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Determination of Transferable Insecticide from Surfaces Painted with Insecticide-Incorporated Paint

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The incorporation of insecticides into household paints to control pests is a relatively novel practice. To obtain the most effective result, paint manufacturers have incorporated some of the most potent contact insecticides into their products. As the efficacy of the paint depends on the ease of transfer of the insecticides to the insects during contact, the most effective formulation could also be the least safe to human. This important safety factor is further compounded by the fact that each manufacturer uses a different formulation which is kept secret. The present study describes a standard procedure to quantitate the amount of insecticides that are transferable from different painted surfaces to samples of moist filter paper during physical contact.

MATERIALS AND METHODS

Sources of paint, insecticides and chemicals: The three insecticides of interest, namely: chlorpyrifos [0,0-diethyl-0-(3,5,6-trichlro-2-pyridyl) thiophosphate], propoxur [2-(1-methylethoxy) phenyl methylcarbamate], and deltamethrin [(S)-a-cyano-3-phenoxybenzyl(1R)-cis-3-(2,2-dibromovinyl)-2,2-dime thylcyclopropanecarboxylate] were gifts from Sundate (Singapore) Proprietary Ltd., Bayer (Singapore) Proprietary Ltd. and Roussel Uclaf, respectively. The emulsion paint containing the three insecticides was supplied by Premacom Trading Singapore. Acetonitrile, methanol and 96% sulphuric acid were purchased from Merck.

Extraction of insecticides from emulsion paint: One gram of emulsion paint was diluted with 2 ml of 100% methanol. The mixture was then vortexed for 1 min and centrifuged at 1840 x g for 30 min. The supernatant was further centrifuged at 265,500 x g for 30 min in a bench-top ultracentrifuge (Beckman TL 100) to obtain supernatant S1.

The pellets were dispersed in 2 ml 100% methanol and the two centrifugation processes repeated to obtain supernatant S2. The procedure was repeated to obtain supernatant S3. The levels of insecticides in S1, S2 and S3 were determined as described below.

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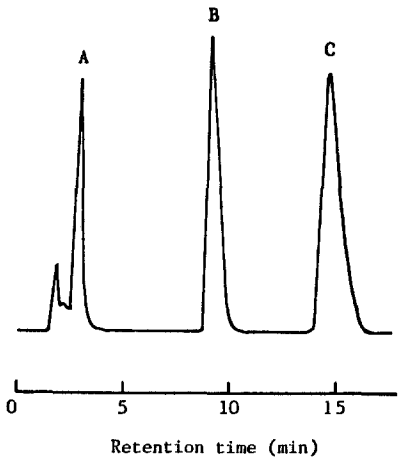

Figure 1. Chromatogram of propoxur (A), chlorpyrifos (B) and deltamethrin (C). 50 µl of a methanol solution containing 0.8 µg of each of the compound were injected onto a HPLC system and the peaks detected at 230 nm (see text for details).

Extraction of insecticides from painted surfaces: A fixed amount of insecticide paint was painted onto a fixed area of an unpainted (raw) wooden and cement surface as a first coat, and as a second coat onto a similar fixed area of a surface pre-painted with ordinary emulsion paint. Two days later, samples of 1-cm Whatman No. 1 filter paper containing various amount of 0.9% saline solution were placed on the painted surfaces. The contact pressure of the 1-cm filter paper with the painted surface was varied by placing different weights (10 to 40 g) on a 1-cm perspex placed over the filter paper. The period of contact varied from 5 to 30 min.

The 1-cm filter paper was then removed and extracted three times with 100 % methanol (0.5 ml per extraction). The three lots of the methanol were consecutively evaporated to dryness at 80 ^{o}C in the same test tube. The residue was then redissolved in 100 µl of chromatographic mobile phase (to obtain a sample solution labeled C1) and the insecticides determined as described below.

Determination of insecticides: The insecticides in the S1, S2, S3 and C1 were determined using high performance liquid chromatography (HPLC) with UV detection as described by Mourot et al. (1979) in their determination of deltamethrin. The HPLC system consisted of a Waters 510 pump, a Waters U6K injector, a Waters variable wavelength UV detector operating at 230 nm, a Waters 740 data module and a Merck LiChrosorb RP-8 5 m column. Isocratic elution was carried out with acetonitrile-1% sulphuric acid (70:30). The flow rate was maintained at 1.1 ml/min.

Reproducibility of determination: Three chromatographic samples each

containing a respective insecticide, namely propoxur, chlorpyrifos and deltamethrin were assayed on five successive days using the above-described HPLC system. The absorption at 230 nm of the eluted peak for each assay was determined. The inter-day coefficient of variation of the method employed to determine each insecticide was found to be 3.5, 2.9, 3.7 % for propoxur, chlorpyrifos and deltamethrin, respectively. Five aliquots of each chromatographic sample were similarly assayed on the same day. From the absorption obtained, the intra-day coefficient of variation was found to be 3.0, 3.1, 3.3 % for propoxur, chlorpyrifos and deltamethrin, respectively.

RESULTS AND DISCUSSION

The chromatogram of the three insecticides is shown in Fig. 1. The contents of the three insecticides in the sample of emulsion paint were fairly close to the values supplied by the manufacturer (see Table 1).

Table 1. Insecticide content of emulsion paint.

Insecticide	Content (in percentage)	
	*Determined Experimentally	Given by Manufacturer
Propoxur	0.17	0.15
Chlorpyrifos	0.42	0.40
Deltamethrin	0.010	0.003

*Combined values of S1 and S2. S3 contained no detectable amount of the insecticide. Each value is the mean of three separate determinations. The standard deviations of each mean is less than 7% of the mean value.

Of the three insecticides, only propoxur was transferred to the moist filter paper in detectable amounts during the contact. The amount transferred varied with the content of saline solution in the filter paper, the time of contact and the contact pressure (see Fig. 2).

In addition, the types and conditions of the surface (i.e. whether wood or cement, painted or non-painted) also affected the transfer of propoxur (see Table 2).

The results show that the transfer of insecticide from surfaces painted with insecticide-incoporated paint to the filter paper during physical contact is affected by common everyday factors like the moisture of paper, the pressure of contact, the time of contact, the type of painted wall, the number of coatings and the area of contact. From our data, the amount transferred varied from no transfer for dry filter paper to 11.2 µg (25.6 % of the amount of insecticide present in the painted area) for 1-cm of filter paper containing 23 µl of saline and under a contact pressure of 30 g per

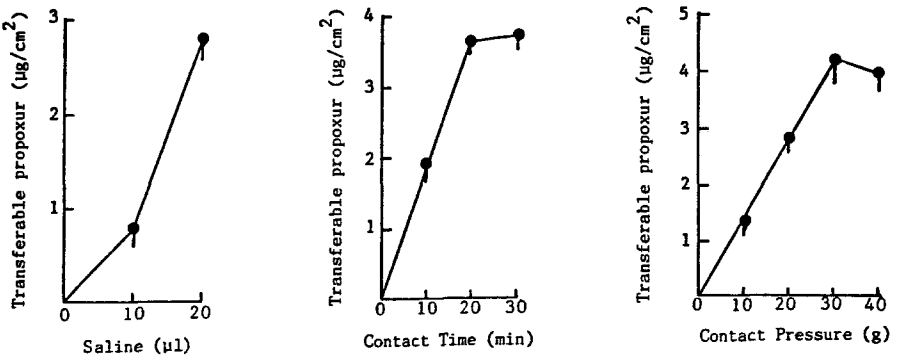

Figure 2. Amount of propoxur transferred to 1 cm of filter paper under different conditions. A: At constant contact pressure (20 g/cm^2) and contact time (10 min) but varying amount of saline in the filter paper. B: At fixed amount of saline in the filter paper (20 mg) and contact pressure (20 g/cm^2) but varying contact time. C: At fixed amount of saline in the filter paper (20 µl) and contact time (10 min) but varying contact pressure. Each bar represents the standard deviation of three separate determinations.

cm^2. Noting that palmar secretion is mainly an aqueous solution of sodium chloride and taking the surface area of an adult palm as 20 cm^2, the amount of propoxur transferable to a moist palm would approximate to 224 µg. Since propoxur is a potent contact insecticide, the danger it poses to human from such easy contamination should not be overlooked.

Table 2. Amount of propoxur transferred to moist filter paper from different surfaces

Contact Pressure (g/cm^2)	Amount Transferred ($\mu g/cm^2$)			
	Wood Surface		Cement Surface	
	1st Coating	2nd Coating#	1st Coating	2nd Coating#
10	0.28 (0.4)	0.32 (0.8)	1.28 (1.8)	5.5 (12.6)
20	1.32 (1.8)	2.00 (5.1)	2.82 (4.1)	8.8 (20.1)
30	0.68 (1.0)	1.10 (2.8)	4.26 (6.1)	11.2 (25.6)
40	2.02 (2.8)	1.06 (2.7)	4.00 (5.8)	4.8 (11.0)

Values in parentheses are percentages of the amount of insecticde present in a 1-cm^2 painted surface. #The insecticide paint was painted as a second coating onto a surface already painted with an ordinary emulsion paint. Each value is the mean of three separate determinations. The standard deviation of each mean is less than 8% of the mean value.

It is of interest to note that chlorpyrifos (present in higher percentage than propoxur in the emulsion paint) and deltamethrin were not transferred in detectable amount to the moist filter paper under the conditions tested. This could be due to the fact that the three insecticides vary very greatly in their solubilities in water (propoxur - 2 g/L at 20 ^{o}C, chlorpyrifos - 2 mg/L at 30 ^{o}C, deltamethrin - 0.2 mg/L at 20 ^{o}C). Another possibility is that in this particular formulation provided by the manufacturer, both chlorpyrifos and deltamethrin in the dried up paint were not accessible to the saline solution.

As secretions of insects are aqueous, we are of the opinion that the stated efficacy of this particular emulsion paint is due mainly to propoxur despite the fact that two other insecticides, namely chlorpyrifos and deltamethrin also form part of the formulation.

The procedure described in this paper is the first of its kind and can reliably be employed by manufacturers of insecticide paint not only to test and improve the quality but also to determine the safety of their products.

REFERENCES

Mourot D, Delephine B, Boisseau J, Gayot G (1979) High-performance liquid chromatography of decamethrin. J Chromatogr 173: 312-414.

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