RESIDENTIAL INDOOR EXPOSURES OF CHILDREN TO PESTICIDES FOLLOWING LAWN APPLICATIONS

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ABSTRACT

Methods have been developed to estimate children's residential exposures to pesticide residues and applied in a small field study of indoor exposures resulting from the intrusion of lawn-applied herbicide into the home. Sampling methods included size-selective indoor air sampling; wipe sampling of floors, sills, tables; the polyurethane foam (PUF) roller for dislodgeable carpet surface residues; and the HVS3 vacuum sampler for floor dust. Personal exposure samples included hand wipes and morning void urine samples.

Pesticide spray drift was found to result in only a relatively minor increase in indoor pesticide levels. Post-application air intrusion from closed house ventilation and the opening and closing of doors and windows increased indoor background levels 6-fold, while track-in by high activity children and pets, and wearing shoes indoors, increased indoor levels by 37-fold. Indoor 2,4-D levels were found to increase continually over a one-week period, with the increase in indoor air levels corresponding to the increased floor dust levels, suggesting resuspension of house dust by human activity. Similar estimates of non-dietary exposure are obtained from models based on 100 mg dust ingestion and surface contact simulated by the PUF roller.

INTRODUCTION

Pesticides are used inside the home and outside on the lawn or garden. After either application, they tend to accumulate indoors in air, house dust, upholstery, on surfaces, and also on children's toys. Pesticides sprayed on the lawn may be tracked indoors, where they can persist for months or years protected from environmental breakdown, as opposed to days outside on the grass. Typically, pesticide concentrations in indoor air and house dust are 10-100 times those found in outdoor air and surface soil. Over the past several years, parallel efforts by the U.S. Environmental Protection Agency (USEPA) have been underway to assess the potential exposures small children may receive from pesticide products used in and around the home. Initial studies [1] suggested that infants and toddlers may receive proportionately larger exposures than adults from both the respiratory and dust ingestion routes. Small children are considered to be the population of highest risk since they spend most of their time indoors. Much of this time is spent in contact with floors, engaging in mouthing of hands, toys, and other objects.

Since these initial findings, several laboratory and field studies have been undertaken. These have involved experiments to determine the dislodgeability of pesticides from surfaces (i.e., their potential for transfer to skin) and investigation of the mechanisms of translocation of pesticide residues from outdoors to indoors and their redistribution within the indoor environment. Specialized tools such as the HVS3 vacuum and the PUF roller were developed and evaluated for determining floor dust loadings and estimating dislodgeable residues [1, 2, 3]. Studies with human subjects have shown that pesticides are dislodged from surfaces by saliva-moistened skin

(as a mouthing child's would often be) much more efficiently than by dry skin [4].

Track-in of lawn-applied pesticides is of particular interest, since many homeowners use pesticides for lawn care and may not use them indoors. The presence of insecticides such as chlorpyrifos and pyrethroids in indoor air and dust suggests primary indoor use, although migration and track-in from perimeter and foundation treatments may also contribute to indoor residues. The presence indoors of 2,4-dichlorophenoxyacetic acid (2,4-D), carbaryl and chlorothalonil, which are applied exclusively outdoors, implies that residues have been transported from outdoors. Previous studies have shown that walking over treated turf as much as one week after application resulted in transfer of residues to carpet dust that were proportional (3-4%) to the dislodgeable residues on the turf [5,6]. Over the past three years, studies have been conducted at 12 households to determine the extent to which lawn-applied 2,4-D may be tracked indoors and disbursed throughout the house following both homeowner and professional applicators lawn treatments.

METHODS

Study homes were single floor with basement (except for one split level), surrounded on all sides by turf, and carpeted in the main living room and a child's bedroom. Each family consisted of two adults, two to three school-age children, and one pet (one home had no pets). Sampling methods included HVS3 vacuum sampling for floor dust residues (2 m^2 area); wipes of solid surfaces such as bare floors, table tops and indoor window sills; dislodgeable residue sampling of carpet surfaces (wetted PUF roller); and air sampling by particle size. Sampling locations were selected inside each home, including a frequently-used entry area, a main living area, dining area, kitchen and child's bedroom that would constitute the primary living areas in any home. Cotton gauze wipes moistened with 2 mL of 70:30 phosphate buffer:acetonitrile were used on indoor surfaces [5]. A medium volume cascade impactor (12 L/min) was used to collect indoor air residues during the period of application (ca. 2 h). Windows and doors to the homes were open at the times of application. For post-application air sampling, four collocated PUF air samplers [7] and provided with inlets for collection of <20 μm (total aerosol), <10 μm, <2.5 μm and <1 μm air particles and fine particle filters were operated for 24-h periods (4 L/min). Two unoccupied model homes were also included to assess the relative importance of spray drift and post-application aerial intrusion relative to track-in.

2,4-D dimethylamine herbicide was applied at each study home either by homeowners or professional applicators. Sampling at each home took place over two one-week periods (pre- and post-application). A 24-h indoor air sample was collected on one day (Day 0) during the pre-application week and on Days 1 and 3 post-application. A 2-h indoor air sample (cascade impactor) was taken during the period of lawn application. Wipe sampling of sills, tables, and bare floors; collection of a carpet surface dislodgeable residue sample; and vacuum sampling of floors all occurred on Day 7 post-application. Deposition coupons on the lawn were used to estimate 2,4-D application rates. An integrated air exchange rate measurement was made during the post-application week. Homeowner treatments were studied during the first year, followed by professional applications in the second. In the third year of the study, dermal (hand) wipe samples were obtained with 2-propanol-wetted gauze [8], along with table wipe samples, and vacuumed floor dust samples on three separate days after the lawn application. First morning void urine samples were also collected on the morning following each dermal wipe sample, so as to ascertain whether urinary excretion of 2,4-D could be tied to microenvironmental levels and/or dermal contaminant levels. The dermal wipe samples and urine samples were collected from the adult applicator and one resident child.

RESULTS

Indoor air concentrations of 2,4-D increased from non-detectable before lawn treatment to 0.2 to 10 ng/m^3 (10 µm inlet) after homeowner application, with about 65% of the total particulate 2,4-D associated with respirable particles (<10 µg/m^3). 2,4-D associated with <1 µm and smaller particles made up 25-30% of the total mass. Detectable residues of 2,4-D were found on all surfaces one week after application. The surface concentration gradient followed the occupant traffic pattern through the house. Post-application floor surface loadings of 2,4-D in the living areas ranged from 1 to 228 µg/m^2 on carpeted floors and 0.2 to 20 µg/m^2 on bare floors, compared to 0 to 0.8 µg/m^2 (median 0.5 µg/m^2) pre-application. About 1% of the 2,4-D in floor dust was dislodgeable (PUF roller wetted with acetonitrile:phosphate buffer) and potentially available for dermal contact. 2,4-D residues on window sills and tables followed a similar traffic gradient, with surface loadings of 0.2 to 20 µg/m^2 (none were detectable before application). In homes in which occupants removed their shoes at the entryway, 2,4-D loadings on floors were typically an order of magnitude lower than in those in which shoes were worn.

The data presented here in Figure 1 show the indoor floor dust loadings of 2,4-D in µg/m^2 for three representative homes one week after homeowner applications to lawns. Home *By* was categorized as a home with high child activity and high pet activity, and Home *Zm* had high child activity, but low pet activity. Home *Rr* had high child and low pet activity, but both adults and children routinely removed shoes at the door when entering from outside (which was not the case for Homes *By* and *Zm*). Note that the concentration gradients inside the homes generally follow the traffic patterns through the houses (carpeting accounted for the apparent gradient shift within Home *Rr*). 2,4-D floor loadings in participating households in which occupants routinely removed shoes were typically 10 to 100 times lower than in those in which shoes were worn. Window sill wipes and air monitoring during spray applications indicated that intrusion of 2,4-D into the home by spray drift was minor compared to track-in. Table-top loadings were approximately one-tenth of floor loadings and resulted from deposition of dust resuspended from the floors by human and pet activity. The same patterns of floor dust distributions were also observed after professional applications (where the applicator did not enter the house), but post-application 2,4-D dust loadings in the living rooms of high child/pet activity homes were reduced by 50-75%.

Indoor air concentrations in ng/m^3 of 2,4-D obtained from collocated sampling with $PM_{2.5}$ and PM_{10} size-selective inlets on Day 3 after homeowner and professional applications were similar to those found on Day 1 and 25-50% of those found during application. Across all homes, indoor levels during the application period were only slightly less during homeowner vs. professional applications. However, mean indoor air levels on Days 1 and 3 after professional application were about half of those after homeowner application and 2,4-D was associated with ultra-fine particles (<1 µm), which accounted for about 75% of the total (inverse of what was found with homeowner treatment). For homeowner applications, only 25% of the indoor air levels could be attributed to intrusion during spraying; in contrast, in a low activity home where the applicator did not wear shoes indoors, 100% of the Day 1 air levels were attributable to 2,4-D spray drift. Post-application air concentrations were roughly proportional to floor dust loadings, supporting the supposition that resuspension of floor dust is responsible for respirable 2,4-D in indoor air. The higher 2,4-D air levels were found in homes with active children and pets, and especially with those where shoes were also worn indoors. Likewise, the homes in which 2,4-D was not

detected in air were those with low levels of activity and/or no shoes worn indoors.

In the third year of the study, biological monitoring was conducted along with more intensive environmental sampling in four of the study homes. Dust levels were monitored pre-application and on Days 1, 3, and 7 post-application. Figure 2 shows the temporal profile of indoor residues in a high-activity (Home *By*).

DISCUSSION

For typical homes, track-in was found to be the most significant route of transport of 2,4-D residues from the lawn indoors. For high activity homes, transport via an indoor-outdoor dog, the applicator's shoes, and by children was estimated to account for about 58%, 25%, and 8% of the indoor residues, respectively. Spray drift and post-application aerial intrusion were minimal contributors (<1%) except for homes in which outdoor shoes were not worn indoors and which had low pet activity. Resuspension of floor dust was the primary source of 2,4-D in indoor air, on table tops, and on window sills.

Relative exposures to 2,4-D via the air route vs. dust ingestion routes could not be accurately estimated since floor dust sampling was conducted several days after air sampling (except for the last year). However, since Day 3 air levels were on average higher than Day 1 levels, airborne 2,4-D was probably due primarily to dust resuspension and would be expected to be at similar or higher levels on Day 7 (when floor dust was collected). Hence for Home *By*, the average daily respiratory dose based on PM_{10} received by children between the ages of 6 months and three years spending 24 hours indoors (avg. 6.4 m^3/d inhalation rate [9]) one week after homeowner application would have been 68 ng/d, while the average dose received from ingestion of 100 mg [9] of floor dust from the living room would have been 100 times higher at 6.7 µg/d. After professional application, these potential exposures were reduced to 16 ng/d vs. 4.2 µg/d, respectively. For Home *Zm*, air and dust route exposures would have been 8.1 ng/d and 2.8 µg/d after homeowner application and 15 ng/d and 0.6 µg/d, respectively, after professional lawn treatment. By contrast, in shoes-off home *Rr*, which had very low post-application dust loadings and relatively low concentrations of 2,4-D in house dust, ingestion of 100 mg of dust would have resulted in the intake of only 0.5 µg/d and respiration to 4.4 ng/d after homeowner treatment. The mean dust contribution over all homes in the study is estimated to have been about 2 µg/d and the mean air route contribution18 ng/d one week after homeowner application compared to 12 ng/d and 1.2 µg/d, respectively, after professional treatment. It should be noted, however, that this comparison may not be valid for the sum of all participating homes since only half the homes participated in both phases of the study, which were also conducted in different years. For a 10 kg child, the average total potential dose via the inhalation and non-dietary ingestion routes would be 0.1 to 0.2 µg/kg/d. This is substantially less than the EPA Reference Dose of 10 µg/kg/d or the World Health Organization's Acceptable Daily Intake value of 300 µg/kg/d. However, these reference doses are not necessarily meant for infants and toddlers.

Use of dislodgeable residue data obtained with the PUF roller, combined with frequency of mouthing activity, may also be used to estimate intake by non-dietary ingestion for comparison with estimates made from ingestion of 100 mg/d of house dust. Assuming that the floor-to-hand transfer efficiency was the same as that for the wetted PUF roller, 10 mouthing events (one whole hand) per hour for 12-h/d [10], and 50% efficiently for removal of 2,4-D by salivation [4], exposure estimates agree quite well with those obtained on the basis of dust ingestion of 100 mg/d for a 10 kg child.

Additional research is underway in occupied homes and in the USEPA test home to examine fate

and transport of pesticides from indoor applications and determine the potential for human exposures. House dust has been fractionated by sieving and aerosol suspension into seven fractions ranging from <4 μm to 500 μm and analyzed to determine the distribution of pesticides as a function of particle size (concentrations increase with decreasing particle size). The mechanics of transfer of particles from outdoors to indoors on shoes, from indoor surfaces to dry and wet skin, and from floors into air by human activity are being investigated. Other studies are underway to better determine surface-to-skin and skin-to-mouth transfer efficiencies, pesticide bioavailability from dust, and the relationship of child activity patterns to residential exposures. Such studies are essential before accurate exposure assessments can be made.

DISCLAIMER

This work has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review process and approved for presentation and publication. Mention of tradenames or commercial products does not constitute endorsement or recommendation for use.

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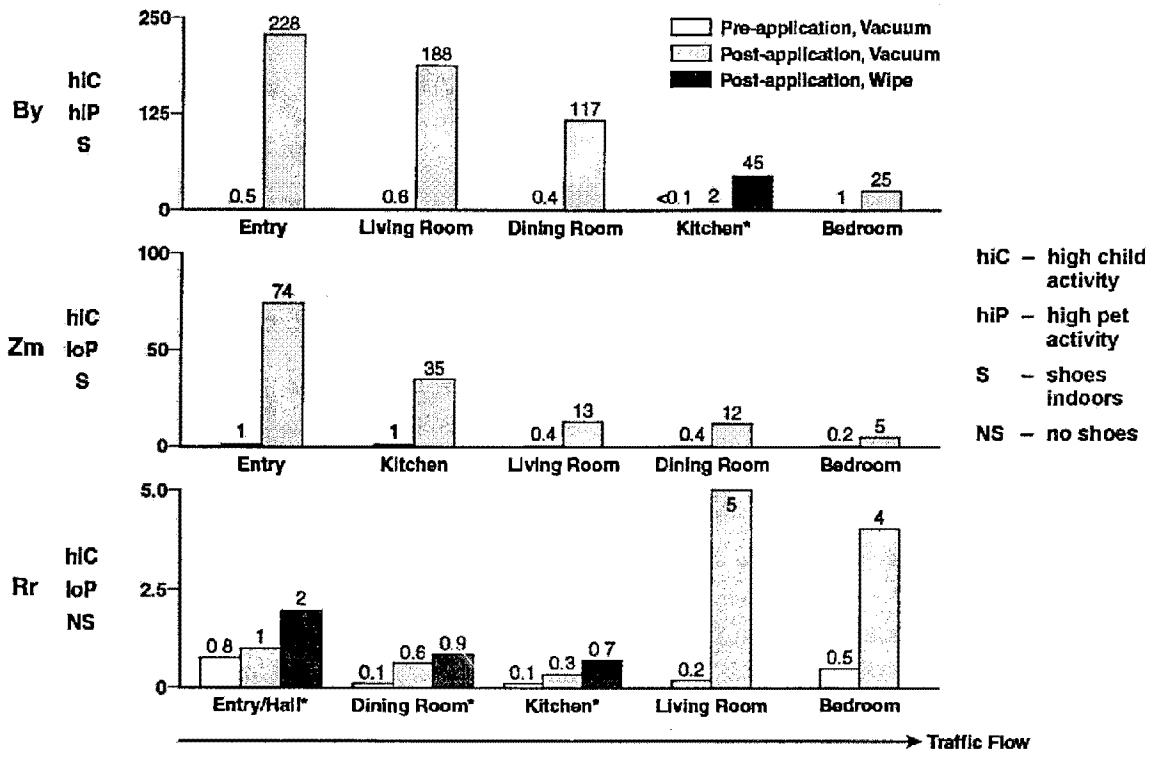

Figure 1. Floor dust loadings in µg/m^2 of 2,4-D in three homes one week after application of 2,4-D to lawn. Rooms marked with an asterisk were not carpeted.

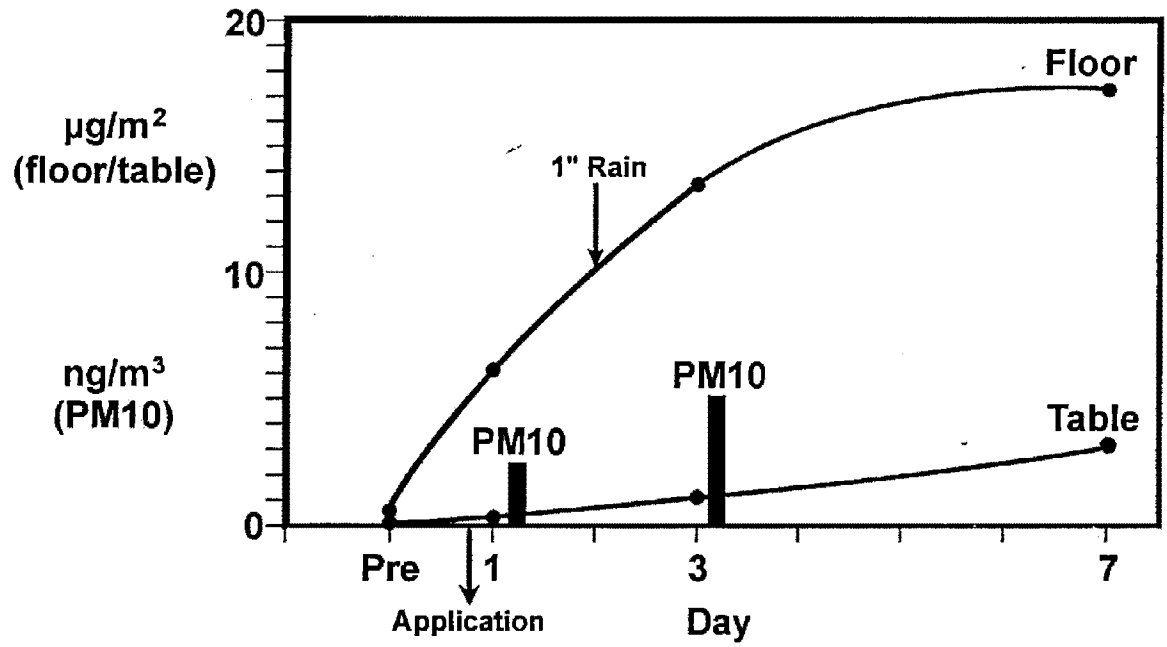

Figure 2. Temporal profile of 2,4-D residues inside a high activity home (*By*) before and after application of 2,4-D to lawn. Note that 2.5 cm of rain fell 24 h after application.

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INTRODUCTION

Pesticides are used inside the home and outside on the lawn or garden. After either application, they tend to accumulate indoors in air, house dust, upholstery, on surfaces, and also on children's toys. Pesticides sprayed on the lawn may be tracked indoors, where they can persist for months or years protected from environmental breakdown, as opposed to days outside on the grass. Typically, pesticide concentrations in indoor air and house dust are 10-100 times those found in outdoor air and surface soil. Over the past several years, parallel efforts by the U.S. Environmental Protection Agency (USEPA) have been underway to assess the potential exposures small children may receive from pesticide products used in and around the home. Initial studies [1] suggested that infants and toddlers may receive proportionately larger exposures than adults from both the respiratory and dust ingestion routes. Small children are considered to be the population of highest risk since they spend most of their time indoors. Much of this time is spent in contact with floors, engaging in mouthing of hands, toys, and other objects.

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(as a mouthing child's would often be) much more efficiently than by dry skin [4].

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METHODS

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RESULTS

Indoor air concentrations of 2,4-D increased from non-detectable before lawn treatment to 0.2 to 10 ng/m^3 (10 μm inlet) after homeowner application, with about 65% of the total particulate 2,4-D associated with respirable particles (<10 μg/m^3). 2,4-D associated with <1 μm and smaller particles made up 25-30% of the total mass. Detectable residues of 2,4-D were found on all surfaces one week after application. The surface concentration gradient followed the occupant traffic pattern through the house. Post-application floor surface loadings of 2,4-D in the living areas ranged from 1 to 228 μg/m^2 on carpeted floors and 0.2 to 20 μg/m^2 on bare floors, compared to 0 to 0.8 μg/m^2 (median 0.5 μg/m^2) pre-application. About 1% of the 2,4-D in floor dust was dislodgeable (PUF roller wetted with acetonitrile:phosphate buffer) and potentially available for dermal contact. 2,4-D residues on window sills and tables followed a similar traffic gradient, with surface loadings of 0.2 to 20 μg/m^2 (none were detectable before application). In homes in which occupants removed their shoes at the entryway, 2,4-D loadings on floors were typically an order of magnitude lower than in those in which shoes were worn.

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Relative exposures to 2,4-D via the air route vs. dust ingestion routes could not be accurately estimated since floor dust sampling was conducted several days after air sampling (except for the last year). However, since Day 3 air levels were on average higher than Day 1 levels, airborne 2,4-D was probably due primarily to dust resuspension and would be expected to be at similar or higher levels on Day 7 (when floor dust was collected). Hence for Home *By*, the average daily respiratory dose based on PM_{10} received by children between the ages of 6 months and three years spending 24 hours indoors (avg. 6.4 m^3/d inhalation rate [9]) one week after homeowner application would have been 68 ng/d, while the average dose received from ingestion of 100 mg [9] of floor dust from the living room would have been 100 times higher at 6.7 µg/d. After professional application, these potential exposures were reduced to 16 ng/d vs. 4.2 µg/d, respectively. For Home *Zm*, air and dust route exposures would have been 8.1 ng/d and 2.8 µg/d after homeowner application and 15 ng/d and 0.6 µg/d, respectively, after professional lawn treatment. By contrast, in shoes-off home *Rr*, which had very low post-application dust loadings and relatively low concentrations of 2,4-D in house dust, ingestion of 100 mg of dust would have resulted in the intake of only 0.5 µg/d and respiration to 4.4 ng/d after homeowner treatment. The mean dust contribution over all homes in the study is estimated to have been about 2 µg/d and the mean air route contribution18 ng/d one week after homeowner application compared to 12 ng/d and 1.2 µg/d, respectively, after professional treatment. It should be noted, however, that this comparison may not be valid for the sum of all participating homes since only half the homes participated in both phases of the study, which were also conducted in different years. For a 10 kg child, the average total potential dose via the inhalation and non-dietary ingestion routes would be 0.1 to 0.2 µg/kg/d. This is substantially less than the EPA Reference Dose of 10 µg/kg/d or the World Health Organization's Acceptable Daily Intake value of 300 µg/kg/d. However, these reference doses are not necessarily meant for infants and toddlers.

Use of dislodgeable residue data obtained with the PUF roller, combined with frequency of mouthing activity, may also be used to estimate intake by non-dietary ingestion for comparison with estimates made from ingestion of 100 mg/d of house dust. Assuming that the floor-to-hand transfer efficiency was the same as that for the wetted PUF roller, 10 mouthing events (one whole hand) per hour for 12-h/d [10], and 50% efficiently for removal of 2,4-D by salivation [4], exposure estimates agree quite well with those obtained on the basis of dust ingestion of 100 mg/d for a 10 kg child.

Additional research is underway in occupied homes and in the USEPA test home to examine fate

and transport of pesticides from indoor applications and determine the potential for human exposures. House dust has been fractionated by sieving and aerosol suspension into seven fractions ranging from <4 μm to 500 μm and analyzed to determine the distribution of pesticides as a function of particle size (concentrations increase with decreasing particle size). The mechanics of transfer of particles from outdoors to indoors on shoes, from indoor surfaces to dry and wet skin, and from floors into air by human activity are being investigated. Other studies are underway to better determine surface-to-skin and skin-to-mouth transfer efficiencies, pesticide bioavailability from dust, and the relationship of child activity patterns to residential exposures. Such studies are essential before accurate exposure assessments can be made.

DISCLAIMER

This work has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review process and approved for presentation and publication. Mention of tradenames or commercial products does not constitute endorsement or recommendation for use.

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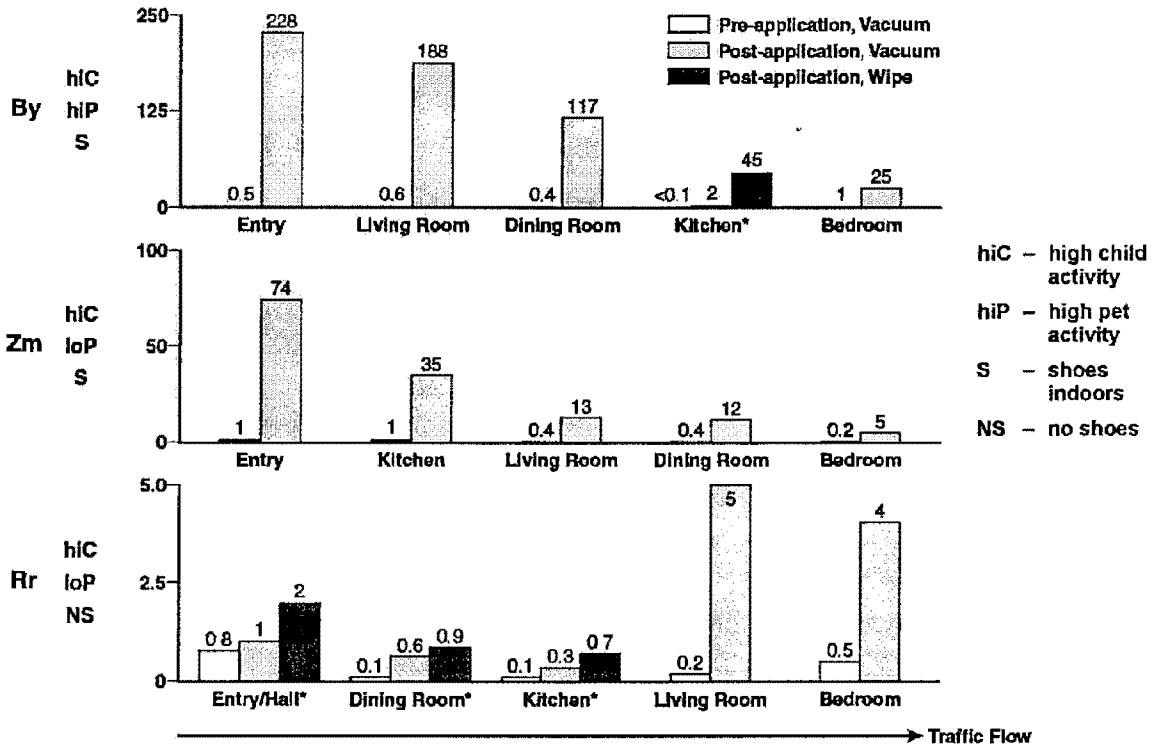

Figure 1. Floor dust loadings in µg/m² of 2,4-D in three homes one week after application of 2,4-D to lawn. Rooms marked with an asterisk were not carpeted.

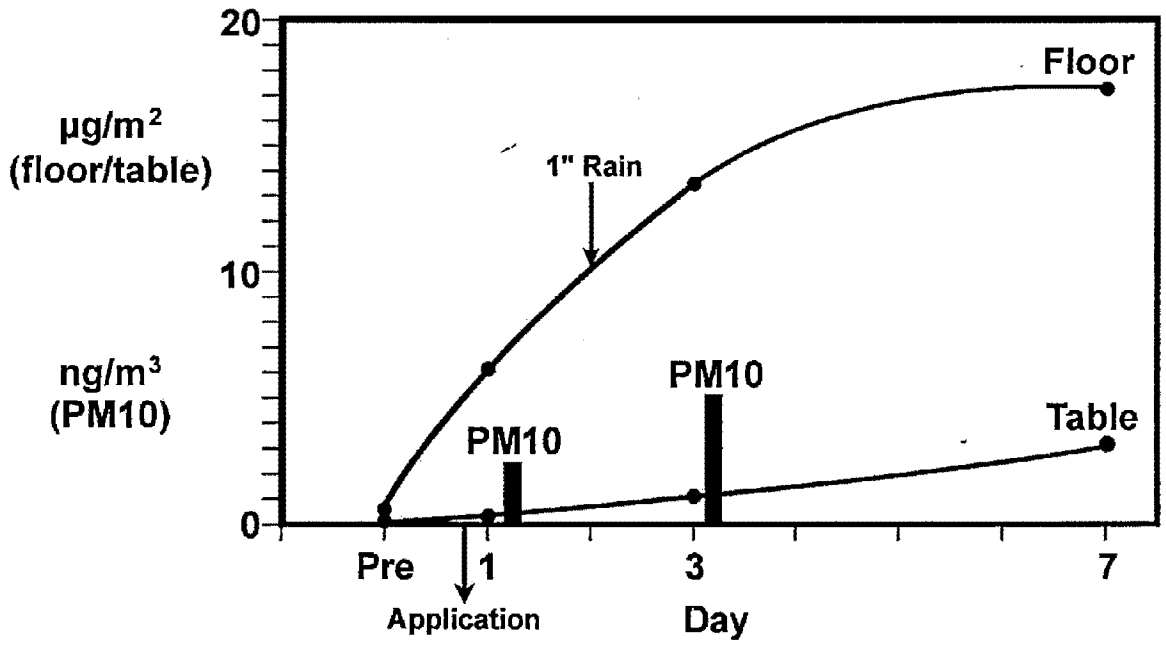

Figure 2. Temporal profile of 2,4-D residues inside a high activity home (*By*) before and after application of 2,4-D to lawn. Note that 2.5 cm of rain fell 24 h after application.

TECHNICAL REPORT DATA

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7. AUTHORS R.G. Lewis and M.G. Nishhioka	8. PERFORMING ORGANIZATION REPORT NO.
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16. ABSTRACT

Methods have been developed to estimate children's residential exposures to pesticide residues and applied in a small field study of indoor exposures resulting from the intrusion of lawn-applied herbicide into the home. Sampling methods included size-selective indoor air sampling; wipe sampling of floors, sills, tables; the polyurethane foam (PUF) roller for dislodgeable carpet surface residues; and the HVS3 vacuum sampler for floor dust. Personal exposure samples included hand wipes and morning void urine samples.

Pesticide spray drift was found to result in only a relatively minor increase in indoor pesticide levels. Post-application air intrusion from closed house ventilation and the opening and closing of doors and windows increased indoor background levels 6-fold, while track-in by high activity children and pets, and wearing shoes indoors, increased indoor levels by 37-fold. Indoor 2,4-D levels were found to increase continually over a one-week period, with the increase in indoor air levels corresponding to the increased floor dust levels, suggesting resuspension of house dust by human activity. Similar estimates of non-dietary exposure are obtained from models based on 100 mg ingestion and surface contact simulated by the PUF roller.

17. KEY WORDS AND DOCUMENT ANALYSIS		
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