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## **Movement of Diazinon Residues into Homes Following Applications of a Granular Formulation to Residential Lawns**

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### **ABSTRACT**

A pilot study was conducted to examine the movement of diazinon following applications of a granular formulation to residential lawns. The objectives included examining the transport and fate of diazinon from an outdoor source to the indoor living areas of six homes, and estimating potential human exposure of six children (age 5-12) living in these homes using site specific data and model default assumptions. Sampling included the collection of the pesticide formulation, soil, particles from doormats, transferable residues from residential turf and indoor flooring, indoor air from living rooms and children's bedrooms, and vacuum dislodgeable dust. Samples were collected from six single family homes located in the Piedmont region of North Carolina between April and August 2001. Environmental samples were collected prior to pesticide application and at days 1, 2, 4, and 8 following the application. Soil concentrations, an indicator of source strength, were highest immediately following the application and declined by an average of 51% by day 8. Transferable residues from turf were determined with the polyurethane foam (PUF) roller and ranged from 0.1 to 970 ng/cm<sup>2</sup> over the study period. Particle-associated residues collected from doormats located at entryways into the home declined from day 2 to day 8 by an average of 75%. Indoor air concentrations in both living rooms and children's bedrooms reached maximal levels from 1 to 2 days following pesticide application and declined over the remainder of the study.

Indoor transferable residue levels from carpeted surfaces were typically below the limit of detection and are reflective of a low efficiency collection technique. Concentrations in vacuum dislodgeable dust were variable over time, but consistently exceeded pre-application concentrations. Results demonstrate that the physical translocation of particle-bound residues and the intrusion of volatilized diazinon contribute to indoor levels. Increased airborne concentrations demonstrate the intrusion of diazinon from the outdoor source. Elevated concentrations in dust suggest the movement and deposition of volatilized and/or particle-bound residues. Model estimates suggest that exposure occurred over the duration of the study and that the estimated absorbed mass declined little over 8 days. In summary, the applications to residential lawns resulted in a sustained increase of diazinon levels above background concentrations inside of all homes. Lawn applications were found to be a source of potential occupant exposure both on treated lawns and inside homes.

## Introduction

Insecticides are commonly applied to residential lawns, school grounds, parks, golf courses, and athletic fields to control for soil borne insects that can damage turf grass and to control terrestrial insect pests such as ants, ticks, fleas and crickets. In the United States (U.S.) a variety of insecticide formulations, spanning multiple chemical classes, may be conveniently purchased by the general public to control insects, fungi and weeds on their personal lawns. Estimates derived from the home and garden market survey conducted in 2000 to 2001 indicate that about 78 million U.S. households used pesticides, spending nearly 1.3 billion dollars to purchase insecticides and applying 888 million pounds of active ingredient (Kiely *et al.*, 2004). The US EPA (1985) reported that about 4.5 million kg of diazinon (O, O-diethyl O-2-isopropyl-6-methylpyrimidin-4-yl phosphorothioate) was used in 1981 on residential lawns and turf farms. In 2001, when this study was performed, the same organophosphate insecticide was ranked as the fourth most commonly used pesticide. Due to emerging health and ecological risks, manufacturers of diazinon agreed to a phase out and cancel all residential use products. Effective as of December 31, 2004 no diazinon products with residential uses were to be registered or sold (US EPA, 2004). Registrations allowing for use of these compounds in and around homes and buildings have been removed and the products are no longer sold to the general public.

Studies have shown that pesticides applied in residential dwellings move from their point of application and contribute to increased indoor concentrations (Wright *et al.*, 1978; Leidy *et al.*, 1993; Gurunathan *et al.*, 1998; Lewis *et al.* 2001; Hore, 2003; Stout and Mason, 2003; Lewis, 2005). The movement or translocation of pesticide residues is dependent on the formulation, the physio-chemical properties of the active ingredient(s), the surfaces onto which the applications are made and human activity patterns around the point of application. Pesticide residues are degraded by factors such as heat, hydrolysis, and microbial activity typically found out of doors. Indoors, where residues are protected from these degradation factors, residues may persist, accumulate (Lewis *et al.*, 1994, Whitmore *et al.*, 1994) and serve as a continued source of exposure to the occupants. Lewis and MacLeod (1982) determined that indoor air concentrations of pesticides may be 10 to 100 times higher than those measured out of doors.

Although less studied, applications to residential lawns and foundations of homes have been shown to result in the movement of pesticide residues away from the point of application and into homes. Drift and subsequent volatilization can contaminate outdoor surfaces and contribute to indoor air and surface concentrations. A professional application of microencapsulated cyfluthrin to the exterior perimeter of homes resulted in the deposition of residues up to 9.1 m from the homes foundations and low level contamination on some indoor surfaces (Stout and Leidy, 2000). Lewis *et al.* (2001) investigated indoor residues insecticide concentrations over 12 days following the application of an emulsifiable concentrate formulation of chlorpyrifos to the exterior perimeter and foundation of a home. They found that the pesticides applied outdoors were transported indoors and deposited onto surfaces. Research conducted by Nishioka *et al.* (1996, 1997, 2001, 2002) substantiated "track-in" as a pathway for the transport of pesticide residues into homes. They showed that foot traffic transported residues from turf treated with the herbicides 2,4-D and dicamba onto carpeting up to 1 week following an application. In a later study, the movement of 2,4-D, chlorpyrifos and dicamba applied as a spray and granular formulation, respectively, to turf was shown to result in increased indoor concentrations. Their findings suggested that rain events and volatilization were important dissipation pathways for these compounds and that "track-in" occurred over the six days following the application. In a third study, 2,4-D was applied to the turf of eleven occupied and two unoccupied homes. Indoor air concentrations and surface loadings were primarily attributed to the re-suspension of residue laden floor dust and subsequent deposition onto surfaces. Results showed that indoor pesticide residues were elevated in the homes with higher occupant activity levels and indoor/outdoor pet dogs, and suggested the residues measured indoors were likely associated with particles tracked-in by the occupants who performed the applications and through the activities of pet dogs. Morgan *et al.* (2001) conducted a one home feasibility study that examined the



role of a pet dog as a vehicle for transporting diazinon residues following a lawn application. Based on questionnaires and recall diaries it was determined that the dog spent more time on the treated turf relative to the occupants and it was hypothesized that the pet was an important mechanism for the transport of residues into the indoor living area of the home.

In the study presented here, we measured diazinon concentrations from various media following an application of a granular formulation of diazinon to residential turf. It expands on the previously mentioned study (Morgan *et al.*, 2001) examining the role of pet dogs as a mechanism for the movement pesticides. The objective of this study was to examine the transport pathway of a semi-volatile insecticide following a granular application to residential turf in six homes, generate input data for human exposure models and to estimate potential human exposure of six children (age 5-12) living in these homes using site-specific data and model default assumptions.

## **Materials and Methods**

From April through August of 2001 six residential homes located in the Piedmont region of North Carolina were monitored following granular applications of diazinon to their residential lawns. Single family households, having one or more adults and children (< 14 years of age) and one indoor-outdoor adult dog and who had previously planned to perform this type application were solicited for participation in the study. Occupants purchased their own commercially-available granular diazinon at local stores and applied the insecticide to their lawns using manually operated rotary spreaders, except for participant 6 who hand broadcast the granular formulation. The applicators were encouraged to read and follow directions as provided on the product labels.

This was a "human observational exposure" study, as defined in 40 CFR Part 26.402. The study protocol and procedures to obtain the assent of the children and informed consent of their parents or guardians were reviewed and approved by an independent institutional review board (IRB) and complied with all applicable requirements of the Common Rule regarding additional protections for children. In addition, the study protocol and procedures were reviewed and approved by an independent Animal Care and Use Committee (ACUC).

Family members recorded in recall diaries their general daily activities. Participant activities were recorded one week prior to the implementation of field sampling and continued on each sampling day throughout the study.

Prior to the application the participant's yards and indoor living areas were measured and graphed on paper. In addition, diagrams of the area treated for each home were prepared prior to the applications to determine the theoretical application rate and actual square meters treated.

Sampling was carried out prior to the application and at 1, 2, 4, and 8 days post-application. Sample types included an aliquot of formulated material collected from the hopper of the spreader or the bag directly, soil cores, turf transferable residues, doormat sweepings at a common occupant/pet entryway, indoor air, vacuum dislodgeable particles (carpet dust) and indoor surface transferable residues from carpeting. Pre- and post-application weights of the formulation were recorded to obtain the total mass applied to the yard.

The aliquots of the granular formulation were placed into 250-mL glass jars using a spatula and nitrile gloved hands. Jars were enclosed in heavy plastic bags to minimize cross contamination with other samples.

Thirty soil samples were collected from thirty different locations within the treated area and composited. The treated area was diagrammed and marked with two bisecting lines that extended to the furthest edges of the treated area. A total of thirty flags designating sample locations were distributed along the two lines. A soil core was collected from within a 30.5 cm radius of each flag at each sampling interval (day). Samples were collected using a T-handle soil probe with a 2.5 cm diameter core. The probe was inserted into the ground to an approximate depth of 1 cm. The soil cores from each home for each sampling interval (day) were aggregated into a 250-mL glass jar capped with a Teflon lined lid.

A single turf transferable residue sample was collected per sampling interval (day) using a polyurethane foam (PUF) roller apparatus and a dry PUF sampling ring (Nishioka *et al.*, 1997; Morgan *et al.*, 2001; ASTM, 2004). The 8-cm wide PUF roller was rolled on the turf at a rate of 10 cm/s over a 2 m distance. Each location sampled was marked on a diagram of the house and lawn and was not re-sampled at any subsequent sampling intervals. Sample locations were within the confines of the area treated by the occupant.

A single doormat sample was collected at days 2, 4, and 8 post-application. Entryway deposits were collected by placing a new, solid black rubber doormat (43 cm X 64 cm) at the door most commonly used to enter and exit the home by the occupants and pets. The entire doormat was vacuumed with a modified hand vacuum fitted with an in-line filter that was hand fabricated from vacuum cleaner bags. The doormat was removed at each sampling interval and replaced by a new unexposed doormat. Vacuuming was accomplished by pulling the intake nozzle over the top doormat surface in a zig-zag pattern across the length of the mat followed by a similar pattern conducted vertically over the mat. Following the systematic vacuuming of the entire top surface of the doormat, the in-line filter was removed and the top folded over to enclose the contents and closed shut with a metal clip. Using gloved hands the filter and its contents were placed into a 250-mL glass jar.

Indoor air was sampled in the room where the participants spent the most time (typically the living room) and in a child's bedroom. Air was sampled from the two locations prior to the application and at 1, 2, 4 and 8 days post

application. Samples were collected using low-flow pumps placed in the center of each room (ASTM 2007). Polyurethane plugs (PUF) enclosed in glass housings were connected to the pumps via PTFE tubing. The PUF plugs were open faced with no particle filtration. The sampling heads were suspended 75-cm from the floor with the inlet positioned downwards. Flow rates were set at 3.8 L/min and each sample was collected over 24-h resulting in a sample volume of 5.5 m<sup>3</sup>. Following the completion of sampling the glass housings containing the PUF sampling, media were capped on both ends and placed into glass jars.

A high volume small surface sampler (HVS3; ASTM, 2006) was used to collect vacuum-dislodgeable dust from 1 m<sup>2</sup> areas from locations in the carpeted areas of living rooms or dens on days. A single location was selected and sampled prior to the application and on days 2, 4, and 8 post application. To determine transferable residues indoors, PUF roller samples were collected from carpeted surfaces in the living room. Locations sampled were not re-sampled on any later days.

In general all samples were placed in appropriately sized pre-cleaned glass jars with PTFE lined lids. The lids were checked for tightness prior to placing into ice chests for storage at reduced temperatures and under darkened conditions for transport to the laboratory. The chain of custody was established and samples were archived at -20 °C until shipped for chemical analysis. Quality assurance consisted of field duplicates, field spikes, and field blanks, and selected results are shown in Table 1.

The samples collected in this study were generally extracted in an automated accelerated solvent extraction apparatus (Dionex ASE 200™) in 100% hexane (analytical grade) at 100 °C and 2000 psi. The extracts were collected in 60 mL glass tubes and concentrated to a volume of 2 mL in a Turbo Vap concentrator set at 42 °C and 20 psi. Sample cleanup was performed using solid phase extraction tubes (0.5 g Florisil, 3 mL capacity, Restek REPREP™). Tubes were conditioned by flushing with 90:10 hexane:acetone. Samples were eluted with 40 mL of 90:10 hexane:acetone. The eluant was reduced to 1 mL and transferred to a 2 mL vial until chemical analysis. Samples were chemically analyzed using gas chromatography and a mass spectrometry (GC-MS). The GC oven was programmed to ramp from 100 °C to 150 °C at a rate of 5 °C/min and held for 2 min. The MS was operated at 70 eV using electron impact ionization in selective ion monitoring mode. Calibrations standards ranged from 50 to 200 ng/mL. Laboratory controls included matrix blanks, spikes and spike duplicates and the use of surrogate standards. The quality assurance criterion for spiked matrices and surrogate recoveries was between 40 and 120% and the data were surrogate corrected. All statistical analyses were performed using fixed effects and mixed-effects models (SAS Proc Reg and Proc Mixed procedures) available with SAS version 8.2 (SAS Institute Incorporated, Cary NC). All measurements were log transformed prior to analysis.



The US EPA/ORD/NERL Stochastic Human Exposure and Dose Simulation model for multimedia, multi-pathway pollutants (SHEDS-multimedia version 3.0) (Glen, 2007; Zartarian et al., 2007) was used to estimate the exposures through the inhalation, non-dietary ingestion, and dermal routes and subsequent intake for children in this study. Briefly, SHEDS simulates individuals from user specified population cohorts by selecting daily sequential time-location-activity diaries from surveys contained in the US EPA's Consolidated Human Activity Database (CHAD; McCurdy *et al.* 2000) relevant to the specific demographic (age, gender, etc.) characteristics of the participants. Each simulated individual is randomly assigned an appropriate activity diary according to demographic characteristics, and the values for each model input parameter (Table 2) are randomly sampled from distributions and inserted into pathway algorithms. An individual's time series of exposure and absorbed mass by pathway is estimated, and the metric of interest (e.g. time-averaged exposure or absorbed dose) is computed for the individual. The process is repeated thousands of times using Monte Carlo simulation to produce a population distribution of exposure or intake (Zartarian *et al.*, 2000; Xue *et al.*, 2006; Zartarian *et al.*, 2006). The SHEDS model has the capability to employ pesticide levels measured in exposure media for individuals within a given scenario (Hore, 2003). The model couples activity data with environmental data using physically based equations and calculates resulting exposures and doses.

This modeling application was used to apportion the per cent contribution of each non-dietary pathway to exposure and absorbed mass for each child participant. The daily inhalation time series absorption profile for each individual was estimated as a product of the actual airborne concentrations ( $\mu\text{g}/\text{m}^3$ ), a basal breathing rate ( $\text{m}^3$  air/day), an activity specific ventilation rate ratio, and the duration of the macro-activity event (day/event). The dermal exposure time series dose profile was calculated as a product of the measured surface concentration (soil and turf transferable outdoors and carpet dust indoors;  $\mu\text{g}/\text{cm}^2$ ), and the dermal transfer coefficients for a given macro-activity ( $\text{cm}^2/\text{h}$ ). In addition to mass loading and removal mechanisms such as hand washing, bathing and hand-to-mouth transfers were also considered by the model. The estimated non-dietary ingestion absorption profile included exposure from hand- and object-to-mouth activities. The dermal exposure for the subjects hands ( $\mu\text{g}$ ) were halved to represent the mass loading to one hand. The mass was adjusted for the fraction of residue on the hand that contacts the mouth per mouthing event and the saliva removal efficiency. The daily absorbed dose ( $\mu\text{g}/\text{kg}/\text{day}$ ) was calculated with a simple pharmacokinetic model in SHEDS using appropriate absorption factors for diazinon. Relevant parameters utilized in the model and their estimates are given in Table 2. The participant's physical attributes used in the SHEDS model are provided in Table 3. The contribution from dietary ingestion was disregarded as part of this modeling exercise.

It is important to note that the assumptions shown in Table 2 are based on literature derived inputs and other defined sources. The reader is cautioned that SHEDS model inputs can be changed and those changes might significantly alter the relative importance of the calculated exposure pathways. In addition, the authors assumed that all diazinon residues were equally available for transfer and uptake whether bound to particles or not.

## **Results and Discussion**

### **Mass Applied and Area Treated**

The homeowners applied the granular diazinon to lawn areas ranging from 139 m<sup>2</sup> (house 3) to 1300 m<sup>2</sup> (house 5) with a mean treated area of 669 m<sup>2</sup> (Table 4). The total amount of granular formulation applied ranged between 2.0 kg (house 1) to 12.7 kg (house 4) with a mean amount of 6.0 kg. The application rates were determined by dividing the mass of formulated material applied by the area treated. The theoretical application rates were based on a labeled rate of 2 lbs/1000 ft<sup>2</sup> or 0.5 g/m<sup>2</sup>. Four out of six homes did not exceed the manufacturer's recommended rates and in fact tended to perform applications at rates below those described on the product label. Two homes (3 and 4) did exceed the recommended label rates, by 2 and 6.5 times, respectively.

### **Concentrations Measured from Soil Cores**

Some homes exhibited background concentrations of diazinon in soil prior to the application (Table 5). Following the granular applications the concentrations increased sharply above background, peaked by day 2 for the majority of homes and declined at rates ranging from 25 to 96% by day eight. Despite the temporal decline the measured concentrations remained high eight days following the application relative to the measured background concentrations. The soil measurements identify a source of diazinon from which residues might emanate. These measurements are in line with the reported half life of diazinon in soil between 7 to 52 days depending on environmental conditions such as soil type, temperature and moisture content (Kuhr *et al.*, 1978; Michel *et al.*, 1997). The 1-cm deep soil cores taken in this study provide a reasonable estimate of the of the total mass present in the soil column, as Kuhr and Tashiro (1978) reported that very little diazinon could be measured deeper than 1.3 cm below the soil surface.

As expected, the total mass applied influenced the measured soil concentrations. A significant association was shown between the (log



transformed) post-application soil concentrations and the (log transformed) mass applied per unit area (ANOVA,  $r^2 = 0.43$ ,  $p = 0.0003$ ). The size of the treated area itself was also a strong indicator of soil concentrations, with the larger the areas associated with lower soil concentrations ( $p = 0.0006$ ). Factors such as the applicator's technique and the spatial distribution of the granules within a confined area might be important elements along with housing factors in future observational studies of potential human exposure.

### **Turf Transferable Residues**

Turf transferable residues concentrations are shown in Table 6. With the exception of one house, diazinon levels prior to applications were typically below the detection limit ( $0.003 \text{ ng/cm}^2$ ). Applications at all homes resulted in an increase in measurable diazinon transferred from turf to the PUF roller. The highest turf transferable residues were typically measured between 1 to 2 days post application and then declined over the remainder of the study. However, in most cases, transferable residues remained up to two orders of magnitude higher than pre-application levels through 8 days. Transferable residue levels declined more rapidly than soil concentrations (Figure 1). The relatively low transferable residue levels observed agree with previous work by Sears (1987) who found that despite the abundant mass of diazinon residue present in the thatch following an application to turf, only 10% could be dislodged using vigorous wiping techniques. Similarly, residues declined rapidly within the first day of application and slowly declined through 15 days post application.

### **Doormat Sweepings**

The doormat was deployed as a simple tool to demonstrate the movement of residues from the source to the principal route of ingress and egress from the residence by the occupants and their pet dogs (Table 7). While doormat levels generally declined between days 2 and 8, the levels measured in the doormat sweepings at one home were actually 32% higher at day 8 than those measured at day 2. This increase did not correspond to any increase in soil concentration or transferable residue levels. In this case, the doormat loading likely was a function of occupant and pet activity during that time. Although no apparent relationship was observed between outdoor soil concentrations and the measured doormat loadings, the levels measured on the doormat provided evidence that residues were deposited at or transferred to the entryway of every single home studied.

### **Indoor Air**

Three homes had measurable background levels of diazinon prior to the lawn application. As was the case for residues in other media, the indoor air concentrations typically declined after day 2. Maximal concentrations were measured at days 1 and 2. The airborne levels were statistically different between all homes ( $p < 0.0001$ ). A statistically significant difference was observed between the living room and bedroom ( $p < 0.05$ ), suggesting that the level of occupancy, by both pets and participants, may affect the concentration in a particular room within the home following an outdoor application (Figure 2). Indoor concentrations were significantly ( $p < 0.05$ ) and positively associated with indoor temperature. These findings suggest that seasonal temperature variations both indoor and outdoor might influence residential exposure to volatile or semivolatile insecticides.

### **Indoor Transferable Residues**

Indoor PUF roller measurements exhibited a low detection frequency (27%) and are not presented. All measurements at two of the homes were below the limit of detection ( $0.006 \text{ ng/cm}^2$ ). The highest value ( $1.4 \text{ ng/cm}^2$ ) was observed at the home with the overall highest soil and turf transferable values (house 3). The PUF roller has been shown to have a low collection (transfer) efficiency of less than 2% for both turf and indoor carpeting (Fortune, 1998a; Fortune, 1998b; Lewis 2005). The combination of low indoor surface loadings and low overall efficiency of the method makes the PUF roller a poor approach for estimating (indoor) transferable residues in an outdoor application scenario. Since the value is a common metric in residential exposure models the utility of the methods would benefit by enhanced collection efficiency.

### **Vacuum Dislodgeable Particles (Carpet Dust)**

The elevated background concentrations of diazinon residues in household carpet dust (Figure 1) may be attributable to previous applications, as the persistence of some organophosphate pesticides in the indoor environments is well documented (Gurunathan et al., 1998; Lewis et al., 1994; Lewis et al., 2006). The highest residue levels were typically measured at 2 to 4 days following the application. Applications to the turf contributed to the diazinon measured in the homes; however, the rate and magnitude of the increase relative to pre-application levels varied among homes. The variability may be associated with housing factors, the sampling location within rooms, or the occupants' movements or activities within, into, and around the home. Once the

contaminant enters the home, indoor dust becomes a sink for diazinon and subsequently serves as a potential source for human exposure.

### **Media Comparisons**

Comparisons among the concentration profiles in the different media suggest a movement of residues from the treated turf to the indoors. Figure 1 illustrates the residue levels from the treated soil, turf transferable (or PUF roller), indoor air and carpet dust measurements. All metrics exhibited an increase in measured concentration above background. Although soil concentration declined gradually over the eight day sampling period, the soil remained a continuous source of residues throughout the study. Transferable turf measurements showed a more rapid decrease beyond two days following the application. Diazinon, which is moderately water soluble, may be expected to migrate into the soil column to a depth of nearly 1.3 cm (Kuhr and Tashiro, 1978) as it is generally watered in following turf applications. Although the soil measurements are reflective of the source strength, the transferable residue levels suggest that the residues become less available for transfer beyond two days, perhaps due to leaching into the soil column. As discussed previously, residues measured from doormats suggest migration to the indoor/outdoor interface of the homes. Based on the vapor pressure of diazinon ( $2 \times 10^{-1}$  mPa at 25 °C [Tomlin, 2006]), volatilization from the application surface into the air, as well as subsequent infiltration of some fraction of the total volatilized mass is expected. In this study, concentrations of diazinon in indoor air increased rapidly (within 24-h following the application) and remained elevated over the 9-day study period. In contrast, carpet dust concentrations generally did not reach maximal concentrations until 2 to 4 days post application. Relative to air, carpet dust displayed a slower rate of intrusion that is probably dependent on tracked-in particles linked to the outdoor activities of the occupants. Additional analysis of the findings shows that doormat loadings were a highly significant predictor of indoor carpet dust ( $p < 0.0001$ ) suggesting a strong relationship between doormat particle deposition and indoor dust loadings in the rooms most commonly occupied by participants and their pet dogs. Soil concentrations, on the other hand, trended towards the prediction of indoor dust loadings but the relationship was not statistically significant.

### **Exposure and Absorbed Mass Estimates for Children**



Figure 3 represents the percentage of estimated absorbed mass apportioned by exposure route (excluding dietary ingestion) before and after the application. Before application when background concentrations are low, the primary route of exposure to diazinon was likely inhalation with an estimated of 49% of total accountable absorbed mass, while hand-to-mouth ingestion potentially accounted for 33% and dermal about 18%. However, following the application hand-to-mouth ingestion is estimated to have increased sharply to 63% of the total, dermal exposure also increased to 35% while inhalation exposure declined to 2%. Examination of the per cent contribution by day (Figure 4) further suggests that the contribution through inhalation becomes nearly negligible immediately after the application. Furthermore, the potential contribution through dermal exposure reached its maximal levels by two days post application followed by a decrease. Potential exposure via the gastrointestinal route remained important over the duration of the study. The relatively large potential intake through non-dietary ingestion is a function of the soil and carpet dust concentration and the transfer coefficient. In light of the attribution of the majority of adsorbed mass to hand-to-mouth ingestion, it should be noted that the values employed for the transfer coefficients are highly uncertain.

The calculated absorbed mass for each participant at each sampling interval is illustrated in Figure 5. Considerable variation can be observed among the six participants. In addition to the amount of diazinon applied and measured diazinon levels, these differences may result from individual activities on the treated turf, the transport of residues into homes and highly variable housing factors. Generally the absorbed mass estimates coincide with the maximal concentrations measured from the turf (both soil and turf transferable).

The total amount of diazinon absorbed from all exposure routes excluding dietary on days following application ranged from 0.271 to 173 ng/day, with a median value of 17.8 ng/day. Since dietary ingestion has been previously reported as representing the dominant route of exposure to diazinon in the absence of any recent application (Morgan et al., 2004), a comparison to dietary exposure is warranted. No dietary samples were collected in this study, and published diazinon ingestion data are sparse. However, Morgan and colleagues (2007) estimated a median dietary intake of approximately 12 ng/day (maximum = 61 ng/day) among 111 children in North Carolina and Ohio enrolled in the CTEPP Study (Morgan *et. al*, 2007), and Moschandreas and colleagues (2001) estimated a median dietary exposure of 0.55 ng/kg/day for Arizona children and adults, corresponding to a range of 9.4 to 20 ng/day for the individuals in this study. The estimated daily amount of diazinon absorbed into the body as a direct result of outdoor turf applications was similar to the amount typically absorbed through dietary ingestion. For the most highly exposed participant, however, the potential contribution due to turf treatment was approximately eight times that of the estimated contribution due to dietary ingestion on days 2 through 8.

## Summary

Insecticide applications to lawns, although uncommon in many countries, are very common in the U.S. This study examined turf applications of granular diazinon, a semivolatile organophosphate insecticide, which has since been deregistered for use on residential lawns. We can derive only rudimentary inferences from these findings due to the small sample size. We anticipated that semivolatile compounds are transported from the source into the indoor living area and that exposure by inhalation would be an important route of exposure. Our results, based on modeling, suggest that the insecticide residues do indeed infiltrate the home, but that inhalation is only a minor potential route of exposure after outdoor lawn applications.

The concentration-time profiles in the various exposure media (Figure 1), particularly for carpet dust suggest that for this type of application, track-in may be the most important mechanism for translocation. The early peak of indoor air concentrations relative to dust concentrations provides evidence of an intrusion of volatilized diazinon.

The utility of the doormat sweeping in establishing a relationship between outdoor and indoor concentrations is equivocal, as the doormat levels are highly predictive of carpet dust concentrations but not predictive of indoor transferable residues levels. Nonetheless the doormats provided supportive evidence that pesticide residues were deposited at least to the outdoor/indoor interface and potentially migrated further into the homes.

SHEDS model estimates suggest that home occupants might experience exposure to this pesticide following outdoor applications. While this exposure may be primarily through non-dietary (i.e. hand-to-mouth) ingestion, the dermal route, and (to a lesser extent) the inhalation route also contribute to the total absorbed mass. The estimated daily absorbed mass declined little over the nine days of the study. It is unlikely that the typical consumer of such products realizes the extent to which the homes become contaminated and the occupants exposed following outdoor applications.

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**Table 1. The limits of detection for spiked matrices, lowest detectable levels for field samples and per cent recoveries for diazinon spiked media.**

<i>Matrix</i>	<i>Matrix Detection Limit</i>	<i>Method Detection Limit</i>	<i>%Recovery Diazinon</i>
Air (PUF)	4.7 ng/PUF	0.9 ng/m <sup>3a</sup>	88
PUF Roller	4.7 ng/PUF	0.003 ng/cm <sup>2b</sup>	75
Soil	0.2 ng/ dry g	1.7 ng/ dry g	95
Formulation	0.6 ng/g	0.6 ng/g	NA <sup>c</sup>
Doormat	0.2 ng/g	0.2 ng/g	74
Carpet Dust	0.6 ng/g	0.1 ng/g	109

<sup>a</sup> calculated based on average flow rate of 3.8 L/min for 24-h. <sup>b</sup> Calculated based on 20 cm X 200 cm sampling area. <sup>c</sup>NA indicates that values were not determined for the matrix.

Table 2. SHEDS model input parameters

Input Parameter	Distribution Type	Frequency	Parameter Estimate <sup>a</sup>			Units	Reference
			$v_1$	$v^2$	$v_3$		
Inhalation absorption factor	Triangle	Person	0.5	0.9	1	[-]	Nolan et al, 1984; Stout and Mason, 2003.
Dermal absorption rate for dust or soil	Uniform	Person	0.001	0.3		1/day	Wester, 1993.
Dermal absorption rate for surface residues	Point	Person	0.03			1/day	Wester, 1993.
GI absorption rate for dust or soil	Point	Person	0.8			1/day	Poet, 2002; Poet 2004.
GI absorption rate for surface residues	Point	Person	0.8			1/day	Poet, 2002; Poet 2004
Soil-skin adherence factor	Point	Person	0			mg/cm <sup>2</sup>	Zartarian <i>et al.</i> , 2003.
Body-surface fractional contact rate	Beta	Hour	3	6.7		1/hr	Zartarian <i>et al.</i> , 2003.
Hand-surface fractional contact rate	Beta	Hour	9.4	33		1/hr	Stout and Mason, 2003
Maximum dermal loading for body	Uniform	Person	0.1	0.7		ug/cm <sup>2</sup>	Stout and Mason, 2003
Maximum dermal loading for hands	Uniform	Person	0.1	2		ug/cm <sup>2</sup>	Zartarian <i>et al.</i> , 2003.
Fraction of body unclothed	Beta	Day	3	6.7		[-]	Zartarian, 2006.
Fraction of surface of single hand that enters mouth	Beta	Person	3.7	25		[-]	Zartarian, 2006.
Dust ingestion rate	Point	Person	1			mg/hr	Ozkaynak, 2005.



Table 2. SHEDS model input parameters (continued)

Input Parameter	Distribution Type	Frequency	Parameter Estimate <sup>a</sup>			Units	Reference
			v <sub>1</sub>	v <sub>2</sub>	v <sub>3</sub>		
Soil ingestion rate	Point	Person	1			mg/hr	Zartarian <i>et al.</i> , 2003
Bathing removal efficiency	Beta	Hour	17.1	5.1	0	[-]	Zartarian <i>et al.</i> , 2003
Mouthing removal efficiency	Triangle	Hour	0	0.16	0.32	[-]	Zartarian <i>et al.</i> , 2003
Hand washing removal efficiency	Beta	Hour	32	22		[-]	Stout and Mason, 2003.
Surface-skin transfer coefficient for body (clothed)	Lognormal	Hour	5900	3.8		cm <sup>2</sup> /hr	Hore, 2003; Stout and Mason, 2003.
Surface-skin transfer coefficient for body (unclothed)	Lognormal	Hour	5900	3.8		cm <sup>2</sup> /hr	Hore, 2003; Stout and Mason, 2003.
Surface-skin transfer coefficient for hand	Lognormal	Hour	2029	5		cm <sup>2</sup> /hr	Stout and Mason, 2003.

<sup>a</sup> distribution (v<sub>1</sub>, v<sub>2</sub>, v<sub>3</sub>); triangle (minimum, mode, maximum.); uniform (minimum, maximum.); normal (mean, standard deviation); lognormal (geometric mean, geometric standard deviation).

**Table 3. Physical attributes of participants that were used as input parameters for the SHEDS model**

<i>House #</i>	<i>Sex</i>	<i>Age</i>	<i>Weight (kg)</i>
1	Female	11	26
2	Female	5	21
3	Male	5	17
4	Female	7	28
5	Female	12	34
6	Male	12	36

**Table 4. Diazinon concentrations measured from commercially available (5% active ingredient [w/w]) granular formulations.**

<i>House</i>	<i>Total Amount of Formulation Applied (kg)</i>	<i>Theoretical Active Ingredient Applied (g)</i>	<i>Actual Area Treated (m<sup>2</sup>)</i>	<i>Calculated Application Rate (g/m<sup>2</sup>)<sup>a</sup></i>
1	2.04	122	1022	0.12
2	5.31	268	465	0.58
3	9.12	456	139	3.28
4	12.7	635	648	0.98
5	9.07	453	1303	0.35
6	2.31	116	437	0.27

<sup>a</sup> The labeled rate is 2 lbs of granular material per 1000 ft<sup>2</sup> of turf providing 9.7 g/m<sup>2</sup>. The theoretical mass of the active ingredient (diazinon) per square meter of turf, based on labeled rates is 0.5 g/m<sup>2</sup>.

**Table 5. Diazinon measured from soil (ng/g) following granular applications to residential lawns.**

<i>House</i>	<i>Day</i>				
	<i>Pre</i>	<i>1</i>	<i>2</i>	<i>4</i>	<i>8</i>
1	<1.7	3800	2070	2150	1680
2	<1.7	19995	14757	12640	14213
3	6.1	72375	74954	55044	54230
4	<1.7	38213	64037	13744	24409
5	183	33230	27986	9200	1070
6	8.3	45015	41753	98968	16078
Avg.		35438	37592	31957	18613

**Table 6. Turf transferable residues measured using the PUF roller following applications to lawns.**

<i>House</i>	<i>Day (ng/m<sup>2</sup>)</i>				
	<i>Pre</i>	<i>1</i>	<i>2</i>	<i>4</i>	<i>8</i>
1	0.19	22.1	51.2	0.43	1.9
2	<0.003	48.6	7.4	10.6	1.7
3	<0.003	NC <sup>a</sup>	970.1	26.7	66.3
4	<0.003	75.1	7.1	2.9	0.5
5	<0.003	0.2	0.2	0.1	0.004
6	<0.003	9.1	6.3	7.2	3.6

<sup>a</sup> NC denote that the sample was not collected.

**Table 7. Diazinon measured from particles vacuumed from door mats (doormat sweepings) of homes following turf applications.**

<i>House</i>	<i>Day (ng/m<sup>2</sup>)</i>		
	<i>2</i>	<i>4</i>	<i>8</i>
1	69934	22274	14353
2	482	1079	719
3	213288	133484	21184
4	720007	442972	72376
5	1711656	220933	410559
6	44574	230337	35356



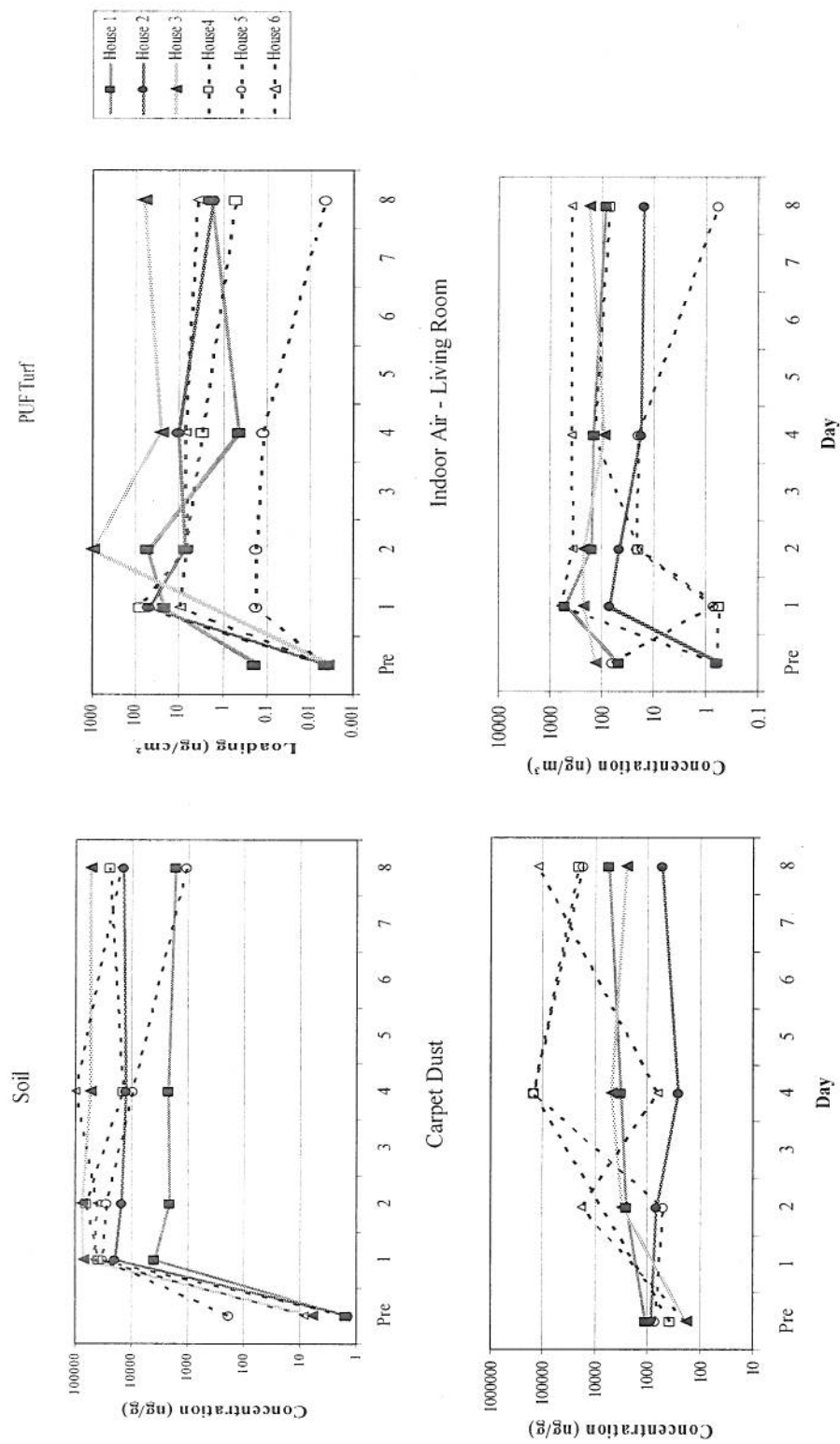


Figure 1. A comparison of temporal changes in diazinon levels from indoor air, carpet dust, soil and transferable residues following an application of a granular formulation to residential lawn.

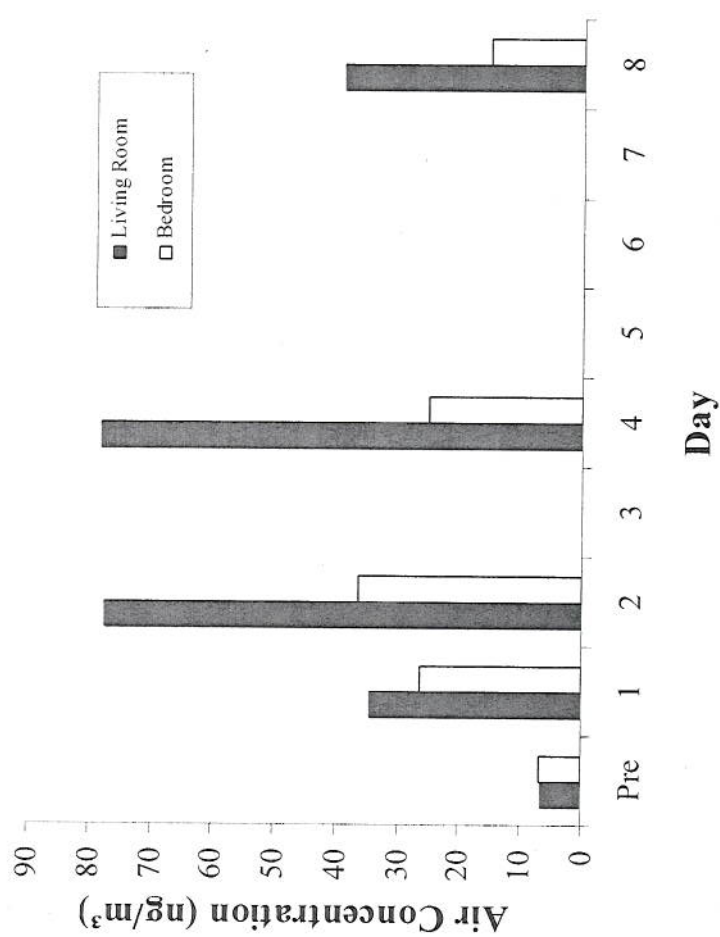


Figure 2. A comparison of the geometric mean of indoor air concentrations measured from the living rooms and bedrooms of six homes following applications of diazinon to residential turf.



Figure 3. The principal routes of the children's exposure to diazinon prior to and following a granular application to residential lawns.



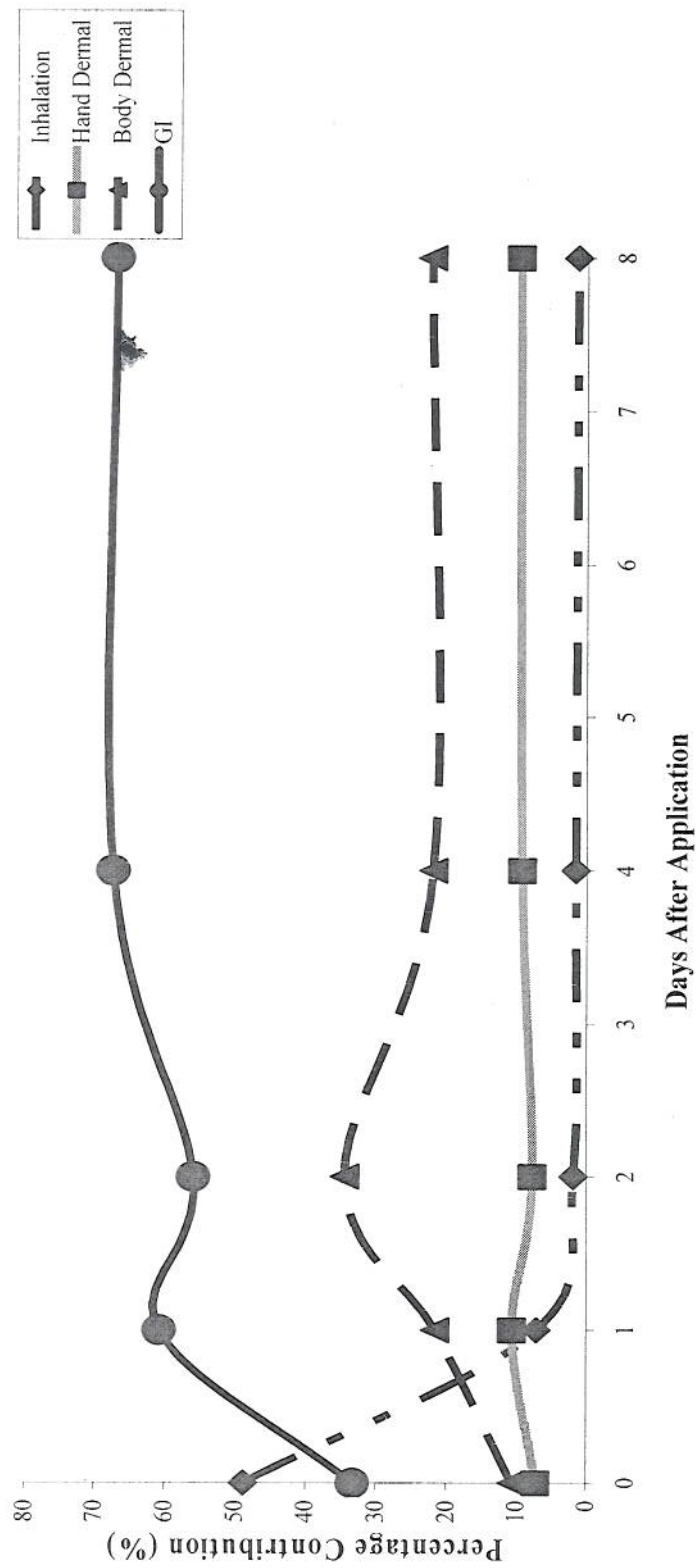


Figure 4. The changes in the contribution of each exposure pathway over time following a granular diazinon application to residential lawns.

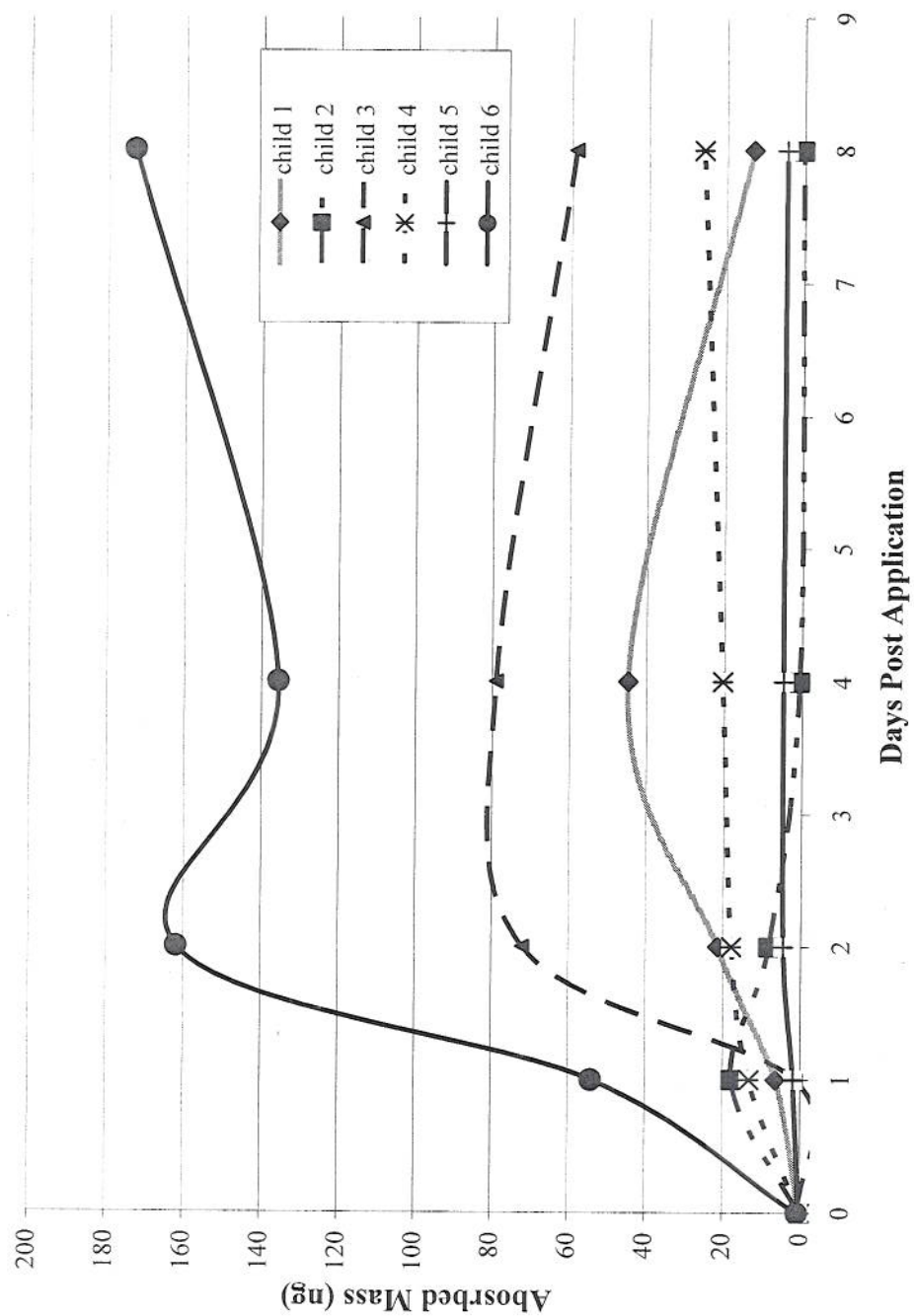


Figure 5. The estimated absorbed mass of diazinon by day for each participant following a granular application to residential lawns.

Figure 1. A comparison of temporal changes in diazinon levels from indoor air, carpet dust, soil and transferable residues following an application of a granular formulation to residential lawn.

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