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Distribution of 2,4-D in Air and on Surfaces inside Residences after Lawn Applications: Comparing Exposure Estimates from Various Media for Young Children

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We collected indoor air, surface wipes (floors, table tops, and window sills), and floor dust samples at multiple locations within 11 occupied and two unoccupied homes both before and after lawn application of the herbicide 2,4-D. We measured residues 1 week before and after application. We used collected samples to determine transport routes of 2,4-D from the lawn into the homes, its subsequent distribution between the indoor surfaces, and air concentration as a function of airborne particle size. We used residue measurements to estimate potential exposures within these homes. After lawn application, 2,4-D was detected in indoor air and on all surfaces throughout all homes. Track-in by an active dog and by the homeowner applicator were the most significant factors for intrusion. Resuspension of floor dust was the major source of 2,4-D in indoor air, with highest levels of 2,4-D found in the particle size range of 2.5–10 µm. Resuspended floor dust was also a major source of 2,4-D on tables and window sills. Estimated postapplication indoor exposure levels for young children from nondietary ingestion may be 1–10 µg/day from contact with floors, and 0.2–30 µg/day from contact with table tops. These are estimated to be about 10 times higher than the preapplication exposures. By comparison, dietary ingestion of 2,4-D is approximately 1.3 µg/day. *Key words*: 2,4-D, indoor air, particle size, pesticide exposure, pesticide transport, residential exposure. *Environ Health Perspect* 109:1185–1191 (2001). [Online 6 November 2001]
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A recent review of occupational studies identified numerous cases in which workplace chemicals such as lead, asbestos, and dichlorobenzidine were transported from the workplace to the home. Analyses to document this transport included measurements made in home areas such as the laundry, in clean clothing drawers, and in house dust (*1*). In some cases, the levels of transported occupational chemicals were sufficiently high to cause an adverse health effect in a resident child or spouse. Other studies of the air and house dust of farmers' and farm workers' homes have shown that pesticide residues are transported from the outside to the indoor environment (*2,3*). In one study, organophosphate insecticides were detected in the house dust of pesticide applicators living adjacent to treated orchards, as well as in house dust of nonapplicator farm workers living more than 50 feet from the orchard, and in nearby homes of families not engaged in agricultural activities (*2*). Spray drift, volatilization, soil/foliar resuspension, track-in on shoes, and/or transport on clothing are assumed to have played important roles in the transport of pesticide residues in these agricultural studies.

Agricultural spray drift and residue resuspension rates have been measured for nonvolatile amine salt formulations of 2,4-dichlorophenoxyacetic acid (2,4-D) and dicamba (*4–6*). Because both mechanisms involve the airborne transport of submicron- to micron-size particles and/or aerosols (*7*), it is reasonable to assume that fine particles containing 2,4-D can be resuspended from residential turf by wind, penetrate the exterior of the home through cracks and crevices, windows, and doors, and be deposited on interior surfaces. Field simulation studies following lawn applications of 2,4-D, chlorpyrifos, and chlorothalonil have shown that residential track-in of pesticide residues can occur, and that walking over treated turf as much as one week after application can transport residues on shoes from turf to carpets (*8,9*).

The study reported here was performed in single-story midwestern homes to determine the occurrence and distribution of 2,4-D residues on surfaces and in air within the home—before, during, and after the lawn application of this herbicide. We used these data to describe quantitatively the effects of transport factors and to estimate potential indoor residential exposures of young children. We took samples at seven occupied homes at which the homeowner had applied 2,4-D to the turf, at four occupied homes that had had commercial applications of the same herbicide, and at two nominally unoccupied homes that had had commercial application (e.g., in one unoccupied home, the builder's agent spent 4 hr/day answering the phone there, but entered the home via the garage).

Experimental Methods

Study design. We made assumptions to link sampling methods with both transport mechanisms and exposure pathways. First, specific sampling methods and sampling locations inside the home could be used to assess the magnitude and relative importance of both transport mechanisms and exposure pathways. Second, spray drift, intrusion of resuspended foliar residues, and track-in would contribute to indoor residue levels. Third, foliar resuspension intrusion might be detectable in indoor air on the third day after application; lacking that, this intrusion would cause detectable and equal deposition to floors, sills, and table tops throughout a house. Fourth, track-in would include residues brought in on the applicator's shoes and clothing as well as residues tracked in on subsequent days, and would produce a residue concentration gradient from the entry point. Finally, in-home particle resuspension could overshadow distinct intrusion mechanisms, but the differences among homes and between occupied and unoccupied homes might be used to disaggregate these effects.

Although bias was potentially introduced into this design by conducting the study at the same homes over 2 years, this approach allowed some control for specific activity patterns and factors that were thought to be important. The sampling scheme for each home, summarized in Table 1, lists the sample collection regimen at each home in the matched 1-week preapplication and 1-week postapplication periods. The day on which the application was made constituted day 1 of the postapplication week. Descriptors of

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important factors for each home and application are given in Table 2. In accordance with U.S. Department of Health and Human Services regulations, the study design, protocol, and informed consent were reviewed and approved by Battelle's Human Subjects Review Board.

Sampling and analysis methods. A four-stage cascade impactor sampler (Delron Research Products, Powell, OH) was used for indoor air sampling during the lawn application. It consisted of a series of stages (glass plates coated with polyethylene glycol 1,000 to limit particle bounce) and a final filter (PTFE-coated glass fiber filter, T60A20; Pall/Gelman, East Hills, NY) separated by impactor jets for the following particle/aerosol sizes: < 1 µm, 1–2 µm, 2–8 µm, and > 8 µm. The outlet critical orifice provided a sampling rate of 12.5 L/min with a 370-W diaphragm pump.

Indoor air samples were taken on the first and third days of each sampling week (day 1, day 3) for 24 hr with four co-located samplers (Model 2500; URG, Chapel Hill, NC), each designed to collect a different air particulate size: < 1 µm, < 2.5 µm ($PM_{2.5}$), < 10 µm (PM_{10}), and total suspended particulate (TSP, generally < 20 µm). Each sampler consisted of an inlet jet and impactor plate for particle size discrimination, 27-mm filter (T60A20; Pall/Gelman), and polyurethane foam (PUF) sorbent trap (27 mm × 76 mm; URG). Impactor plates were oiled with 50 µL of silicone oil (Dow-Corning 704; Dow-Corning, Midland, MI). Samplers were located within the breathing zone height, 1.1 m above the floor, separated from each other by 45 cm, and operated at 4 L/min. Pumps were placed in a ventilated polystyrene foam box. The volume of sound produced by the sampler pumps was low enough for families to talk and watch television in the same room.

We determined air exchange and infiltration rates using the Brookhaven National Laboratory (BNL) Air Infiltration Measurement System, which employs small diffusive perfluorocarbon tracer sources and small diffusive samplers (*10*). Sources and samplers were deployed throughout the homes at the time of lawn application and retrieved at the conclusion of the one week sampling period. The 3-zone model was used by BNL in these analyses.

The collection of floor dust by HVS3 vacuum, PUF Roller, and wipe methods have been detailed previously (*8,11*). Sampling for residues on window sills and table tops was similar to that used for bare floors (*11*). We used a cotton gauze wipe (one-half of a Johnson & Johnson SOF-WICK dressing sponge) moistened with 2 mL of a "sweat simulant" (70:30 phosphate buffer:acetonitrile) to collect residues from these surfaces. The surface was wiped once in a single

Table 1. Sampling sequence over two 1-week periods at each home: preapplication week, application time, and postapplication week.

Day/sample and sequence[a]	Air volume or area sampled	Room
Day 1 (application)[b]		
2-hr indoor air	1.76 m^3	Liv
Day 1 (pre- and postapplication weeks)[c]		
24-hr indoor air	5.76 m^3	Liv
Day 3 (postapplication week)		
24-hr indoor air	5.76 m^3	Liv
Day 8 (pre- and postapplication weeks)		
Window sill wipe	Area available	Liv, Din, Kit, Bed
Table wipe	0.08 m^2	Liv, Din, Kit, Bed
Bare floor wipe	0.2 m^2; adjacent to vacuum area	Ent, Din, Kit (as available)
Carpet surface dislodgeable residues	0.48 m^2; perimeter of vacuum area	Liv
Vacuumed dust; bare floor or carpet	1-2 m^2; as available	Ent, Liv, Din, Kit, Bed

Abbreviations: Bed, child's bedroom; Din, dining area; Ent, most frequently used entrance; Kit, kitchen; Liv, primary living area; Pre, preapplication; Post, postapplication.
[a]Day 8 samples collected in the order listed; vacuumed floor dust was collected in the bedroom first, then in reverse order of anticipated loading. [b]Samples collected during application. [c]Samples collected in both preapplication week and in postapplication week.

Table 2. Descriptors for families and homes.

	Home						
Descriptor	A	B	C (X)[a]	D (Y)[b]	E	F	G
No. of adults	2	2	2(1)	2 (0)	2	2	2
No. of children	3	3	3	3	3	3	2
Ages of children, male	8, 10		5, 7, 9	5	7, 10		5, 10
Ages of children, female	6	6, 11, 14		11, 13	12	5, 10, 12	
Child activity level[c]	High	Moderate[d]	High	Low	High	Low	Low
Pets	Dog	None	2 Cats	Dog	Dog	Cat	Dog
Pet activity area[e]	In/out		In/out	In/out	In	Out	Out
Pet activities	Runs with children		Sedate	Old; sedate	House kennel	Garage kennel	
Pet activity for track-in	High	Low	Low	Low	Low	Low	Low
Applicator shoes indoors	Yes	No	Yes	No	No	No	No
Family's shoes worn indoors	Yes	Yes	Yes	Yes	No (Year 2 sometimes)	No (Year 2 sometimes)	Yes
Air infiltration, m^3/hr							
Homeowner	247	407	289	249	254	127	300
Commercial	117	831	(203)	(70)	78	177	NA
Air exchange, L/hr							
Homeowner	0.5	0.7	0.6	0.6	0.6	0.3	0.6
Commercial	0.3	1.4	(0.2)	(0.1)	0.2	0.4	NA
2,4-D on turf, mg/m^2							
Homeowner	43	19	51	56 10, back yard	73, front yard 40, back yard	217, front yard	9
Commercial	46	56	(48)	(48)	45	44	NA
Spray technique							
Homeowner	Hose end	Pump	Hose end	Hose end	Hose end	Hose end	Hose end
Commercial	Hose end	Hose end	(Hose end)	(Hose end)	Hose end	Hose end	NA

NA, not applicable; home not used in commercial applicator study.
[a]Applicable data for unoccupied home X in commercial applicator study listed in parentheses. [b]Applicable data for unoccupied home Y in commercial applicator study listed in parentheses. [c]High activity defined by observations: two boys close in age, share a bedroom, have friends in neighborhood, run and jostle; low activity defined by observations: separate bedrooms, not observed playing together. [d]Three additional children, ages 8 (male), 8 (male), and 11 (female) at this house after school for 2 hr. [e]Areas around house where pet spends time.

direction, the wipe was then folded to the inside, and the surface was wiped a second time, orthogonal to the first direction of wipe. The entire flat surface of a window sill was wiped. Instead of sampling homeowners' table tops, we placed an 850 cm^2 laminate square on each designated table surface on day 1 of each week for subsequent wipe sampling on day 8.

Deposition coupons were pinned lightly to the lawn in 3 locations just before application. These consisted of a full SOF-WICK dressing gauze backed by aluminum foil. After application, the gauze was placed in an extraction tube, and the foil backing was rinsed into this container.

Chemical analysis methods. A similar extraction and cleanup methodology was applied to all matrices, albeit scaled to the size of the sample type. The basic methodology is presented below, with variations as listed in Table 3.

Each sample was spiked with 3,4-D as a surrogate recovery standard (SRS) at a level similar to that expected for 2,4-D: 100 ng for air samples (filters, PUF, plates), surface wipe samples, and carpet dislodgeable residue samples, and 500 ng for dust samples. Samples were extracted with 70:30 acetonitrile:phosphate buffer (0.1 M sodium acid phosphate) at pH 3. We extracted wipe, filter, and impactor plate media using sonication for 10 min; dust samples were sonicated for 10 min and centrifuged, and 80% of the extract was removed. PUF samples (air and dislodgeable residue sleeves) were extracted in an appropriately sized, zippered polyethylene bag by squeezing the solvent through the PUF.

We added distilled/deionized water to the extract, adjusted the pH to 12 with 1M NaOH, and partitioned the extract twice with *n*-hexane. Rotary evaporation removed excess acetonitrile from the PUF sample extracts (80 mL for air PUF and 400 mL for PUF Roller sleeve), after adjusting to pH 12. Emulsions were broken at the interface of dust extracts using either NaCl, a few drops of Antifoam A (Aldrich, Milwaukee, WI), and/or by chilling the separatory funnel for a few minutes. After discarding the hexane, we added water and used a C18 solid-phase extraction (SPE) method for further cleanup (*8,11*).

The SPE eluate was concentrated to near dryness. The internal standard (IS) 2,6-D was added at the same level as the SRS; the extract was then adjusted to 1 mL with 5% methanol in methyl-*t*-butyl ether and then methylated with diazomethane (*8,11*). We analyzed multilevel calibration standards concurrently with samples. Samples that exceeded the calibration range were diluted, respiked with IS, remethylated, and reanalyzed.

Sample extracts were analyzed with gas chromatography/electron capture detection (GC/ECD; Hewlett Packard 5890 GC; Agilent Technologies, Palo Alto, CA). Chromatographic conditions included the following: 60 m DB-5 column [0.25 mm inner diameter, 0.25 µm film thickness (Agilent Technologies)]; temperature program 100–150°C at 6°C/min, 150–215°C at 2°C/min, and 215–300°C at 25°C/min. We conducted confirmation analyses using gas chromatography/mass spectrometry with similar chromatographic conditions and full scan electron impact ionization.

Method validations. Recoveries of 2,4-D and dicamba (a second herbicide acid contained in the commercial formulations used) from the various sampling media were generally 85–95%, and are summarized in Table 4. Retention and distribution between filter and PUF sorbent of both free acids and amine salts during 24-hr air sampling at 4 L/min with room temperature air and varying levels of humidity are also detailed in Table 4. The free acids migrated from filter to PUF sorbent at both 50% and 80% relative humidities (RH). In contrast, the amine salts, though water-soluble, remain largely (> 80%) on the filter.

Average percentage recoveries for SRS 3,4-D in field samples were 90 ± 19 (n = 52) for cascade impactor samples; 91 ± 17 (n = 104) for URG air filter samples; 83 ± 20 (n = 172) for surface wipe samples; 88 ± 19 (n = 115) for floor dust samples; 86 ± 24 (n = 24) for surface dislodgeable residue PUF Roller samples. Field spike recoveries of 2,4-D were 111 ± 33% (n = 23) for wipes; 83 ± 10 (n = 7) for air filters; and 71 ± 12% (n = 3) for PUF sleeves.

Table 3. Variations in extraction/cleanup methods for differing media analyzed.

Sample type	Extraction solvent[a]	Extraction method	First water addition	Hexane partition	Second water addition
Air filter	5 mL × 2	Sonicate	100 mL	20 mL × 2	100 mL
Air PUF	30 mL × 4	Squeeze	80 mL	20 mL × 2	70 mL
Impactor plate	5 mL × 2	Sonicate	100 mL	20 mL × 2	100 mL
Impactor filter	10 mL × 2	Sonicate	100 mL	20 mL × 2	100 mL
Surface wipe	20 mL × 2	Sonicate	360 mL	20 mL × 2	0 mL
PUF roller sleeve	150 mL × 4	Squeeze	150 mL	25 mL × 2	0 mL
Floor dust (bulk)	25 mL	Sonicate	100 mL	20 mL × 2	80 mL

Variation in standard procedure, scaled to size of sample matrix.
[a]Solvent volume added and number of repeats of extraction with that volume; extraction solvent is 70:30 acetonitrile:phosphate buffer at pH = 3.

Table 4. Recoveries of herbicide acids from sampling media.

	Free acid standard			Amine salt formulation		
Matrix	Dicamba (0.5 µg)	2,4-D (1 µg)	3,4-D (0.1 µg)	Dicamba (0.1 µg)	2,4-D (1 µg)	3,4-D (1 µg)
Recovery of spike, %						
Air filter (n = 3)	86 ± 2	90 ± 2	99 ± 6	90 ± 1	93 ± 1	95 ± 4
Air PUF (n = 3)	84 ± 3	86 ± 3	88 ± 3	93 ± 1	90 ± 1	95 ± 4
Impactor plate (n = 2)	82 ± 3	83 ± 2	88 ± 1	92 ± 2	93 ± 1	91 ± 1
Surface wipe (n = 2)	68 ± 3	86 ± 1	87 ± 1	NT	NT	NT
PUF Roller (n = 2)	84 ± 6	105 ± 4	105 ± 2	NT	NT	NT
Dust (n = 3)	87 ± 2	84 ± 9	93 ± 6	NT	NT	NT
Deposition coupon (n = 2)	NT	NT	NT	86 ± 3[a]	89 ± 1[a]	NT
Retention and distribution with 24-hr air sampling, %[b]						
RT; 50% RH: filter	26 ± 3	72 ± 2		81 (n = 1)	82 ± 1	
PUF	57 ± 5	21 ± 1		22 (n = 1)	ND	
Sum	83	93		103	82	
RT; 80% RH: filter	13 ± 1	67 ± 1		77 ± 1	85 ± 3	
PUF	83 ± 1	30 ± 1		12 ± 2	ND	
Sum	96	97		89	85	

Abbreviations: ND, not detected; NT, not tested; RH, relative humidity; RT, room temperature, ~20°C.
[a]Spike level was 6.5 µg of dicamba and 65 µg of 2,4-D. [b]Analytes spiked onto filter and then air drawn through sampler for 24 hr.

Results and Discussion

2,4-D in indoor air by particle size. We detected no 2,4-D (< 0.2 ng/m^3) in the preapplication indoor air samples. Because windows and doors were open at all homes during applications (except at unoccupied homes), we anticipated spray drift intrusion. The mean and range of indoor air 2,4-D concentrations in $PM_{2.5}$ and PM_{10} particle sizes during and following homeowner and commercial applications are shown in Figure 1. Several trends are evident: *a*) With homeowner applications, there is about a 3-fold decrease in average 2,4-D levels between the 2-hr application time on day 1 and the integrated 24 hr of day 1; *b*) with commercial application, there is about a 2-fold decrease in average 2,4-D levels between the application

time and that full day of sampling; *c*) within each application method, the average levels on days 1 and 3 are remarkably similar, although the ranges are considerably different; *d*) the levels in indoor air during the application time appear to be about 3-fold lower with the commercial application, compared with the homeowner application; and *e*) the 2,4-D levels on $PM_{2.5}$ are similar on days 1 and 3 of both application methods (~1.5 ng/m^3), but there is about a 2-fold difference in the levels associated with PM_{10} (4 ng/m^3 vs. 2 ng/m^3). As shown in Figure 2, the average 2,4-D level in the three larger particle sizes was lower by about a factor of 2 during commercial applications than during homeowner applications. For the < 1 µm particle size, the average levels of 2,4-D were similar at all times (~1 ng/m^3), except during the commercial application. This difference in the 2,4-D levels on the < 1 µm particles for the two applications may have been caused by slightly different collection protocols. For the homeowner applications, a consistent air sample collection time of 2 hr was used. Because this sampling time exceeded the time required for application, most homeowners completed spray application and reentered the home before the cascade impactor sampler was stopped. For this reason, with homeowner application, some of the indoor 2,4-D that appeared during application may have been carried in by particles released from the homeowner's clothing. The consistency in 2,4-D on the < 1 µm particles in all homes on day 1 and day 3 after application may also indicate the foliar resuspension intrusion mechanism.

After the applications, approximately 70% of the total indoor airborne 2,4-D was associated with inspirable particles (i.e., particles < 10 µm, PM_{10}); of that inspirable material, approximately 30% with homeowner application and 80% with commercial application was respirable (i.e., particles < 2.5 µm, $PM_{2.5}$). Among homes, the major differences between the postapplication indoor 2,4-D air levels were caused by levels on particles > 2.5 µm. These differences stem from variation in familial activity, which is, in turn, related to the amount of 2,4-D tracked in by family members and pets. Examination of the postapplication day 1 and day 3 air data on a home-by-home basis shows that the higher 2,4-D air levels were associated with homes with active children and pets, and especially with those where shoes were also worn indoors. Similarly, the homes with lower 2,4-D air levels tended to be those with low levels of activity and/or no shoes worn indoors.

2,4-D on tables, window sills, and floors. Residues of 2,4-D were detected in all preapplication floor dust samples (wipe and vacuum) except the bare floor wipe samples in the two unoccupied homes. The range of preapplication levels of 2,4-D in floor dust was fairly narrow, generally 0.2–1.0 µg/m^2. Residues of 2,4-D were detected in all postapplication floor dust samples, except the kitchen floor of one unoccupied home, and the increases in the 2,4-D loadings (µg/m^2) one week postapplication were readily apparent. Postapplication loadings ranged from approximately 1 to 200 µg/m^2. Dicamba was detected in these samples as well, in the same ratio to 2,4-D as the formulation, 10% of the 2,4-D level; this compound will not be discussed further.

In occupied homes, track-in was the dominant mechanism for contribution of residues to floors. In occupied homes, we

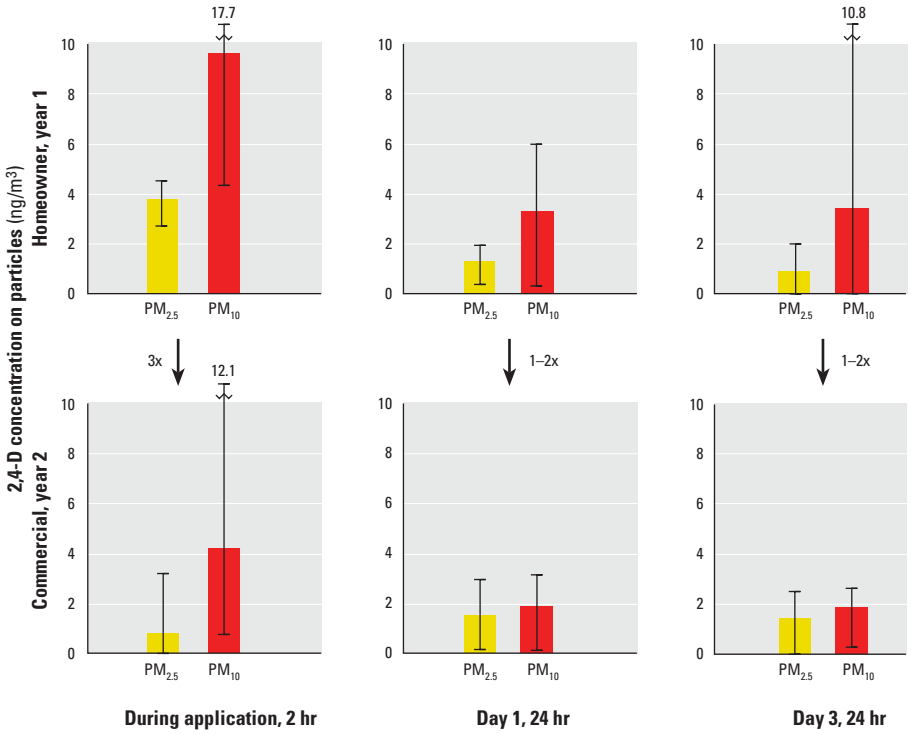

Figure 1. Indoor 2,4-D air levels on $PM_{2.5}$ and PM_{10} particles during and after homeowner and commercial lawn applications.

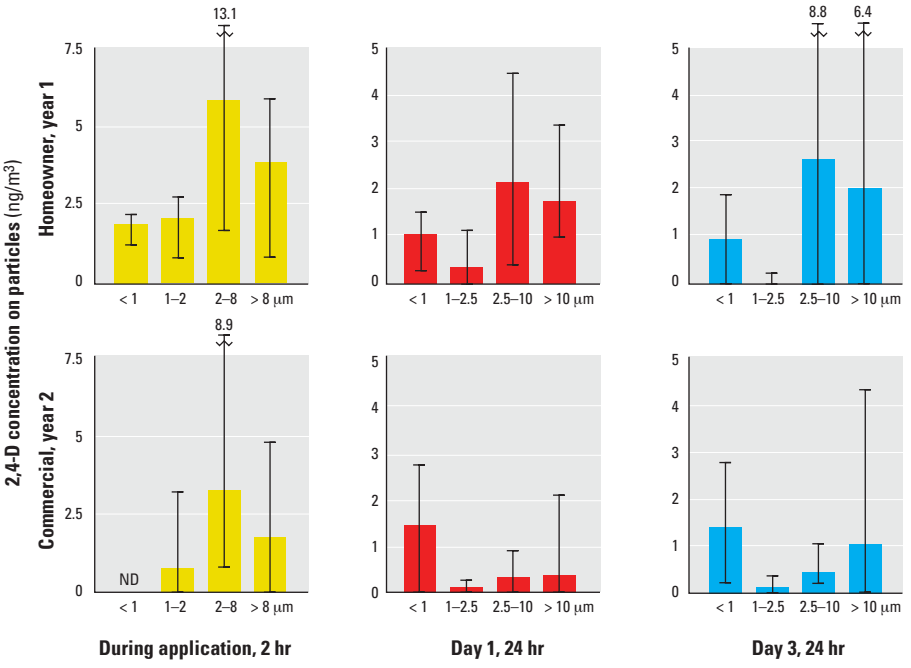

Figure 2. Indoor air concentrations (ng/m^3) of 2,4-D on four different particle sizes during and after homeowner and commercial lawn applications.

observed a concentration gradient in 2,4-D that followed the traffic density pattern within the home. This gradient in 2,4-D levels from high to low was evident whether calculated on the basis of 2,4-D surface loading (μg/m^2) or 2,4-D dust concentration (μg/g), and is consistent with our expectation of track-in from external sources. On the basis of particular activity characteristics of each home, it was possible to disaggregate the 2,4-D surface loadings into the contributions of multiple transport mechanisms, notably the dog, children, their shoes, and the applicator's shoes when worn inside (*11*).

During the preapplication period, we detected 2,4-D at low levels (< 1 μg/m^2) on 10% of the table tops (4 out of 40) and on 25% of the window sills (10 of 42). At the end of the postapplication period, we detected 2,4-D on sills and table tops in all homes (except sill and table surfaces in one unoccupied home) at levels considerably higher than preapplication levels. The ranges of postapplication surface loadings on floors, tables, and window sills in each home are listed in Table 5. In most homes the 2,4-D levels on sills and tables showed a gradient similar to that seen for the floor loadings, from high to low with the direction of traffic through the home. In those homes exhibiting pronounced gradients (e.g., homes A, B, and C), the 2,4-D loadings on tables and sills were approximately 10% and 8%, respectively, of the floor loadings. The observed traffic-dependent gradients in table and sill surface 2,4-D loadings, combined with the higher levels of activity in these homes, strongly implied dust resuspension within the home as the major source of 2,4-D residues on sills and tables. The 10-to-1 ratio here of floor-to-table 2,4-D surface loadings closely mirrors the reported 10-fold difference in activity-dependent dust resuspension rates: 10^{-4}/hr for low activity and reading and 10^{-3}/hr for normal traffic and play (*1*). The assumption here is that most 2,4-D starts out on the floor, but over time, in high activity homes, it is resuspended and then settles out partly on sills and tables. In the one home that was carpeted throughout (thus having a similar surface for dust resuspension in all rooms), postapplication 2,4-D floor loadings were highly correlated with both sill and table loadings, r^2 = 0.82 and 0.95, respectively.

The 2,4-D surface loadings in the main living area and the 2,4-D air concentrations were compared, and Pearson correlations among these matrices are listed in Table 6. Correlations are high (r^2 > 0.85) between surface loading and 2,4-D TSP and PM_{10} concentrations, and less so between surface loadings and 2,4-D $PM_{2.5}$ concentrations. These results are consistent with reports where deposition of larger particles contributed more to surface loadings than smaller particles (*12*).

In homes characterized as having low child and pet activity and/or homes where shoes were not worn indoors, a gradient in 2,4-D loading on the sills, and to some extent on the tables, was barely evident (e.g., homes E and F with homeowner application). In these homes, the 2,4-D loadings on floors, sills, and tables were comparable and generally in the range of 1–2 μg/m^2. The relatively consistent levels of 2,4-D on all surfaces of these low-activity homes, and the fact that residents removed outdoor shoes before entering, suggests that airborne intrusion mechanisms were the major contributory mechanisms in such homes for indoor 2,4-D levels. Air exchange rates in these low-activity homes were also relatively similar (0.2–0.6 L/hr).

The contributions of household activity descriptors and transport mechanisms to the levels of 2,4-D on floors, sills, and tables in the main living area were determined using a multivariate analysis of variance (ANOVA). The factors are listed in Table 7. As noted there, spray drift and foliar resuspension intrusion, listed together under airborne particle intrusion, accounted for 1% of the total 2,4-D on floors in homes with substantial track-in mechanisms. The applicator's shoes contributed significantly to floor loadings but little to levels on sills and tables. However, the resuspension of floor residues by active children and dogs was important. In fact, about 60% of the residues on the living area floor and 80% of the residues on tables and sills (explanatory power of the ANOVA model) may be attributable to the dog in a

Table 5. Range of postapplication 2,4-D surface loadings along traffic gradient (μg/m^2).

Occupancy, home	Application	Carpet[a]	Bare floor[b]	Table	Window sill
Occupied					
A	Homeowner	228–25	23	27–6.4	22–4.8
A	Commercial	76–32	7.9	10–3.2	8.2–2.6
B	Homeowner	74–5.3	NS	5.1–2.1	3.4–1.7
B	Commercial	24–5.2	NS	2.5–1.9	1.8–1.2
C	Homeowner	70–27	9.2–2.5	3.1–1.7	3.8–1.1
D	Homeowner	17–4.5	0.7–0.3	2.0–1.4	2.0–0.6
E	Homeowner	5.0–3.6	1.6–0.6	4.8–1.3	1.4–0.9
E	Commercial	20–5.0	2.6–1.4	4.8–0.8	3.9–0.5
F	Homeowner	4.0[c]–1.2	0.2	3.5–0.5	1.9–0.8
F	Commercial	6.5–4.4	2.2	1.3–0.9	5.7–0.5
G	Homeowner	3.1– < 0.1	<0.01	2.2–0.3	0.8–0.5
Unoccupied					
X	Commercial	1.9–0.8	1.0–0.8	0.8– < 0.02	0.2– < 0.02
Y	Commercial	0.5–0.05	1.0	ND, <0.02	ND, <0.02

Abbreviations: ND, not detected; NS, not sampled; no bare floors in designated sampling areas.
[a]Carpet dust collected with HVS3. [b]Wipe collection from bare floors. [c]Highest floor loading was in bedroom of child who cuts neighbors' grass; other floors, and sill and table loadings, were approximately equal at all locations except Din table.

Table 6. Pearson correlations between 2,4-D air particulate levels on day 3 and 2,4-D surface loadings in the living area.

Surface	TSP	PM_{10}	$PM_{2.5}$
Table	0.96	0.90	0.46
Window sill	0.93	0.87	0.44
Floor	0.89	0.88	0.45

Pearson correlation: 2,4-D living-area surface loading (μg/m^2) and 2,4-D air level (ng/m^3).

Table 7. Contributions of transport mechanisms to 2,4-D loadings on living-area surfaces.

	Contribution to 2,4-D surface loading (μg/m^2)			Distribution of total loading (%)		
Transport	Floor	Table	Sill	Floor	Table	Sill
Air intrusion[a]	1.7	1.7	1.7	1	7	8
Applicator's shoes worn indoors	51.2	~0	~0	27	~0	~0
High-activity children with shoes	16.7	3	2.1	9	12	10
Low-activity children with shoes	1.7[b]	~0.3[b]	~0.1[b]	—[b]	—	—
High-activity dog	117.5	21	18	63	81	82
Sum	187	26	22	100	100	100

[a]Airborne intrusion includes spray drift (~0 μg/m^2), closed home intrusion through cracks (0.3 μg/m^2), and open house intrusion via opening/closing of doors and windows (1.4 μg/m^2). [b]Included for comparison with high-activity children; value not included in the sum or distribution.

home where all high activity and track-in mechanisms are found.

Turf application rates. The manufacturer-suggested lawn application rate (if assumed equal to the deposition rate) of 80 mg/m^2 for 2,4-D was rarely achieved. Most deposition rates were 30–70 mg/m^2, and many homeowners deliberately applied less in child play areas. Note that the home with the highest deposition rate (217 mg/m^2 in the front yard and 40 mg/m^2 in the back yard) had lowest levels indoors through careful control of track-in. The deposition rate with commercial application was uniform, 48 ± 4 mg/m^2 at the homes.

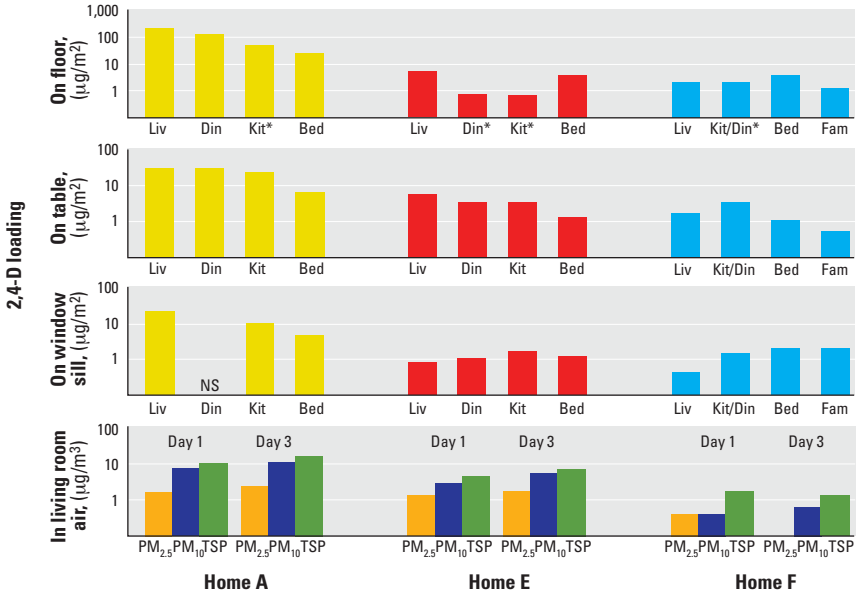

Figure 3. Spatial distribution of 2,4-D in homes of varying lawn application rates, air exchange rates, and activity patterns. Abbreviations: Bed, child's bedroom; Din, dining area; Kit, kitchen; Liv, living area. Room order follows traffic pattern in the home.
*Bare floor.

Table 8. Contributing factors for 2,4-D transport.

Factor	Home A	Home E	Home F
2,4-D lawn deposition rate (mg/m^2)	43 (front and back yard)	73 (front yard) 10 (back yard)	217 (front yard) 40 (back yard)
Air infiltration rate (m^3/hr)	247	254	127
Family activity pattern			
Child activity level	High	High	Low
Pet activity level	High	Low	Low
Family shoes worn indoors	Yes	No	No
Applicator shoes worn indoors	Yes	No	No

Table 9. Potential postapplication nondietary ingestion (NDI) of 2,4-D for a young child (macroactivity and microactivity estimation approaches).

Parameter or NDI exposure	Median value	Maximum value
Bulk Liv floor dust (HVS3 collection) (µg/g)	10.0	67.3
Bulk Liv floor dust (HVS3 collection) (µg/m^2)	12.7	188
Liv carpet surface dust (PUF Roller collection) (µg/m^2)	0.100	1.69
Liv table surface dust (wipe collection) (µg/m^2)	2.69	24.0
NDI: macroactivity and bulk floor dust contact (µg/day)[a]	1.00	6.73
NDI: microactivity and floor surface contact: total (µg/day)	0.012	2.17
Object to mouth (OtM) component[b]	0.004	1.75
Hand to mouth (HtM) component[c]	0.008	0.417
NDI: microactivity and table surface contact: total (µg/day)	0.226	27.8
Object to mouth (OtM) component[d]	0.116	24.8
Hand to mouth (HtM) component[e]	0.110	2.96

Liv, living area.
[a]NDI = dust 2,4-D concentration µg/g × 100 mg dust ingestion/day. [b]OtM = median or maximum transfers to mouth per hour × 12 hr × median or maximum dislodgeable surface dust µg/m^2 × object area of 10 cm^2 × 100% removal in mouth. [c]HtM = median or maximum transfers to mouth per hour × 12 hr × median or maximum dislodgeable surface dust µg/m^2 × hand area of 0.008 m^2 × 10% removal for thumb suck. [d]OtM = median or maximum transfers to mouth per hour × 12 hr × median or maximum table surface dust µg/m^2 × object area of 10 cm^2 × 100% removal in mouth. [e]HtM = median or maximum transfers to mouth per hour × 12 hr × median or maximum table surface dust µg/m^2 × 50% transfer × hand area of 0.008 m^2 × 10% removal for thumb suck.

Effects of activity patterns on postapplication indoor levels. Figure 3 shows the postapplication levels of 2,4-D in three homes after homeowner application. This figure presents the 2,4-D levels on floor, table, and sill surfaces, and the 2,4-D air levels on day 1 and day 3. To accompany Figure 3, we have listed in Table 8 the values for other contributory factors of transport: the level of 2,4-D on the lawn, the air infiltration rate of the home, and the descriptions of family activity patterns. As shown in Figure 3 and Table 8, the household with the highest lawn application rate (F) had the lowest air exchange rate and indoor residue levels, and occupants consistently removed shoes upon entering the house. In a home with high child activity and a no-shoes policy (E), indoor surface residues were also low. In contrast, the home with an active dog and children and shoes worn indoors had significantly higher indoor levels despite application rates and air exchange rates equivalent to home E. The role of high activity is also evident in the increases in indoor 2,4-D air levels day 1 and day 3 in homes A and E. It appears, then, that homeowners can limit a large portion of 2,4-D intrusion into the house through a strict "no outdoor-shoes worn indoors" policy. Control over track-in by a dog is considerably more difficult, but may be accomplished with creative approaches to control and restricted access.

Estimating indoor exposure. We used three scenarios to estimate potential postapplication nondietary ingestion exposures of a 1- to 2-year-old child in these homes, using macroactivity and microactivity approaches that have been discussed recently (*13–15*). The first scenario is a macroactivity approach that assumes 100 mg ingestion of dust per day, regardless of child activities (*16*), and the assumption that the bulk dust from the living room floor, collected with the HVS3, is the dust that is ingested. The second scenario is a microactivity approach that combines the carpet surface dislodgeable residue loading from the PUF Roller with frequency rates for object-to-mouth and hand-to-mouth activities (*17*). We assumed a scenario where the palm side of a child's hand is in constant contact with a carpet (or soft toy) surface such that the residues on the hand are equivalent to the measured carpet dislodgeable residues; either the thumb or the toy is mouthed, with removal efficiencies of 100% for the toy (*18*) and 10% for the hand (which is approximately the same as 100% removal from a thumb, which has 10% of the area of the hand) (*19*). The third exposure scenario is similar to the second one, except that the hand is in contact with the table surface (or smooth toy with equivalent

Table 10. Potential preapplication and postapplication exposures for a young child.

Exposure pathway	Preapplication exposure		Postapplication exposure	
	Median, µg/day (% of total)	Maximum, µg/day (% of total)	Median, µg/day (% of total)	Maximum, µg/day (% of total)
Inhalation[a]	< 0.002 (< 1)	< 0.002 (< 1)	0.030 (1)	0.150 (2)
Nondietary ingestion[b]	0.075 (5)	0.592 (30)	1.000 (41)	6.730 (76)
Dermal penetration[c]	0.008 (1)	0.062 (3)	0.105 (4)	0.705 (8)
Dietary ingestion[d]	1.286 (94)	1.286 (66)	1.286 (53)	1.286 (14)
Total	1.370	1.941	2.421	8.871

[a]Inhalation = 8.7 m^3/day × 2,4-D concentration in TSP ng/m^3. [b]Nondietary ingestion = 100 mg dust ingestion per day × 2,4-D dust concentration µg/g. [c]Dermal penetration = 0.563 m^2 × 31% exposed skin × 0.5 mg dust/cm^2 × dust concentration µg/g × 1.2% absorption of 2,4-D from soil (*13,16*). [d]Dietary ingestion = average 2,4-D intake for non-nursing child < 1 year (*17*).

loading) throughout the day, rather than the carpet surface, and the dislodgeable residue loading is that established by the table wipe measurement. The median and maximum values for various 2,4-D loadings in the living areas of these homes are listed in Table 9, together with the three estimates of potential postapplication nondietary ingestion exposure. The median exposure estimates based on macroactivity and microactivity contact with floor dust (scenarios 1 and 2) differ by almost a factor of 100; contact with the table dust (scenario 3) is about 20% of the macroactivity (scenario 1) estimate. Microactivity-based estimates of exposure suggest that contact with a table surface may produce higher exposures than contact with a carpeted floor surface (0.226 µg/day vs. 0.012 µg/day for median exposures; 28 µg/day vs. 2.2 µg/day for maximum exposure).

Table 10 shows potential preapplication and postapplication exposures for young children. This table includes the four exposure pathways, with median and maximum values estimated using this study's input 2,4-D concentrations for inhalation, nondietary, and dermal penetration. We derived the mean value for the dietary ingestion using the Dietary Exposure Potential Model [(DEPM), version 3.3.2; database options selected for input concentrations of 2,4-D in various foods were the California Pesticide Monitoring Database, 1986–1993; FDA's Compliance and Surveillance Monitoring Program, 1992–1994; and food consumption from the National Health and Nutrition Examination Survey (NHANES) III database, 1998] (*20*). As shown, median exposures in the preapplication period are dominated by dietary ingestion; in the postapplication period, dietary ingestion accounts for about 53%, with the remainder attributable to nondietary ingestion (41%) and dermal penetration (4%). For the maximum exposure scenario, nondietary ingestion accounts for 30% of preapplication exposure and 76% of postapplication exposure.

The World Health Organization's acceptable daily intake for 2,4-D is 300 µg/kg/day, and the U.S. EPA Integrated Risk Information System (IRIS) reference dose (RfD) is 10 µg/kg/day, or 100 µg/day for a 10-kg child (*21*). The data presented here suggest that children are not exposed at levels exceeding the IRIS RfD.

The inferences drawn here are limited by the relatively small number of homes studied. However, to the extent that these homes represent the general population, we can deduce that familial factors (children, pets, and shoes) have a greater effect on indoor residential exposures than application factors. Because exposure must be assessed definitively through the monitoring of biologic markers, studies must be conducted to compare 2,4-D levels in residents' urine with microenvironmental measurements.

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