

## Influence of Termite Treatment in the Home on the Chlordane Concentration in Human Milk

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**Abstract.** An increase of chlordane contamination in humans after termite treatment of their homes was investigated by analyzing human milk samples of breast-feeding women. Chlordane in human milk was analyzed by gas chromatography-mass spectrometry (GC-MS) and the concentrations were compared between women living in termite-treated and non-treated houses. Polychlorinated biphenyls (PCB) were also analyzed on the same samples and the result was discussed in relation to the concentration of chlordane.

Mean concentrations of six chlordane components of the exposed group were three times higher on the average than those of the control group. When the duration of dwelling was less than one year, however, chlordane levels of the treated group were similar to those of the control.

A proportional correlation was observed between the concentration of total chlordane and the duration (years) after the termite treatment of the home. The pattern of chlordane components observed in the milk of the exposed group resembled that of technical chlordane, a commercial product. Chlordane compounds were so stable and persistent in the treated homes that continuous accumulation into the dwellers persisted for at least five years after treatment.

The levels of total chlordane in the milk samples of the control group were very low compared to PCB levels. However, when duration of dwelling after termite treatment expanded for two years or longer, chlordane in the milk increased to a level similar to PCB.

A close correlation between concentrations of chlordane and PCB of the non-treated group was observed. This fact suggested that a main source of chlordane exposure for the general population was marine products. However, in the case of the

treated group, chlordane exposure was inferred to be derived from the inhalation of a contaminated atmosphere in the homes or absorption of chlordane via the skin or uptake of foods contaminated in the homes.

Chlordane is a viscous, light yellow to amber-colored liquid. Technical chlordane is a mixture of at least 26 different components (WHO 1984a). The main components are trans-chlordane ( $24 \pm 2\%$ ), cis-chlordane ( $19 \pm 3\%$ ),  $\gamma$ -chlordene (9%), heptachlor (7%), trans-nonachlor (7%) (Sovocool *et al.* 1977). Chlordane has been used for approximately 40 years as a broad spectrum contact insecticide, mainly on non-agricultural crops and animals. In Japan, use of the chemical has been permitted only for the control of termites and wood-boring beetles. Chlordane is so persistent in the environment and stable chemically that it maintains its effects for approximately fifteen years after treatment.

The acute toxicity of chlordane in rats is estimated to be similar to that of DDT. Signs of poisoning in various animal species are neurotoxic manifestations such as disorientation, tremors, and convulsions (WHO 1984a). WHO (1984b) rated the toxicity of chlordane as "moderately hazardous". Most of its metabolites are slightly to moderately toxic, with the exception of highly toxic oxychlordane (acute oral  $LD_{50}$  for rat: 19.1 mg/kg body weight). The acute lethal dose of technical chlordane for man is estimated to be 25-50 mg/kg body weight (WHO 1984a). No adverse effects have been reported in occupationally exposed workers.

In Japan, the consumption of chlordane has increased rapidly since 1980. Subsequently, its envi-

ronmental contamination became a serious social task. Recently, the Environmental Agency of Japan conducted an investigation on the contamination of chlordane in the environment, and reported that the concentration of the chemical in marine organisms was increasing. Based on animal experiments carried out for the evaluation of chronic toxic effects of chlordane, the Japanese government decided to prohibit the use of the chemical in 1986.

Chlordane vaporizes gradually in the domestic atmosphere of treated homes (major use of chlordane is termite control) for over ten years. It will accumulate in the dweller's body by the inhalation of vapor, by the intake of foods which adsorbed the vapor, or by skin contact of the chemical.

Data are available on the levels of chlordane in human milk of breast-feeding women whose homes have not been treated with the chemical for termite control (Mes and Davies 1978; Strassman and Kuntz 1977; Barnett *et al.* 1979; Savage *et al.* 1976, 1981). Until now, however, there are few data on dwellers in homes treated with chlordane, except the report of Kawano and Tatsukawa (1982), which reported a relatively high concentration of oxy-chlordane in one blood sample. With these reports, it is still difficult to relate the chlordane treatment of the home to the increase of the levels of chlordane contamination in man.

Our study was conducted to clarify the problem by using human milk samples collected in the Osaka Prefecture from 1984 to 1985. Chlordane levels in the milk of breast-feeding women were analyzed by GC-MS and the results of subjects living in the treated house were compared with that of a non-treated group. Polychlorinated biphenyls were also determined and the results were discussed in relation to the concentration of chlordane.

## Experimental

### Reagents

Column chromatography for the partial purification of chlordane in samples was made with Florisil® (60–100 mesh, Katayama Chemical). Florisil® (60–100 mesh, Wako pure Chemical Industries) was used for the purification of PCBs. Reagents used for the analysis were all special analytical grade for PCBs and pesticides (Wako Pure Chemical Industries). Six chlordane components in a technical chlordane of "Tech. Ref. Std. chlordane 100%" (Velsicol Chemical Corp., Lot. No. b-8113) were determined for the pattern analysis of chlordane residues.

### Samples

Human milk samples from 197 breast-feeding women were collected in Osaka prefecture from 1984 to 1985. Milk fat of the samples was extracted by means of the official procedure for

PCB analysis of Japan (Ministry of Health and Welfare Japan 1972) and kept at 5°C until used. Exposed group consisted of sixteen samples from women whose homes were treated with chlordane. As a control, seven were selected randomly among 181 human milk samples.

### Extraction and Clean-up

Chlordane Analysis: After milk fat was removed by Florisil® dry column chromatography (Wakimoto and Tatsukawa 1972; Yakushiji *et al.* 1979), organochlorine compounds were extracted with *n*-hexane. The extracts were concentrated to 3 to 5 ml and pouted into a glass column (30 cm × 15 mm I.D.) containing 15 g of Florisil® activated for 15 hr at 130°C. Chlordane compounds were eluted with 120 ml of *n*-hexane/ether (9:1, v/v). The eluates were concentrated to 0.5 ml with a Kuderna Danish concentrator.

In a preliminary study, three chlorinated hydrocarbons, *p,p'*-DDD, tetrachlorobiphenyls and 6-hexachlorocyclohexane (δ-HCH) were examined for use as an internal standard for chlordane analysis. Retention times of these compounds on capillary gas chromatography were different from all of the chlordane components. However, large quantities of unknown contaminants whose retention time was the same as *p,p'*-DDD and tetrachlorobiphenyls were detected in the sample extracts. Therefore, δ-HCH was chosen as an internal standard and added to the sample extracts. The concentration of 6-HCH was standardized at 200 µg/L in both sample extracts and standard solutions.

PCB Analysis: PCBs were extracted according to the official procedure of Japan (Ministry of Health and Welfare Japan 1972), partly improved (Yakushiji *et al.* 1978). After the milk fat sample was saponified with alcoholic potassium hydroxide, PCBs were extracted with *n*-hexane and cleaned up by Florisil® column chromatography.

### Instruments

Chlordane Analysis: A double focusing mass spectrometer (Jeol JMS DX-300) coupled to a data system JMA 3100/3500, with a Hewlett-Packard model HP5710A capillary gas chromatograph equipped splitless injector was used for selected ion monitoring of chlordane.

Chromatographic conditions were as follows; a methylsilicone fused silica capillary column (Durabond® DB-I, film thickness 0.25 µm, 30 m × 0.32 mm I.D.) programmed from 150°C to 310°C at 16°C/min; injection temperature 250°C; carrier gas (helium) at a flow rate of 20 ml/min.

Mass spectrometer operating conditions were as follows; temperatures (°C): separator 297, chamber 200; ionization voltage 70 eV; accelerating voltage 6.5 kV.

The monitored ions (*m/z*) were 217 3-HCH (internal standard); 303 and 305 γ-chlordene; 353 and 355 heptachlor epoxide (HCE); 373 trans-chlordane and cis-chlordane; 387 and 389 oxychlordane; 407 trans-nonachlor and cis-nonachlor.

PCBs Analysis: A Varian 2100 gas liquid chromatograph equipped with a nickel-63 electron-capture detector, fitted with 2% OV-1 (Gaschrom Q 100-120 mesh, 1.8 m × 2.0 mm I.D.) was used for analyses of PCBs. Temperatures of detector, injector and column was adjusted to 210°C, 210°C and 250°C, respec-

Table 1. Means of the concentration ( $\mu\text{g/kg}$ ) of chlordane and PCB in milk fat of lactating women

|                        | N <sup>a</sup> | Age<br>(years) | Panty | Y.A.T. <sup>b</sup><br>(years) | $\gamma$ -<br>chlor-<br>dene | trans-<br>chlor-<br>dane | cis-<br>chlor-<br>dane | oxy-<br>chlor-<br>dane | trans-<br>non-<br>achlor | cis-<br>non-<br>achlor | total<br>chlor-<br>dane <sup>c</sup> | HCE <sup>f</sup> | PCB |
|------------------------|----------------|----------------|-------|--------------------------------|------------------------------|--------------------------|------------------------|------------------------|--------------------------|------------------------|--------------------------------------|------------------|-----|
| Non-exposed<br>control | 7              | 26.3           | 1.7   | —                              | 3.1                          | 1.5                      | 2.1                    | 19.3                   | 25.6                     | 6.4                    | 58                                   | 21.1             | 261 |
| Exposed <sup>d,e</sup> | 15             | 28.7           | 1.8   | 1.8                            | 11.0                         | 7.9                      | 5.9                    | 33.6                   | 84.9                     | 19.1*                  | 162                                  | 54.9*            | 374 |
| group 1                | 3              | 29.7           | 2.0   | 0 < Y.A.T. < 1                 | 0.2                          | 0.4                      | 2.0                    | 10.4                   | 19.2                     | 4.4                    | 37                                   | 20.9             | 344 |
| group 2                | 6              | 29.8           | 2.0   | 1 $\leq$ Y.A.T. < 2            | 10.0*                        | 5.9***                   | 6.0*                   | 26.2                   | 67.9***                  | 17.6**                 | 134***                               | 43.5**           | 375 |
| group 3 <sup>e</sup>   | 6              | 27.0           | 1.5   | 2 $\leq$ Y.A.T.                | 17.3                         | 13.6                     | 7.7                    | 52.5*                  | 134.9*                   | 28.0**                 | 254*                                 | 83.2**           | 389 |

<sup>a</sup> Number of samples<sup>b</sup> Years after termite treatment of house<sup>c</sup> Total concentration of chlordane compounds except for heptachlor epoxide (HCE)<sup>d</sup> Group 1, 2, and 3 are combined<sup>e</sup> One sample ("Y.A.T." was eight years) was rejected (see text)<sup>f</sup> HCE = heptachlor epoxide\*  $P < 0.05$ . \*\* $P < 0.01$ . \*\*\* $P < 0.001$ ; significance level against control

tively. The carrier gas was nitrogen at a flow rate of 30 ml/min. PCB components whose retention times were greater than *p,p'*-DDE were quantified by using Kanechlor<sup>®</sup> mixture (Kanechlor<sup>®</sup> 300, 400, 500, 600 = 1:1:1:1, w/w) as the standard. The Ugawa method (Ugawa *et al.* 1973) was used for the quantitation of PCBs.

## Results and Discussion

Chlordane components in human milk were statistically higher in the exposed group than in the control group (Table 1). Levels of cis-nonachlor and HCE increased when the period (in years) after the termite treatment of the house was longer. To indicate the duration of exposure in the house where the termite treatment was carried out with chlordane, the exposure period (in years) is expressed with the term of "Y.A.T." which stands for the year after the treatment. Mean concentration of total chlordane in the milk of exposed women whose exposure was two years or longer (group 3) was 4.4 times higher than that of the control group.

Heptachlor epoxide (HCE) is a metabolite of heptachlor and technical chlordane contains approximately 7% heptachlor. Before its use for termite control, however, heptachlor had been used as an insecticide for agricultural purposes. These two origins of HCE contamination confused the source of HCE exposure, particularly for the exposed group. Therefore, the concentration of HCE was not added to the total chlordane in this paper.

Three major chlordane residues in human milk, namely oxychlordane, trans-nonachlor and cis-nonachlor, have been measured in several countries. These data for the exposure of general population are summarized in Environmental Health Criteria 34 (WHO 1984a). Levels of chlordane in

human milk in Canada (Mes and Davies 1978) and the USA (Strassman and Kuntz 1977; Barnett *et al.* 1979; Savage *et al.* 1976, 1981; FAO/WHO 1981) are higher than those in Japan. Miyazaki *et al.* (1985) analyzed chlordane residues in milk samples of Japanese women. Mean concentrations of oxychlordane, trans-nonachlor and cis-nonachlor were 16–20, 24–45 and 4–7  $\mu\text{g/kg}$ , respectively. Our measurements for nonexposed control group (Table 1) were within the same range as reported by Miyazaki *et al.* (1980, 1985, 1986a).

Technical chlordane is a multicomponent insecticide, and the results of our analysis are shown in Figure 1. Miyazaki *et al.* (1986b) reported that the pattern of chlordane residues in river water and sea water were quite similar to that of technical chlordane. Moderate changes of the component patterns in freshwater fish, marine shellfish and seawater fish were observed at each stage of the food-chain. Major changes were the increase in the relative percentages of trans-nonachlor, oxychlordane and cis-nonachlor, and the decrease of trans- and cis-chlordane. This tendency was even clearer in human milk samples (Miyazaki *et al.* 1986a).

Our results agreed with Miyazaki's in the changes of the component pattern. The patterns in human milk of both control and exposed group were different from that of technical grade chlordane. Relative percentages of major components of the technical chlordane, trans-chlordane, cis-chlordane and  $\gamma$ -chlordane, were very small compared to those of technical chlordane (Figure 1). These components are presumed to be metabolized relatively easily and to disappear rapidly from the human body. This tendency has been reported in animal experiments (Tashiro and Matsumura 1977, 1978; Barnett and Dorough 1974).

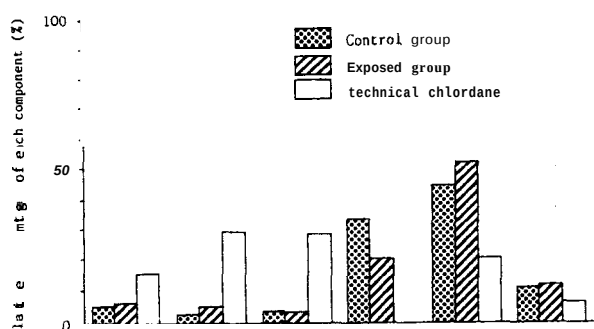


Fig. 1. Composition of chlordane compounds detected in the control and the exposed group. Relative percentages in total chlordane (58  $\mu\text{g/kg}$  for the control group, 162  $\mu\text{g/kg}$  for the exposed group) were calculated by using the mean concentration in Table 1.

The ratio of oxychlordane to total chlordane (relative %) of the exposed group was less than that of the control group. However, the ratios of *trans*- and *cis*-nonachlor to total chlordane of the exposed group were greater than that of the control group. Oxychlordane does not exist in technical chlordane, and is a metabolite formed in a living cell. Brimfield and Street (1979) proposed metabolic routes for *cis*- and *trans*-chlordane and the formation of oxychlordane in rat microsomes. Our results confirmed the reported trends. In another investigation, oxychlordane was biotransformed in experimental animals from *trans*-nonachlor via *trans*-chlordane (Tashiro and Matsumura 1978). In humans, however, it was deduced that *trans*-nonachlor is very slowly metabolized to oxychlordane via *trans*-chlordane (Kawano and Tatsukawa 1982; Miyazaki et al. 1986a). On the basis of the reported results, the pattern of chlordane components of the exposed group is more likely to be that of a technical chlordane when compared to the control group. Namely, *trans*-nonachlor is highly accumulated probably through domestic exposure to the technical chlordane in the home, and since the rate of biotransformation of *trans*-nonachlor into oxychlordane is very slow, the percentage of oxychlordane resulted in a smaller value.

Similar results were reported on the ratios between DDT residues in human milk. The ratio of DDE to DDT is lower in countries where DDT is still being used for pest control than in countries where its use has been severely restricted (Slorach and Vaz 1983). When the use of DDT is restricted, the level in vegetable foods falls rapidly. Human

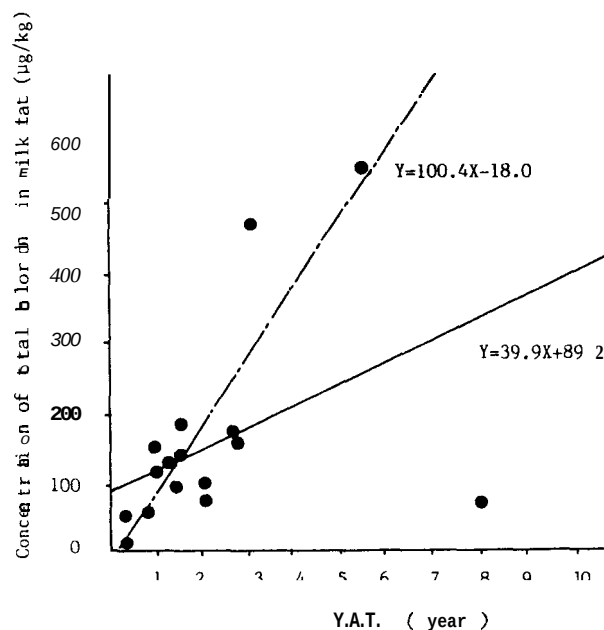


Fig. 2. Correlation between the concentration of total chlordane and the period after termite treatment (Y.A.T.). Linear regression equations were obtained for all the data (smaller slope line) or for the data after one sample was rejected (greater slope line). The data for eight years was rejected

exposure to its metabolite DDE continues, however, through the consumption of animal foods (e.g. fish) which continue to accumulate this metabolite from the environment longer after the prohibition of DDT. DDT is also metabolized gradually to DDE in the human body. The difference in the ratio of chlordane components between the control and the exposed group in this report indicates that the dwellers in the treated homes are exposed to chlordane which has been applied to the house. On the contrary, relative percentages of chlordane components found in the milk of the control group indicate that the major source of contamination is a biological material in foods.

Concentrations of total chlordane were compared to those of PCB, which was analyzed as a representative of organochlorine contaminants in human milk. Mean concentration of total chlordane in the control group was less than  $\frac{1}{4}$  of the mean PCB level (Table 1). However, it increased to a similar level to PCB when the period of exposure extended into two years or more.

Correlations between the concentration of total chlordane and the period of exposure are shown in Figure 2. When a linear relationship is assumed, a correlation coefficient ( $r$ ) for all the data was 0.417. The situation became more clear ( $r = 0.866$ ) when one result for the longest residence time (eight years) was excluded. This indicates that chlordane

**Table 2-1.** Correlation coefficient matrix between each item for the control group

|                               | Age    | Parity | Y.A.T. <sup>a</sup> | $\gamma$ -chlor-dene | trans-chlor-dane | cis-chlor-dane | oxy-chlor-dane | trans-non-achlor | cis-non-achlor | total chlor-dane <sup>b</sup> | HCE   | PCB |
|-------------------------------|--------|--------|---------------------|----------------------|------------------|----------------|----------------|------------------|----------------|-------------------------------|-------|-----|
| <b>Age</b>                    | —      |        |                     |                      |                  |                |                |                  |                |                               |       |     |
| Parity                        | 0.595  | —      |                     |                      |                  |                |                |                  |                |                               |       |     |
| Y.A.T. <sup>a</sup>           | —      | —      | —                   |                      |                  |                |                |                  |                |                               |       |     |
| $\gamma$ -chlordene           | -0.112 | -0.032 | —                   | —                    |                  |                |                |                  |                |                               |       |     |
| trans-chlordane               | -0.195 | 0.010  | —                   | 0.972                | —                |                |                |                  |                |                               |       |     |
| cis-chlordane                 | 0.005  | -0.319 | —                   | 0.766                | 0.743            | —              |                |                  |                |                               |       |     |
| oxychlordane                  | -0.106 | -0.508 | —                   | 0.373                | 0.392            | 0.864          | —              |                  |                |                               |       |     |
| trans-nonachlor               | -0.397 | -0.412 | —                   | -0.845               | -0.805           | -0.634         | -0.205         | —                |                |                               |       |     |
| cis-nonachlor                 | 0.822  | 0.431  | —                   | -0.084               | -0.239           | -0.200         | -0.404         | -0.280           | —              |                               |       |     |
| total chlordanes <sup>b</sup> | -0.492 | -0.833 | —                   | -0.159               | -0.127           | 0.346          | 0.720          | 0.502            | -0.575         | —                             |       |     |
| HCE                           | 0.380  | -0.164 | —                   | -0.096               | -0.045           | 0.476          | 0.680          | -0.059           | -0.064         | 0.440                         | —     |     |
| PCB                           | -0.156 | -0.732 | —                   | -0.236               | -0.242           | 0.198          | 0.488          | 0.459            | -0.091         | 0.785                         | 0.492 | —   |

<sup>a</sup> Years after the termite treatment of house<sup>b</sup> Total concentration of chlordane compounds except heptachlor epoxide (HCE)**Table 2-2.** Correlation coefficient matrix between each item for the exposed group

|                               | Age    | Parity | Y.A.T. <sup>a</sup> | $\gamma$ -chlor-dene | trans-chlor-dane | cis-chlor-dane | oxy-chlor-dane | trans-non-achlor | cis-non-achlor | total chlor-dane <sup>b</sup> | HCE    | PCB |
|-------------------------------|--------|--------|---------------------|----------------------|------------------|----------------|----------------|------------------|----------------|-------------------------------|--------|-----|
| <b>Age</b>                    | —      |        |                     |                      |                  |                |                |                  |                |                               |        |     |
| Parity                        | 0.454  | —      |                     |                      |                  |                |                |                  |                |                               |        |     |
| Y.A.T. <sup>a</sup>           | -0.184 | -0.017 | —                   |                      |                  |                |                |                  |                |                               |        |     |
| $\gamma$ -chlordene           | -0.147 | 0.189  | 0.761               | —                    |                  |                |                |                  |                |                               |        |     |
| trans-chlordane               | -0.059 | 0.346  | 0.686               | 0.880                | —                |                |                |                  |                |                               |        |     |
| cis-chlordane                 | -0.143 | 0.187  | 0.741               | 0.950                | 0.789            | —              |                |                  |                |                               |        |     |
| oxychlordane                  | -0.020 | 0.162  | 0.916               | 0.850                | 0.833            | 0.812          | —              |                  |                |                               |        |     |
| trans-nonachlor               | -0.040 | 0.280  | 0.849               | 0.889                | 0.925            | 0.856          | 0.955          | —                |                |                               |        |     |
| cis-nonachlor                 | 0.072  | 0.282  | 0.808               | 0.637                | 0.746            | 0.617          | 0.884          | 0.853            | —              |                               |        |     |
| total chlordanes <sup>b</sup> | -0.042 | 0.264  | 0.866               | 0.904                | 0.924            | 0.867          | 0.970          | 0.996            | 0.870          | —                             |        |     |
| HCE                           | -0.189 | 0.034  | 0.590               | 0.552                | 0.628            | 0.441          | 0.712          | 0.634            | 0.722          | 0.666                         | —      |     |
| PCB                           | 0.072  | -0.465 | -0.063              | -0.135               | -0.086           | -0.236         | -0.090         | -0.087           | -0.143         | -0.105                        | -0.248 | —   |

<sup>a</sup> Years after the termite treatment of house<sup>b</sup> Total concentration of chlordane compounds except heptachlor epoxide (HCE)

compounds are so persistent in the environment and stable in the human body that the accumulation continues for at least five or six years after the treatment of the house. As for the sample which was rejected, we have no reason to explain the low value. It is to be determined whether the atmospheric concentration of chlordane in the houses suddenly decreases after five years. Further investigations are necessary in this area.

Tables 2-1 and 2-2 show matrices of correlation coefficient between each item. A close relationship was noted between the concentrations of total chlordane and PCB in the control group ( $r = 0.785$ ). Each component of chlordane was less clearly related to PCB. In another investigation conducted on human milk of residents in Osaka from 1972 to 1977, a proportional correlation was

observed between the levels of PCB and pesticides, such as HCH, *p,p'*-DDT, *p,p'*-DDE (Yakushiji et al. 1979). The relationship suggested that PCB and the organochlorine pesticides were taken into the human body through similar foods. It is generally accepted that the major source of contamination of organochlorine compounds for the general population is the ingestion of foods. Intakes from drinking water and air are deemed to be negligible (Bergland 1972; Kolbye 1972). Watanabe et al. (1979) conducted a surveillance of dietary PCB intake of ordinary Japanese women, and indicated that approximately 70 to 90% of PCB was derived from eating marine fish. Jelinek and Corneliussen (1976) have also reported that fresh water fish are the major source of PCB exposure in the U.S.A. From the data, it was deduced that the main source of chlor-

Table 3. Mean value of the concentration ( $\mu\text{g/kg}$ ) of chlordane and PCB in milk fat of lactating women of first and second or more childbirth

|                         | N <sup>a</sup> | Age<br>(years) | average<br>Parity | Y.A.T. <sup>b</sup><br>(years) | $\gamma$ -<br>chlor-<br>dene | <i>trans</i> -<br>chlor-<br>dane | <i>cis</i> -<br>chlor-<br>dane | <i>oxy</i> -<br>chlor-<br>dane | <i>trans</i> -<br>non-<br>achlor | <i>cis</i> -<br>non-<br>achlor | total<br>chlor-<br>dane <sup>c</sup> | HCE <sup>f</sup> | PCB  |
|-------------------------|----------------|----------------|-------------------|--------------------------------|------------------------------|----------------------------------|--------------------------------|--------------------------------|----------------------------------|--------------------------------|--------------------------------------|------------------|------|
| Group (I) <sup>d</sup>  | 10             | 27.0           | 1.0               | 1.8                            | 7.2                          | 4.3                              | 4.2                            | 27.3                           | 57.3                             | 15.4                           | 116                                  | 53.5             | 441  |
| Group (II) <sup>e</sup> | 13             | 30.1           | 2.4               | 2.5                            | 12.7                         | 9.9                              | 6.7                            | 36.2                           | 100.5                            | 22.4                           | 188                                  | 51.8             | 312* |

<sup>a</sup> Number of samples<sup>b</sup> Years after the termite treatment of house<sup>c</sup> Total concentrations of chlordane compounds except heptachlor epoxide (HCE)<sup>d</sup> Mothers of first childbirth<sup>e</sup> Mothers of second or more childbirth<sup>f</sup> HCE = heptachlor epoxide\*:  $P < 0.05$ 

dane for the control group was marine products in their diet.

On the other hand, the concentration of total chlordane showed no correlation to that of PCB in the exposed group (Table 2-2). This indicates that the source of chlordane contamination was different from that of PCB. A good correlation is observed among chlordane components. The concentration of six chlordane components is also related to exposure time, as shown in Figure 2. From the data, the main source of chlordane in the exposed group was not from the intake of fish. The route of contamination for the exposed group was inferred partly to be from inhalation in the home of a contaminated atmosphere and partly to be absorption via the skin. Another inference may be the adsorption of chlordane to dietary materials during the period of storage in the kitchen.

The tendency that the main source of chlordane was not fish intake in the exposed group is also apparent in Table 3, where the results of the exposed group are classified into two groups by the parity number. The mean concentrations of PCB in milk fat obtained from mothers after the second or more childbirth ( $312 \mu\text{g/kg}$ ) was significantly lower than that from mothers after the first childbirth. It has been recognized that the excretion of PCB via breast milk is a major excretory route. In general, a 30 to 50% decrease in the PCB level of mother's milk is observed after five to six months of breast-feeding (Yakushiji et al. 1982). The half-life of the concentration of PCB in human milk was estimated to be approximately eight months (Yakushiji et al. 1978). In the present study on chlordane, however, such a tendency was not observed. Contrary to the case of PCB, total chlordane measured in the samples of group (II) was such higher than the group (I) (Table 3). Milk chlordane levels of the exposed group were strongly influenced many years after the treatment of the house for termite control.

Thus, it was clearly demonstrated that the chlor-

dane treatment of a house increased the contamination level of human milk. This study should arouse interests in the toxicological and environmental areas of chlordane pollution, especially in the treatment of houses. Further studies are strongly recommended.

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