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CONTAMINATION OF COMMERCIAL MILK BY ORGANOCHLORINE PESTICIDES IN LIBYA

(With 2 Tables)

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تلوث الحليب التجاري المنتج في ليبيا بالمبيدات العضوية الكلورية

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أجريت هذه الدراسة لمعرفة مستوى التلوث ببعض المبيدات العضوية في 6 علامات تجارية من الحليب المتداول بالمنطقة الغربية من الجماهيرية الليبية . استخدم جهاز الكروماتوغرافي (GLC) الغازي المزود بكاشف الكتروني (ECD) لقياس هذه المركبات. أوضحت نتائج هذا البحث أن 40 عينة من 59 كانت ملوثة بواحد أو أكثر من المبيدات العضوية الكلورية كما ثبت أيضاً أن كل من Lindane و HCB هما أكثر الملوثات التي تم الكشف عنها وذلك بمعدل تلوث بلغ 37% لكل مركب. كما تم الكشف عن المركبات العضوية الأخرى بمعدل تلوث بلغ 68% للـ ppDDD و 5% للـ ppDDD ، 169 لـ ppDDT. وفيما عدا بعض العينات التي كانت درجة تلوثها ضئيلة فإن بقايا هذه المركبات كان غالباً عند مستوى أعلى من المسموح به. كذلك أن معظم العينات التي وجدت ملوثة بـ HCB كانت تحتوي على تركيزات أعلى من القيم المسموح بها.

SUMMARY

Contamination of Libyan commercial milk obtained from six different sources by some chlorinated pesticides was studied using gas-liquid chromatography with electron-capture detector. The results of this investigation revealed that out of 59 analyzed samples, 40(67.8%) were contaminated by one or more chlorinated

pesticides. Moreover, HCB and lindane were the most prevalent compounds with contamination frequency of about 37 % in both cases. Other organochlorine contaminants were also found at the frequency of 6.8 % for aldrin, 10 % for pp'-DDE, 5 % for pp'-DDD and 16.9% for pp'-DDT. Except in some cases where the contaminants were only detected in traces, residues of these were often present at concentrations higher than the permitted tolerance residue levels. The public health significance of chlorinated pesticide residues were discussed.

Key words: Commercial Milk - Contamination - Pesticides - Libya.

INTRODUCTION

Food-chain contamination by chemicals has drawn increasing concern from hygienists and toxicologists. This problem become very important particularly in "developing" countries where pesticides are largely used in agriculture and in disease vector control. These compounds, especially organochlorine pesticides, are known to be highly persistent in the environment and, thus, are susceptible to present a potential hazard to public health due to the harmful biological effects (UHNAK *et al.* 1986; TANABE *et al.* 1990; DEIANA and FATICHENTI, 1992).

In Libya, tremendous amounts of pesticides have been used for more than one decade as a result of agricultural development. For example, the quantity of pesticide used for agriculture purposes was estimated to 2,200 tons from 1973 to 1975 (TAHER *et al.* 1978) and to about 6,000 tons from 1976 to 1980 (National Statistic Board, 1985). These quantities are relatively high as compared to those applied in other countries like Morocco where agricultural activities are more intensive (KESSABI *et al.* 1990).

The aim of this paper is to report some preliminary results related to the assessment of pesticide contamination of commercial milk in Libya. Such investigations are almost lacking in this country.

MATERIAL and METHODS

A total of 59 milk samples representing six different sources of milk manufactured in the north western region of Libya were collected from August to September 1993. Milk was first stored in ice boxes and sent shortly to the Hassan II Institute of Agriculture and Veterinary Medicine of Rabat (Morocco), where it was kept frozen until extraction and chemical analysis.

The extraction of chlorinated pesticides was accomplished according to the method described by Veierov and Aharonson (1980), which is slightly modified. Briefly, pesticides residues were isolated from liquid milk by liquid-liquid partitioning using concentrated sulfuric acid and petroleum ether in conical funnels. Purification was accomplished by silica gel adsorption and the detection and quantification of residues was performed by a gas-liquid chromatography system (hp 3750 A) with an electron capture detector and a calculator integrator (hp 3390 A). Calibration was done by standard dilutions ranging from 10 to 500 ppb depending on the organochlorine standards which were the following : hexachlorobenzene (HCB), lindane, aldrin, dieldrin, endrin, p,p'-dichlorodiphenyldichloroethylene (p,p'-DDE), p,p'-dichlorodiphenyldichloroethane (p,p'-DDD) and p,p'-dichlorodiphenyltrichloroethane (p,p'-DDT).

RESULTS

The results of this investigation indicate that out of 59 analyzed samples, 40 (67.8 %) were contaminated by one or several chlorinated pesticides. The frequency of this contamination differed according to the nature of pesticide ranging from 37.3 % for HCB and lindane to 5 % for p,p'-DDD with 6.8 % for aldrin, 10 % for p,p'-DDE and 16.9 % for p,p'-DDT (Table 1).

Mean residual levels as well as the range (minimum and maximum) of the detected pesticides are mentioned in Table 2. The calculated overall mean concentration values were 61.9 for HCB, 187.1 ppb for lindane and 178.8 ppb for aldrin. The corresponding values for DDT and its derivatives were found to be 84.0 ppb for p,p'-DDE, 17.5 for p,p'-DDD and 76.2 ppb for p,p'-DDT. It has to be noticed that, in this study, dieldrin was not detected in any of the analyzed samples. A comparison of the contamination level among the six different sources of milk is difficult to make in this situation. However, we may consider from data of table 2 that milk samples issued from Ogba and airport plants are relatively the most contaminated followed by milk obtained from university experimental station and that of the private shop: AlArabi.

DISCUSSION

It is evident from the obtained results that most of the analyzed milk commercialized in Libya was contaminated by chlorinated pesticides. The contamination by HCB and lindane and, to a lesser extent, by aldrin and DDT derivatives may be related to the use of these chemicals in agriculture, animal production sector and in public health. For example, out of the total quantity of

pesticides used from 1976 to 1980, 2,000 tons consisted of insecticides including lindane, aldrin and dieldrin (National Statistic Board, 1985). Lindane is the most widely used insecticide in the treatment of external parasitic diseases in animals. Hexachlorobenzene is frequently used as a fungicide and it is also a contaminant of other organochlorine molecules (Venant and Richou, 1981) or derived from many industrial activities (Frank and Ripley, 1990). Furthermore, the accumulation of HCB in the environment is well studied and this chemical is known to be very persistent in the food-chain (Uhnak *et al.* 1986). Similar findings concerning the relatively high contamination frequency of HCB and lindane have also been reported in Moroccan dairy (Kessabi *et al.* 1990) and human milk (Benazzou, 1992). Although dieldrin is used in Libyan agriculture since 1977, it was not detected in any of the analyzed milk samples. This phenomenon may be due to the fact that residues of this compound were destroyed by the acid treatment used to clean up the samples (UNEP/IOC/IAEA, 1986).

In reviewing the detected levels, it become evident that certain samples contained residues of pesticides which are higher than those authorized in milk by the Swedish regulation (Vaz, 1993) and the Codex alimentarius (Codex Alimentarius, 1989). In fact, 73 % of the contaminated milk samples had residues of HCB higher than 20 ppb which is permitted in Sweden and 36.4 % of these samples contained residues of p,p'DDT above the Codex alimentarius tolerated value which is 50 ppb. The detected residues of lindane were mostly very high since 91 % of the residue concentrations are equal to or greater than the maximum tolerance level of this compound in milk which is 10 ppb. This situation is undesirable because of the potential for human exposure especially children. It has been found that HCB intake in children up to 3 years of age was seven times that in adults (Uhnak *et al.* 1986).

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Table 1: Occurrence of organochlorine pesticides in Libyan milk collected from different sources.

	HCB	Lindane	Aldrin	pp'-DDE	pp'-DDD	pp'DDT
OGBA (n=10)						
Number (*)	4	2	1	1	0	1
%	40	20	10	10	0	10
UNIVERSITY (n=12)						
Number	4	8	0	1	0	1
%	33.3	66.6	0	8.3	0	8.3
AIRPORT (n=12)						
Number	4	4	1	1	0	2
%	33.3	33.3	8.3	8.3	0	16.6
AL ARABI (n=13)						
Number	6	4	0	3	3	6
%	46	30.8	0	23	23	46
HERRA (n=10)						
Number	3	4	2	0	0	0
%	30	40	20	0	0	0
EVAPORATED (n=2)						
Number	1	0	0	0	0	0
%	50	0	0	0	0	0
TOTAL (n=59)						
%	37.3	37.3	6.8	10.2	5.1	16.9

(*) = Number of positive samples

**Table 2: Contamination levels (ppb) of Libyan milk by organochlorine pesticides
(T indicates trace amount found).**

	HCB	Lindane	Aldrin	pp'-DDE	pp'-DDD	pp'DDT
OGBA						
mean	86.7	717.5	172.4	262.0	-	20.7
range	12.8-186	119-1316	0.0-172.4	0.0-262	-	0.0-20.7
UNIVERSITY						
mean	45.1	144.9	-	13.7	-	14.0
range	T-57.9	T-233.3	-	0.0-13.7	-	0.0-14.0
AIRPORT						
mean	76.3	102.9	244.0	20.8	-	223.8
range	T-102.7	T-206.0	0.0-244.0	0.0-20.8	-	117.0-330.7
AL ARABI						
mean	52.8	96.1	-	61.7	17.5	61.6
range	T-77.1	32.5-156.6	-	34.5-89.0	T-21.0	T-200.0
HERRA						
mean	51.5	154.7	149.5	-	-	-
range	32.3-77	29.5-375	126.7-172	-	-	-
EVAPORATED						
mean	102.7	-	-	-	-	-
range	T-186.0	-	-	-	-	-
TOTAL						
mean	61.9	187.1	178.8	84.0	17.5	76.2
Range	T-186.0	T-1316	172.4-244.0	T-262.0	T-21.0	T-340.7

