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Feasibility Study of the Potential for Human Exposure to Pet-Borne Diazinon Residues Following Lawn Applications

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Diazinon (O,O-diethyl-O-[2-isopropyl-6-methylpyrimidin-4-yl]phosphorothioate) is a broad spectrum organophosphorus insecticide commonly used to control a variety of pest insects (ticks, grubs, ants, and fleas) on lawns (Tomlin, 1994; Earl *et al.* 1971). Recently, Stout II (1998) showed an association between applications of insecticides to the exterior perimeter of residential dwellings and residues measured inside the homes. His results suggest that pesticide residues intrude readily into living areas by translocation as vapors and/or by track-in, depending on the pesticides' physical characteristics. Nishioka *et al.* (1997) showed that foot traffic through pesticide (2,4-D) treated turf was a significant mechanism for transport of this lawn-applied pesticide into homes. In a subsequent examination of homes receiving pesticide applications to turf, Nishioka *et al.* (1999) found that carpet dust collected from homes having high child and pet activity had greater levels of pesticide residues than homes having low child and pet activity. These findings suggest that familial factors, such as the activities of children and specifically pets, might serve as an important vehicle for transport of turf-applied pesticides into dwellings. Nishioka's results also suggest that the activity of pets greatly exceeds all other routes of transport for such residues. Furthermore, residues tracked into the home by indoor/outdoor pets, such as family dogs, may be deposited onto surfaces and volatilize or be resuspended into air, potentially exposing occupants. Pets may also transfer pollutants to humans through direct intimate contact (for example, petting, playing, kissing, licking and resting on laps).

The objectives of this study were 1) to investigate the potential for an indoor/outdoor pet dog to transport and translocate diazinon residues into a residence following a lawn application, 2) to determine if intimate contacts between a pet dog and occupants resulted in measurable exposures, and 3) to determine if a pet dog could be a good indicator of exposure following a lawn application of diazinon.

MATERIALS AND METHODS

The study was conducted at a single-family dwelling in Chatham County, North Carolina starting October 18 and ending November 2, 1999. A family of four with an indoor/outdoor dog, who intended to apply diazinon granules to their lawn, was recruited to participate in the study. The children were both female, ages 6 and 10. The dog (*Canis familiaris*) was a one year old, female Australian Shepherd. Family members recorded in recall diaries their general daily activities at the residence starting seven days before application and ending on the fifteenth day after application. The mother also recorded the daily activities of the dog in a recall diary for the same time period.

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The homeowner purchased a commercial formulation of 5% diazinon granules (Real Kill®). Prior to application, researchers randomly dispersed 30 orange stake flags in the yard and placed a petri dish (id=13.7 cm) next to each one. The petri dishes were used to calculate the deposition loading of diazinon over the treated area. The grass varied from 5 cm and 10 cm in length in the front and backyard, respectively. The homeowner used a clean broadcast spreader to apply diazinon to the turf. An aliquot of the formulation was collected from the hopper of the spreader. The sample was transferred into a 250 mL wide-mouth glass jar fitted with a Teflon-lined lid. The petri dishes were gathered and the granules aggregated into a similar jar. The homeowner applied 11 kg of diazinon granular formulation to approximately a 1245 m^2 area of the lawn.

Measurements made outside the house were soil cores, transferable residues from turf, and entryway deposits. Inside the home, airborne residues and vacuum sweepings were collected in the living room, which was the main activity area of the family. Hand wipes were collected from the mother and her two children. Fur clippings, fur wipes, and paw wipes were collected from the dog. The samples were collected approximately three hours before application (pre) and on days 0 (day of application), 3, 9, and 15 after application, except for the entryway deposits and vacuum sweeping samples. Entryway deposits were collected on days 3, 9, and 15 after application, and vacuum samples were collected on days pre, 3, 9, and 15 after application.

The 24-hr air samples were collected using the method described by Lewis *et al.* (1994), which placed the inlet port of the cartridge 75 cm above and oriented toward the floor (a child's approximate breathing height) and an average flow rate of 3.8 L/min. The polyurethane foam roller (PUF) method (Nishioka *et al.* 1997) was used to collect transferable residues from turf. The PUF roller apparatus with a dry PUF sampling ring was rolled on the turf at a rate of approximately 10 cm/s for a 2 m distance. Soil cores (id=25 mm) were collected to measure diazinon residues at a depth of approximately 10 mm with a T-handle soil probe near each orange stake flag. The thirty soil cores were combined in a glass jar. Entryway deposits were collected by placing a new black rubber doormat (43 x 64 cm) at the doorway receiving the highest pet and human foot traffic. The doormat was shaken onto aluminum foil and deposits transferred into a jar. Doormats were replaced at each successive sampling interval. The high-volume surface sampler (HVS3) method (ASTM, 1994) was used to collect vacuum sweepings from 1 m^2 areas of the level loop carpet in the living room. Samples were collected from areas in the living room receiving high pet and human foot traffic.

The hand wipe method by Geno *et al.* (1996) was used to collect transferable residues from the surfaces of the left and right hands of the mother and her two children. The method was slightly modified by presoaking two dressing sponges (Johnson and Johnson SOF-WICK®, 10 x 10 cm, 6 ply) with 20 mL of 2-propanol for each subject. The mother and oldest child performed their own hand wipes before bedtime on days 0, 3, 9, and 15 after application. The mother wiped the hands of the youngest child. The surface area of the hands was estimated by measuring the length and width of each side of a subject's hand. Paw wipes were prepared by presoaking four dressing sponges with 40 mL of 2-propanol. The researcher rubbed each paw (footpads and between toes) with a single dressing sponge and transferred it into the original glass jar containing an additional 50 mL of 2-propanol. The surface area of the front and back paws of the dog was calculated by dipping the paws in non-toxic, red paint (Children's Tempera Paint) and applying the paws to white paper to create a measurable imprint. Fur wipes were prepared by presoaking one dressing sponge with 10 mL of 2-

propanol. An aluminum template with a 7.5 cm x 7.5 cm slot (56 cm^2) was pre-rinsed with 2-propanol, wiped dry, and placed on the chest near the front left leg. The fur was rubbed twice in each direction (up, down, back, forth) with the dressing sponge inside the 56 cm^2 area of the template, and transferred back into the original jar containing an additional 50 mL of 2-propanol. The template in successive intervals was placed on non-sampled areas of the fur. A smaller aluminum template with a 5.0 cm by 5.0 cm slot (25 cm^2), pre-cleaned with 2-propanol, was placed on the chest near the right front leg. Fur was cropped inside the template with solvent rinsed scissors at the base of the hair shaft and transferred into a jar. Immediately after collection, all sample jars were further sealed with Teflon tape and placed in an ice chest at a reduced temperature. Samples were stored at -20°C until chemical analysis. Relative humidity, temperature, and rainfall were reported.

Samples in general were extracted using either Soxhlet or shake extraction techniques. Sample clean-up was performed as necessary. Soil, pesticide formulation, and petri dish samples were analyzed using a HP 5890 Gas Chromatograph (GC), equipped with a split-splitless injector Flame Photometric Detector and a DB-1701 30 m x 0.53 mm id column with a 1 μm film thickness. The remaining samples were analyzed with a HP 6890 GC equipped with a HP 5973 mass selective detector operated in selected ion monitoring mode. Separation was performed using a DB-5.625 30-m x 25-mm id column for the analysis. Field duplicates were collected for air, transferable residues, soil, and fur clipping samples (Table 1). Field blanks were collected for air, transferable residues, HVS3 vacuum sweepings and hand wipe samples. Only the hand wipe field blanks contained detectable level (~10 ng) of diazinon. Field spikes consisted of 1.0 μg/μl and 10 ng/μl of diazinon in 10% ether/hexane. Extraction and clean-up procedures were performed on all samples. Lab spikes were added before the extraction of diazinon in the air, transferable residues, and hand wipe samples. The recoveries were good, ranging from 80% to 120%, except for the vacuum sweepings, which gave recoveries of 143%.

RESULTS AND DISCUSSION

The average temperature and relative humidity were 20°C/60% inside and 13°C/81% outside the home. It rained once during the study, which produced an inch of rain on the night of application (Day 0). The deposition loading of diazinon granules on the lawn (petri dishes) was 0.8 g/m^2. The theoretical loading of diazinon based on the bag's label was 0.5 g/m^2, and the calculated loading based on the weight applied (11 kg) was 0.6 g/m^2. These results showed that the petri dish method slightly overestimated the actual loading of diazinon on the lawn.

Table 1 shows the diazinon residues measured in the samples taken at the residence. Diazinon residues in the soil declined by more than 55% by day 15 (10.1 μg/g) after application. Transferable residues from the turf were as high as 1967.1 $\mu g/m^2$ on day 0, however, they declined to slightly above background levels (14.6 $\mu g/m^2$) by day 15. These residues probably would have been higher, particularly on day 0, if it had not rained the night of application. Entryway deposits on the doormats were 135.0 μg/g on day 3, but declined to 5.1 μg/g by day 15. Airborne residues in the living room were at least 50 times above background levels at 0, 3, 9, and 15 days post-application. In the living room, HVS3 vacuum sweepings collected from the carpet were six times greater than background levels at all sampling intervals. These data showed that diazinon residues were being physically tracked in by the pet and humans, and re-distributed through the indoor air. A gradient of diazinon residues was found from the soil to the carport entryway and into the living room of the home. Nishioka *et al.* (1994)

showed a similar gradient occurring with 2,4-D residues into residences after lawn applications of this pesticide. In addition, subjects could continue to be exposed to diazinon, since residues in the soil remained as high as 10.1 μg/g on day 15. The children had higher levels of diazinon residues on their hands than did the mother on days 0 and 3 after application (Table 1). These were as high as 0.35 and 0.30 μg for Child A (oldest) and Child B (youngest), respectively. However, Child B was exposed to the greatest amount of diazinon (0.94 ng/cm^2) on day 3 as indicated by the surface loading of residues on the hands (Table 2). The activities of the children, such as washing their hands less often or playing with the dog, might have contributed to their higher exposures to diazinon (Table 3). The oldest child had 0.33 μg of diazinon residues on her hands on the day before application. The time-activity diary for the oldest child revealed that the hand wipe sample was taken after school, which suggests that exposure to diazinon might have occurred there. Overall, results showed dermal deposition of diazinon on the hands of the mother and the two children throughout the study.

Table 1. Residues detected at the residence from various matrices following a granular diazinon application to residential turf.

Sample	Unit	Sampling Interval (d)				
		Pre	0	3	9	15
Soil	μg/g	<0.06	27.60 22.10 [a]	11.80 18.30 [a]	17.40	10.10
Doormat	μg/g	----	----	135.00	28.70	5.09
HVS3 Vacuum Sweepings (carpet)	μg/g	0.55	----	4.28	3.59	3.56
Transferable Residues (turf)	μg/m^2	<0.28	1967.11 446.05 [a]	30.26 26.38 [a]	22.30	14.61
Air (living room)	μg/m^3	<0.001	0.18 0.18 [a]	0.07 0.06 [a]	0.05	0.05
Hand Wipe (mother)	μg	0.04	0.17	0.08	0.04	0.08
Hand Wipe (10 yr old)	μg	0.33	0.35	0.29	0.03	0.09
Hand Wipe (6 yr old)	μg	<0.004	0.19	0.30	0.07	0.05
Paw Wipe	μg	0.01	2.27	1.39	0.27	0.55
Fur Wipe	μg	0.006	0.16	0.30	0.11	0.08
Fur Clippings	μg/g	0.20	0.29 0.68 [a]	0.47	0.41	0.60

[a] Duplicate samples

The dog was allowed outside the home about seven times a day for a total of 3-4 hours. The dog stayed outside primarily in the front yard and on the porch. Inside the home, the dog spent the majority of its time in the living room and kitchen while the occupants were awake. The dog had diazinon residues on its paws as high as 2.27 μg (23.16 ng/cm^2) and 1.39 μg (14.18 ng/cm^2) on days 0 and 3 post-application, respectively (Table 1), and residues remained 55 times greater than background levels by day 15. Transferable residues compared to the total extractable amount of diazinon on fur was greatest at 5.43 ng/cm^2 on day 3 and declined to 1.43 ng/cm^2 by day 15 (Table 2). However, transferable residues on the fur were 14 times greater than background levels. In addition, the dog had a higher frequency and duration of spending time outside (>2 hours) on a daily basis than the family members, likely resulting in its higher exposure to diazinon.

The dog was exposed to greater amounts of diazinon compared to the occupants. These results suggest that the dog is a good vehicle for the uptake, transfer, and translocation of residues into the home and is likely to expose occupants to these residues through intimate contacts. Unfortunately, a direct association between pet contacts and the transfer of residues to the occupants remains unclear. Additional studies are planned to clarify further the role of pet dogs in translocating residues and the resulting potential for human exposure.

Table 2. Estimated loadings (ng/cm^2) of diazinon on the hands of the three occupants and on the paws and fur of the pet dog.

Sample (ng/cm^2)	**Sampling Interval (d)**				
	Pre	**0**	**3**	**9**	**15**
Hand Wipe (Mother)	0.07	0.31	0.15	0.07	0.14
Hand Wipe (Child A)	0.66	0.69	0.57	0.07	0.19
Hand Wipe (Child B)	<0.01	0.60	0.94	0.23	0.16
Paw Wipe	0.11	23.16	14.18	2.80	6.64
Fur Wipe	0.10	2.89	5.43	2.00	1.43
Fur Clippings	1.98	5.74	7.01	5.08	5.90

Table 3. The general daily activities of the mother and her two children that were recorded in recall diaries.

Activity	**Adult** (Mother)	**Child A** (10 yr. old)	**Child B** (6 yr. old)
Hand washing events?			
Mean	9	2	2
Median	9	2	2
Work/play?			
A. Outside (Yes/No/NA)	11/11/1	10/12/1	11/12/0
How long (min)?			
Mean	70	31	39
Median	30	30	30
C. Where?	Front Yard	Various	Front & Back Yard
Did you pet the dog?			
A. Yes/No/NA	22/0/1	22/0/1	21/2/0
B. How many times?			
Mean	9	9	4
Median	10	9	3
Total time (min)			
Mean	16	6	23
Median	3	2	5
Spent the most time indoors?	Kitchen	Living Room	Living Room

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