EPA/600/A-97/075

Comparison of Commercial vs Homeowner Application for Transport of Lawn-Applied Herbicide 2,4-D into Homes

Marcia Nishioka, Charles Hines, Marielle Brinkman, Hazel Burkholder
Battelle Memorial Institute
505 King Avenue; Columbus, OH, USA 43201

Robert G. Lewis
National Exposure Research Laboratory MD-77
U.S. EPA; Research Triangle Park, NC, USA 27711

ABSTRACT

Homeowners are participating in a three year study to measure the migration and transport of lawn-applied herbicide acids 2,4-D (2,4-dichlorophenoxyacetic acid) and dicamba into the home, the spatial distribution of these residues in the home, and the temporal profile of exposures of residents to these residues. Associated activity patterns that are being considered in the study design as contributing factors for transport and exposure include homeowner vs commercial application, activity levels of children and pets, and wearing vs removal of outdoor shoes indoors. Sample types being collected include air, surface wipes, floor dust, dislodgeable carpet surface residues, handwipes and urine. All sample types are collected pre- and post-application, over two one-week periods at each home. Studies have included eight occupied and two unoccupied homes.

With both homeowner and commercial application, track-in is the most significant transport factor contributing residues to the inside of the home. Reductions of 2-10 fold in surface concentrations ($\mu g/m^2$) of 2,4-D and dicamba on floors, sills, and tables in main living areas of the home were found with commercial application, relative to homeowner application. This reduction is most likely due to the fact that the commercial applicator, unlike the homeowner, does not enter the home with contaminated clothing and shoes after application. Levels of 2,4-D in the child's bedroom were very similar, irrespective of application, indicating the significance of their activity patterns for track-in. The changes in floor dust levels were proportional to changes in the PM10 air levels of 2,4-D, suggesting dust resuspension in the home as a contributing factor for air levels and inhalation exposures.

INTRODUCTION

Agricultural pesticide studies have documented pesticide transport and translocation through plants, soil, water, and air (1,2). Although pesticide applications are designed to deposit the formulation onto a target plant, soil or insect surface, inadvertent translocation and deposition often occurs via spray drift, soil/foliar resuspension, and/or volatilization (3). An additional transport mechanism that has been identified for the residential environment is "track-in" or transport of residues into the home on shoes and feet after walking over treated turf (4). Recent field simulation studies of track-in have demonstrated a correlation between turf dislodgeable residues and carpet residue levels (5).

Measurements of pesticide levels in indoor air and house dust have led researchers to

conclude that segments of the U.S. population may undergo chronic exposure due to residential pesticide applications (6). In some cases, the presence of insecticides such as chlorpyrifos can be ascribed to the indoor use of whole room foggers and sprays. The presence of semivolatile organochlorine pesticides, such as chlordane and DDT, indoors appears to be due to infiltration and/or volatilization into the home of pesticides originally applied to foundations. However, the presence *indoors*, of non-volatile pesticides such as 2,4-D and chlorothalonil that are used exclusively outdoors, and often at a distance from the home, strongly suggests the role of mechanisms such as track-in that involve particle transport.

Transport of pesticides into the home carries potential implications for chronic human exposure. Carpets, house dust, and home furnishings become long-term sinks for the pesticides. The common environmental weathering factors such as wind, rain, soil microbes, and sunlight are not available for degradation. Residues on floors and surfaces can become a source of exposure for young children through hand-to-mouth non-dietary ingestion or via inhalation of resuspended dust.

This manuscript describes selected results from studies in which sampling regimens have been developed to elucidate the relative importance of various residential transport mechanisms. The analyses have provided important information regarding both the spatial and temporal distributions inside the home of pesticide residues following lawn application.

SAMPLING AND ANALYSIS

Sampling has been carried out at 8 homes which routinely apply the post-emergent herbicide Trimec, or equivalent, that contains dicamba, mecoprop, and 2,4-D. With the exception of one split level home, all homes were single-story ranch design, had turf on at least three sides of the home, and a lawn area of 0.125-1 acre. All homes had two or three school-age children and one pet (one exception). Sampling was carried out at each home throughout a one-week pre-application period for background measurements, during the herbicide application for turf deposition and spray drift, and during a one-week post-application period. Deposition coupons on the lawn were used to assess application rate. A cascade impactor was run inside the home during the lawn herbicide application to collect the indoor component of spray drift aerosol in the following particle size ranges: <1 μm, 1-2 μm, 2-8 μm, and >8 μm.

The one-week pre-application and post-application periods had the same collection regimen, consisting of 24 hr air sampling, wipe sampling of sills and tables, and vacuum sampling of floors. The air sampling was performed with four co-located 4 L/min samplers consisting of a T60A20 Teflon-coated glass fiber filter and PUF (polyurethane foam) sorbent (URG-2500) with an inlet for particle size discrimination of either <1 μm, <2.5 μm, <10 μm or <20 (total) μm. Air samples were acquired on the first day (day one; day of application for post-application week) and on the second day after application (day three). The wipe samples of four table and four window sill surfaces, and vacuum samples from five separate floor areas were collected on day seven. The floor surfaces typically included an entry area, the main living room, dining area, kitchen and a child's bedroom. The sill and table surfaces, to the extent possible, included areas in the living room, dining area, kitchen and child's bedroom. Each sill and table surface was wiped with a Johnson & Johnson SOF-WICK gauze wipe that was moistened before use with a small amount of a "sweat simulant", a 70:30 phosphate buffer:acetonitrile solution. Each surface was wiped twice in opposite

directions. The floor dust samples were collected from 2 m^2 areas using the HVS3 vacuum sampler (Cascade Stack Sampling Systems, Inc.), which is designed for controlled collection of floor dust particles $>5\mu m$ (7). In addition to these samples, an integrated seven day air exchange rate measurement (Brookhaven Laboratory) was collected at each home during the post-application week.

The extraction, cleanup and analysis methods were similar for all matrices and differed mainly in volume needed for extraction of each sampling medium. The basic method included sonication extraction in a 30:70 phosphate buffer: acetonitrile solution, hexane partition at high pH, C18 SPE cleanup, methylation with diazomethane, and GC/ECD analysis. The method has been reported in detail elsewhere (5). The surrogate recovery standard (SRS) was 3,4-D, and the internal standard for quantification was 2,6-D (note both are positional isomers of 2,4-D and expected to act similarly). Quantification was based on linear regression from multi-point calibration solutions prepared for each matrix over the expected analyte concentration range.

RESULTS AND DISCUSSION

The recoveries of analytes dicamba and 2,4-D, in both free acid and salt form, and recovery of the SRS 3,4-D (free acid only) from the various sampling media are listed in Table 1. As shown there, recoveries were essentially quantitative (>85%), with minor exception, from all media, and slightly higher for the salt form relative to the free acid. In addition, 3,4-D appears to function as an excellent SRS, in that its recovery very closely mirrors the recoveries of analytes.

Measurement of spiked analytes on the filter/PUF sampling system following sampling for 24 hrs at room temperature showed that analytes were fully retained in either free acid form or salt form. Except at high relative humidity, analytes were retained on the filter.

The data presented here in Figures 1-3 show the comparisons between Year 1 (homeowner application) and Year 2 (commercial applicator) for two representative homes for selected sample types. Home A was categorized as a home with high child activity and high pet activity, and Home B had low levels of both child and pet activity. In addition, adults and children in Home B routinely removed shoes at the door when entering from outside; this practice was not observed in Home A.

In Figure 1 are shown the floor dust surface loading of 2,4-D in four areas of each home on Day 7 post-application . Three distinct trends were noted for the study homes in comparing homeowner vs commercial application, and these trends are shown with these two homes. First, for homes such as A, where shoes are not removed at the door, the commercial application resulted in substantially lower surface loadings of 2,4-D in floor dust, by a factor of 3-4X, in main living rooms of the house. Presumably, a significant proportion of the track-in 2,4-D came in on the clothing and shoes of the homeowner/applicator. Second, in all homes, represented here by Homes A and B, the floor dust surface loading of 2,4-D was nearly identical in the child's bedroom irrespective of the application method. We assume that the activity patterns of the child are largely responsible for the track-in seen in their room, and, that child track-in component is not altered substantially by the application method. In those homes where shoes are removed routinely, the psycho-social aspects of participation in a study may be somewhat in evidence. Homeowners report that it is difficult to enforce consistently a "no shoes indoors" policy, and it is the lessening of reminders that may be responsible for the 2-3X higher levels in the second year of the study. In Home B,

the bedroom sampled belonged to the child who cut neighbors' grass, and this activity may help to explain the higher levels there, relative to the rest of the house, in the first year study.

The levels of 2,4-D on table surfaces in Homes A and B are shown in Figure 2. Comparing data in Figures 1 and 2 for Home A, we note that the difference between floor and table loadings ($\mu g/m^2$) was roughly a factor of 10. In a manner analogous to the floor dust 2,4-D surface loadings of Home A, there was also a 2-3X reduction in table surface 2,4-D levels between homeowner and commercial applications. The decline in 2,4-D table levels that mirrors the floor levels as a function of the traffic pattern through the house tends to indicate that the majority of 2,4-D on tables here results from resuspended floor dust. In contrast, the data for Home B are somewhat more difficult to rationalize with respect to the floor dust levels, especially in light of the similarity in floor and table loadings, the similarity in table loadings throughout the house, and the lower overall activity level in the home. These data suggest that a substantial portion of the table surface 2,4-D in Home B resulted from airborne intrusion and settling. Note that in Year 1, the preparation and mixing of 2,4-D before application took place outside the kitchen window.

The levels of 2,4-D in the indoor air two days after application for Homes A and B are shown in Figure 3. The levels of 2,4-D in the PM10 air particles are roughly proportional to the levels in the floor dust. Thus, where floor dust levels were lower in Year 2 with respect to Year 1 for home A, the air levels in PM10 particles were also lower. The same trend was observed for Home B. These data may tend to support the assumption that resuspension of floor dust is responsible for the PM10 levels of 2,4-D in indoor air. The analyses of the co-located samplers for PM2.5 and PM1 show that the PM2.5 levels of 2,4-D are comprised almost exclusively composed of 2,4-D on particles <1 μm. These fine particles may be associated with long term resuspension of 2,4-D from foliar surfaces and subsequent drift. Given the similarity in air infiltration rates for all homes, it is not surprising to find similar 2,4-D PM2.5 levels in the homes.

CONCLUSIONS

The data collected and presented here suggest that in-home ingestion and inhalation of dust residues may contribute to children's exposures to pesticides in general, and 2,4-D in particular. Following application of 2,4-D to a lawn, we have measured 2,4-D in the air and on floor, sill, and table surfaces throughout each home. There was a 3-4X reduction to floor levels and a 2-3X reduction in table surface levels for professional application, relative to homeowner application, and this is believed to be due primarily to the fact that the applicator's contaminated clothing and shoes are not brought into the home after application. The low levels of 2,4-D on sills and tables in low activity homes may indicate the role of foliar resuspension and intrusion, as indicated also by the presence of 2,4-D on particles that are <1 μm, on the second day after application.

Acknowledgment

Battelle acknowledges support of this work under U.S. EPA Cooperative Agreement CR-822082. It has been subjected to Agency review and approved for publication. Mention of tradenames or commercial products does not constitute endorsement or recommendation for use.

REFERENCES

1) Glotfelty, D.E.; J Air Pollut Control Assoc **1978**, 28, 917-921.
2) Stanley, C.W.; Barney, J.E.; Helton, M.R.; Yobs, A.R.; Environ Sci Tech **1971**, 5, 430-435.
3) Glotfelty, D.E.; Schomburg, C.J.; McChesney, M.M.; Sagebiel, J.C.; Seiber, J.N.; Chemosphere **1990**, 21, 1303-1314.
4) Lewis, R.G.; Fortmann, R.C.; Camann, D.E.; Arch Environ Contam Toxicol **1994**, 26, 37-46.
5) Nishioka, M.G.; Burkholder, H.M.; Brinkman, M.C.; Gordon, S.M.; Lewis, R.G.; Environ Sci Tech **1996**, 30, 3313-3320.
6) Whitmore, R.W.; Immerman, F.W.; Camann, D.E.; Bond, A.E.; Lewis, R.G.; Schaum, J.L.; Arch Environ Contam Toxicol **1994**, 26, 47-59.
7) ASTM. Standard Practice for Collection of Dust from Carpeted Floors for Chemical Analysis D5438-93. In Annual Book of ASTM Standards, Vol 11.03; Phil., 1994.

Table 1. Recovery of Analytes and Surrogate Recovery Standard from Sampling Media

		Recovery of Spiked Analyte, %		
		Free Acid		
Sampling Medium	n	Dicamba (0.5 μg)	2,4-D (1 μg)	3,4-D /SRS (1 μg)
T60A20 Filter-air	3	90	93	92
URG PUF Sorbent-air	3	84	86	88
Impactor Plate w/PEG 1000	2	82	83	88
J&J Gauze Wipe-surfaces	2	68	86	87
Vacuumed House Dust	3	87	84	93
PUF Roller Sleeve	2	84	105	105

		Amine Salt Formulation		
Sampling Medium	n	Dicamba (0.1 μg)	2,4-D (1 μg)	3,4-D /SRS (1 μg-free acid)
T60A20 Filter	3	86	90	94
URG PUF Sorbent	3	93	90	95
Impactor Plate w/PEG 1000	2	92	93	91
PUF Roller Sleeve	2	NT	89	NT

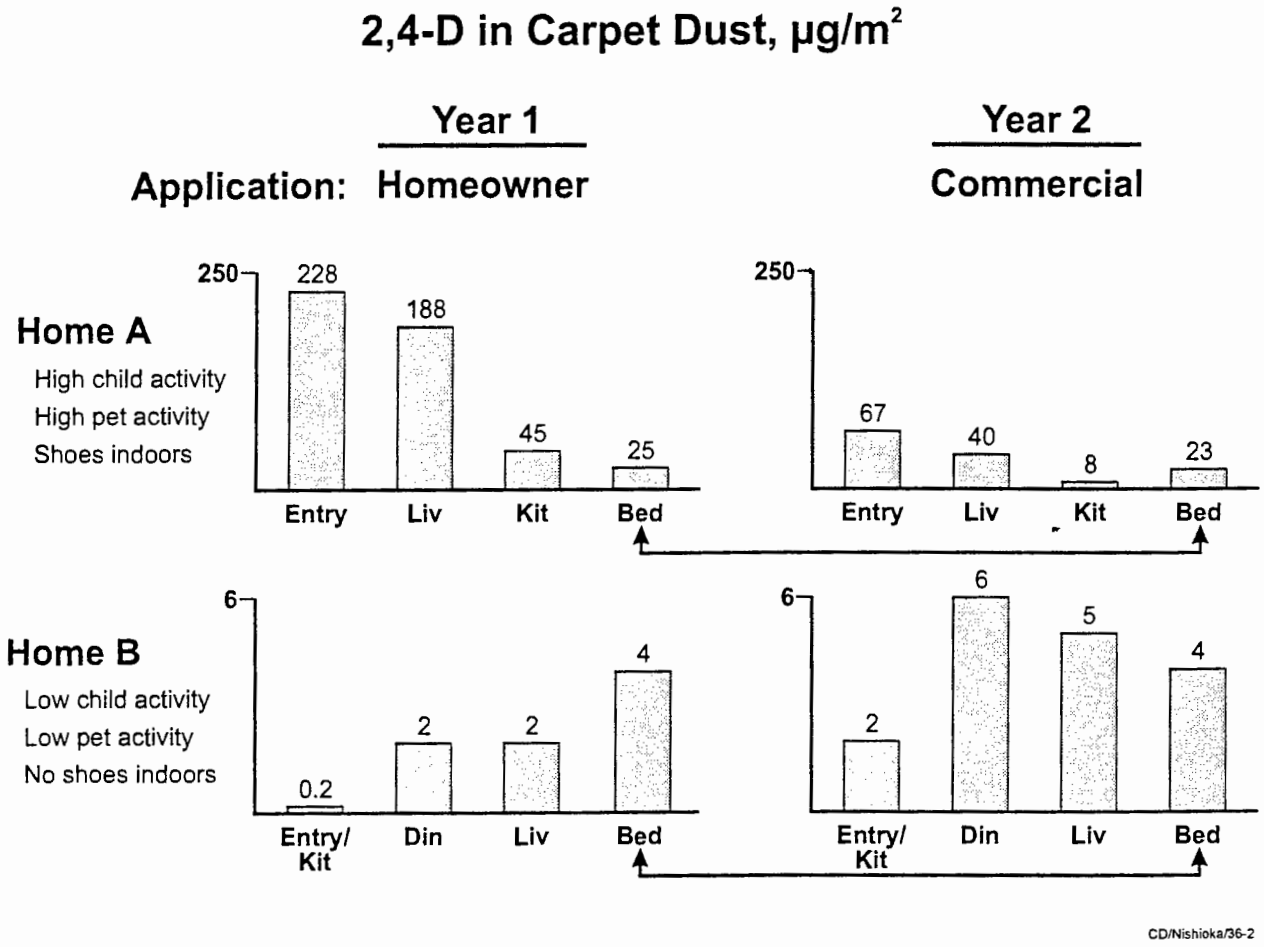

Figure 1. 2,4-D in House Dust for Homeowner and Commercial Application

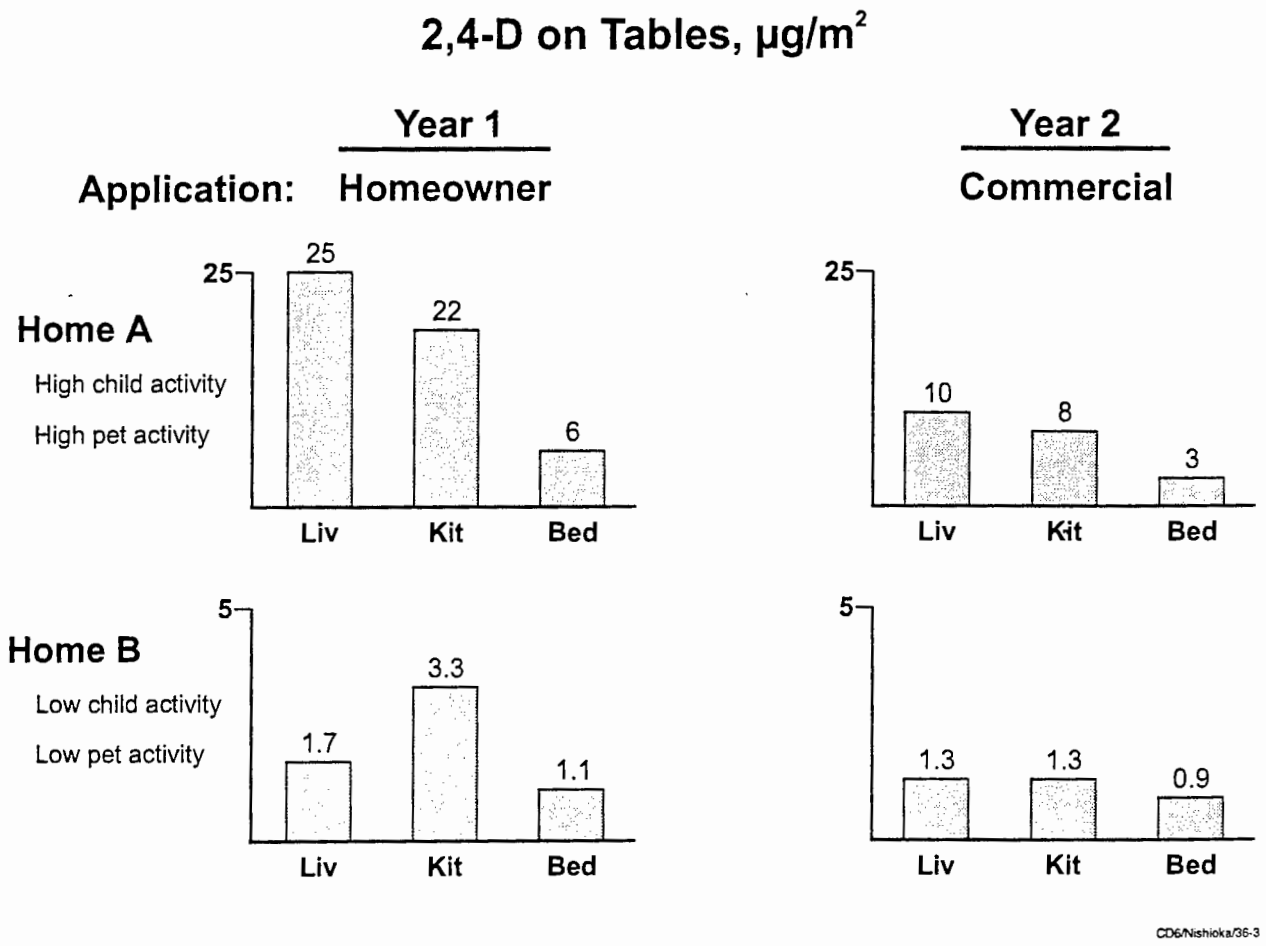

Figure 2. 2,4-D on Table Surfaces for Homeowner and Commercial Application

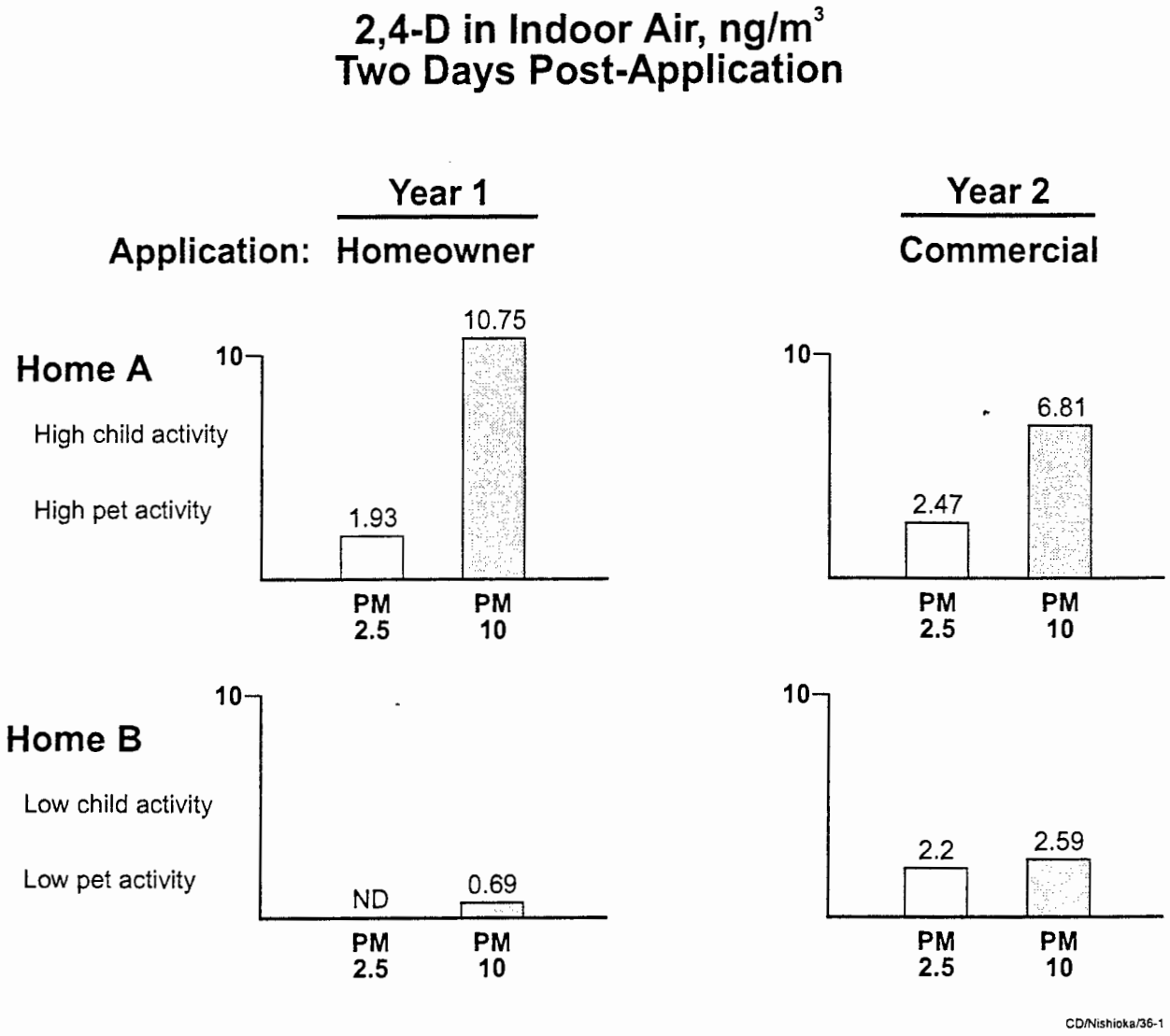

Figure 3. 2,4-D in Indoor Air, Two Days Post-Application to Lawn

TECHNICAL REPORT DATA

1. REPORT NO.	2.	
EPA/600/A-97/075		

4. TITLE AND SUBTITLE

Comparison of Commercial vs Homeowner Application for Transport of Lawn-Applied Herbicide 2,4-D into Homes

5. REPORT DATE

6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)

[1]Marcia Nishioka, Charles Hines, Marielle Brinkman, Hazel Burkholder, and [2]Robert G. Lewis

8. PERFORMING ORGANIZATION REPORT NO.

9. PERFORMING ORGANIZATION NAME AND ADDRESS

[1]Battelle Memorial Institute
505 King Avenue, Columbus, OH 43201

[2] U.S. Environmental Protection Agency
National Environmental Research Laboratory
Research Triangle Park, NC

10. PROGRAM ELEMENT NO.

11. CONTRACT/GRANT NO.

12. SPONSORING AGENCY NAME AND ADDRESS

U. S. Environmental Protection Agency
National Environmental Research Laboratory
Research Triangle Park, NC

13. TYPE OF REPORT AND PERIOD COVERED

14. SPONSORING AGENCY CODE

15. SUPPLEMENTARY NOTES

16. ABSTRACT

Homeowners are participating in a three year study to measure the migration and transport of lawn-applied herbicide acids 2,4-D (2,4-dichlorophenoxyacetic acid) and dicamba into the home, the spatial distribution of these residues in the home, and the temporal profile of exposures of residents to these residues. Associated activity patterns that are being considered in the study design as contributing factors for transport and exposure include homeowner vs commercial application, activity levels of children and pets, and wearing vs removal of outdoor shoes indoors. Sample types being collected include air, surface wipes, and urine. All sample types are collected pre- and post-application, over two one-week periods at each home. Studies have included eight occupied and two unoccupied homes.

With both homeowner and commercial application, track-in is the most significant transport factor contributing residues to the inside of the home. Reductions of 2-10 fold in surface concentrations ($\mu g/m^2$) of 2,4-D and dicamba on floors, sills, and tables in main living areas of the home were found with commercial application, relative to homeowner application. This reduction is most likely due to the fact that the commercial applicator, unlike the homeowner, does not enter the home with contaminated clothing and shoes after application. Levels of 2,4-D in the child's bedroom were very similar, irrespective of application, indicating the significance of their activity patterns for track-in. The changes in floor dust levels were proportional to changes in the PM10 air levels of 2,4-D, suggesting dust resuspension in the home as a contributing factor for air levels and inhalation exposures.

17. KEY WORDS AND DOCUMENT ANALYSIS

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

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