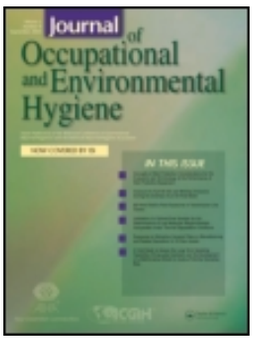

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Pesticide Contamination Inside Farm and Nonfarm Homes

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Twenty-five farm (F) households and 25 nonfarm (NF) households in Iowa were enrolled in a study investigating agricultural pesticide contamination inside homes. Air, surface wipe, and dust samples were collected. Samples from 39 homes (20 F and 19 NF) were analyzed for atrazine, metolachlor, acetochlor, alachlor, and chlorpyrifos. Samples from 11 homes (5 F and 6 NF) were analyzed for glyphosate and 2,4-Dichlorophenoxyac etic acid (2,4-D). Greater than 88% of the air and greater than 74% of the wipe samples were below the limit of detection (LOD). Among the air and wipe samples, chlorpyrifos was detected most frequently in homes. In the dust samples, all the pesticides were detected in greater than 50% of the samples except acetochlor and alachlor, which were detected in less than 30% of the samples. Pesticides in dust samples were detected more often in farm homes except 2,4-D, which was detected in 100% of the farm and nonfarm home samples. The average concentration in dust was higher in farm homes versus nonfarm homes for each pesticide. Further analysis of the data was limited to those pesticides with at least 50% of the dust samples above the LOD. All farms that sprayed a pesticide had higher levels of that pesticide in dust than both farms that did not spray that pesticide and nonfarms; however, only atrazine and metolachlor were significantly higher. The adjusted geometric mean pesticide concentration in dust for farms that sprayed a particular pesticide ranged from 94 to 1300 ng/g compared with 12 to 1000 ng/g for farms that did not spray a particular pesticide, and 2.4 to 320 ng/g for nonfarms. The distributions of the pesticides throughout the various rooms sampled suggest that the strictly agricultural herbicides atrazine and metolachlor are potentially being brought into the home on the farmer's shoes and clothing. These herbicides are not applied in or around the home but they appear to be getting into the home para-occupationally. For agricultural pesticides, take-home exposure may be an important source of home contamination.

Keywords children, house dust, pesticide exposure, surface wipe

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Farmers are the biggest users of pesticides, applying approximately 1.2 billion pounds in 1999; herbicides accounted for the largest proportion of this amount with approximately 534 million pounds applied.[1] A wide variety of agricultural pesticides are used on farms: herbicides, crop insecticides, livestock insecticides, fungicides, and fumigants. Crop herbicides are used the most, with approximately 50% to 93% of farmers reporting their use.[2–4]

Farm families may be exposed to pesticides through home contamination even though they may not participate in farming activities involving pesticide use. Residential environments in proximity to farm operations where pesticides are used may be contaminated through a variety of routes including airborne spread and tracking of contaminated soil into the home, and through deposition on the clothing of applicators. Indirect inhalation and dermal exposure of families to pesticides may occur through redistribution of pesticides via indoor air to surfaces (due to volatilization/condensation and resuspension/settling). A study by Lewis et al.,[5] which collected air, dust, and surface wipe samples, documents rapid translocation of diazinon and chlorpyrifos within the home following indoor and outdoor home applications. Families are also exposed to pesticides through food and in homes that have been sprayed with pesticides.

The potential for exposure of children living on farms to pesticides is a serious concern. Several studies have found an association between *in utero* and postnatal household pesticide

exposure and childhood leukemia.[6–8] Differences in children's physiology, behavior patterns, and hygiene may result in significantly greater exposures to environmental contaminants than adults.[9] Small children spend much of their time on the floor or ground and are very likely to come into contact with pesticide residues on carpets or uncovered floors when playing inside, and yard dirt when playing outside. Children may also be more susceptible than adults to the toxic effects of pesticides, due to the sensitivity of developing organ systems.[10] Older children, through their increased mobility and ability to assist with farm work, may have opportunities for direct pesticide exposure. Although the public health importance of preventing injury to farm children has been well recognized, the hazards of exposure to pesticides and other chemicals to children in the farm environment have received relatively little attention.

Studies have found that farm homes have a greater frequency of detectable residues of pesticides and higher concentrations of pesticides in dust than in reference homes, potentially leading to greater exposure to pesticides among family members.[11–13] Pesticide urine concentrations among the children of farmers and farm workers have been shown to be elevated when compared with children of nonfarm families.[11,14] These studies generally investigated organophosphate and other insecticides. To date, no studies investigating herbicide contamination in farm homes have been conducted.

Recent EPA-funded studies have shown that transport of lawn-applied pesticides in the residential environment can lead to elevated levels of those pesticides in the home within a short time after application. For example, Nishioka et al.[15,16] measured the distribution of the herbicide 2,4-D in homes within a week of a lawn application and showed that transport mechanisms were dominated by track-in from active dogs, the homeowners' contaminated shoes, and the children's shoes when worn indoors. Lewis et al.[5] found that chlorpyrifos residues in indoor air and in carpet dust were higher within a few days of an exterior residential application than before the application and suggested that track-in was the principal source of these residues.

To date studies investigating pesticide exposures among farm families have focused on insecticides, particularly organophosphates, while herbicides studies have not been reported. Studies investigating track-in of herbicides have been so far confined to residential applications. The primary purpose of this article is to investigate farm home pesticide contamination to seven pesticides, six of which are herbicides, and to describe the sources of pesticide contamination in farm homes. A comparison of pesticide contamination will be made among farm homes and reference homes. This article offers unique information on pesticide exposure among farm families not previously studied by investigating herbicide exposure, four of which are not used residentially, which offers insight into paraoccupational exposure pathways in the home.

METHODS

Study Population

In the spring and summer of 2001, 25 farm (F) households and 25 nonfarm (NF) households in Iowa were enrolled in a study investigating agricultural pesticide contamination inside homes. Participant recruitment has been described previously.[17] Briefly, participants were recruited from participants of the Agricultural Health Study in Keokuk and Mahaska counties, the Keokuk County Rural Health Study in Iowa, and by word of mouth. To be eligible for the study, each home had to have at least one child 8 years old or younger. In addition, nonfarm homes had to be located on land that was not used for farming and had no person in the home working in agriculture or commercial pesticide application.

The farm homes had to be using during the spring of 2001 at least one of the 7 target pesticides: atrazine, acetochlor, alachlor, chlorpyrifos, metolachlor, glyphosate, and 2,4-D. These pesticides were selected because of their extensive use in Iowa agriculture. Six of the pesticides are corn or soybean herbicides, while chlorpyrifos is an insecticide used on corn. The study protocol was reviewed and approved by the Institutional Review Boards of the National Institute for Occupational Safety and Health, University of Iowa, and the National Cancer Institute.

Sample Collection

Between May and August 2001, each home was visited on two occasions. The first visit was shortly after a spraying event, and the second visit was approximately 4 weeks later (mean 4 weeks, range 3 to 5 weeks). A three-part questionnaire was administered to either parent at each home on the first visit. The information was updated on the second visit. Part 1 dealt with parental information. Part 2 dealt with child information and included questions about whether children handled pesticides, performed other farm chores, or had access to treated fields. Part 3 dealt with household information, including residential pesticide use in and around the home and proximity of the house to treated fields.

In addition to the three-part questionnaire, a fourth part was administered to the principal farmer in the farm homes about those factors that may influence home contamination, including farm activities, agricultural pesticide use, crops, agricultural practice, and use of personal protective equipment (PPE) since the start of the growing season and throughout the study period. The questionnaire gathered information from the start of the 2001 growing season until the last home visit and generally reflected the early 2001 growing season. With respect to home, yard, and garden use of residential pesticides, homeowners were asked about their use in the month and year prior to the first visit and the month between visits. Data on the pesticide applied, farm practices, farm demographics, and household pesticide use have been reported previously.[17]

Environmental samples, including surface wipe, dust, and air were collected at each visit. Wipe samples were collected from the steering wheel and driver's seat of the primary family vehicle and from the kitchen counter, top of the washing machine, and various rooms with hard surface floors inside the home. Dust samples were collected from carpet where available, including wall-to-wall carpet, area rugs, or floor mats from the entranceway, father's change area, laundry room, child's bedroom, and child's playroom. When floors from these rooms did not have a carpet or rug, a wipe sample was collected. A single 24-hour air sample was collected from the living room of each home and an additional 24-hour air sample was collected outside, near the home. Dust and wipe samples from 39 homes (20 F and 19 NF) were analyzed for atrazine, metolachlor, acetochlor, alachlor, and chlorpyrifos. Dust and wipe samples from 11 homes (5 F and 6 NF) were analyzed for glyphosate and 2,4-D. All air samples were analyzed for atrazine, metolachlor, acetochlor, alachlor, chlorpyrifos, and 2,4-D.

Dust samples were collected from carpets using a high-volume small surface sampler (HVS3, CS3 Inc., Sandpoint, Idaho) using the ASTM *Standard Practice for Collection of Dust from Carpeted Floors for Chemical Analysis*.[18]

The wiping procedure consisted of sampling a 1 ft × 1 ft (900 cm^2) area using two 4-inch × 4-inch (103.2 cm^2) Johnson & Johnson SOF-WICK sponges sequentially. The first sponge was moistened with 10 mL of 100% isopropanol and four adjacent but slightly overlapping wipes of approximately 8 cm width were taken in one direction. The sponge was folded after each pass so that a clean surface was available for each wipe. The sponge was placed in an amber glass jar covered with a Teflon-lined cap. The second sponge was then moistened with 10 mL of 100% isopropanol and four more adjacent 8 cm wide wipes were taken in a similar manner but in a direction perpendicular to the first wipe. The second sponge was added to the jar containing the first sponge.

For the steering wheel, one sponge was wrapped around the steering wheel and half the wheel was wiped. The sponge was folded in half and the second half of the steering wheel was wiped in the same manner. The procedure was repeated with the second sponge starting with the second half of the steering wheel. The sponges were treated the same way as above. Polyurethane foam (PUF) moistened with 6 mL of isopropanol was used in the same manner to sample for glyphosate and 2,4-D.

Air samples were collected for 24 hours on OSHA Versatile Sampler (OVS-2) sorbent tubes (SKC, Eighty Four, Pa.) containing XAD-2 resin with an 11 mm quartz fiber pre-filter and polyurethane foam. The nominal flow rate for the sampling pump was 1 L/min. Pumps were pre- and post-calibrated each sampling day with the OVS-2 media in line using a DryCal DC-Lite (Bios International, Butler Park, N.J.).

Approximately one field blank sample for every 20 air and wipe samples was submitted for analysis along with the field samples. The blank samples were handled in the same manner as the field samples. All blank samples were below the analytical limit of detection (LOD) for all pesticides tested.

All samples were transported from the field in a cooler and transferred to a refrigerator where they were stored for a few days until shipment to the laboratory. At the laboratory, samples were stored in a freezer from 3 to 6 months until analysis.

Sample Analysis

The limits of detection and recovery efficiencies are reported in Table I.

Air

The OVS-2 tubes were separated into two sections with the front ring, filter, and front resin section placed in one 4 mL vial, and the middle PUF separator and back resin section placed in another 4 mL vial. Each vial was desorbed with 2 mL of diazomethane desorption solution.

Wipe

The SOF-WICK sponges were desorbed in their shipping containers with 40 mL of isopropanol, after which an aliquot of each sample was poured into a GC vial for analysis. Liquid standards were used for quantitation. The PUF sponges were desorbed in their shipping containers with 75 mL of methanol. All air and the SOF-WICK sponge wipe samples were analyzed using a gas chromatograph equipped with an electron capture detector using a 30 m DB-1701 column programmed from 130–270°C. The PUF samples were analyzed using a gas chromatograph equipped with an electron capture detector using a 30 m DB-608 column programmed from 90–270°C.

Dust

Dust analyses for acetochlor, atrazine, metolachlor, alachlor, chlorpyrifos (nonacidic or neutral pesticides) included extraction of 0.5 g aliquots of the dust with 12 mL of 1:1 (v:v) hexane:acetone using sonication. The extract was cleaned up

TABLE I. Pesticide Detection Limits by Sample Type

	Air (ng/sample)		Surface Wipe (ng/sample)		Dust (ng/g)[A]	
Pesticide	LOD	% Recovery	LOD	% Recovery	LOD	% Recovery
Atrazine	200	90–100	4000	100	1.5	98
Metolachlor	10	90–100	100	99	0.7	86
Chlorpyrifos	10	90–100	80	103	1.5	83
Acetochlor	10	90–100	100	100	1.5	80
Alachlor	10	90–100	40	100	1.5	82
Glyphosate	NA	NA	400	90–100	0.7	91
2,4-D	200	90–100	700	90–100	0.7	104

[A]Based on an extracted sample of 0.5 g of dust for atrazine, metolachlor, chlorpyrifos, acetochlor, alachlor, and glyphosate and 1.0 g of dust for 2,4-D. NA = not applicable.

using sequential elution on a silica SPE cartridge. Samples were analyzed using GC/MS in the multiple ion detection mode with a 30 m DB-1701 column programmed from 160–280°C.

Analyses for 2,4-D (acidic pesticide) included sonication extraction of 1 g of dust with 25 mL of a 70:30 (v:v) mix of acetonitrile and 0.1 M sodium acid phosphate buffer at pH = 3.[19] The extract was further cleaned prior to analysis using a C18 SPE cartridge. The extract was derivatized using diazomethane. The extracts were analyzed as described above, using a 30 m RTx-5 ms column programmed from 180–280°C.

Analyses for glyphosate (acidic pesticide) included sonication extraction of 0.5 g of dust with 12 mL of deionized water following addition of isotopically labeled glyphosate and acidification of the dust with concentrated H_3PO_4. The extract was partitioned with neutral solvents for cleanup, then evaporated to a small volume under vacuum and then lyophilized overnight. The residue was derivatized with a 2:1 mixture of trifluoroacetic acid and trifluoroethanol, and then extracted into dichloromethane. Samples with chromatographic interferences to glyphosate were further cleaned up using sequential elution on a silica SPE cartridge. Samples were analyzed using GC/MS in the multiple ion detection mode with a 30 m RTx-5 ms column programmed from 120–180°C.

Data Analysis

Pesticide levels were reported in ng/sample for air and surface wipe samples and in ng/g for dust samples by the analyzing laboratories and were not corrected for recovery efficiency. The LOD varied by pesticide and sample type (Table I). The LODs for the dust samples were based on an extracted sample of 0.5 g of dust for the neutral pesticide and glyphosate analyses and 1 g of dust for the 2,4-D analysis. Eighty percent of the dust samples provided at least these amounts. The rest of the samples contained less than 0.5 g of dust. These low mass samples were analyzed in all instances, with the exception of one dust sample that produced sufficient dust only for the 2,4-D analysis. However, the LODs would be higher for these samples due to the smaller amounts of dust. Therefore, LODs for samples less than 0.25 g were adjusted proportionally: the LOD for a sample with mass <0.0625 g was adjusted by a factor of 10, a sample with mass <0.125 g by a factor of 4, and a sample with mass <0.25 g by a factor of 2, unless all other full mass samples from the household were also nondetects.

The number of air samples with detectable levels of pesticide was small, therefore additional analyses were not performed on these samples. The percent of wipe and dust samples above the LOD were computed separately for farm and nonfarm samples. Rates of detection were compared between farm and nonfarm samples using generalized estimating equations (GEE) methods to account for the correlated nature of samples taken within the same household. The GENMOD procedure in SAS, which fits generalized linear models, was used to compute the odds ratio for detecting a positive sample for farm homes versus nonfarm homes. Models specified a logit link function, an exchangeable correlation matrix, and household as a repeated effect.

For surface wipe samples, pesticide levels reported in ng/sample were standardized to ng/cm^2 using the area associated with each sample. Since less than half the wipe samples had analytes present above the LOD, only the range of the detectable samples was reported. For dust samples, pesticide levels reported as "below the LOD" were replaced with one-half of the LOD[20] prior to analysis. Pesticide levels in dust reported as ng/g were standardized to ng/cm^2 using the total mass (in grams) and area associated with each sample. Both the distributions of the pesticide concentration in dust (ng/g) and the pesticide concentration in carpet (ng/cm^2) were highly skewed to the right, therefore a natural log transformation was applied to these concentrations prior to statistical analysis. The geometric mean (GM) and geometric standard deviation (GSD) were reported only when at least 50% of the samples overall were above the LOD.

Since each household was sampled on two visits and more than one dust sample was obtained at each visit, resulting in correlated dust samples, mixed-effects models were used to determine statistical significance. In these models, household was treated as a random effect and group (farm, nonfarm), visit (visit 1, visit 2), and room (child's bedroom, child's playroom, laundry room, father's change room, and entranceway) were treated as fixed effects. For farm households, crop spray records were used to determine whether the pesticide was sprayed in the 7 days preceding the visit (yes, no). The 7-day cutoff was intended to focus on more recent pesticide applications, rather than applications that occurred more than 1 week prior to the visit.

Household covariates (Table II) included the age of the home (<60, ≥60 years); home/lawn/garden sprayed with pesticide in the last month/year (yes, no); the age of the carpets (<8, ≥8 years); frequency of carpet vacuuming (<once per week, ≥once per week); own a dog (yes, no); own a cat (yes, no); presence of doormats (yes, no); and proximity to farm fields (<0.5, ≥0.5 mile). Cutpoints for the age of the home, carpets, and frequency of vacuuming were selected to divide the households approximately equally. Crop demographics, pesticide use and application practices, use of PPE, and children's farm activities were presented previously.[17]

Additional covariates were tested one at a time after adjusting for group, spray status, visit, and room. All mixed models were fit using the MIXED procedure in SAS assuming a compound symmetric covariance structure. Model residuals were assessed for departures from normality. For dust samples obtained from farm homes, estimates of within- and between-household variability, after adjusting for spray status, visit, and room, were computed using the MIXED procedure in SAS assuming a compound symmetric covariance structure.

All significance testing was performed at the 0.05 level of significance. When comparing geometric means for more than two categories, p-values were adjusted using the Tukey-Kramer adjustment for multiple comparisons. All statistical

TABLE II. Household Covariates

Variable	Household Type	
	Nonfarm (n = 25)	Farm (n = 25)
Age of home (years)		
median (range)	30 (1–111)	84 (<1–139)
Percent of homes sprayed with insecticides in the		
last month	12%	40%
last year	28%	52%
Percent of lawns treated with pesticides in the		
last month	28%	12%
last year	38%	32%
Percent of gardens sprayed with pesticides[A]		
in the last month	17%	14%
in the last year	33%	43%
Age of carpet (years)		
median (range)	6 (1–40)	10 (<1–40)
Vacuum <1 time per week, %	24%	21%
Own a dog, %	40%	76%
Own a cat, %	48%	68%
Have doormats, %	68%	80%
Percent of homes <0.5 miles from farm fields	44%	100%

[A]Limited to 12 nonfarm and 21 farm homes that reported having a garden.

analyses were performed using SAS system software, version 8.2 (SAS Institute, Inc., Cary, N.C.).

RESULTS

Air Samples

A total of 99 indoor and 98 outdoor air samples were obtained and analyzed for atrazine, metolachlor, chlorpyrifos, acetochlor, alachlor, and 2,4-D. Eighty-nine percent of the indoor air and 99% of the outdoor air samples were below the LOD for the six pesticides tested. Chlorpyrifos was detected in indoor air samples taken from six farm (range: 0.04–0.23 $\mu g/m^3$) and two nonfarm homes (range: 0.01–0.05 $\mu g/m^3$); acetochlor was detected in one indoor air sample taken from a farm home (0.04 $\mu g/m^3$). All indoor air samples were below the LOD for atrazine, metolachlor, alachlor, and 2,4-D. None of the homes had detectable levels in any of the air samples taken outside the home except for one farm home that had a single sample positive for metolachlor (0.1 $\mu g/m^3$).

Wipe Samples

A total of 203 house and 153 vehicle wipe samples were obtained for the neutral pesticide analysis and 82 house and 48 vehicle wipe samples were obtained for the glyphosate/2,4-D analysis (Table III). A majority of these samples were below the LOD for the pesticides tested. For house wipe samples, atrazine was detected in only a single nonfarm sample, metolachlor in only 4 farm samples, and acetochlor in only 7 farm samples. All house wipe samples were below the LOD for alachlor, glyphosate, and 2,4-D. Chlorpyrifos was the most commonly detected pesticide in both house (F 23% vs. NF 22%) and vehicle wipe samples (F 21% vs. NF 7.9%, odds ratio [OR] = 3.1, 95% confidence interval [CI] = 1.04–9.1). Acetochlor was detected more often in farm vehicle wipe samples (F 13% vs. NF 5.3%) and metolachlor was detected significantly more often in farm vehicle wipe samples (F 12% vs. NF 1.3%, OR = 9.8, 95% CI = 1.1–87). Atrazine and alachlor were rarely detected and glyphosate and 2,4-D were never detected in vehicle wipe samples.

Dust Samples

A total of 295 dust samples (sample mass: range = 0.01–204 g, median = 2.8 g) were obtained from carpet (sample area: range = 0.3–7.4 m^2, median = 1.1 m^2) inside the homes. After adjusting for visit and room, farm homes had a significantly higher geometric mean carpet dust loading than nonfarm homes (F 2.7 vs. NF 1.5 g/m^2, p-value = 0.026). The unadjusted geometric mean concentration (ng/g) of each pesticide sampled in dust was higher in farm homes compared with nonfarm homes, although only significantly for atrazine and metolachlor. The difference, however, becomes even more apparent when standardizing for area sampled (ng/cm^2), due to the fact that farm homes had more dust than nonfarm homes.

A total of 230 dust samples from 20 farm and 19 nonfarm homes were analyzed for atrazine, metolachlor, chlorpyrifos, acetochlor, and alachlor (Table III). Compared with the wipe samples, dust samples were more likely to detect pesticide residues with a majority of the dust samples above the LOD for atrazine, metolachlor, chlorpyrifos, glyphosate, and 2,4-D. A pesticide residue was detected significantly more often in dust samples from farm homes compared with nonfarm homes for atrazine (OR = 9.4, 95% CI = 3.5–25) and metolachlor (OR = 2.1, 95% CI = 1.1–3.9). Detection rates were similar between farm homes and nonfarm homes for chlorpyrifos in dust samples. A total of 65 dust samples from five farm and six nonfarm homes were analyzed for glyphosate and 2,4-D (Table III). Glyphosate was detected marginally more often in dust samples from farm homes, while 2,4-D was detected in every dust sample.

Dust samples were categorized as belonging to a nonfarm home, a farm home that did not apply the pesticide in the 7 days preceding the visit, and a farm home that applied the pesticide in the 7 days preceding the visit. Acetochlor and alachlor were excluded from this analysis since greater than 50% of the dust samples for these pesticides were below the LOD. Geometric means, after adjusting for visit and room, for each of these groups are presented in Table IV. Atrazine and metolachlor were significantly higher in dust from farm homes that reported applying these pesticides in the 7 days preceding the visit compared with farm homes that did not apply these pesticides and nonfarm homes. In addition, the concentration of atrazine in dust was significantly higher in farm homes that did not

TABLE III. Wipe and Dust Samples Greater than or Equal to the Limit of Detection

Sample Type Pesticide	Nonfarm Number of Samples	Nonfarm n > LOD(%)	Nonfarm Range[A]	Farm Number of Samples	Farm n > LOD(%)	Farm Range[A]	Odds Ratio[B] e^{β}	Odds Ratio[B] 95% CI
House wipe								
atrazine	95	1 (1.1%)	160	108	0 (0%)	—	—	—
metolachlor	95	0 (0%)	—	108	4 (3.7%)	0.85–8.5	—	—
chlorpyrifos	95	21 (22%)	0.22–3.8	108	25 (23%)	0.32–25	1.1	0.44–2.8
acetochlor	95	0 (0%)	—	108	7 (6.5%)	0.32–2.5	—	—
alachlor	95	0 (0%)	—	108	0 (0%)	—	—	—
glyphosate	39	0 (0%)	—	43	0 (0%)	—	—	—
2,4-D	39	0 (0%)	—	43	0 (0%)	—	—	—
Vehicle wipe								
atrazine	76	0 (0%)	—	77	3 (3.9%)	38–410	—	—
metolachlor	76	1 (1.3%)	5.2	77	9 (12%)	9.8–680	9.8	1.1–87
chlorpyrifos	76	6 (7.9%)	0.43–11	77	16 (21%)	0.23–9.3	3.1	1.04–9.1
acetochlor	76	4 (5.3%)	1.3–6.2	77	10 (13%)	0.79–39	2.7	0.68–11
alachlor	76	1 (1.3%)	3.3	77	2 (2.6%)	1.2–1.3	2.0	0.19–21
glyphosate	26	0 (0%)	—	22	0 (0%)	—	—	—
2,4-D	26	0 (0%)	—	22	0 (0%)	—	—	—
Dust								
atrazine	114	30 (26%)	0.0017–0.077	116	91 (78%)	0.00039–17	9.4	3.5–25
metolachlor	114	59 (52%)	0.00073–1.3	116	80 (69%)	0.0011–9.8	2.1	1.1–3.9
chlorpyrifos	114	92 (81%)	0.00021–3.6	116	97 (84%)	0.00049–10	1.2	0.37–3.6
acetochlor	114	17 (15%)	0.00054–1.4	116	34 (29%)	0.00086–2.6	2.1	0.84–5.5
alachlor	114	5 (4.4%)	0.00027–0.012	116	12 (10%)	0.00085–0.046	2.3	0.51–11
glyphosate	33	28 (85%)	0.0012–13	31	31 (100%)	0.0081–2.7	—	—
2,4-D	33	33 (100%)	0.0041–1.9	32	32 (100%)	0.00099–5.3	—	—

Notes: Samples from 20 farm and 19 nonfarm homes were analyzed for atrazine, metolachlor, chlorpyrifos, acetochlor, and alachlor. Samples from five farm and six nonfarm homes were analyzed for glyphosate and 2,4-D.

[A] Range of samples greater than or equal to the LOD (ng/cm^2), reported to two significant figures. The ng/cm^2 value for dust was calculated by multiplying the ng/g value reported by the laboratory by the amount (g) of dust collected per cm^2 of carpet sampled.

[B] e^{β} = the odds ratio, defined as the odds of a farm sample being above the LOD divided by the odds of a nonfarm sample being above the LOD, obtained from the GENMOD procedure in SAS assuming an exchangeable correlation matrix.

apply atrazine compared with nonfarm homes. Chlorpyrifos and glyphosate were higher, but not significantly, in dust from farm homes that applied these pesticides in the 7 days preceding the visit compared with farm homes that did not apply them and nonfarm homes. However, there were only two farms that reported having sprayed chlorpyrifos prior to a visit, and one nonfarm, located within a quarter of a mile of a farm and in proximity to a field, had unusually high levels of glyphosate in dust ($n = 7$, GM = 2100 ng/g). If this nonfarm is excluded, then farm homes that sprayed glyphosate within 7 days preceding the visit had significantly greater concentrations of glyphosate in dust than nonfarm homes.

The spray effect analysis for 2,4-D included dust samples when 2,4-D was applied to crops in the 30 days preceding the farm-visit since there were only 2 farm-visits where 2,4-D was applied to crops in the 7 days preceding the visit. 2,4-D levels were similar in dust from farm homes that sprayed 2-4,D in the 30 days preceding the visit compared with farm homes that did not spray 2-4,D. 2,4-D was higher, but not significantly, in farm homes compared with nonfarm homes. Acetochlor was applied to crops at only five farms prior to visits; alachlor was not applied to crops at any of the farms, and since less than 50% of the dust samples were above the analytical LOD for both acetochlor and alachlor, additional analyses were not performed for these pesticides.

The distributions of five of the pesticides in the homes are shown in Table V. In general, for atrazine and metolachlor, the entranceway, father's change area, and laundry room had the highest levels of pesticide in dust for farm homes that sprayed these pesticides within the 7 days preceding sampling, whereas in nonfarms, the entranceway and child's bedroom had the highest pesticide levels in dust. Chlorpyrifos levels in dust were similar in all rooms but highest in the child's bedroom for both farm and nonfarm households. A room effect could not be assessed in farm homes that sprayed chlorpyrifos in the 7 days preceding the visit due to sample size limitations. Glyphosate levels in dust were highest in the child's bedroom for both farm and nonfarm homes, while 2,4-D concentrations in dust

TABLE IV. Dust Sample Results from the Spray Effect Analysis

Pesticide Spray Category	Number of Samples	% > LOD	Pesticide Residue in Dust (ng/g)				Pesticide Residue in Carpet (ng/cm^2)			
			GM	GSD	Adjusted GM[A]	95% CI	GM	GSD	Adjusted GM	95% CI
Atrazine										
nonfarm	114	26	2.3	6.0	2.4[B,C]	1.1–5.1	0.00035	12	0.00035[D,E]	0.00018–0.00068
narm—no spray	58	64	16	11	26[B,F]	11–59	0.0042	16	0.0055[D,G]	0.0025–0.012
narm—spray within 7 days	58	93	170	11	94[C,F]	41–220	0.048	12	0.039[E,G]	0.017–0.087
Metolachlor										
nonfarm	114	52	5.7	14	5.9[H]	3.2–11	0.00088	24	0.00089[I,J]	0.0043–0.0018
farm—no spray	95	64	9.9	13	12[K]	6.0–22	0.0032	23	0.0037[I,L]	0.0017–0.0078
farm—spray within 7 days	21	90	310	20	240[H,K]	69–840	0.042	32	0.043[J,L]	0.0099–0.19
Chlorpyrifos										
nonfarm	114	81	30	8.5	33	14–80	0.0046	14	0.0050	0.0021–0.012
farm—no spray	111	83	39	11	41	17–96	0.011	13	0.012	0.0051–0.029
farm—spray within 7 days	5	100	73	2.1	106	17–680	0.011	4.7	0.021	0.0018–0.23
Glyphosate										
nonfarm	33	85	140	21	110	21–610	0.023	34	0.018	0.0022–0.15
farm—no spray	18	100	920	2.1	1000	140–7400	0.22	2.9	0.28	0.024–3.2
farm—spray within 7 days	13	100	1100	2.4	1300	180–9700	0.33	5.0	0.45	0.039–5.3
2,4-D										
nonfarm	33	100	330	3.3	320	100–1000	0.056	4.9	0.053	0.013–0.22
farm—no spray	20	100	340	2.7	850	240–3100	0.082	3.7	0.20	0.038–1.0
farm—spray within 30 days	12	100	1700	4.0	730	190–2800	0.41	14	0.25	0.045–1.4

Notes: All results reported to two significant figures. Samples reported as below the LOD were assigned half LOD prior to statistical analysis.
[A] Adjusted for visit (visit 1, visit 2) and room (child's bedroom, child's playroom, laundry room, father's change room, and entranceway).
[B,C,D,E,G,H,K,J] Tukey-Kramer adjusted p-value <0.001.
[F,L] Tukey-Kramer adjusted p-value <0.01.
[I] Tukey-Kramer adjusted p-value <0.05. Means with the same letter are significantly different.

were highest in the entranceway for nonfarms and highest in the change area for farms.

After adjustingfor visit, room, and spray status, none of the pesticides were related to any of the household covariates except for atrazine. The concentration of atrazine in dust was significantly higher in farm homes only that reported using an insecticide inside the home within the year prior to sampling after adjusting for visit, room, and spray status. For dust samples from farm households, the effects of agricultural practices on pesticide concentration were also evaluated. However, due to small sample sizes for each pesticide and a lack of variability among some practices, only a limited analysis of atrazine was performed. Atrazine, applied to crops at 16 out of 20 farms in the neutral pesticide analysis, was applied by the farmer at 10 farms and by a custom applicator at 6 farms. Higher atrazine levels in household dust were observed at farm visits where atrazine was applied by the farmer compared to farm visits where atrazine was applied by a custom applicator (adjusted GM 370 vs. 27 ng/g, p-value = 0.0013). Farmers self applying atrazine reported similar spray practices, so it was not possible to perform tests of significance for many of these variables. Higher atrazine levels in household dust were marginally associated with the use of a closed cab; however, after adjusting for the number of acres sprayed, the difference was not significant (adjusted GM closed cab 610 vs. open cab 290 ng/g, p-value = 0.46).

For each dust sample, the pesticide concentration in dust (ng/g) was converted to a pesticide loading on the carpet

TABLE V. Dust Sample Results from the Room Effect Analysis

Pesticide Group	Number of Samples	Room in House (Adjusted GM, ng/g)[A]				
		Child's Bedroom	Child's Playroom	Laundry Room	Father's Change Area	Entranceway
Atrazine						
nonfarm	114	2.7	2.0	1.3[B]	0.85[C]	4.5[B,C]
farm—no spray	58	8.9[D]	15	24	76[D]	35
farm—spray within 7 days	58	100[E]	140	530	740[E]	340
Metolachlor						
nonfarm	114	41[F,G,H]	1.8[F,I]	0.50[G,J]	0.40[H,K]	15[I,J,K]
farm—no spray	95	30[L,M]	6.5	1.4[L,N]	3.0[M]	23[N]
farm—spray within 7 days	21	55	9.2	1200	1400	350
Chlorpyrifos						
nonfarm	114	52[O]	32	33	12[O]	31
farm	116	77[P,Q]	22[P]	39	56	22[Q]
Glyphosate						
nonfarm	33	510	8.6	260	60	260
farm	31	1500[R]	1400[S]	NA	1400	550[R,S]
2,4-D						
nonfarm	33	450	120[T]	83	270[U]	740[T,U]
farm	32	660	610	1300	1600	850

[A] All results reported to two significant figures. Samples reported as below the LOD were assigned half LOD prior to statistical analysis. Geometric mean is adjusted for visit (visits 1, 2). Significance testing was performed within each group.
[B–E,M,N,P,S–U] Tukey-Kramer adjusted p-value <0.05.
[F–H,J,K] Tukey-Kramer adjusted p-value <0.0001.
[I,L,O,Q,R] Tukey-Kramer adjusted p-value <0.01. Means with the same letter are significantly different.
GM = geometric mean; NA = not available.

(ng/cm^2) using the mass and area associated with each sample. The effect of spraying in this analysis was similar to the spray effect in the pesticide concentration in dust analysis (Table IV). The rooms were not equally dusty, however, with the entranceway having the highest amount of dust per unit area compared to the other rooms. Consequently, pesticide loadings on the carpet in the entranceway tend to be higher than loadings from the other rooms. For example, in farm homes that sprayed atrazine in the 7 days preceding the visit, atrazine levels in carpet dust (ng/g) were higher, although not significantly, in the father's change room compared with the entranceway (least squares geometric mean (LSGM) = 740 vs. 340 ng/g, respectively). However, after standardizing to unit area (ng/cm^2), atrazine loadings on the carpet were higher at the entranceway compared to the father's change area (LSGM = 0.59 vs. 0.16 ng/cm^2, respectively).

The within-household (GSD_w) and between-household (GSD_b) variance components expressed as geometric standard deviations for five of the pesticides are provided in Table VI for both pesticide concentration in dust (ng/g) and pesticide concentration in carpet (ng/cm^2) for dust samples from farm households. Variance components, computed after adjusting for visit, room, and spray status, were not markedly changed by the addition of other exposure determinants to the model.

DISCUSSION

While there are a few studies that have investigated the take-home pesticide issue and pesticide home contamination in rural and agricultural environments, most of these studies have examined organophosphate pesticides, while this study looked at several pesticides not generally measured in these previous studies. Chlorpyrifos has been studied frequently and can serve as a benchmark.

In a study conducted by the Centers for Disease Control and Prevention for the Arizona Department of Health Services, dust was collected from 152 homes and 25 schools and tested for the presence of 43 pesticides.[21] Chlorpyrifos had a GM of 113 ng/g. Curl et al.[22] collected 156 house dust samples from farm worker households and found a GM level of 50 ng/g for chlorpyrifos. This compares with chlorpyrifos concentrations of 40 and 30 ng/g for farm and nonfarm homes, respectively, in our study.

Farm homes in this study are clearly more contaminated than nonfarm homes. Other studies have found similar results. Simcox et al.[13] measured pesticide levels in house dust of both farm homes and reference homes and found that farm homes had significantly higher levels of pesticide in dust. Bradman et al.[12] found higher pesticide levels in dust between farm worker homes and nonfarm worker homes.

TABLE VI. Within- and Between-Household Variance Components for Pesticide Levels in Dust Samples Obtained from Farm Households

Sample Type Pesticide	Number of Farms	Number of Samples	Within-household		Between-Household	
			GSD_w^A	%[B]	GSD_b^C	%[B]
Dust (ng/g)						
Atrazine	20	116	4.6	45	5.4	55
Metolachlor	20	116	10.4	81	3.1	19
Chlorpyrifos	20	116	3.8	33	6.5	67
Glyphosate	5	31	1.8	77	1.4	23
2,4-D	5	32	2.1	22	4.2	78
Dust (ng/cm^2)						
Atrazine	20	116	7.2	84	2.4	16
Metolachlor	20	116	16.2	86	3.1	14
Chlorpyrifos	20	116	4.7	37	7.5	63
Glyphosate	5	31	3.2	67	2.2	33
2,4-D	5	32	3.7	45	4.2	55

Note: Variance components were computed using the MIXED procedure in SAS after adjusting for visit, room, and spray status.
[A] GSD_w = estimated geometric standard deviation of the within-household distribution.
[B] Percent of the random effect variance attributable to that source.
[C] GSD_b = estimated geometric standard deviation of the between-household distribution.

Differences in pesticide levels in dust seen between nonfarm homes, farm homes that did not spray the pesticide, and farm homes that did spray the pesticide were greater for the strictly agricultural pesticides (e.g., atrazine and metolachlor) vs. pesticides that have both residential and agricultural uses (chlorpyrifos, glyphosate, and 2,4-D). This would be expected since these latter pesticides are commonly used in residential settings. Chlorpyrifos, glyphosate, and 2,4-D appear to be ubiquitous in both the nonfarm and farm homes. Better than 80% of the dust samples in both farm and nonfarm homes had detectable levels of these pesticides. This finding is comparable to other literature reports. Chlorpyrifos, for example, was detected in 81% of dust samples in Yuma County, Arizona[21] and in 98% and 82% of dust samples from agricultural and nonagricultural families respectively.[13] It is interesting to note that chlorpyrifos was one of the most frequently detected pesticides in the current study but was applied at only two farms.[17]

One potential source of pesticides in farm homes results from farmer take-home mechanism. When atrazine or metolachlor was applied to crops on the farm, concentrations of these pesticides tended to be higher in dust in the entranceway, laundry room, and change room—rooms where dirt would be tracked in or where the farmer's clothes would be deposited. These pesticides have agricultural uses only, and therefore would not be used in or around the house. Chlorpyrifos, glyphosate, and 2,4-D all have residential uses so that contamination may have multiple sources. This is supported by our finding that both farm homes and nonfarm homes have a high percentage of detectable samples for these pesticides and their more even distribution throughout the homes. The higher levels of atrazine and metolachlor in the entranceway, laundry room, and change room would suggest that the farmer is bringing them home on clothing and shoes, supporting the notion of take-home pesticide exposure. Other studies have suggested the take-home pathway as the primary mechanism for contamination of the indoor environmen.[12,13,22–24] Acetochlor, which was sprayed by only a few farmers, and alachlor, which was not sprayed at all, were not detected frequently enough to allow this analysis.

Spray drift as a source of pesticide residue cannot be ruled out. Even though both the indoor and outdoor air samples were largely nondetectable, they were taken a few days after an application, by which time pesticide in the air may have settled out. Koch et al.[25] found that organophosphate metabolite levels in children's urine samples were higher during the spray season in an agricultural region in the absence of parental work contact or residential proximity to treated fields. The authors suggest that spray drift may account for some of the observed increases.

Several factors that were anticipated to be associated with pesticide levels in dust were investigated for their affect on the pesticide levels. Only the use of an insecticide inside a farm home was found to be associated with atrazine in dust. Since atrazine is a herbicide, this association does not make sense and may be spurious, since farm homes that sprayed with an insecticide in the last year were also more likely to have sprayed atrazine prior to both visits. Age of carpet, frequency of carpet vacuuming, the presence of door mats, the age of the homes, and the presence of pets were not associated with pesticide levels in dust.

It is unclear why none of these variables were associated; however, testing for an association between the pesticide level

and some of the household covariates was complicated due to confounding with farm/nonfarm status or confounding with spray status. For example, all farms that sprayed the pesticide may have had "old" carpet. In a simulated pesticide track-in study by Nishioka et al.,[19] 2,4-D levels in dust were lower when a door mat was present. In another study, the presence of a high activity dog was shown to be significantly correlated with 2,4-D levels in indoor house dust.[16] The authors warn though that the sample sizes were small and caution should be exercised in interpreting the results. One reason why the door mats may not have reduced pesticide levels in dust in our study is that they may have acquired a high pesticide and dust loading after only a short time of use, becoming a reservoir for contamination as opposed to an element for reducing contamination. This speculation should be investigated further. In the case of dog activity, dogs on the farms were outdoor dogs only. In only one case in the farm homes with dogs did the dog spend time both indoors and outdoors.

Distance to a treated field did not correlate with pesticide levels in dust in nonfarm homes. Distance to a treated field was not analyzed for the farm homes, since all farm homes were reported to be within a quarter of a mile of a treated field. It may be that the distance categories (<1/4 mile, 1/4 mile to 1/2 mile, 1/2 mile to 3/4 mile, 3/4 mile to 1 mile, >1 mile) may not have permitted detection of differences. Simcox et al.[13] found that pesticide levels in dust decreased with increasing distance, but considered much smaller distances (<50 ft, 50–200 ft, >200 ft). In Yuma County, Arizona, however, although an association between pesticide levels in dust and proximity to treated fields was not investigated, the authors did investigate urine metabolite levels and proximity to a treated field and did not find an association.[21]

Most of the analyses in this article focused on the pesticide concentrations in dust. The wipe and air samples were not a particularly useful sample media for evaluation of low-level pesticides in homes in this study. This may largely be the result of higher LODs for the wipe and air samples in this study. Indeed, the dust sample values are orders of magnitude lower than the wipe values, which are likely due to the better limits of detection for the dust analysis. Another factor may be the sampling method, particularly the use of polyurethane foam for the wipe samples for 2,4-D and glyphosate. Of the dust samples analyzed for 2,4-D and glyphosate, 100% and 94% had detectable levels of 2,4-D and glyphosate, respectively. These pesticides were not detected in any wipe sample.

One apparent problem with using PUF for wipe sampling is that PUF does not hold liquid very well. The amount of isopropanol added to the PUF had to be reduced to 6 mL from the 10 mL added to the SOF-WICK sponges. Even so, the isopropanol would run off the PUF, leaving it fairly dry when wiping. Because of this, it is likely that not as much pesticide residue would be picked up from the surface. Further investigation is needed to confirm this assumption.

There are a few limitations to the analyses. Chlorpyrifos was not sprayed very often, so it is difficult to draw conclusions about the spray effect for chlorpyrifos. In the glyphosate/2,4-D analysis, there were only five farms and six nonfarms available for the analysis. As a result of the small number of homes, the differences seen were not statistically significant. Only the acid form of 2,4-D was analyzed in the samples. In some farm-homes the ester form of 2,4-D may have been applied resulting in an underestimate of 2,4-D contamination. Testing some of the household covariates for a relationship with pesticides was difficult due to confounding with farm/nonfarm status or confounding with spray status. The LODs for the wipe and air samples are substantially higher than the LODs for the dust samples, making it difficult to compare the sample media. Lastly, for the dust data analysis, we considered the effect of spraying in the 7-day period preceding the visit. The choice of 7 days, although somewhat arbitrary, was intended to focus on more recent pesticide applications.

CONCLUSION

Farm homes have more pesticide residue inside than non-farm homes, and farms that spray a particular pesticide are more likely to have higher levels of that pesticide inside the home than other homes. This is particularly apparent for the strictly agricultural herbicides atrazine and metolachlor. While these herbicides are not applied in or around the home, they appear to be getting into the home paraoccupationally. It appears for agricultural pesticides that take-home exposure may be an important source of home contamination.

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