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Selected Pesticide Residues in House Dust from Farmers' Homes in Central New York State, USA

A. T. Lemley,[1] A. Hedge,[2] S. K. Obendorf,[1] S. Hong,[1] J. Kim,[1] T. M. Muss,[2] C. J. Varner[2]

[1] Department of Textiles and Apparel, College of Human Ecology, MVR Hall, Cornell University, Ithaca, NY 14853-4401, USA
[2] Department of Design and Environmental Analysis, College of Human Ecology, MVR Hall, Cornell University, Ithaca, NY 14853-4401, USA

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Pesticides can enter the indoor environment in several ways: by being tracked in from outdoors on shoes, clothing, and the fur of animals; by airborne outdoor particles entering indoors; by indoor application for pest control; and by vapor infiltration from treated foundations and crawl spaces. Once indoors, human exposure to these pesticides can occur through respiration of vapor or particles (Roinestad et al., 1993), dermal absorption, and ingestion from hand to mouth (Hawley, 1985). Pesticides in house dust are known to persist for longer time periods than outdoors because they are protected from degradation by sunlight, moisture, temperature extremes, and most microbial action (Lewis et al., 1994). A definitive pilot study of nine homes in North Carolina (Lewis et al., 1994) evaluated methods of monitoring for pesticide exposure of small children in residential settings. Analyzing for 30 target pesticides, of which 23 were detected in the study, they found that concentrations and frequency of pesticide occurrence were greater in carpet dust than in outdoor soil. Simcox et al. (1995) found similar results in a 59 home study in Washington state. These pesticides in house dust may increase the potential human exposure by direct contact and by inhalation of airborne particles after surface disturbance (Bradman et al. 1997, Fenske et al., 1990, Roberts and Dickey 1995). Sampling house dust gives an estimate of the potential for chronic exposure to pesticides in the home and may serve as an indicator of chronic pesticide exposure (Roberts and Dickey 1995).

The purpose of this study is to analyze the levels of 18 pesticides in house dust samples in 10 farmers' homes. This central New York pilot study was done in order to establish information on pesticide exposure in rural homes and to test methods for future, larger studies. The analytical method was developed for the detection of pesticides that are commonly used in agriculture in central New York State. Method development included extraction procedures and gas chromatography/mass spectrometry analysis (Hong et al. 2001). Based upon the range of pesticides used in agriculture in this region, 14 non-acid pesticides and 4 acid pesticides that have high potential for being present in residential houses were selected as the pesticide residues for analysis in the present study. Settled house dust samples were collected in three ways: by placing filter paper in petri dishes set in living rooms, by wiping smooth surfaces with a moist filter paper, and by vacuuming floor carpets and rugs.

Correspondence to: A. T. Lemley

MATERIALS AND METHODS

House dust samples were collected during the summer of 1999 in ten farmer households in Cortland and Tompkins Counties, New York State, six of whom were New York State certified rural pesticide applicators. The homes of New York State certified pesticide applicators in this study were randomly selected from those who had agreed by phone to participate. A list of farms was compiled based on geographic location, type of agriculture, proximity to crops, and pesticide use. Each home was visited twice with a one-week interval in order to collect two sets of samples. At the time of the first visit there was no control over either when the carpet areas had last been vacuumed or the efficiency of the vacuum cleaner; consequently, a second visit at a constant time interval was included to determine whether or not repeated vacuuming reduced contaminant levels. Characteristics of the homes that were studied are summarized in Table 1. None of the homes had young children as residents and none had air conditioning.

Table 1. House characteristics

House Characteristics	Range
House age range	9 – 140 years
House size	90 – 250 m^2
Type of construction	Wood frame
Farm sizes	100 - 600 ha
Distance from fields	15 – 450 m
Number of homes near corn	8
Number of homes near hay	5
Number of homes near soybeans	2
Number of homes near wheat	2
Number of homes near rye	2
Number of homes near oats	1
Number of homes with porch	7
Number of homes with porch rug	1
Size of living room floor	7.5 – 21.0 m^2
Area of room covered by furniture	0 – 40 %
Age of carpet	1 – 33 years
Carpet pile depth	2 – 8 mm
Number of homes with carpet that had been steam cleaned/shampooed at some time	3

On the first visit to the house, the areas of the room and of textile floor covering in the living room were measured, and the room layout, with dimensions, was sketched. Floor dust was collected from the living room carpet or rug by making four passes with a motorized vacuum cleaner (Hoover Windtunnel mach 6.9) with a high efficiency filtration bag (<0.1 μm pore size) over the entire uncovered textile floor area, excluding regions underneath furniture. The vacuum cleaner was motorized to control for collection speed and domestic to simulate occupant

behavior. Pesticides in dust samples collected in this manner compare well with those collected using high volume surface sampling methods (Colt et al., 1998). Immediately upon completion of vacuuming, the bag was removed, sealed, placed inside an airtight bag, and placed in a cooler for transport to the laboratory. This same vacuuming procedure was repeated for the second visit to each house one week later. Between each specimen collection, the dust path in the vacuum was rinsed three times with 70% aqueous methanol to prevent cross-contamination.

Surface dust wipe samples were taken in the living room of each house. Two contiguous locations, with an approximate area of $0.031 m^2$ (~30 cm by ~10.2 cm), usually a windowsill or table, were selected for replicate sampling and were measured. At each sampling time, two wet filter papers, one moistened with water and the other moistened with aqueous methanol solution, were wiped across the sample surface. The filter papers were sealed immediately in an airtight bag and placed in a cooler for transport to the laboratory. When it was demonstrated that both wipe methods gave similar results, filter papers were combined for analysis.

Dry filter paper (Whatman No. 1, Maidstone, England) was placed in an open petri dish (7.5 cm diameter, with an area of 44.18 cm^2). On the first visit to each house, two dishes were placed in the living room on a flat surface (usually on a shelf or on a table where it would not be disturbed) and were retrieved at the second visit one week later. The dish and filter paper were then immediately sealed in an airtight bag and placed in a cooler for transport to the laboratory.

Fourteen non acid and four acid pesticides were extracted and analyzed as wipe or dust samples using methods developed specifically for this project (Hong et al. 2001). The non acid pesticides were methamidophos, carbaryl, atrazine, methyl parathion, alachlor, 2,4-D-butyl ester, pendimethalin, metolachlor, diazinon, malathion, tetramethrin, trifluralin, resmethrin, and chlorpyrifos. The acid pesticides were picloram, 2,4-D-acid, dicamba, and mecoprop.

The non acid pesticides were first extracted from dust with ethyl acetate:cyclohexane (3:1) which was replaced with dichloromethane. The extract was filtered and collected for further cleanup with Size Exclusion Chromatography(SEC) using a high-resolution SEC polyvinyl benzene/polystyrene column (EnvirosepABC column) protected by an EnvirosepABC guard column (Phenomenex, Torrance, CA) on a HP1090 HPLC (Agilent Technologies, Sunnyvale, CA) equipped with diode array detector (DAD). Detailed SEC conditions can be found in Hong et al. (2001). The effluent was collected manually and condensed for GC/MS analysis. Filter paper samples were extracted with acetone for GC/MSD analysis. Pesticides were analyzed on a HP5890 Series II gas chromatograph coupled to a HP 5971A MS (Agilent Technologies, Sunnyvale, CA). Operating conditions are detailed in Hong et al. (2001). Pesticides were identified by matching both characteristic MS fragment ions and chromatographic retention times.

Acid pesticides were extracted three times from dust with distilled water and $Ca(OH)_2$ (weight ratio of dust:$Ca(OH)_2$ was adjusted to 1:0.1). The pH was adjusted to 1 to 2 and the effluent was cleaned up by SPE with a polyvinylbenzene/polystyrene cartridge (Oasis HLB 6 mL, Waters Co. Milford, MA). The cartridge was washed with distilled water at pH 2 and eluted with methanol in MTBE. The solution was extracted with diethyl ether and dried, followed by addition of methanol and trimethylsilyl diazomethane to methylate the carboxylic acid pesticides.

Filter paper samples were extracted with acidified acetone (3 mM H_3PO_4) The extract was condensed, methylated, and injected into the GC in the same way as dust extract except SPE cleanup was not used. Optimized GC/MS conditions were similar to those for non-acid pesticides (Hong et al.,2001).

RESULTS AND DISCUSSION

Six settled dust samples from the petri dishes were available for pesticide analysis. Results are shown in Table 2. Atrazine was detected in all settled dust samples; 2,4-D butyl ester and resmethrin were each detected in only one sample. No other pesticides were detected. Analyses were performed on 6 wipe samples from the first visits and 9 wipe samples from the second visits (Table 3). Atrazine was detected in all but one sample. Chlorpyrifos was found in 2 samples, and diazinon, 2,4-D butyl ester, methyl parathion, picloram and tetramethrin were found in one sample each.

Table 2. Detection of three pesticides in settled dust samples ($\mu g\ m^{-2}$)

Pesticide	Frequency (n)	Range	Mean	Standard error
Atrazine	6	4.6 to 31.0	12.7	1.6
2,4-D butylester	1	1.5	1.5	--
Resmethrin	1	9.5	9.5	--

Results for the pesticides found in the carpet dust samples from the two visits are shown in Table 4. The pesticides methamidophos and trifluralin were not detected in any of the carpet dust samples. Four pesticides (carbaryl, metolachlor, picloram and 2,4-D acid) were detected in every carpet dust sample. Sixteen pesticides were detected in at least one carpet dust sample.

Both wipe samples and settled dust samples should provide samples of natural airborne particle deposition either from outside or as redistribution of carpet dust. Pearson's correlation coefficients were used to test associations between the first visit surface dust wipe samples, the second visit surface dust wipe samples, and the second visit settled dust samples. Correlations between settled dust samples and wipe samples were significant (Table 5). Levels of pesticides in settled dust samples correlated with levels in surface dust wipe samples for both visits, but were more strongly correlated with levels in surface dust wipe samples for the first visit.

Table 3. Detection of pesticides in the surface dust wipe samples for first and second visits ($\mu g\ m^{-2}$)

Pesticide	First Visit[a]					Second Visit[b]				
	Frequency	Minimum	Maximum	Mean	Standard Error	Frequency	Minimum	Maximum	Mean	Standard Error
Atrazine	6	2.1	76.1	18.1	4.8	8	6.8	92.6	25.3	3.7
Chlorpyrifos	2	32.6	37.4	34.8	3.4	2	16.0	27.7	21.9	4.1
Diazinon	1	6.9	6.9	6.9	----	3	1.0	19.6	9.3	3.1
Tetramethrin	1	4.5	4.5	4.5	----	0	ND	ND	ND	--
Picloram	1	2.4	2.4	2.4	----	0	ND	ND	ND	--
Methyl	1	1.5	1.5	1.5	----	0	ND	ND	ND	--
2,4-D butyl	1	0.7	0.7	0.7	----	1	0.6	0.6	0.6	----

[a] Data on first visit wipes were only available for 6 total households.
[b] Data on second visit wipes were only available for 9 total households.

Table 4. Levels of pesticides in carpet dust samples from ten rural houses ($\mu g\ m^{-2}$)

Pesticide	First Visit					Second Visit				
	Freq	Min	Max	Mean	Std Error	Freq	Min	Max	Mean	Std Error
Metolachlor	10	0.1	10.0	2.6	1.1	10	0.1	8.7	1.8	0.9
2-4-D acid	10	0.0	9.3	1.2	0.9	9	0.1	3.3	1.0	0.4
Picloram	10	0.1	3.4	0.7	0.4	9	0.1	2.6	0.4	0.3
Carbaryl	8	0.1	5.2	1.3	0.6	9	0.1	10.9	3.5	1.3
Atrazine	7	0.1	20.1	4.8	2.7	7	0.1	35.3	6.2	4.9
Mecoprop	7	0.0	0.6	0.1	0.1	7	0.0	1.6	0.4	0.2
Diazinon	6	0.2	27.4	6.4	4.3	7	0.1	5.7	1.3	0.8
Dicamba	6	0.0	2.7	0.8	0.4	3	0.0	1.3	0.6	0.4
2,4-D butyl ester	5	0.8	65.7	17.2	12.4	3	0.1	1.9	0.7	0.6
Methyl parathion	5	0.3	52.0	12.5	10.0	2	0.1	0.1	0.1	0.02
Alachlor	5	1.2	18.1	5.1	3.3	8	0.1	30.1	8.4	4.2
Tetramethrin	4	0.1	12.1	3.5	2.9	4	0.1	3.4	1.5	0.7
Resmethrin	4	1.7	7.0	4.5	1.4	4	0.2	12.3	4.0	2.8
Malathion	3	0.2	0.9	0.5	0.2	5	0.1	11.7	2.8	2.2
Chlorpyrifos	3	0.4	4.2	1.7	1.3	4	0.1	14.7	4.1	3.6
Pendimethalin	1	0.2	0.2	0.2	--	1	0.2	0.2	0.2	--
Methamidophos	0	ND	ND	ND	--	0	ND	ND	ND	--
Trifluralin	0	ND	ND	ND	--	0	ND	ND	ND	--

Pesticide levels in carpet dust samples from the first visit and the second visits were compared but no significant correlation was observed (Table 5). Also, the correlation between surface dust wipe samples and the carpet dust sample from the second visit was not statistically significant.

The amount of pesticides in the first visit carpet dust samples may have been affected by the cleaning behavior of the occupants, such as the frequency of surface cleaning, vacuuming or steam cleaning of the carpet. Cleaning behavior (frequency of surface cleaning, number of times floor vacuuming/week, frequency of steam/hot-water cleaning) was assessed by interview and these data were correlated with first visit total pesticide levels for the surface dust and carpet dust, but there were no significant correlations. The type of carpet that was sampled in each house was recorded but there were six different categories of carpet: cut pile, loop pile, cut and loop pile, jacquard, rug, and plush shag, which yielded too few examples per category for any statistical analyses.

Results for this random sample of rural homes of pesticide applicators and farmers in rural central New York show that multiple pesticides can be detected in house dust. Of the eighteen pesticides analyzed in each floor dust sample, 16 pesticides were detected in at least one household sample, and 12 pesticides were found in at least half of the homes. Of the pesticides analyzed in settled dust samples and wipe samples, 8 pesticides were detected in at least one wipe sample or settled dust sample, but only 2 pesticides were found in more than 1 home.

Atrazine was the most frequently detected pesticide in settled dust and surface dust wipe samples in these homes. It was also detected in carpet dust for both visits for 7 of the 10 houses. Atrazine mean levels were higher for the settled and surface dust wipe samples than for the carpet dust, although a greater number of pesticides were detectable in the carpet dust than in the settled or surface dust wipe samples. This suggests that one primary route of entry for atrazine is in airborne dust, even if tracking in on footwear is also a route of entry. Atrazine is widely used on the crops grown in this agricultural area. If atrazine were deposited due to redistribution from carpet dust resulting from human activity, it would be expected that other pesticides found in carpet dust would also be found more frequently in settled and surface dust wipe samples, similar to results obtained by others for 2,4-D and chlorpyrifos (Nishioka et al, 2001; Lewis et al, 1999). But this was not the case. Since there were no young children living in these homes, redistribution of particulates from this source of activity is not likely, but several of the homes had pets. Also, the presence of atrazine in settled and surface wipe dust is unlikely to have occurred through volatilization, since it is several orders of magnitude less volatile than other pesticides found in the carpet such as 2,4-D acid (3.85×10^{-8} vs. 1.1×10^{-5} kPa, respectively) (Tomlin, 1997).

These samples were collected in the summer time, and none of the homes had air conditioning; thus windows were probably open much of the time, providing a source for airborne particulates. The fact that pesticides were not found in settled and surface dust in most homes but were found in carpet dust suggests that they

remained in the carpet over time from either airborne settling or from tracking and that those found in settled and surface dust were brought in on airborne particles. Pesticide levels in settled and surface dust were correlated, which also supports the theory that pesticides may gain entry indoors on airborne particles (Nishioka et al. 1996). The presence of most pesticides in the second carpet sample, but not in the settled or surface wipe dust samples, implies that the carpet retains pesticides regardless of source.

Table 5. Correlations between pesticide levels in dust samples

Correlation between	Pearson's r	Pr>|r|
Settled dust & second wipe sample	+0.406	<0.0001
Settled dust & first wipe sample	+0.701	<0.0001
First & second wipe sample	+0.103	0.3321
First & second carpet dust sample	+0.124	0.1567
Second wipe & second carpet dust sample	+0.025	0.7731

These results are similar to those of other researchers (Lewis et al. 1994, Bradman et al. 1997, Colt et al. 1998), including the concentration of pesticide in the dust. However, different pesticides were present probably due to differences in regional use. We found atrazine, an herbicide used extensively on the crops of these central New York farms. Lewis and coworkers collected their data in the Raleigh-Durham-Chapel Hill area of North Carolina from a non-agricultural sample, and they most frequently found chlordane, chlorpyrifos, dieldrin, heptachlor, and pentachlorophenol. In an urban Washington DC area sample, Colt et al. (1998) found high frequency of chlordane and pentachlororphenol. In this urban sample, they did not detect atrazine, but they did find the herbicide, 2,4-D. In indoor samples, Whitmore et al. (1994) found high frequency of chlorpyrifos, propoxur, and diazinon in their Jacksonville, FL samples. While in their Springfield/Chicopee, MA samples, the higher frequencies were observed for propoxur and ortho-phenylphenol. In rural, agricultural samples in California, Bradman et al. (1997) found diazinon and chlorpyrifos in multiple households. We have found suggestive evidence that pesticides in house dust may be tracked indoors and that heavily used agricultural pesticides may also enter on airborne dust particles. Future research should further investigate other factors that could influence both the transport into and the redistribution of pesticides in the indoor environment, such as different kinds of textile or smooth floor surfaces, the role of shoe removal, travel distance within the house, cleaning practices, and the presence of pets and children.

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