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Changing Patterns of Cow's Milk Contamination with Organochlorine Compounds in Israel (1976–1986)

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Environmental pollution with organochlorine insecticides (OCIs) and polychlorinated biphenyls (PCBs) has decreased due to restriction and/or banning of these compounds (Abbot et al. 1985; Schmitt et al. 1985; Weisenberg et al. 1985; Holt et al. 1986). However, several recent reports indicate that pollution with OCIs and PCBs still does exist, and may be of public and environmental health significance even in developed countries (Kim 1984; Sawhney and Hankin 1985; Brunn et al. 1985; Rogan et al. 1986).

Cow's milk is one of the most important media where organochlorine compound (OCC) levels are monitored (Wedberg et al. 1978; Frank et al. 1979; Futianos et al. 1985). On the one hand, milk and its products play a central role in human nutrition and, on the other, highly lipophilic OCCs can accumulate in fat-rich milk products.

In our previous study (Pines and Cucos 1984), we reported a significant decrease in OCC content in both farm and market milk in Israel between the mid-1970s and early 1980s. The clearest decrease was seen in the cases of DDT, dieldrin and lindane, but not as much as for heptachlor epoxide (HE) and PCBs. The present study was undertaken to follow up the observed trends and to determine whether cow's milk contamination with OCC presently poses a public health problem in Israel.

MATERIALS AND METHODS

The overall study included 42 samples of farm cow's milk (1976 - 10 samples, 1983 - 14, 1986 - 18) and 41 samples of market milk (1976 - 10 samples, 1983 - 16, 1986 - 15). Sampling was performed at random. To limit the effect of uncontrolled confounding factors

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(geographical or seasonal), farm samples in 1983 and 1986 were selected from the same milk farm during the same month (April). Market milk samples were collected during the same month as farm milk samples.

Determination of OCC residue levels in milk included milk lipid extraction and separation of OCIs from PCBs (Armour and Burke 1970), followed by gas chromatography (Packard - 7400 with an electron-capture detector). The detailed procedure of OCC measurement, as performed in this laboratory, is described elsewhere (Polishuk et al. 1977). In line with reported mass- spectrometric studies (Greichus et al. 1974), individual chloropbiphenyl peaks were grouped and subsequently identified as tetra-, penta- and hexa-chlorobiphenyls (CBs), using Aroclor 1254 as a standard. All the analyses were performed applying identical methodology by the same chemist; therefore, interlaboratory and interobserver variations in results (Lawton et al. 1985) have been minimized. Statistical procesing of the data included calculation of the mean residue levels for each compound during each sampling period and standard error of mean (SEM). The difference between means for different years was tested with Student's t-test. The 1983 and 1986 findings were also compared by Fisher's exact test (FET). No outliers were excluded. Undetected OCC residues were treated as zero values.

RESULTS AND DISCUSSION

During 1976-1983, statistically significant reductions of the residue levels of total DDT, lindane and dieldrin took place in both farm and market milk samples, while HE decreased in market samples only. During the following 3-year period, further reductions took place in DDT, lindane and dieldrin residues in farm milk samples, as well as reductions in DDT and dieldrin levels in market milk samples. All the above decreases were statistically significant by both Student's t-test and FET (Table 1).

The changes in the pattern of DDT analogs are especially noteworthy (Table 2, Fig. 1): p,p'-DDT and o,p'-DDE pratically disappeared in the 1986 samples of both market and farm milk. Other DDT analog levels showed a continuous decrease from 1983 to 1986, except that of p,p'-DDE in the farm milk samples, which remained stable, and p,p'-DDD in market milk samples, which significantly increased in 1986 as compared with 1983. This discrepancy between the trends of p,p'-DDT residues in farm and market cow's milk samples remains to be elucidated. Disappearance of p,p'-DDT from milk and the increase in the share of of p,p'-isomers

Table 1. Changes in some organochlorine insecticide milk residues (mean ± SEM) ng residue/g whole milk

Year of sampling	Total DDT[a]	Lindane	Dieldrin	Heptachlor epoxide
		Farm milk samples		
1976[b]	43.7 ± 5.00	5.5 ± 1.74	4.5 ± 1.01	2.3 ± 1.23
1983	13.7 ± 1.15*** @@@	1.5 ± 0.14* @@@	2.3 ± 0.18* @@@	2.9 ± 0.38
1986	6.6 ± 0.79***	0.4 ± 0.06**	0.7 ± 0.07**	2.2 ± 0.18
		Market milk samples		
1976[b]	87.7 ± 6.61	15.7 ± 1.55	9.8 ± 1.17	11.1 ± 3.11
1983	8.9 ± 1.37*** @	0.7 ± 0.12***	1.8 ± 0.33*** @@	2.1 ± 0.37**
1986	4.9 ± 0.60***	0.7 ± 0.09***	0.8 ± 0.14***	1.9 ± 0.23**

[a]Sum of p,p'- and o,p'-DDT analogs calculated as DDT.

[b]Data calculated from Nutsathapana (1976).

Significance testing (Student's t):
1983 or 1986 vs. 1976: *p < 0.05, **p < 0.01, ***p < 0.001;

1986 vs. 1983: @p < 0.05, @@p < 0.01, @@@p < 0.001.

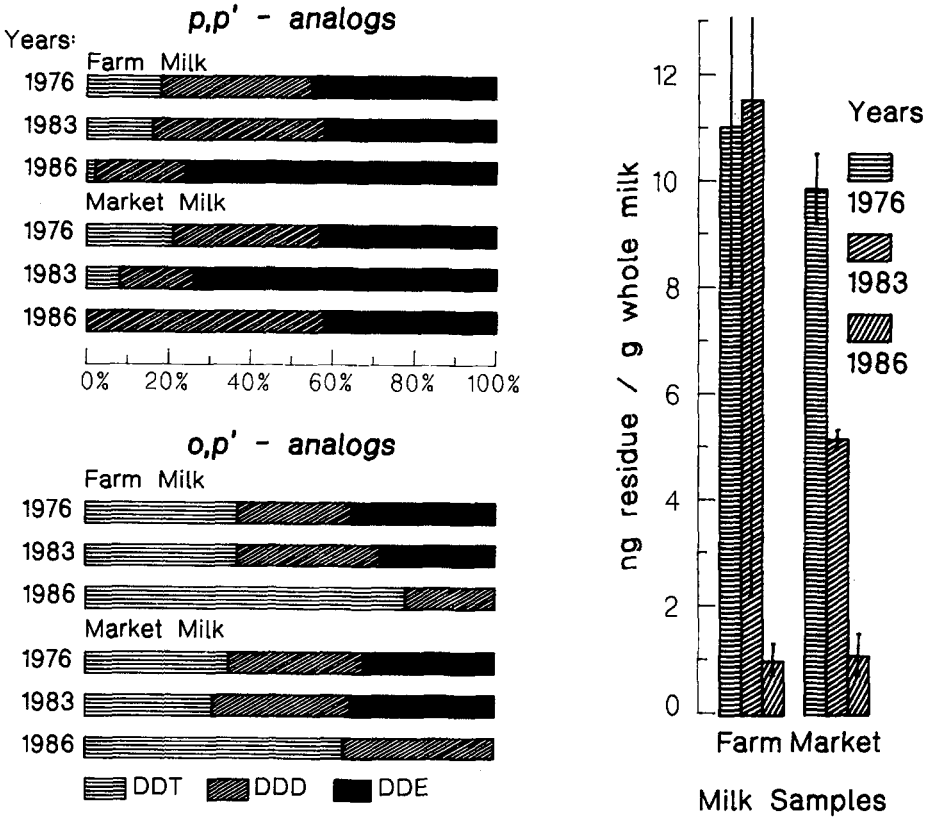

Figure 1 Changes with time in the distribution pattern of p,p'- and o,p'-DDT analogs in the farm and market milk samples.

Figure 2 Changes with time in the total PCBs concentration in the farm and market milk samples.

accounted for by DDE reflect differences in chemical stability and metabolization rates of these compounds: the latter indicates mainly past exposure to this insecticide, while the former reflects current exposure (Peterson 1979).

Unlike OCIs, residues of PCBs in milk underwent a steep decrease in the 1983-1986 period, while during the preceding period, this decrease was less pronounced and observed only in market milk samples (farm samples exhibited significant reductions in total PCBs only after exclusion of outliers) (Fig. 2). The pattern of CB congeners changed in parallel to those of p,p'-DDT analogs, with complete disappearance of less stable and quickly metabolized tetra-CBs, indicative of current exposure, and a relative increase in the proportion of slower biodegradable, highly chlorinated penta- and hexa-CBs, characteristic of past exposure (Wolf et al. 1982)

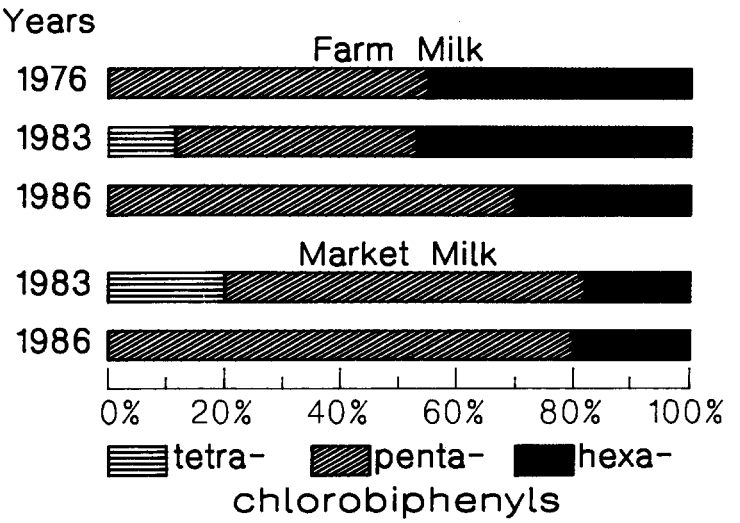

Figure 3 Changes with time in the chlorobiphenyl distribution pattern in the farm and market milk samples (data for market milk samples for 1976 are unavailable).

In the 1976 series of milk samples, those from the market exhibited OCC residues two to five times higher than those form the farm. This might have resulted either from atypically low contamination of the farm where 1976 samples were picked, or from strongly biased sampling of market milk during that period. As shown in our previous study, a large range of variation exists between the average OCC concentrations in various grades of milk (Pines and Cucos 1984). In the 1983 and 1986 sample series, the differences between market and farm residue levels were small, although statistically significant for some compounds.

A drastic decrease in the OCI contamination observed during the 11-year period of investigation is clear evidence of successful enforcement of regulatory measures aimed at abolishing use of the OCIs for crop protection and for control of vector diseases in Israel. Such measures were initiated for DDT in 1970, dieldrin in 1975, lindane in 1978, and heptachlor in 1980. No such regulations exist concerning the use of PCB's, although in the first half of the 1980's, the Israeli Electric Company voluntarily discontinued use of PCB-containing transformers and other equipment. An obvious effect of the latter measure on cow's milk contamination emerged, albeit later than the ban on OCI use. Further support regarding the effect of regulatory measures is provided by resemblance of qualitative and quantitative changes in OCC residue patterns in cow's milk, and in blood samples of the general Israeli

Table 2. Changes in DDT analog milk residues (mean ± SEM ng residue/g whole milk)

	Year of sampling	DDT	DDD	DDE	Sum of the respective isomer analogs[a]
		Farm milk samples			
p,p'-isomers	1976[b]	$4.9 \pm 0.76^{***}$	$9.2 \pm 2.34^{**}$	$11.6 \pm 1.42^{***}$	$28.1 \pm 3.10^{***}$
	1983	0.9 ± 0.21	$2.1 \pm 0.29^{**@@}$	$2.1 \pm 0.77^{***}$	$5.6 \pm 0.55^{***}$
	1986	-[c]	0.9 ± 0.19	3.2 ± 0.43	4.7 ± 0.59
o,p'-isomers	1976[b]	$5.8 \pm 0.76^{**}$	3.8 ± 1.26	5.0 ± 1.87	$15.6 \pm 2.47^{**}$
	1983	$2.9 \pm 0.34^{***@@}$	$2.6 \pm 0.23^{*@@@}$	2.1 ± 0.19	$8.1 \pm 0.71^{**}$
	1986	1.5 ± 0.30	0.4 ± 0.9	-[c]	$1.9 \pm 0.31^{***@@@}$
		Market milk samples			
p,p'-isomers	1976[b]	$12.3 \pm 2.43^{***}$	$18.5 \pm 2.59^{***}$	$22.0 \pm 2.37^{***}$	57.4 ± 4.81
	1983	0.3 ± 0.07	$0.6 \pm 0.10^{***@@}$	$2.4 \pm 0.36^{***@@}$	$3.6 \pm 0.49^{***}$
	1986	-[c]	1.7 ± 0.17	1.2 ± 0.21	$3.2 \pm 0.30^{***}$
o,p'-isomers	1976[b]	$10.6 \pm 1.68^{***}$	$8.7 \pm 1.83^{***}$	$9.0 \pm 1.36^{***}$	$30.3 \pm 4.52^{***}$
	1983	$1.6 \pm 0.26^{***@@}$	$1.6 \pm 0.31^{***@@@}$	1.7 ± 0.32	$5.3 \pm 0.92^{***}$
	1986	1.0 ± 0.32	0.6 ± 0.20	-[c]	$1.7 \pm 0.44^{**@@@}$

[a] Calculated as DDT

[b] Data calculated from Nutsathapana (1976)

[c] In all but one sample, residue below the detection limits

Significance testing (Student's t): 1983 or 1986 vs. 1976: $^{*}_{@}p < 0.05$, $^{**}_{@@}p < 0.01$, $^{***}_{@@@}p < 0.001$;
1986 vs. 1983 : $^{@}p < 0.05$, $^{@@}p < 0.01$, $^{@@@}p < 0.001$.

population during the same period (Pines et al. in press).

The currently observed levels of OCC residues in both farm and market milk samples are well below the maximum residue limits for OCI pollution of whole milk established by FAO/WHO (1983) as well as the FDA tolerance for PCBs in milk fat (US FDA, 1979). This fact, together with cautious extrapolation of the observed downward trend in cow's milk OCC residues, enables us to conclude that the current level of cow's milk contamination by the above reported compounds is very low in Israel and may hardly represent any measurable threat to human health.

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