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Breast Milk Excretion Kinetic of *β*-HCH, pp′DDE and pp′DDT

S. M. Waliszewski · G. Melo-Santiesteban · R. Villalobos-Pietrini · S. Gómez-Arroyo · O. Amador-Muñoz · M. Herrero-Mercado · O. Carvajal

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Abstract Breast milk is considered the most important route in the elimination of deposited organochlorine pesticides in a mother's body. The equilibrium of organochlorine pesticides in the human body considers the elements of internal transport processes, the equilibrium pattern between pesticides and tissue fat contents, and the mobilization of lipids and lipoproteins among body parts. The aim of this study was to determine organochlorine pesticide levels in breast milk samples from the 4th to the 30th day of lactation and the trend in their concentration time so as to forecast the time tendency of residue levels and the pesticide excretion pattern. Milk samples were taken from forty participants and analyzed by GLC-ECD. The organochlorine pesticide residues determined in the breast milk samples during lactation decreased: *β*-HCH from 0.095 to 0.066 mg/kg, pp′DDE from 1.807 to 1.423 mg/kg and pp′DDT from 0.528 to 0.405 mg/kg, at the characteristic rate for each compound. The obtained results compared with the calculated fits of forecasts were parallel and did not exhibit significant differences. The newborn baby exposed during lactation had organochlorine pesticide residues whose levels decreased permanently. The levels depended not only on the breast milk nutrition, but also on the total environmental exposures which included air pollution as a significant contamination source.

Keywords DDT · HCH · Breast milk · Excretion

S. M. Waliszewski (✉) · G. Melo-Santiesteban · M. Herrero-Mercado · O. Carvajal
Institute of Forensic Medicine, University of Veracruz, SS Juan Pablo II s/n, 94290 Boca del Río, Veracruz, Mexico
e-mail: swal@uv.mx

R. Villalobos-Pietrini · S. Gómez-Arroyo · O. Amador-Muñoz
Center of Atmospheric Sciences, UNAM, Mexico City, Mexico

Persistent organochlorine pesticides (DDT and HCH) have been beneficial for humans in combating vectors that transmit serious diseases and in protecting crops. The past use and the biological persistence of these pesticides in the environment have originated their presence in all elements of the environment and the food chain. Due to their volatility and widespread propagation, the main contemporary route of organochlorine pesticides for human exposure consists in the inhalation of contaminated air (Alegría et al. 2005) and the consumption of contaminated food of animal origin. Because of their lipophilic nature and high persistence, organochlorine pesticides accumulate in lipophilic human body parts, particularly in lipid-rich tissues such as adipose tissue. The contamination rate of humans depends mainly on individual exposure and accumulation which is influenced by local soil and air pollution, diet, duration of exposure, age, capacity for elimination by metabolism, and breast milk production. The monitoring of human milk could be used as an indicator to understand the rate of accumulation of such pesticides and to assess the rate of their excretion.

Psychological and medical studies have underlined the benefits of nursing which raises immunological defenses and provides a healthier development of the baby. Parallel findings have increased concern about the excretion of drugs and environmental contaminants contained in breast milk, since it is considered the main route for eliminating deposited organochlorine pesticides from a mother's body (Jensen and Slorach 1991; Sonawane 1995; Cupul-Uicab et al. 2008).

For the assumption as to the chemical equilibrium of DDT and HCH and their deposition in the human body, we have considered the elements of internal transport processes as well as the equilibrium pattern between pesticides and tissue fat contents. The model describes the distribution of persistent chemicals within the body and the mobilization of lipids and lipoproteins among body parts. The liposolubility rate is a major factor influenced by rates of accumulation and elimination from tissues and organs (Juchau 1983; Brown and Lawton 1984). The model in our study assumes that there exist equal concentrations in the body parts of both sizes and that the differences depend on the content and quantity of lipids which participate in the biological formation processes.

Individual variations in the organochlorine pesticide contents among samples are influenced by environmental pollution factors, body deposition of the pesticides and dietary habits. The aim of this research was to determine organochlorine pesticide levels in breast milk samples from the 4th to the 30th day of lactation as well as the trend in pesticide concentration time so as to forecast the time tendency of residue levels and the pesticide excretion pattern.

Materials and Methods

The human breast milk samples were collected at the 4th, 6th, 10th, 15th, 20th, 25th and 30th day postpartum from forty volunteer mothers admitted for delivery. The volunteers had lived a minimum of 5 years in Veracruz or its suburban zone. The samples, about 30 mL of breast milk, were taken from the participants in their home by manual express and put into a chemically clean glass bottle; then they were transported to the laboratory. There, the milk samples were centrifuged to separate the fat which was stored at −25°C until its analysis. The milk fat samples were transferred to a mortar and ground with sodium sulfate to obtain a coarse powder (Waliszewski et al. 2004). The ground sample was then transferred to a chromatographic column 1 cm ID and 50 cm long, and the fat containing organochlorine pesticides was extracted with 150 mL of hexane. The eluate was concentrated with a rotary evaporator to approximately 30 mL. Ten milliliters of the concentrated extract were transferred into a previously weighed round-bottomed flask of 50 mL, and the solvent was rotary evaporated to determine the fat content gravimetrically. Concentrated extract containing a maximum of 500 mg of fat was transferred into a 10 mL tube with stopper, and 1 mL of concentrated sulfuric acid was added (Waliszewski et al. 2008). The tube was stopped up and shaken vigorously, then left to reach an adequate phase separation. The supernatant was dried by passing it through a 3–5 g layer of sodium sulfate and washed with hexane. The extract with rinses was rotary evaporated to a few drops and quantitatively transferred to a volumetric tube in which the final volumes were adjusted to 1.0 mL.

The analyses were carried out in a gas chromatograph Varian model 3400CX equipped with a ^{63}Ni electron capture detector. For organochlorine pesticide separation, the procedure was as follows: a fused silica column J&W Scientific DB-608 30 m × 0.32 mm ID with a 0.83 μm thick film was used at a program temperature of 193°C (7 min)–250°C at 6°C/min and held for 20 min; carrier gas was nitrogen at 27 cm/min and a manual split/splitless sample injection of 1 μL was applied.

All breast milk samples were analyzed to determine the most frequently appearing organochlorine pesticides: HCB, α,β,γ-HCH, pp′DDE, op′DDT and pp′DDT. To determine the quality of the analytical method, we performed a recovery study on blank cow fat samples overspiked in ten replicates and obtained 88–96% recovery. The minimum detection limits for the analyzed residues on fat basis were: 0.001 mg/kg for HCB, α,γ-HCH and 0.002 mg/kg for β-HCH, pp′DDE, op’DDT and pp′DDT.

Differences among the organochlorine pesticide concentrations in the breast milk fat samples were examined at the 4th, 6th, 10th, 15th, 20th, 25th and 30th day after delivery calculating the basic statistic (mean and standard deviation of mean). We used the linear time trend model ($y_t = \beta_o + \beta_{1t} + e_t$) which compares variability in the time period and forecasts the time trend of residues. Residue levels were calculated using statistical software Minitab 12.

Results and Discussion

The presence of only β-HCH, pp′DDE and pp′DDT was detected in 100% of the samples analyzed. The results as to organochlorine pesticides in milk samples to avoid the influence of the circadian cycle on fat production are expressed on a fat basis as mg/kg, summarized in Table 1.

In human milk production, the body contributes approximately 75% of endogenous fat, and the rest proceeds from the diet (Jensen 1989, 1999). Changes in the composition of breast milk which occur postpartum as time progresses have led to classifying the milk into three types. Colostrum is first and generally believed to last from birth until about the 5th day postpartum. Transitional milk begins from the 6th day and continues up to the 15th day, and mature milk begins to excrete from the 15th day until weaning. The average fat content of colostrums is 2.9%, of transitional milk 3.6% and of mature milk 3.9% (Jensen 1989). The cholesterol, protein and phospholipids contents of milk decrease as lactation progresses from colostrum to mature milk. The fatty acid (triacylglycerols) content increases from colostrums 3.06% to mature milk at 4.01% as do the free fatty acids (Pons et al. 2000; Minda et al.

Table 1 Mean and standard deviation ($x \pm$ SD) of organochlorine pesticide concentrations (mg/kg on fat basis) in breast milk fat samples (n = 40) during lactation

Days after delivery	β-HCH ($x \pm$ SD)	pp′DDE ($x \pm$ SD)	pp′DDT ($x \pm$ SD)
4	0.095 ± 0.060	1.807 ± 0.597	0.528 ± 0.423
6	0.090 ± 0.058	1.695 ± 0.641	0.499 ± 0.414
10	0.084 ± 0.056	1.656 ± 0.540	0.479 ± 0.417
15	0.079 ± 0.054	1.567 ± 0.509	0.458 ± 0.403
20	0.074 ± 0.051	1.501 ± 0.485	0.436 ± 0.396
25	0.069 ± 0.048	1.449 ± 0.478	0.415 ± 0.388
30	0.066 ± 0.045	1.423 ± 0.473	0.405 ± 0.385

2004; Sala-Vila et al. 2005; Kent 2007; Rudolph et al. 2007).

Differences in persistence of the studied organochlorine pesticides showed that their bioaccumulation rate and the environmental pollution alter the concentrations of these pesticides accumulated in the human body due to the magnitude of exposure. Thus, the organochlorine pesticide excretion in the human body makes the more persistent pesticides predominate over the lipoproteins (colostrum) that bind the lipophilic compounds with greater force. The most important factors influencing persistent pesticide excretion into breast milk are molecular weight, lopophility and protein binding capacity (Waliszewski et al. 1999a, b). Thus, the quantity of organochlorine pesticide transferred via lactation depends on the affinity of a pesticide with the milk constituents during lactation (Yu et al. 2007).

The observed correlation between milk fat compositions is expressed by a reduction of persistent organochlorine pesticide concentrations quantified in milk fat samples during lactation and a decreasing contamination rate from colostrum to mature milk. The statistical time trend model was applied to determine the time trend of organochlorine pesticide concentrations and provide forecasts.

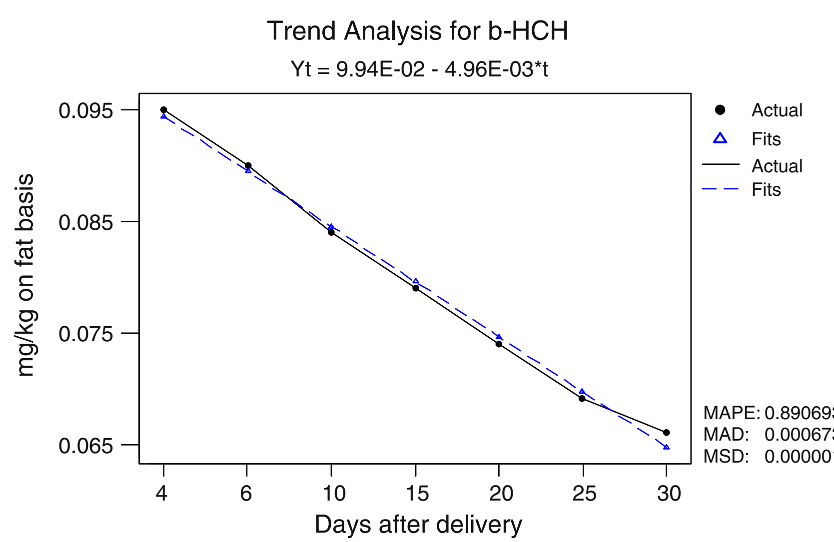
Figure 1 presents β-HCH tendency during lactation, indicating that the concentrations decreased from 0.099 mg/kg at a rate of 0.0049 mg/kg per day with low variability expressed by MSD (Mean Squared Deviation) that did not exceed 0.02%.

Figure 2 illustrates pp′DDE results of time trend tendency during lactation. These results evidenced that the excretion rate of pp′DDT initiated with 1.842 mg/kg and decreased 0.064 mg/kg daily during the study period, with low variability expressed by MSD that did not exceed 0.6%.

Figure 3 presents pp′DDT time trend during lactation. The excretion rate indicates that a pp′DDT concentration decreased from 0.543 mg/kg with a daily reduction of 0.021 mg/kg during the study period and presented low variability expressed by MSD that did not exceed 0.08%.

In conclusion, the organochlorine pesticide residues (β-HCH, pp′DDE and pp′DDT) determined in breast milk samples during lactation revealed decreased quantities at a rate characteristic of each compound. The obtained results compared with the calculated fits of forecasts were parallel and did not exhibit significant differences. Moreover, the results indicate that the newborn baby exposed to

Fig. 1 Time tendency of β-HCH during lactation



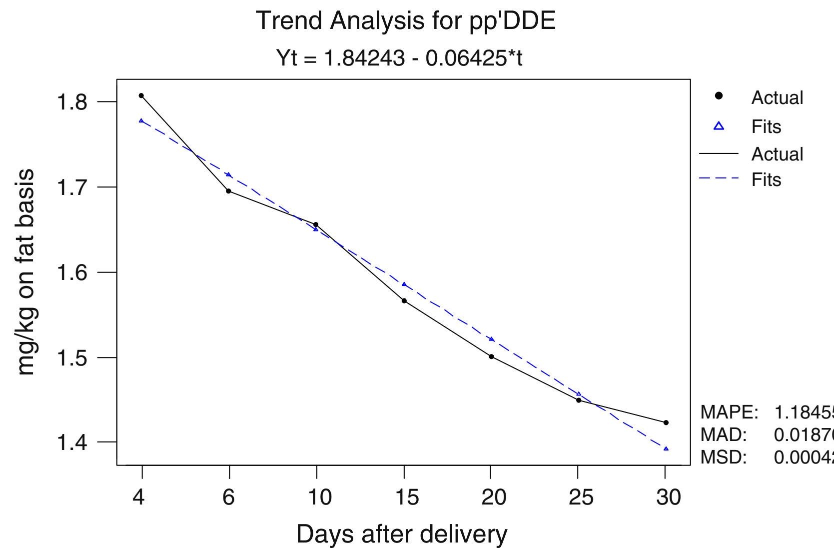


Fig. 2 Time tendency of pp′DDE during lactation

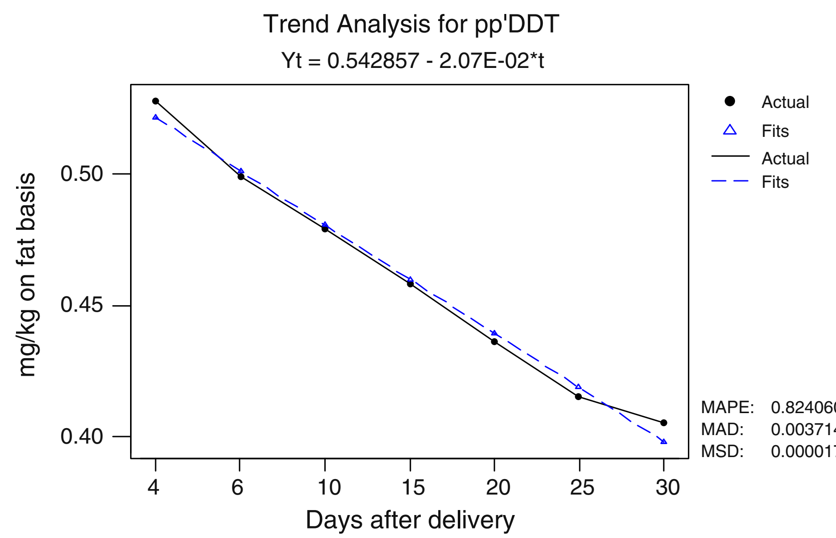


Fig. 3 Time tendency of pp′DDT during lactation

organochlorine pesticide residues during lactation had levels which decreased permanently. Thus, the rate of organochlorine pesticide residues accumulated in the body of a newborn baby nursed by breast milk, does not depend only on the breast milk contents but on the total environmental exposure which also includes air pollution as a significant contamination source (Scholtz and Bidleman 2006).

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