

## **House Treatment with Organochlorine Pesticides and Their Levels in Human Milk—Perth, Western Australia**

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In a survey carried out during 1979/80 (Stacey et al. 1984) to examine the effect Government controls on the use of organochlorine pesticides had caused on their residue levels in human milk in Western Australia it was noted that while the levels of HCB and DDT had decreased the level of dieldrin had shown a substantial increase (5-9 ng/g). A statistical analysis of the results showed a strong correlation between the dieldrin levels and the treatment of houses for their protection against "white ants" (termites of the Isoptera family). All new houses in Perth, Western Australia must be treated and many householders take out a contract, with the pest control agencies, which frequently results in an annual treatment of the house and/or its surroundings. At the time of the 1979/80 survey aldrin was a commonly used pesticide for this purpose and the results suggested that its use had contributed to some of the high levels of dieldrin found in breast milk.

The present survey was undertaken in an attempt to determine whether this type of treatment did in fact contribute to these high levels of dieldrin.

As in previous surveys (Stacey and Thomas 1975, Stacey et al. 1984) the samples of breast milk were supplied by the Western Australian Branch of the Nursing Mothers Association of Australia who the authors wish to thank for their continued support.

### **MATERIALS AND METHODS**

Commencing in November 1980 fourteen donors from various parts of the Perth metropolitan area were asked to supply samples of breast milk (40ml if possible) on a monthly basis. In cases where the house was treated after the initial sample had been collected the donor was requested to supply more frequent samples.

The samples were to be taken over a number of feeds and at various stages of the feed. This was to avoid unduely low or high values as a result of the pre or post feed effects noted in the previous survey (Stacey et al. 1984). All samples were kept frozen until required for analysis.

Each donor was also asked a series of questions relating to the treatment programme of the house or houses in which they had lived during the previous years. The results of this questionnaire are given in Table 1.

Table 1. History of Treatment of Houses Lived in by Donors\*

Donor No.	Last sprayed prior to survey	Pesticide	Sprayed yearly	Comments
1.	1 Month	Heptachlor	Yes	
2.	2 Months	Aldrin	Yes	
3.	12 Months	Aldrin	Yes	House was sprayed with chlordane the day after 1st sample taken.
4.	5 Weeks	Heptachlor	No	Previous spraying 2.5 years before.
5.	10 Days	Heptachlor	Yes	Shifted interstate after 2nd sample
6.	4 Months	Aldrin	Yes	
7.	1 Month	Heptachlor	Yes	Partial spraying 6mth before with Aldrin.
8.	1 Week	Heptachlor	Yes	Stopped lactating after 1st sample.
10.	7 Months	Heptachlor	Yes	House sprayed with Heptachlor after 3rd sample.
11.	3 Months	N.O.P.	No	Last sprayed 3yrs prior.
12.	12 Months	N.O.P.	Yes	House sprayed with NOP after 1st sample.
13.	4 Months	Heptachlor	No	
14.	4 Months	Heptachlor	Yes	

N.O.P. = Non organochlorine pesticide

\* The rate of application of pesticide can vary with the type of treatment however the Standards Association of Australia in their Australian Standards AS2057-1981 (buildings under construction) and AS2178-1978 (existing buildings) recommend a rate of application not less than 150 L/m<sup>2</sup> of soil with any of the following water emulsions; aldrin (0.5%), chlordane (1%), dieldrin (0.5%), heptachlor (0.5%) or chlordane/heptachlor (0.5%/0.25%).

Each sample was homogenised and the fat extracted from 30ml (where available) using the single extraction method described in the

Pesticide Analytical Manual of the Food and Drug Administration, U.S. Department of Health, Education and Welfare (1968). A modified Moat's column clean up was employed using a florisil column eluted with 20%  $\text{CH}_2\text{Cl}_2$ /hexane, hexane and acetonitrile. The acetonitrile residue was further eluted from a  $\text{MgO}$ /celite column with hexane.

Analyses were performed using the following instrument parameters:

Chromatograph: Varian model 1440  
Detector: Concentric tube; electron capture, tritium  
Column: Glass, 2m, packed with equal parts of 5% QF-1 and 5% DC-200 on 100-120 mesh Varaport 30. For confirmation; glass, 2m, packed with 1.5% OV-17 on 100-120 mesh Varaport 30.

In addition to gas chromatography, peak identifications were confirmed by T.L.C. using  $\text{AgNO}_3$  incorporated alumina. Results were corrected to 100% recovery. Standards were injected after every five samples. This method detected organochlorines at a sensitivity level of 0.001ppm.

## RESULTS AND DISCUSSION

The initial aim of this survey was to test the conclusion, of the previous survey (Stacey et al. 1984) that the high levels of dieldrin found in some samples of human milk in Western Australia were related to the use of aldrin in protecting houses against attack by white ants. This was to be achieved by monitoring the dieldrin levels in the milk of several donors whose houses had recently been treated with aldrin. However, since the completion of the previous survey, there had been a movement away from the use of aldrin (although still legal) towards heptachlor, chlordane and the organophosphates. The survey was therefore extended to include chlordane, heptachlor and heptachlor epoxide.

Since these had not been included in the earlier surveys there were no figures for comparison, however some interesting results were obtained with two of the donors (No.s 3 and 10).

Unlike the previous surveys the donors this time were not a random group but were biased to include only those whose houses had been recently treated (12 months or less). Thus it was no surprise that the mean level for dieldrin was higher than the 1979/80 survey (Table 2). The donors come from both old and new areas of the metropolitan area and the mean value (13 ng/g) of dieldrin fell between the two groups New/Treated (16 ng/g) and Old/Treated (11 ng/g) from the previous survey.

In only three of the 14 cases studied (Donors No.2,6 and 9) had aldrin been used in the most recent treatment. Apart from the very high value of the first sample of No.2 and the low value for the last sample of No.6 it appeared that the levels of dieldrin continue to rise until the seventh or eighth month after treatment

(Table 4) at which time they level off and start to decline. The levels of other donors who have previously had their houses treated (Table 1 and 3) suggested that the decline in level of dieldrin could be rather slow.

Table 2. Mean Values of 74 Samples from 14 Donors (Values Expressed in ng/g, Whole-milk Basis).

<u>Pesticide</u>	<u>Range</u>	<u>Mean</u>	<u>Std Deviation</u>
HCB	1- 33	9 ( 8)	7
BHC	0- 4	1 ( 1)	0
Chlordane	0- 66	6	13
Heptachlor	0- 13	1	2
Heptachlor Epoxide	0- 29	4	5
Dieldrin	2- 35	13 ( 9)	7
Total DDT	3-159	42 (46)	32

Results in parenthesis are for the 1979-80 survey

Table 3. Mean Levels of Organochlorine Pesticides in Milk of Individual Donors (Values Expressed in ng/g, Whole-milk Basis)

<u>Donor No.</u>	<u>No of Sample</u>	<u>HCB</u>	<u>BHC</u>	<u>Chlor- dane</u>	<u>Hept- achlor</u>	<u>Heptachlor Epoxide</u>	<u>Diel- drin</u>	<u>DDE</u>	<u>DDT</u>	<u>TOTAL DDT</u>
1.	4	12	1	2	Tr	3	14	27	8	38
2.	6	15	2	2	Tr	3	14	50	5	61
3.	7	3	2	32	Tr	2	10	15	3	20
4.	6	4	1	2	Tr	2	9	18	3	23
5.	2	13	4	3	Tr	7	26	104	23	139
6.	6	7	1	2	Tr	3	21	28	6	37
7.	6	10	2	2	1	5	10	33	5	42
8.	1	5	1	2	Tr	1	19	40	5	50
9.	3	5	1	1	Tr	2	19	17	3	22
10.	14	15	1	4	2	11	13	48	10	63
11.	4	9	1	1	1	2	7	17	4	23
12.	7	5	1	3	1	2	9	18	5	25
13.	4	8	Tr	2	1	2	8	16	3	21
14.	4	4	1	8	Tr	3	16	44	8	57

Tr = trace below 1 ng/g

Although there had been a trend away from the use of aldrin it had been used extensively in the past and donors whose houses had been treated annually indicated that previous treatments had been with aldrin. The fact that the dieldrin levels for this same group were all equal to or greater than the mean value of the previous survey

(9 ng/g) further strengthened the connection between house treatment with aldrin and high levels of dieldrin in the breast milk.

Table 4. Dieldrin Levels in Donors Where Aldrin Most Recent Pesticide Used (Values Expressed in ng/g, Whole-milk Basis)

Donor No.	Dieldrin levels in months after treatment								
	2 mths	3 mths	4 mths	5 mths	6 mths	7 mths	8 mths	9 mths	
2	26	9	12	14	11	14			
6			19	21	26	31	21	8	
9						16	24	16	

The only donors with dieldrin values equal to or less than the previous mean were No.4 (9 ng/g), No.11 (7 ng/g), No.12 (9 ng/g) and No.13 (8 ng/g). All indicated that aldrin had not been used to treat their houses for at least 2 years (Table 1).

Results of the survey tend to strengthen the previous contention that the use of aldrin for the irradiation of and protection against termites contributed to the high levels of dieldrin in some samples of breast milk in Western Australia.

Although it was not possible, with the donors available, to monitor the levels of dieldrin before and after the treatment with aldrin, there were two donors (Nos 3 and 10), where organochlorines other than aldrin had been used, with whom a monitoring of this type was achieved. With donor No 3 the pesticide used was chlordane while heptachlor was used in the case of No.10. The levels of the pesticides or their metabolites in the milk showed significant rises soon after treatment with the peak levels occurring approximately a month later. (Table 5 and 6). The levels returned to near pre-treatment values by the 4th or 5th month. Comparing these results with Table 4 suggested that the increase and decrease in levels for both chlordane and heptachlor epoxide are more rapid than for aldrin.

Table 5. Chlordane Levels in Donor No. 3 (Values Expressed in ng/g Whole-milk Basis)

Sample No.	Sampling time relative to treatment	Chlordane
1.	3 days prior	Tr
2.	1 week after	63
3.	3 weeks after	66
4.	7 weeks after	64
5.	11 weeks after	26
6.	15 weeks after	2
7.	19 weeks after	2

Tr = trace less than 1 ng/g

Table 6. HCB, Heptachlor and Heptachlor Epoxide Levels in Donor No. 10 (Values Expressed in ng/g, Whole-milk Basis)

Sample No.	Sampling time relative to treatment	HCB	Heptachlor	Heptachlor Epoxide
1.	3 months prior	3	-	1
2.	2 months prior	8	Tr	4
3.	1 month prior	7	Tr	3
4.	1 day after	6	13	2
5.	3 days after	22	2	4
6.	1 week after	18	Tr	8
7.	2 weeks after	17	Tr	8
8.	3 weeks after	13	2	13
9.	4 weeks after	22	3	29
10.	5 weeks after	23	4	29
11.	7 weeks after	21	2	21
12.	9 weeks after	18	1	16
13.	12 weeks after	17	Tr	7
14.	15 weeks after	9	Tr	3

Tr = trace less than 1 ng/g

With donor No.10 the rise and fall in the level of heptachlor epoxide was accompanied by a similar effect in the HCB level. This suggested that the spray used contained not only heptachlor but some HCB as well. No confirmation of this could be obtained from the pest control agency.

Houses with concrete floors were treated by injecting the pesticide into the soil around the house to form a barrier while those with wooden floors were also sprayed under the house. As well most fences and sheds were sprayed. Although it was recommended that the inhabitants vacate the house during the treatment and for some hours afterwards this was not always possible. To our knowledge no organochlorine pesticide was used inside the house thus reducing the risk of absorbing by contact with treated surfaces. This suggests that breathing the vapours over a period of time may be the main route of contamination. As no levels in the air were taken during this survey it was not possible to confirm this.

The above data, however, showed a clear connection between the levels of organochlorine pesticides or their metabolites in human milk with their use in and around the house. This suggests that the main source of pesticide residue in the body may not always be from food but may in fact come from sources, not connected to agriculture, which previously were considered to have little or no effect on levels in the body. In reference to the small percentage of pesticide that finds its way to the crop during spraying Crosby (1984) posed the question "Where does it all go?" It would appear that this question is just as relevant when applied to other forms of treatment such as the use of pesticides in or around the house.

When authorities are formulating controls on pesticide usage they should therefore address themselves to this question and consider it in light of the various ways in which pesticides can be taken into the body.

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