Contents lists available at ScienceDirect





# **Regulatory Toxicology and Pharmacology**

journal homepage: www.elsevier.com/locate/yrtph

# Deposition and spatial distribution of insecticides following fogger, perimeter sprays, spot sprays, and crack-and-crevice applications for treatment and control of indoor pests

James J. Keenan<sup>a</sup>, John H. Ross<sup>b</sup>, Vincent Sell<sup>c</sup>, Helen M. Vega<sup>a</sup>, Robert I. Krieger<sup>a,\*</sup>

<sup>a</sup> Personal Chemical Exposure Program, Department of Entomology, University of California, Riverside, CA 92521, United States <sup>b</sup> Gem Quality Risk, Inc. 5233 Marimore, Carmichael, CA 95608, United States <sup>c</sup> Washburn & Sons, 807 Center Street, Riverside, CA 92507, United States

#### ARTICLE INFO

Article history: Received 23 November 2009 Available online 22 May 2010

Keywords: Pesticide deposition Indoor pesticide Fogger Exposure assessment Chlorpyrifos Pyrethroid

#### ABSTRACT

The indoor surface deposition and distribution of insecticides applied as foggers, baseboard or perimeter sprays, spot sprays and crack-and-crevice sprays represent distinct pathways of potential unintentional and unavoidable residential pesticide exposure of children and adults. Fogger, perimeter spray, crack-and-crevice, and spot sprays using registered commercial products were studied using three 5-part deposition plates positioned in unoccupied residences in Riverside, CA. Pesticide active ingredients included permethrin, chlorpyrifos, cyfluthrin, cypermethrin, and deltamethrin. Horizontal distribution factors of 100% (total release fogger in a small room), 50% (perimeter spray), 15% (crack-and-crevice), and 2% (spot spray) were assigned based upon application of selected commercial products by a licensed pest control operator and investigators who participated in these studies. This research reduces uncertainties associated with assessing human exposure following different application methods.

© 2010 Elsevier Inc. All rights reserved.

# 1. Introduction

Indoor surface deposition of insecticides applied as foggers, baseboard or perimeter sprays, spot sprays and crack-and-crevice sprays represent pathways of potential unintentional and unavoidable pesticide exposure of children and adults (Krieger et al., 2001). Knowledge of the spatial distribution of surface residues is required to develop responsible risk assessments of children's exposure to pesticides that may include considering aggregate and cumulative exposures as required by the Food Quality Protection Act (FQPA, 1996). If default assumptions impute uniform insecticide distribution over the entire surface of treated rooms, unreasonable exposure estimates result, particularly when the uncertainties of indirect hand-to-mouth exposure are included in the exposure estimates of children (USEPA, 2008).

Common methods of indoor insecticide application used by professionals and residents alike include fogger, baseboard or perimeter sprays, spot sprays, and crack-and-crevice sprays. A crack-and-crevice application done by a resident as per label instructions (30 cm from the target using an aerosol can) using over-the-counter products or by a licensed professional applicator using a low pressure handwand will most likely differ vastly in deposition and distribution and thus, in potential exposure to children and adults.

Using existing Standard Operating Procedures (SOPs) of the USEPA (USEPA, 1997 revised 2001), a crack-and-crevice treatment of a home with a 0.5% permethrin product leads to an estimated indoor surface residue of 7.5  $\mu$ g/cm<sup>2</sup> while a 6 oz fogger application (0.5% permethrin, 57 m<sup>3</sup> room) leads to an indoor surface residue of 4.8  $\mu$ g/cm<sup>2</sup> (USEPA, 2006) (Table 1). These standard assumptions are used to estimate surface residues and along with EPA default algorithms can be used to estimate dose (USEPA, 1997 revised 2001) (Table 1). Surface residue defaults calculated from EPA's SOPs give no indication of distribution, which will be vastly different due to the directed sprays of perimeter, crack-and-crevice, and spot applications as compared with the total surface application of the fogger. Distribution differences will invariably lead to exposure differences due to accessibility of pesticide residues.

Human exposure can occur through inhalation, ingestion, and dermal pathways. Air monitoring has shown that insecticide air levels decline rapidly following indoor insecticide applications (Ross et al., 1992). This is likely due to the relatively low vapor pressure of active ingredients and relatively large particle size of products registered for home use. Dermal exposure is the primary route of pesticides used indoors (Ross et al., 1990, 1991; Krieger et al., 2001). Deposition and distribution studies can help characterize potential dermal exposure to children and adults indoors.

<sup>\*</sup> Corresponding author. Fax: +1 951 827 5803.

E-mail address: bob.krieger@ucr.edu (R.I. Krieger).

<sup>0273-2300/\$ -</sup> see front matter  $\circledcirc$  2010 Elsevier Inc. All rights reserved. doi:10.1016/j.yrtph.2010.05.003

Table	1
-------	---

Surface	residues	in a	3.1	m	< 3.1	m	test	room	and	default	dose	rates	following	g indoor	pesticide	applications.
---------	----------	------	-----	---	-------	---	------	------	-----	---------	------	-------	-----------	----------	-----------	---------------

Experimental	Residue (	ug/cm²)			EPA default exposure (mg/day)					
applications	0–8 cm from wall	8–16 cm from wall	16–24 cm from wall	24–32 cm from wall	32–40 cm from wall	Mean (0–40 cm Whole from wall) room <sup>a,b,c</sup>		Potential dermal dose (6 year child) <sup>d</sup>	Adjusted potential dermal dose using distribution factor <sup>e</sup>	
Fogger	4.2	4.2	4.1	3.8	4.4	4.1	4.1	9.9	9.9	
Perimeter (Mean of four applications) <sup>f</sup>	7.1	4.9	4.0	2.3	0.92	3.8	1.8	4.2	2.1	
Crack-and-crevice	14.6	0.12	0.1	0.03	0.02	3.0	1.4	3.3	0.49	
Spot spray <sup>g</sup>	15.0	0.54	0.19	0.12	0.01	3.2	0.23	0.56	0.01	
EPA default applications										
Perimeter spray <sup>h</sup>	7.5	7.5	7.5	7.5	7.5	7.5	7.5	18	9	
Crack-and-crevice <sup>h</sup>	7.5	7.5	7.5	7.5	7.5	7.5	7.5	18	2.7	
Fogger <sup>i</sup>	4.8	4.8	4.8	4.8	4.8	4.8	4.8	11.5	11.5	

<sup>a</sup> Residue whole room for perimeter and crack-and-crevice application = mean residue (0–40 cm from wall) \* 42,368 cm<sup>2</sup> (surface area of the test room within 40 cm of the wall) 92,903 cm<sup>2</sup> (surface area of the 10  $\times$  10 ft room).

<sup>b</sup> Residue whole room for Spot application = residue (0-40 cm from wall on three deposition plates) \* 6840 cm<sup>2</sup> (surface area of the three deposition plates) 92,903 cm<sup>2</sup> (surface are of the 10 × 10 ft room).

<sup>c</sup> Residue whole room for fogger application (*R*<sub>w</sub>) = average of all measurements because distribution was the same throughout whole room.

<sup>d</sup> Calculated using default values of 6000 cm<sup>2</sup>/h transfer coefficient, 8 h per day, 5% transferability from carpet.

<sup>e</sup> Potential dermal dose calculated using distribution factors of 100, 50, 15, and 2% floor coverage were assessed for fogger, perimeter spray, crack-and-crevice, and spot applications, respectively.

<sup>f</sup> Mean of four applications (0.17% chlorpyrifos: heavy, 0.17% chlorpyrifos: light, 0.03% deltamethrin, 0.015% cyfluthrin).

<sup>g</sup> Two sprays in a corner, under a window, and along an unobstructed wall.

<sup>h</sup> Crack-and-crevice permethrin surface levels following a 0.5% permethrin application (USEPA, 2006).

<sup>i</sup> Fogger application permethrin surface levels following a 0.5% permethrin, 6 oz fogger application (USEPA, 2006).

This study including our experience with monitoring residential exposure presents research to evaluate the surface deposition and spatial distribution of commercial insecticides used as foggers, perimeter sprays, spot sprays, and crack-and-crevice sprays in unoccupied residences in Riverside, California. The procedures used during this work represent a means to gauge the relative exposure potential of these important application technologies under controlled testing conditions. Deposition and distribution studies following routine application procedures can be used in the characterization of the potential dermal exposure of children and adults indoors. Since deposition and the spatial distribution of residues are principally determined by the physical characteristics of pesticide sprays rather than by their chemical nature, we propose that a generic database can be developed to represent the magnitude of surface spray residues ( $\mu g/cm^2$ ) associated with particular application methods. These data also represent the feasibility of a novel approach for improvement of indoor pesticide exposure assessment which acknowledges important characteristics of insecticide application procedures. Lack of uniform definition and understanding of use practices is currently a serious impediment to exposure assessment and risk characterization.

# 2. Materials and methods

A licensed professional applicator (PCA) applied pyrethroids to an unoccupied room of a house in Riverside, CA. Crack-and-crevice, spot, and fogger applications were also made by investigators in a 3.1 m  $\times$  3.1 m carpeted test room. The PCA applied Suspend<sup>®</sup> SC insecticide (0.03% deltamethrin), Tempo<sup>®</sup> WP Ultra (0.015% cyfluthrin), and Lorsban<sup>®</sup>-4E (0.17% chlorpyrifos) as perimeter (baseboard) sprays. The PCA performed both "heavy" and a "light" pesticide applications (usually based on the apparent needs of the client) in the house.

Study personnel applied Raid<sup>®</sup> Max Roach Killer 7 (0.05% deltamethrin) as a crack-and-crevice spray in the test room using an applicator wand supplied at purchase by the manufacturer. In the crack-and-crevice application, pesticide was applied directly into the cracks-and-crevices around the perimeter of the test room. Study personnel applied Raid<sup>®</sup> Ant and Roach Killer 17 – Country Fresh (0.1% cypermethrin) as six spot applications along the baseboard of the test room. Each spray lasted 1 s, two sprays to each deposition plate. A total release Raid<sup>®</sup> Concentrated Deep Reach<sup>™</sup> Fogger containing 1.7% cypermethrin was discharged in the center of the test room according to the Directions for Use. Applications were performed on consecutive days. Cross-contamination due to airborne pesticide from a previous application would be negligible, and because foam boards were not re-used, there was no concern about surface cross contamination.

Deposition was measured on a large sheet of chromatography paper attached to a foam board (57 cm  $\times$  46 cm CHM 1 Whatman filter paper mounted with double-sided tape on Elmers<sup>®</sup> Foam Board) designated a "deposition plate." Deposition plates also included Water and Oil Sensitive Papers (Teejet<sup>®</sup>) affixed to the foam board at 0, 12.5, 25, 37.5, and 50 cm from the wall. The deposition plates were positioned firmly against the wall and were located in a corner (position 1), under a large window (position 2), and along an unobstructed wall (position 3) in the house and in a separate test room. After application of pesticides, water and oil sensitive papers were photographed (data not shown). The foam boards were collected and the CHM 1 papers were cut into five strips that were each 8 cm wide (456 cm<sup>2</sup> each). Photographs and deposition plate collection was performed immediately following crack-andcrevice, spot, and perimeter pesticide applications. Collection of deposition plates and photographs occurred 4.5 h post-fogger release, according to product specific re-entry times. Strips were stored frozen and extracted overnight using a Soxhlet apparatus and 300 mL of ethyl acetate. Extracts were analyzed for active ingredients using GCMS.

### 3. Results and discussion

Dermal exposure is the primary exposure route of semi-volatile chemicals used indoors and deposition and distribution studies following routine application procedures can help characterize this exposure (Ross et al., 1990, 1991; Krieger et al., 2001). Physical characteristics of pesticide sprays and the actions of applicators determine the deposition and spatial distribution of residual pesticide. We examined several application scenarios using three pyrethroid pesticides and chlorpyrifos, an organophosphate pesticide. For comparison, whole floor residues ( $R_w$ ) were calculated for a



Fig. 1. Chlorpyrifos deposition following perimeter applications. Chlorpyrifos deposition (µg/cm<sup>2</sup>) at indicated distances from wall (cm) following subjectively "heavy" and "light" applications indoors in an unoccupied residence.

3.1 m  $\times$  3.1 m room. The discharge of a cypermethrin fogger in the small room (30 m<sup>3</sup>) represented relatively uniform surface deposition of cypermethrin. The widespread fogger deposition pattern contrasted sharply with the very restricted deposition patterns of crack-and-crevice sprays, spot sprays, and perimeter sprays that were also evaluated under the same conditions. The testing protocol permitted direct comparison of the exposure potential of common alternate spray technologies used in residential pest treatment and control.

# 3.1. Fogger application

Total release foggers are often used indoors by residents to treat and control ants, fleas, roaches and other domestic insects. A 40 s discharge from a Raid<sup>®</sup> Concentrated Deep Reach<sup>TM</sup> Fogger releases about 0.7 g of active ingredient as an aerosol. As a result insecticide residues in the  $\mu$ g/cm<sup>2</sup> range are deposited on open surfaces within the home. Skin contact with these surfaces can result in contacttransfer of low levels of chemical to the skin as demonstrated by a number of post-application re-entry exposure monitoring studies with human volunteers. A fogger application in a relatively small, confined space yielded a relatively uniform distribution of cypermethrin. This application served as a reference point for the comparison of the spatial distribution pattern of other application types.

The Personal Chemical Exposure Program (Krieger et al., 2001) has done extensive monitoring of family exposures to chlorpyrifos and cypermethrin foggers. In those studies, deposition of pyrethrins (PY), piperonyl butoxide, and *N*-octyl bicycloheptene dicarboximide (MGK 264) were measured on  $\alpha$ -cellulose coupons positioned 2, 4, and 6 m from the fogger contained a mean  $3.7 \,\mu\text{g/cm}^2$  PY. Reconstruction of the spatial deposition of the three components in the aerosol demonstrated decreased deposition of each component of the aerosol as distance from the fogger increased, irrespective of active ingredient (Selim and Krieger, 2007). Typically, in a large open setting an obvious gradient is formed outward from the fogger since larger particles carrying more chemical fall out in closer proximity to the release point.

Air levels post-fogger release in the current study were 0.02  $\mu$ g/m<sup>3</sup> at 4.5 h post-application (the product specific re-entry time). Air levels were negligible as potential exposure sources as compared with surface levels. Average chemical residue perpendicular from the wall to 40 cm was 4.1 ± 1.2  $\mu$ g/cm<sup>2</sup>. Similarly, average

levels of cypermethrin found within 8 cm of the wall were  $4.2 \pm 0.8 \ \mu g/cm^2$  and levels between 32 and 40 cm from the wall were  $4.4 \pm 1.3 \ \mu g/cm^2$  (Fig. 2). Chemical residue on an additional fourth deposition plate located in the center of the test room was  $3.1 \pm 0.1 \ \mu g/cm^2$ . Distance from the fogger in the small test room had little or no effect on cypermethrin deposition, contrary to what has been observed in the past with foggers in a larger open space (Selim and Krieger, 2007).

In the smaller test room, a well-mixed aerosol was created from the fogger that was placed in the center of the room. Cypermethrin was evenly distributed to the floor. In this respect, fogger applications differ from all other methods of application that rely on directed sprays examined in this paper. This supports our proposal that deposition and spatial distribution are principally determined by the type of pesticide application (i.e. fogger vs. crack-and-crevice) and the actions of the applicator (i.e. heavy vs. light applications).

# 3.2. Perimeter application

A perimeter application performed by a PCA is sometimes considered interchangeable or equivalent to a crack-and-crevice application by some persons constructing exposure assessments in regulatory organizations. An earlier situational monitoring study (Krieger et al., 2001) has demonstrated the need for distinguishing perimeter sprays from spot or crack-and-crevice applications. The investigators in the 2001 study expected a limited "crack-and-crevice" application based on their own previous use of over-thecounter products. Instead they received a much more general perimeter (or even an area spray in some places) based upon their discussion with the applicator at the time the work was to be done and our much later interview of the applicator. The results of the biomonitoring for that application revealed low level exposures that were greater than the extremely low levels that follow actual crack-and-crevice applications (Krieger et al., 2001). This confusion underscores the need for more uniform classification of application technologies since each carries distinct human exposure potential.

In the present research a perimeter application is a spray directed at the baseboards but spreading for as much as half a meter up the wall and along the floor. Lorsban<sup>®</sup>-4E (0.17% chlorpyrifos) was applied to the residence in Riverside, CA by a PCA via perimeter application. The PCA described making a "heavy" and a "light" level of pesticide application based upon the apparent needs of the



**Cypermethrin Deposition Following Fogger Application** 

**Fig. 2.** Cypermethrin deposition following total release fogger application. Pyrethroid surface residues ( $\mu$ g/cm<sup>2</sup>) following fogger application. Deposition plates at indicated distances from wall (cm) were located in a corner (1), under a window (2), and along an unobstructed wall (3) in a 30 m<sup>3</sup> test room.

client in discussion with study staff. The two applications were indistinguishable in regards to time spent per wall and study staff could not visually distinguish any difference in pesticide coverage.

Following the "heavy" application of chlorpyrifos, deposition within 0–40 cm of the wall was  $4.4 \pm 4.3 \ \mu\text{g/cm}^2$  (Fig. 1). Following the "light" application of chlorpyrifos, deposition within 0–40 cm of the wall was  $4.5 \pm 2.8 \ \mu\text{g/cm}^2$ . Total deposition following heavy and light application of chlorpyrifos was nearly identical (Fig. 1).

Although average deposition was nearly identical, the spatial distribution of the chlorpyrifos residue differed between the two application levels. Residues inside of 8 cm were  $10.6 \pm 1.6$  and  $7.9 \pm 1.0 \ \mu g/cm^2$  following "heavy" and "light" applications, respectively. Levels decreased to  $0.7 \pm 0.9$  and  $3.0 \pm 2.3 \ \mu g/cm^2$  for heavy and light, respectively, between 24 and 32 cm from the

wall. Levels between 32 and 40 cm from the wall were below  $1.0 \ \mu g/cm^2$  for both applications (Fig. 1). The light application by the PCA resulted in spray that was less concentrated on the baseboard. However, a light application would likely lead to higher potential human exposure due to higher levels of pesticide further from the wall.

A light perimeter application of Suspend<sup>®</sup> SC insecticide (0.03% deltamethrin) and Tempo<sup>®</sup> WP Ultra (0.015% cyfluthrin) were applied by the PCA to the same house in Riverside, CA. Following the application of deltamethrin and cyfluthrin, deposition within 0–40 cm of the wall averaged  $5.1 \pm 2.3$  and  $1.4 \pm 1.1 \,\mu$ g/cm<sup>2</sup>, respectively. Levels inside of 8 cm were  $8.2 \pm 2.5$  and  $1.9 \pm 0.7 \,\mu$ g/cm<sup>2</sup> for deltamethrin and cyfluthrin, respectively. Levels between 32 and 40 cm from the wall decreased to  $2.4 \pm 1.5$  and





**Fig. 3.** Deltamethrin deposition following perimeter spray application. Pyrethroid surface residues ( $\mu$ g/cm<sup>2</sup>) on deposition plates at indicated distances (cm) from wall in a corner (1), under a window (2), and along an unobstructed wall (3) of an unoccupied residence.

 $0.2\pm0.3\,\mu g/cm^2$  for deltamethrin and cyfluthrin, respectively (Fig. 3).

Although deposition levels were very different following these two applications of pyrethroids, distribution patterns were similar for light applications of chlorpyrifos, deltamethrin, and cyfluthrin. Total floor levels were influenced more by percent active ingredient and the actions of the PCA than by the particular chemical characteristics of the products.

#### 3.3. Crack-and-crevice application

In a crack-and-crevice application, pesticide is applied directly into the cracks-and-crevices of a house. Little pesticide residue occurs on the floor or wall adjacent to the targeted site. The homeowner, using over-the-counter products can perform true crack-and-crevice applications. Raid<sup>®</sup> Max Roach Killer 7 (0.05% deltamethrin) was applied in a carpeted test room. Deltamethrin deposition within 0–40 cm of the wall was  $3.0 \pm 6.2 \,\mu\text{g/cm}^2$ . Levels inside of 8 cm were  $14.6 \pm 7.0 \,\mu\text{g/cm}^2$ . Levels decreased to  $0.02 \pm 0.007 \mu \text{g/cm}^2$  between 32 and 40 cm from the wall (Fig. 4). Average levels of deltamethrin 0-40 cm from the wall were similar between crack-and-crevice and perimeter applications but distribution differed greatly (Table 1). Levels of deltamethrin between 8 and 16 cm from the wall following crack-and-crevice application were below 1.0  $\mu$ g/cm<sup>2</sup> (Fig. 4). Greater than 98% of the deltamethrin deposited within 8 cm of the baseboard using crack-andcrevice application.

Crack-and-crevice and perimeter applications as defined here have historically been considered to be crack-and-crevice applications in a regulatory setting. We propose that these are clearly different application types that result in separate and distinctive spray deposition and distribution patterns. Each of those is linked with significantly different human exposure potential requiring separate human exposure assessments for risk characterization. We propose that a typical crack-and-crevice application by a PCA be termed a *perimeter application* and be regulated as such. Applications made from a pressurized container using an extension tube result in virtually no splash-back deposition and insignificant human exposure whether applied by a PCA or homeowner (Keenan, 2007).

#### 3.4. Spot application

Spot applications involve pesticide sprayed either directly onto the insect or in places where residents saw, or would likely see, insects. Study personnel applied Raid<sup>®</sup> Ant and Roach Killer 17 – Country Fresh (0.1% cypermethrin) as six spot applications along the baseboard of the test room. Each spray lasted 1 s, two sprays to each deposition plate. Deposition within 0–40 cm of the wall was  $3.2 \pm 6.2 \ \mu g/cm^2$ . Levels inside of 8 cm were  $15 \pm 1.2 \ \mu g/cm^2$ , which accounts for about 95% of the total spray deposition from the spot applications. Levels decreased to  $0.01 \pm 0.005 \ \mu g/cm^2$  between 32 and 40 cm from the wall (Fig. 5). Spot applications, although seemingly similar to crack-and-crevice with regards to deposition levels and distribution patterns actually deposit significantly less pesticide and would lead to much lower potential human exposures.

Whole room residue ( $R_w$ ;  $\mu g/cm^2$ ) is the site specific residue resulting from each application type (Table 1). R<sub>w</sub> for a fogger application is the average deposition ( $\mu g/cm^2$ ) multiplied by the surface area of the whole room (92,900 cm<sup>2</sup>) divided by the surface area of the whole room. R<sub>w</sub> for perimeter and crack-and-crevice applications is the average deposition within 0-40 cm of the wall multiplied by the surface area of the room within 0-40 cm of the wall (42,368 cm<sup>2</sup>) and divided by the surface area of the whole room.  $R_w$  for spot application is the average deposition within 40 cm of the wall multiplied by the surface area of three deposition plates (6840 cm<sup>2</sup>) and divided by the surface area of the whole room (Table 1). Fogger, perimeter, crack-and-crevice, and spot spray have  $R_w$  values of 4.1, 1.8, 1.4, and 0.23  $\mu$ g/cm<sup>2</sup>, respectively. As expected, fogger application resulted in the highest total amount of pesticide applied. Fogger surface levels are higher than previously measured levels in residential monitoring due to the small size of the test room used (Krieger et al., 2001). Perimeter and crack-and-crevice application resulted in remarkably similar  $R_{\rm w}$  values, but the distribution patterns were very different within 0-40 cm of the wall. In crack-and-crevice applications over 98% of





**Fig. 4**. Deltamethrin deposition following crack-and-crevice application. Pyrethroid surface residues (µg/cm<sup>2</sup>) on deposition plates at indicated distances (cm) from wall in a corner (1), under a window (2), and along an unobstructed wall (3) of an unoccupied residence.





**Fig. 5.** Cypermethrin deposition following spot application. Pyrethroid surface residues ( $\mu$ g/cm<sup>2</sup>) on deposition plates at indicated distances (cm) from wall in a corner (1), under a window (2), and along an unobstructed wall (3) of an unoccupied residence.

the insecticide was delivered to the 0–8 cm coupon with only trace amounts of 'splash back' recovered from the remainder of the test field.

Residue levels were similar within 0–40 cm space from the walls following each type of application (Table 1). The differences in pesticide distribution are most apparent 0–8 cm from the wall and 32–40 cm from the wall. Fogger distributed low levels of pesticide to all surfaces (Table 1; Fig. 2). Perimeter spray deposited higher levels near the walls (Table 1; Figs. 1 and 3). Crack-and-crevice and spot applications deposited over 98% of the pesticide within 8 cm of the wall (Table 1, Figs. 4 and 5). These different distribution patterns result in distinctly different patterns of potential human exposure. Since most activities in residences do not occur where walls and floor coverings are joined, opportunity for human contact, transfer, and absorption are lower than those resulting from fogger applications which distribute residue over the entire indoor surface (Table 1).

USEPA (2006) default algorithms are used as screening levels to predict potential human exposure from pesticide residue levels and to help identify exposure issues of regulatory importance. If uniform distributions of the whole room surface residues are assumed following fogger, perimeter spray, crack-and-crevice spray, and spot spray (Table 1) the potential dermal dose to a 6-year-old child would be 9.9, 4.2, 3.3, and 0.56 mg/day, respectively (Table 1). If the insecticide spray distributions are ignored in forming screening levels of resident exposure, the resulting dermal doses are unreasonably high and unrelated to measured patterns of deposition. Such estimates are substantially higher than exposures measured using biomonitoring following fogger and crack-and-crevice application of pyrethroids (Keenan, 2007). In our unpublished biomonitoring studies, exposures following crack-and-crevice application of cypermethrin (0.00027 mg/kg) did not exceed background levels in any subject (n = 10).

Default algorithms or screening level estimates must consider application type, which has a much greater impact on residue levels than the chemical characteristics of the active ingredient. Data supports our proposal for a generic database developed to represent the magnitude of surface spray residues ( $\mu$ g/cm<sup>2</sup>) following different types of indoor pesticide applications. These experimental data in Table 1 can be adjusted to reflect differences in relative amount of active ingredient in a particular type of application.

Default exposure algorithms have a conversion factor for transferable residue from different flooring materials (hard and soft), but historically they do not address the inherent differences in distribution following different application types. Fogger application of pyrethroid pesticides was uniform on the floor and, furthermore, we assumed 100% coverage of the floor. As such, the current regulatory default algorithm is valid. Hypothetically, a distribution factor of 1 would be applied to fogger estimates to reflect total floor coverage. However, perimeter, crack-and-crevice, and spot application of pesticides to a 3.1 m  $\times$  3.1 m room deposited significant amounts of pesticide on only 50%, 15%, and 2% of the floor, respectively. These percentages would decrease as room sizes increased making these initial hypothetical estimates very conservative. We suggest differences in surface coverage be reflected in default algorithms. For example, inserting an experimental distribution factor of 15% floor coverage for a crack-and-crevice application would change the hypothetical potential dose to a young child from 3.2 to 0.49 mg/day (Table 1). Using this experimental distribution factor, potential dose estimates will reflect the important relative differences in distribution of pesticide following fogger, perimeter spray, crack-and-crevice, and spot applications. We would caution that for perimeter, spot and crack-and-crevice spray, another typical use practice not accounted for in hypothetical exposure estimates is the tendency to spray only a portion of the four sides of a room; i.e., there are limited ingress points for pests in any given room, and not every room in a house is typically treated.

### 4. Conclusion

Each application type produced a surface residue, but the residues differed sharply in deposition and distribution. Relative to the general distribution of residue following fogger applications, perimeter, crack-and-crevice, and spot applications resulted in less total chemical residue and limited distribution to within 0–40 cm of the wall. Crack-and-crevice and spot applications deposited high levels of pesticide directly at the target site, within 8 cm of the baseboard. Experimental distribution factors would reflect these differences and improve first tier indoor pesticide exposure assessments. Crack-and-crevice application of pyrethroid pesticides appear to be the most effective application type when one is trying

to decrease potential exposure and maintain efficacy of treatment. When materials are applied at the wall a limited amount of spray is released. Mass balance considerations and data shown in Table 1 make assumptions of widespread horizontal distribution of residue untenable.

Deposition and distribution were principally determined by the physical characteristics of pesticide sprays and the actions of the applicators. Although these tests were performed in controlled environments and results might differ slightly in occupied residential settings, it is clear that the manner in which applications are made is a critically important determinant of potential human exposure. This aspect of safe use can be incorporated into training and labeling as well as clarifying consumer expectations about pest treatment and control. These deposition and spatial distribution data support the proposal for a generic database representing the magnitude of surface spray residues ( $\mu g/cm^2$ ) irrespective of active ingredient, but proportional to the amount of active ingredient relative to the amount used experimentally in this study. While EPA has recently proposed changes in their SOPs that are based on unpublished data, these changes have not yet been implemented (EPA, 2009). This generic database should qualify whether a directed spray application is a perimeter application by a PCA or a crackand-crevice application by a member of the public or PCA due to clear differences in pesticide distribution. This study presents the feasibility of this novel approach for improvement of indoor pesticide exposure assessment.

#### Acknowledgments

Western Exterminators Company, Riverside, CA is gratefully acknowledged for supply of commercial products used in this research. Anasthasia Krieger is thanked for her generous assistance and continued support.

#### References

FQPA (Food Quality Protection Act) 1996. Public Law 104-170, Aug. 2, 1996.

- Keenan, J.J., 2007. Potential exposures of children and adults to cypermethrin and other pyrethroid insecticides following treatment and control of indoor pests. Ph. D. Dissertation. University of California Riverside. Riverside, CA. p. 284.
- Krieger, R.I., Bernard, C.E., Dinoff, T.M., Ross, J.H., Williams, R.L., 2001. Biomonitoring on persons exposed to insecticides used in residences. Ann Occup Hyg 45, S143–S153.
- Ross, J., Thongsinthusak, T., Fong, H., Margetich, S., Krieger, R., 1990. Measuring potential transfer of surface pesticide residue generated from indoor fogger use: an interim report. Chemosphere 20, 349–360.
- Ross, J., Fong, H., Thongsinthusak, T., Margetich, S., Krieger, R., 1991. Measuring potential transfer of surface pesticide residue generated from indoor fogger use: using the CDFA roller method interim report. Chemosphere 22, 975–984.
- Ross, J.H., Fong, H., Thongsinthusak, T., Krieger, R.I., 1992. Experimental method to estimate indoor pesticide exposure to children. In: Guzelian, P.S., Henry, C.J., Olin, S.S. (Eds.), Symposium on Similarities and Differences of Adults and Children: Implications for Risk Assessment. ILSI Press, Baltimore, MD, pp. 226– 241.
- Selim, S., Krieger, R.I., 2007. Indoor human pyrethrins exposure: contact absorption, metabolism, and urine biomonitoring. In: Krieger, R., Seiber, J., Ragsdale, N. (Eds.), Assessing Exposures and Reducing Risks to People from the use of Pesticides. ACS Series Publication 951. Oxford University Press, pp. 125–140.
- USEPA, 1997. (revised 2001). Science Advisory Council for Exposure Policy Number 12: Recommended Revisions to the Standard Operating Procedures (SOPs) for Residential Exposure Assessments. Revised February 22, 2001. U.S. Environmental Protection Agency, Office of Pesticide Programs, Washington, DC.
- USEPA, 2006. Reregistration Eligibility Decision (RED) for Permethrin. Case No. 2510. U.S. EPA 738-R-06-017. Office of Prevention, Pesticides, and Toxic Substances. Washington, DC.
- USEPA, 2008. Child-Specific Exposure Factors Handbook (Final Report) 2008. U.S. Environmental Protection Agency, Washington, DC., EPA/600/R-06/096F, 2008.
- USEPA (U.S. Environmental Protection Agency), 2009. Draft Technical Guidelines Standard Operating Procedures for Residential Pesticide Exposure Assessment. Submitted to the FIFRA Scientific Advisory Panel For Review and Comment October 6–9, 2009. Office of Pesticide Programs Office of Prevention, Pesticides, and Toxic Substances U.S. Environmental Protection Agency Washington, DC.