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Levels of Organochlorine Pesticides Residues in Human Adipose Tissue, Data from Tabasco, Mexico

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Abstract The objective of this study was to determine the levels of organochlorine pesticides HCB, α-β-γ-HCH, *p,p′*-DDE, *o,p′*-DDT and *p,p′*-DDT in 150 adipose tissue of inhabitants of Tabasco, Mexico. The following pesticides were detected: *p,p′*-DDE in 100 % of samples at mean 1.034 mg/kg; *p,p′*-DDT in 96.7 % at mean 0.116 mg/kg; *o,p′*-DDT in 78.7 % at mean 0.022 mg/kg and β-HCH in 58.0 % at mean 0.049 mg/kg. The pooled sample was divided according to sex of donors (75 female and 75 male). Significantly higher levels of all organochlorine pesticides in females were found. The sample was divided into three age's ranges (15–28, 29–45 and 46–84 years). The mean and median levels of β-HCH, *p,p'*-DDE and Σ-DDT increase significantly ($p < 0.05$) from the first to the second and third group. The presence of organochlorine pesticide residues in Tabasco inhabitants is still observed, indicating sources of exposure to the pesticides.

Keywords Organochlorine pesticides · Adipose tissue · Tabasco Mexico

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The use of organochlorine pesticides caused human exposure to their residues through different routes, which have been identified by environmental and human monitoring studies (Lucena et al. 2007; Aulakh et al. 2007). Organochlorines are a group of pesticides that have provided great benefits in the eradication of various pest diseases in agriculture and in the elimination of vectors transmitting mortal diseases for humans. Used for these purposes in Mexico until 1999, DDT (1,1,1-trichloro-2,2-bis(4-chlorophenyl) ethane) and HCH (hexachlorocyclohexane) have been sprayed extensively in public health programs to combat the spread of disease-transmitting vectors. These pesticides are characterized by chemical stability, a lipophilic nature and a propensity to bioaccumulation in the environment and food chain. Due to their persistence long after application and susceptibility to long-range transport routes, they remain in the environment as ubiquitous contaminants (Daba et al. 2011). Sprayed pesticides volatilize and move from particles on the ground and surface soils, exposing humans that have not lived in contaminated areas or manipulated these compounds (Martınez-Salinas et al. 2011; Santiago and Cayetano 2011). Another exposure source is consumption of foods that contain organochlorine residues (Borchers et al. 2010). DDT, banned in developed countries, is still used today in many countries where malaria is a public health problem, such as in Africa, Asia, and Latin America (Anonymous WHO 2006). Consistent with this usage history, populations of these countries generally should have higher tissue residue levels than those in Europe and the USA (Dhananjayan et al. 2012).

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 tend to decrease, compared to the previous surveys and this trend will continue

(Daglioglu et al. 2010; Waliszewski et al. 2010; Herrero-Mercado et al. 2010). They have declined in the overall population, especially in the younger part of the population, which is exposed to lower concentrations of pesticides for a shorter period, but have held steady in older people (Herrero-Mercado et al. 2010; Waliszewski et al. 2011). Nevertheless, decades after they were banned, they are still detected in samples of the human body. For example, DDT was banned in Mexico in 1999 but its breakdown product DDE and insecticide *p,p′*-DDT are still detected in human samples (Waliszewski et al. 2011). 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 and climatic places in Mexico and when compared between sexes, levels were generally higher in males than in females (Waliszewski et al. 2011).

Due to its high lipid affinity, organochlorine pesticides are stored in the fats of tissues of most organs (Waliszewski et al. 2003). They enter the circulatory system and are transported via the lipid and protein components of blood serum and deposit in adipose tissue according to the partition coefficients between blood and adipose tissue lipids (Herrero Mercado et al. 2011). The determination of serum and adipose tissue levels are commonly used as biomarkers of exposure in monitoring studies evaluating the concentrations related to regional and country variability (Sawada et al. 2010; Eskenazi et al. 2009).

The state of Tabasco is located in the southeast of Mexico, bordering the states of Campeche, Chiapas and Veracruz with the Gulf of Mexico to the north and the country of Guatemala to the south and east. The environment of the state consists of extensive low lying floodplains, mountains and valleys. Most of the territory is covered in tropical rainforest and wetlands. There are four principle ecosystems in the state: tropical rainforest, tropical savannah, beaches and wetlands. Tabasco has a hot tropical climate, with the Gulf of Mexico having significant influence on weather patterns. Over 95 % of the state's territory has a hot, wet climate. The rest is hot and semi-humid, and is located in the far northeast of the state. The average annual temperature is 27°C with high temperatures averaging 36°C, mostly in May, and lows of 18.5°C, which occur in January. Unlike many parts of Mexico, Tabasco has abundant year-round precipitation. Rain occurs all year but is particularly heavy from June to October. The flat areas of the state are subject to frequent flooding, thus these areas are endemic spaces for malaria. To prevent its propagation, they were treated with DDT at a rate of 2 g/m^2 from 1956 to 1999. The subsequent human contamination is influenced by local environmental pollution, duration of exposure, age, diet, capacity for elimination by metabolism, and number of nursed infants (Czaja et al. 1997; Laden et al. 1999). Thus, the monitoring of human adipose tissue from Tabasco serves as an indicator for understanding the biological specificity in the behavior of organochlorine pesticides in the tropical humid environment, as well as for assessing their participation in environmental pollution.

Materials and Methods

During 2011 and 2012, 150 human adipose tissues were taken by autopsy carried out at the Medical Forensic Services of Villahermosa, Tabasco. The donors had died from natural or accidental causes. All participants' families were asked for consent to participate in the study and donate the adipose tissue sample for the monitoring study. The dated adipose tissue samples were labeled with donor origin, sex and age. The samples were stored in glass jars, immediately frozen, and kept at −25°C until analysis.

The organochlorine pesticide residue determinations were performed according to a previously described method (Waliszewski et al. 2010). All of the samples were analyzed for HCB, α, β, γ-HCH, *p,p′*-DDT, *o,p′*-DDT, and *p,p′*-DDE. The minimum detection limits for the residues were as follows: 0.001 mg/kg for HCB; 0.002 mg/kg for the α-β-γ-HCH isomers and *p,p′*-DDE; and 0.003 mg/kg for *p,p′*-DDT and *o,p′*-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 from 88 % to 93 % recovery.

Concentrated sulfuric acid used in the cleanup step degrades the ubiquitous phthalate esters that interfere in the GC-ECD identification of organochlorine pesticides, permitting their accurate determination (Waliszewski et al. 2008).

Gas chromatography was conducted with a Varian model 3800 (Palo Alto, CA, USA) equipped with a ^{63}Ni electron-capture detector. The operating conditions were as follows: the capillary chromatography column from J&W Scientific (Folsom, CA, USA) was a DB-608 with a 30-m, 0.32-mm inner diameter (i.d.) and 0.83-µm film thickness; the temperature program was 193°C (7 min) to 250°C at 6°C/min, hold for 20 min; the carrier gas was nitrogen at 6.3 mL/min and a 1 µL aliquot was injected in a splitless mode.

Statistical calculations were conducted using statistical software Minitab version 12. Concentrations of organochlorine pesticide (milligrams per kilogram on fat base) were expressed as frequencies, arithmetic means

(X) ± standard deviations (SD), medians, and geometric means (GM). The resulting concentrations were used to determine the significance of categorical factors on pesticide levels by the variability among samples, pairing them to identify differences among means by applying the Student′s t test and among medians by the Mann–Whitney test. These tests were applied between sexes and after dividing these groups into tertiles to associate sex and age as a determinant factor of exposure.

Results and Discussion

Routine biomonitoring may be desirable for the efficiency valuation of risk management options and efficacy of environmental and health policies. For organochlorine pesticides, average exposures may not reflect peak exposures arising through infrequent exposure episodes. Invasively collected matrices such as adipose tissue for human biomonitoring are toxicologically relevant alternatives for many of the biomarkers currently determined. Moreover, a well-informed choice of matrix can provide an added value for human biomonitoring, offering opportunities to study additional aspects of exposure to organochlorine pesticide residues and effects of short- and long-term toxicokinetics, the change of exposure over time, or the monitoring of selected communities (Smolders et al. 2009; Waliszewski et al. 2010).

During the study, only the presence of *β*-HCH, *p,p′*-DDE, *o,p′*-DDT and *p,p′*-DDT were detected, thus only these compounds are discussed. Table 1 summarizes results from 150 adipose tissue samples expressed as frequency, mean ± standard deviations of mean (SD), median and geometric mean (GM), all expressed on lipid base (mg/kg) of organochlorine pesticides. *p,p′*-DDE was found in 100 % of the samples analyzed, whereas *p,p′*-DDT, *o,p′*-DDT and *β*-HCH were presented in 96.7 %, 78.7 % and 58.0 %, respectively.

p,p′-DDE was found at a higher mean concentration of 1.034 mg/kg on lipid base. This metabolite of insecticide *p,p′*-DDT is followed by its maternal compound *p,p′*-DDT that had a mean concentration of 0.116 mg/kg. The results for *β*-HCH and *o,p′-DDT* indicated lower concentrations of 0.049 and 0.022 mg/kg, respectively. A comparison of results 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 influences the mean values presented. The interpretation of DDT′s concentrations indicates antique exposures to DDT and the predominance of *p,p′*-DDE as the principal contaminant for the Tabasco inhabitants. The results reveal the existence in Tabasco of specific points of extreme levels of contamination with DDT and the accumulation of the compound in the human body originating from a contaminated environment.

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

Pesticide	Frequency (%)	X ± SD	Median	GM
β-HCH	58.0	0.049 ± 0.145	0.012	0.012
p,p′-DDE	100.0	1.034 ± 0.864	0.877	0.535
o,p′-DDT	78.7	0.022 ± 0.033	0.015	0.015
p,p′-DDT	96.7	0.116 ± 0.171	0.058	0.058
Σ-DDT		1.164 ± 0.967	0.969	0.714
Age		39.0 ± 15.8	36.0	35.9

To observe the possible influence of sex as a discriminatory factor for organochlorine pesticide levels, the pooled sample was divided according to sex of donors (75 female and 75 male, Table 2). To look at differences in organochlorine pesticide levels between sexes, the samples were paired. The results demonstrate statistically significant ($p < 0.05$) higher mean and median levels in females for all organochlorine pesticides, which correlate with levels determined in Turkey (Daglioglu et al. 2010). The results are surprising when compared to previous monitoring studies conducted in Puebla and Veracruz (Waliszewski et al. 2010; 2012) where due to male alimentary habits, higher organochlorine pesticide levels in males were detected. In general, during the study, sex of Tabasco residents was a determinant factor for organochlorine pesticide contamination levels. This fact points to different exposures and accumulations of these organochlorine pesticides in the bodies of residents exposed to the Tabasco environment and by the consumption of foods that contains the residues.

Epidemiological studies evaluate the health effects of organochlorine pesticides. They quantify the effects of time of exposure and elimination rate to support the importance of age of a person as a contributing factor to age-related increases or permanence of the levels among populations where these pesticides have been used relatively recently or where population exposures are rapidly declining (Wolff et al. 2007). Thus, to determine if organochlorine pesticide levels in monitored human adipose tissues are dependent on the age of the monitored person, the pooled sample was divided according to age into an ordered distribution of three parts, each containing a third of the population, and mean, standard deviation of mean and median tertiles of pesticide levels were calculated (Table 3). The *β*-HCH and *p,p′*-DDE levels increase from first to second and to third tertile, and mean and medians are statistically different ($p < 0.05$) (Fig. 1). The increase of concentrations for insecticide *p,p′*-DDT was noted, but the increase was not

Table 2 Comparison of organochlorine pesticide levels (mg/kg on fat basis) between sexes (M–male, F–female)

Pesticide	Frequency (%)	Ranges	X ± DE	Median
β-HCH	M 34/75 = 45.3	0.002–0.091	0.019 ± 0.023*	0.009
	F 53/75 = 70.7	0.002–1.067	0.068 ± 0.183*	0.017
p,p′-DDE	M 75/75 = 100	0.050–5.007	0.872 ± 0.837*	0.605*
	F 75/75 = 100	0.014–3.665	1.196 ± 0.866*	1.044*
o,p′-DDT	M 56/75 = 74.7	0.003–0.056	0.014 ± 0.012*	0.011*
	F 62/75 = 82.7	0.003–0.346	0.029 ± 0.043*	0.021*
p,p′-DDT	M 73/75 = 97.3	0.005–0.691	0.083 ± 0.117*	0.034*
	F 72/75 = 96.0	0.004–0.994	0.149 ± 0.207*	0.082*
Σ-DDT	M	0.055–5.089	0.963 ± 0.910*	0.630*
	F	0.017–4.088	1.365 ± 0.986*	1.142*
Age	M	15–79	40.9 ± 14.3	41.0
	F	15–84	37.1 ± 17.0	40.0

* Differences statistically significant ($p < 0.05$)

Table 3 Organochlorine pesticide levels (mg/kg on fat basis) in tertiles according to age of participants

Pesticide	First tertil 15–28		Second tertil 29–45		Third tertil 46–84	
	X ± SD	Median	X ± SD	Median	X ± SD	Median
β-HCH	0.009 ± 0.012*	0.003*	0.088 ± 0.203	0.017*	0.065 ± 0.136*	0.031*
p,p′-DDE	0.735 ± 0.532*	0.648*	0.837 ± 0.703*	0.736*	1.529 ± 1.055*	1.348*
*o,p′*DDT	0.020 ± 0.012	0.020	0.028 ± 0.056	0.014	0.019 ± 0.014	0.014
*p,p′*DDT	0.108 ± 0.167	0.057	0.117 ± 0.189	0.050	0.123 ± 0.160	0.060
Σ-DDT	0.855 ± 0.730*	0.657*	0.968 ± 0.840*	0.775*	1.668 ± 1.144*	1.412*

* Differences statistically significant ($p < 0.05$)

statistically significant ($p > 0.05$) (Fig. 1). Although the existence of an especially contaminated person′s independence of age can be observed. o,p′-DDT changes within three tertiles were not ordered and not significant. Σ-DDT levels, due to predominance of *p,p′*-DDE, increase statistically significant ($p < 0.05$) with age. In conclusion, the more persistent organochlorine pesticides accumulate during the lifetime, especially *β*-HCH and *p,p′*-DDE, in which concentrations in human adipose tissue increase with the age of monitored persons. Due to the prohibition of use of the pesticide DDT since 1999 in Mexico, concentrations of the *p,p′*-DDT and its isomer *o,p′-DDT* remain stable without any significant fluctuations among three age groups. From the results its gradual decrease in concentrations and final disappearance from Mexican environment can be prognosticated.

The results from analyzing the data to compare organochlorine pesticide levels between Veracruz population (Waliszewski et al. 2011) and this study are presented in Table 4. Minor concentrations of all organochlorine

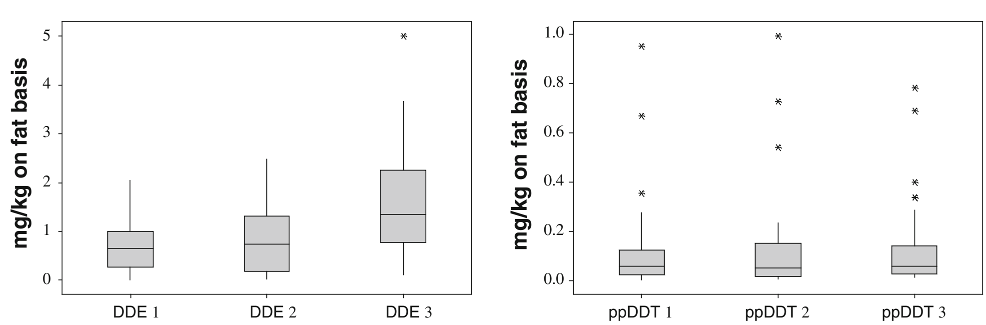

Fig. 1 Fluctuation of *p,p′*-DDE and *p,p′*-DDT levels (mg/kg on fat basis) in tertiles according to age of participants

Table 4 Comparison of organochlorine pesticide levels (mg/kg on fat basis) between Tabasco and Veracruz inhabitants

Pesticide	X ± SD Tabasco	Median Tabasco	X ± SD Veracruz	Median Veracruz
β-HCH	0.049 ± 0.145	0.012*	0.056 ± 0.072	0.034*
p,p′-DDE	1.034 ± 0.864*	0.877*	1.790 ± 1.436*	1.336*
o,p′-DDT	0.022 ± 0.033	0.015	0.024 ± 0.022	0.017
p,p′-DDT	0.116 ± 0.171*	0.058*	0.247 ± 0.552*	0.106*
Σ-DDT	1.164 ± 0.967*	0.969*	2.051 ± 1.774*	1.488*
Age	39.0 ± 15.8	36.0	45.1 ± 15.9	42

* Differences statistically significant ($p < 0.05$)

pesticides in the Tabasco population are noted. When the statistical test was applied to compare differences among means and medians, only *o,p′*-DDT showed no difference in concentrations ($p > 0.05$). Other organochlorine pesticides in Tabasco inhabitants are lower than that of Veracruz inhabitants. The data reveals higher Veracruz inhabitants exposure to these pesticides.

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