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Levels of Airborne Chlordane and Chlorpyrifos in Two Plenum Houses: Saranex S-15 as a Vapor Barrier

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In the 1960's the wood industry developed a plenum-type structure that used a down-flow furnace and air-conditioning unit to distribute heated or cooled air to a plenum underneath a wood floor from which air was circulated throughout the house by air vents in the floor or baseboards. The most efficient means of protecting these structures from termite damage is the treatment of the footings and the soil adjacent to the foundation with termiticides such as chlordane or chlorpyrifos. However, several studies have shown that chlordane residues are measureable in conventional houses treated with chlordane by subslab injection or post-construction exterior ditching (Lillie 1981; Wright and Leidy 1982). Because treatment of plenum houses involves the application of termiticide both inside and outside of the plenum air space, attention in the present study was directed to the use of a plastic film barrier to impede the movement of volatilized termiticides from treated soil into the plenum air space. The results of pilot studies in this laboratory, which examined films of polyethylene (1 and 6 mils thick) polyvinylidene chloride and polyethylene (Saranex S-14, 2 mils thick), and polyamide (Capran 77-C, 1 mil thick) for their effectiveness as barriers to chlordane and chlorpyrifos penetration, indicated that the Saranex film was the most effective of these. For this reason it was selected as the film barrier to be used in this study of plenum houses which were constructed, treated and monitored to determine its effectiveness in preventing vapors of the termiticides chlordane and chlorpyrifos from appearing in the ambient air of the house.

MATERIALS AND METHODS

Two plenum houses (24 x 40 ft), each containing two rooms or units separated by a standard wall, were constructed especially for this study. One house was constructed in Athens, GA, on poorly drained clay soil and the second house was constructed in Gainesville, FL, on well-drained sandy soil. Prior to the construction of the houses, soil samples (3 x 24 in cores) from each site were taken near the four corners of the two proposed structures and analyzed for the presence of chlordane and chlorpyrifos. Following the de-

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termination that these samples contained residues of these termiticides below 10 ppb, construction was begun. After the construction of the wood foundations of each house had been completed, the soil along the inside and outside perimeters of the foundation walls and along both sides of the interior bearing wall of each unit was treated with chlordane or chlorpyrifos at a rate of 4 gals/10 linear ft per ft of depth. The gravel footings were treated with termiticide at the rate of 1.25 gals/ft^3 of footing. Technical chlordane in emulsifiable concentrate (Gold Crest C-100, Velsicol Chemical Corp.; 8 lbs/gal) and chlorpyrifos in emulsifiable concentrate (Dursban TC, Dow Chemical; 4 lbs/gal) were diluted with water to give a 1% emulsion according to the label instructions.

Immediately after termiticide treatment, 80 in-wide sheets of Saranex S-15 film (Dow Chemical; 4 mils thick) were placed over the entire plenum area and the ends draped over and secured to the top of the foundation studs along the perimeter of each unit. Each film sheet was overlapped along its length by at least one foot of the width of an adjacent sheet. During the final construction phase, Saranex S-15 film sheets were secured to the full area of both sides of the center wall separating each unit before the installation of the dry wall.

Each unit in both houses was independently heated and cooled. The inside temperature was maintained between 70 and 79°F in the Athens house (except for a 2-month period between November 1985 and January 1986 when the thermostat was set at 65°F) and between 70 and 81°F in the Gainesville house during the study period.

Air sampling was conducted at zero time (the earliest possible time following completion of the structure), at weekly intervals for the first month, at monthly intervals from month 1 to month 4, and at bimonthly intervals from month 6 to month 18 (Athens house) or to month 14 (Gainesville house). The chlordane unit of the Athens house was also sampled at month 21.

Air sampling tubes (Orbo-44, Supelco), containing 150 mg of Chromosorb 102 sorbent which was separated into 100 and 500 mg sections by glass wool, were used to sample air in the two living units and in the air outside the house at a point approximately 30 ft from and directly adjacent to the dividing wall of the structure. During sampling, the sorbent tubes were attached to Millipore vacuum pumps set at a flow rate of 4 L/min for 2 hrs. Two pumps were located in each unit. One pump was positioned approximately ten ft from the outside door and the other, ten ft from the dividing wall between the two units. At the beginning of each sampling period all pumps were started within a period of 6 min in the following order: the two pumps in the chlordane unit, the two pumps in the chlorpyrifos unit, and the single outside pump. After one hour, one blank sorbent tube was opened in each of the two units and immediately capped. Also at this time temperature and humidity readings were made inside both units and outside with a sling psychrometer. Barometric pressure was also recorded at this

time. At the end of the two-hour period, the sampling was terminated in the same order as it had been started. The flow rate of air through the sampling tubes was recorded before the sampling was terminated and where this flow had changed from the initial 4 L/min setting, the average of the two flow rates was used to compute the total air volume sampled. Sampling tubes were immediately capped and stored at 0°C until analysis.

At the time of analysis, each sampling tube was scored with a file, broken, and the contents of the 100 mg sorbent portion were emptied into a 50-mL Erlenmeyer flask. Fifty mL of hexane (Pesticide grade, Fisher Scientific) were added to each flask and the contents were sonicated for 30 min After sonication, the sorbent was separated from the hexane by filtration through a sintered glass funnel. Each Erlenmeyer flask was washed three times with 10 mL of hexane and the washes were added to the filtrate of sorbent extract. The filtrate was then reduced by evaporation in Kuderna-Danish flasks to approximately 5-10 mL. One μL of the concentrated sample was injected onto a capillary column (OV-101 on fused silica, 0.2 mm x 12 m) for analysis on a Hewlett-Packard 5880A or 5890 gas chromatograph (^{63}Ni electron capture detector; oven temperature program 70°C to 205°C at 5°C/min; splitless injection with a 45 sec delay; detector temperature 300°C, injector temperature 175°C and 5% argon/methane carrier flow of 1 mL/min).

Chlordane, a mixture of chlorinated hydrocarbons, was analyzed for six of its components, Compound C, heptachlor, Compound E or γ-chlordene, γ- or trans-chlordane, α- or cis-chlordane, and α- or trans-nonachlor (Brady and Atallah 1986). Standards of these compounds were generously provided by Velsicol Chemical Corp.

RESULTS AND DISCUSSION

Figures 1 and 2 illustrate the results of analyzed air samples taken from the Gainesville, FL, and Athens, GA, plenum houses during the first 428 and 684 days, respectively, following termiticide application. The values on these graphs represent primarily Compound C and heptachlor (Table 1). The detection of heptachlor reflects the presence of this compound in Goldcrest chlordane (listed as approximately 18% by weight). The highest concentration of total chlordane components in the Gainesville, FL, plenum house (2.09 $\mu g/m^3$) was in the chlordane unit air samples taken 204 days following the application of termiticide and in the chlordaneunit air samples from the Athens, GA, plenum house (2.04 $\mu g/m^3$) taken 684 days following the application of termiticide.

Air samples collected from both units of the Gainesville, FL, house during the first two sampling periods (37 and 44 days post-treatment) an from both units of the Athens, GA, house during the first six sampling periods (76, 83, 90, 97, 104, and 132 days post-treatment) contained no measureable residues of chlordane components or chlorpyrifos. During the period of this study no compound E, trans-chlordane, cis-chlordane or trans-nonachlor was detected in the chlordane or chlorpyrifos units of the Athens, GA,

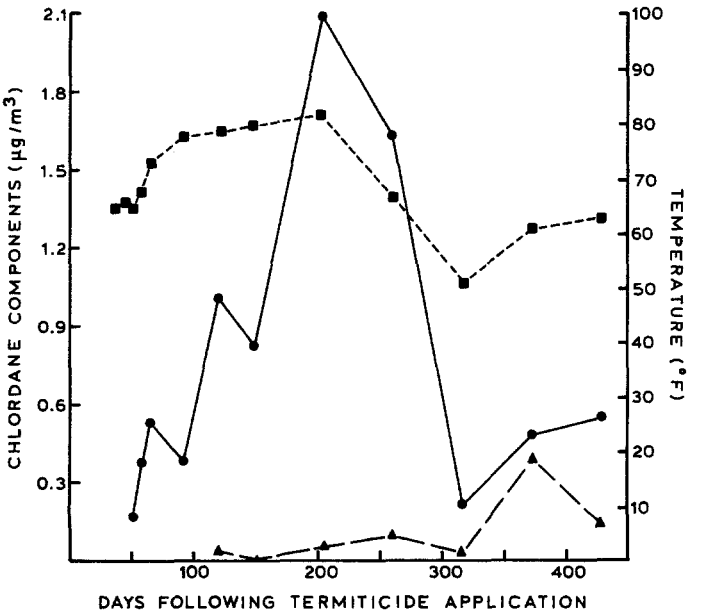

Figure 1. Chlordane (total of six components) in the air of the Gainesville, FL. plenum house. Chlordane unit, ●——●, chlorpyrifos unit, ▲——▲, outside ambient temperature (mean of preceding 7 days), ■-----■.

house. The detection of chlordane components in the chlorpyrifos unit (Figures 1 and 2 and Table 1) shows the crossover of volatilized chlordane from the chlordane unit to the chlorpyrifos unit. This crossover may have occurred in the gravel footing area between the two units or through the dividing wall above and/or below the floor.

Although there are no published values of airborne chlordane and chlorpyrifos in the living space of plenum houses where a film barrier has not been used following termiticide treatment, a recent study of airborne chlordane in the living space of houses in which the underfloor crawl space areas were treated with this termiticide provides some data with which the results of the present study may be compared. In the absence of a plenum vapor barrier, airborne chlordane would be expected to be present in greater amounts in the living space of chlordane-treated plenum houses where underfloor air is constantly circulated throughout the living space by the heating and cooling system than in the living space of chlordane-treated crawl space houses where the underfloor air is not similarly circulated. In 12 houses in which crawl space areas were treated with a 1% emulsion of chlordane according to label instructions 1 to 5 years prior to measurement, airborne chlordane levels (excluding heptachlor) were greater than 1.0 $\mu g/m^3$ in 11 out of 12 kitchen areas and in 10 out of 12 bedroom areas surveyed (range, 1.6-5.7 $\mu g/m^3$; Leidy et al. 1985). It is

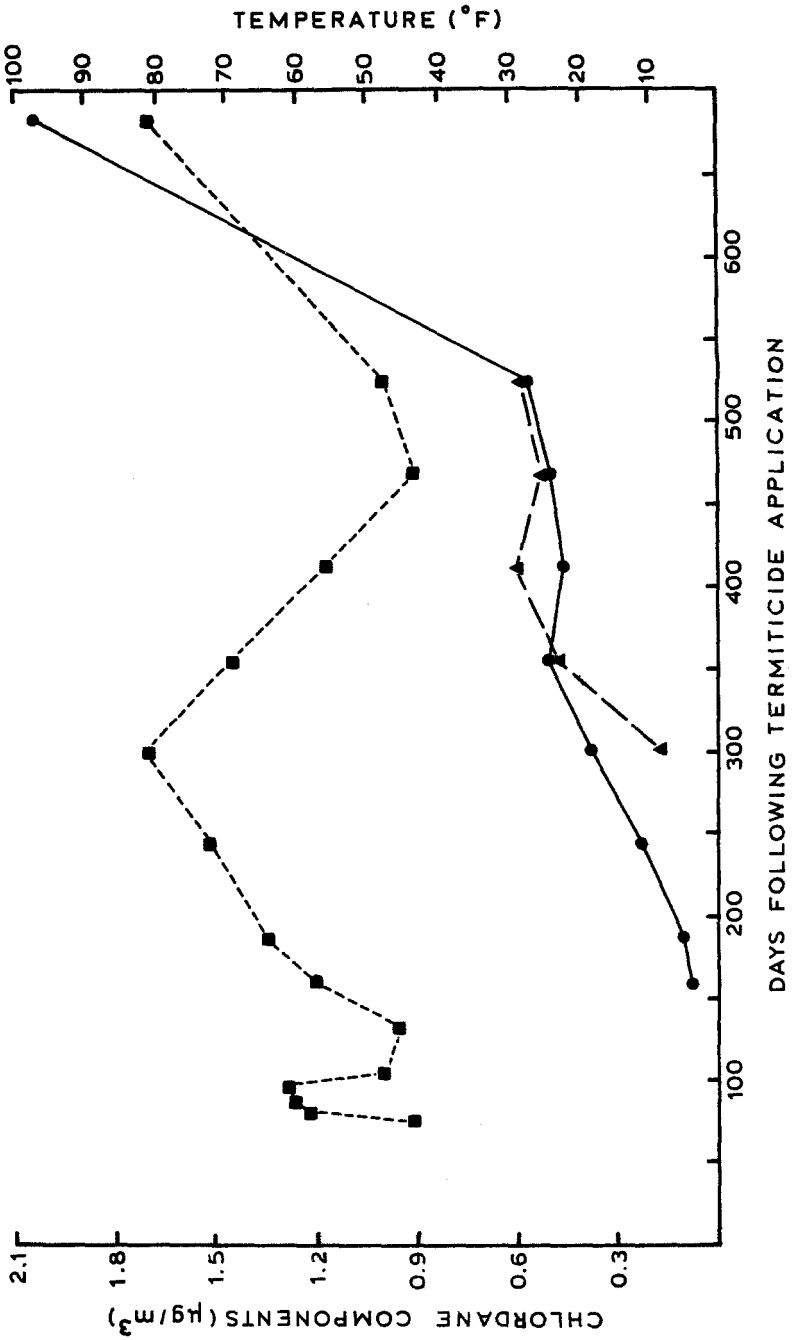

Figure 2. Chlordane (total of six components) in the air of the Athens, GA. plenum house. Chlordane unit, ●——●, chlorpyrifos unit, ▲——▲, outside ambient temperature (mean of preceding 7 days), ■----■.

Table 1. Concentrations of Chlordane Components and Chlorpyrifos in the Air of Plenum Houses in Gainesville, FL and Athens, GA.

CHLORDANE UNIT

Days	Location	Cmpd C	Heptachlor	Cmpd E	trans CD	cis CD	trans nonachlor	C
50	Gainesville	*	0.169	*	*	*	*	
57	Gainesville	0.069	0.334	*	*	*	*	
64	Gainesville	0.097	0.437	*	*	*	*	
92	Gainesville	0.097	0.289	*	*	*	*	
120	Gainesville	0.172	0.452	0.012	0.335	0.013	0.024	0.
149	Gainesville	0.250	0.572	*	*	*	*	
204	Gainesville	0.465	1.277	0.107	0.138	0.058	0.049	0.
260	Gainesville	0.333	0.883	0.097	0.188	0.074	0.054	
316	Gainesville	0.105	0.337	a	*	*	*	
372	Gainesville	0.153	0.331	a	*	*	*	
428	Gainesville	0.164	0.388	a	*	*	*	a
160	Athens	*	0.081	*	*	*	*	
188	Athens	0.033	0.072	*	*	*	*	
244	Athens	0.066	0.167	*	*	*	*	
300	Athens	0.088	0.295	*	*	*	*	
356	Athens	0.146	0.369	*	*	*	*	
412	Athens	0.084	0.380	*	*	*	*	
468	Athens	0.126#	0.383	a	*	*	*	
524	Athens	0.141	0.427	a	*	*	*	
684	Athens	0.345	1.691	a	*	*	*	

CHLORPYRIFOS UNIT

Days	Location	Cmpd C	Heptachlor	Cmpd E	trans CD	cis CD	trans nonachlor	C
50	Gainesville	*	*	*	*	*	*	
57	Gainesville	*	*	*	*	*	*	
64	Gainesville	*	*	*	*	*	*	
92	Gainesville	*	*	*	*	*	*	
120	Gainesville	*	0.037	*	*	*	*	
149	Gainesville	*	*	*	*	*	*	
204	Gainesville	*	*	*	0.025	0.015	0.029	0.
260	Gainesville	0.034	0.067	*	*	*	*	0.
316	Gainesville	*	0.065	a	*	*	*	
372	Gainesville	0.095#	0.289	a	*	*	*	
428	Gainesville	0.015	0.133	a	*	*	*	
160	Athens	*	*	*	*	*	*	
188	Athens	*	*	*	*	*	*	
244	Athens	*	*	*	*	*	*	
300	Athens	*	0.168	*	*	*	*	
356	Athens	0.117	0.365	*	*	*	*	
412	Athens	0.094	0.567	*	*	*	*	
468	Athens	0.040	0.498	a	*	*	*	a
524	Athens	0.053	0.538	a	*	*	*	a
684	Athens	a	a	a	a	a	a	a

* - Below detection limits.
a - Not determined.
- One sample only.

interesting to note that these values are clearly greater than those measured in the present study where the sum of chlordane components other than heptachlor never exceeded 1.0 $\mu g/m^3$ in either house.

The results of an ongoing study by Dow Chemical Company of chlorpyrifos concentrations in the living space of five plenum houses following plenum treatment with this termiticide show the presence of airborne chlorpyrifos in four of five houses (0.07-1.38 $\mu g/m^3$) 7 to 90 days following plenum treatment (personal communication, Dr. D. Barber, Dow Chemical Co.). Also, a recent study of sixteen houses, eight of which were crawl-space houses treated with 1% chlorpyrifos showed average airborne chlorpyrifos concentration ranging from 0.73 to 2.08 $\mu g/m^3$ in living spaces sampled from the time of application to 2 years following application (Leidy and Wright 1987). In the present study, chlorpyrifos was never detected in the Athens, GA, house and was detected on only three occasions in the Gainesville, FL, house at levels ranging from 0.037 to 0.175 $\mu g/m^3$.

No chlordane components or chlorpyrifos was detected in air samples taken outside the Gainesville, FL, plenum house. Air samples taken outside the Athens, GA, plenum house contained chlordane components on only two occasions: day 524 following initial treatment, heptachlor at 0.134 mg/m^3; and day 684 following initial treatment, Compound C at 0.009 $\mu g/m^3$ and heptachlor at 0.025 mg/m^3.

There was a statistically significant correlation ($p<0.05$, $r=0.998$; Pearson's correlation coefficient) between the average ambient outside temperature for the seven days preceding the sampling date and the ln of the concentration (ng/m^3) of chlordane components ($T=a + be^c$, where T=absolute temperature, a and b are constants, and $c=ng/m^3$) in the chlordane unit of the Athens, GA, plenum house when the values for the last three sampling dates were analyzed. There was no such similar significant relationship between ambient outside temperature and chlordane components in the chlordane unit of the Gainesville, FL, plenum house or in the chlorpyrifos units of either house.

The range of averaged recoveries of chlordane components and chlorpyrifos added to 100 mg of resin removed from a previously unopened resin tube and carried through the analytical procedure was 82.6 to 104.6%. The amount of the termiticides used in recovery experiments ranged from 0.5 ng to 100 ng of individual chlordane components and chlorpyrifos. These quantities are equivalent to those that would be measured in tubes collecting air containing 1.04 to 208 ng of termiticide/m^3 and flowing through a sampling tube at a rate of 4 L/min.

The results of the present study demonstrate that the use of a Saranex S-15 4-mil film barrier as described in this study effectively inhibited penetration of chlordane and chlorpyrifos into the air space of the two plenum houses studied such that detect-

able airborne levels of these termiticides and their components were well below the interim guideline levels recommended by the Committee on Toxicology of the National Academy of Sciences (1982). The interim guideline levels for continuous exposure have been set at 10.0 $\mu g/m^3$ for chlorpyrifos, 2.0 $\mu g/m^3$ for heptachlor, and 5.0 $\mu g/m^3$ for chlordane (the sum of cis- and trans-chlordane isomers).

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