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Levels of Organochlorine Pesticides in Beijing Human Milk, 1998

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During recent years, investigators have reported that human milk is often contaminated with organochlorine pesticides (OCP) and their metabolites (Albers et al, 1996; Cok et al, 1996; Czaja et al, 1997; Dogheim et al, 1996; Mahmoud et al, 1992; Newsome et al, 1995; Prachar et al 1993; Saleh et al, 1998; Vassilios Schinas et al, 2000). Contamination has resulted from the increased use of these compounds in agriculture and in animal husbandry. These materials have low water solubility, high environmental persistence, and are easy to be bioaccumulative in food chain. Breast milk is at the top of the food chain and one of the good markers for the determination of environmental pollution created by OCP.

Monitoring of OCP levels in the human milk is of particular importance as infants and little children fall into the high-risk category. Infants and little children do not have fully developed detoxification mechanism yet, their immune systems are immature, and their organs are in the process of rapid growth (Czaja et al, 1997; Cornacoff et al, 1988).

The objective of the present work was to identify and quantify OCP and their metabolites in human milk in the Chinese Capital of Beijing, and obtain additional information about DDT and BHC present pollution by comparing their levels with those in previous studies.

MATERIALS AND METHODS

Analytical grade solvents were used after distillation in an all-glass apparatus. Standards of HCB, α-BHC, β-BHC, γ-BHC, p, p'-DDE, p, p'-DDT were obtained from Chinese Center for Standard Materials and Technology.

60 human milk samples were collected in December 1998 from healthy donors living in four different regions of Beijing including Dongcheng, Xicheng, Xuanwu and Chongwen for at least five years. The age of mothers ranged from 22 to 30. Quantities between 40 and 50mL samples were taken by manual expression and kept in glass wide-mouth bottles of about 50 mL capacity, frozen at -20℃ until analysis. Each mother completed a questionnaire to provide personal

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information such as number of births, smoking, occupation, diary habits and place of residence.

Before extraction, samples were melted in 50 ℃ water bath and then homogenized. 10g of milk sample were mixed with 20mL of petroleum ether/acetone 1:1(v/v), then centrifuged until separation. The upper phase containing fat was transferred into a weighed tube and the lower phase was re-extracted twice with an additional 10mL of petroleum ether. The pooled extracts were evaporated to dryness under a stream of nitrogen and the fat content was weighed. The fat was redissolved in 5mL of petroleum ether and purificated by 5mL conc. H_2SO_4, and then centrifuged. 1μL sample was injected into gas chromatograph for analysis.

Gas chromatographic analysis was performed using a GC-9A Gas chromatograph equipped with ^{63}Ni electron capture detector and a 2m glass column packed with Chromosorb WHP coated by 1.5% OV-17 and 2% QF-1. The temperature of the injector and the detector were maintained at 230℃. The column was held at 200 ℃. The carrier gas was nitrogen at a flow rate of 40mL/min. Quantitative analysis was achieved by comparing the area of peaks with those of their respective standards of known concentrations. Residue levels are expressed as mg/kg extracted fat.

Detection limits for α-BHC, β-BHC, γ-BHC, HCB, p, p'-DDE, o, p-DDT, p, p-DDD, p, p'-DDT, were 0.005, 0.02, 0.005, 0.005, 0.04, 0.07, 0.07, 0.07mg/kg, respectively. Recoveries were in the range of 80-120% for this method.

RESULTS AND DISCUSSION

It had been 15 years in 1998 since OCP are prohibited for use in agriculture in Beijing in 1983. As these compounds are slowly degraded biologically, they tend to persist in the environment, and accumulate in the animal and human food chain. Sixty milk samples from 60 mothers in Beijing in China were analyzed for OCP. Average concentrations of OCP in the mother's milk samples (fat basis, median) are presented in Table 1. The total DDT residues in human milk are 2.04 mg/kg, which were contributed from the p, p'-DDT (0.24mg/kg) and p, p-DDE (1.72mg/kg). The total BHC residue in women's milk is 1.18mg/kg, which is from β-BCH. The level of HCB in human milk is 0.039mg/kg. In 1998, average levels of p, p'-DDT, p, p'-DDE, and total DDT residues decreased 85.3, 71.8 and 73.5% from 1983, respectively. β-HCH and total BHC residues also decreased 87.8 and 88.3%, respectively. Thus since OCP are prohibited for use in agriculture in Beijing in 1983, the total levels of these chemicals in human body has decreased.

The numbers of positive samples of OCP in the mother's milk samples (fat basis, median) in 1998 and 1983 are given in Table 2. In 1983, almost all DDT and BHC isomers and all DDT metabolites were found in investigated human milk samples.

But in 1998, p, p'-DDT was found in only 77% milk samples and p, p'-DDE was found in 100% samples. β-BHC was also found in all milk samples in 1998. These results show the number of OCP contaminants in human milk samples is decreasing. The ratio of the concentration of p, p'-DDE to that of p, p'-DDT in woman milk samples also increased from 3.59 in 1983 up to 7.34 in 1998.

Table 1. Average concentrations of OCP in the mother's milk samples (mg/kg fat basis)(Median)

Year	n	Milk fat(%)	p, p'-DDT	p, p'-DDE	∑-DDT	β-BHC	∑-BHC	HCB
1983	50	3.05	1.63	5.89	7.71	9.66	10.1	-
1998	60	3.62	0.24	1.72	2.04	1.18	1.18	0.039

Table 2. Positive samples of OCP in the mother's milk samples (Median).

Compounds	n(1998)	Positive Samples(%)	n(1983)	Positive Samples(%)
p, p-DDE	60	100	50	100
p, p'-DDT	46	77	50	100
p, p-DDT	0	0	50	100
p, p'-DDD	0	0	49	98
α-BHC	7	12	50	100
β-BHC	60	100	50	100
γ-BHC	1	2	50	100

Table 3 presents the decreasing levels of β-BHC and ∑-DDT in mother's milk samples in China and Germany from 1983 to 1998(Huifang Yu et al, 2000).

Table 4 lists reported mean concentrations of OCP in breast milk (mg/kg milk fat) in various countries (Tian Lan et al 1993). The level of DDT and β-BHC in Beijing woman milk samples in 1998 is higher than that in other developed countries in 1990s. For example, since DDT was prohibited for use in 1977 in Spain and in 1970 in Sweden, average concentrations of p, p'-DDE in human milk have decreased to 0.604mg/kg in 1991, which were lower than that in Beijing, where DDT has been banned for use for 15 years. This suggested that human milk contamination resulting from OCP residues in environment is still high in China.

Table 3. Comparison of decrease of β-BHC and ∑-DDT in mother milk samples between China and Germany (milk fat, mg/kg).

Contents	Beijing			Germany		
	1983	1998	% Decrease	1983	1998	% Decrease
∑-DDT	7.71	2.04	73.5	1.45	0.202	86
β-BHC	9.66	1.18	87.8	0.28	0.036	87

Table 4. Reported mean concentrations of OCP in breast milk (mg/kg milk fat) collected in various countries.

Country/City	year	p,p'-DDT	p,p'-DDE	∑-DDT	β-BHC	HCB
Beijing	1998	0.24	1.72	2.04	1.18	0.039
Germany	1997	-	-	0.20	0.04	0.065
Spain	1991	-	0.604	-	-	-

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