Bull Environ Contam Toxicol (2008) 80:255–259
DOI 10.1007/s00128-007-9356-7

Organochlorine Pesticides and Polychlorinated Biphenyls Levels in Human Milk from Chelem, Yucatán, México

Juan Pablo Rodas-Ortíz · Victor Ceja-Moreno · R. L. González-Navarrete · Jorge Alvarado-Mejía · Marissa E. Rodríguez-Hernández · Gerardo Gold-Bouchot

Received: 26 October 2007 / Accepted: 26 December 2007 / Published online: 12 January 2008

Abstract Organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in human milk from Chelem, Yucatan, Mexico were analyzed. Relatively high levels of *p,p′*-DDE, γ-chlordane, β-hexacyclohexane (β-HCH) and PCB congeners 170, 28, and 44 were found. Concentration profiles by OCP groups followed the next order: ΣDDTs > ΣChlordanes > ΣHCHs > ΣChlorobenzenes > ΣDrins. Total OCPs showed a decreasing tendency with number of births (primipara and multipara and age ranks) but these differences were not significant. ΣDDT levels were lower than in other studies in Mexico, but 36% of the samples exceeded the JMPR-FAO/WHO acceptable daily intake (ADI). About 60.53% of samples exceeded the ADI value for heptachlors.

Keywords Human milk · POPs · Organochlorine pesticides · PCBs · Yucatán México

Persistent organic pollutants (POPs) are a vast group of compounds (i.e. DDT, hexachlorobenzene, hexachlorocyclohexanes, heptachlor, and dieldrin, among others), that are manufactured as pesticides, and also industrial products or byproducts such as polychlorinated biphenyls (PCBs) and dioxins. They have been used since the 1940s, and although their use has been prohibited in many countries, they are widely distributed in the environment. POPs are persistent, toxic and lipophilic, and therefore bioaccumulate and biomagnify at all levels of food chains, affecting top predators and humans. DDT exposure increases the risk of breast cancer (Cohn et al. 2007); relations between PCBs, hexachlorobenzene and chlordanes with testicular cancer (Hardell et al. 2007), fetal growth retardation and lowered birth weight (Siddiqui et al. 2003; Rylander et al. 2000), among others have been reported. Breast milk has been used to determine organochlorine pesticides (OCPs) and PCBs levels in humans as indicators of long-term exposure because it is easy to obtain, since it is a non-invasive method, and indicates the contaminant levels in maternal fat. Moreover, it allows to evaluate the obvious concern of exposure of suckling infants to these chemicals.

Yucatan state is located in Eastern Mexico, in a potentially susceptible malaria and dengue zone. From 1959 DDT was used in Mexico in campaigns for the control of vectors, and in small amounts in agricultural zones. From 1987 its use was restricted exclusively to combat the malaria vector, but in 2000 its use was substituted by pyrethroids. In this way, from 1988 to 1999 a total amount of 33 tons of technical DDT (75% purity) in Yucatan were sprayed (Secretaria de Salud 2000). Moreover, other organochlorine pesticides such as dieldrin were also used for vector control. The goals of present study were to determine the levels of OCPs and PCBs in breast milk from Chelem, Yucatan; to evaluate differences in POPs levels according to birth number and age rank, and to compare levels of OCPs obtained in this study with the acceptable daily intake (ADI) (JMPR-FAO/WHO 2006) and other studies in different countries and Mexico.

J. P. Rodas-Ortíz · V. Ceja-Moreno · G. Gold-Bouchot (✉)
CINVESTAV Unidad Mérida, Km. 6 Antigua Carretera a Progreso, Merida, Yucatan CP 97310, Mexico
e-mail: ggold@mda.cinvestav.mx

R. L. González-Navarrete · J. Alvarado-Mejía · M. E. Rodríguez-Hernández
Unidad Interinstitucional de Investigación Clínica y Epidemiológica de la Facultad de Medicina, Universidad Autónoma de Yucatán, Avenida Itzáes No.498, Merida, Yucatan CP 97000, Mexico

Materials and Methods

Human milk samples were obtained from 38 breast-feeding mothers between December 2005 and February 2006 from Chelem in Yucatan, Mexico; samples (20 mL) were collected 4–21 weeks post-partum into amber glass bottles with Teflon caps and frozen (−20°C) until analysis. Organochlorine pesticides and PCBs were extracted and quantified by the following method. To 10 mL of milk sample, internal standards (200 ng of PCB 103) were added and then extracted three times with 50 mL of hexane for 3 min with a tissue mixer. The supernatant was filtered and humidity eliminated with anhydrous sodium sulfate. The extract was concentrated to 6 mL and cleaned with 16 g of partially deactivated Florisil (activated to 600°C for 4 h and deactivated partially with 5% w/w water) packed in a glass column and using anhydrous sodium sulfate at the top and bottom of the column. The first fraction was eluted with 70 mL hexane and the second fraction was eluted with 50 mL of hexane-dichloromethane (7:3 v/v). A total of 26 OCPs and 22 PCBs were quantified by gas chromatography using a Hewlett Packard 5890 Series II gas chromatograph, equipped with an electron capture detector and with a 30 m × 0.25 mm (0.25 μm film thicknesses) Ultra 2 capillary column. These compounds were identified and quantified using standards from Ultra Scientific. Blanks and spiked blanks were analyzed simultaneously with each set of samples for quality control and assurance. OCPs and PCBs concentrations were converted to a milk fat basis assuming that human milk is 3.5% fat (Burke et al 2003b); concentrations were reported as mean ± standard deviation, and their frequency in the samples were expressed as percentages. Estimated daily intake (EDI) (Jensen 1983) for some OCPs were calculated assuming 5 kg body mass, average 0.8 kg/day of milk intake, and a proportion of lipid in milk of 0.035, for comparison with ADI values established by JMPR-FAO/WHO (2006). Differences in POP levels in primipara and multipara women, and age ranks from mothers, were evaluated by nonparametric Kruskal–Wallis ANOVA ($p < 0.05$).

Results and Discussion

Table 1 shows OCPs and PCBs concentrations in breast milk. High *p,p′*-DDE levels (3041.36 ± 5056.83 ng/g lipid) and high frequency in the samples (84%) were found. This is the major metabolite from commercial DDT. Although it is not used in vector control since year 2000 (Secretaria de Salud 2000), high levels in this study demonstrate its persistence in the environment. A similar concentration was reported by Waliszewski et al. (2002) for *p,p′*-DDE (3,230 ng/g lipid), in human milk from 1997 to 1999 in Veracruz, Mexico. DDT isomers and metabolites in human milk are a very important risk factor in suckling health because they have been classified as possibly carcinogenic to humans (2B) by the International Agency for Research on Cancer (IARC 1991); moreover its endocrine disruptor potential has been demonstrated. In order to evaluate whether DDT exposure is caused by its recent use in the study zone, the DDE/DDT ratio was used. Ratios higher than 1:2 indicate a non-recent use (Harris et al. 1999) and it could be due to diet (food contaminated with residues).

Concentrations of γ-chlordane (925.10 ± 1038.92 ng/g lipid) and heptachlor (576.51 ± 240.31 ng/g lipid) were the highest in the chlordane group. Lindane (γ-hexacyclohexane) has been a widely used pesticide in Mexico (in agriculture, cattle ranching, and pharmaceutical industry); but high concentrations (612.31 ± 701 ng/g lipid) and highest frequencies in samples (66%) of β-HCH were found. This is due to the fact that Lindane can be converted to the beta isomer (β-HCH) in the environment (INE 2004) and it is more persistent. In general, 1,2,3,4-tetrachlorobenzene (1,2,3,4-TCB) was the most frequently detected OCP (89%) in the samples, but the mean concentration (304.40 ± 476.50 ng/g lipid) was lower than for other OCPs; its sources in the environment could be dielectric fluids spills, it was also used as insecticide and as intermediary in the manufacture of herbicides and defoliants (IPCS 1991).

The concentration profile of OCPs by groups followed the next order DDTs > Chlordanes > HCHs > Chlorobenzenes > Drins and their mean concentrations can be observed in Table 1 (Endosulfan II, Mirex and pentachloroanisole were not grouped). PCBs 195, 8 and 29 were the most frequent congeners (36.84%, 28.95% and 26.32% respectively), and the highest mean concentrations were for PCBs 170, 28 and 44 (1607.81 ± 750.87; 1264.71 ± 454.23; 1155.29 ± 235598 ng/g lipid respectively); is important to remark that PCBs have been classified as probably carcinogenic to humans (2A) by IARC (1987). Differences by age ranks and number of births, from OCPs and PCBs levels were tested, and significant differences ($p > 0.05$) were not found. An increasing tendency of ΣPCBs levels with age is observed (Fig. 1b), possibly due to bioaccumulation. In the case of the number of births (primipara and multipara), a slight decrease tendency in multiparas is observed. Total OCPs levels in primipara mothers are higher than multiparas (Fig. 1a), because during lactation, the endogenous fats (with accumulated contaminants) were mobilized and incorporated in the mammary gland as a component of breast milk; therefore a POPs depuration and elimination mechanism from the mothers is lactation.

p,p′-DDE and *p,p′*-DDT levels in this study were slightly lower than the study of Waliszewski et al. (2002) in Veracruz, Mexico (see Table 2); it is important to

Table 1 OCPs and PCBs levels (ng/g lipid) ± standard deviation and frequency (%) in human milk samples from Chelem, Yucatan Mexico

OCPs	Mean ± SD (ng/g lipid)	%	PCBs	Mean ± SD (ng/g lipid)	%
1,2,4,5-TCB	520.04 ± 563.70	55	PCB 8	687.93 ± 512.73	29
1,2,3,4-TCB	304.40 ± 476.50	89	PCB 18	443.00 ± 327.96	24
Pentachlorobenzene	178.86 ± 70.29	37	PCB 28	1264.71 ± 1454.23	18
Hexachlorobenzene	91.61 ± 49.12	55	PCB 29	25.57 ± 22.93	26
ΣChlorobenzenes	676.27 ± 701.00		PCB 44	1155.29 ± 2355.98	13
α-HCH	308.26 ± 74.12	24	PCB 52	73.48 ± 27.69	13
β-HCH	612.31 ± 694.75	66	PCB 66	3.52	3
γ-HCH	376.41 ± 151.15	18	PCB 87	212.43 ± 191.44	16
δ-HCH	245.39 ± 126.13	53	PCB 101	85.75 ± 59.92	5
ΣHCHs	753.67 ± 722.76		PCB 105	372.44	3
α-Chlordane	259.90 ± 128.59	5	PCB 110	59.15 ± 39.29	21
γ-Chlordane	925.10 ± 1038.92	16	PCB 118	ND	
Heptachlor	576.51 ± 240.31	58	PCB 128	ND	
Heptachlor epoxide	154.60 ± 129.83	47	PCB 138	38.72 ± 19.15	16
trans-nonachlor	269.33 ± 62.88	34	PCB 153	111.86 ± 35.97	8
cis-nonachlor	250.16 ± 154.53	24	PCB 170	1607.81 ± 750.87	16
ΣChlordanes	974.62 ± 742.61		PCB 180	ND	
Aldrin	282.03 ± 139.25	13	PCB 187	63.84 ± 35.44	8
Dieldrin	296.65 ± 85.92	29	PCB 195	577.90 ± 260.47	37
Endrin	290.69 ± 43.63	5	PCB 201	ND	
ΣDrins	404.20 ± 205.79		PCB 206	90.01 ± 30.00	5
o,p′-DDE	132.88 ± 83.69	42	PCB 209	ND	
o,p′-DDD	51.03 ± 35.84	47	Σ PCBs	1541.25 ± 1563.18	
p,p′-DDD + *o,p′*-DDT	126.85 ± 95.76	32			
p,p′-DDE	3041.46 ± 5056.83	84			
p,p′-DDT	209.75 ± 123.09	29			
ΣDDTs	3064.63 ± 4996.62				
Endosulfan II	277.39 ± 123.14	29			
Mirex	204.59 ± 74.50	8			
Pentachloroanisole	101.61 ± 62.96	76			
ΣOCPs	5123.06 ± 5372.77				

TCB, tetrachlorobenzene; HCH, Hexacyclohexane; ND, not detected

remark that the chronological levels obtained by Waliszewski et al. (1996, 1999, 2002) have a decreasing tendency (from 5,017 to 3,230 ng/g lipid of *p,p′*-DDE in three studies carried out between 1996 and 2002), that coincides with the restriction and prohibition years for the use of DDT in vector control in Mexico. *p,p′*-DDE and *p,p′*-DDT levels were relatively high, only lower than the results from Thailand (Stuetz et al. 2001) and Jordan (Nasir et al. 1998). For Hexachlorobenzene (HCB), β-HCH and γ-HCH our results were higher than those reported by Waliszewski et al. (1996; 1999). Heptachlor levels were higher than those from Thailand (Stuetz et al. 2001), Jordan (Nasir et al. 1998) and Taiwan (Chao et al. 2006).

The ADI is the amount of a chemical that can be ingested daily over the lifetime of an individual without an appreciable health risk to the consumer, on the basis of all known studies. In Table 3, EDI levels of human milk

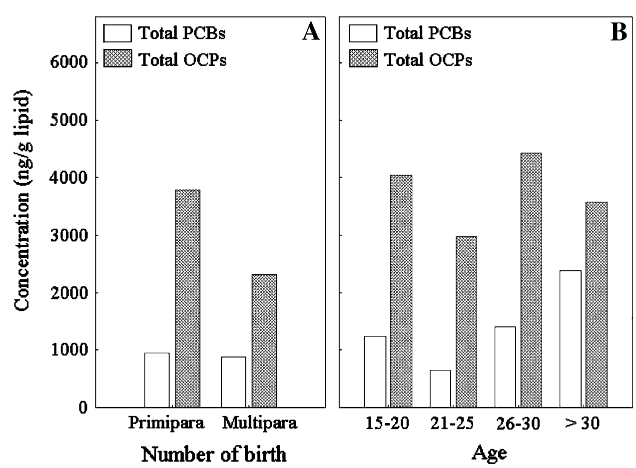

Fig. 1 Medians of total PCBs and OCPs (ng/g lipid) in human milk from Yucatan Mexico, for number of births (**a**) primipara (n = 12) and multipara (n = 26) and for age ranks (**b**) 15–20 (n = 7 8), 21–25 (n = 17), 26–30 (n = 7), and >30 (n = 6)

Table 2 Means of some organochlorine pesticides in human milk (ng/g lipid) form several countries

Country	References	n	Year	*p,p′*-DDE	*p,p′*-DDT	HCB	γ-HCH	β-HCH	α-Chlordane	Aldrin	Dieldrin	Heptachlor	Heptachlor epoxide
Thailand[a]	1	25	1998	8,214	2,597	146	103	NA	NA	NA	NA	126	177
Jordan	2	411	1998	1,950	4,400	350	710	NA	460	860	1,410	500	190
Taiwan	3	36	2000–2001	310	23	NA	2	2	10	ND	ND	3	4
Indonesia	4	10	2003	400	90	60	ND	90	NA	10	NA	NA	NA
UK	5	54	2001–2003	150	6	17	1	15	0.3	NA	NA	NA	NA
Tunisia	6	87	2002–2003	2,421	1,015	260	8	50	NA	NA	59	NA	NA
Russia[b]	7	17	2003–2004	600	50	100	0	800	NA	NA	NA	NA	NA
Japan[b]	8	38	2001–2004	330	13	14	NA	110	NA	NA	NA	NA	NA
Mexico[c]	9	43	1994–1995	5,017	1,271	47	22	561	NA	ND	ND	ND	ND
Mexico[c]	10	60	1997–1998	3,997	651	25	2	61	NA	NA	NA	NA	NA
Mexico[c]	11	112	1997–1999	3,230	510	NA	NA	NA	NA	NA	NA	NA	NA
This study		38	2006	3,041	210	92	376	612	260	282	297	577	155

NA = not analyzed; ND = not detected

[a] Results converted to ng/g lipid assuming 3.5% of lipid in human milk (Burke et al. 2003b)

[b] Primipara

[c] Studies realized in Veracruz, Mexico

[1] Stuetz et al. (2001); [2] Nasir et al. (1998); [3] Chao et al. (2006); [4] Burke et al. (2003a); [5] Kalantsi et al. (2004); [6] Ennaceur et al. (2007); [7] Tsydenova et al. (2007); [8] Kunisue et al. (2006); [9] Waliszewski et al. (1996); [10] Waliszewski et al. (1999); [11] Waliszewski et al. (2002)

Table 3 Acceptable daily intake (ADI) and means of estimated daily intake (EDI) of some OCPs found in breast milk from Chelem, Yucatan Mexico

OCP	ADI	EDI mean	Maximum levels	% > ADI
ΣDDTs[b]	10[a]	26.816	175.237	36.84
Dieldrin + aldrin	0.1	3.145	8.470	34.21
ΣHeptachlors[c]	0.1	5.884	11.240	60.53
Endrin	0.2	2.544	2.813	5.26
γ-HCH (lindane)	5	3.294	6.265	2.63

ADI and means of EDI are expressed in μg/kg day

[a] Value modified in 2000 from 20 to 10 μg/kg day (JMPR-FAO/WHO 2006)

[b] ΣDDTs = *p,p′*-DDT + *p,p′*-DDE + *p,p′*-DDD + *o,p′*-DDT + *o,p′*-DDE + *o,p′*-DDD

[c] ΣHeptachlors = Heptachlor + Heptachlor epoxide

from this study and ADI values for some OCPs (JMPR-FAO/WHO 2006) are compared. Heptachlor + heptachlor epoxide levels exceed ADI levels in most of the samples (60.53%); mean EDI level (5.884 μg/kg-day) was much higher than the ADI value (0.1 μg/kg-day). Σ-DDTs levels exceed the ADI for 36.84% of the samples. ADI values are based on the ingestion lifetime and that breast milk is consumed only on the first years of life. But it is necessary to indicate that this short period of the life is very critical, and possible adverse effects can appear in the long-term.

Our findings indicate that OCPs and PCBs were found in high levels in human milk from Yucatan Mexico, and a possible depuration mechanism of OCPs from the mothers can be the lactation. Further studies are necessary, for determining contaminant sources, and to evaluate if the POPs ingested during lactation could be causing adverse effects in the long run in children.

Acknowledgments We thank Dr. Luis René Antonio Capurro Filograsso for reviewing an earlier version and Jorge Arturo Dominguez Maldonado for his technical assistance. This project was funded by the University of Yucatan Priori-Uady program (project FMED-05-003).

References

Burke ER, Holden AJ, Shaw IC (2003a) A method to determine residue levels of persistent organochlorine pesticides in human milk from Indonesian women. Chemosphere 50:529–535

Burke ER, Holden AJ, Shaw IC, Suharyanto FX, Sihombing G (2003b) Organochlorine pesticides residues in human milk from primiparous women in Indonesia. Bull environ Contam Toxicol 71:148–155

Chao H-R, Wang S-L, Lin T-Ch, Chung X-H (2006) Levels of organochlorine pesticides in human milk from central Taiwan. Chemosphere 62:1774–1785

Cohn BA, Wolff MS, Cirillo PM, Sholtz RI (2007) DDT and breast cancer in young women: new data on the significance of age at exposure. Environ Health Perspect doi:10.1289/ehp.10260

Ennaceur S, Gandoura N, Driss MR (2007) Organochlorine pesticide residues in human milk of mothers living in northern Tunisia. Bull Environ Contam Toxicol 78:325–329

Hardell L, Van Bavel B, Lindström G, Carlberg M, Dreifaldt AC, Wijkström H, Starkhammar H, Eriksson M, Hallquist A, Kolmert T (2007) Increased concentrations of polychlorinated biphenyls, hexachlorobenzene and chlordanes in mothers of men with testicular cancer. Environ Health Perspect 111:930–934

Harris CA, O'Hagen S, Merson GHJ (1999) Organochlorine pesticide residues and polychlorinated biphenyls in human milk in the United Kingdom 1997–1998. Human Exp Toxicol 18:602–606

IARC (1987) Monographs on the evaluation of carcinogenic risk to humans, overall evaluations of carcinogenicity: an updating of IARC vol 1–42, supplement 7. International Association of Research on Cancer, Lyon, France pp 350–354

IARC (1991) Monographs on the evaluation of carcinogenic risk to humans, Vol 53. Occupational exposure in insecticide application, and some pesticides. International Association of Research on Cancer, Lyon, France pp 179–249

INE (2004) Lindano en México, Instituto Nacional de Ecología, México 2004

IPCS (1991) Chlorobenzenes other than hexachlorobenzene. Geneva, World Health Organization, International Programme on Chemical Safety Environmental Health Criteria 128

Jensen AA (1983) Chemical contaminants in human milk. In: Gunther FA, Gunther JD (eds) Residue reviews – residues of pesticides and others contaminants in the total environment, Vol 89. Springer Verlag, New York

JMPR-FAO/WHO (2006) Inventory of IPCS and other WHO pesticide evaluations and summary of toxicological evaluations performed by the joint meeting on pesticide residues (JMPR) through 2006. Joint FAO/WHO meeting on pesticide residues (JMPR) publications

Kalantsi OI, Martin FL, Thomas GO, Alcok RE, Tang HR, Drury SC, Carmichael PL, Nicholson JK, Jones KC (2004) Different levels of polybrominated diphenyl ethers (PBDEs) and chlorinated compounds in breast milk form two UK Regions. Environ Health Perspect 112:1085–1091

Kunisue T, Muraoka M, Ohtake M, Surdayanto A, Minh NH, Ueno D, Higaki Y, Ochi M, Tsydenova O, Kamikawa S, Tonegi T, Nakamura Y, Shimomura H, Nagayama J, Tanabe S (2006) Contamination status of persistent organochlorines in human breast milk from Japan: recent levels and temporal trend. Chemosphere 64:1601–1608

Nasir K, Bilto YY, Al-Shuraiki Y (1998) Residues of organochlorine hydrocarbon insecticides in human milk of Jordanian women. Environ Poll 99:141–148

Rylander L, Strömberg U, Hagmar L (2000) Lowered birth weight among infants born to women with a high intake of fish contaminated with persistent organochlorine compounds. Chemosphere 40:1255–1262

Secretaría de Salud (2000) Situación actual de la malaria y uso del DDT en México. Centro Nacional de Salud Ambiental y Centro de Vigilancia Epidemiológica, Secretaría de Salud, Diciembre 2000

Siddiqui MKJ, Srivastava A, Srivastava SP, Mehrotra PK, Mathur N, Tandom I (2003) Persistent chlorinated pesticides and intra-uterine foetal growth retardation: a possible association. Int Arch Occup Environ Health 76:75–80

Stuetz W, Prapamontol T, Erhardt JG, Classes HG (2001) Organochlorine pesticides in human milk of a Hmong hill tribe living in northern Thailand. Sci Tot Environ 273:53–60

Tsydenova OV, Surdayanto A, Kajiwara N, Kunisue T, Batoev VB, Tanabe S (2007) Organohalogen compounds in human breast milk from Republic of Buryatia, Russia. Environ Poll 146:225–232

Waliszewski SM, Pardio-Sedas VT, Chantiri JN, Infanzon RM, Rivera J (1996) Organochlorine pesticide residues in human breast milk from tropical areas in Mexico. Bull Environ Contam Toxicol 57:22–28

Waliszewski SM, Aguirre AA, Infanzon RM, Benitez A, Rivera J (1999) Comparison of organochlorine pesticide levels in adipose tissue and human milk of mothers living in Veracruz, Mexico. Bull Environ Contam Toxicol 52:685–690

Waliszewski MT, Bermudez ST, Infanzon RM (2002) Niveles de DDT en tejido adiposo materno, suero sanguíneo y leche de madres residentes en Veracruz, México. Revista internacional de contaminación ambiental 18(001):17–25