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Pesticide Exposure of Children in an Agricultural Community: Evidence of Household Proximity to Farmland and Take Home Exposure Pathways[1]

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Children's exposure to organophosphorus (OP) pesticides in an agricultural community in central Washington State was determined. Spot urine and hand wipe samples were collected from 109 children 9 months to 6 years of age, as were house dust samples, and wipe samples from various surfaces. Children were categorized based on parental occupation (agricultural vs nonagricultural) and on household proximity to pesticide-treated orchards. Median house dust concentrations of dimethyl OP pesticides in homes of agricultural families were seven times higher than those of reference families (1.92 vs 0.27 μg/g; $P < 0.001$). Median pesticide metabolite concentrations in agricultural children were five times higher than those in reference children (0.05 vs 0.01 μg/ml; $P = 0.09$). Median pesticide concentrations in housedust ($P = 0.01$) and metabolite concentrations in urine ($P = 0.01$) from agricultural families were significantly higher in the children living near treated orchards (within 200 ft or 60 m) than those living more distant. Ten of 61 agricultural children had detectable OP pesticide levles on their hands, whereas none of the reference children had detectable levels. These findings indicate that children living with parents who work with agricultural pesticides, or who live in proximity to pesticide-treated farmland, have higher exposures than do other children living in the same community

Key Words: **pesticides; organophosphates; children; urinary metabolites; house dust.**

INTRODUCTION

Children can be exposed to pesticides and other hazardous chemicals through multiple pathways and by multiple routes. Diet is considered the primary exposure pathway for most pesticides, with drinking water and residential contact contributing to aggregate exposure in some cases (NRC, 1993; ILSI, 1998). Children in agricultural communities may be at increased risk from pesticide exposure if they live near pesticide-treated farmland, or if their parents come into contact with pesticides in the workplace. Thus, an evaluation of children's exposures in these communities requires evaluation of these two additional pathways.

Orchard pesticide treatments can result in deposition of pesticides beyond the application site (MacNeil and Hikichi, 1986; Clark *et al*., 1991; Fox *et al*., 1993). Household proximity to treated farmland may increase children's exposure if pesticides drift onto residential property or other areas in which children are active (Richter *et al*., 1992). In some cases the distinction between farmland and residence is blurred, as when a home is in the midst or on the boundary of an orchard. In other cases children may use fields as play areas, with or without the knowledge of their parents. All of these circumstances might lead to exposures that would not be captured by current aggregate exposure assessments.

If parents or other family members work with pesticides, chemicals may be brought into the home on work boots, tools, work clothing, or on the skin. Vehicles used for work may be used to transport the family, also contributing to this para-occupational or take home pathway. Numerous studies have demonstrated that children of exposed workers have significantly higher exposures to workplace chemicals

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than control children (NIOSH, 1995; Whelan *et al*., 1997; Piacitelli *et al*., 1997). Several studies have reported associations between parental occupational pesticide use and childhood leukemia (Loewengart *et al*., 1987; Shu *et al*., 1988; Buckley *et al*., 1989; Leiss and Savitz, 1995). Children of agricultural families appear to have a high potential for pesticide exposure, particularly when their parents are engaged in activities such as pesticide mixing, application, and intensive hand labor in treated fields. Studies have suggested that children of workers who are exposed to agricultural chemicals have increased risks of childhood cancers (Savitz and Chen, 1990; O'Leary *et al*., 1991; Daniels *et al*., 1997). It therefore seems clear that the potential health risks associated with pesticide exposure within this subpopulation require more thorough investigation.

A 1992 study in central Washington state found that children of agricultural families had higher potential exposures to organophosphate (OP) pesticides in soil and house dust than those living in nonagricultural families, suggesting that both proximity to farmland and parental occupation can contribute to increased environmental concentrations (Simcox *et al*., 1995). In 1995 a more comprehensive evaluation of children's exposure was conducted in the same community, including biological monitoring. A preliminary report of this study (Loewenherz *et al*., 1997) focused on concentrations of a single urinary metabolite of the OP pesticides (dimethylthiophosphate) for a subset of the study population (children of pesticide applicators). The present article expands upon this earlier analysis by utilizing a larger sample size, and including environmental measurements. Its purpose is to better characterize what will be referred to as the *household proximity* and *take home* exposure pathways, and to determine their contributions to children's exposure as estimated by measurement of OP pesticide concentrations in children's home environments and dialkylphosphate metabolite concentrations in children's urine.

METHODS

Study Design and Population

The study was cross-sectional in design, and took place in central Washington state in the vicinity of Wenatchee (Douglas and Chelan counties). The primary industry of the region is tree fruit, with many small family orchards. This area was chosen because of periodic OP pesticide use on tree fruit and because of the prevalence of agricultural employment.

Agricultural family selection was based on the following eligibility criteria: at least one child 6 years old or younger, and at least one family member employed as an orchardist–applicator or farm worker. Reference family eligibility criteria were that no family member's work involved contact with agricultural pesticides, and that the residence was located more than one-quarter mile (about 400 m) from any pesticide-treated orchard. In this study pesticide applicators were individuals responsible for pest management for a specific orchard, and who therefore conducted periodic spraying for pest control as well as other farm management tasks. The study population did not include applicators who provided commercial services to growers, or whose primary work activity was pesticide spraying. Farm workers in this study were individuals who conducted orchard hand labor tasks such as irrigation, thinning and pruning, and harvesting.

Agricultural families were identified through a health services agency, a grower organization, and the state's cooperative extension program. Reference families were contacted through the same service organizations mentioned above, and included staff members and their neighbors. The University of Washington Human Subjects Review Committee approved the study procedures associated, and all subjects provided their informed consent.

A total of 109 children and 76 homes were sampled May through July 1995. An initial visit included administration of an interview and collection of the following samples: a urine void and isopropanol hand wipe from each participating child, a house dust sample from a carpeted area in the house, and wipe samples from a noncarpeted floor surface, the steering wheel of the vehicle, and the worker's boots. Interviews were conducted in either Spanish or English as appropriate, and included questions regarding frequency and extent of occupational and residential pesticide use, hygienic practices, housekeeping practices, proximity to pesticide-treated orchards, and child activity. All homes were visited a second time, 3–7 days after the first visit, and involved a short interview, and collection of urine and hand wipe samples. Two dimethyl OP pesticides commonly used during the May–July spraying season for tree fruit production in this region were targeted for analysis: azinphos methyl (CAS No. 86-50-0; trade name Guthion) and phosmet (CAS No. 732-11-6; trade name Imidan). Two diethyl OP pesticides were also selected for analysis, and these data will be reported separately.

Proximity Exposure Pathway

House dust was considered to be the best medium through which to characterize the accumulation of pesticides in residences due to nearby agricultural use. House dust samples were collected using the HVS-3 (Roberts *et al*., 1991), a vacuum system designed specifically for house dust sampling, from the main entrance/living area of the home if carpeted, or from the area where the children played most frequently in the home. An area of 45×137-cm (a total of 6165 cm^2) divided longitudinally into three strips was marked with masking tape for sampling. The HVS-3 sampler was placed at the first strip, and pushed from the beginning to the end of the strip in 4 s. Each strip was sampled back and forth four times.

Take Home Exposure Pathway

Investigation of take home exposure involved wipe sample collection from the steering wheel of the vehicle used for travel to work, work boots, and noncarpeted indoor floors where children play. Residues on the steering wheel were considered indicative of worker skin and clothing contamination; residues on work boots were thought to indicate potential for track-in; residues on noncarpeted floors were considered indicative of recent track-in.

Two sterile $4'' \times 4''$ all-cotton gauze pads wetted with 1–2 mL of 100% isopropanol were used to wipe the noncarpeted floor area with a 50×50-cm metal template and a sequence of three vertical and three horizontal strokes. A similar procedure was used to wipe the toe area of the work boots of a worker at home at the time of sampling using a 5×5-cm template. After wiping the boots with three vertical strokes, the same gauze pad was folded so that a fresh surface was used for the second wipe of three horizontal strokes. The same procedure was used for the steering wheel, except that instead of using a template, samples were collected from the top half of the steering wheel using one continuous stroke.

Personal and Biological Exposure Sampling

Hand wipe sampling was performed in order to measure pesticides on the skin. One gauze pad was used for the palm and the back of each hand and a second pad for the fingers, so that the entire surface of the hand was wiped. Both hands were wiped, so a total of four gauze pads were used and placed in a prelabeled jar and treated as one sample. A single urine void was collected at each visit from each child. Samples were obtained using either a urine collection bag (Lil'Katch; General Medical Corp., Richmond, VA) for the nontoilet-trained child, or a commode insert (Specipan; Baxter Scientific, McGaw Park, IL) for the toilet-trained child. If the field staff could not collect samples at the time of the visit, a parent was given a collection apparatus and instructions, and samples were picked up within 24 h of the void. Timing of the sample in these cases was at the convenience of the family.

Laboratory Methods

All samples were brought to the field laboratory in Wenatchee in an ice chest, where they were processed and stored at $-10^{\circ}C$, and were later transported to the analytical laboratory in Seattle with dry ice and stored at $-20^{\circ}C$ until analysis. House dust samples were first sieved in a 100-mesh (150 μm) stainless-steel sieve for 6 min. The sieved samples were extracted in 50 ml of acetone for 1 min using a sonicator, centrifuged at 2500 rpm for 8 min, concentrated under purified nitrogen stream, and solvent-exchanged into 1 ml of cyclohexane. The samples were subject to further cleanup in a series of filtration, gel permeation chromatography, concentration, and evaporation steps prior to gas chromatography analysis with a mass selective detector in selected ion monitoring mode for four targeted pesticides: chlorpyrifos, ethyl parathion, phosmet, and azinphos methyl (Simcox *et al*., 1995). All gauze pad samples were extracted with 50 ml of ethyl acetate on a shaker table for 30 min.

Urine samples were analyzed for OP pesticide metabolites including dimethylphosphate (DMP), dimethylthiophosphate (DMTP), and dimethyldithiophosphate (DMDTP), which result from OP pesticides with dimethyl moieties, such as azinphos methyl and phosmet. Urine samples were processed by azeotropic distillation in acetonitrile, centrifuged, evaporated, and derivatized with pentafluorobenzylbromide prior to gas chromatography analysis with a flame photometric detector in the splitless mode (Loewenherz *et al*., 1997). Creatinine concentrations were also measured to identify abnormal samples.

Extraction of azinphos methyl and phosmet in gauze pads was complete ($117 \pm 39\%$ recovery for azinphos methyl and $101 \pm 12\%$ recovery for phosmet), but was less complete in house dust ($68 \pm 10\%$ for azinphos methyl and $60 \pm 9\%$ for phosmet). The extraction efficiencies of DMTP and DMDTP averaged $80 \pm 9\%$ and $62 \pm 6\%$, respectively. Pesticide concentrations in house dust samples and metabolite concentrations in urine samples were adjusted

by these extraction efficiencies. No adjustment was made for wipe samples.

Data Analysis

The limits of quantitation (LOQs) for DMP, DMTP, DMDTP, azinphos methyl, and phosmet varied among analytical batches. Samples with detectable concentrations but below the respective LOQs were assigned one-half the value of LOQ. Median values were lower than mean values in most cases, suggesting a skewed distribution of dimethyl OP pesticide concentrations in house dust and urinary metabolites. Statistical analyses, therefore, were performed using nonparametric tests in SPSS (SPSS, Inc., 6.1.1, Chicago, IL).

DMTP and DMDTP were selected as the biomarkers of dimethyl OP pesticide exposure because DMP measurements were found to be inconsistent across batches and were not considered reliable. DMTP and DMDTP levels for visit 1 and visit 2 from each child were averaged and converted to their molar equivalents, summed to produce a single dialkylphosphate molar concentration, and then multiplied by the molecular weight of azinphos methyl and phosmet (both 317 g/mol), as in the equation

$$OP_U = [C_{DMTP}/MW_{DMTP} + C_{DMDTP}/MW_{DMDTP}] * MW_{OP}, \quad (1)$$

where OP_U = dimethyl OP pesticide metabolite concentration (μg/ml); C_{DMTP} = DMTP concentration (μg/ml); C_{DMDTP} = DMDTP concentration (μg/ml); MW_{DMTP} = molecular weight of DMTP (142 g/mol); MW_{DMDTP} = molecular weight of DMDTP (158 g/mol); MW_{OP} = molecular weight of either azinphos methyl or phosmet (317 g/mol).

A similar equation was used to calculate a dimethyl OP pesticide equivalency concentration in house dust using azinphos methyl and phosmet concentration data,

$$OP_D = [C_{phosmet}/MW_{phosmet} + C_{azinphos\,methyl}/MW_{azinphos\,methyl}] * MW_{OP}, \quad (2)$$

where OP_D = dimethyl OP pesticide house dust concentration (μg/g); $C_{phosmet}$ = phosmet house dust concentration (μg/g); $C_{azinphosmethyl}$ = azinphos methyl house dust concentration (μg/g); MW_{OP} = molecular weight of either azinphos methyl or phosmet (317 g/mol).

In order to remove the within-household dependence for families with more than one child participating in the study, a focus child was selected from those families based on the following criteria: having completed the two spot urine samplings and having acceptable creatinine measurements for both samples. Random selection was then made for families with more than one child meeting the above criteria.

RESULTS

Agricultural and Reference Family Comparisons

Participating families consisted of 49 applicator, 13 farm worker, and 14 reference families. Thirty-one families had more than one participating child. There were 72, 19, and 18 children living in pesticide applicator, farm worker, and reference families, respectively. Pesticide applicator and farm worker families have been combined for some analyses as agricultural families due to the small sample size of the farm worker group. (Our earlier report of DMTP concentrations (Loewenherz *et al*., 1997) included 48 of these 49 pesticide applicator families and the 14 reference families).

Azinphos methyl and phosmet concentrations in house dust for applicator, farm worker, agricultural and reference homes are presented in Table 1. Pesticide applicator families had the highest median house dust concentrations of azinphos methyl, phosmet, and dimethyl OP pesticides, followed by farm workers and reference families, but applicator and farm worker values were not statistically different (Whitney *U*–Wilcoxon Rank Sum *W* test, $P > 0.1$) When all agricultural families were compared to reference families, median values were significantly different (Kruskal–Wallis one-way ANOVA, $P < 0.001$, for azinphos methyl and dimethyl OP pesticides: $P = 0.02$ for phosmet). Median house dust concentrations of azinphos methyl, phosmet, and dimethyl OP pesticides were 6.7, 1.6, and 7.1 times higher in agricultural than in reference families, respectively. The boxplot in Fig. 1 illustrates that most agricultural families showed higher dimethyl OP levels in house dust than did reference families.

Twenty-three families (22 agricultural and 1 reference) sampled in this study also participated in our 1992 study (Simcox *et al*., 1995). Figure 2 compares the 1992 dimethyl OP pesticide house dust concentrations with those of the current study. The four homes with the highest concentrations in 1992 had lower levels in 1995, and concentrations decreased over time for 16 of the 23 families (Wilcoxon Matched-Pairs Signed-Ranks test, $P = 0.04$).

TABLE 1
OP Pesticide Concentrations in House Dust, and Urinary Metabolites Levels in Focus Children of Applicator, Farmworker, and Reference Families

Pesticide or metabolite	Applicator families ($n = 49$)	Farmworker families ($n = 13$)	Ag families[a] ($n = 62$)	Reference families ($n = 14$)
House dust (μg/g)[b]				
Azinphos methyl				
Mean (SD)	2.06 (2.3)	1.47 (1.5)	1.94 (2.19)	0.29 (0.35)
Median	1.06[f]	0.75[f]	1.0[g]	0.15[g]
Range	0.04–9.2	0.3–5.3	0.04–9.2	0.01–1.1
Phosmet				
Mean (SD)	1.23 (2.5)	0.14 (0.1)	1.01 (2.27)	0.09 (0.04)
Median	0.15[f]	0.11[f]	0.14[h]	0.09[h]
Range	0.01–14.6	0.03–0.3	0.01–14.6	0–0.2
Dimethyl OPs[c]				
Mean (SD)	3.29 (3.2)	1.61 (1.6)	2.95 (3.0)	0.37 (0.37)
Median	2.36[i]	0.92[i]	1.92[g]	0.27[g]
Range	0.2–15.1	0.4–5.5	0.2–15.1	0.01–1.3
Urine (μg/ml)[d,e]				
DMTP				
Mean (SD)	0.04 (0.05)	0.03 (0.04)	0.04 (0.04)	0.02 (0.04)
Median	0.03[f]	0.02[f]	0.02[i]	0.005[i]
Range	0–0.2	0–0.1	0–0.2	0–0.1
DMDTP				
Mean (SD)	0.005 (0.01)	0.002 (0.003)	0.004 (0.009)	0.003 (0.005)
Median	0[f]	0[f]	0[f]	0[f]
Range	0–0.04	0–0.007	0–0.04	0–0.02
Dimethyl metabolites[c]				
Mean (SD)	0.1 (0.1)	0.07 (0.08)	0.09 (0.11)	0.06 (0.09)
Median	0.06[f]	0.05[f]	0.05[i]	0.01[i]
Range	0–0.6	0–0.2	0–0.6	0–0.3

[a] Ag families are a combination of applicator and farm worker families.
[b] House dust sample limits of quantitation (LOQs) were 0.013 μg/g for phosmet, and varied from batch to batch for azinphos methyl (0.013–0.056 μg/g). Data were adjusted by extraction efficiencies (see Methods).
[c] Dimethyl OP pesticide (μg/g) and dimethyl OP metabolite (μg/ml) concentrations were calculated using molar-equivalent method (see Methods).
[d] DMTP and DMDTP limits of quantitation were 0.02 and 0.04 μg/ml, respectively. Data were adjusted by the extraction efficiencies (see Methods).
[e] Average values of visit 1 and visit 2 of focus child for each family.
[f] No significant difference: Whitney U–Wilcoxon Rank Sum W test, $P \geq 0.1$.
[g] Significant difference: Whitney U–Wilcoxon Rank Sum W test, $P < 0.001$.
[h] Significant difference: Whitney U–Wilcoxon Rank Sum W test, $P = 0.02$.
[i] Marginal significant difference: Whitney U–Wilcoxon Rank Sum W test, $P = 0.07$ for dimethyl OPs in house dust and DMTP, and $P = 0.09$ for dimethyl metabolites.

Approximately 67% of urine samples (both visits 1 and 2) collected from applicator and farm worker children contained detectable DMTP levels, while this was the case for only 53% of the reference children. The overall frequency of detection of DMDTP was 13%. DMTP and dimethyl OP metabolite concentrations tended to be higher in applicator children when compared to farm worker children, but the differences were not statistically significant (Table 1). When these two groups were combined as agricultural children, median DMTP and dimethyl OP metabolite concentrations were four to five times higher than those in reference children (Whitney U–Wilcoxon Rank Sum W test, $P = 0.07$ for DMTP; $P = 0.9$ for dimethyl OP metabolite). Median levels of DMDTP in applicator, farm worker, and reference children were below the limit of detection, and assigned values of zero. The difference in urinary dimethyl OP metabolite concentrations between agricultural and reference children is shown in Fig. 2.

Proximity

The agricultural families were categorized by distance from a nearby orchard that had been treated

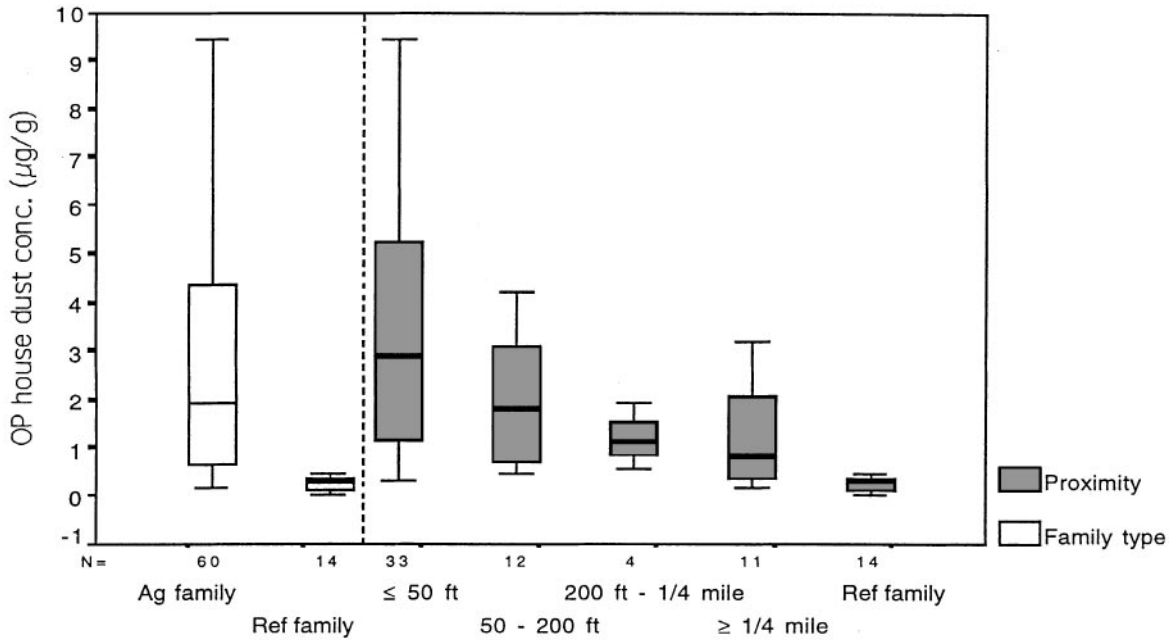

FIG. 1. Dimethyl OP pesticide concentrations in house dust of agricultural and reference families, and grouped by proximity to a pesticide-treated orchard. Group concentrations and trend with proximity showed a significant difference ($P < 0.001$ and $P = 0.04$, respectively. Box plot key: the harizontal lines in each plot represent 10th, 25th, 50th, 75th, and 90th percentiles, bottom to top.

with pesticides. Thirty-five of these families lived within 50 feet (15 m), 12 lived between 50 and 200 feet (15–60 m), 4 lived between 200 feet and 1/4 mile (400 m) away, and 11 lived more than 1/4 mile away from a pesticide-treated orchard. By definition, all of the 14 reference families lived more than 1/4 mile away from a pesticide-treated orchard. Figures 1 and 3 show boxplots for dimethyl OP pesticide concentrations in house dust, and the OP metabolite levels in agricultural and reference children, grouped by proximity, respectively.

When proximity to an orchard was categorized as ≤ 200 ft and >200 ft (Table 2), azinphos methyl and dimethyl OP pesticide concentrations were

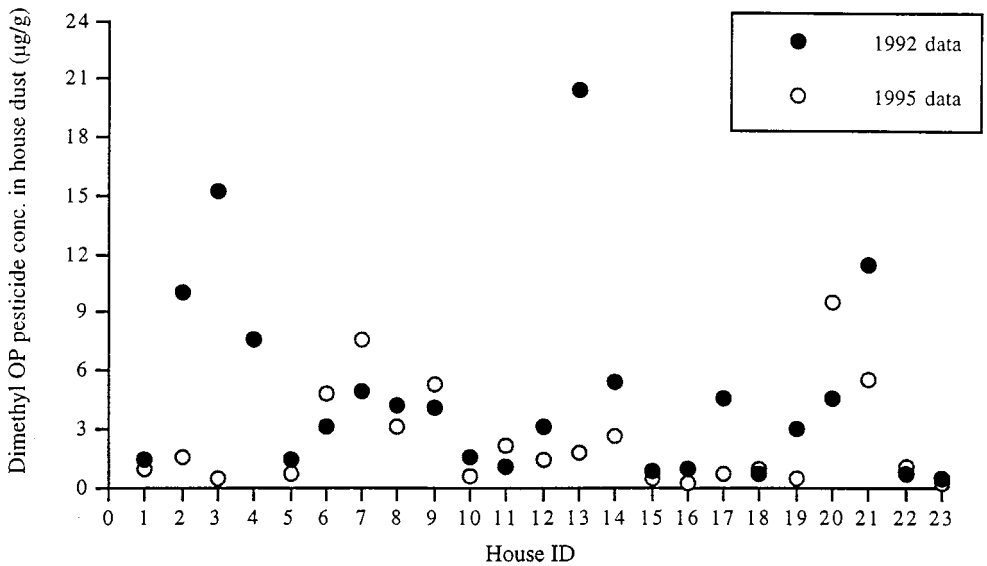

FIG. 2. Dimethyl OP pesticide concentrations (μg/g) in house dust in 23 families who participated in both 1992 and 1995 studies. Concentrations in 1995 were significantly lower than 1992 concentrations ($P = 0.04$).

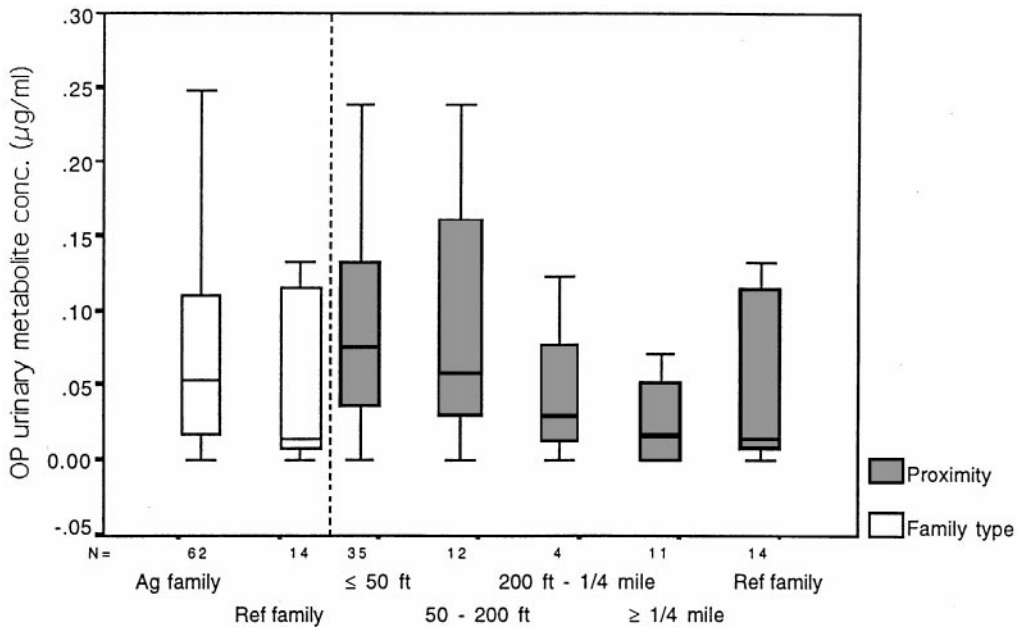

FIG. 3. Dimethyl OP pesticide metabolite levels in urine of agricultural and reference focus children, and grouped by proximity to a pesticide-treated orchard. Group concentrations and trend with proximity showed a marginal significant difference ($P = 0.09$ and $P = 0.10$, respectively). Box plot key: the horizontal lines in each plot represent 10th, 25th, 50th, 75th, and 90th percentiles, bottom to top.

found to be significantly higher in the house dust of proximate homes (Whitney *U*–Wilcoxon Rank Sum *W* test, $P < 0.01$ for azinphos methyl; $P = 0.01$ for dimethyl OP pesticides); the DMTP and dimethyl OP metabolite concentrations in the urine of children from proximate homes were also elevated (Whitney–*U*–Wilcoxon Rank Sum *W* test, $P = 0.01$ for both). Using the four proximity categories for agricultural families presented in Fig. 1, a test of slope for the linear regression line indicated a decreasing trend in OP pesticide housedust concentrations ($y = -0.68x + 4.24$; $P = 0.04$). The same trend was evident for dimethyl OP metabolites ($y = -0.2x + 0.13$) (Fig. 3), but the relationship was less significant ($P = 0.1$). Including reference family data as a fifth category significantly strengthened these trends (housedust concentrations, $P < 0.01$; metabolite concentrations, $P = 0.06$).

Take Home Exposure Pathway

Table 3 presents summary statistics for azinphos methyl and phosmet on children's hands, parents' work boots, vehicle steering wheels, and noncarpeted floors. The percentages of quantifiable dimethyl OP pesticide levels ranged from 3 to 33% and all were collected from agricultural families. Neither hand nor environmental wipes collected from reference families had detectable levels of either of the target OP pesticides.

Ten of the 61 agricultural focus children had measurable dimethyl OP pesticide (either azinphos methyl or phosmet) concentrations in their hand wipe samples, and all of them lived within 200 feet of a pesticide-treated orchard. For 38 of the agricultural families one or more environmental wipe samples had measurable dimethyl OP pesticide levels. Wipe samples taken from parents' work boots showed the highest frequency of detection of dimethyl OP pesticides, followed by samples taken from vehicle steering wheels. Samples collected from the noncarpeted floor showed the lowest detection frequency. Thirty-two of the 38 agricultural families with measurable levels lived within 200 feet (28 lived within 50 feet) of a pesticide-treated orchard.

A Mann–Whitney *U* test was used to evaluate differences in OP pesticide house dust concentrations between agricultural families living more than 1/4 mile from treated farmland and reference families (see the final two boxplots in Fig. 1). The test was performed to remove the effect of proximity from the analysis of the take home exposure pathway. OP pesticide residues in house dust were found to be significantly higher in the agricultural family homes

TABLE 2

OP Pesticide House Dust Concentrations in Agricultural Households and Dimethyl OP Urinary Metabolite Levels in Focus Agricultural Children for Residences >200 ft or ≤ 200 ft from Orchards

Pesticide or metabolite	Proximity ≤ 200 ft	Proximity > 200 ft
House dust (μg/g)	($n = 45$)	($n = 15$)
Azinphos methyl		
Mean (SD)	2.2 (2.2)	1.3 (2.1)
Median	1.3[a]	0.49[a]
Range	0.3–9.2	0.04–8.3
Phosmet		
Mean (SD)	1.2 (2.6)	0.45 (0.6)
Median	1.14	0.12
Range	0.01–15	0–2.0
Dimethyl OP		
Mean (SD)	3.4 (3.1)	1.7 (2.2)
Median	2.6[b]	0.87[b]
Range	0.3–15	0.01–8.4
Urine (μg/ml)	($n = 47$)	($n = 15$)
DMTP		
Mean (SD)	0.04 (0.05)	0.02 (0.03)
Median	0.03[c]	0.01[c]
Range	0–0.2	0–0.1
DMDTP		
Mean (SD)	0.005 (0.01)	0.002 (0.004)
Median	0	0
Range	0–0.04	0–0.01
Dimethyl OP metabolite		
Mean (SD)	0.1 (0.11)	0.04 (0.07)
Median	0.07[d]	0.02[d]
Range	0–0.6	0–0.3

[a] Significant difference: Whitney *U*–Wilcoxon Rank Sum *W* test, $P = 0.008$.

[b] Significant difference: Whitney *U*–Wilcoxon Rank Sum *W* test, $P = 0.014$.

[c] Significant difference: Whitney *U*–Wilcoxon Rank Sum *W* test, $P = 0.009$.

[d] Significant difference: Whitney *U*–Wilcoxon Rank Sum *W* test, $P = 0.01$.

when compared to the reference family homes ($P = 0.02$).

House Dust and Urinary Metabolite Comparison

A marginally significant association was found between house dust concentrations (azinphos methyl, phosmet, or combined dimethyl OP pesticides) and urinary metabolite concentrations for either agricultural family data or for all data (Fig. 4 illustrates all data). The Spearman *rho* was 0.35, resulting in an estimated R-squared value of 0.12 (Spearman Rank Correlation, $P = 0.09$). Parametric analyses using a linear regression model, log-linear model, or log–log model produced similar results.

Residential Activities

Data were gathered through parental interviews regarding children's behavior, family hygienic practices, or residential pesticide use. Analysis of these data failed to reveal any significant associations between these practices and either pesticide house dust concentrations or pesticide metabolite concentrations (Tables 4a–4c). Parents were asked about their children's behavior such as time spent outdoors, hand washing before each meal, hand-to-mouth activity, and frequent thumb-sucking. In general, higher dimethyl OP metabolite concentrations were found among agricultural children whose parents gave positive responses to these questions; however, no statistically significant relationships were found (Table 4a).

Questions regarding parental hygienic practices focused on the presence of doormats, the wearing of work shoes and work clothes in the house, laundering practices, and vacuuming frequency (Table 4b). Lower dimethyl OP metabolite levels among agricultural children were found in families who removed shoes at the door and work clothes before entering the house than those who did not, but these relationships were not statistically different. Results related to use of doormats and separating family laundry from work clothes tended in the opposite direction, but again no significant differences were evident. Vacuuming frequency also did not appear to be associated with differences in dimethyl OP metabolite concentrations in the agricultural children.

Participating families were also asked about their residential pesticide use (Table 4c). Sixteen percent of agricultural families treated household pets (both dogs and cats) with pesticides. No differences were found in OP metabolite concentrations related to this practice. Those who answered negatively to questions related to pet or indoor pesticide treatments appeared to have higher OP pesticide house dust concentrations, but the differences were not significant. Treatment of lawn and garden was not associated with differences in either metabolite or house dust concentrations. In summary, none of the interview data were significantly related to the exposure measures evaluated in this study.

DISCUSSION

Concern over children's pesticide exposure and consequent health risks has increased in recent years. The recent National Research Council report called for investigation of exposure from all pathways and sources (NRC, 1993). This study

TABLE 3

OP Pesticide Concentrations on Agricultural Children's Hands, and on Boots, Vehicle Steering Wheels, and Noncarpeted Floors in Agricultural Households[a]

	Azinphos methyl	Phosmet	Dimethyl OPs
Children's hands ($n = 61$) (μg/two hands)[b]			
Mean	0.07	0.01	0.07
Median	0	0	0
SD	0.2	0.04	0.2
Range	0–1.2	0–0.3	0–1.2
Frequency (%)[f]	8 (13)	2 (3)	10 (16)
Parent's work boots ($n = 60$) ($\mu g/cm^2$)[c]			
Mean	0.03	0.07	0.09
Median	0	0	0
SD	0.06	0.34	0.37
Range	0–0.3	0–2.6	0–2.9
Margin (%)	20 (33)	17 (28)	27 (45)
Steering wheel ($n = 55$) ($\mu g/cm^2$)[d]			
Mean	0.001	0.001	0.002
Median	0	0	0
SD	0.002	0.004	0.005
Range	0–0.01	0–0.02	0–0.03
Frequency (%)	5 (9)	4 (7)	6 (11)
Noncarpeted floor ($n = 55$) ($\mu g/cm^2$)[e]			
Mean	0.001	0.001	0.002
Median	0	0	0
SD	0.004	0.003	0.005
Range	0–0.02	0–0.02	0–0.02
Frequency (%)	3 (5)	2 (4)	5 (9)

[a] All samples collected from reference families were nodetectable, so this table reports data from agricultural families only; wipe sample limits of quantitation (LOQs) were 0.65 μg/sample for phosmet, and varied from batch to batch for azinphos methyl (1.25–3 μg/sample).

[b] Values represent the average of samples from visits 1 and 2, and for focus children only.

[c] Boots were wiped using a 5×5-cm template to outline the toe area of work boot.

[d] Wipe samples were taken from the top half of the steering wheel, an estimated area of 550 cm^2.

[e] A 50×50-cm template was used to wipe the floor near the entryway, if it was not carpeted, or a noncarpeted area where the child often played.

[f] Frequency = number of families (or focus children) with quantifiable OP pesticide concentrations (either azinphos methyl or phosmet); percentages in parentheses.

demonstrates that children of agricultural families can be exposed to pesticides through pathways other than diet, drinking water, and residential pesticide use. A clear effect of proximity to treated farmland was found both through measurement of pesticide residues in the home and by biological monitoring. Persuasive evidence was also found for the existence of a take home exposure pathway, since housedust concentrations were elevated in those agricultural family homes more than 1/4 mile from farmland, and pesticide residues were detected on work boots, steering wheels, and children's hands for many of the agricultural families.

This study represents a continuing effort to better characterize children's OP pesticide exposures, and is novel in its use of biological monitoring data to complement environmental concentration measurements for exposure pathway analysis. Our previous report (Loewenherz *et al.*, 1997) focused on DMTP levels for children of pesticide applicators, while this more complete report includes a group of farm worker children, and extensive environmental sampling. In the earlier report, a significant difference in DMTP concentrations between applicator children and reference children was found. The magnitude of the difference between agricultural and reference children urinary metabolite levels observed here was similar (four- to fivefold), but the statistical significance was reduced ($P = 0.03$ in 1997 study; $P = 0.07$ in current study). The farm

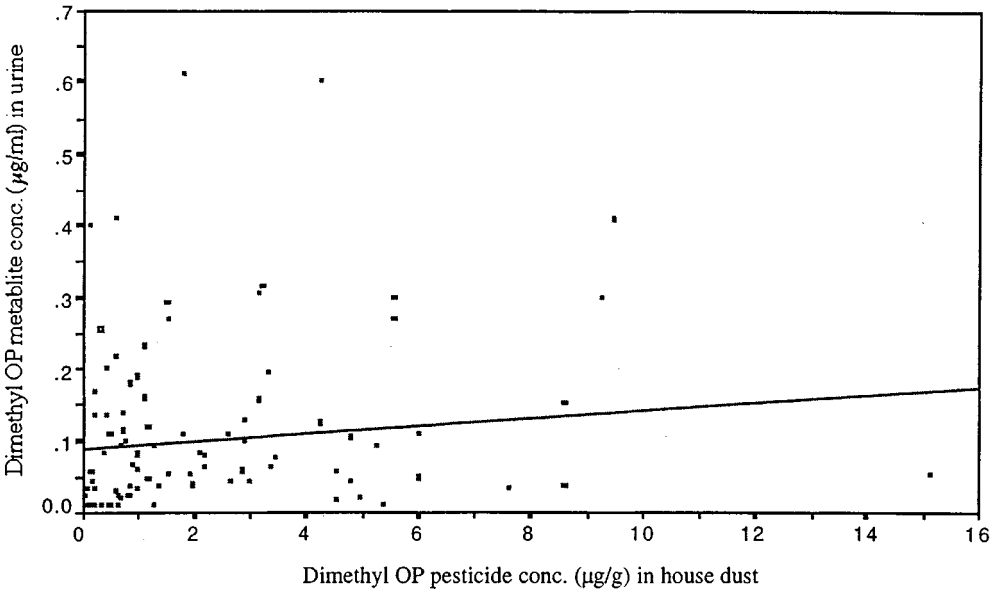

FIG. 4. Scatter plot of dimethyl OP pesticide concentrations in house dust of agricultural families and dimethyl OP metabolite levels in urine of agricultural children ($n = 60$). Spearman *rho* of 0.35 was marginally significant ($P = 0.09$).

worker children's exposures fell between those of the applicator and reference children groups, so including them as part of the agricultural family group appears to have diluted the original finding. Similarly, the earlier report demonstrated an effect of proximity on the DMTP metabolite concentration for the applicator children. This finding is confirmed by analysis of the full study population. The clear difference in house dust levels between agricultural and reference families, and differences related to proximity category further support the conclusion that agricultural children have elevated exposures compared to reference children in the same communities.

It is important to note several aspects of this study that limit the generalization of its findings. First, all environmental and biological samples were collected from May to July, a period that coincided with active spraying of azinphos methyl and phosmet for coddling moth control in this region. Thus, the values reported here may not be representative of year-round exposure for these

TABLE 4a
Child Behavior Questions and the Corresponding Median Dimethyl OP Metabolite Concentrations in Agricultural Focus Children

Question	OP metabolite concentration (μg/ml) Positive response[a]	Negative response[a]	*P* value
How many hours/day are children outdoors?			
Less than an hour	0.05 (31)		0.8[b]
Between 1 and 4 hours	0.05 (47)		
More than 4 hours	0.06 (22)		
Do children wash their hands before meals?	0.09 (69)	0.05 (31)	0.2[c]
Do children have hand-to-mouth activity?	0.05 (69)	0.06 (31)	0.6[c]
Do children suck their thumbs?	0.09 (16)	0.05 (84)	0.6[c]

[a] Percentage response in parentheses.
[b] Kruskal–Wallis one-way ANOVA.
[c] Whitney *U*–Wilcoxon Rank Sum *W* test.

TABLE 4b

Family Hygienic Practice Questions and the Corresponding Median Dimethyl OP Metabolite Concentrations in Agricultural Children or Dimethyl OP Concentrations in House Dust in Agricultural Families

	OP metabolite concentrations (μg/ml)			OP house dust concentration (μg/g)		
Question	Positive response[a]	Negative response[a]	*P* value	Positive response[a]	Negative response[a]	*P* value
Do household members remove shoes at the door?	0.04 (60)	0.07 (40)	0.2[b]	1.5	2.1	0.8[b]
Are there doormats outside the main entrance?	0.07 (89)	0.03 (11)	0.3[b]	1.8	2.9	0.6[b]
Do household members wear work clothes in the house?	0.07 (38)	0.05 (62)	0.2[b]	2.7	1.5	0.2[b]
Do work clothes mix with family laundry?	0.05 (68)	0.08 (32)	0.8[b]	1.4	3.1	0.4[b]
How frequently is the carpet vacuumed?						
More than once a week	0.07 (59)			1.5		
Once a week or less	0.05 (35)		0.3[b]	2.6		0.6[b]
No answer	0.08 (6)			2.3		

[a] Percentage response in parentheses.
[b] Whitney *U*–Wilcoxon Rank Sum *W* test.

children. Second, both azinphos methyl and phosmet are metabolized to DMP, DMTP, and DMDTP, and then excreted in urine, but only DMTP and DMDTP data were used in these analyses. Therefore, the calculated dimethyl OP metabolite concentrations are likely underestimates of the total dialkylphosphate concentrations. Our more recent studies suggest that DMP represents approximately 37% of total dimethyl OP metabolites in the urine.

TABLE 4c

Residential Pesticide Use Questions and the Corresponding Median Dimethyl OP Metabolite Concentrations in Agricultural Children and Dimethyl OP Concentrations in House Dust in Agricultural Families

	Dimethyl OP metabolite conc. (μg/ml)			Dimethyl OP house dust conc. (μg/g)		
Question	Positive response[a]	Negative response[a]	*P* value	Positive response[a]	Negative response[a]	*P* value
Are household pets treated with pesticides?	0.07 (16)	0.05 (84)	0.6[b]	0.7	2.1	0.1[b]
Has your house been treated with OPs since January 1995?	0.05 (31)	0.05 (69)	0.6[b]	1.1	2.1	0.3[b]
Has your lawn ever been treated with OPs?	0.06 (31)	0.05 (69)	0.7[b]	2.6	1.8	0.7[b]
Have you ever used OPs in your garden?	0.08 (29)	0.05 (61)	0.9[b]	2.1	1.9	0.8[b]

[a] Percentage response in parentheses.
[b] Whitney *U*–Wilcoxon Rank Sum *W* test.

The presence of elevated pesticide concentrations in the house dust of agricultural family homes requires further study. Pesticides probably reach homes near treated farmland through spray drift. Airblast applications, which are common in tree fruit orchards, are known to produce measurable drift up to a distance of 200 ft (Fox *et al*., 1993). Small particles are likely to travel greater distances, and soil in the orchards may be resuspended and move off site. The fact that azinphos methyl, a pesticide registered for agricultural use only, was detected in every study home — as it was in 1992 (Simcox *et al*., 1995) —is clear evidence that such pesticides move beyond the targeted application area. Identification of the specific mechanisms through which pesticide residues enter the home in this community will require additional investigations.

Most agricultural families in this study lived in close proximity to treated farmland, so it was not possible to determine the relative importance of the proximity and take home exposure pathways. It is clear, however, that pesticide residues were present on the work boots and in the vehicles of some agricultural workers, while such residues were not found among any of the reference family participants. Hygienic practices such as use of doormats, removal of work shoes and work clothes before entering the home, separation of work clothes for laundering, and frequent vacuuming are often included as common sense recommendations in farm worker education programs. Our analysis failed to demonstrate that these practices reduce either pesticide house dust concentrations or children's biological levels, and even the trends were inconsistent. However, our findings were based on parental self-reports, and the validity and reliability of these reports could not be ascertained. These results suggest that such questions may not be useful as predictors of residential contamination levels or body burdens.

It appears that both azinphos methyl and phosmet levels in house dust have been decreasing over time in most of the households that were sampled in 1992 and in 1995. At the conclusion of the 1992 study we sent out letters to participating families with results and public health recommendations, followed by meetings in the community to answer questions regarding the study and concerns about pesticide exposure in children. These educational activities were aimed at helping families reduce pesticide exposures among their children and around their homes. This intervention may have contributed to the reduction in house dust concentrations.

While we have emphasized the finding of a difference between OP pesticide exposures in agricultural and reference children, it is also worth noting that a majority of the reference children had detectable dimethyl OP pesticide metabolities. The use of a class-specific biomonitoring assay like the dialkylphosphate metabolites rather than a compound-specific assay (e.g., *para*-nitrophenol for parathion) extends the integrative capacity of the analysis, but at the cost of specificity. The data collected in this study do not allow identification of specific pathways for the reference population. For example, diet is likely to have been an important contributor to metabolite concentration, as it has been reported that many OP pesticides, including azinphos methyl, can be found on fresh fruits and vegetables (USDA, 1997).

Nonetheless, the presence of OP pesticides in reference family house dust suggests that environmental residues from agricultural pesticide use contribute to total body burden for children in agricultural communities. The lack of a clear association between house dust concentrations and biological levels suggests that residential exposure pathways for young children are complex, and require additional data related to children's behavior. More comprehensive exposure assessment studies that include both exposure pathway and biological monitoring approaches will be needed to specify sources and to identify mitigation strategies for this population.

CONCLUSION

Children living with parents who work with agricultural pesticides, or who live in proximity to pesticide-treated farmland have higher exposures than do other children living in the same community. These children thus have additional exposure pathways beyond diet, drinking water, and residential pesticide use, the pathways considered common to all children. Further research efforts should be directed toward determination of the health risks that these exposure levels represent, and toward a better understanding of the importance of all exposure pathways in agricultural communities.

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