

Presence of Organochlorine Pesticides, Polychlorinated Biphenyls, and Mercury in Spanish Human Milk Samples

G. Baluja, L. M. Hernandez, Ma. J. González, and Ma. C. Rico

Instituto de Química Organica, Juan de la Cierva, 3. Madrid-6, Spain

Organochlorine pesticide residues have been detected in human milk in various countries, among these are : England (EGAN et al. 1965), Belgium (HEYNDRIKX & MAES 1969), Germany (ACKER & SCHULTE 1970), Russia (GRACHEVA 1970), U.S.A. (DYMENT et al. 1971), Holland (TUINSTRA 1971), Poland (KONTEK et al. 1971), Sweden (WESTOO & NOREN 1972), Guatemala (WINTER et al. 1976), and Canada (MES & DAVIES 1979). Polychlorinated biphenyls have also detected in Germany (ACKER & SCHULTE 1970), Sweden (WESTOO & NOREN 1972), U.S.A. (SAVAGE et al. 1977) and Canada (MES & DAVIES 1979). Mercury was also detected in Sweden (WESTOO 1973, PITKIN 1976) and Japan (HARADA 1968, WAKATSUKI 1973, FUJITA & TAKABAKATE 1977). The objective of the present study has been to investigate the levels of organochlorine pesticides, polychlorinated biphenyls and mercury in whole breast milk of Spanish mothers.

MATERIALS AND METHODS

A total of twenty milk samples was collected during the period of April to July 1981 from lactating mothers from Madrid (Spain) by manual extraction and stored at -20°C until time of analysis.

For the analysis of organochlorine compounds, samples of whole breast milk (5g) were dispersed in anhydrous sodium sulfate and the mixture extracted with hexane:acetone (40:60) in a Soxhlet apparatus for six h.

The REYNOLDS (1969) procedure was used to cleanup the extract and to separate polychlorinated biphenyls from most of the organochlorine insecticides.

The organochlorine residues were identified by the conventional methods of EC-GLC. For quantitation a mixture of 5% DC-200 and 7.5% QF-1 on 80-100 mesh Chromosorb W HP was used; temperature parameters were: column 200°C, detector 210°C, injector 215°C; nitrogen flow rate: 67 mL/min.

Simultaneously, 10 g of whole breast milk were digested with nitric and sulfuric acids and the mercury quantitated by the method of reductive aeration with stannous chloride and flameless atomic absorption spectrophotometry (UTHE 1972).

RESULTS AND DISCUSSION

The data in Table 1, expressed in parts per million, show the average residue levels, ranges, standard deviation and percentage of samples with residues ≥ 0.001 $\mu\text{g/g}$ in whole breast milk samples collected in the period of April to July of 1981.

Table 1. Organochlorine compounds and total mercury (ppm) in the whole breast milk of Spanish mothers residing in Madrid.

	a) Average	Maximum observed	Minimum observed	S.D.	% of samples with residues ≥ 0.001 $\mu\text{g/g}$
γ -HCH	0.019	0.043	0.001	0.012	100
H. epox.	0.004	0.014	0.000	0.004	77
Dieldrin	0.003	0.014	0.000	0.002	46
pp'-DDE	0.17	0.25	0.052	0.062	100
pp'-TDE	0.003	0.008	0.000	0.002	54
pp'-DDT	0.083	0.15	0.027	0.038	100
PCBs	0.25	0.32	0.19	0.038	100
Hg	0.0095	0.019	0.0009	0.0055	100

a) Averages are based on the number of samples with residues ≥ 0.001 $\mu\text{g/g}$, except for mercury levels;
H. epox. = Heptachlor epoxide

All of the twenty samples analyzed showed measurable quantities of γ -HCH, pp'-DDE, pp'-DDT, polychlorinated biphenyls and mercury. In addition, a large majority of the samples showed detectable heptachlor epoxide, dieldrin and pp'-TDE residues.

Although laboratory differences in methodology, sample origin and collection time make direct comparisons difficult, it can be noteworthy, however, to relate these residue concentrations with others of the same nature stated in a previous publication (POZO et al. 1979). According to those results, it may be assumed that in a period of two years a decreasing concentration is observed in the residue levels of α - and γ -HCH, chlordane and heptachlor, but an increase is noted in the accumulation of heptachlor epoxide, dieldrin, pp'-DDE, pp'-TDE and pp'-DDT residues (Table 2).

Therefore, the pesticide levels in whole human milk found in this study ranged from one fourth (γ -HCH) up to eight times more (total DDT) than the legal limits for organochlorine compounds established by FAO/WHO (1972) for the acceptable daily intake in human beings.

Table 2. Trends of organochlorine pesticides residues in Spanish human milk.

Compound	Average $\mu\text{g/g}$ whole milk	
	Year of publication	
	1979	1981
α -HCH	0.001	0.001
γ -HCH	0.040	0.019
Chlordane	0.0003	ND
Heptachlor	0.039	0.001
Heptachlor epoxide	0.0003	0.003
Dieldrin	0.0005	0.003
pp'-DDE	0.11	0.17
pp'-TDE	0.001	0.003
pp'-DDT	0.065	0.083

Studies on DDT levels in the newborn's blood suggest that there may exist a correlation between high levels of DDT in their blood and infant's health, and the mother's milk is an important source of infant nutrition.

In Table 3, pp'-DDE, pp'-DDT (pesticides very frequent in different studies), total DDT and PCB levels in whole human milk of various countries are given. The average content of total DDT in Spanish milk (0.25) is about the middle of the DDT range typically found in many European and American countries. Data on polychlorinated biphenyls are not so abundant as DDT, but those values shown in Table 3 indicate that PCBs in Spanish milk are higher than individual organochlorine pesticides and also higher than PCB levels reported by other countries.

With regard to the mean of total mercury content in Spanish breast milk (0.0095) it appears to be higher than in Sweden, where WESTOÖ (1973) reported 0.00093 ppm and PITKIN (1976) a range of 0.0008-0.0016 ppm. On the other hand, mercury concentration in milk of healthy mothers from the Minamata area of Japan was reported by a mean of 0.063 ppm (HARADA 1968). However, WAKATSUKI (1973) reported a range of 0.50-0.54 ppm in breast milk of mothers from an agricultural district of Japan but FUJITA and TAKABAKATE (1977) have found a mean of 0.0036 (\pm 0.0022) ppm in the mother's milk sampled in two hospitals of Tokyo.

Table 3. Comparison of pp'-DDE, pp'-DDT, total DDT and PCBs mean levels in whole human milk of various countries (ppm).

Country	Year	PCBs	pp'-DDE	pp'-DDT	DDT _{total}	Ref.
Guatemala	1974				0.38	a
Poland	1971*		0.20	0.08	0.28	b
Spain	1981	0.25	0.17	0.08	0.25	c
Russia	1970*		0.10	0.10	0.20	d
Belgium	1969*		0.07	0.05	0.12	e
England	1964		0.07	0.04	0.12	f
Germany	1970*	0.10	0.08	0.03	0.11	g
USA	1971*		0.08	0.02	0.11	h
USA	1977*	0.04				i
Sweden	1972	0.02	0.06	0.02	0.08	j
Holland	1969		0.03	0.02	0.05	k
Canada	1975	0.01	0.03	0.01	0.04	l

* Year published; a = WINTER et al. (1976); b = KONTEK et al. (1971); c = This study; d = GRACHEVA (1970); e = HEYNDRIKX & MAES (1969); f = EGAN et al. (1965); g = ACKER & SCHULTE (1970); h = DYMENT et al. (1971); i = SAVAGE (1977); j = WESTOO (1972); k = TUINSTRA (1971); l = MES & DAVIES (1979).

REFERENCES

- ACKER, L. and E. SCHULTE: Deut. Lebensm. Rundsch. 66, 385 (1970).
- DYMENT, P.G., L.M. HEBERTSON, E.D. GOMES, J.S. WISEMAN and R. HORNABROOK: Bull. Environm. Contam. Toxicol. 6, 532 (1971).
- EGAN, H., R. GOULDING, J. ROBURN and J.O.G. TATTON: Brit. Med. J. 2, 66 (1965).
- FAO/WHO: Pesticide residues in food. Report of the 1971 Joint Meeting of the FAO Working Party of Experts on Pesticide Residues and the WHO Expert Group on Pesticides Residues. FAO Agricultural Studies No 8 (1972).
- FUJITA, M. and E. TAKABAKATE: Bull. Environm. Contam. Toxicol. 18, 205 (1977).
- GRACHEVA, G.B.: Vopr. Pitan 29, 75 (1970).
- HARADA, M.: Minamata Report, Japan, Kumamoto Univ. 73, (1968).
- HEYNDRIKX, A. and R. MAES: J. Pharm. Bel. 24, 459 (1969).
- KONTEK, M., S. KUBACKI, S. PARADOWSKI and J. WIERZCHOVIEC-KA: Pediat. Pol. 46, 183 (1971).
- MES, J. and D.J. DAVIES: Bull. Environm. Contam. Toxicol. 21, 381 (1979).
- PITKIN, R.M.: Proc. Soc. Exp. Biol. Med. 148, 523 (1975).
- POZO, R., A. HERRERA, L.M. POLO, R. LOPEZ, M. JODRAL and J. IGLESIAS: Rev. Esp. Ped. 35, 94 (1979).

REYNOLDS, L.M.: Bull. Environm. Contam. Toxicol. 4, 128
 (1969).
 SAVAGE, E.P.: National Technical Information Services,
 Accession No PB 284393/As, Springfield, Va. (1977).
 TUINSTRA, L.G.M.T.: Neth. Milk Dairy J. 25, 24 (1971).
 UTHE, J.F., J. SALOMON and B. GRIFT: J. Ass. Offic.
 Anal. Chem. 55, 583 (1972).
 WAKATSUKI, R. Environmental Pollution and Health Hazards,
 Tokyo, Kodansha 275 (1973).
 WESTOO, G. and K. NOREN: Var Foeda 24, 41 (1972).
 WESTOO, G.: Var Foeda 25, 122 (1973).
 WINTER, M., M. THOMAS, S. WERNICK, S. LEVIN and M.G.
 FARVAR: Bull. Environm. Contam. Toxicol. 16, 652
 (1976).