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Indoor Air Concentration of Chlordane, and Its Seasonal Variation

F. Asakawa, F. Jitsunari, N. Takeda, Y. Manabe, S. Suna, I. Fukunaga

Department of Hygiene and Public Health, Kagawa Medical School, 1750-1, Ikenobe, Miki-cho, Kagawa 761-07, Japan

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Chlordane has been used mainly as a termiticide for more than 20 years in Japan. It has been detected in marine products and human breast milk (Miyazaki et al. 1980a, b; Yamagishi et al. 1981). Accordingly, the Japanese government banned the use of chlordane in September, 1986, because of its properties of non-reactivity and bioaccumulation, and its potential for deleterious effects on human health as a result of continuous exposure. Based on the criteria in the guidelines for assessment of carcinogenic risk of the United States Environmental Protection Agency (USEPA), chlordane was classified as a probable human carcinogen (Group B2) in 1988.

In Japan, chlordane was previously directly applied in residences and on residential real estate, as well as being used as a termiticide on wood materials during the manufacture of veneer board (Nishimoto 1983) . It is estimated that the total amount of chlordane applied was quite large. Accordingly, in residential environments in which chlordane or chlordane-treated materials were used, harmful indoor air pollution due to the volatilization of residual chlordane may be present. Several investigations conducted in the United States have revealed a wide distribution of chlordane concentrations, which in most cases are within 10 $\mu g/m^3$(Livingston and Jones 1981 ; Wright and Leidy 1982 ; Leidy et al. 1985). However, in Japan, no data for chlordane concentration are available, even though long-term monitoring of indoor air chlordane concentration is necessary to verify the safety of residential living circumstances.

The authors have developed a diffusion sampler monitoring method (Jitsunari et al. 1987) . In the present study, we investigated the indoor air chlordane concentration in residences in Japan and its seasonal variation.

MATERIALS AND METHODS

Technical chlordane consists of almost 20 different chemical compounds. In this study, we measured the concentration of the 5 compounds (*trans* and *cis* chlordane, *trans*- and *cis*- nonachlor, and heptachlor) which are the main components of technical chlordane.

Correspondence to: F. Asakawa

In measuring the air chlordane concentration, the diffusion sampler (PRO-TEK™ G-AA, DuPont) method reported in a previous study (Jitsunari et al. 1987) was used. This method is based on Fick's first law that M=SR x C x T, where M =collected weight (μg) , SR = sampling rate (m^3/day) , C = concentration (μg/m^3), and T= sampling time (days) . The value of SR when samples were obtained without a protective bilateral cover was 0.04610 for heptachlor, 0.02400 for *trans*-chlordane, 0.02210 for *cis*-chlordane, 0.02020 for *trans*-nonachlor, and 0.00686 for *cis*-nonachlor.

Analysis of chlordane was performed as follows:
The activated charcoal was removed from the sampler, and placed in a 2-ml vial. Then, 1.5 ml of benzene was added to the vial as the extraction solvent.
The vial was capped and then shaken gently for 0.5~1 hr. The benzene extract was then poured into a sampling bottle.
This solution was analysed and quantified by gas chromatography (GC) . The collected weight (M) of each compound was determined and used to calculate the respective air concentrations(C).

The gas chromatograph (Shimazu GC-7A, ECD) was attached to a data recorder (Shimazu, Chromatopack C-R2A). The column (3 mm i.d x 2 m) was packed with 2% OV-1 (80/100 mesh Gas-chrom Q) . The column temperature was 190 °C, and the inlet temperature was 240 °C.

Reagents and organic solvents were of high purity and suitable for trace analysis. For the standard solutions of chlordane compounds, heptachlor and *trans*- and *cis*-chlordane from Wako Pure Chemical, and *trans*- and *cis*- nonachlor from Nanogen Co., were used. Each solutions was prepared with n-hexane.

For the investigation of air concentration of chlordane, 12 houses were selected. The number of years since chlordane had been applied as a termiticide was 1 to 5 years in 9 houses. One house which had never been treated with chlordane as a termiticide and 2 old houses in which a termiticide had never been used were used as controls. Air samples were acquired at 3 locations in each house by suspending a sampling unit under the first floor, in a room on the first floor, and in a room on the second floor. Samples were obtained for a 1 - month period in August.

In addition, outdoor air chlordane concentration was monitored near the house built 1 year previously without the use of a termiticide.

Indoor air concentration of chlordane in 5 of the houses was monitored for a one-year period from September, 1986, to August, 1987. In House A, 5 years had elapsed since termiticide treatment because of termite damage, in House B, 3 years had elapsed since preventive termiticide treatment, in House C, 1 year had elapsed since termiticide treatment because of termite damage, in House D, no chlordane had been applied, and House E was an old house to which a termiticide had never been applied. Sampling units were fixed under the floor and in a room on the first floor, and exchanged every month. Outdoor air chlordane concentration was also continuously monitored.

RESULTS AND DISCUSSION

Table 1 shows the indoor air chlordane concentration at each location in the houses. No chlordane was detected in the old house built without the use of termiticide. On the other hand, chlordane was clearly detected in the houses in which termiticide had been applied. The chlordane concentration ranged from 0.14 to 32 $\mu g/m^3$ under the floor, 0.041 to 2.0 $\mu g/m^3$ on the first floor, and 0.032 to 2.1 $\mu g/m^3$ on the second floor. In general, houses which had been treated with termiticide within the previous year showed high indoor air chlordane concentration. One house which was treated with termiticide 5 years ago, still showed detectable chlordane concentration, which was 9.9 $\mu g/m^3$ under the floor, 1.0 $\mu g/m^3$ on the first floor, and 0.43 $\mu g/m^3$ on the second floor. These results indicate that indoor air chlordane concentration remains detectable for a considerably long period. The finding was that chlordane concentration was highest under the floor, followed by on the first and second floors, respectively. This may be due to the fact that chlordane termiticide treatment was generally applied mainly under the floor.

In the house which had been built without termiticide, little chlordane was detected under the floor, but considerably more was detected on the first floor, even though no chlordane was detected in the outdoor air. These results suggest the presence of another indoor source of chlordane. The floor boards of this house consisted of lauan veneer board, which constitutes 90 % of the veneer board used in Japan, and is usually treated with various pesticides during its production because of its vulnerability to insect damage (Nishimoto 1983). We therefore consider that the chlordane detected in this house originated from the lauan veneer boards for floor and other building materials.

The results of this investigation indicate that chlordane in indoor air originates from indoor chlordane residue by the process of volatilization. Moreover, to elucidate the scope of contamination of indoor air by chlordane more clearly, further continuous monitoring is required.

Table 1 Chlordane concentration (total for 5 compounds) detected in the ambient indoor air of twelve houses

Number of years after termiticide application	No. of houses sampled	Under the floor [a]	1st floor [a]	2nd floor [a]
4~5 years	3	7.3±2.9	0.44±0.40	0.24±0.14
2~3 years	3	4.0±3.6	0.102±0.066	0.068±0.045
~1 year	3	17±11	0.85±0.80	0.85±0.87
~1 year [b]	1	0.025	0.27	0.079
Controls [c]	2	N.D.[d]	N.D.[d]	N.D.[d]

a : Mean with standard deviation, $\mu g/m^3$

b : House built within the previous year without temiticide

c : Old houses built without termiticide

d : None detected at the quantitative detection limit of 0.005 $\mu g/m^3$

Figure 1 shows the monthly concentrations for 5 chlordane compounds and for total chlordane (total for 5 compounds) in Houses A-D. House C showed the highest chlordane concentration, followed by House A. The chlordane detected in House D was suspected to originate from the veneer floor boards. No chlordane was detected outside House D in any season. No chlordane was detected in House E.

The chlordane concentration under the floor showed almost a linear decrease from summer to winter, while that on the first floor showed a peak in July, decrease in August, and increase in October, followed by decrease. While there was some difference between the variation of chlordane concentration under the floor and that on the first floor, it showed seasonal variation at each location. The range and mean seasonal variation under the floor and on the first floor, respectively, were as follows; House A: 1.04 ~ 12.5 $\mu g/m^3$ (5.41 $\mu g/m^3$) and 0.17 ~ 1.51 $\mu g/m^3$ (0.68 $\mu g/m^3$) ; House B: 0.58 ~ 11.9 $\mu g/m^3$ (4.90 $\mu g/m^3$) and 0.09 ~ 0.38 $\mu g/m^3$ (0.24 $\mu g/m^3$) ; House C: 8.73 ~ 34.4 $\mu g/m^3$ (21.6 $\mu g/m^3$) and 0.59 ~ 3.77 $\mu g/m^3$ (1.96 $\mu g/m^3$) ; and, House D: not detected and 0.11 ~ 0.43 $\mu g/m^3$ (0.23 $\mu g/m^3$) . The concentration of chlordane under the floor showed greater variance than that on the first floor in most cases.

Of the 5 chlordane compounds, *ttans*-chlordane showed the highest concentration in both locations in each house, and accounted for 45% of the mean total chlordane concentration under the floor and 35% of that on the first floor. The heptachlor concentration in Houses C and D was almost the same as that of *trans*-chlordane. On the other hand, the *cis*-chlordane concentration was comparatively low in both locations in each house. All chlordane compounds showed similar seasonal variations.

Figure 2 shows the average monthly temperature in the prefecture. The pattern of seasonal variation of indoor air chlordane concentration, particularly of that under the floor, was strongly correlated ($p<0.01$) with that of average monthly temperature recorded in the prefecture (Table 2). This finding supports the hypothesis that air chlordane concentration is associated with the volatilization of chlordane residue, which is a function of the air temperature (Guish and Rouch 1986 ; Norback et al. 1990). The stronger correlation of temperature with the concentration under the floor than with that on the first floor is thought to be due to the influence of such factors as air conditioning and air ventilation in winter and summer.

The results of the present study indicate that chlordane concentration must be monitored over the course of a year, or at least in the summer and winter, to obtain a more complete data set.

It is difficult to evaluate pesticide concentration findings obtained in various countries, since methods of termiticide treatment, building structural composition, and life-styles vary. The chlordane concentrations of indoor air reported in several studies conducted in the United States were 0.4~264 $\mu g/m^3$ (Livingston and Jones 1981) , <3.5 $\mu g/m^3$ in 86% of the homes studied (Wright and Leidy 1982) , and 0.05~9.90 $\mu g/m^3$ (Leidy et al. 1985).

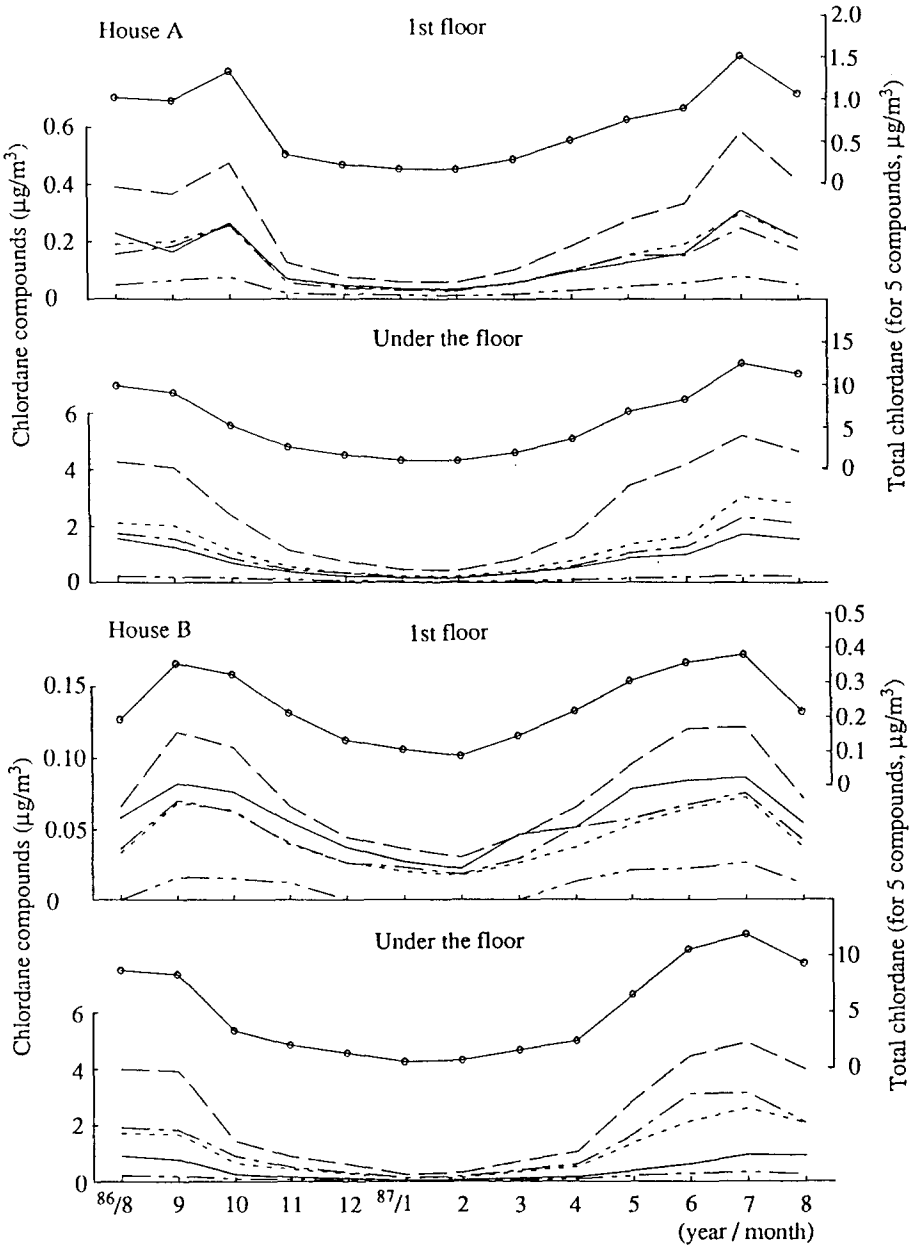
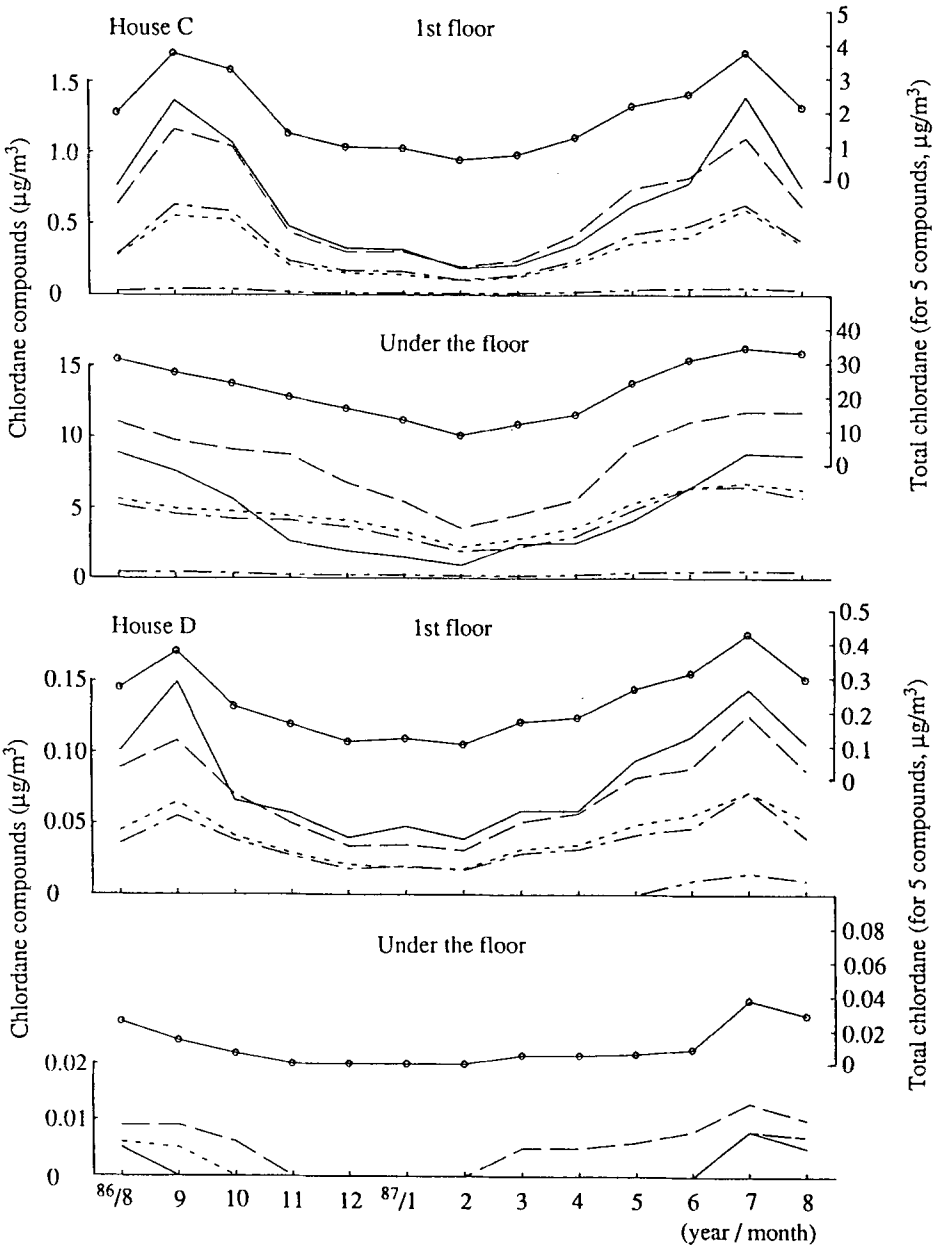


Figure 1 Monthly concentrations of chlordane in house ambient indoor air

total chlordane
heptachlor
trans-chlordane
cis-chlordane
trans-nonachlor
cis-nonachlor


House C
1st floor
Under the floor
House D
1st floor
Under the floor
Chlordane compounds (μg/m³)
Total chlordane (for 5 compounds, μg/m³)
(year / month)
86/8
9
10
11
12
87/1
2
3
4
5
6
7
8

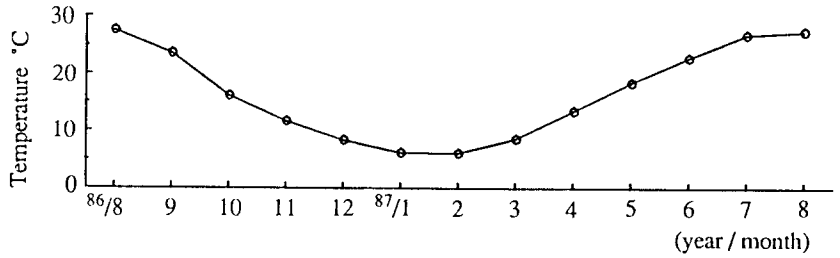


Figure 2 Average monthly temperature in the prefecture

Table 2 Correlation between temperature and indoor air chlordane concentration (total for 5 compounds)

House	Under the floor	1st floor
A	0.988**	0.876**
B	0.957**	0.829**
C	0.959**	0.836**
D	-	0.937**

** : $p<0.01$

The occupants of the houses we studied did not report any adverse health effects. However, Menconi et al. (1988) found a statistical relationship between air chlordane concentration and the incidence of such diseases as migraine, bronchitis, and sinusitis by comparing the incidences in homes with <1, 1~5, and >5 $\mu g/m^3$ chlordane concentration. In 1979, the USA National Academy of Sciences issued a temporary safety limit of 5 $\mu g/m^3$ for indoor air chlordane concentration, but did not assure that no biological effects would occur in humans exposed at below this level.

In a United States study of outdoor air chlordane concentration,which has not been monitored in Japan, the maximum value recorded at 10 locations was 7.3 ng/m^3 (average 0.4 ng/m^3) (Kutz 1983). Indoor air chlordane concentration is therefore about 1000 times higher that outdoors. The average time that a person spends at home is generally 10~20 hours a day, and is particularly long for infants and the elderly. Although the Japanese government banned the use of chlordane in September, 1986, our results indicate that chlordane is present in houses treated with termiticide and / or containing veneer board, and suggest that continuous exposure will continue in the future.

We conclude that chlordane is detected in the indoor air of houses which have been treated with termiticide, as well as houses constructed with veneer board treated with chlordane. Seasonal variation of indoor air chlordane concentration which was high in summer and low in winter, was strongly correlated with the average monthly air temperature in the prefecture. The concentration of chlordane was

highest in the house which had been most recently treated with termiticide.

Since the USEPA classified chlordane as a Group has B_2 carcinogen, which, even at low concentration, can induce chronic health effects in those continuously exposed for a long period, the issue of chlordane in previously treated houses cannot be ignored.

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