gawaad and Shams El Dine 1989, Abdel Gawaad 1985, and Ezz, et al., 1991a.b).

High levels of different chlorinated hydrocarbon pesticides were also reported in milk in USA Rogan et al., (1986) and Bakken and Seip (1976); in Japan by Inoue et al., (1979); in Spain by Lora et al., (1979); in Israel by Weisenberg et al., (1980); in Costarica by Umana and Constenla (1984) and in Egypt by Ezz et al., (1991 a).

The global perspective of organochlorine pesticide residues in dairy milk as compiled by the GEMS/food programme reveals from the data submitted by reporting countries, that, in general, milk contains the highest residue levels compared to any other food group. However, these residues are generally below MRL's with a few exceptions and are slowly declining in most developed countries such as USA, Canada and Netherlands as well as some developing countries. There is no evidence of changes in these levels with time as the general trend is maintained except for Germany, Japan and some developing countries, where the level is increasing at a high rate. (GEMS, 1991).

The organochlorine pesticides detected in this study are in accordance with previous studies conducted in Egypt in general for most pesticides. However, residues for some pesticides, that were not used extensively in Egypt at any time, were detected at an exceptionally high level in both packed and bulk milk samples. This could only be explained by the contamination of animal feeds and concentrates used. As the relation between levels of chlorinated hydrocarbons in feed and that in cows milk is linear. However at lower levels in feed the level in milk could be as much as double or more that in feed (Witt et al., 1966 and Matsumura 1976). These feeds and concentrates are being imported from countries that used or may be are still using these pesticides.

Another possible explanation for the aforementioned contamination, especially when the high levels are mainly in packed milks is that such residues are present in the imported powder milk being added and recombined with fresh milk in some packed milk factories and possibly in some of the bulk milk as well.

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