Bull. Environ. Contam. Toxicol. (2003) 71:148–155
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DOI: 10.1007/s00128-003-0142-x

Bulletin of Environmental Contamination and Toxicology

Organochlorine Pesticide Residues in Human Milk from Primiparous Women in Indonesia

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Received: 15 September 2002/Accepted: 11 April 2003

Humans bioaccumulate organochlorine (OC) pesticides throughout their lifetime, due to the persistence of these compounds in the environment and their high lipophilicity. Human exposure occurs by direct exposure from pesticide use and indirect exposure from residues in food. OC residues are very slowly metabolised and excreted in humans, indeed a high proportion of OC intake becomes associated with body fat and is stored in adipose tissue for very long periods. It is thought that some residues accumulated throughout a woman's lifetime are mobilised during lactation and excreted in her milk. Schutz et al. (1998) reported that 90% of chlorinated hydrocarbons ingested by the mother may be excreted in her milk.

Primiparous women have the potential to excrete greater amounts of OC residues in their milk accumulated during their lifetime, than multiparous women who may have excreted the majority of their accumulated residues during previous lactations. Studies by Kostyniak et al. (1999), Spicer and Kereu (1993) and Stuetz et al. (2001) demonstrated that primiparous women have higher residues of OC pesticides in milk than women nursing their second or third child. However, some studies have found no significant difference between pesticide residues in milk and parity (Dagher et al. 1999; Duarte-Davidson et al. 1994; Torres-Arreola et al. 1999). The current study aimed to determine the concentration of OC pesticide residues in the milk of primiparous women from Jakarta, an urban metropolis and the Puncak Mountains, a rural area of West Java where the main agriculture is vegetable production associated with high pesticide use. Despite most OC pesticides' receiving worldwide bans in the 1970's, there is evidence (particularly for DDT) of their continued use in this region of Indonesia (Shaw et al. 2000).

MATERIALS AND METHODS

Human milk samples were collected from primiparous mothers between February and May 2000 from health centres in Jakarta (n = 35) and the Puncak (n = 35). All mothers who participated in the study gave their informed consent. Mothers were asked to complete a questionnaire to obtain details about the mother and infant, area of residence, employment, smoking habits and dietary information. Samples were collected at the mother's convenience. The mothers were nursing their first child and were aged 17 to 33 years (mean 22.5 ± 3 years).

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The analytical method of Burke et al. (2003) was used to analyse OC's in the human milk samples. Samples were extracted using hexane:acetone (2:1v/v) and cleaned up on a florisil cartridge. Analysis was by gas chromatography with electron capture detection (GC-ECD). The samples were analysed for the following pesticides: *p,p'*-DDT (1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethane), *p,p'*- DDE (1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethene), dieldrin, HCB (hexachlorobenzene), β-HCH (hexachlorocyclohexane) and γ-HCH. A validation sample was analysed at the beginning and end of each batch and the samples contained an internal OC standard (aldrin). The limit of detection (LOD) in milk fat was 0.01 mg kg^{-1}. The LOQ based on milk fat was 0.01 mg kg^{-1} for *p,p'*-DDT, *p,p'*-DDE and dieldrin, 0.03 mg kg^{-1} for HCB and γ-HCH and 0.04 mg kg^{-1} for β-HCH. Where residues were below the LOQ, a value of LOQ/2 was assigned. The analyte recoveries from spiked milk samples were in the range 53 to 109%.

Potential correlations between residue levels in milk fat and questionnaire data were tested using Spearman's Rank (P=0.05). The Mann Whiney U test was used to investigate significant differences between urban and rural samples.

The infants' estimated daily intake (EDI) of OC residues from human milk were calculated based on the average 5 kg infant consuming 800 mL of milk day^{-1} (Jensen 1983).

RESULTS AND DISCUSSION

There was no significant difference (P=0.05) between the fat content of the samples and their area of collection. The mean fat content in urban samples was 3.4 ± 1.5% (1.6 – 7.4) and 3.7 ± 1.6% (1.0 – 6.9) in rural samples. Jensen (1983) reported that the mean fat content of human milk samples was between 2.6 and 4.5% and individual values of 1.2 to 12.1% were possible.

OC residues in human milk from primiparous Indonesian women are shown in Table 1. Only *p,p'*-DDT and *p,p'*-DDE were found above the LOQ in all of the samples. DDE was the dominant analyte detected in the majority of samples. The mean DDE:DDT ratio is greater in urban than rural areas, but the difference is not significant. The third most frequently detected residue from the rural area was dieldrin (69% of samples), compared to β-HCH (54% of samples) in Jakarta. The levels of OC residues found were generally low in a world context (Shaw et al., 2000).

There was no significant difference in the residue levels detected between rural and urban sampling areas. Significant positive correlations were found for the concentration of *p,p'*-DDE (P = 0.01) and dieldrin (P = 0.05) and the age of the mother, suggesting that older mothers have had a greater cumulative exposure to *p,p'*-DDE and dieldrin throughout their lifetime. This is expected and corresponds with previous findings Dagher et al. (1999); Duarte-Davidson et al. (1994); Mussalo-Rauhamaa et al. (1988).

Table 1. OC pesticide residues in human milk from urban and rural areas of Indonesia (mg kg^{-1} milk fat). ΣDDT = *p,p'*-DDT + *p,p'*-DDE.

	Urban (n = 35)			Rural (n = 35)		
Residue	% > LOQ	Mean ± σ	Range	% > LOQ	Mean ± σ	Range
HCB	17	0.04± 0.07	<0.03-0.30	17	0.03 ± 0.03	<0.03-0.14
γ- HCH	11	0.02± 0.02	<0.03-0.09	3	0.02 ± 0.04	<0.03-0.27
β- HCH	49	0.10± 0.13	<0.04-0.52	40	0.08 ± 0.10	<0.04-0.32
Dieldrin	40	0.02± 0.02	<0.01-0.11	69	0.02 ± 0.02	<0.01-0.12
p,p'-DDE	100	0.39± 0.90	0.02-3.69	100	0.18 ± 0.56	0.01-3.37
p,p'-DDT	100	0.07± 0.08	0.02-0.44	100	0.05 ± 0.04	0.01-0.25
ΣDDT	100	0.46± 0.94	0.02-3.89	100	0.02 ± 0.57	0.02-3.50
DDE:DDT		4.9 ± 8.7	0.2-38.3		2.5 ± 4.2	0.5-25.6

No significant correlations were found between residues levels and mother's height and weight or duration of lactation. The duration of lactation has been reported to influence levels of OC pesticide residues detected. During lactation women excrete OC residues mobilised from their body fat stores. Therefore, if accumulated pesticides are excreted in milk, it is anticipated that as the duration of lactation increases, residue levels decline. Evidence of this decline has been identified by Duarte-Davidson et al. (1994) and Kostyniak et al. (1999). However, not all studies have detected this trend (Mussalo-Rauhamaa et al. 1988).

Food frequency questionnaires and diet types (e.g. high fish consumers) are often used to assess the dietary intakes of persistent pesticides. Some reports have identified correlations between diet and levels of OC pesticides in the diet. Not surprisingly women who consume greater amounts of foods containing animal fats have been found to contain higher residue levels in their milk (Dagher et al. 1999; Torres-Arreola et al. 1999). On the other hand some studies have not identified any trends or significant correlations between consumption of animal products and residue levels in milk (Duarte-Davidson et al. 1994; Kalra et al. 1994; Kostyniak et al. 1999; Mussalo-Rauhamaa et al. 1988). Our study did not show any correlations or trends between frequency of food consumption and OC pesticides. Stuetz *et al.,* (2001) investigated pesticide residues in human milk from Northern Thailand, where the diet is similar to Indonesia (predominantly rice, fruit and vegetables) and found no correlation between dietary habits and pesticide residue levels.

It has been suggested (Shaw et al., 2000) that the levels of OC pesticides in human milk fat can be used to compare the degree of environmental contamination in different countries (Table 2). The highest levels of *p,p'*-DDT and *p,p'*-DDE were recorded in Central Java, Indonesia by Noegrohati et al. (1992) where DDT had been used in malaria eradication programmes. High residues have also been reported by Kalra et al. (1994) where DDT had been widely used in agriculture and public health. In the present study, residues were lower than other regions of SE Asia and similar to Papua New Guinea, where no reported exposure to DDT has occurred. Our results perhaps indicate that women have been exposed to low levels of DDT and DDE through environmental contamination but that direct exposure from agricultural and household use has been minimal. The DDE:DDT ratio in Indonesia indicates environmental exposure to DDT, similar to India. The results from Indonesia are higher than other areas of SE Asia, but the values are much lower than Australia, New Zealand and the UK. This suggests that DDT may have been used recently in Indonesia.

Dieldrin residues have not been reported in human milk from other SE Asian countries. The levels detected in Indonesia were lower than those reported from the UK and Australia. These residues are likely to reflect environmental contamination from past use.

HCB is an OC fungicide and industrial process intermediate. Most reports of levels of HCB in human milk are from developed countries and tend to exceed values from the few reports in less developed countries, probably due to its past release into the environment from industry. Very high residues were reported from Australia (max 7.6 mg kg^{-1}) (Quinsey et al. 1994). Indonesian women had higher residues than those from Japan and New Zealand, but were comparable to the UK. The mean value of HCB in human milk from Thailand was higher than Indonesia.

β-HCH is an isomer of HCH, which is more persistent than the other isomers. The highest levels in human milk were reported from India, this is likely to reflect the high usage of lindane and technical HCH in this country. Levels from Indonesia are comparable to the UK and Vietnam. Information on the past and current usage of HCH isomers in Indonesia is not available.

The Joint FAO/WHO Meeting on Pesticide Residues (JMPR) and the Joint FAO/WHO Expert Committee on Food Additives (JECFA) have established the acceptable daily intake (ADI) of various pesticides (Table 3). The EDI of OC residues by infants in Indonesia are given in Table 3.

Some infants in both areas of Indonesia exceeded the ADI for HCB, dieldrin and total DDT, but the majority of infants ingest levels of OC pesticides, that were below the ADI levels. The ADI is calculated from adult animal experiments and it is possible that this cannot be directly extrapolated to human infants who are likely to have different susceptibilities to environmental chemicals. However, the safety factor included in the calculation of the ADI should account for this variability. In addition, the ADI is based on a lifetime exposure, but infants only consume breast milk for a small proportion of their lives. In a risk context it is unlikely that that the OC intake will be less harmful than not breastfeeding.

The results from this study demonstrate that the levels of OC pesticide residues in human milk from Indonesia are generally low, even though a previous study have

Table 2. Mean and maximum OC pesticide residues in human milk (mg kg^{-1} milk fat) from around the world.

	DDT		DDE		Dieldrin		HCB		β-HCH		γ-HCH		DDE: DDT
Country	Mean	Max	Mean	Max	Mean	Max	Mean	Max	Mean	Max	Mean	Max	
Australia	0.23	0.96	0.96	3.9	0.16	0.19	0.41	7.6	0.35	4.4	0.11	0.48	4.27
India	7.18	27.00	10.00	44.25	N/A	N/A	N/A	N/A	4.37	22.46	0.21	0.71	1.39
Indonesia*	0.06	0.44	0.28	3.69	0.02	0.12	0.03	0.30	0.09	0.52	0.01	0.27	4.67
Indonesia	12.20	41.60	15.80	54.70	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	1.30
Japan	0.02	0.12	0.27	1.00	N/D	N/D	0.01	0.02	0.21	0.97	-	-	15.17
NZ	0.026	0.49	0.62	1.49	0.02	0.04	0.01	0.027	0.02	0.08	0.001	0.01	24.34
PNG	0.42	1.20	0.45	1.65	N/D	N/D	N/D	N/D	N/D	N/D	N/D	N/D	1.07
Thailand#	2.60		8.21		N/A	N/A	0.15		N/A	N/A	0.10		3.16
UK	0.04	0.80	0.43	4.00	0.05	0.35	0.04	0.33	0.07	0.75	0.04	0.20	10.75
Vietnam	1.40	3.10	3.40	7.60	N/A	N/A	N/A	N/A	0.14	0.33	0.02	0.14	2.43

(Australia (Quinsey et al. 1994); India (Kalra et al. 1994) Indonesia* (this study); Indonesia (Noegrohati et al. 1992); Japan (Konishi et al. 2001); NZ (New Zealand) (Bates et al. 2001); PNG (Papua New Guinea) (Spicer and Kereu, 1993); Thailand (Stuetz et al. 2001); UK (Harris et al. 1999); Vietnam (Nakamura et al. 1994). N/A = not analysed, N/D = not detected above LOD, # results have been converted to milk fat assuming a fat content of 3.5%).

Table 3. The EDI (mg kg^{-1} day^{-1}) of pesticide residues from mother's milk for infants in urban and rural areas of Indonesia and the ADI values.

		HCB	*ΣHCH*	*Dieldrin*	*ΣDDT*
ADI		0.0006[d]	0.008	0.0001	0.02
Urban	Mean	0.0002	0.0005	0.0001	0.0022
	Max	0.0019	0.0034	0.0003	0.0277
	% > ADI	9	0	14	3
Rural	Mean	0.0001	0.0005	0.0001	0.0009
	Max	0.0008	0.0023	0.0004	0.0078
	% > ADI	3	0	14	0

(ΣHCH = βHCH + γHCH. [d] this value was withdrawn in 1976 and a replacement has not been released).

suggested higher values (Shaw et al., 2000) this was based on a low number of samples (n=4) and may reflect the variability in exposure. However, due to the traditional low consumption of animal fats and more recent problems of malnutrition among the population, it is likely that body fat stores and associated lipophilic contaminants have been mobilised in primiparous women before the onset of lactation. Therefore, contaminants in milk are at least in part due to recent dietary exposure. On a global scale, the residues found are comparable to regions of Europe and Oceania and do not indicate high exposure through their use in agriculture and public health. However, the DDE:DDT ratio does indicate recent use of DDT rather than environmental exposures to DDE. The effect of OC pesticides on the suckling infant in Indonesia is probably of less concern than malnutrition, sanitation and food born illness. If the government bans on persistent OC pesticides are enforced, levels of these compounds in the environment, food and consequently human milk will not increase further, as has occurred in developed countries.

Acknowledgments. We thank the British Council for funding the academic link between the University of Central Lancashire (UK) and the Universitas Indonesia.

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