Movement and Deposition of Pesticides within Residences after Interior and Exterior Applications.

Robert G. Lewis
U.S. Environmental Protection Agency, National Exposure Research Laboratory, MD-44, Research Triangle Park, NC 27711

Christopher R. Fortune and Fredrick T. Blanchard
ManTech Environmental Technology, Inc., P.O. Box 12313, Research Triangle Park, NC 27709

David E. Camann
Southwest Research Institute, P.O. Box 28510, San Antonio, TX 78228

ABSTRACT

In a study begun in 1999, the Environmental Protection Agency (EPA) is investigating the temporal and spatial distributions of pesticides applied by homeowners and commercial applicators for indoor crack and crevice and exterior perimeter treatments. In each participating household, pesticide levels in indoor air at 10-cm and 75-cm above the floor (child's breathing zone) were measured pre- and post-application (0-14 days), along with dermal- and vacuum-dislodgeable floor residues, deposition on table tops and dinnerware, absorption by surrogate food, and residues on children's hands and toys. Surface soil residues were measured in the case of perimeter application. Monitoring devices and methods used include the URG 2500 air samplers (PM 2.5 inlet), the HVS3 vacuum sampler, the PUF Roller, hand wipes, and various techniques for deposition monitoring. Results from the study demonstrate the nature and magnitude of translocation of pesticides from the areas of application to surfaces accessible for human contact. This paper presents data from two indoor applications of diazinon (O,O-diethyl O-[6-methyl-2-(1-methylethyl)-4-pyrimidinyl] phosphorothioate, CAS No. 333-41-5) and one outdoor (perimeter) application of chlorpyrifos [O,O-Diethyl-O-(2-isopropyl-6-methyl-4-pyrimidinyl) phosphorothioate, CAS No. 2921-88-2]. The post-application redistribution of the pesticides within the home and the exposure potentials via various routes (respiration, dermal contact, ingestion) are discussed.

INTRODUCTION

Pesticides may be periodically introduced into indoor air for pest control by direct application (e.g., insect sprays and bombs). They are also applied outdoors on lawns, in gardens, or around the foundation of the house to control weeds and insect damage. Most pesticides applied indoors are semivolatile (saturation vapor pressures between 10^{-2} kPa and 10^{-8} kPa) and vaporize from treated surfaces (e.g., carpets and baseboards). Both semi- and nonvolatile pesticides can be resuspended into air on particles by human and pet activity. Insecticides and herbicides applied outside the home can intrude into the indoor living spaces by vapor penetration or spray drift and be tracked inside by humans and pets. Whether used inside the home or outside on the lawn or garden, pesticides accumulate on indoor surfaces (particularly in carpet dust), in food, on

dinnerware, and on children's toys were they may present exposure risks to humans, especially small children.

Previous studies have investigated the movement, or redistribution, of pesticides applied indoors to air, floors and other indoor surfaces.[1,2,3,4] Other studies have examined the transport of pesticides applied outside the residence to the interior.[5,6,7] Several residential pesticide monitoring studies have addressed exposures to small children, but few of these have involved measurements made on children or their toys.[2,7,8,9] In this study, we have conducted a broad, multi-media residential monitoring effort, including the first concurrent measurements on children's hands and toys. In this paper, we report the results obtained from an intensive monitoring program conducted in one home over two week periods after two pesticide applications separated by seven weeks.

METHODS

Study Design

The study home (designated as Home 2) was a split-level frame house in central North Carolina built in 1961. The ground-floor level was built on a foundation with crawl space and the upper level was over a furnished half-basement. The house was surrounded on all sides by yard that was mostly turf covered in front and mostly bare soil in back. The main living area, or family room, was on the ground level and had a standard exterior door to the back yard and a sliding glass door to an adjoining screened porch, a double window, and a large entryway to the dining area and adjacent kitchen. The family room had a floor area measuring 4.4-m x 4.8-m and was fully carpeted with wall-to-wall plush nylon carpeting. The 3.2-m x 3.5-m kitchen area had vinyl flooring, Formica®-covered counter tops, wooden cabinets, a doorway to the carport, and a single window over the sink. One side of the kitchen was fully open to a 3.4-m x 3.5-m dining area, which was carpeted and connected to the family room through a 2.4-m wide entryway. The monitored bedroom (that of a four year-old boy) was on the upper level, had a hardwood floor (3.0-m x 3.6-m), on which was a 74-cm x 127-cm throw rug, and a double window. A hardwood-floored hallway, partially covered with a throw rug, led from the staircase to this bedroom, the bedroom of a two year-old girl, and the master bedroom, as well as to a bathroom, all of which had entrances from the hallway. Another bathroom was connected to the interior of the master bedroom and a half bath was located in the half-basement, near the bottom of the staircase. The home was equipped with a central heating, ventilation and air conditioning (HVAC) system. The furnace was fueled with natural gas.

On August 5, 1999, the homeowner applied diazinon as an aerosolized emulsifiable concentrate (EC) (Ortho Home Defense® Hi-Power® Brand Roach, Ant & Spider Killer, 0.5% a.i., Monsanto Co., San Ramon, CA) to the cracks and crevices in the kitchen (between cabinets and floors, and inside the cabinet under the kitchen sink), in the family room (baseboards only), and three bathrooms of the house (Application 1). The total quantity of diazinon applied indoors was estimated to be about 0.5 g by weighing the aerosol can before and after use. A similar formulation, with much higher diazinon concentration (Spectracide® Diazinon Multi-Purpose Insect Spray Concentrate, 25% a.i., Spectrum Group, St. Louis, MO), was also applied on the same day to the full outside perimeter of the house. About 320 mL of the concentrate, or approximately 75 g of diazinon, was applied to a 1-m-wide band of soil around the perimeter of

house and on the exterior walls of the house up to 1 m above ground level. The perimeter treatment also included the exterior doorway thresholds.

The indoor application was by pressurized aerosol spray can, while a hose-end sprayer was used for the outside application. The interior and exterior of the house had been treated with diazinon in the same manner three months earlier, shortly after the owner purchased the residence. Multi-media monitoring was conducted one day before Application 1 (pre-application) and one, four, eight, and twelve days post-application.

On September 23, 1999, the owner once again applied the diazinon formulation indoors in the same manner as before, but used an EC formulation of chlorpyrifos (Spectracide® Dursban® Multi-Purpose Insecticide Spray Concentrate, 6% a.i., Spectrum Group, St. Louis, MO) for the perimeter treatment (Application 2). Approximately 315 mL of formulation (19 g of chlorpyrifos) was applied. The pre-application sampling was performed one week prior to the pesticide treatments and the post-application monitoring schedule for Application 2 was the same as that for Application 1.

Sampling Strategy

Most of the sampling methods used in this study have been previously reported. Air sampling was performed using EPA Compendium Method TO-10A (ASTM D 4861), which employed a URG 2500-25A (URG, Chapel Hill, North Carolina) model self-contained air sampling cartridge comprised of a 2.5 μm inlet, 2.2-cm quartz-fiber particle filter and a 2.2-cm x 7.6-cm polyurethane foam (PUF) vapor trap.[10, 11, 12] Air was pulled through the sampling cartridge at 3.8 L/min for 24 h with a Du Pont Model P-4000 pump (Du Pont Co., Kennett Square, PA). Although these pumps, which are no longer manufactured, are exceedingly quiet during operation, they were placed in Styrofoam®-insulated ice chests to assure that the occupants of the home were not disturbed. Vacuumable carpet dust was collected with the HVS3 dust sampler (CS-3, Inc., Sandpoint, Idaho) following ASTM standard practice D 5438.[2, 11] Skin-dislodgeable residues were estimated using the PUF Roller technique according to ASTM standard practice D 6333.[2, 11] Table-top deposition was determined by wiping a 30.5-cm x 30.5-cm Formica® sheet (placed for 24 h on a table or chest) with two cotton gauze sponges (Sof-Wick® dressing sponges, 10-cm x 10-cm, 6-ply, Johnson & Johnson Medical, Inc., Arlington, Texas), each wetted with 10 mL of 98% 2-propanol (pesticide quality).[13] The sheets were double-wiped sequentially with two sponges, using nitrile rubber gloves and lateral wiping motions in perpendicular directions. Floor wipes were performed in the same manner with the aid of a 30.5-cm x 30.5-cm template. Child hand wipes were conducted according to a procedure previously reported, using the same type of gauze sponges and wetting agent.[14] The child's toy was a 12-cm tall, semi-rigid plastic doll in the likeness of a Teletubby® character (Hasbro, Inc., Pawtucket, RI). It was wiped in the same manner as the children's hands. To determine the potential contamination of dinnerware and food stored in kitchen cabinets, two devices were used: (1) a 20-cm glazed ceramic dinner plate placed in a closed cabinet and wiped with 2-propanol-wetted gauze in a manner similar to that employed for the deposition sheets; and (2) a 5.5-cm x 7.6-cm solid PUF cylinder placed in a cabinet with grains and cereals. The latter served as a passive air sampler to estimate vapor sorption by foods. Soil samples, collected before and after Application 2, were obtained by scraping 30.5-cm x 30.5-cm surface areas (to a depth of ca. 0.5-cm) with the aid of a template.

Air samples were collected over 24-h periods at 10-cm and 75-cm above floor level in the family room near the door to the back yard and in the 4-year-old boy's bedroom on the second level. (Note: For safety considerations, the air samplers could not be operated in the 2-year-old girl's bedroom, which was adjacent to the boy's room.) The air sampling cartridges were assembled in the laboratory and transported to and from the field in sealed polyethylene bags. A ring stand was used to support the cartridges in horizontal orientations at the proper heights above the floor. The pumps were calibrated with an electronic bubble flow calibration unit (Mini-Buck Model M-5, A. P. Buck, Inc., Orlando, FL) and flow rates measured before and after each use. Duplicate samples were taken in the family room only on Day 1, Day 4, and Day 12. The pre-application air sampling was carried out the day before Application 1, but occurred one week before Application 2 due to an interruption caused by adverse weather conditions. A Formica sheet was placed on a table in the family room near the air sampling assembly and another was positioned on a chest of drawers in the boy's bedroom at the beginning of each air sampling period to determine deposition over the same 24-h period. At the same time, the test dinner plate was placed in the kitchen cabinet normally used for storing dinnerware and the PUF food surrogate was placed in the kitchen cabinet used to store cereals, flour, etc., both for the 24-h exposure period. Likewise, the toy was given to the female child at the beginning of each 24-h sampling period, was sampled (wiped) in the home at the end of that period to recover pesticide residues, and was replaced with a duplicate toy. The deposition sheets and dinner plate were also wiped in the home. Air, deposition and food exposure sampling were begun about four hours after each application (generally about 2:00 p.m.) and repeated four, eight, and twelve days later. All other samples (HVS3, PUF Roller, floor wipes, and hand wipes) were collected at the end of the air and deposition sampling periods.

HVS3, PUF Roller, and floor wipe samples were collected at several locations within the home. The primary location for the collection of dislodgeable residues was the family room. Three separate 1.0 m^2 areas of the plush carpeted floor were selected and marked off using a metal template and paper adhesive tape to outline the sample areas. Standard protocols were used to collect both the surface-dislodgeable residues (PUF roller method) and the vacuum-dislodgeable residues (HVS3 sampler method). The only protocol departure involved the collection of composite carpet dust samples and PUF Roller samples on three of the five days of testing in each study period. For those samples, each of the three HVS3 sample areas was split into three equal sections, and the HVS3 sampler was operated so that one of the three sections (left, center, or right) was vacuumed on each sampling day to yield a total area of 1.0 m^2 sampled. The PUF Roller samples that were collected on those days were similarly composited, in that two back and forth passes over a 1.0 m length were made over the carpet in areas adjacent to each of the three test areas using a single PUF Roller ring; thus resulting in six passes, 1.0 m in length, and covering a total surface area of 228 cm^2. PUF roller samples were also collected in the same manner from three separate 1.0 m long areas of a throw rug located in the male child's bedroom.

The floor wipe procedure used was similar to the procedure described above for the collection of the other surface wipe samples; i.e., two propanol-wetted gauze pads were used to wipe the surface in perpendicular directions. Duplicate floor wipe samples were collected from the kitchen floor before and on Days 1 and 4 after Application 2. Single floor wipe samples were collected in the child's bedroom on every sampling day during both study periods. In all cases,

care was taken to ensure that there was no overlap of the sample areas wiped during a given round of testing.

The wipe samples of the children's hands were collected at the convenience of the parents, taking into account the child's activities on the test day. The children's hand wipe samples were usually collected at a time near the end of the other sampling procedures. Fresh nitrile gloves were used for each child, and a separate gauze pad was used for each hand. One clean surface of a folded-over pad was used first to wipe the back of the hand and fingers, then the opposing surface of the pad was used to wipe the palm side of the hand and fingers. The pad was then unfolded and turned inside out, and this fresh surface was used to thoroughly clean the inside surfaces between the fingers and the thumb. This process was then repeated using a fresh treated gauze pad to wipe the other hand.

A duplicate of the PUF plug food surrogate served as a field blank. A clean field blank was taken to the home at the beginning of each sampling period, placed in the food storage cabinet, and immediately returned to its transport container (glass jar with a PTFE-lined lid, sealed with PTFE tape). At the end of each sampling period, the inlets and outlets of the air sampling cartridges were plugged and the cartridges resealed in plastic bags for shipment to the laboratory. The HVS3 carpet dust sample was returned in the sealed cyclone catch bottle. The PUF Roller media and PUF food surrogate were resealed in the same PTFE-lined glass jars used for transport to the field. Gauze wipes were immediately placed in individual glass jars to which an additional 50 mL of 2-propanol was added. All samples were shipped on dry ice to San Antonio, Texas for analysis.

Analytical Methods

Carpet dust samples were sieved to exclude particles larger than 150 μm (16-56% of mass) as specified in ASTM standard D 5438.[11] Soil samples were sieved to exclude particles larger than 2 mm (15-60% of soil mass), a 30 g aliquot separated for analysis, and a 2 g aliquot was taken to determine moisture content (16 to 38% water content). The sieved dust (≤2 g) and wet soil (30 g) aliquots, as well as the PUF air, roller and food surrogate samples were each spiked with terphenyl-d_{14} as a recovery surrogate. These samples were subjected to Soxhlet-extraction with 6% diethyl ether in *n*-hexane for 16 h (air filter and PUF plug combined), and the extracts concentrated to 1 mL (air), 10 mL (other PUF), or 2.5 to 10 mL (dust and soil). 2-Propanol-wetted gauze sponges used for surface (floor, table top, dinner plate) and hand wipes were likewise spiked with terphenyl-d_{14}, shaken for 30 min on a mechanical shaker, shaken twice more with 1:1 diethyl ether:hexane for 1 min (after decanting the prior solvent), squeezed, and all solvent fractions combined for concentration to 10 mL (5 mL for hand and floor surface wipes) by means of an N-Evap concentrator (Organomation Associates, Berlin, MA). One mL aliquots of the dust, soil, PUF, and hand wipe extracts were passed through a Florisil® (Floridin Corp., Tallahassee, FL) column and adjusted to 2 mL with 10% diethyl ether in hexane.

Diazinon and chlorpyrifos concentrations were determined by GC/MS using either a Hewlett-Packard 6890/5973 (Palo Alto, CA) or a ThermoQuest MD800 (Austin, TX) instrument operated in the selected ion monitoring mode. A 30 m x 0.25 mm i.d. (5% phenyl)-methylpolysiloxane (DB-5.625) column was used as the GC analytical column. The GC/MS instrument was scanned

to monitor two selected ions per analyte to achieve low level detection. Quantification was performed using d_{12}-labeled polycyclic aromatic hydrocarbons as internal standards. The percent relative standard deviation (%RSD) for each analyte was maintained within 30% during the initial five-point standard calibration. A continuing calibration standard was processed at the beginning and end of each sequence of 15 samples. The percent difference of each analyte in the mid-level standard was generally maintained within 40% of the initial calibration value during continuing calibrations. Soil concentrations were reported per g dry weight, based on a separate dried aliquot.

The nominal analyte detection limit used in this study was one-third of the analyte level in the lowest standard of the initial 5-point calibration curve. When both the analyte and an interfering compound coeluted, the entire peak was quantified and the analyte amount reported as ≤ the peak amount. Usual detection limits were 0.002 μg for air samples, surrogate food PUF and Application 1 PUF roller samples; 0.010 μg for dust and soil samples; 0.021 μg for floor wipes; 0.041 μg for hand wipes and Application 2 PUF roller samples; and 0.010 μg for other wipes.

Mean analyte recoveries for air (PUF + filter), surrogate food PUF, and 2-propanol gauze wipes were 93%, 94% and 92%, respectively, for chlorpyrifos, and 88%, 92%, and 87% for diazinon.

RESULTS AND DISCUSSION

First Application

The first application occurred in summer (August 5) and the monitoring period extended from August 3 to August 17. The weather during the period was unusually hot and dry. The mean outdoor temperature at the nearest weather station was 27.2°C (mean high 35.0°C, mean low 20.5°C), the mean relative humidity (RH) was 65% (range 23-80%), and only 4.5-cm of rainfall was recorded throughout the course of the study. Outdoor climatological measurements taken on site during the study agreed well with those reported by the local weather service. Indoor temperatures at the time of sample collection (usually mid-afternoon) ranged from 25.3 to 27.6°C (mean 27.2) and the indoor RH was between 38% and 56% (mean 48.5%). Windows and doors to the home were closed and the air conditioning was on during virtually the entire monitoring period.

The monitoring results from Application 1 are presented in Table 1. Air levels of diazinon in the family room, which received crack and crevice treatment, increased about 20-fold from less than 0.1 $\mu g/m^3$ on the day before the application to 1.5 $\mu g/m^3$ for the 24-h period following application (Day 1). Day 1 air concentrations in the upstairs bedroom of the 4-year-old male child were less than one third as high as those in the family room, at about 0.4 $\mu g/m^3$, showing a post-application increase of 12- to14-fold. Air concentrations declined over the next 12 days to about 20% of Day 1 levels in both rooms. Family room carpet dust collected with the HVS3 vacuum sampler 24 h after the application (Day 1) exhibited 8- to 10-fold increases in both the diazinon surface loading (4.1 $\mu g/cm^2$ vs. 0.5 $\mu g/cm^2$ pre-application) and diazinon concentration (1.2 μg/g vs. 0.1 μg/g). These levels remained about the same on Day 4 (4.0 $\mu g/cm^2$; 1.4 μg/g), but declined to 1.0 $\mu g/cm^2$ and 0.7 μg/g by Day 12. A single carpet dust sample taken on Day 8 from the ground-floor living room, which was not treated, revealed lower diazinon loadings and concentrations (0.58 $\mu g/m^2$ and 0.65 μg/g, respectively). Dust loadings were also lower in this

room (1.2 g/m^2 vs. 4-5 g/m^2 in the family room), which largely accounted for the lower diazinon loadings.

The highest floor diazinon loadings were found on the hardwood floor in the child's upstairs bedroom, with monitored levels highest on Day 4 at 9.4 μg/cm^2, approximately four times the pre-application level. However, the floor wipe data should not be compared to the HVS3 and PUF Roller data as the wipes likely removed nearly 100% of the diazinon residue, while the HVS3 and PUF Roller removal efficiencies were much lower.

The PUF Roller results reflect the likely potential contact transfer of floor residues to dry skin. The dislodgeable residues, as determined by the PUF Roller method, from the family room carpet were 10% to 15% of the diazinon loadings measured with the HVS3. The PUF Roller/HVS3 ratio was comparable to that reported in other studies.[2, 6, 13, 17] PUF Roller measurements taken on Day 1 of the throw rug in the child's bedroom were about one-third those found on the family room carpet.

Deposition of diazinon over a 24-h period onto the Formica sheet placed on a table top about 70 cm above the family room floor resulted in a loading of 2 μg/m^2 on Day 1 (vs. non-detectable pre-application levels). The rate of daily deposition slowly declined thereafter, with deposition on Day 12 amounting to about 35% of that observed on Day 1. Deposition on the sheet placed on top of the chest of drawers in the child's bedroom (1.2-m height) was lower than that observed downstairs (ca. 0.7 μg/m^2 on Days 1 and 4), likely due to a combination of factors: (1) the room was not treated, (2) the absence of dust-generating carpeting, and (3) the increased height of the deposition sheet above the floor. Deposition on the dinner plate in the closed kitchen cabinet (about 1.8 m above the floor and 1 m above the countertop) was detectable only on Day 1. The single dinner plate observation was less than two times the detection limit; therefore, its relatively high loading compared to other surfaces (ca. 2.1 μg/m^2) may not be meaningful. The PUF cylinders, which were used as food surrogates, were located in another closed cabinet on the opposite side of the kitchen from the dinner plates (ca. 1.6 m above floor level) and exhibited nearly the same level of diazinon (ca. 0.4 μg) for each 24-h exposure period throughout the study, although a 50% increase was observed on Day 1. A possible explanation for this behavior is that the exposed surfaces of PUF became saturated at about 0.4 μg, even when exposed to the relatively low pre-application air levels of diazinon. Note that the diazinon levels in the daily PUF food surrogate field blanks ranged from <0.003 to 0.004 μg, ruling out contamination.

No diazinon residues were found on the children's hands or the toy prior to the application. After the application, residues well above the detection limit were found on both children's hands, with levels on the male child's hands five to 10 times those found on the female's on Days 1 and 4. Hand size was a minor factor in the higher residues found on the boy's hands, which were less than 20% larger than the girl's (combined surface areas of both hands, ca. 380 cm^2 vs. ca. 310 cm^2, respectively). Based on visual observations made by the field sampling personnel and the child activity diary kept by the mother, however, it was apparent that the boy spent more time on the floor, which likely accounted for his higher hand residues. Hand residues for both children dropped considerably by Day 8 and were undetectable (<0.04 μg) by 12 days post-application. Residues on the surface of the toy, which was given to the two-year-old girl, were comparable to those found on her hands. The total surface area of the toy was estimated to

be 200-250 cm^2, comparable to that of a small child's hands. Field observations and activity diary data indicate that the girl played with the toy often and it is likely to have come into contact with many of the same surfaces that she contacted. Therefore, surface loadings were similar to those found on the female child's hands.

As can be seen from the data plotted in Figure 1, the diazinon air concentrations and dust loadings in the family room show similar temporal profiles over the 12-day period, all peaking on Day 1. The toy residues follow this same pattern, but residues on the girl's hands were highest on Day 4. The child activity diary showed that the girl had been off the premises for more than 2.5 h immediately prior to collection of the hand wipe sample on Day 1, which may explain why her hand loadings were lower that day than on Day 4, when she had been playing in the family room for nearly 2 h immediately prior to sample collection.

Application 2

The second application took place in the fall (September 23). Pre-application monitoring was conducted about one week in advance of the application (on September 15) and post-application monitoring was begun on September 24. The weather conditions were much cooler and damper during the fall post-application period than during the summer period. The mean outdoor temperature was 18.7°C (mean high 24.2°C, mean low 12.6°C), mean RH 84% (range 60-100%), and 33.6-cm of rainfall occurred as a result of a hurricane. Indoor climatological conditions were essentially identical to those outdoors, with temperatures at the time of sample collection ranging from 21.1 to 24.5°C (mean 23.0) and the indoor RH 53% to 87% (mean 66.3%). The windows to the domicile were frequently opened during this period, with little or no use of air conditioning and no heating employed.

The monitoring data for diazinon (applied indoors) in Application 2 are presented in Table 2 and the data for chlorpyrifos (applied outdoors) are shown in Table 3. Pre-application indoor air concentrations of diazinon in the treated family room and untreated child's bedroom were elevated relative to the summer pre-application levels, due presumably to accumulation from prior treatments. Day 1 indoor air concentrations were about half of those measured after Application 1, most likely due to the windows being opened; however, post-application air levels on subsequent days were similar to those observed in summer.

Carpet dust samples in the fall were significantly lower in diazinon loadings ($\mu g/cm^2$), but comparable to the summer levels in concentration ($\mu g/g$). Two possible reasons for the lower diazinon loadings were: (1) lower quantities of vacuumable dust (18 to 35% of that found during the summer) due to the fact that the same areas had been carefully vacuumed seven weeks earlier, as well as the likelihood of less indoor-outdoor traffic because of the rainy weather, and (2) reduced diazinon track-in due to the fact that the outdoor application of the insecticide had taken place 50 days earlier. Also, the increase in diazinon carpet loadings from pre-application to Day 1 was only about half that observed after Application 1 and loadings appear to have fallen more rapidly, both likely due to a lower level of track-in. The PUF roller likewise picked up less diazinon after Application 2. Floor wipe values from the upstairs bedroom floor, on the other hand, were as high or higher than those observed after Application 1, suggesting that the primary source of those residues in both cases was indoor aerosol and vapor deposition. Table top deposition in both the family room and bedroom were comparable after both applications.

More incidents of deposition on the dinner plate were detected after Application 2, but residues were still small and often close to the detection limit. Again, the PUF plug food surrogate exhibited relatively constant residues throughout the study (0.2 to 0.4 µg, vs. <0.002 to 0.003 µg for the blanks). Wipes of the kitchen floor (30.5-cm x 30.5-cm areas) were performed in the fall monitoring event only. Diazinon loadings there were much higher than those observed in other rooms, ranging up to 262 µg/m^2 on Day 4. The relatively high pre-application loading on the kitchen floor may reflect residual from Application 1. The analyses of the second set of collocated samples from the kitchen floor by a different laboratory using a similar analytical method yielded a lower pre-application loading for diazinon (shown in parentheses in Table 2), but the overall results were reasonably comparable considering the expected non-uniformity of the incidental residues. Analyses of perimeter soil samples (surface scrapings from 30.5-cm x 30.5-cm areas) revealed diazinon residues in the 0.3 to 2.3 µg/g range remaining from Application 1. These residues appeared to be declining rapidly between the pre-application and Day 4 monitoring, most likely due to the occurrence of more than 33 cm of rain during the period. Soil levels remained somewhat constant after Day 4.

Diazinon residues on the children's hands were lower after Application 2. Unfortunately, a sample from the 4-year-old boy could not obtained on Day 1, and two of the other samples were compromised by leakage of the sample container. None-the-less, the highest observed diazinon skin residue (1 µg on Day 8) was on the boy's hands, as before.

Monitoring data for chlorpyrifos are presented in Table 3. Analysis of surface soil near the foundation revealed post-application chlorpyrifos concentrations of 3 to 19 µg/g (20 to 200 µg/m^2), comparable to the diazinon loadings found on the kitchen floor), with the highest measurement occurring on Day 1. These residues may have been higher and persisted longer during times of normal rainfall. The rainfall between the time of perimeter treatment and Day 4 amounted to 5.7 cm, followed by another 8.2 cm of rainfall between Day 4 and Day 8.

Despite the fact that the chlorpyrifos formulation was applied outdoors, however, concentrations in indoor air, dust and on solid surfaces generally increased after the application. Air concentrations in the family room were more than double pre-application levels (0.09 vs. 0.04 µg/m^3) and appeared to have risen slightly by Day 4. Little, if any, increases were observed in bedroom air residues. In both cases, concentrations were much lower than those found for diazinon. Carpet loadings of chlorpyrifos increased on Days 1 and 4 by about the same magnitude as those for diazinon, further suggesting that track-in was the principal source of these residues. Bare-floor chlorpyrifos residues in the boy's bedroom were as high or higher than the diazinon residues, suggesting that they were tracked in from outdoors by the child. The same was true for deposition residues found on the chest of drawers in that room, which may be attributed to the settling of re-suspended tracked-in residues. Dislodgeable residues (PUF Roller) of chlorpyrifos on the throw rug in the bedroom were also detectable on Day 1, while no diazinon was detectable on any day. No dislodgeable residues of either pesticide above the detection limits were found on the hallway throw rug outside the bedroom door. Family room table-top residues of chlorpyrifos were higher than those for diazinon, again suggesting resuspension of tracked in outdoor residues. The PUF Roller showed quantifiable levels of chlorpyrifos and diazinon on the family room carpet on Day 1 only. Comparisons of air and carpet dust residues in the family room are shown graphically in Figure 2.

Chlorpyrifos loadings on the kitchen floor were substantially lower than those for diazinon, and were comparable to those found on the floor of the child's bedroom. Positive results for chlorpyrifos on the dinner plate were observed on every day, both before and after Application 2. Although these levels were relatively low, they were, surprisingly, higher than those found for diazinon. However, chlorpyrifos is ubiquitous in the residential environment.[1, 2, 16, 18] In contrast to the dinner plate, the chlorpyrifos residues in the PUF plug food surrogate were minimal, but positive (0.01 to 0.04 μg compared to <0.003 to 0.007 μg for the field blanks).

As was the case with the dinner plate, chlorpyrifos residues on the child's toy were higher than those for diazinon. This finding was reflected in residues found on the girl's hands. In fact, Day 1 chlorpyrifos measurement on her hands represented the only instance in both the summer and fall applications in which a pesticide residue in excess of 1 μg was found on the girl's hands. The lowest chlorpyrifos residue on the girl's hands (0.13 μg), found on Day 12, corresponded to the highest level found on the toy (1.04 μg). The child's activity diary revealed the doll was not played with at all on Day 12, suggesting that the higher residues were the result of vapor adsorption or atmospheric deposition and remained on the toy because there was no opportunity for contact transfer to other surfaces. The highest chlorpyrifos contamination of the boy's hands (taking into account that there were only two reliable measurements) occurred before the perimeter application. The source of this residue could not be ascertained, but the boy was in kindergarten that day until shortly before the sample collection.

CONCLUSIONS

This study is believed to be one of the most intensive multi-media monitoring efforts conducted in a single household following both indoor and outdoor applications. The methods employed performed well except for the food surrogate, which appeared to saturate at relatively low exposure levels. This effort has clearly demonstrated, as have previous studies, that pesticides applied inside the home are transported to non-targeted surfaces and untreated rooms. It has also shown that pesticides applied outdoors are transported to the indoors, where they are re-distributed into the indoor air and onto various interior surfaces. In the first phase of this study (Application 1), diazinon was applied both indoors and to the outside of the house. Therefore, the resultant indoor residues undoubtedly arose from both sources. Analysis of data from Application 2 suggests that the indoor air levels of diazinon observed after Application 1 were primarily influenced by redistribution of residues from indoor treatment, while track-in from the perimeter treatment may have contributed significantly to carpet residues. After Application 2, most of the indoor chlorpyrifos contamination observed was the result of track-in or vapor/drift intrusion from outside, although background residues from historical use likely contributed to the overall burden.

Except for Day 1, the air levels of diazinon found in the child's breathing zone (10 to 75 cm above the floor) in this study were not significantly higher than the mean summer indoor air concentrations reported for about 140 homes in Jacksonville, Florida by the Non-occupational Pesticide Exposure Study (NOPES).[18] That study, conducted by the EPA during 1986-88, found diazinon in 83% of the Jacksonville homes monitored at mean indoor air concentrations of 0.42 $\mu g/m^3$ in summer, 0.19 $\mu g/m^3$ in spring and 0.09 $\mu g/m^3$ in winter. Chlorpyrifos air levels found in the current study were significantly lower than those reported by NOPES in Jacksonville, where the insecticide was found in the indoor air of 100% of the participating homes in summer

at a mean concentration of 0.37 μg/m^3; 88% in spring at 0.20 μg/m^3, and 96% in winter at 0.12 μg/m^3. However, the mean indoor air concentration at 75 cm reported by Lewis et al.[2] for chlorpyrifos found in 100% of nine homes monitored in central North Carolina in the fall of 1990 was only 0.08 μg/m^3, which is similar to that reported here. Diazinon was rarely encountered in the 1990 North Carolina study, being found in only two of five samples collected in one of the nine participating homes.

Chlorpyrifos is ubiquitous in the residential environment and the pre-application residues found in this study likely reflect use and track-in prior to purchase of the home by the participating family. Intrusion of chlorpyrifos into the home following the perimeter treatment, however, probably accounted for most of post-application indoor residues. Despite the treatment, the chlorpyrifos carpet loadings and dust concentrations observed in this study were lower than those reported by Lewis et al.[2] for the aforementioned North Carolina study (1.3 μg/m^2 and 1.6 μg/g, respectively). Chlorpyrifos was one of the major contaminants found in a composited carpet dust collected in 1996 from 25 homes located in the same area as Home 2, with concentrations ranging from 0.54 μg/g on the coarse fraction (<2 mm mean particle diameter) to 4.5 μg/g on respirable fraction (<4 μm).[16] Diazinon was not found in this sample at concentrations above the detection limits of 0.05 μg/g. In carpet dust vacuumed with the HVS3 from 362 homes in nine northern and midwestern states, Camann and Buckley[19] found chlorpyrifos in 61% of the samples at a median concentration of 0.54 μg/g (among positive samples) and loading of 0.82 μg/m^2 and diazinon in 21% of the samples at very similar median levels (0.52 μg/g and 0.74 μg/m^2). The National Institute of Standards and Technology Standard Reference Material® (SRM) 2583, an indoor dust standard collected from households, cleaning services, hotels, and motels located in North Carolina, Maryland, Ohio, and New Jersey, has been shown to contain 0.90 μg/g of diazinon and 0.54 μg/g of chlorpyrifos.[16] It can be deduced from previous studies[6,7] that the major contributor to chlorpyrifos in the indoor environment in this study was track-in by the residents. The rainy weather, along with the fact that the homeowner did not wear his shoes indoors after the application and the absence of indoor-outdoor pets undoubtedly minimized track-in.

There are few reports in the published literature that offer data for comparison with the other measurements performed in this study. Lewis et al.[2] used an earlier model of the PUF Roller with water-moistened sampling media in the aforementioned North Carolina children's exposure study. Despite the higher carpet loadings and presumed higher sampling efficiency, they reported mean dislodgeable residues of chlorpyrifos from family room carpets in six of nine residences of 0.11 μg/m^2, compared to <0.17 to 0.22 μg/m^2 in this study. The maximum value reported by Lewis et al. was 0.32 μg/m^2 for a home that had received a crack and crevice treatment with chlorpyrifos two days earlier. The latter value is similar to that obtained for diazinon one to four days post-application in the current study. Several researchers have studied pesticide deposition on table tops and on aluminum plates or Petri dishes,[1,7,21,22] but none of the published studies were sufficiently similar to the current effort for reasonable comparison of results. Leidy et al.[22] found residues of 3,500 μg/m^2 of diazinon and 4,600 μg/m^2 of chlorpyrifos on Formica sheets placed in non-targeted areas of unoccupied dormitory rooms one day after aerosol spray crack and crevice treatments with 1% and 0.5% formulations, respectively. While these diazinon residues cannot be directly compared with our findings for the family room of

House 2 due to differences in room size and the intensity of the dormitory treatment, as well as the higher concentration of a.i. applied. However, the Leidy et al. results would be expected to be somewhat comparable to the kitchen floor residues observed here, but the deposition amounts they observed were much greater than we observed.

Employing the same methods used here, EPA recently reported that table top deposition of re-suspended floor dust contaminated with tracked-in 2,4-dichlorophenoxyacetic acid dimethylamine salt used as a herbicide on lawns was about 25% of floor loadings in the same room.[21] In this study, table top residues of diazinon after Application 1 were 30-70% of the carpet loadings and often higher than carpet loadings after Application 2, although the deposition data appear less reliable in the fall study. In the case of diazinon, much of the table top residues may have resulted from volatilized pesticide, rather than deposition of re-suspended floor dust.

A subset homes located throughout Arizona participating in EPA's National Human Exposure Assessment Survey (NHEXAS) were subjected to multi-media sampling and the results recently published.[23] Diazinon was found in 53% of the house dust samples at <0.02 to 50.5 $\mu g/m^2$ (<0.02-66.2 $\mu g/g$); indoor air, 63%, <0.002-20.5 $\mu g/m^3$; hand wipes (primarily adults, both hands), 32%, <0.01-18.4 μg; and foundation soil (2.5 cm depth), 37%, <0.007-7 $\mu g/g$. Chlorpyrifos was more prevalent and generally at higher concentrations: house dust, 88%, <0.004-48.5 $\mu g/m^2$ (median 0.14 $\mu g/m^2$; 90th percentile 2.2); indoor air, 65%, <0.003-3.2 $\mu g/m^3$ (0.008 $\mu g/m^3$; 0.085 $\mu g/m^2$) ; hand wipes, 36%, <0.03-544 μg (0.03 μg; 2.6 μg); and foundation soil, 48%, <0.001-85 $\mu g/g$ (<0.03 $\mu g/g$; 0.13 $\mu g/g$). Home 2 in the current study would fall between the 50th and 90th percentiles, except for foundation soil, which exhibited higher residues because it received direct treatment.

Child hand residues in this study were relatively high compared to other studies. Diazinon residues found on the 2-year-old girl's hands on Day 1 through Day 8 (<0.04 to 0.30 μg after Application 1 and <0.04 to 0.07 μg after Application 2) were generally higher than levels for several pesticides determined by Lewis et al.[2] using 2-propanol hand washes on 1.5 to 3.5 year-old children residing in homes not recently treated (0.004 - 0.1 μg; mean 0.08 μg). They were, however, similar to residues measured by Geno et al.[14] for children of farmers in Minnesota using the same hand wipe procedure employed here. Residues found on the 4-year-old boy's hands were much higher than most of the observations reported in the literature (3 of 7 post-application measurements exceeded 1 μg). The only report of pesticide residues on children's hands in excess of 1 $\mu g/m^2$ known to us is that of Camann et al.[23], who found the herbicide metolachlor at 1.3 to 1.7 μg levels on the hands of two children, aged 3 and 5.5 years, residing on two Iowa farms the day after application.[24]

Diazinon residues on the toy ranged from 0.06 to 0.11 μg after Application 1 and ≤0.03 to 0.05 μg after Application 2, similar to residue levels on the hands of the child (girl) who played most with the toy. Chlorpyrifos levels on the toy after Application 2 were much higher (0.10 to 0.97 μg) than those of diazinon, as were the chlorpyrifos residues on the girl's hands (0.13 to 1.27 μg). These levels were much lower than those reported by Gurunathan et al.[24] for deposition on unhandled plastic toys left in apartments treated with chlorpyrifos (mean peak value 11.5 $\mu g/cm^2$). In that study, however, the entire toy was extracted with solvent to recover the

pesticide residue and the authors' assumed, probably incorrectly, that the residues were confined to the outside surface.

For exposure assessment purposes, EPA estimates that a child aged one to two years breathes 6.8 m^3 of air per day.[20] Therefore, a child of this age would have received a respiratory dose of about 6.5 µg of diazinon on Day 1 after Application 1 and a corresponding dose of 3.1 µg after Application 2 had the child spent half of the time in the family room and half in the bedroom. The Day 12 respiratory exposure estimate following this scenario would have been 1.5 µg for either application. Inhalation exposure to chlorpyrifos would have been much lower: 0.6 µg on Day 1 and 0.4 µg on Day 12 after Application 2. The average breathing rate for children aged three to five suggested by EPA is 8.3 m^3/d, which more closely approximates that of the child occupants of Home 2. Consequently, the children in this study would have potentially inhaled about 1.2 times the above quantities of pesticides (7.6 µg and 3.7 µg for diazinon on Day 1 for the two applications; 0.7 µg for chlorpyrifos on Day 1 after the second application).

The average dust and soil ingestion rate for small children is estimated by EPA to be 100 mg/d.[20] At this rate, the maximum quantity of diazinon that a child in Home 2 may have ingested after either application is only 0.14 µg/d. A similar dust ingestion rate for chlorpyrifos may be calculated after Application 2. Ingestion of 100 mg of perimeter soil, on the other hand, could have resulted in the mean intake of 1.8 µg of chlorpyrifos on Day 1 and 0.3 µg on Day 12. Had weather conditions been more favorable, the potential exposure from soil ingestion would likely have been much higher.

Data obtained from the PUF Roller, designed to estimate dermal contact exposure of a 9 kg child to pesticide residues on floor surfaces, suggests that a child of that weight in the study home could potentially pick up 0.1 to 0.6 µg/m^2 diazinon or chlorpyrifos on dry skin. Saliva-moistened skin has been shown to be 2 to 8 times more efficient than dry skin at dislodging freshly-applied pesticide residues from carpet surfaces.[17] Therefore, a small child with wet, sticky hands on the family room floor on Day 1 could pick up as much as 4 µg of pesticide residue per m^2 of carpet contacted. Recent studies have demonstrated that human saliva removes 40-60% of freshly-applied chlorpyrifos and other pesticides from human skin.[17] Assuming the residues found on the children's hands were similarly removable and rapidly replenished, the 2-year-old girl could have ingested 0.02 to 0.09 µg of diazinon per mouthing event during the week following the summer application, should she have placed her whole hand in her mouth. Following the same scenario, the 4-year-old boy would have ingested 0.02 to 0.4 µg per mouthing event. Since children under two engage in frequent mouthing activities, it is conceivable that a such a child in this household could have ingested up to several micrograms per waking hour in such a manner.

In summary, the combination of inhalation, dermal contact and non-dietary oral exposure to diazinon in House 2 on Day 1 after Application 1 may have resulted in a total potential exposure of 10 to 15 µg, or 0.7 to 1.1 µg/kg for the 2-year-old girl (13.5 kg) and 0.6 to 0.8 µg/kg for the 4-year-old boy (18.2 kg). Exposure rates averaged over two weeks should have been 50% or less of the Day 1 rate. Potential exposures to diazinon and chlorpyrifos following Application 2 appeared to be significantly less. These estimates are far lower than the exposure estimates of Gurunathan et al.[25] for a 3- to 6-year-old child living in an apartment that received crack and

crevice treatment with chlorpyrifos (356 µg/kg/d peak; 208 µg/kg/d averaged over two weeks) and below the EPA reference dose of 3 µg/kg/d for chronic oral exposure (RfD) to chlorpyrifos.[26] No reference dose has been established for diazinon.

After normal pest control treatments, pesticides deposit onto or are transported to floors and other surfaces, to dinnerware and food stored in closed cabinets, to children's toys, and are picked up on children's hands. Whereas these residues dissipate rapidly from the treated surfaces after pesticide applications, they are usually detectable (sometimes at relatively high levels) for months after the application. Although accurate assessment of the human exposure risks associated with pesticide use in and around the home is still difficult, it is apparent that residents of treated domiciles may be exposed to pesticide residues in areas other than those treated. Small children, who are more likely to come into intimate contact with both intentionally and incidentally-contaminated surfaces, are generally at highest risk of exposure.

ACKNOWLEDGMENTS

The authors thank Dr. William D. Ellenson of ManTech Environmental Technology for technical support; and Michelle Ortiz, James Voos, and Tina Smeal of Southwest Research Institute for sample extraction and analysis. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

This work has been wholly or in part funded by the United States Environmental Protection Agency under contract 68-D5-0049 to ManTech Environmental Technology, Inc. It has been subjected to Agency review and approved for publication.

REFERENCES

1. Leidy, R.B.; Wright, C.G.; Dupree Jr., H.E. In *Pesticides in Urban Environments*; Racke, K.D.; Leslie, A.R., Eds.; ACS Symposium Series 522; American Chemical Society: Washington, D.C., 1993; pp. 283-296.

2. Lewis, R.G.; Fortmann, R.C.; Camann, D.E. *Arch. Environ. Contam. Toxicol.* **1994**, *26*, 37-46.

3. Koehler, P.G.; Moye, H.A. *J. Econ. Entomology* **1995**, *88*,1684-1689.

4. Lu, C.; Fenske, R.A. *Environ. Sci. Technol.* **1998**, *32*, 1386-1390.

5. Yeary, R.A.; Leonard, J.A. In *Pesticides in Urban Environments*; Racke, K.D.; Leslie, A.R., Eds.; ACS Symposium Series 522; American Chemical Society: Washington, D.C., 1993; pp. 275-281.

6. Nishioka, N.G.; Burkholder, H.M.; Brinkman, M.C.; Lewis, R.G. *Environ. Sci. Technol.* **1999**, *30*,1359-1365.

7. Lewis, R.G.; Nishioka, M.G. In *Indoor Air '99: Proceedings of the 8th International Conference on Indoor Air Quality & Climate*; Edinburgh, Scotland, 1999; Paper No. 436.

8. Fenske, R.A.; Black, K.G.; Elkner, K.P.;Lee, C-L.; Methner, M.M.; Soto. R. *Am. J. Public Hlth.* **1990**, *80*, 689-693.

9. Simcox, N.J.; Fenske, R.A.; Wolz, S.A.; Lee, I-C.; Kalman, D.A. *Environ. Health Perspec.* **1995**,*103*, 1126-1134.

10. *Compendium of Methods for the Determination of Toxic Organic Chemicals in Ambient Air*; U. S. Environmental Protection Agency. Office of Research and Development: Cincinnati, Ohio. 1996; EPA-600/R-96/010b (http://www.epa.gov/ttn/amtic/airtox.html).

11. *Annual Book of ASTM Standards*, Vol. 11.03; American Society for Testing and Materials: West Conshohoken, Penn. 1999.

12. Lewis, R. G. In *Indoor Air Quality Handbook*; Spengler, J.D.; Samet, J.M.; McCarthy, J.F., Eds.; McGraw-Hill: New York, N.Y., 2000 (in press).

13. *Transport of Lawn-Applied 2,4-D from Turf to Home: Assessing the Relative Importance of Transport Mechanisms and Exposure Pathways*; US Environmental Protection Agency: Research Triangle Park, N.C. 1999; EPA-600/R-99/040.

14. Geno, P.W.; Camann, D.E.; Harding, H.J.; Villalobos, K.; Lewis, R.G. *Arch. Environ. Contam. Toxicol.* **1996**, *30*, 132-138.

15. Hsu, J.P.; Wheeler, Jr., H.G.; Schattenberg, III, H.J.; Camann, D.E.; Lewis, R.G.; Bond, A.E. *J. Chromatogr. Sci.* **1988**, *26*, 181-189.

16. Lewis, R.G.; Fortune, C.R.; Willis, R.D.; Camann, D.E.; Antley, J.T. *Environ. Health Perspect.* **1999**, *107*, 721-726.

17. Lewis, R.G. In *Occupational and Incidental Residential Exposure Assessment*; Worgan, J.P.; Franklin, C.A., Eds.; Wiley & Sons, Ltd.: Sussex, U.K., 2000 (in press).

18. Whitmore, R.W., Immerman, F.W., Camann, D.E., Bond, A.E., Lewis, R.G., Schaum, J.L. *Arch. Environ. Contam. Toxicol.* **1994**, *26*, 47-59.

19. Camann D.E., Buckley J.D. In *Abstracts of the Joint Conference of the International Society of Environmental Epidemilogy/International Society of Exposure Analysis*; Research Triangle Park, NC, 1994; Abstract 141.

20. *Exposure Factors Handbook*; U. S. Environmental Protection Agency, Office of Research and Development: Washington, DC: 1997; EPA-600/P-95/002F.

21. *Transport of Lawn-Applied 2,4-D from Turf to Home: Assessing the Relative Importance of Transport Mechanisms and Exposure Pathways*; U.S. Environmental Protection Agency, National Exposure Research Laboratory: Research Triangle Park, NC: 1999 EPA-600/R-99/040.

22. Wright, C.G., Leidy, R.B., Dupree H.E. *Bull. Environ. Contam. Toxicol.* **1984**, *32*, 259-264.

23. Gordon, S.D., Callahan, P.J., Nishioka, M.G., Brinkman, M., O'Rourke, M.K., Lebowitz, M.D., Moschandreas, D. *J. Exp. Anal. Environ. Epidem.* **1999**, *9*, 456-470.

24. Camann, D.E., Clothier, J.M., Kuchibhatla, R.V., Bond, A.E. In *Measurement of Toxic and Related Air Pollutants: Proceedings of the EPA/A&WMA International Symposium*: Air & Waste Management Association: Pittsburgh, PA: 1995; Publication VIP-50, pp. 548-554.

25. Gurunathan, S., Robson, M., Freeman, N., Buckley, B., Roy, A., Meyer, R., Bukowski, J., Lioy, P.J. *Environ. Health Perspect.* **1998**, *106*, 9-16.

26. US EPA. *Integrated Risk Information System*; U. S. Environmental Protection Agency: Washington, DC: 1999 (http://www.epa.gov/iris/).

Key Words

Pesticide, residential, exposure, multi-media, children, diazinon, chlorpyrifos

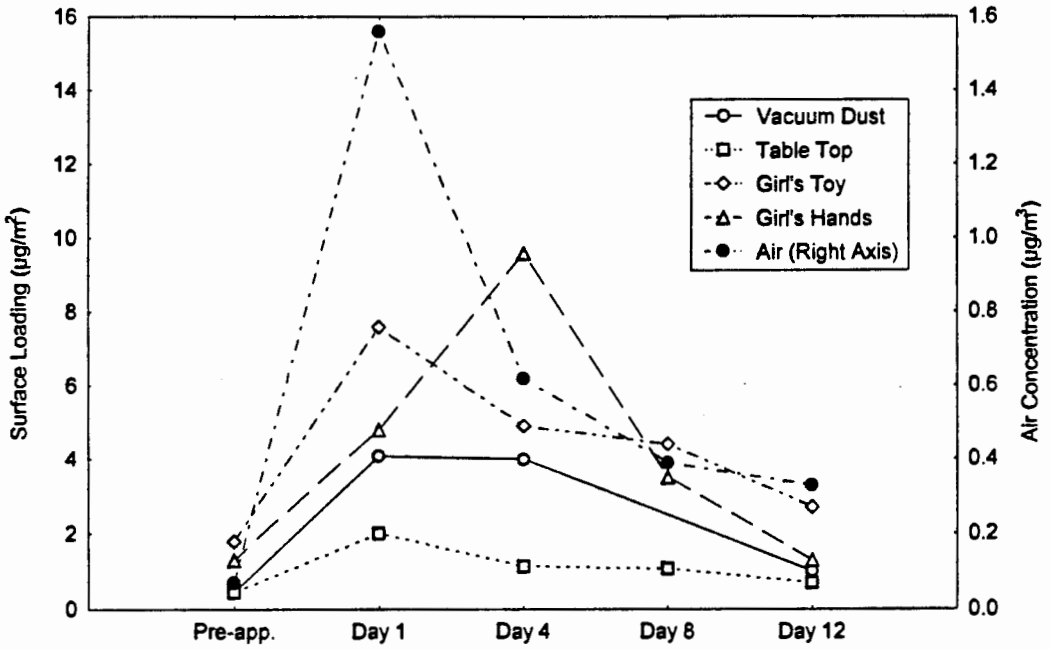
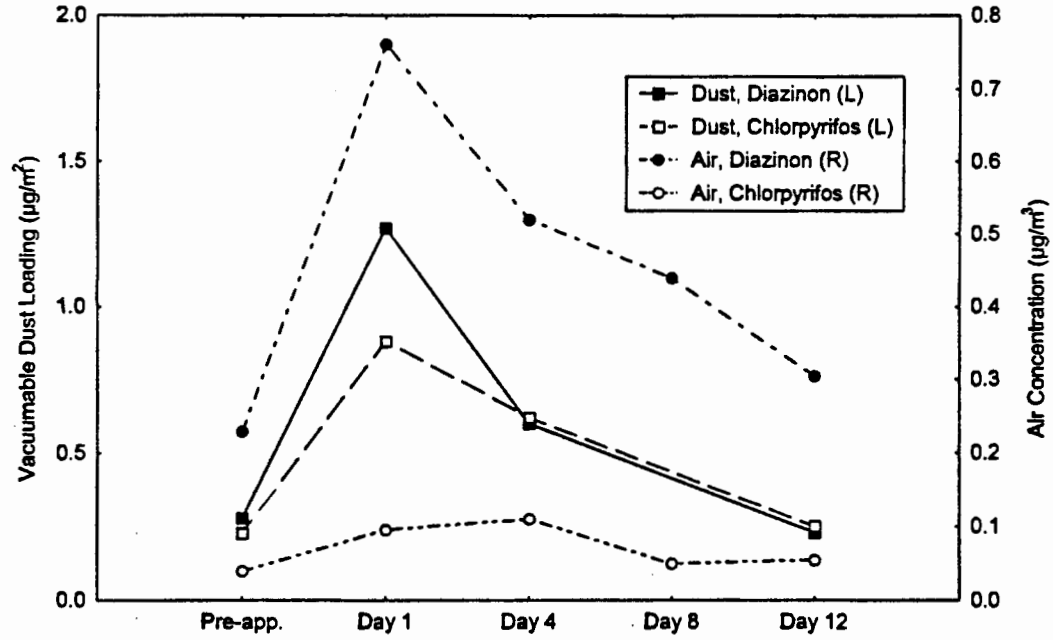


Figure 1. Diazinon levels in Home 2 and before and after the Application 1 During August 1999. Note: Preapplication values for table top, toy, and hand residues were all below the quantitation limits.



Figur **e 2.**
Air and carpet dust residues of diazinon and chlorpyrifos in the family room of Home 2 before and after Application 2 during September - October 1999.

Table 1. Diazinon levels, Home 2, first application, August 3-17, 1999.

Sample Type	Room[a]	Pre-app.	Day 1	Day 4	Day 8	Day 12
Air, $\mu g/m^3$, 75-cm	Family	0.07	1.56	0.62	0.39	0.33
10-cm		0.08	1.49	0.51	0.39	0.29
Air, $\mu g/m^3$, 75-cm	Bedroom	0.03	0.42	0.11	0.12	0.13
10-cm		0.03	0.36	0.12	0.16	–
Carpet dust, $\mu g/m^2$	Family	0.46	4.08	4.00	–	1.01
$\mu g/g$		0.12	1.20	1.42	–	0.68
PUF roller, $\mu g/m^2$	Family	0.03	0.61	0.37	–	0.16
PUF roller, $\mu g/m^2$	Bedroom	–	0.22	–	–	0.12
Floor wipe, $\mu g/m^2$	Bedroom	2.25	4.22	9.44	8.01	5.35
Table top, $\mu g/m^2$	Family	<0.45	2.01	1.13	1.08	0.71
Table top, $\mu g/m^2$	Bedroom	<0.45	0.67	0.73	<0.45	<0.45
Dinner plate, μg	Kitchen	<0.04	0.07	<0.04	<0.04	<0.04
$\mu g/m^2$		<1.23	1.99	<1.23	<1.23	<1.23
Food surrogate, μg	Kitchen	0.44	0.60	0.44	0.44	0.40
Girl's toy, μg	NA	<0.04	0.17	0.11	0.10	0.06
$\mu g/m^2$		<1.8	~7.6	~4.9	~4.4	~2.7
Girl's hands, μg	NA	<0.04	0.15	0.30	0.11	<0.04
(Age 2) $\mu g/m^2$		<1.3	~4.8	~9.6	~3.5	<1.3
Boy's hands, μg	NA	<0.04	1.29	1.46	0.11	<0.04
(Age 4) $\mu g/m^2$		<1.0	~34	~38	~2.8	<1.0

[a] Family = main living area on ground level (treated); Bedroom = 4-year-old male child's room on upper level; Kitchen = Food storage and preparation room on ground level (treated)

NA = not applicable

– = Not sampled

Table 2. Diazinon levels, Home 2, second application, September 15-October 5, 1999.

Sample Type	Room[a]	Pre-app.	Day 1	Day 4	Day 8	Day 12
Air, µg/m³, 75-cm	Family	0.23	0.84	0.42	0.48	0.30
10-cm		0.23	0.68	0.62	0.40	0.31
Air, µg/m³, 75-cm	Bedroom	0.14	0.13	0.14	0.20	0.12
10-cm		0.14	0.15	0.13	0.14	0.11
Carpet dust, µg/m²	Family	0.28	1.27	0.60	–	0.23
µg/g		0.43	1.05	0.79	–	0.91
PUF roller, µg/m²	Family	<0.18	0.35	<0.18	<0.18	<0.18
PUF roller, µg/m²	Bedroom	<0.18	<0.18	–	–	<0.18
Floor wipe, µg/m²	Bedroom	11.0	4.41	~21*	5.70	8.72
Floor wipe, µg/m²	Kitchen	56.3 (20)	76.1 (60)	262 (150)	–	–
Table top, µg/m²	Family	0.84	~1.8*	1.01	0.68	≤0.67
Table top, µg/m²	Bedroom	≤0.24	0.90	≤0.28	≤0.26	≤0.32
Dinner plate, µg	Kitchen	0.01	0.04	≤0.02	0.04	≤0.02
µg/m²		0.31	0.81	≤0.60	0.81	≤0.60
Food surrogate, µg	Kitchen	0.42	0.36	0.43	0.26	0.19
Foundation soil, µg/g	NA	2.33	1.10	0.38	0.30	0.31
µg/m²		25.1	11.8	4.09	3.23	3.34
Girl's toy, µg	NA	≤0.03	0.05	0.05	≤0.04	≤0.05
µg/m²		≤1.3	~2.2	~2.2	≤1.8	≤2.2
Girl's hands, µg	NA	~0.1*	0.06	0.06	0.07	<0.04
(Age 2) µg/m²			~1.9	~1.8	~2.4	<1.3
Boy's hands, µg	NA	0.09	–	~0.1*	1.06	<0.1*
(Age 4) µg/m²		~0.2			~27	

[a] Family = main living area room on ground level (treated); Bedroom = 4-year-old male child's bedroom on upper level; Kitchen = food storage and preparation room on ground level (treated)

NA = not applicable; – = Not sampled; *Analytical result adjusted for sample leakage; ( ) Numbers in parentheses for collocated samples analyzed by a second laboratory

Table 3. Chlorpyrifos levels, Home 2, second treatment, September 15-October 5, 1999.

Sample Type	Room[a]	Pre-app.	Day 1	Day 4	Day 8	Day 12
Air, μg/m³, 75-cm	Family	0.04	0.10	0.10	0.06	0.05
10-cm		0.04	0.09	0.12	0.04	0.06
Air, μg/m³, 75-cm	Bedroom	0.05	0.07	0.07	0.05	0.05
10-cm		0.06	0.08	0.06	0.05	0.05
Carpet dust, μg/m²	Family	0.23	0.88	0.62	–	0.25
μg/g		0.35	0.72	0.81	–	1.01
PUF roller, μg/m²	Family	≤0.21	0.23	<0.18	–	≤0.21
PUF roller, μg/m²	Bedroom	≤0.26	0.41	–	–	<0.18
Floor wipe, μg/m²	Bedroom	12.2	7.96	~27*	11.8	22.4
Floor wipe, μg/m²	Kitchen	11.7 (10)	19.5 (30)	17.1 (10)	–	–
Table top, μg/m²	Family	16.6	~9*	3.34	12.2	3.01
Table top, μg/m²	Bedroom	3.44	15.4	2.48	3.34	12.1
Dinner plate, μg	Kitchen	0.10	0.21	0.06	0.29	0.04
μg/m²		3.10	6.70	1.70	8.53	1.25
Food surrogate, μg	Kitchen	0.02	0.03	0.04	0.01	0.03
Foundation soil, μg/g	NA	1.91	18.6	2.87	1.83	3.42
μg/m²		20.6	200	30.9	19.7	36.8
Child's toy, μg	NA	0.18	0.21	0.97	0.10	1.04
μg/m²		~ 8.1	~ 9.5	~ 43	~ 4.5	~ 46
Girl's hands, μg	NA	~ 0.4*	1.27	0.18	0.24	0.13
(Age 2) μg/m²		~ 14	~ 48	~ 5.8	~ 7.7	~ 4.2
Boy's hands, μg	NA	1.91	–	~0.2*	0.21	~0.8*
(Age 4) μg/m²		~50			~5.4	

[a] Family = main living area room on ground level (treated); Bedroom = 4-year-old male child's bedroom on upper level; Kitchen = food storage and preparation room on ground level (treated)

NA = not applicable; – = Not sampled; *Analytical result adjusted for sample leakage; ( ) Numbers in parentheses for collocated samples analyzed by a second laboratory

NERL-RTP-HEASD-00-134

TECHNICAL REPORT DATA

1. REPORT NO.	2.	
EPA/600/A-00/035		

4. TITLE AND SUBTITLE	5.REPORT DATE
Movement and Deposition of Pesticides within Residences after Interior and Exterior Applications	6.PERFORMING ORGANIZATION CODE

7. AUTHOR(S)	8.PERFORMING ORGANIZATION REPORT NO.
R. G. Lewis, C. R. Fortune, F. T. Blanchard, and D. E. Camann	

9. PERFORMING ORGANIZATION NAME AND ADDRESS	10.PROGRAM ELEMENT NO.
U. S. EPA/ORD/NERL, Research Triangle Park, NC 27711 ManTech Environmental Technology, RTP, NC 27709 Southwest Research Institute, San Antonio, TX 78228	80201F
	11. CONTRACT/GRANT NO. 68-D5-0049

12. SPONSORING AGENCY NAME AND ADDRESS	13.TYPE OF REPORT AND PERIOD COVERED
National Exposure Research Laboratory U. S. Environmental Protection Agency Research Triangle Park, NC 27711	Conference Paper
	14. SPONSORING AGENCY CODE EPA/600/09

15. SUPPLEMENTARY NOTES

16. ABSTRACT

In a study begun in 1999, the Environmental Protection Agency (EPA) investigated the temporal and spatial distributions of pesticides applied by homeowners and commercial applicators for indoor crack and crevice and exterior perimeter treatments. In each participating household, pesticide levels in indoor air at 10-cm and 75-cm above the floor (child's breathing zone) were measured pre- and post-application (0-14 days), along with dermal- and vacuum-dislodgeable floor residues, deposition on table tops and dinnerware, absorption by surrogate food, and residues on children's hands and toys. Surface soil residues were measured in the case of perimeter application. Monitoring devices and methods used include the URG 2500 air samplers (PM 2.5 inlet), the HVS3 vacuum sampler, the PUF Roller, hand wipes, and various techniques for deposition monitoring. Results from the study demonstrate the nature and magnitude of translocation of pesticides from the areas of application to surfaces accessible for human contact. This paper presents data from two indoor applications of diazinon and one outdoor (perimeter) application of chlorpyrifos. The post-application redistribution of the pesticides within the home and the exposure potentials via various routes (respiration, dermal contact, ingestion) are discussed.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b.IDENTIFIERS/ OPEN ENDED TERMS	c.COSATI

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS *(This Report)*	21.NO. OF PAGES
	20. SECURITY CLASS *(This Page)*	22. PRICE