Bull. Environ. Contam. Toxicol. (1990) 45:840–846

Bulletin of Environmental Contamination and Toxicology

Volatilization and Redistribution of Propoxur from a Formulation Used for Control of the German Cockroach

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Redistribution of toxicants is probably the most important aspect of volatilization formation during or after pesticide applications (Hislop 1987). New surfaces may be contaminated by condensation of volatilized toxicants (Ebeling et a. 1974). However, little is known concerning the impact of exposures of urban pests to toxicants that are redistributed after an insecticide application. Presumably the effects would be sublethal. Sublethal exposures of an insect may cause behavioral responses (Haynes 1988) and, possibly, the development of behavioral resistance (Gould 1984; Lockwood et al. 1984).

The purpose of the present study is to investigate propoxur volatilization and redistribution from a commerical propoxur formulation used to control the German cockroach, Blattella germanica (L.). A propoxur-in-oil formulation was used because dispersal induced by vapors of this particular formulation was due mainly to the active ingredient (Wooster and Ross 1989). Dispersal was rapid in response to vapors of the complete formulation but that to vapors of the solvent system (placebo) was relatively slow and less extensive. The present experiments were conducted similarly to those on dispersal. The dispersal apparatus was described by Wooster et al. (1990).

MATERIALS AND METHODS

A modified technique for capturing volatiles in polyurethane foam plugs by passing air continuously over samples in a closed system (Turner et al. 1977; Petruska et al. 1985) was combined with the use of radiolabelled propoxur to estimate air concentration and condensation. The air scrubbing system used to

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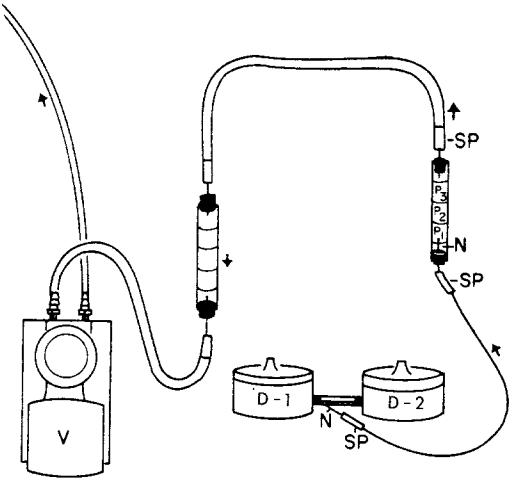

Figure 1. Air scrubbing system used to capture radiolabelled propoxur attached to an apparatus used to study dispersal of cockroaches induced by volatiles of a propoxur oil formulation. D-1 and D-2: dish-1 (aggregation) and dish-2 (dispersal) of the dispersal apparatus; V - vacuum pump; SP - syringe parts; P1,2,3, - polyurethane plugs; N - needle. Arrows indicate direction of air flow from dish-1 through the air scrubbing system.

capture radiolabelled propoxur was attached to a dispersal test apparatus by an 18 gauge (ga) (2 mm i.d.) needle (Fig. 1). The needle was pushed through a screen in a tube connecting two parts of the apparatus, the "aggregation dish" (dish-1) in which cockroaches were exposed to pesticide vapors and the "dispersal dish" (dish-2) into which they could move to escape the vapors (Wooster and Ross 1989). The needle tip was placed just inside an opening leading from dish-1 into the connecting tube. Air flow from the needle tip was measured by an air flow meter and adjusted by use of a hose clamp. The needle was connected to a syringe tube (1 cc TB Plastipak Luer-Lok). The open end of the tube was connected to a piece of Tygon tubing. The Tygon tube led to another syringe tube which was connected to an 18 ga needle that was pushed through a neoprene stopper. The stopper-needle combination was used to seal one end of a glass tube (10 cm long by 1.3 cm diam) that contained three polyurethane plugs (3 cm long by 1.3 cm diam). For safety, vapors from dish-1 were passed through another glass tube containing polyurethane plugs before passing into a vacuum air pump. The plugs were prewashed in hexane and dried before placing them in the tube. A glass plate (20 cm by 20 cm) was placed underneath a screened opening in

the tube connecting dish-1 and dish-2 dishes to collect possible deposits from vapors vented from the connecting tube. Both dishes were covered with a Petri dish that had been modified by attaching a glass tube (3 cm long; 2 mm i.d.) projecting upward from the center.

Vapors were introduced into dish-1 by placing a filter paper collar (1 cm by 43 cm) impregnated with 300 ul of a 1% propoxur-in-oil formulation (Octogon Process Inc. Lot C-4043) near the top, as in the dispersal experiments (Wooster and Ross 1989). The formulation was spiked with carbonyl-^{14}C-propoxur (Mobay Chemical Corp., S.A. 17.98 mCi/mole) in benzene at the rate of 5 uCi per 300 ul. After placement of the collar, a plug blocking the opening from dish-1 into the connecting tube was removed, the lid on the dish replaced, and the pump turned on. A flow rate of 150 ml/min was selected. The turnover rate in dish-1 was 6.6 volumes/30 min. Pesticide volatiles were scrubbed from the air and trapped in the foam plugs. Scrubbing was done at 30 minute intervals for 2 hr. The pump was turned off at the end of each interval and a clean tube with three fresh plugs was placed in line. Propoxur trapped in the polyurethane plugs during each 30 min interval was extracted by soaking in hexane (30 ml/plug). At the end of 2 hr, the apparatus was disassembled. The separate parts were rinsed in 30 ml of hexane, placed individually in 30 ml of hexane, and soaked for a minimum of 30 min. Hexane extracts from the plugs and apparatus parts were concentrated down to 1 ml using a nitrogen flow evaporator at 40^{o} C. A 100 ul aliquot from each 1 ml concentrate was placed in 8 ml of scintillation fluid (Ecoscint) and counted in a scintillation counter (Beckman LS 3150T). Sample counts were corrected for background and quenching. Three 1 cm x 1 cm samples of each filter paper strip were placed in 8 ml of scintillation fluid and counted to determine any residual activity following hexane extraction of the filter paper. Air and surface propoxur concentrations were estimated on the basis of the specific activity of recovered radiolabelled propoxur to that applied in the propoxur solution. The experiments were conducted at 22 $\pm$ 3^{o}C and 70 $\pm$ 5% relative humidity.

The above procedures were used in an experiment on container surface concentrations, except that air scrubbing was not done.

RESULTS AND DISCUSSION

Table 1 shows the amount of ^{14}C-propoxur recovered from the test apparatus when the air scrubbing system was used. Most of the propoxur recovered was from the

filter paper collar (61.6%). Propoxur condensed on the top, sides, and base of the dish-1 (aggregation dish). The highest estimate of condensation was 152 ng/cm^2 from the sides of dish-1. That from the base of the dish was 34 ng/cm^2, followed by the 10 ng/cm^2 on the top. Very little radiolabelled material was recovered from dish-2 (dispersal dish).

Table 1. Estimated deposits of propoxur volatilized from a 300 ul volume of a radiolabelled 1% propoxur-in-oil formulation applied to a filter paper collar in an apparatus used for testing vapor-induced dispersal with air scrubbing (150 ml/min for two hours)[a].

Recovery sites[b]	Mean % of total DPM (± SD)[c,d]	Total wt (ug) from formulation	Area (cm^2)	Estim. conc. (ng/cm^2)
dish-1				
top	0.04 ± 0.028	1.17	113	10
sides	1.15 ± 0.800	34.38	226	152
base	0.13 ± 0.001	3.81	113	34
screens	0.02 ± 0.001	0.48	[e]	[e]
tube	0.001± 0.0001	0.03	30	1
glass plate	<0.001	[e]	400	[e]
dish-2				
top	<0.001	[e]	113	[e]
sides + base	0.001 ± 0.0001	0.03	339	0.1
screens	<0.001	[e]		
dish-1 total	1.33 ± 0.200	39.87	452	88
dish-2 total	0.002 ± 0.0001	0.06	452	0.1
filter paper	61.61 ± 5.000	1,848.30	130	1,4217

[a]See Figure 1 for details; data obtained from air scrubbing found in Table 3.
[b]dish-1 - aggregation dish; dish-2 - dispersal dish; screens - strips of screen wire that served as harborage in prior dispersal experiment; glass plate - under screened opening in tube connecting dish-1 and dish-2.
[c]DPM - disintegrations per minute. Total recovery of radiolabelled propoxur in this experiments was 63 ± 5%.
[d]Values are averages based on 3 replicates.
[e]Not detectable at <0.001 total DPM.

Table 2 shows the redistribution of propoxur without air scrubbing. Highest recovery was from the sides of dish-1, followed by the base, top and screens. Most of

Table 2. Estimated deposits of propoxur volatilized from a 300 ul volume of a radiolabelled 1% propoxur-in-oil formulation in an apparatus used for testing vapor-induced cockroach dispersal without air scrubbing.

Recovery sites[a]	Mean % of total DPM (± SD)[b,c]	Total weight (ug)	Area (cm^2)	Estim. conc. (ng/cm^2)
dish-1				
top	0.04 ± 0.030	1.05	113	9
side	0.62 ± 0.182	18.45	226	82
base	0.84 ± 0.039	2.52	113	22
screens	0.02 ± 0.012	0.69	—[d]	—[d]
tube	0.01 ± 0.003	0.01	30	5
glass plate	0.001 ± 0.0001	0.03	400	0.1
dish-2				
side + base	0.002 ± 0.0001	0.06	339	0.2
screens	<0.001	—[d]	—[d]	—[d]
totals:				
dish-1	0.76 ± 0.220	22.80	452	50
dish-2	0.01 ± 0.005	0.24	452	0.5
filter paper	65.20 ± 3.500	1,955.85	130	15,045

[a]See Table 1, footnote "b".
[b]DPM - disintegrations/minute. Total recovery of radiolabelled propoxur in this experiment was 67 ± 3%.
[c]Values are averages based on 4 replicates.
[d]Not detectable at <0.001% total DPM.

the propoxur was again on the filter paper collar. The projected amount of propoxur condensation was lower in dish-1 than when the air scrubbing system was used. A lower air flow over the filter paper and consequently less volatilization than when the air scrubbing system was used accounts for the lower amount of propoxur in dish-1. Condensation in dish-2 was very low in both experiments, but the total amount of propoxur condensed in dish-2 was higher in absence of the air scrubbing. Air scrubbing drew off the vapors before they moved into dish-2.

The above experiments show that propoxur was redistributed onto previously uncontaminated surfaces, although the amounts were very low. Nevertheless, this type of environmental contamination should not be overlooked in planning pest control strategies. Conceivably, the amounts might be sufficient to repel cockroaches, causing them to avoid a lethal exposure.

Table 3. Estimated air concentrations of propoxur from a 300 ul volume of a radiolabelled 1% propoxur-in-oil formulation applied to a filter paper collar in an apparatus used for testing vapor-induced cockroach dispersal with air scrubbing[a].

Time[b]	Mean DPM (± SD)[c,d]	% total DPM	Estim wt (ng)[e]	Air conc (pg/ml)
30	2306±1040	0.023	660	146
60	613±295	0.006	180	40
90	453±391	0.004	120	26
120	370±216	0.004	90	20
Total	3742±1540	0.037	1050	232

[a]See Table 1 for data on surface deposits of propoxur.
[b]30 min intervals.
[c]DPM - disintegrations per minute.
[d]Means from 4 replicates.
[e]From a volume of 4500 ml of air scrubbed at each 30 min interval (total air volume scrubbed = 18 l).

Table 3 shows the estimated air concentrations of ^{14}C-propoxur recovered from dish-1 for each 30 minute scrubbing interval. The concentration for the first 30 min interval was 146 pg/ml, but it declined rapidly to 40 pg/ml (60 min), followed by 26 and 20 pg/ml for the remaining intervals (90 and 120 min, respectively). In view of these results, it is not surprising that most dispersal of cockroaches exposed to vapors of the propoxur-in-oil formulation was over within the first 60 minutes (Wooster and Ross 1989).

The estimated air concentrations of propoxur (Table 3) are an order of magnitude smaller than those of permethrin and pthalthrin that cause knockdown of German cockroaches (Petruska et al. 1985). The estimates give an approximation of air concentrations that caused dispersal in the prior experiment with the propoxur-in-oil formulation (Wooster and Ross 1989). Evidently, cockroach dispersal can be induced by very minute air concentrations of a pesticide, such as might cause an aggregation to disperse before being reached by a lethal dosage.

Acknowledgments. The research was supported in part by the National Pest Control Association. We thank D. G. Cochran for critical reading of the manuscript and Elizabeth Watson and Nancy Boles for technical assistance. Mobay Inc. and Johnsons's Wax provided the propoxur formulations and Mobay the sample of radiolabelled propoxur.

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Received February 1, 1990; accepted June 4, 1990.