

## Organochlorine Pesticide Contaminants in Human Adipose Tissue Collected in Ankara (Turkey) 1991–1992

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Many studies have shown the extreme persistence of organochlorine pesticide (OCP) contaminants in the environment, their accumulation in human tissues, and their toxicity. Their concentrations in adipose tissues of human populations are good markers in determining the extent of exposure and in evaluating the hazard(Jensen 1983, Kutz et al., 1991). Most countries have conducted initial monitoring programs to determine OCP residues in their human populations (Kashimoto et al., 1989, Robinson et al., 1990, Mes et al., 1990).

In Turkey, OCP residues have been monitored in the Turkish population by carrying out national surveys at given time intervals since 1976 (Kayaalp et al., 1979, Karakaya and Özalp 1987, Karakaya et al., 1987). This paper reports on the part of the results of the latest monitoring program in terms of residue levels in the body fat of residents of Ankara and draws a possible trend of the disappearance of restricted OCP.

## MATERIALS AND METHODS

Human adipose tissue samples were taken during surgical operations in Gazi University and Ankara State Hospitals from 60 patients living in Ankara at least 5 years. Each patient had been interviewed information regarding sex, age, weight, height, smoking and dietary habits. The sample consists of 25 men and 35 women, aged between 15 and 81, with a mean of 45.5. Most the donors except eighteen were nonsmokers. All of the subjects were mixed food consumers. The mean of donors' body mass index (SD) was 26.3 (4.3). The adipose samples were deep-frozen (-20°C) immediately tissue until analysis.

Standards of HCB,  $\alpha$ -BHC,  $\beta$ -BHC,  $\gamma$ -BHC, pp'DDE, pp'DDT, op'DDE, heptachlor and heptachlor epoxide were obtained from U.S. Environmental Protection Agency (EPA).

100 mg samples were ground with anhydrous sodium sulphate by using a tissue grinder, op'DDE, confirmed not to be present in samples, was used as an internal standard and also added to the adipose tissue before extraction. Then tissue samples were extracted with 4x2x2 ml of n-hexane. The combined n-hexane extract was evaporated to dryness to determine extractable fat content (Smrek and Needham 1982). Using 8 ml n-hexane as the solvent, the sample was cleaned up with 1-2 ml of conc.sulphuric acid (Murphy 1972, Wickström et al., 1983).

Gas chromatography analysis was done on a Packard 438 equipped with  $^{63}\rm Ni$  EC detector. The column used for quantitation was a 25 m  $\times$  0.25 mm fused silica capillary column cp Sil 8 from Chrompack. The operating conditions were: injector temperature 260°C; detector 320°C; column 80°C initial with 1 min. hold 10°C/min to 280°C. A 25 m long glass capillary column cp Sil 5 was also used. Peak heights were used as the basis for quantification. Residue levels are expressed as mg/kg extracted fat (ppm).

Recoveries from a fortified sample at a 0.2 ppm each level were in the range of 80.3-92.1 % on this method, including internal standard. Results were not corrected for the percentage recovery.

As heptachlor is unusual finding in humans, we refer to the residue as heptachlor-like.

The different sets of data were examined for significant difference (p<0.05) by Student's-t test. Correlation analysis were done by simple linear regression.

## RESULTS AND DISCUSSION

The residue levels of OCP are shown in Table 1. It can be seen that residues of HCB,  $\beta$ -BHC, pp'DDE, pp'DDT and heptachlor epoxide are found to be major contaminants in the samples analyzed. All the OCP show higher values in females than in males, but the only compounds that show significant differences are HCB,  $\beta$ -BHC, pp'DDT and heptachlor epoxide. Although it is difficult to reach conclusions, however, these results are in concordance with those obtained by other authors and are indicatives of the existence of some sex related factor in the accumulation of OCP residues (Mes et al., 1982, Camps et al., 1989, Kutz et al., 1991).

A general trend of increasing OCP concentration with increasing age appears to exist. Residue levels of HCB and  $\beta$ -BHC increase significantly with selected age groups. Although the same is true for pp'DDE and  $\Sigma$ DDT levels, there is no significant difference between age

groups. These findings are consistent with previous studies which have shown that continued exposure results in higher levels of these chemicals in human adipose tissue because of bioaccumulation (Kutz et al., 1991).

Smokers did not have more OCP compounds in their fat tissues compared with nonsmokers. On the contrary, high residue levels of  $\beta\text{-BHC}$  and pp'DDT in human fat samples were significantly more frequent in nonsmokers compared with smokers. This might indicate that the residue levels in adipose tissue reflect relatively recent exposure of humans to OCP. As previoully reported, tobacco is of minor importance as a source of OCP compounds (Mussalo-Rauhamaa et al., 1984, Abbott et al., 1985).

Table 1. Chlorinated hydrocarbon residues in adipose tissue of Ankara citizens (The mean levels expressed as mg/kg extracted fat basis)

Compound	Mean (S.D.)	Range	Frequency of residue (%)
нсв	0.164 (0.22)	(0.008-1.135)	100.0
α-BHC	0.014 (0.06)	(ND≃-0.428)	11.7
β-BHC	1.522 (1.03)	(0.271-5.276)	100.0
γ-BHC	0.002 (0.01)	(ND-0.062)	3.3
Σ BHC <sup>b</sup>	1.539 (1.04)	(0.271-5.325)	
pp'-DDE	3.722 (3.59)	(0.722 - 24.813)	) 100.0
pp'-DDT	0.275 (0.32)	(0.007-1.889)	100.0
Σ DDT=	4.42 (4.16)	(0.838-28.886	)
'Heptachlor'	0.021 (0.04)	(ND-0.181)	51.7
Heptachlor		•	
epoxide	0.062 (0.06)	(ND-0.432)	91.7

<sup>■</sup>N.D. = under the limit of detection

Our results indicated that the donors' body mass index (weight/height²) did not correlate with residue levels of OCP compounds in human adipose tissue.

HCB is found in all the fat tissue samples, despite the fact that it has not been ever since 1959 allowed in Turkey for agricultural uses (Göcmen et al., 1989).

By comparison with published data from other countries the values we obtained for HCB (0.164 ppm) are lower than those found in Italy (2.26 ppm) and Spain (2.99 ppm) in which HCB has never been massively used as fungicide (Focardi et al., 1986, Camps et al., 1989).

 $<sup>^{</sup>b}\Sigma BHC = \alpha - BHC + \beta - BHC + \delta - BHC$ 

 $<sup>^{\</sup>circ}\Sigma DDT = 1.115 \times pp'DDE + pp'DDT$ 

<sup>&#</sup>x27;Heptachlor'=Heptachlor-like

Our HCB levels given in this paper are, however, higher than those found in some American population (0.037 ppm). HCB is known to have been used as fungicide and also to have been released in the environment due to industrial activities in U.S.A.(Robinson et al.,1990).

studies, describe that HCB is released in Several environment by the discharge of plastic or as products of the chlorinated hydrocarbons and may reach agricultural land by waste compost and atmospheric fallout. HCB is also a contaminant of other widely pesticides (Tobin 1986). HCB levels in our study difficult to interpret because of its diverse and not known sources, however, HCB may still appear as contaminant of various pesticides which are also widely used in our country. It is unfortunate that no previous data on HCB levels in human adipose tissue exist Ankara region. These would be of great interest compared to the present data.

Traces of 'heptachlor', was detected in the half of the samples. However, its metabolite, heptachlor epoxide was clearly detected in most of the samples analyzed. The presence of heptachlor epoxide in Turkish human adipose tissue has not been reported earlier. The heptachlor epoxide levels in human body might have originated as an oxidation product from heptachlor as pesticide used in our country until 1985. Heptachlor epoxide levels found are lower than those reported from Germany, Italy, Japan and U.S.A.(Kutz et al., 1991).

A positive correlation (p=0.0001) is found between the concentrations of the pp'DDE,  $\beta$ -BHC and HCB contaminants. This may indicate that these compounds are perhaps obtained from the same dietary sources, and this has also been reported by other authors (Ferrer et al., 1992).

In Turkey, beginning from 1979, restrictions are imposed on OCP and since 1985 usage has been prohibited except endosulfan, toxaphene and diflubenzuron. Figure 1 shows the trends for the mean levels of the main residues, pp'DDE, pp'DDT and  $\Sigma BHC$ , over the 15 years that these surveys have been carried out in Ankara region.

Comparison of results shows that the downward trend for pp'DDT continues, as previously observed (Karakaya and Uzalp 1987). The decreases in concentrations probably reflect the continuing prohibitions on the use of DDT by our country. This observation is also supported by the significant decrease in the mean concentrations of pp'DDE, while no significant decrease in the mean levels of  $\Sigma BHC$  is observed during the period from 1984 to 1992 (Figure 1).

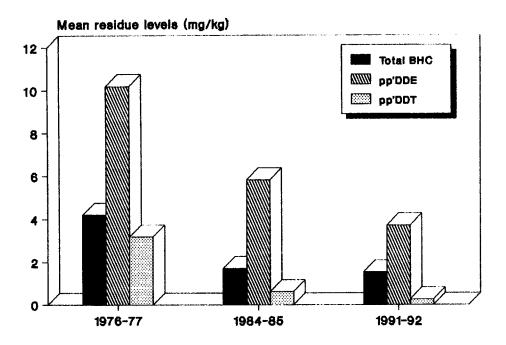


Figure 1. Mean levels of residues of OC compounds in human adipose tissue from Ankara over the period 1976-1992, based on the following data:

Year	Mean	residue	level	(mg/kg)
	ΣΒΗС	pp'DDE	pp'DDT	References
1976-77	4.2ª	10.2	3.2	Kayaalp et al (1979)
1984-85	1.72	5.83	0.62	Karakaya and Özalp (1987)
1991-92	1.54	3.72	0.27	Present study

a: as Y-BHC

In Table 2, the levels of pp'DDE and pp'DDT in human adipose tissue found in the present study are compared with those of other countries. By comparison with published data from other countries, levels of pp'DDE and pp'DDT from Ankara region are similar to those of most countries in recent years with the exception of

India, where pp'DDT is probably still used. When the use of pp'DDT ceases, human exposure to this compound decreases initially fairly rapidly. However, exposure to its persistent metabolite pp'DDE is still occurs due to continued exposure mainly via foodstuffs of animal origin. Thus the ratio of DDE and DDT in the human body will increase when the use of DDT ceases (Slorach and Vaz 1983). In our study, DDT and DDE correlated significantly (r=0.456, p<0.05) and DDE/DDT ratio is 13.77, which is as high as those found in most European countries where DDT use has been prohibited since 1970's (Table 2).

Table 2. Mean levels (mg/kg extracted fat) of pp'DDE and pp'DDT compounds in human adipose tissues collected in different countries

Country	Year	pp'DDE	pp'DDT	DDE/DDT	References
Canada	1979-81	3.26	0.16	20.3	Williams et al (1984)
India	1982	9.39	9.66	0.97	Ramachandran et al (1984)
Italy	1983-84	7.35	0.83	8.85	Focardi
United Kingdom	1982-83	1.3	0.11	11.81	et al (1986) Abbott et al (1985)
U.S.A.	1987-88	0.68=	0.29=	2.34	Adeshina et al
Turkey	1991-92	3.72	0.27	13.77	(1990) Present study

<sup>&</sup>lt;sup>a</sup>Geometric mean

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