Bull Environ Contam Toxicol (2011) 86:289–293
DOI 10.1007/s00128-011-0204-4

Organochlorine Pesticide Gradient Levels Among Maternal Adipose Tissue, Maternal Blood Serum and Umbilical Blood Serum

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Received: 6 October 2010 / Accepted: 24 January 2011 / Published online: 3 February 2011

Abstract The objective of the present study was to determine levels and calculate ratios of copartition coefficients among organochlorine pesticides *β*-HCH, *pp*′DDE, *op*′DDT and *pp*′DDT in maternal adipose tissue, maternal blood serum and umbilical blood serum of mother-infant pairs from Veracruz, Mexico. Organochlorine pesticides were analyzed in 70 binomials: maternal adipose tissue, maternal serum and umbilical cord serum samples, using gas chromatography with electron capture detection (GC-ECD). The results were expressed as mg/kg on fat basis. *p,p*′-DDE was the major organochlorine component, detected in every maternal adipose tissue (0.770 mg/kg), maternal serum sample (5.8 mg/kg on fat basis) and umbilical cord blood sample (6.9 mg/kg on fat basis). *p,p*′-DDT was detected at 0.101 mg/kg, 2.2 mg/kg and 5.9 mg/kg respectively, according to the order given above. *β*-HCH was detected at 0.027 mg/kg, 4.2 mg/kg and 28.0 mg/kg respectively. *op*′DDT was detected only in maternal adipose tissue at 0.011 mg/kg. The copartition coefficients among samples identify significant increases in concentrations from adipose tissue to maternal blood serum and to umbilical blood serum. The increase indicated that maternal adipose tissue released organochlorine pesticides to blood serum and that they are carried over to umbilical cord blood.

Keywords Organochlorine pesticides · Adipose tissue · Blood

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The discovery of insecticide properties of DDT (1,1,1-trichloro-2,2-bis(4-chorophenyl)ethane) and HCH (hexachlorocyclohexane), that occurred over a half century ago, provided great benefits by protecting plants from pests and combating the spread of disease-transmitting organisms. Since 1956, Mexico has been spraying DDT and other organochlorine pesticides in sanitary actions to combat malaria. Since 1999, due to the persistence of and inhabitant's exposure to DDT, the Public Health Program in Mexico has replaced DDT and organochlorine pesticides with pyrethroids.

The uses of DDT and HCH throughout the decades lead to investigations of the magnitude of their residues in all elements of the human environment. These pesticides with low or intermediate volatility, when deposited upon superficial soils, volatilize and tend to distribute uniformly throughout the environment (Alegria et al. 2008; Wong et al. 2008). The monitoring studies revealed the permanence of organochlorine pesticide residues in all elements of the environment, a fact that led the governments of many countries to ban DDT and restrict the maximum accepted residue levels in foodstuffs. Since 2005, due to the reappearance of malaria, the World Health Organization has again recommended DDT as the insecticide of choice for malaria vector combat in tropical areas.

Human beings are part of the environment and are contaminated with the same chemicals which are present in surrounding air, water, food, consumer articles, etc. The exposure to organochlorine pesticide residues occurs as a consequence of their persistence in soil, air, plants and food. They accumulate in superficial soils, volatilize from the deposition and expose inhabitants to their vapors (Alegria et al. 2008; Wong et al. 2008). Another source of exposure is food consumption principally of animal origin that previously accumulated and metabolized the residues of organochlorine pesticides. The lipid-rich tissues accumulate lipophilic environmental pollutants by virtue of physiological cellular component interactions, identifying them as bioindicators of body burden. Thus, the determination of levels in human tissues, such as adipose or blood serum, can reflect the magnitude of local environment pollution (Waliszewski et al. 2010).

Environmental exposure to persistent organochlorines can provide an internal dose to the female reproductive tract, as these compounds have been found in human adipose tissue, maternal blood serum and umbilical cord serum, providing evidence of exposure to critical tissues during important windows of early development (Meeker et al. 2009). Of special concern are the potential effects of organochlorine pesticide on the fetus caused by a higher sensitivity to environmental toxicants compared to adults. The ability to assess fetus exposures to environmental chemicals via the transplacental route is crucial for developing a complete picture of early-life exposures to those chemicals (Sapbamrer et al. 2008). Conventional wisdom regarding environmental exposures to persistent pesticides presumes declines in concentrations (expressed as wet weight) over time and that the origin of those chemicals lies in a mother's cumulative exposure over her lifetime. The important implications stemming from these lines of thought are that assessments of childhood exposures during the first part of life should incorporate the carrying over of mother—fetus concentrations.

The objective of the present study was to determine and compare the levels of organochlorine pesticides in maternal adipose tissue, maternal blood serum and umbilical blood serum taken during cesarean delivery and compare the concentrations among the sample groups and calculate the copartition coefficients among compartments.

Materials and Methods

Seventy selected volunteer mothers admitted for delivery during 2009 were asked to donate about 5 g of abdominal adipose tissue, about 5 mL of blood and umbilical cord blood for determination of organochlorine pesticide levels. The donors of mean 27 years old originated from the state of Veracruz (Herrero Mercado et al. 2010a, 2010b). The volunteers signed their consent to participate in the study. The maternal blood samples were taken before cesarean delivery and the umbilical cord blood samples were taken after it was cut. The blood samples were then centrifuged to separate blood cells from the serum. The samples were stored in glass jars, immediately frozen, and kept at −25°C until analyzed.

Chemical analyses of organochlorine pesticides were performed according to a previously detailed method (Waliszewski et al. 2004a). Gas chromatographic determinations of selected organochlorine pesticides (HCB, α-,β-,γ-HCH, *pp*′DDE, *op*′DDT and *pp*′DDT) were conducted with a Varian model 3400CX gas chromatograph equipped with a ^{63}Ni electron-capture detector. The operating conditions were as follows: separations were done on capillary column J&W 30-m × 0.32-mm inner diameter and 0.83 μm film thickness, the temperature program was 193 (7 min)–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. The minimum detection limits for the residues analyzed were as follows: for adipose tissue 0.002 mg/kg on fat basis; for blood serum samples, 0.2 mg/kg on fat basis and 0.1 μg/L on wet weight. To determine the quality of the methods, a recovery study was performed on 10 spiked replicates of blank cow adipose tissue and blood samples, which presented contamination levels below the detection limits. The fortification study was done at 0.05 mg/kg for adipose tissue and for blood serum samples at 2.0 mg/kg on fat basis and 1.0 μg/L on wet weight and showed mean values from 83 to 94% for recovery. The standard deviation and coefficient of variation were below 10, indicating excellent repeatability of the method. Total serum lipids were determined calorimetrically with phosphovanillin according to the method recommended by Hycel de Mexico using a commercial kit for clinical laboratories.

Statistical calculations were conducted using statistical software Minitab version 12. Concentrations of organochlorine pesticide (mg/kg on fat base and μg/L on wet weight) were expressed as frequencies, arithmetic means and medians. The resulting concentrations were then used: (1) 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 differences among medians by applying the Mann–Whitney test at $\alpha = 0.05$ and (2) To calculate the copartition factors for maternal adipose tissue—maternal blood serum and umbilical blood serum and maternal blood serum—umbilical blood serum.

Results and Discussion

Lipophilic organochlorine pesticides are considered to have the ability to transfer through the placental barrier. The equilibrium models for these pesticides in the human body during pregnancy describe the partitioning rate between maternal compartments and fetus tissues, especially umbilical cord blood (Waliszewski et al. 2000). Since organochlorine pesticides reside principally in lipids and their solubility does not differ significantly among lipids in the human body (Longnecker et al. 1999; Waliszewski et al. 2004b), it is indicated that the concentration for mother and fetus depends on the lipid content in the compartments and liposolubility of the pesticide. Therefore, organochlorine pesticides would normally pass through the placental barrier via passive diffusion. Posterior metabolism in the fetus of accumulated residues is low due to the poor detoxification mechanisms of the developing fetus.

During the monitoring study, only the presence of β-HCH, *pp*′DDE, *op*′DDT and *pp*′DDT in adipose tissue and β-HCH, *pp*′DDE, and *pp*′DDT in blood serum were detected, thus only these pesticides are discussed. The summarized results of organochlorine pesticides levels from 70 monitored samples, expressed as frequencies, mean and standard deviations of mean (SD) and medians are presented in the following tables.

In Table 1, the lower levels and higher frequencies of β-HCH were determined in maternal adipose tissue. The levels increased significantly in maternal blood serum and umbilical blood serum. The obtained results indicate statistically significant differences ($p < 0.05$) between means and medians for MAT/MBS, MAT/UBS and MBS/UBS. Calculations of mean copartition coefficients disclose the following values: 0.007 for MAT/MBS, 0.002 for MAT/UBS and 0.483 for MBS/UBS, evidencing that β-HCH concentration increased 155 times from adipose tissue to maternal blood serum and 1037 times from maternal adipose tissue to umbilical blood serum. From maternal blood serum to umbilical blood serum, β-HCH concentration increased only 6.6 times. These results show a constant carry-over of the pesticide through the placental barrier and a low metabolism and specific accumulation in the fetus body.

Table 2 displays analytical results of three compartments of *pp*′DDE analyses. The mean and median results increased visually from maternal adipose tissue to blood serum lipids and to umbilical blood serum lipids. Applied statistical tests to show mean and median statistical differences among samples of MAT/MBS and MAT/UBS were significant ($p < 0.05$) and not significant ($p > 0.05$) for MBS/UBS. This demonstrates the existence of *pp*′DDE equivalent equilibrium between mother and fetus blood serum lipids. The calculation of mean copartition coefficients yields the following: 0.198 for MAT/MBS; 0.165 for MAT/UBS; and 0.974 for MBS/UBS. The *pp*′DDE concentrations increased 7.5 times from maternal adipose tissue lipids to maternal blood serum lipids, 8.9 times from maternal adipose to umbilical cord blood and only 1.2 times from maternal blood to umbilical blood. To observe the behavior of *pp*′DDE levels, the regression analyses applied showed no correlation for MAT/MBS ($r^2 = 63.9\%$) or MAT/UBS ($r^2 = 74.7\%$), and good correlation for maternal and fetal blood serum (MBS/UBS) ($r^2 = 93.2\%$) *pp*′DDE levels.

Table 3 presents comparisons of *pp*′DDE results expressed as µg/L serum on wet weight basis. The concentrations comparing maternal and fetus serum levels decreases from mother to fetus, manifesting a different behavior of concentrations from that expressed on fat basis. The observation is in concordance with the levels reported by Mustafa et al. (2010) for other organochlorine pesticides. Calculating the copartition coefficient for the maternal fetus relation, MBS/UBS (3.3 ± 2.1), it does explain the decrease of 3.3 times in the concentration of *pp*′DDE from maternal to fetus blood serum. The obtained

Table 1 β-HCH levels (mg/kg) on fat basis in binomial mother—fetus

Sample	Frequency	X ± SD	Median
Maternal adipose tissue (MAT)	63/70	0.027 ± 0.034	0.015
Maternal blood serum (MBS)	14/70	4.2 ± 4.8	2.3
Umbilical cord blood serum (UBS)	3/70	28.0 ± 9.3	23.3

Table 2 *pp*′DDE levels (mg/kg) on fat basis in binomial mother—fetus

Sample	Frequency	X ± SD	Median
Maternal adipose tissue (MAT)	70/70	0.770 ± 0.927	0.433
Maternal blood serum (MBS)	70/70	5.8 ± 13.5	2.4
Umbilical cord blood serum (UBS)	70/70	6.9 ± 13.7	3.1

Table 3 *pp*′DDE levels (µg/L) on wet weight basis in binomial mother—fetus

Sample	X ± SD	Median
Maternal blood serum (MBS)	3.7 ± 5.8	2.0
Umbilical cord blood serum (UBS)	1.3 ± 1.9	0.7

results are in agreement with those obtained by Barraza-Vázquez et al. (2008) and Pathak et al. (2008). To evaluate differences between mean and medians levels, both the *t*-test and Mann–Whitey tests indicated $p < 0.05$ and the sample groups as different.

The observed differences in behavior of organochlorine pesticide blood serum levels between mother and fetus are originated from different lipid contents in maternal and fetus serum samples. The values revealed mean maternal serum lipids of 795 mg/L versus 231 mg/L in umbilical cord blood. The calculated mean copartition coefficient for lipids was 4.09, a value that correlates with for 3.3 of *pp*′DDE copartition coefficient between maternal fetus concentrations.

Table 4 presents results of *pp*′DDT levels obtained from three compartment samples analysis. Likewise other organochlorine pesticides, *pp*′DDT levels increase in a similar way from maternal adipose tissue to maternal blood serum and to umbilical blood serum. The *pp*′DDT levels increased from MAT to MBS 21.8 times, from MAT to UBS 58.4 times and from MBS to UBS 2.7 times. Comparing the results from three sample groups, the *t*-test and Mann–Whitney tests presented: MAT/MBS and MAT/UBS as different ($p < 0.05$); and MBS/UBS as not different ($p > 0.05$). The calculations of mean copartition coefficients indicated MAT/MBS = 0.164, MAT/UBS = 0.422 and MBS/UBS = 1.097. The copartition coefficient obtained for maternal blood serum versus umbilical blood serum is based solely on three umbilical blood samples containing the pesticide with a standard deviation of 0.764, that possibility inherent bias.

Table 4 *pp*′DDT levels (mg/kg) on fat basis in binomial mother—fetus

Sample	Frequency%	X ± SD	Median
Maternal adipose tissue (MAT)	70/70	0.101 ± 0.256	0.042
Maternal blood serum (MBS)	17/70	2.2 ± 2.5	1.1
Umbilical cord blood serum (UBS)	3/70	5.9 ± 5.2	7.4

Table 5 Σ-DDT levels (mg/kg) on fat basis in binomial mother—fetus

Sample	X ± SD	Median
Maternal adipose tissue (MAT)	0.880 ± 1.142	0.487
Maternal blood serum (MBS)	6.4 ± 14.7	2.6
Umbilical cord blood serum (UBS)	7.2 ± 15.0	3.1

Table 5 summarizes results of total DDT's determined in the sample groups. The copartition factors between compartments display MAT/MBS and MAT/UBS as different ($p < 0.05$) and MBS/UBS as not different ($p > 0.05$). The applied regression test showed: MAT/MBS ($r^2 = 73.4\%$) and MAT/UBS ($r^2 = 74.7\%$) were not correlated and a good correlation for MBS/UBS ($r^2 = 93.1\%$). The mean copartition coefficients were as follow: 0.221 for MAT/MBS, 0.193 for MAT/UBS and 0.881 for MBS/UBS.

The pregnant woman's exposure to persistent organochlorine pesticides is of much interest because of exposures to the fetus and potential subsequent health effects. During pregnancy, the disposition of organochlorine pesticides is affected by physiologic changes, such as increased renal perfusion, increased volume of distribution, and increased serum lipids. The increase of serum triglyceride concentration can cause the redistribution of organochlorine pesticides stored in adipose tissue to the blood compartments. The significance of understanding the changes of these pesticide levels during gestation is so that one can to appropriately classify the fetus's level of exposure based on the sampling period during gestation (Longnecker et al. 1999). Because these pesticides tend to accumulate in the body over a person's lifetime, women's ages should play a role in their exposure to these pesticides (Herrero Mercado et al. 2010a). First pregnancy is considered to be a reliable exposure index source among pregnancies because of the lack of confounding variables, such as parity and prior breast-feeding (Wang et al. 2009) although the differences were not significant among pregnant women in Veracruz (Herrero-Mercado et al. 2010a).

In conclusion, surprisingly, the organochlorine pesticide levels differ depending on the manner of expression. If the results are expressed on serum wet weights, resulting concentrations decreased from maternal to fetal blood serum, whereas when expressed on fat basis the concentrations increased from maternal adipose tissue to maternal blood serum lipids and to fetal blood serum lipids (Fukata et al. 2005; Jimenez Torres et al. 2006; Ostrea et al. 2009) and the copartition coefficients increased significantly among the compartments (Wang et al. 2008, 2009).

Acknowledgments The authors express their appreciation to Winston Smith of the Peace Corps México for his help in editing this article.

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