Pesticides in Household Dust and Soil: Exposure Pathways for Children of Agricultural Families

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Children of agricultural families are likely to be exposed to agricultural chemicals, even if they are not involved in farm activities. This study was designed to determine whether such children are exposed to higher levels of pesticides than children whose parents are not involved in agriculture and whose homes are not close to farms. Household dust and soil samples were collected in children's play areas from 59 residences in eastern Washington State (26 farming, 22 farmworker, and 11 nonfarming families). The majority of the farm families lived within 200 feet of an operating apple or pear orchard, whereas all reference homes were located at least a quarter of a mile from an orchard. Four organophosphorous (OP) insecticides commonly used on tree fruit were targeted for analysis: azinphosmethyl, chlorpyrifos, parathion, and phosmet. Samples were extracted and analyzed by gas chromatography/mass selective detection. Pesticide concentrations in household dust were significantly higher than in soil for all groups. OP levels for farmer/farmworker families ranged from nondetectable to 930 ng/g in soil (0.93 ppm) and from nondetectable to 17,000 ng/g in dust (17 ppm); all four OP compounds were found in 62% of household dust samples, and two-thirds of the farm homes contained at least one OP above 1000 ng/g. Residues were found less frequently in reference homes, and all levels were below 1000 ng/g. Household dust concentrations for all four target compounds were significantly lower in reference homes when compared to farmer/farmworker homes (Mann-Whitney U test; $p < 0.05$). These results demonstrate that children of agricultural families have a higher potential for exposure to OP pesticides than children of nonfarm families in this region. Measureable residues of a toxicity I compound registered exclusively for agricultural use (azinphosmethyl) were found in household dust samples from all study homes, suggesting that low-level exposure to such chemicals occurs throughout the region. Children's total and cumulative exposure to this pesticide class from household dust, soil, and other sources warrants further investigation. *Key words*: agriculture, azinphosmethyl, children, chlorpyrifos, household dust, insecticides, organophosphates, parathion, pesticides, phosmet, soil. *Environ Health Perspect* 103:1126–1134 (1995)

Concern about residential pesticide exposures among children has increased recently with the reported associations between residential pesticide use and childhood leukemia (*1–3*). Substantial research has focused on pesticide exposure after indoor and lawn applications (*4–8*), and a recent study demonstrated that individuals who contact treated indoor surfaces can absorb measurable amounts of the compound through the skin (*9*). In cases of residential misapplication, exposures have resulted in pesticide-related illnesses (*10,11*). Studies designed to characterize children's exposure to pesticides in the general population indicate that the largest number of pesticides and the highest concentrations are found in household dust compared to air, soil, and food (*12,13*). However, few of these studies have been conducted in or near agricultural regions, where one might expect relatively higher exposures for residents due to both residential and agricultural pesticide use.

Children of farmers and agricultural field workers are likely to have a high potential for pesticide exposure, even if they are not involved in farm activities related to exposure. Pesticide exposure could occur from a number of sources such as contaminated soil, dust, work clothing, water, and food, or through drift, the deposition of a pesticide off target. In many agricultural communities, residential home sites are close to or surrounded by fields or orchards. Pesticides can be tracked into the home on shoes or by pets and become part of a household dust "reservoir." Pesticide residues in indoor environments are not subject to degradative environmental processes such as sun, rain, and soil microbial activity, and may thus persist longer in the house than in outdoor soil.

Household dust and yard soil are considered significant sources of exposure to pesticide residues and other toxicants for small children and toddlers (*13*). Young children spend a large portion of their time on the floor or ground and can easily come in direct contact with yard soil or dust by putting hands and objects in their mouths frequently and thereby ingesting soil or dust. Studies using tracer elements to quantify soil ingestion have estimated that children in the United States can ingest from 10 to 1300 mg of soil/day; in children with a pica history the level can reach 5000 mg/day (*14–17*). EPA investigators estimated the potential health risks to children for the soil and dust pathway to be 12 times that of adults (*18*).

Government reporting of pesticide poisoning cases is one indicator of the hazards or risks associated with pesticide use on the farm or in the home. In 1991, 39% of pesticide incidents reported to all agencies in Washington State were agriculturally related (*19*). One case that demonstrates the potentially serious nature of post-application exposures involved a 20-month-old child who developed acute poisoning from ingesting ethyl parathion-contaminated soil. However, present reporting data do not allow assessment of the overall prevalence or severity of chronic exposures to pesticides for children in agricultural settings. Reliance on such statistics is limited by at least three factors: 1) reported cases generally involve only acute intoxications (subacute or chronic effects are likely to remain unreported), 2) even acute cases may not be recognized or reported consistently by physicians as pesticide related, and 3) cases tend to provide little information for exposure mitigation. Thus, properly focused environmental sampling represents a more reliable and preventive approach for investigating public health concerns related to children's exposure to pesticides in agricultural and residential settings.

Organochlorine and arsenical compounds were the first pesticide classes studied in the home environment, due primarily to their widespread use, persistence, and chronic health effects (*20–23*). However, during the past 20 years there has been a dramatic increase in the use of less persis-

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tent but more acutely toxic organophosphorus (OP) pesticides. Acute effects of OP exposure are well known, but chronic effects are not well characterized, and available information pertains primarily to adults (*24–28*). Thus, major gaps exist in our knowledge of the health effects of chronic pesticide exposure in children (*29*). No published studies have examined the neurotoxic effects of low-level pesticide exposure to children.

The primary objective of this study was to evaluate the potential for chronic exposures of children to pesticides in and around the homes of farmers and agricultural workers. The study had two specific aims: to determine to what extent household dust and surface soil from children's play areas contain agricultural pesticides, and to determine if children of agricultural families live in homes that contain higher levels of pesticides than homes of nonfarm children. An attempt was also made to identify risk factors for elevated residential pesticide levels in the study population.

Methods

Study design. This study employed a cross-sectional environmental sampling strategy during the 1992 pesticide spray season. Targeted residences were those of agricultural families, including both farmers and nonseasonal farmworkers, and nonagricultural reference families. Sampling goals were to collect household dust using a vacuum sampler from carpeted entryways and indoor play areas and to collect surface soil from outdoor play areas at each residence. The greater Wenatchee area in eastern Washington State was chosen for study because its residents are engaged predominantly in agricultural production of tree fruits, including apples, pears, and cherries.

Four OP pesticides commonly used during the spray season were targeted for analysis: azinphosmethyl [*O,O*-dimethyl *S*-(4-oxo-1,2,3-benzotriazin-3(4H)-ylmethyl)-phosphorodithioate (CAS no. 86-50-0)], phosmet [*N*-(mercaptomethyl)-phthalimide *S*-(*O,O*-dimethylphosphorodithioate (CAS no. 732-11-6)], chlorpyrifos [*O,O*-diethyl *O*-(3,5,6-trichloro-2-pyridyl) phosphorothioate (CAS no. 2921-88-2)], and ethyl parathion [*O,O*-diethyl *O*-*p*-nitrophenyl phosphorothioate (CAS no. 56-38-2)]. These pesticides were identified as the most commonly used OPs for apple production. Parathion registration was canceled for use in orchards in 1991 by the U.S. Environmental Protection Agency due to its high acute toxicity and the frequency of reported poisonings nationwide, but continued use of existing stock was allowed through the 1992 spray season.

Recruitment. Participating families were recruited from Chelan and Douglas counties with the assistance of several commercial and social service organizations. Service organizations recruited farmworkers by mailing letters to their members that described the study and asked interested families to contact the organization or the university directly. Reference families were also recruited using these procedures; several of the reference families included an employee of the service organizations. Farmers were sent a similar letter through the Washington Growers Clearinghouse Association. When a positive response was received, the family was contacted by phone and screened for eligibility. All procedures involving human subjects were reviewed and were approved by the University of Washington Human Subjects Review Committee before the study began.

Farmer and farmworker family selection was based on the following eligibility criteria: at least one child between the ages of 1 and 6 years and at least one family member living in the home employed as an orchardist, fieldworker, and/or pesticide applicator. Reference family eligibility factors were: no family member working in the farm industry, no family member having direct contact with agricultural pesticides, and the residence situated more than one-quarter mile from a commercial orchard or crop. Although most farmer and reference families were of Caucasian background, the majority of farmworkers were Hispanic.

Soil sampling and analysis. Participating families were asked to identify their children's outdoor play areas, including sandboxes, front and back lawns, and driveways. Five locations within these designated play areas were chosen for sampling. A 26 cm × 26 cm template was placed on the ground, and the top 0.5–1 cm soil layer was scraped with the edge of a 5-inch stainless-steel spatula. The five samples were composited for each home, transported on dry ice, and stored at -20°C. Samples were analyzed within 12 months of collection.

Samples were thawed to room temperature and sieved through a 425-µm stainless mesh to remove large nonsoil debris. Wet samples were dried in a desiccator for 5–16 hr. A portion of each sieved sample was submitted to the University of Washington Forest Research Laboratory for determination of moisture content. All samples contained <10% moisture at the time of extraction.

A sonication method was adapted from Nigg (*30*) and is described in detail elsewhere (*31*). Five-gram soil samples were pre-wet with 400 µl distilled water and refrigerated at 4°C for 15–18 hr. We added 50 ml acetone and sonicated the soil at 20 kHz for 1 min in an ultrasonic processor with a 0.5-inch tapped horn (Heat Systems-Ultrasonics, Inc., Farmingdale, New York). The clear supernatants were separated from soil solids and evaporated to near dryness under a purified nitrogen stream and then partitioned between hexane (2 ml) and water (40 ml). The hexane layer was separated and dried over anhydrous sodium sulfate.

We prepared standard OP solutions at 1 mg/ml of each analyte in acetone using neat materials (≥98% purity) purchased from Chem Service (West Chester, Pennsylvania). Further dilutions were made in hexane to prepare OP calibrant solutions. We used 1 ng/ml tributylphosphate as a GC internal standard in all samples. Quantification of the target OPs was performed by GC/mass selective detection (MSD), in selected ion monitoring mode using a Hewlett-Packard gas chromatograph 5890A series II equipped with 5971 mass selective detector and a 15-m × 0.25-mm i.d. J&W capillary column with 0.25 µm DB-1701 bonded phase. Selected ions were acquired for each analyte; two confirmation masses, and one mass (typically the most abundant in that compound's electron impact mass spectrum) for quantitation.

We determined the analytical limit of detection (LOD) by running analytical standards in solvent (no matrix effect). The method limit of quantitation (MLOQ) was determined by running analytical standards in a soil extract (matrix effect). Relative ion intensities and simultaneity were used to confirm each positive detection. Samples with quantitation ion response, but without qualifier ion response were defined as having concentrations below the MLOQ. Samples with no ion response were designated as below the limit of detection. These limits are specified in Table 1. In most cases the LOD and MLOQ were similar. Extraction of the OP compounds from soil was virtually complete, with extraction efficiencies ranging from 90% to 110%. Final OP concentration results were reported as nanograms of pesticide per gram soil, without correcting for the minimal moisture content of the soil.

Household dust sampling and analysis. Household dust was collected using the high-volume, small-surface sampler (HVS-3; Cascade Stamp Sampling Systems, Bend, Oregon) from two carpeted or rug-covered areas in each home: 1) 3 ft inside the main entryway, and 2) in an area where children commonly played. The HVS-3 is a cyclone-equipped vacuum sampler developed for U.S. EPA, which collects small

Table 1. Instrument limits of detection (LOD), method limits of quantitation (MLOQ), and extraction efficiencies for analysis of targeted organophosphorus insecticides in soil and household dust by GC/mass selective detector[a]

	LOD[b] (ng/ml)		MLOQ[c] (ng/g)		Extraction efficiency (%)[d]	
Insecticide	Soil	Dust	Soil	Dust	Soil	Dust
Azinphosmethyl	11	16	32	40	90 (10)	77 (17)
Chlorpyrifos	13	20	11	17	92 (9)	72 (14)
Phosmet	11	10	7	12	98 (11)	73 (8)
Ethyl parathion	13	16	34	11	110 (14)	106 (20)

[a]Instrument type: HP 5890A series II, with mass spectrum detector, in selected ion mode.
[b]Instrument LOD determined with analytical standards in solvent (no matrix effect); determined separately under instrument conditions used for analyzing soil and conditions for dust.
[c]MLOQ determined by spiking soil or dust extracts to account for matrix effects.
[d]Values are means with SDs in parentheses. For soil, n = 12: six samples fortified with organophosphorus mix at 100 ng/g soil and six samples at 500 ng/g soil. For dust, n = 7: four samples fortified with organophosphorus mix at 250 ng/g dust and three samples at 650 ng/g dust.

particles (>5 μm) in a teflon catch bottle (*32*). A measured area on the rug or carpet was sampled according to standard procedures described in the HVS-3 operation manual, with a target sample weight of 5 g. Samples were transported on dry ice and stored at -20°C and analyzed within 12 months of collection.

Samples were sieved through a 150-μm stainless mesh to remove large nondust debris, hair, and carpet fibers, and to yield the smaller-diameter particles shown to adhere more readily to the hands (*33*). Analyzing solvent-extracted dust proved to be much more difficult than analyzing soil, due in part to analytical interference by waxy substances and other organic components of the dust. Procedures used for dust were modifications of those described above for soil, with the addition of a filtration step and a gel permeation chromatography (GPC) clean-up procedure before GC/MSD analysis.

We pooled the two sieved dust samples from each house and sonicated 2.5 g portions in 50 ml of acetone for 1 min. Acetone extracts were concentrated under a purified nitrogen stream, solvent exchanged into cyclohexane, and filtered through 0.45-μm polytetrafluoroethylene membrane filters (Gelman Sciences, Ann Arbor, Michigan) to remove fine dust particles and precipitate. The resultant 1.5 ml cyclohexane extracts were applied to a 20-cm × 2-cm i.d. GPC column (Bio-Beads S-X3, Bio-Rad Laboratories, Richmond, California) and eluted with cyclohexane. After discarding an initial volume of 48–52 ml (depending on column), 230 ml of eluant was collected, concentrated using Kuderna-Danish flasks with Snyder columns over a hot water bath, and evaporated to 2 ml under a purified nitrogen stream. The analysis of target OPs in household dust was performed by GC/MS as described for soil, again using tributylphosphate as an internal standard and with standard OP calibrant solutions diluted in cyclohexane. The LOD and MLOQ concentrations for dust were similar and did not differ greatly from those for soil, as indicated in Table 1. Extraction of ethyl parathion from dust was complete, but for the other three OP compounds extraction efficiencies ranged from 72% to 77%. Final OP concentrations were adjusted by these values.

Quality assurance. Blank samples were prepared from solvent-rinsed laboratory-grade sand, carried into the field on each day of sampling, and processed along with the field samples. No targeted analytes were detected in the 19 field blanks (16% of field samples). Field spike samples were prepared by spiking the same sand with the target OP compounds. Samples were carried into the field on each day of sampling and processed with field samples. Results were inconsistent, ranging from 15% to 83% recovery of target analytes. Sand was used in the absence of a standard "clean" dust or soil medium at the time of the field study. It is unclear whether results from the spiked sand samples are due to pesticide instability in storage or use of this particular spiking medium. As there was doubt that sand was a representative matrix, field sample results were not adjusted by field spike recoveries. Further work on storage stability of these types of samples is needed. Reagent blanks were included during the extraction and analysis procedures; no targeted analytes were detected (n = 3, or 2.5% of field samples).

Participant interviews. Participants were asked about occupational pesticide use, frequency of both residential and agricultural pesticide use in and around the home during the past 6 months, and proximity of their homes to orchards. Pesticide registration numbers were collected whenever possible for verifying the active ingredient for home pesticide products. Family members who reported applying pesticides were asked about their personal protective equipment use and laundering of work clothes. Additional questions gathered information about vacuuming frequency, number of days since last vacuum cleaning, routine removal of shoes at the door, use of door mats, and presence of an indoor/outdoor pet. The survey instruments used for this study were largely adapted from EPA's Nonoccupational Pesticide Exposure Study (*12*) and the National Cancer Institute/EPA Farm Occupational Exposure Study (*34*). Interviews were conducted in Spanish when appropriate.

Statistical methods. Median values were lower than mean values in nearly all cases, suggesting skewed distribution of the residue data. Log_{10} transformation yielded approximately log-normal distributions in some but not in all groups. Therefore, nonparametric statistical tests were used to analyze the data whenever possible, including the Wilcoxon Signed-Rank, Mann-Whitney U, Kruskal-Wallis, and Spearman Rank Correlation tests. Analysis of variance tests were performed on some of the log_{10}-transformed data. Concentrations which fell below the method limit of quantitation (<MLOQ) were assigned one-half the MLOQ for statistical purposes.

Results

Families recruited and sampled included 26 farming families, 22 farmworker families, and 11 reference families. The average age of the farmers and farmworkers was 33 years; all had at least one young child (1–6 years). The average number of persons per household employed in the tree fruit industry was 1.0 for farming families and 1.8 for farmworker families.

Pesticide Use

Participants in the farmer study group who owned and/or managed orchards (23 of the 26 farming families) were surveyed regarding the use of pesticides during the 1992 spray season (January 1–July 1): 91% (21/23) reported using at least one of the target OP compounds, and 65% (15/23) reported the use of more than one target OP compound. Azinphosmethyl was the most commonly used OP, reported by 83% (19/23) of respondents. Chlorpyrifos was used by 57% (13/23), phosmet by 22% (5/23), and parathion use was reported by only 1 responding farmer (4%) during the 1992 spray season. Azinphosmethyl was the OP most recently sprayed, with applications ranging from 1 to 3 weeks before sampling, phosmet was used 1–4 weeks before sampling, chlor-

pyrifos 2–3 months before sampling, and parathion use was reported several months prior to sampling.

Of the 28 agriculturally employed study subjects who reported direct involvement with pesticide application, all but one (97%) reported using some personal protective equipment when applying OP pesticides, including rain suits, gloves, boots, and ventilated spray helmets with face shields. Eighty-two percent (23/28) reported leaving protective equipment outside the home, usually in a barn or shed. Eighty-nine percent (25/28) reported washing work clothes worn beneath protective equipment (jeans, shirts) after each pesticide application.

Analysis of the active ingredients reported by homeowners who used pesticide products in the home or on their lawn indicated that residues in soil and household dust samples were due primarily to agricultural use and not to home use of pesticides. One reference family reported application of chlorpyrifos to their lawn by a professional service 1 month before sampling. Soil from this reference home had a greater chlorpyrifos concentration (39 ng/g) than those found in the majority of agricultural family homes.

Soil and Household Dust

Table 2 provides the mean, median, range, and frequency of detection of each compound from soil samples by study group. A large fraction of samples had nondetectable levels (<LOD) of one or more of the targeted pesticides; many additional samples exhibited some ion response, but were below the <MLOQ. As stated previously, all such samples were assigned a value of one-half the MLOQ for statistical purposes. In soil samples from farmer/farmworker families (henceforth called Ag families), levels of the four target insecticides ranged from nondetectable to 930 ng/g, with one or more target compounds found in 58% of soils. For reference homes, residues in soil ranged from nondetectable to 39 ng/g, exceeding the MLOQ only twice (two homes had quantifiable levels of chlorpyrifos).

Household dust sampling results are presented in Table 3. In Ag family homes, levels of the four target analytes ranged from nondetectable to 17,100 ng/g. All four targeted insecticides were found in quantifiable levels in 62% of these homes (30/48). Two-thirds of the homes (32/48) had concentrations >1000 ng/g (>1 ppm) for one or more of the target compounds. Azinphosmethyl was quantified in 100% of the dust samples from agricultural residences. For reference families, OP concentrations ranged from nondetectable to 820 ng/g. Only one sample contained all four target analytes. Azinphosmethyl and phosmet were quantified in all reference household dust samples.

Median household dust levels of the target analytes were 17–100 times higher than soil levels, whether looking at the paired results from all study families or from the Ag families alone (Wilcoxon Signed-Rank test: $p<0.0001$). The box plots in Figure 1 indicate the distribution of pesticide concentrations in soil and dust samples from Ag families. Despite the high numbers of nondetectable residues in soil, paired outdoor (soil) and indoor (dust) values for the Ag families were significantly correlated for all pesticides (Spearman's rank correlation test; see Table 4). For reference families a significant correlation was observed for parathion only.

Table 2. Organophosphorus pesticide concentrations in soil (ng/gm)[a]

Pesticide	Ag families[b] (*n* = 48)	Reference families (*n* = 11)	Ag families: Farmers (*n* = 26)	Ag families: Farmworkers (*n* = 22)
Azinphosmethyl				
Mean	60	<32	84	<32
Median	<32*	<32*	<32	<32
Range	ND–814	ND–<32	ND–814	ND–172
Frequency (%)[c]	20 (42)	0 (0)	13 (50)	7 (32)
Phosmet				
Mean	26	<7	38	11
Median	<7	<7	<7	<7
Range	ND–332	ND–<7	ND–332	ND–101
Frequency (%)	8 (17)	0 (0)	5 (19)	3 (14)
Chlorpyrifos				
Mean	17	11	18	14
Median	<11	<11	<11	<11
Range	ND–234	ND–39	ND–234	ND–152
Frequency (%)	11 (23)	2 (18)	6 (23)	5 (23)
Ethyl parathion				
Mean	<34	<34	46	<34
Median	<34	<34	<34	<34
Range	ND–932	ND–<34	ND–932	ND–<34
Frequency (%)[c]	1 (2)	0 (0)	1 (4)	0 (0)

[a]Method limits of quantitation (MLOQ) in soil (ng/g): azinphosmethyl, 32; phosmet, 7; chlorpyrifos, 11; parathion, 34; ND, nondetectable; values <MLOQ assigned one-half MLOQ for statistical analysis.
[b]Ag families group combines the data from the farmers and farmworkers groups.
[c]Frequency = number of families with quantifiable sample concentrations (>MLOQ); percentages in parentheses.
*Significantly different concentrations; Wilcoxon signed-rank test, $p = 0.04$.

Agricultural and Reference Family Comparisons

A comparison of OP pesticide concentrations in household dust for Ag and reference families indicated that Ag families had significantly higher concentrations of azinphosmethyl ($p = 0.001$), chlorpyrifos ($p = 0.01$), and parathion ($p = 0.02$) (Mann-Whitney *U* test). Phosmet levels also appeared to be elevated ($p = 0.07$). Median values for azinphosmethyl, phosmet, and chlorpyrifos were 3–5 times higher, while parathion was 13 times greater. A significant difference in pesticides levels between soil samples from agricultural and reference homes was apparent only for azinphosmethyl (Wilcoxon Signed-Rank test: $p = 0.04$). This compound was used in many orchards 1–3 weeks before the sampling period.

Occupational Comparisons within Agricultural Family Groups

Median household dust concentrations for the Ag family groups tended to be higher in homes of farmers than in homes of farmworkers for azinphosmethyl, chlorpyrifos, and parathion, but levels were higher for phosmet in the farmworker homes (Table 3). However, differences between the two groups were statistically significant only for parathion (Mann-Whitney *U* test: $p = 0.0007$). Ag families were also grouped as "applicators" or "nonapplicators," based on reported direct handling of OP pesticides. Median dust concentrations were significantly higher in homes of applicators versus nonapplicators for chlorpyrifos and parathion (Mann-Whitney *U* test: $p = 0.02$ and $p = 0.0003$, respectively). Azinphosmethyl levels also tended to be higher for the applicators, but phosmet levels were similar across these two groupings.

A 2 × 2 contingency analysis was performed to test the null hypothesis that these two methods of occupational classification were independent: farmer/farmworker ($n = 26$ and $n = 22$); applicator/nonapplicator ($n = 28$ and $n = 20$). Results indicated a statistically significant association between the

Table 3. Organophosphorus pesticide concentrations in household dust (ng/g)[a]

Pesticide	Ag families[b] (n = 48)	Reference families (n = 11)	Farmers (n = 26)	Farmworkers (n = 22)	Applicators (n = 28)[c]	Nonapplicators (n = 20)
Azinphosmethyl						
Mean	1870	330	2090	1620	1955	1758
Median	1100*	283*	1320	951	1225	769
Range	170–11,270	134–816	171–6520	180–11,270	171–6520	179–11,270
Frequency (%)[d]	48 (100)	11 (100)	26 (100)	22 (100)	28 (100)	20 (100)
Phosmet						
Mean	2080	227	1700	2540	2108	2137
Median	519	185	415	519	523	523
Range	<12–17,100	73–658	<12–14,500	19–17,100	6–17,100	6–14,496
Frequency (%)	46 (96)	11 (100)	24 (92)	22 (100)	27 (96)	19 (95)
Chlorpyrifos						
Mean	429	168	506	338	514	318
Median	267*	53*	372	172	395‡	156‡
Range	<17–3585	<17–483	<17–3585	40–2180	8–3585	40–2182
Frequency (%)	47 (98)	9 (82)	25 (96)	22 (100)	27 (96)	20 (100)
Ethyl parathion						
Mean	365	76	591	98	516	161
Median	154*	<11*	310†	20†	273‡	<11‡
Range	<11–2786	<11–425	<11–2786	<11–440	<11–2786	<11–1847
Frequency (%)	33 (69)	3 (27)	22 (85)	11 (50)	25 (89)	9 (45)

[a]Method limits of quantitation (MLOQ) in dust (ng/g): azinphosmethyl, 40; phosmet, 12; chlorpyrifos, 17; parathion, 11; values <MLOQ assigned one-half MLOQ for statistical analysis.
[b]Ag families group combines the data from the farmers and farmworkers groups.
[c]Applicators and nonapplicators are groups within the Ag family group, based on whether orchard workers were engaged in pesticide handling (mixing, loading, application).
[d]Frequency = number of families with quantifiable sample concentrations (>MLOQ); percentages in parentheses.
*Signficant difference across groups: azinphosmethyl, $p = 0.001$; chlorpyrifos, $p = 0.01$; parathion, $p = 0.02$ (Mann-Whitney U test). †Signficant difference across groups: parathion, $p = 0.0007$ (Mann-Whitney U test). ‡Signficant difference across groups: chlorpyrifos, $p = 0.02$; parathion, $p = 0.0003$ (Mann-Whitney U test).

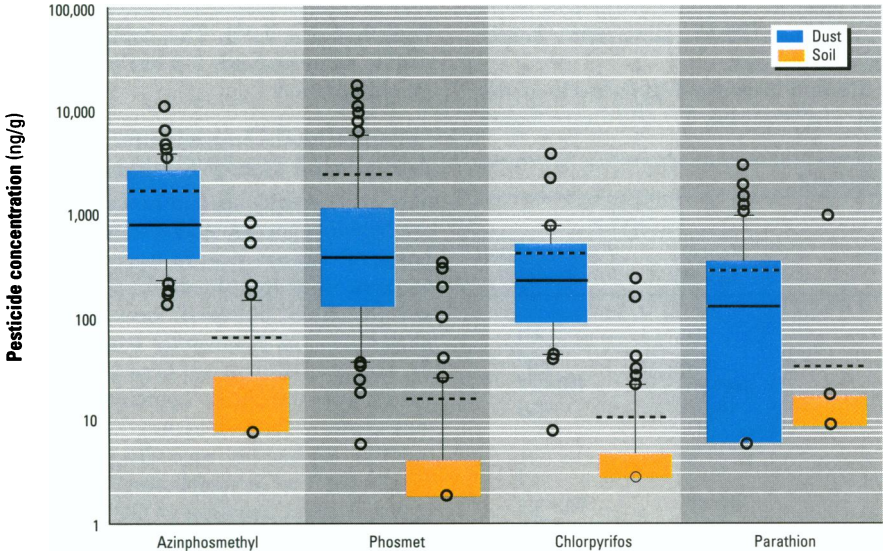

Figure 1. Box plots comparing organophosphorus pesticide concentrations in soil and household dust samples from agricultural families (farmers and farmworkers), plotted on a $\log_{10}$ scale. From the bottom to the top, the box lines in the figure represent 10th, 25th, 50th, 75th, and 90th percentiles, respectively. Circles represent outliers, and the horizontal dotted lines represent the mean concentration.

Table 4. Spearman rank correlation coefficients (r) between household dust and soil organophosphate concentrations

	Ag families (n = 48)		Reference families (n = 11)	
Pesticide	r	p	r	p
Azinphosmethyl	0.49	0.001	0.05	0.87
Phosmet	0.67	<0.0001	0.23	0.48
Chlorpyrifos	0.52	0.0003	0.40	0.21
Ethyl parathion	0.35	0.02	0.81	0.01

two grouping variables, with 73% of farmers categorized as pesticide applicators and 59% of farmworkers categorized as nonapplicators (chi-square test: $p = 0.02$).

Orchard Proximity

Ag family respondents categorized the proximity of their homes to any commercial orchards as <50, 50–200, or >200 ft. Thirty-three of 48 Ag families lived within 50 ft of an orchard, 7 families lived between 50 and 200 ft, and 8 families lived more than 200 ft from an orchard. By definition, all of the 11 reference homes were >1/4 mile from a commercial orchard. Nonparametric analysis of variance of Ag family data revealed a tendency for median OP concentrations in dust to decrease with increasing distance from an orchard. However, a significant difference was seen across the three proximity categories only for parathion (Kruskal-Wallis: $p = 0.005$). Due to the small numbers of subjects in the 50–200 ft and >200 ft groups, these groups were combined into a category of >50 ft from an orchard and compared again to homes <50 ft from an orchard. The box plots in Figure 2 show this distribution of OP household dust concentrations from Ag family homes with respect to proximity. Mean and median levels were higher in the proximate group for all four OP compounds, with significant differences observed for azinphosmethyl and parathion (Mann-Whitney U test: $p = 0.04$ and 0.005, respectively). Including the reference family data in this analysis strengthened the trend, with OP concentrations decreasing at increasing distance from an

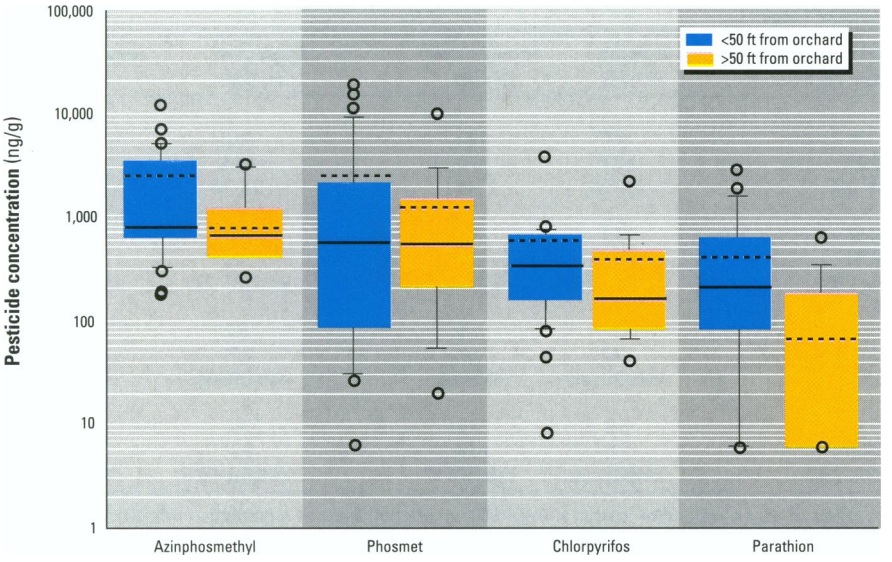

Figure 2. Box plots of organophosphorus pesticide concentration in agricultural family household dust samples comparing groups whose homes are <50 ft or >50 ft from a commercial orchard, plotted on a $\log_{10}$ scale. From the bottom to the top, the box lines in the figure represent 10th, 25th, 50th, 75th, and 90th percentiles, respectively. Circles represent outliers, and the horizontal dotted lines represent the mean concentration.

orchard for azinphosmethyl, chlorpyrifos, and parathion (Kruskal-Wallis: p = 0.0001, 0.02, and 0.001, respectively).

The eight Ag families who lived more than 200 ft from an orchard were distributed unevenly across the groups tested above. To determine if nonproximity to orchards confounded these analyses, tests for significant differences between farmers/farmworkers and applicators/nonapplicators were repeated excluding those eight families, but the outcome of the analyses were unchanged.

Further analysis was performed to determine if an association existed between proximity to orchards in categories of <50 ft (n = 32) and >50 ft (n = 15), and the occupational classifications of farmer (n = 26) and farmworker (n = 22). A significant association was observed between the two grouping variables (chi-square: p = 0.04), with 65% of those living <50 ft of an orchard categorized as farmers and 67% of those living >50 ft categorized as farmworkers. As indicated above, occupation and pesticide application activities were also interrelated grouping variables. However, an additional analysis of these variables demonstrated that pesticide application activity and homesite orchard proximity were not associated groupings (chi-square: p>0.05).

Analyses of variance were performed to determine which one or combination of these three interrelated variables might best explain the variability in household dust OP concentrations for Ag families: proximity (<50 ft or >50 ft), occupation (farmer or farmworker), and applicator or nonapplicator status. One-way analysis of variance (ANOVA) of $\log_{10}$-transformed data revealed significant differences between the categories of all three variables for parathion (p <0.001 for proximity, occupation, and applicator status). No statistical differences were seen between categories of these variables for the other pesticides. Two-way ANOVAs with parathion concentrations of dust showed that the variables "proximity" and "applicator status" were not interactive and that each explained a significant component of variability in OP dust levels between the groups (proximity: p = 0.002, applicator: p = 0.004, proximity*applicator: p = 0.82). When two-way ANOVAs included the variable "occupation," the difference in levels of OPs between farmers and farmworkers varied whether looking at applicators or nonapplicator status, or living <50 ft or >50 ft from an orchard; i.e., when the occupation was paired with either applicator status or proximity, there was interaction, and the variables could not be considered independent in predicting OP household dust level.

Surface Loading and Track-in

Surface loading levels are defined as mass per unit surface area, in this case micrograms of OP pesticide per square meter of carpet. On average, a larger surface was sampled in the reference family homes than in the Ag family homes (6.1 m^2 vs. 4.1 m^2), suggesting differences in dust concentrations. Average (± SD) dust loadings across the three study groups were 8.2 ± 6.4 µg/m^2 for farmer, 14.9 ± 13.4 µg/m^2 for farmworker, and 4.4 ± 2.9 µg/m^2 for reference families. OP loading levels are summarized in Table 5. Loading levels across groups follow the same patterns as described previously for OP concentrations in household dust. Ag families were again divided into applicators and nonapplicators to determine if mass loading levels differed between the two groups. Significant differences between the two groups were observed for chlorpyrifos and parathion (applicators>nonapplicators; Mann-Whitney U Test: p = 0.04, and p = 0.002, respectively).

Questions pertaining to variables affecting pesticide loading in homes, including track-in behavior, cleaning activities, and orchard proximity, were answered as indicated in Table 6. No significant differences in OP loading levels were found for any of these questionnaire variables, even after adjusting for the number of days since participants had last vacuumed (Mann-Whitney U test: p>0.05). Multiple regression analysis of these variables also failed to show any significant relationships.

Discussion

This study reports residential levels of agricultural chemicals in a farming region across both agricultural and nonagricultural households. The sample population included both farmers and farmworkers, most of whom lived on orchard property, where OP pesticides are sprayed frequently. As such, the study population would appear to approximate a "maximally exposed" group, at least in the tree fruit regions of North America. This study had a potential for selection bias because participation was voluntary and self-selected. Studies which focus on health and safety often attract participants with concerns for these issues. However, we have no evidence to suggest that the study families were unrepresentative of families in the region.

As expected, significantly higher levels of OPs were found in homes of Ag families than in those of reference families. Much higher levels of pesticides were found in household dust, where chemicals are not degraded or dispersed by environmental factors such as rain, sun, and soil microbial activity. These results are consistent with other reports of the persistence of pesticides in indoor environments (*12,13,20,22*).

Despite low pesticide concentrations in soil, significant correlations were observed between paired outdoor and indoor levels, suggestive of common sources for pesticide contamination of soil and household dust. In

Table 5. Organophosphorus mass loading results (µg pesticide/m² carpet)[a]

Pesticide	Ag families[b] (n = 48)	Reference families (n = 11)	Farmers (n = 26)	Farmworkers (n = 22)	Applicators (n = 28)[c]	Nonapplicators (n = 20)
Azinphosmethyl						
Mean	16.6	1.4	16.6	16.7	19.3	13.7
Median	9.9	0.83	10.7	8.0	14.4	5.8
Range	0.8–878	0.39–3.18	0.8–88	1.1–51	0.8–88	1.3–51
Phosmet						
Mean	27.1	0.91	18.4	36.1	26.8	27.5
Median	3.0	0.94	2.1	8.4	5.2	2.5
Range	<MLOQ–289	0.21–1.93	<MLOQ–289	0.2–222	<MLOQ–289	<MLOQ–164
Chlorpyrifos						
Mean	4.8	0.59	4.1	5.4	5.7	3.5
Median	1.9	0.47	1.62	2.0	2.7*	1.2*
Range	<MLOQ–27.7	<MLOQ–1.62	<MLOQ–25	0.09–28	<MLOQ–24.7	0.12–27.7
Ethyl parathion						
Mean	3.9	0.35	5.2	2.4	5.1	2.2
Median	1.2	<MLOQ	2.5	0.57	2.7*	0.05*
Range	<MLOQ–20.4	<MLOQ–2.43	<MLOQ–20	<MLOQ–17	<MLOQ–20.4	<MLOQ - 17.0

[a]Mass loading (µg/m²) = concentration of pesticide (ng/g) × grams of dust collected/m² carpet × 1 µg/1000 ng. Method limits of quantitation (MLOQ) in dust (ng/g): azinphosmethyl, 40; phosmet, 12; chlorpyrifos, 17; ethyl parathion 11; values <MLOQ assigned one-half MLOQ for statistical analysis.
[b]Ag families group combines the data from the farmers and farmworkers groups.
[c]Applicators and nonapplicators are groups within the Ag family group, based on whether orchard workers were engaged in pesticide handling (mixing, loading, application).
*Significant differences across groups: chlorpyrifos, $p = 0.04$; parathion, $p = 0.002$ (Mann-Whitney U test).

Table 6. Behavioral variables related to pesticide track-in

Question (n = 59; agricultural and reference families)	Positive response (%)
Do family members remove shoes at the door?	28
Are there walk-off mats outside main entries?	69
Is there a pet that goes in and out of the house?	33
How frequently are children's indoor play areas vaccuumed?	
>Weekly	40
Weekly	45
<Weekly	16
How far is the house from a commercial orchard? (n = 48 agricultural families)	
<50 ft	69
50–200 ft	15
>200 ft	17

contrast to trends for lead and arsenic contamination in these same samples (*35*), soil OP levels appear to be poor predictors of the magnitude of dust OP contamination due to degradation in the outdoor environment.

Pesticide concentrations in reference homes were much lower than those in Ag homes, yet it was surprising how frequently agricultural OP compounds were detected in dust samples for reference families. Due to the prevalence of orchards in the Wenatchee region, it was difficult to find volunteers who met the reference family inclusion criteria for reference homes. Although all reference families did live >0.25 mile from an orchard, many were within 0.5 mile. It therefore appears likely that those who reside in an agricultural region such as Wenatchee will have measurable pesticide residues in their homes regardless of personal pesticide use.

The significant relationship between proximity to orchards and concentration of azinphosmethyl in dust for the Ag families may reflect the fact that azinphosmethyl was the most recently sprayed and most commonly used OP compound (reported by 83% of farmers). Azinphosmethyl was the most frequently detected insecticide in household dust (100%) and soil (21%) among all the residences, including the reference homes. Chlorpyrifos had been applied 2–3 months previously by 57% of the surveyed farmers, but elevated soil residues were not found on these farms. The small number of subjects in each proximity category limits interpretation of these results. Phosmet was used by only 22% of farmers, limiting the possibility of detecting differences between groups with respect to proximity.

The interrelationship of three possible categories of the Ag family participants—homesite orchard proximity, occupation, and applicator status—complicated our analysis. The finding that the proximity and applicator status variables were not interactive for parathion levels in household dust suggests that either of these variables is predictive for elevated OP concentrations in dust.

Previous studies have suggested that toxicants carried or tracked into the home accumulate and may concentrate in dust, particularly in carpeted homes (*13,35,37*). Although the data presented here demonstrate substantial accumulation of pesticides in agricultural family homes, we were unable to identify specific exposure pathways such as track-in on shoes or by pets in this study. Pathway identification may have been confounded by variables such as type of carpeting, type of vacuum cleaner, composition of dust, or recall bias in self-reported information.

Dust sampling was conducted with the HVS-3 vacuum, a relatively new tool for environmental sampling. This method has been used to demonstrate that certain interventional measures can reduce the mass loading of contaminants in carpeted homes (*36*), but it is unclear whether the loading values obtained with the HVS-3 are representative of residues available to young children. A previous report in nine homes compared the HVS-3 technique with a polyurethane foam roller weighted to simulate the pressure applied to a surface by a crawling child (*13*). Mass loading results obtained by the two methods were correlated, but the levels from the HVS-3 were 4–12 times higher than those obtained by the foam roller. Methods used by other investigators in studies demonstrating correlations between mass loading and children's exposures have included wipe sampling and the use of a low-flow,

hand-held suction device (*37–39*). The more powerful suction of the HVS-3 may render results obtained with this technique more susceptible to confounding by carpet age, vacuum type, and frequency of cleaning. Further studies are needed to determine the representativeness of surface sampling techniques for estimating children's exposures.

The highest pesticide concentration found in any sample was 17 ppm (17,100 ng/g; phosmet in household dust), and the greatest total OP concentration measured in dust was 21.5 ppm (21,549 ng/g; sum of four OP compounds without regard to relative toxicity). A hazard evaluation was conducted for acute health risks among children living in study homes and indicated that acute intoxications from OP pesticide exposure through soil and contact with dust were unlikely. The hazard evaluation included use of toxicity data for the four OP compounds studied, a standard EPA soil contact transfer factor of 200 mg/day for children 1–6 years old (*18*), and the total OP dust concentration values from this study. A more detailed analysis of potential exposure to multiple OP compounds in these residential environments will be reported elsewhere.

Conclusions

Investigations of environmental and occupational health hazards normally proceed through the steps of recognition, evaluation, and control. This study has identified a potential hazard for young children residing in homes on or near sites of agricultural pesticide use by documenting environmental concentrations of four OP pesticides. In particular, it appears that children are likely to be exposed simultaneously to several pesticides that are not registered for residential use and that have the same mechanism of toxicity. Additional work is needed to evaluate children's exposure to agricultural pesticides in these settings, and, if necessary, to develop appropriate interventions to mitigate exposures. Carefully designed longitudinal or interventional studies will be needed to more adequately identify risk factors associated with the introduction of contaminants into the home. Biological monitoring based on urine sample collection may serve as an appropriate and noninvasive means of sampling exposure among small children.

Proximity to spray areas appears to have been the predominant, though not the only, factor responsible for elevated pesticide concentrations in household dust in this study. A number of variables still need to be assessed before it is possible to accurately estimate children's exposure from the dust/soil pathway, such as track-in, children's activity patterns, surface-to-skin contact/transfer rates for pesticides, dust/soil ingestion rates, and percutaneous uptake. Further investigation is warranted to address cumulative exposure to the multiple OP compounds found in these environments, rather than the traditional approach of focusing on a single compound for regulatory purposes.

Several strategies are available to reduce the risk potential of pesticide contamination in the home. A high percentage of participants in this study reported the use of full protective equipment while spraying and indicated that they did not bring this equipment into the home. These prudent work practices should be encouraged. Furthermore, programs designed to assist families with preventing or reducing indoor contaminants have been implemented in urban areas, especially for lead, and can be implemented in rural areas as well. Recommendations to reduce residential contaminants include improved home hygiene and personal hygiene measures, such as removal of shoes at the door, use of door mats, improved vacuuming techniques, and frequent washing of children's hands. The use of greater precautions when applying pesticides close to homes and a change in the practice of situating homes within orchard spray regions might also be considered. Finally, a change at the policy level to reduce the use of pesticides in the home and in surrounding agricultural areas would represent a strategy of primary prevention of pesticide exposure. The Environmental Protection Agency and the U.S. Department of Agriculture have recently proposed a Pesticide Use Reduction Initiative, which has as one of its goals the establishment of integrated pest management on 75% of active agricultural lands in 5 years. Policies such as this are very likely to affect pesticide contamination in the home, thereby reducing potential exposure to children and other family members.

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