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Monitoring of Organochlorine Pesticide Residue Levels in Adipose Tissue of Veracruz, Mexico Inhabitants

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Abstract The objective of the present study was to monitor the levels of organochlorine pesticides HCB, α-β-γ-HCH, *pp*′DDE, *op*′DDT and *pp*′DDT in 150 adipose tissue samples of Veracruz, Mexico inhabitants. In analyzed samples, the following pesticides were detected: *p,p'*-DDE in 100% of the samples at mean 1.643 mg/kg; *p,p'*-DDT in 99.3.% of the samples at mean 0.227 mg/kg; β-HCH in 97.3% of the samples at mean 0.063 mg/kg; and *op*′DDT in 93.3% of the samples at mean 0.022 mg/kg. Comparing mean, median and geometric mean concentrations of organochlorine pesticides shows a decrease in values from mean to median and to geometric mean which points out a prevalence of lower concentrations among the total samples and the existence of occasional cases of extreme exposure expressed in range values. The pooled samples divided according to sex, showed only significant differences of *pp*′DDE median concentrations between sexes. The other organochlorine pesticides indicated no statistical differences between sexes, including the *pp*′DDE/*pp*′DDT ratio. The samples grouped according to age, showed that the third tertile was more contaminated for both sexes, indicating age as a positively associated factor with organochlorine pesticide levels in adipose tissue of Veracruz inhabitants. Comparing organochlorine pesticide levels between 2008 and 2010 years, a decreased tendency for β-HCH, *pp*′DDE, Σ-DDT and *pp*′DDE/*pp*′DDT ratio levels was observed.

Keywords Organochlorine pesticides · Adipose tissue · Human

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Recently, researchers have found that levels of organochlorine pesticides contaminate humans are lower than in previous years (Lucena et al. 2007). The new data provide the most comprehensive picture to date of exposure levels to these persistent organic pollutants. Concentrations of all organochlorine pesticides detected in monitored persons are lower than in the previous surveys and it is a trend that will continue (Mueller et al. 2008, Daglioglu et al. 2010, Waliszewski et al. 2010, Herrero-Mercado et al. 2010). Despite the statistical shortcomings, researchers are able to make some cautious comparisons over time. For example, levels of organochlorine compounds have declined in the overall population, especially in the younger part of the population, but have held steady in older people (Herrero-Mercado et al. 2010). This is consistent with today's younger people being exposed to lower levels of volatilized compounds from the environment (Lakind et al. 2009).

Many persistent compounds can still be detected in samples of the human body decades after they are banned. For example, DDT was banned in Mexico in 1999 but its breakdown product DDE and insecticide *pp*′DDT are still detected in human samples (Waliszewski et al. 2010). This continuing occurrence may result at least in part from inhaled vapors, which originated from superficial soils (Martínez-Salinas et al. 2011) and consumption of foods that contain organochlorine residues (Borchers et al. 2010). With the new data, researchers can better understand how and why organochlorine pesticide levels vary within certain populations. For example, lower total levels exist among Mexicans living in different geographic places in Mexico and when compared within sexes, levels were generally higher in females than in males (Waliszewski et al. 2010).

A number of studies have reported positive and negative associations between Body Mass Index (BMI) and organochlorine pesticide levels (Perry et al. 2005, Wolff et al. 2007). In the Veracruz population of pregnant women (Herrero-Mercado et al. 2010) this relationship was negative. In epidemiological studies evaluating the health effects of organochlorine pesticides, statistical approaches considered BMI as a potential confounder (Bradman et al. 2007). Pharmacokinetic models seeking to quantify the effects of BMI, time of exposure and elimination rate of organochlorine pesticides support the importance of age of a person as a contributing factor to age-related increases of organochlorine pesticide levels among populations where these pesticides have been used relatively recently or where population exposures are rapidly declining (Wolff et al. 2007).

Estimated dietary intake of DDT has dropped significantly during the last 20 years. Environmental and biological monitoring studies in the United States demonstrate that use restrictions were successful in lowering human exposure to DDT (Patterson et al. 2009), however, nearly all U.S. residents have measurable serum *pp*′DDE levels, whereas *pp*′DDT is detected in only 5–10% of the population (Eskenazi et al. 2009). Recent studies in Mexico report low concentrations of DDT and DDE in vegetables and foods of animal origin (Waliszewski et al. 2003a, 2003b, 2004a, 2004b, 2008).

Few reports are available that establish the extent of organochlorine pesticide residue levels in areas where they were applied only for malaria or ectoparasites control. Moreover, there are few studies that implicate sanitary actions as a principal source of human exposure. The aim of this study was to determine organochlorine pesticide levels in human adipose tissue of Veracruz inhabitants in order to establish biomarkers of environmental exposure to organochlorine pesticides.

Materials and Methods

Human adipose samples (approximately 5 g) from abdominal cavities of 150 randomly selected persons (75 females and 75 males) were taken during 2010 by autopsy at the Forensic Medicine Services of Veracruz. The donors originated principally from Veracruz City and from the state of Veracruz. The samples were stored in glass jars previously washed with sulfuric acid, immediately frozen, and kept at –25°C until analyzed. The dated samples were labeled with donor data.

The analyses of organochlorine pesticide residues in human adipose tissue were performed by gas chromatography with ECD according to a method previously described (Waliszewski et al. 2004c; 2010). All of the samples were analyzed for HCB, α, β, γ-HCH, *pp*′DDT, *op*′DDT and *pp*′DDE. The minimum detection limits for the residues analyzed were as follows: 0.001 mg/kg for HCB, 0.002 mg/kg for the HCH isomers and *pp*′DDE, and 0.003 mg/kg for *pp*′DDT and *op*′DDT. To determine the quality of the method, a recovery study was performed on 10 spiked replicates of blank cow fat samples, which presented contamination levels below the detection limits. The fortification study, done at 0.01–0.03 mg/kg levels, depending on the pesticide, showed mean values for recovery from 90 to 95%. The standard deviation and coefficient of variation were below 10, indicating excellent repeatability of the method.

Statistical calculations were conducted using the statistical software Minitab Version 12. Concentrations of organochlorine pesticide (mg/kg on fat base) were expressed as frequencies, arithmetic means, medians and geometric means (GM). The significance of categorical factors on pesticide levels was determined by: 1) using the variability among samples; 2) pairing them to identify differences among means by applying the Student's t-test and; 3) using differences among medians by applying the Mann–Whitney test at $\alpha = 0.05$.

Results and Discussion

The mean age of the monitored population was 44.8 years (range 15–86). Divided according to sex, the mean for females was 44.8 years (range 25-86), and for males it was 44.6 years (range 15–83). Comparing both sexes, no statistically significant mean age difference ($p > 0.05$) was observed. The median age was 42 years for the total population and for the female and male population separately it was also 42 years for each.

During the monitoring study, only the presence of β-HCH, *pp*′DDE, *op*′DDT and *pp*′DDT were determined, thus only these compounds are discussed. Table 1 summarizes results from 150 adipose tissue samples expressed as frequencies, ranges, mean ± standard deviations of mean (SD), median and geometric mean (GM) levels, all expressed on lipid base (mg/kg) of organochlorine pesticides. *pp*′DDE was found in 100% of the samples analyzed, whereas *pp*′DDT, *β*-HCH and *op*′DDT were presented in 99.3, 97.3 and 93.3%, respectively, of the samples.

pp′DDE was found at a higher mean concentration of 1.643 mg/kg on lipid base. This compound is followed by the insecticide *pp*′DDT that had a mean concentration of 0.227 mg/kg. The results for *β*-HCH and *op*′DDT indicated lower concentrations of 0.063 and 0.022 mg/kg, respectively. A comparison of mean, median and geometric mean concentrations of organochlorine pesticides shows a decrease of values from mean to median and to geometric mean, which points out a prevalence of lower concentrations among the total samples and the existence of occasional cases of extreme exposure that are indicated by the range values. The calculated *pp*′DDE/*pp*′DDT ratios, which reflect the age of exposure, reveal a mean value of 13.53 that decreases when expressed as a median and GM. The ratio indicates antique exposures and the predominance of *pp*′DDE, which is a metabolite of *pp*′DDT, as the principal contaminant for the Veracruz inhabitants. The data presented in Table 1 indicates the existence in Veracruz of specific points of contamination with DDT and the accumulation of the compound in the human body by inhalation of contaminated air and consumption of contaminated foods associated with specific alimentary habits.

To observe the possible influence of sex as a discriminatory factor for organochlorine pesticide levels, the pooled sample was divided according to the sex of donors (75 female, Table 2; and 75 male, Table 3). In order to look for differences in organochlorine pesticide levels between sexes, the samples were paired. The results demonstrate only significantly higher male levels of the *pp*′DDT median value ($p = 0.047$). Other organochlorine pesticide concentrations indicated no statistical differences ($p > 0.05$) between sexes, including the *pp*′DDE/*pp*′DDT ratios. In general, during the monitoring study, sex was not observed to be a determinant factor for organochlorine pesticide contamination levels. This fact points to a uniform exposure for inhabitants, caused only by living in the urban Veracruz environment.

To determine if organochlorine pesticide levels in monitored human adipose tissue can depend on the age of the monitored person, each sex group (female and male) was divided according to age into an ordered distribution of three parts, each containing a third of the population, and then the frequencies, mean and median tertiles of pesticide levels were calculated (Table 4). In the female group, *β*-HCH levels increase from first to second and third tertile, and means and medians are statistically different ($p < 0.05$). The increase of mean values from the first to third tertile for *pp*′DDE, *op*′DDT, *Σ*-DDT and *pp*′DDE/

Table 1 Organochlorine pesticide levels (mg/kg) in human adipose tissue from Veracruz inhabitants (n = 150)

Pesticide	Frequency	Ranges	Mean ± SD	Median	GM
β-HCH	146/150	0.001–0.443	0.063 ± 0.078	0.037	0.019
pp′DDE	150/150	0.042–7.564	1.643 ± 1.465	1.233	1.071
op′DDT	140/150	0.002–0.160	0.022 ± 0.022	0.016	0.033
pp′DDT	149/150	0.019–3.502	0.227 ± 0.488	0.086	0.134
Σ-DDT		0.099–9.200	1.887 ± 1.762	1.323	1.248
pp′DDE/*pp*′DDT		0.88–45.83	13.53 ± 9.62	12.18	10.23
Age		15–86	44.8 ± 14.2	42	42.5

Table 2 Organochlorine pesticide levels (mg/kg) in female adipose tissue (n = 75)

Pesticide	Frequency	Ranges	Mean ± SD	Median	GM
β-HCH	74/75	0.002–0.443	0.070 ± 0.083	0.039	0.036
pp′DDE	75/75	0.098–7.564	1.497 ± 1.489	1.105	0.936
op′DDT	70/75	0.002–0.160	0.021 ± 0.042	0.016	0.097
pp′DDT	75/75	0.019–2.957	0.207 ± 0.419	0.076	0.097
Σ-DDT		0.164–9.200	1.723 ± 1.745	1.175	1.104
pp′DDE/*pp*′DDT		0.88–40.92	13.51 ± 9.82	12.52	9.67
Age		25−86	44.6 ± 12	42	43

Table 3 Organochlorine pesticide levels (mg/kg) in male adipose tissue (n = 75)

Pesticide	Frequency	Ranges	Mean ± SD	Median	GM
β-HCH	72/75	0.001–0.365	0.056 ± 0.073	0.034	0.026
pp′DDE	75/75	0.042–6.497	1.790 ± 1.436	1.336	1.225
op′DDT	70/75	0.006–0.128	0.024 ± 0.022	0.017	0.018
pp′DDT	74/75	0.030–3.502	0.247 ± 0.551	0.106	0.121
Σ-DDT		0.099–8.179	2.051 ± 1.774	1.488	1.410
pp′DDE/*pp*′DDT		1.01–45.83	13.55 ± 9.49	12.04	10.32
Age		15−83	45 ± 16	42	42

Table 4 Organochlorine pesticide levels (mg/kg) in tertiles according to age for female and male groups

	First			Second			Third		
Pesticide	Freq	X ± SD	Median	Freq	X ± SD	Median	Freq	X ± SD	Median
Female									
β-HCH	25/25	0.035 ± 0.042	0.026	25/25	0.086 ± 0.079	0.058	24/25	0.089 ± 0.107	0.044
pp′DDE	25/25	1.012 ± 0.737	0.829	25/25	1.447 ± 1.022	1.348	25/25	2.030 ± 2.168	1.110
op′DDT	22/25	0.014 ± 0.007	0.013	24/25	0.019 ± 0.013	0.017	24/25	0.027 ± 0.034	0.015
pp′DDT	25/25	0.194 ± 0.298	0.064	25/25	0.179 ± 0.330	0.094	25/25	0.249 ± 0.585	0.071
Σ-DDT		1.219 ± 0.967	0.921		1.646 ± 1.190	1.598		2.305 ± 2.533	1.175
DDE/*pp*′DDT		10.30 ± 8.47	8.02		14.39 ± 9.60	13.22		15.85 ± 10.77	14.29
Age		32.6 ± 4.0	33.0		42.3 ± 1.9	42.0		58.8 ± 9.4	56.0
Male									
β-HCH	23/25	0.025 ± 0.042	0.010	25/25	0.044 ± 0.051	0.027	24/25	0.099 ± 0.094	0.055
pp′DDE	25/25	1.090 ± 1.366	0.699	25/25	2.037 ± 1.433	2.184	25/25	2.242 ± 1.283	1.897
op′DDT	21/25	0.026 ± 0.026	0.016	25/25	0.022 ± 0.024	0.015	24/25	0.023 ± 0.018	0.019
pp′DDT	24/25	0.146 ± 0.306	0.059	25/25	0.294 ± 0.686	0.104	25/25	0.296 ± 0.592	0.143
Σ-DDT		1.254 ± 1.660	0.750		2.353 ± 1.800	2.255		2.546 ± 1.642	2.073
DDE/*pp*′DDT		10.46 ± 6.97	8.00		14.13 ± 9.63	12.42		15.95 ± 10.89	13.21
Age		28.1 ± 6.1	28.0		43.4 ± 6.2	42.0		63.6 ± 6.9	62.0

Table 5 Comparison of the organochlorine pesticide levels (mg/kg) in human adipose tissue between 2008 and 2010

Pesticide	Mean ± SD 2008	Mean ± SD 2010	Median 2008	Median 2010
β-HCH	0.072 ± 0.126	0.063 ± 0.078	0.029	0.037
pp′DDE	2.364 ± 1.953*	1.643 ± 1.465*	1.829*	1.233*
op′DDT	0.022 ± 0.017	0.022 ± 0.022	0.020	0.016
pp′DDT	0.192 ± 0.219	0.227 ± 0.488	0.114*	0.086*
Σ-DDT	2.589 ± 2.104*	1.887 ± 1.762*	2.006*	1.323*
pp′DDE/*pp*′DDT	15.63 ± 12.8	13.53 ± 9.62	13.25	12.18
Age	42.9 ± 17.4	44.8 ± 14.2	41	42

* significant differences

pp′DDT ratio were statistically significant ($p < 0.05$), whereas the median values among three tertiles and the increase in the levels were not statistically significant ($p > 0.05$). In the male group the increases for the mean and median β-HCH, *pp*′DDE, *pp*′DDT, Σ-DDT and *pp*′DDE/*pp*′DDT ratio from the first to third tertile were statistically significant ($p < 0.05$), whereas the increases from the second to third tertile were not significant

Table 6 Comparison of the organochlorine pesticide levels (mg/kg) in female adipose tissue between 2008 and 2010

Pesticide	Mean ± SD 2008	Mean ± SD 2010	Median 2008	Median 2010
β-HCH	0.129 ± 0.211*	0.070 ± 0.083*	0.069*	0.039*
pp′DDE	2.494 ± 2.184*	1.497 ± 1.489*	1.767*	1.105*
op′DDT	0.020 ± 0.010	0.020 ± 0.022	0.018	0.014
pp′DDT	0.200 ± 0.196	0.207 ± 0.420	0.123*	0.076*
Σ-DDT	2.736 ± 2.348*	1.723 ± 1.745*	1.889*	1.175*
pp′DDE/*pp*′DDT	14.92 ± 11.65	13.51 ± 9.82	12.41	12.52
Age	44.1 ± 13.9	44.6 ± 12.4	44	42

* significant differences

Table 7 Comparison the organochlorine pesticide levels (mg/kg) in male adipose tissue between 2008 and 2010

Pesticide	Mean ± SD 2008	Mean ± SD 2010	Median 2008	Median 2010
β-HCH	0.051 ± 0.064	0.056 ± 0.072	0.026	0.034
pp′DDE	2.321 ± 1.887*	1.790 ± 1.436*	1.879*	1.336*
op′DDT	0.023 ± 0.019	0.024 ± 0.022	0.020	0.017
pp′DDT	0.189 ± 0.228*	0.247 ± 0.552*	0.113	0.106
Σ-DDT	2.539 ± 2.036*	2.051 ± 1.774*	2.074*	1.488*
pp′DDE/*pp*′DDT	15.86 ± 12.30	13.55 ± 9.49	13.63	12.04
Age	42.5 ± 18.5	45.1 ± 15.9	40	42

* significant differences

($p > 0.05$). In conclusion, the organochlorine pesticides accumulated during the lifetime reveals higher concentrations in human adipose tissue in older monitored persons.

The results from analyzing the data to compare organochlorine pesticide levels between year 2008 (Waliszewski et al. 2010) and this study are presented in Table 5. A decreased tendency for β-HCH, *pp*′DDE, Σ-DDT and the *pp*′DDE/*pp*′DDT ratio in the 2010 study and an increase in mean and decrease in median *pp*′DDT levels can be observed. The ages of monitored persons in the two groups were not statistically different for means and medians, and the groups are considered as equal.

Both monitored groups (2008 and 2010) divided according to sex, indicated for female (Table 6), a statistically significant decrease ($p < 0.05$) of mean and median β-HCH, *pp*′DDE and Σ-DDT values, whereas *op*′DDT mean and median and mean *pp*′DDT levels were not different ($p > 0.05$). *pp*′DDT median concentrations decreased significantly.

Analyzing Table 7 for males, there is a decreased tendency for both periods for *pp*′DDE mean and median, *pp*′DDT mean and Σ-DDT mean and median, with concentration differences that are statistically significant ($p < 0.05$). Other organochlorine pesticides concentrations showed no statistically significant differences ($p > 0.05$).

In conclusion, during the monitoring study of adipose tissue from Veracruz inhabitants, the presence of organochlorine pesticide residues are still observed. Differences in concentrations related to sex of donors, in general were not significant. The higher concentrations of organochlorine pesticides correspond to older persons. These persons are environmentally and alimentary exposed to organochlorine pesticides that accumulate in adipose tissue and are stored during a lifetime. Analyzing the time trend from 2008 to 2010, a decrease of mean and median concentrations was noted, indicating a gradual disappearance of these compounds from the Veracruz environment since they were banned in 1999 in Mexico.

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