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Multimedia measurements and activity patterns in an observational pilot study of nine young children

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A pilot observational exposure study was performed to evaluate methods for collecting multimedia measurements (air, dust, food, urine) and activity patterns to assess potential exposures of young children to pesticides in their homes. Nine children (mean age = 5 years) and their caregivers participated in this study, performed in the Duval County, Florida, in collaboration with the Centers for Disease Control and Prevention and the Duval County Health Department. For all nine children, the total time reported for sleeping and napping ranged from 9.5 to 14 h per day, indoor quiet time from 0 to 5.5 h per day, indoor active time from 0.75 to 5.5 h per day, outdoor quiet time from 0 to 1.5 h per day, and outdoor active time from 0.5 to 6.5 h per day. Each home had one to three pesticide products present, with aerosols being most common. Pesticide inventories, however, were not useful for predicting pesticide levels in the home. Synthetic pyrethroids were the most frequently identified active ingredients in the products present in each home. Fifteen pesticide active ingredients were measured in the application area wipes (not detected (ND) to 580 ng/cm^2), 13 in the play area wipes (ND-117 ng/cm^2), and 14 in the indoor air samples (ND-378 ng/m^3) and the socks (ND-1000 ng/cm^2). *Cis*-permethrin, *trans*-permethrin, and cypermethrin were measured in all nine homes. Chlorpyrifos was measured in all nine homes even though it was not reported used by the participants. All urine samples contained measurable concentrations of 3-phenoxybenzoic acid (3-PBA). The median 3-PBA urinary concentration for the nine children was 2.2 μg/l. A wide variety of pesticide active ingredients were measured in these nine homes at median concentrations that were often higher than reported previously in similar studies. These data highlight the need for additional observational studies in regions where pesticides are used in order to understand the factors that affect young children's exposures and the education/mitigation strategies that can be used to reduce children's exposures.
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Introduction

Data on the factors affecting children's exposures and activities are inadequate to sufficiently assess residential exposures to environmental contaminants (Cohen Hubal et al., 2000a, b). Although recent research efforts have collected much needed data to improve our understanding of the potential exposures of young children in their homes, child care centers, and other environments (Zartarian et al., 1995, 1997, 1998; Bradman et al., 1997, 2007; Byrne et al., 1998; Gurunathan et al., 1998; Landrigan et al., 1999; Adgate et al., 2000, 2001; Fenske et al., 2000b, 2002; Freeman et al., 2001; Lu et al., 2001; MacIntosh et al., 2001; Pang et al., 2002; Clayton et al., 2003; Curl et al., 2003; Duggan et al., 2003; Shalat et al., 2003; Wilson et al., 2003, 2004; Hore et al., 2005; Morgan et al., 2005; Perera et al., 2006), it is not clear what factors most influence a child's potential exposure to pesticides.

Children's physiological characteristics may influence their exposures to pesticides in their environment either by affecting their rate of contact with various media or altering the exposure–uptake relationship. Children's behavior and the ways that they interact with their environment may also influence their potential exposures to pesticides in their environment. Developmental stage, physical activity, diet and eating habits, gender, socioeconomic status, and race/ethnicity are the factors that have been identified as potentially influencing a child's potential exposure to pesticides (Cohen Hubal et al., 2000a, b). Understanding these factors is important in evaluating a child's aggregate exposure to pesticides and identifying which factors most influence a child's potential exposure to pesticides. For example, is hand–to–mouth activity a factor that influences a child's indirect ingestion exposure?

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In addition to the child's physiological and behavioral characteristics, the physico-chemical characteristics of the pesticide and the activities in the household may also influence a child's potential exposure to pesticides. For example, a house that is cleaned on a daily basis will have a much lower pesticide residue concentration and potential for contact with pesticides than a house that is cleaned infrequently. Likewise, the pesticide residue concentration and potential for exposure to a bait is much less than to a fogger because of more widespread dispersion by a fogger in the home (Williams et al., 2006).

Researchers have evaluated whether residential location is a factor that influences a child's potential exposure to pesticides. Differences in urinary metabolite concentrations and housedust, soil, and handwipe concentrations for the organophosphorus pesticides (OPs) and their nonspecific metabolites have been found between children living in close proximity to agricultural areas and children living in suburban or urban areas in Washington State (Simcox et al., 1995; Loewenherz et al., 1997; Lu et al., 2000).

Researchers have also investigated whether the type of diet is a factor that influences a child's potential exposure to pesticides. Using a protocol that substituted an organic diet for a child's conventional diet for a 5-day period, Lu et al. (2006a) showed that young children's exposures to the agricultural pesticides chlorpyrifos and malathion can be significantly reduced. Lu et al. (2006a) reported that the median urinary concentrations of the specific metabolites for chlorpyrifos and malathion decreased to nondetectable levels after the introduction of the organic diet and remained at nondetectable levels until the child's conventional diet was re-introduced. Conversely, Lu et al. (2006b) used the same protocol for a pyrethroid study and concluded that residential pesticide use is the most important factor influencing a child's potential exposure to pyrethroid pesticides. Despite research efforts, data gaps still remain on the factors that most influence a child's potential exposure to pesticides.

This pilot study was one component of a large collaborative research study conducted by the United States Environmental Protection Agency (EPA), the Centers for Disease Control and Prevention (CDC), and the Duval County Health Department (DCHD) to evaluate young children's (4 to 6 years) potential exposures to current-use pesticides in the residential environment. All three organizations worked jointly on the design of the study with CDC conducting limited biomonitoring screening to assess young children's exposures to current-use pesticides in Jacksonville, FL, USA; DCHD evaluating environmental screening methods to identify elevated pesticide levels in homes; and EPA conducting an observational study to evaluate sampling and analysis methods for estimating aggregate and cumulative exposures to pesticides in residences under real-world conditions during normal day-to-day activities. Each organization had responsibility for a different aspect of the project. CDC provided overall project management, analysis of the urine samples, and data reporting on the biomonitoring screening component; DCHD was responsible for the environmental screening component of the study and served as the community liaison; and EPA was responsible for the observational study. EPA defines observational exposure measurement studies as studies that measure people's exposures to chemicals in their everyday environments during their normal daily activities. These studies involve measurements of chemicals in environmental media; collection of information about the study participants, their homes, their work environments, and their activities; and collection of personal exposure and biomarker measurements.

The EPA component of the study involved collection of multimedia samples and activity pattern data for young children in residential environments. This paper describes the collection methods, measured pesticide concentrations, and activity pattern data for the nine children who participated in the observational study.

Materials and methods

Recruitment of Participants

DCHD recruited participants for the main biomonitoring study at six of the health care centers operated by DCHD when they visited a center for routine health care during the summer and fall of 2001. A convenience sample of 201 children and their caregivers participated in the biomonitoring screening component of the study. From this total, 42 children and their caregivers participated in the environmental screening assessment conducted by DCHD. The nine children and their caregivers who participated in the observational measurement study were selected from the participants in the environmental screening assessment based on their willingness to participate in this component of the study and their reported frequent use of spray-type pesticide products in the pesticide inventory. Participant consent was obtained for each segment of the study. This was an observational research study, as defined in 40 CFR Part 26.402. The study protocol and procedures to obtain the assent of the children and informed consent of their parents or guardians were reviewed and approved by three independent institutional review boards (IRBs) and complied with all applicable requirements of the Common Rule regarding additional protections for children (Subpart D).

Field Protocol

We completed two visits with each participating family. During the first visit, we obtained the assent of the child and informed consent of his/her parents or guardians; completed the pesticide inventory; provided instructions on completing the time–activity diary and a completed example; provided

the cotton socks, duplicate diet collection containers, urine specimen collection cups and instructions on their uses, and collection schedules; and set up the indoor and outdoor air samplers. During the second visit, we reviewed the time–activity diary for completeness, collected all samples, and provided monetary compensation to the participants. The activity collection instruments and multimedia samples collected in this study included the time–activity diary, pesticide inventory, indoor and outdoor air samples, surface wipe samples, clothing (socks), duplicate diet, and urine samples.

Activity Collection Instruments

Time–Activity Diary

Each caregiver completed a 24-h time–activity diary for his/her participating child during the observational study. The diary was divided into two time periods, with time period one including the time from when the child woke up to lunch time and time period two including from lunch time to when the child went to sleep. Identical information was asked in each time period and included the time indoors, outdoors, and away from home; locations occupied; surfaces contacted (carpet/rugs, hard floor, upholstered furniture, bedding, grass, dirt, paved surfaces, wood, other); activities that the child engaged in; activity level (active play, quiet play, napping/sleeping); and type of clothing worn (long-sleeved clothing, short-sleeved clothing, legs and ankles covered, legs partially covered).

Pesticide Inventory

A pesticide inventory was completed by the field technician and the primary caregiver by inspection of containers and use of a recall survey. Information was collected for the brand name, type of product, EPA registration number, label copyright year, date last used, frequency of use in the last 6 months, purpose for use, and where applied in the home. Information was also collected for any professional applications and/or for products that may have recently been used, but for which there were no longer containers of the product in the home.

Multimedia Samples

Indoor and outdoor air samples were collected at each participant's home during a 24-h monitoring event with a pre-cleaned polyurethane foam (PUF) plug attached to a constant-flow battery powered pump operating at a flow rate of approximately 3.8 l/min. Indoor air samples were collected in the main play area of the house from a height approximating the child's breathing zone (1 m as measured from the floor), while outdoor samples were collected from the front yard at a height of 1.5 m, similar to the method used by Bradman et al. (2007).

One surface wipe sample was collected from the main play area of the house and another surface wipe sample was collected from an area where the caregiver reported past pesticide application(s). Wipe samples were collected from a 929 cm^2 area on a hard surface using Johnson & Johnson SOF-WICK™ dressing sponges wetted with 10 ml of isopropanol (Tulve et al., 2006a).

Duplicate diet samples were collected during the 24-h monitoring period. To collect the duplicate diet, each caregiver was asked to monitor the amount of food consumed by the participating child during breakfast, lunch, dinner, and snack times, and place an exact copy of the foods consumed into the solid food and liquid food sampling containers (Thomas et al., 1997; Bradman et al., 2007). The food sampling containers were stored in the participant's refrigerator until the field technicians arrived at the home to collect the samples.

Socks (80% cotton/20% polyester, Cuddly Soft Brand, Childrenswear Centres, Piscataway, NJ, USA, www.childrenswearcentre.com) were used to estimate the amount of pesticide residue that could be on the participating child's skin after normal play activities. Children wore the socks for a 1-h or longer period while at home and engaged in normal play activities. The caregiver was asked to record the time the socks were put on and the time that the socks were taken off.

Each participating child collected a morning void urine sample during the 24-h monitoring period. The procedure used to collect the sample has been published previously by the Centers for Disease Control and Prevention (CDC, 2003). The urine sample was stored in a cooler with frozen ice packs until the field technicians arrived at the home to collect the environmental samples. All data were collected in October, 2001.

Multi-Residue Analysis Method

Environmental samples were analyzed by a multi-residue method that involved solvent extraction, solid-phase extraction (SPE) clean-up, and analysis by gas chromatography/mass spectrometry (GC/MS) (Tulve et al., 2006a). Details for the extraction and clean-up of the various matrices can be found in Bradman et al. (2007). All solvents were high purity and purchased from Fisher Scientific. The method included the analysis of 22 organophosphate pesticides, 13 synthetic pyrethroid pesticides, two natural pyrethroid pesticides, one synergist (piperonyl butoxide), and one phenyl pyrazole (fipronil). The method included the addition of surrogate recovery standards (SRSs) representative of the major compound classes. The internal standard (IS) method of quantification was used for all analytes and SRSs, and analyte concentrations were adjusted by the appropriate SRS recovery (Tulve et al., 2006a).

Wipe samples were extracted using accelerated solvent extraction (ASE) with dichloromethane (DCM), concentrated, solvent exchanged into hexane, loaded onto a silica SPE cartridge (1 g BakerBond, JT Baker) and eluted with a

series of solvents (hexane (3 ml), 15% diethyl ether in hexane (two aliquots of 6 ml each), DCM (6 ml), 20% acetone in ethyl acetate (three aliquots of 6 ml each)), concentrated again, and fortified with IS (Tulve et al., 2006a). Laboratory and field quality control samples were processed in sequence with the field samples.

Air samples were extracted using ASE with acetone:hexane (10:90, v:v), then processed in a manner identical to that described for the wipes (Bradman et al., 2007).

A 5×5 cm section of the sock corresponding to the ball of the participant's foot was cut from the sock for analysis. This sock sample was extracted using ASE with DCM. Laboratory and field quality control samples were prepared with 5×5 cm sock sections. Due to a laboratory error, the field spike and blank samples were not available for analysis.

A 10 g aliquot of the homogenized food sample was fortified with SRS (5 ng), equilibrated, and combined with Extrelute (5 g, EM Sciences). This sample was homogenized with acetonitrile (100 ml), centrifuged, and decanted into a separatory funnel containing a saturated NaCl solution (30 ml). This process was repeated twice for each sample. After partitioning, the aqueous layer was discarded. After drying over sodium sulfate, the extract was solvent exchanged into acetone and then eluted through a series of SPE cartridges (1 g alumina, 1 g Envicarb, 1 g aminopropyl) with acetone: toluene (four aliquots of 6 ml of 3:1, v:v). The resulting extract was eluted a second time through this same series of SPE cartridges with acetone:toluene (two aliquots of 6 ml of 3:1, v:v), concentrated, re-diluted with hexane, and eluted through a silica SPE cartridge using hexane (3 ml) and 60% ethyl ether in hexane (6 ml). This final extract was used for analysis.

A seven-point calibration curve of analytes and SRSs was prepared that spanned the concentration range of 2.5–50 $\times$ the instrument detection limit of each analyte. Samples and standards were analyzed using an Agilent/HP 6890 gas chromatograph/5973 mass selective detector in the multiple ion detection mode using an embedded standard approach in which the standards were interspersed with the field samples within the run sequence. For samples where an analyte(s) exceeded the maximum calibration concentration by $>15\%$, the solution was diluted, re-spiked with IS, and re-analyzed (Tulve et al., 2006a).

Urine samples were extracted and analyzed using published methods (Baker et al., 2004; Olsson et al., 2004; Bradman et al., 2007).

In developing the database for analysis, the wipe samples were blank- and surrogate-recovery corrected.

Results

Quality Control Data

The performance of the multi-residue analysis method has been previously discussed in Tulve et al. (2006a). For this manuscript, the quality control data for the 12 synthetic pyrethroid pesticides (*cis*-permethrin, *trans*-permethrin, cyfluthrin, cypermethrin, *cis*-allethrin, *trans*-allethrin, delta/tralomethrin, esfenvalerate, *lambda*-cyhalothrin, bifenthrin, sumithrin, tetramethrin), two natural pyrethroid pesticides (pyrethrins I and II), an organophosphate pesticide (chlorpyrifos), a synergist (piperonyl butoxide) and a phenyl pyrazole (fipronil) will be presented since these were the pesticide products measured in the homes.

The method detection limit (MDL) was based on instrumental performance only, as equivalent clean-up was achieved with all matrices. The MDL was determined as the analyte level giving 3:1 S:N in a wipe-extract fortified with a known amount of analyte just before the GC/MS analysis. The method detection limits (MDLs) ranged from 0.002 to 0.016 ng/cm^2 for wipe samples, 0.4 to 3 ng/m^3 for air samples, 0.08 to 0.8 ng/cm^2 for sock samples, and 0.02 to 0.4 ng/g for duplicate diet samples (Table 1).

Three field matrix blanks and eight laboratory matrix blanks were analyzed with the 18 field wipe samples. Concentrations (mean $\pm$ SD, ng/cm^2) of the target analytes in the field wipe matrix blanks were as follows: pyrethrin II (0.01 ± 0.01), *cis*-permethrin (0.02 ± 0.04), *trans*-permethrin (0.01 ± 0.01), chlorpyrifos (0.03 ± 0.05), and piperonyl butoxide (0.02 ± 0.04). For the laboratory wipe matrix blanks, the concentrations (mean $\pm$ standard deviation, ng/cm^2) were: *cis*-allethrin (0.002 ± 0.01), cypermethrin (0.2 ± 0.5), *cis*-permethrin (0.01 ± 0.02), *trans*-permethrin (0.2 ± 0.4), chlorpyrifos (0.004 ± 0.01), and piperonyl butoxide (0.1 ± 0.1).

Three field matrix blanks and three laboratory matrix blanks were analyzed with the 18 field air samples. Concentrations (mean $\pm$ SD, ng/m^3) of the target analytes in the field PUF matrix blanks were as follows: pyrethrin II (1.9 ± 3.4), *cis*-permethrin (1.5 ± 0.12), *trans*-permethrin (2.2 ± 0.9), and tetramethrin (0.5 ± 0.43). For the laboratory PUF matrix blanks, the concentrations (mean $\pm$ SD, ng/m^3) were: bifenthrin (0.3 ± 0.5), pyrethrin II (0.4 ± 0.7), *cis*-permethrin (1.2 ± 1.0), *trans*-permethrin (1.2 ± 1.1), and tetramethrin (0.1 ± 0.1).

The laboratory solvent method blanks for the food analyses only showed trace levels of piperonyl butoxide (equivalent to 0.02 ng/g) in one blank. The only analytes detected in the laboratory food matrix blanks were *cis*-permethrin and *trans*-permethrin at 0.19 and 0.11 ng/g, respectively.

Although the pesticide levels in the wipe blanks were low, they were measurable and statistically different from zero. As a result, all wipe samples were corrected for field blank levels. No other sample media were corrected by the corresponding field blank levels.

For each medium type, corresponding laboratory spike samples were prepared and processed. For the eight wipe samples, the mean recovery averaged $108 \pm 64\%$ and the

Table 1. Method detection limits (MDLs) and detection frequencies (DFs) for the target analytes in multimedia samples ($N = 9$).

Analyte	Outdoor air (ng/m^3)		Indoor air (ng/m^3)		Wipe, Appl Area (ng/cm^2)		Wipe, Play Area (ng/cm^2)		Sock (ng/cm^2)		Food (ng/g)	
	MDL	DF (%)	MDL	DF (%)	MDL	DF (%)	MDL	DF (%)	MDL	DF (%)	MDL	DF (%)
Pyrethroids and Pyrethrins												
cis-Allethrin	1.0	0	1.0	33	0.005	56	0.005	33	0.20	33	0.2	0
trans-Allethrin	1.0	0	1.0	33	0.005	56	0.005	33	0.20	33	0.2	0
Bifenthrin	1.0	0	1.0	11	0.005	11	0.005	22	0.20	22	0.02	33
Cyfluthrin[a]	1.2	0	1.2	11	0.006	33	0.006	11	0.24	0	0.1	22
λ-Cyhalothrin	1.0	0	1.0	0	0.005	22	0.005	11	0.20	11	0.04	0
Cypermethrin[a]	1.2	22	1.2	22	0.006	89	0.006	67	0.24	67	0.1	67
Deltamethrin	3.0	0	3.0	0	0.016	22	0.016	0	0.60	11	0.4	11
Esfenvalerate	1.4	0	1.4	0	0.008	11	0.008	0	0.28	22	0.2	0
cis-Permethrin	1.0	100	1.0	89	0.005	78	0.005	67	0.80	80 ($N = 8$)	0.02	78
trans-Permethrin	1.0	100	1.0	89	0.005	78	0.005	78	0.20	100	0.02	78
Pyrethrin I	2.0	0	2.0	44	0.011	11	0.011	0	0.40	0	NR	0
Pyrethrin II	0.8	0	0.8	11	0.011	0	0.011	0	0.16	0	NR	0
Sumithrin	1.0	0	1.0	11	0.005	0	0.005	33	0.20	44	0.04	11
Tetramethrin	0.4	0	0.4	22	0.002	11	0.002	33	0.08	22	0.04	22
Other												
Piperonyl Butoxide	1.0	33	1.0	89	0.005	75 ($N = 5$)	0.005	67	0.20	78	0.04	100
Chlorpyrifos	1.0	56	1.0	100	0.005	78	0.005	56	0.40	100	0.04	100
Fipronil	1.0	0	1.0	0	0.005	33	0.005	22	0.20	33	NM	NM

Abbreviations: Appl, application; DF, detection frequency; MDL, method detection limit; NR, not recovered; NM, not measured.
[a]Detection limit when all chromatographically resolved isomers are detected.

SRS recoveries were $69 \pm 26\%$ for fenchlorphos and $71 \pm 21\%$ for $^{13}C_6$-*trans*-permethrin. For the three air samples, the mean recovery averaged $78 \pm 33\%$ and the SRS recoveries were $83 \pm 5\%$ for fenchlorphos and $88 \pm 4\%$ for $^{13}C_6$-*trans*-permethrin. For the two reference food samples, the mean recovery averaged $84 \pm 20\%$, and the SRS recoveries were $83 \pm 16\%$ for fenchlorphos and $98 \pm 50\%$ for $^{13}C_6$-*trans*-permethrin.

Nine wipe and three air samples were spiked with 11 target pesticides for use as field controls. The recoveries ranged from 74 to 131% for the wipe sample field controls and 62 to 119% for the air sample field controls.

Participant Demographics

Nine children (five males, four females; M = 5 years; SD = 1 year) and their caregivers participated in this component of the study. Six children attended school during the monitoring period (ranging from 7.5 to 9.5 h per day). For all nine children, the total time reported for sleeping and napping ranged from 9.5 to 14 h per day, indoor quiet time ranged from 0 to 5.5 h per day, indoor active time ranged from 0.75 to 5.5 h per day, outdoor quiet time ranged from 0 to 1.5 h per day, and outdoor active time ranged from 0.5 to 6.5 h per day. In situations where the time reported in the time–activity diary did not sum to 24 h, we estimated the hours based on the information available in the time–activity diary.

Distribution of Pesticides in the Residences

Table 2 presents the results of the information collected in the pesticide inventory for each participant. At the time of the study, each home had one to three products present, with aerosols being the most common. All homes reported recent and frequent pesticide use (Table 2; pesticide inventory questions: *When was the last time it was used?* and *In the last 6 months, how often was it used?*). Roaches and ants were the most commonly reported bug problems. All participants reported applying a pesticide in the kitchen, and most reported applying a pesticide in more than one room of the house. Synthetic pyrethroids were the most frequently identified active ingredients in products present in each home at the time of the study.

Table 3 presents the pesticide concentrations in the air, wipe, sock, and duplicate diet (food) samples for 17 pesticides measured in the homes. Fourteen of the 17 pesticides were detected in the indoor air samples, whereas cypermethrin, *cis*-permethrin, *trans*-permethrin, tetramethrin, piperonyl butoxide, and chlorpyrifos were the only compounds measured in the outdoor air samples. Both the application area wipes and the play area wipes had numerous pesticide residues measured, 15 and 13 pesticides, respectively. Of the pesticides reported in this study, cyfluthrin and the natural pyrethrins (pyrethrins I and II) were the only pesticides not detected in the sock samples. Numerous pesticide residues were also measured in the food samples (Table 3).

Table 4 highlights four pesticides that were measured in all homes (*cis*-permethrin, *trans*-permethrin, cypermethrin, chlorpyrifos). Chlorpyrifos was detected in the highest concentrations in the indoor air in six (Homes 1, 2, 6, 7, 8, 9) of the nine homes (Table 4) even though it was not

Table 2. Information collected in the pesticide inventories from the nine homes.

Home	Type	When was the last time it was used?	In the last 6 months, how often was it used? (times per month)	What was the pesticide used for?	Where was the pesticide applied?	Active ingredients in product
1	Aerosol	1 day ago	8	Roaches, ants	Kitchen	Propoxur, pyrethrins I and II, cyfluthrin, piperonyl butoxide
1	Aerosol	4 days ago	8	Roaches, ants	Kitchen, bathroom	Propoxur, pyrethrins I and II, cyfluthrin, piperonyl butoxide
2	Aerosol	2 days ago	2	Roaches	Kitchen, bathroom	Cypermethrin, imiprothrin
3	Aerosol	1 day ago	4	Roaches, ants	Kitchen, living room, family room, bedroom	Cypermethrin, imiprothrin
4	Aerosol	1 day ago	12	Roaches	Kitchen, living room, bedroom, bathroom	Tralomethrin, d-*trans*-allethrin
4	Fogger	30 days ago	<1	Roaches	All rooms	Permethrin, sumithrin, piperonyl butoxide
4	Fogger	30 days ago	<1	Roaches	All rooms	Permethrin, sumithrin, piperonyl butoxide
5	Aerosol	1 day ago	4	Roaches, ants	Kitchen, living room, bathroom	Cypermethrin, imiprothrin
6	Aerosol	1 day ago	5	Flies, ants, mosquitoes	Kitchen, front yard, living room	Pyrethrins I and II, piperonyl butoxide
6	Aerosol	30 days ago	10	Mosquitoes	On body	DEET
6	Bait station	7 days ago	<1	Roaches	On countertops, in cabinets	—[a]
7	Pump spray	1 day ago	1	Roaches, ants	Kitchen, living room	—[a]
7	Aerosol	1 day ago	8	Roaches, ants	Kitchen, living room	Cypermethrin, imiprothrin
8	—[a]	1 day ago	60	Roaches	Kitchen, bathroom	—[a]
8	Aerosol	—[a]	—[a]	—[a]	—[a]	Tralomethrin, d-*trans*-allethrin
9	Granules	14 days ago	<1	Ants	Backyard	—[a]
9	Aerosol	3 days ago	1.25	Roaches, ants	Kitchen, bathroom	Permethrin, pyrethrins I and II, piperonyl butoxide

[a]Information not available.

reported used in the last 6 months. For the indoor samples (indoor air, application area floor wipes, play area floor wipes, socks), the cypermethrin concentrations measured in homes 3 and 5 were much higher than the other pesticide residues measured in the same media in these homes. In general, the concentrations of the three pyrethroids measured in the wipe samples are higher than the concentrations of chlorpyrifos measured in the wipe samples, suggesting that the pyrethroids are more likely to be bound to a surface or dust particles (the exceptions are Homes 1 and 2). All four pesticides were measured on the socks with four homes (Homes 3, 4, 8, 9) having all pesticide residues detected. Five homes (Homes 1, 2, 5, 6, 8) had all four pesticides measured in the food samples.

Table 5 shows a comparison of the pesticide concentrations in the multimedia samples across three recent studies where numerous pyrethroid and OP pesticides were measured. Where comparable, the median pesticide residue concentrations in the air, application area floor wipes, and food were usually higher for this study as compared to the Bradman et al. (2007) study or the CTEPP (A Pilot Study of Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants) study (Morgan et al., 2004).

Each participating child provided a morning urine sample. All urine samples contained measurable concentrations of 3-phenoxybenzoic acid (3-PBA), *cis*/*trans*-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane carboxylic acid (*cis*/*trans*-DCCA), and 3,5,6-trichloro-2-pyridinol (TCPy) (Table 6). In addition, all nine children had measurable levels of 4-fluoro-3-phenoxybenzoic acid (4F-3-PBA) in their urine, with four children having 4F-3-PBA metabolite concentrations slightly less than the limit of detection (0.05 μg/l) reported in Table 6. The TCPy concentrations are within the range of TCPy concentrations reported by Morgan et al. (2005), but the median TCPy concentration of 9.8 μg/l is near the 90th percentile (10.7 μg/l) for the 6–11 year old age group in the Third National Report on Human Exposure to Environmental Chemicals (NHANES) (CDC, 2005). Becker et al. (2006) reported a 4F-3-PBA median concentration of less than 0.1 μg/l from a sample size of 395 participants in

Table 3. Pesticide concentrations in multimedia samples ($N = 9$)[a].

Analyte	Outdoor Air (ng/m^3)		Indoor Air (ng/m^3)		Wipe, Appl Area[b] (ng/cm^2)		Wipe, Play Area (ng/cm^2)		Sock[c] (ng/cm^2)		Food (ng/g)	
	p50	max	p50	max	p50	max	p50	max	p50	max	p50	max
Pyrethroids and pyrethrins												
Cis-Allethrin	—[d]	—[d]	—[d]	74	0.20	31	—[d]	0.11	—[d]	180	NM	NM
trans-Allethrin	—[d]	—[d]	—[d]	38	0.09	15	—[d]	0.05	—[d]	96	NM	NM
Bifenthrin	—[d]	—[d]	—[d]	3.0	—[d]	2.5	—[d]	0.05	—[d]	68	—[d]	1.3
Cyfluthrin[e]	—[d]	—[d]	—[d]	5.5	—[d]	10	—[d]	3.4	—[d]	—[d]	—[d]	3.6
λ-Cyhalothrin	—[d]	—[d]	—[d]	—[d]	—[d]	3.7	—[d]	0.12	—[d]	14	—[d]	—[d]
Cypermethrin[e]	—[d]	19	—[d]	100	6.9	580	0.68	18	8.7	1000	2.3	9.5
Delta/Tralomethrin	—[d]	—[d]	—[d]	—[d]	—[d]	1.8	—[d]	—[d]	—[d]	13	—[d]	13
Esfenvalerate	—[d]	—[d]	—[d]	0.32	—[d]	0.7	—[d]	—[d]	—[d]	2.6	—[d]	—[d]
cis-Permethrin	2.1	2.3	2.0	92	0.24	42	0.04	9.8	7.7	130	0.29	13
trans-Permethrin	2.5	10	3.1	130	0.34	67	0.05	14	1.4	180	0.22	22
Pyrethrin I	—[d]	—[d]	—[d]	12	—[d]	1.4	—[d]	—[d]	—[d]	—[d]	—[d]	—[d]
Pyrethrin II	—[d]	—[d]	—[d]	0.91	—[d]	—[d]	—[d]	—[d]	—[d]	—[d]	—[d]	—[d]
Sumithrin	—[d]	—[d]	—[d]	4.2	—[d]	—[d]	—[d]	1.36	—[d]	31	—[d]	0.11
Tetramethrin	—[d]	0.15	0.15	63	—[d]	4.6	—[d]	0.28	—[d]	30	—[d]	0.13
Other												
Piperonyl Butoxide	—[d]	3.1	7.4	378	1.4	264	0.35	117	7.9	85	0.36	1.1
Chlorpyrifos	3.8	6.6	20	85	0.39	3.1	0.006	2.3	2.2	5.1	0.38	7.4
Fipronil	—[d]	—[d]	—[d]	—[d]	—[d]	2.3	—[d]	0.03	—[d]	4.3	NM	NM

Abbreviations: Appl, application; NM, not measured; p50, median; max, maximum.
[a]Analyte concentrations presented only when detectable levels were measured in one or more media.
[b]$N = 8$ for piperonyl butoxide application area wipe sample measurement.
[c]$N = 5$ for *cis*- and *trans*-permethrin sock sample measurements.
[d]At this percentile, all values were below the detection limit.
[e]Sum of all chromatographically-resolved isomers.

which less than 1% of the samples were greater than or equal to the limit of quantification (LOQ = 0.1 μg/l). The 3-PBA and *trans*-DCCA median concentrations (2.2 μg/l and 1.1 μg/l, respectively) fall within the 95% confidence interval of the 90th percentile (1.34–2.69 μg/l for 3-PBA and 1.03–1.68 μg/l for *trans*-DCCA) for the age group 6–11 years (NHANES). The median concentration of *cis*-DCCA (0.62 μg/l) falls within the 95% confidence interval (0.49–0.87 μg/l) of the 95th percentile for the age group 6–11 years (NHANES) (CDC, 2005).

Table 7 presents a Spearman rank sum correlation matrix for cypermethrin, *cis*-permethrin, *trans*-permethrin, and chlorpyrifos. For cypermethrin, significant correlations existed among all parameters evaluated (except the food and creatinine-corrected 3-PBA) and indoor and outdoor air, application area floor wipes, and the sock samples, whereas no relationship existed between food and the play area floor wipes and any of the parameters evaluated (except the play area floor wipes and total DCCA). For *cis*-permethrin and *trans*-permethrin, significant correlations existed among indoor air, application area floor wipes, play area floor wipes, and the sock samples. A significant association also existed between food and 3-PBA (*cis*-permethrin and *trans*-permethrin) and food and *cis*-DCCA (*cis*-permethrin). For chlorpyrifos, the only significant association was between the application area floor wipes and the sock samples.

Discussion

We collected multimedia samples in homes for a subset of children who participated in a larger biomonitoring survey. Measurable levels of pyrethroids, OPs, a synergist, and a phenyl pyrazole were detected in indoor and outdoor air, application and play area floor wipes, socks, and food samples. Metabolites corresponding to the parent pesticides were also measured in the children's urine.

We also collected time–activity information for the nine participating children. Using a paper diary, each caregiver recorded location and activity information, time spent on various surface types (carpet/rugs, hard floor, upholstered furniture, bedding, grass, dirt, paved surfaces, wood, other), and the type of clothing worn, while engaged in that particular activity. In an attempt to make the diary as easy as possible for the participants, the diary was divided into morning and afternoon sections. We asked the participants to complete the diary in real time.

One participant child (Home 5) spent approximately 27% of his time outdoors, as compared to the others who averaged 6.4%. The total average outdoor time for all children was 9%. Sleeping averaged 47% of the children's time and indoor time awake at home averaged 21%. Total time away from home averaged 23% for all children, but averaged 34% for the six children (homes 2, 3, 4, 6, 8, 9), who were in

Table 4. Concentrations of selected pesticides measured in all homes.

Pesticide	Home ID Number								
	1	2	3	4	5	6	7	8	9
Outdoor Air (ng/m^3)									
Cypermethrin	—[a]	—[a]	19.2	—[a]	3.2	—[a]	—[a]	—[a]	—[a]
cis-Permethrin	1.9	2.3	2.2	2.1	2.3	1.7	1.7	2.2	1.8
trans-Permethrin	4.6	5.7	10.2	2.1	3.0	1.8	1.7	2.5	2.1
Chlorpyrifos	5.1	4.2	6.6	—[a]	4.9	3.8	—[a]	—[a]	—[a]
Indoor Air (ng/m^3)									
Cypermethrin	—[a]	—[a]	64.8	—[a]	105	—[a]	—[a]	—[a]	—[a]
cis-Permethrin	1.9	2.0	2.2	93	1.8	2.0	1.9	—[a]	5.6
trans-Permethrin	3.1	6.0	6.4	134	1.9	2.1	1.8	—[a]	6.6
Chlorpyrifos	32.4	84.9	18.4	18.3	20.4	28.1	9.8	45.5	12.6
Application area surface wipe (ng/cm^2)									
Cypermethrin	—[a]	6.9	580	39	198	0.8	3.7	72	0.8
cis-Permethrin	0.1	0.2	1.7	42	0.2	0.5	—[a]	—[a]	32
trans-Permethrin	0.2	0.3	3.5	67	0.1	0.7	—[a]	—[a]	45
Chlorpyrifos	0.2	0.7	3.1	0.4	—[a]	0.3	—[a]	2.4	0.5
Play area surface wipe (ng/cm^2)									
Cypermethrin	—[a]	—[a]	18	0.4	10	0.7	1.0	1.0	—[a]
cis-Permethrin	—[a]	0.01	2.7	0.9	0.04	0.7	—[a]	Missing	10
trans-Permethrin	<0.01	0.03	2.2	1.5	0.05	1.2	—[a]	0.02	14
Chlorpyrifos	—[a]	0.04	2.3	—[a]	0.01	<0.01	0.5	0.01	—[a]
Socks (ng/cm^2)									
Cypermethrin	—[a]	2.9	173	70	408	—[a]	—[a]	2.5	0.3
cis-Permethrin	Missing	—[a]	3.8	64	—[a]	—[a]	—[a]	0.1	0.6
trans-Permethrin	0.6	0.5	5.8	90	0.4	1.1	0.4	0.1	0.8
Chlorpyrifos	0.7	1.0	2.5	0.9	0.4	0.8	1.3	0.2	0.2
Food (ng/g)									
Cypermethrin	1.5	7.6	5.4	—[a]	3.8	9.5	—[a]	2.3	—[a]
cis-Permethrin	0.3	12.9	—[a]	—[a]	0.1	0.3	0.4	0.1	0.5
trans-Permethrin	2.1	22.1	—[a]	—[a]	0.2	0.2	0.2	0.2	0.5
Chlorpyrifos[b]	2.7	1.9	1.0	15	0.2	1.8	0.5	0.7	0.6

[a]Not detected in media.
[b]Sum chlorpyrifos and chlorpyrifos methyl concentrations in solid food.

school at the time of the observation period. Our results are similar to other published results on how and where children spend their time (Schwab et al., 1992; Silvers et al., 1994; Elgethun et al., 2003).

The paper diary did not work very well. The participants found the morning and afternoon sections difficult to complete when the field technicians did not arrive in the morning. Furthermore, too many questions needed to be answered with text and the hours did not always sum to 24. During the second visit (as described in the field protocol), the field technicians spent a fair amount of time helping the caregivers complete the time–activity diary. Based on these observations, the time–activity diary was redesigned and pilot tested (Tulve et al., 2007).

We used simple regression to analyze the responses to the questions in the pesticide inventory (application types, days since last use, frequency of use, and rooms treated) in an attempt to determine which questions may be the most useful in predicting pesticide levels in the home. The only association with logged total surface residue loading (μM/cm^2, summed over all measured pyrethrins and pyrethroids) was found with the number of types of rooms treated (P-value = 0.06). The questions were specific to each pesticide product inventoried. These inventories may adequately capture the products in the home at the time of the inventory, but they are not useful for collecting information on other pesticide products used but discarded or never stored in the home. As a result, the inventories were not useful for predicting pesticide levels in the home. Sexton et al. (2003) also reported that their questionnaire screening approach was ineffective in identifying households with higher levels of the individual target pesticides.

Table 5. Comparison of the pesticide concentrations in multimedia samples across recent studies.

Sample	Pesticide	Study[a]							
		This Study		Bradman et al. (2007)		CTEPP-NC		CTEPP-OH	
		Median	Max	Median	Max	Median	Max	Median	Max
Air[b] (ng/m^3)	*cis*-Permethrin	2.0	92	<0.56[c]	8.2	0.58	34.4	<0.39[c]	5.39
	trans-Permethrin	3.1	130	<0.56[c]	11	0.36	40.9	<0.33[c]	6.8
	Cypermethrin	<1.2[c]	100	<28[c]	380	NM	NM	NM	NM
	Chlorpyrifos	20	85	11	36	6.21	391	1.67	98
Wipe[d] (ng/cm^2)	*cis*-Permethrin	0.24, 0.04	9.8	0.10	1.7	0.044	0.87	0.009	5.2
	trans-Permethrin	0.34, 0.05	14	0.23	3.6	0.04	1.01	0.0094	5.18
	Cypermethrin	6.9, 0.68	18	<0.22[c]	2.8	NM	NM	NM	NM
	Chlorpyrifos	0.39, 0.006	2.3	0.046	0.2	0.007	0.21	0.003	3.86
Food (ng/g)	*cis*-Permethrin	0.29	13	<4.5[c]	<4.5[c]	<0.08[c]	80.7	<0.08[c]	560
	trans-Permethrin	0.22	22	<2.9[c]	<2.9[c]	<0.08[c]	70.4	<0.08[c]	448
	Cypermethrin	2.3	9.5	NM	NM	NM	NM	NM	NM
	Chlorpyrifos	0.38	7.4	<1.4[c]	<1.4[c]	0.19	19.7	0.19	3.51

Abbreviation: NM, not measured.
[a]This Study: $N=9$; Bradman et al. (2007)'s study: $N=20$; CTEPP-NC: $N=128$ (air), $N=28$ (wipe), $N=129$ (food, *cis*-permethrin, and chlorpyrifos), $N=128$ (food, *trans*-permethrin); CTEPP-OH: $N=125$ (air), $N=21$ (wipe), $N=125$ (food).
[b]Indoor air.
[c]Reported method detection limit.
[d]This study wipe sample: application area wipe, play area wipe; Bradman et al. (2007)'s study wipe sample: floor wipe collected from a central location, usually the kitchen or dining area near the boundary with a carpeted floor; CTEPP-NC and CTEPP-OH wipe samples: hard floor surface wipes collected from an indoor floor where the children spent most of their time.

Table 6. Limits of detection (μg/l), detection frequencies (%), and concentrations (μg/l) for the target analytes in urine samples ($N=9$ children).

Analyte	Parent Compound[a]	LOD (μg/l)	DF (%)	p50	p75	Max
Pyrethroids						
3-Phenoxybenzoic acid (3-PBA)	Cypermethrin, Deltamethrin, Permethrin	0.52	100	2.2	29	99
4-Fluoro-3-phenoxybenzoic acid (4F-3-PBA)	Cyfluthrin	0.05	56	0.09	0.26	1.7
cis-3-(2,2-Dichlorovinyl)-2,2-dimethlycyclopropane carboxylic acid (*cis*-DCCA)	Cyfluthrin, *cis*-Cypermethrin, *cis*-Permethrin	0.19	100	0.62	3.9	43
trans-3-(2,2-Dichlorovinyl)-2,2-dimethlycyclopropane carboxylic acid (*trans*-DCCA)	Cyfluthrin, *trans*-Cypermethrin, *trans*-Permethrin	0.21	100	1.1	6.9	61
Organophosphates						
3,5,6-Trichloro-2-pyridinol (TCPy)	Chloryrifos	0.40	100	9.8	15	21

[a]Only parent compounds relevant to the environmental sampling are included.

Floor wipe samples had the largest variety of pesticides detected, as compared to the other media. Outdoor air had the least number of pesticides detected. With the discontinued registration on the use of chlorpyrifos and diazinon in the indoor residential environment, pyrethroids are being increasingly used in the indoor environment. Although chlorpyrifos was not reported used, it was measured in all media, suggesting that it has a long residence time in the indoor environment (Fenske et al., 2000a, b). Piperonyl butoxide is added as a synergist to many pyrethroid mixtures for products used in the residential environment so it was not surprising to find it in all media (Selim et al., 1999).

Cypermethrin, *cis*-permethrin, *trans*-permethrin, and chlorpyrifos were measured in all nine homes. For homes 3 and 5, cypermethrin was measured in the highest concentration in all indoor media (indoor air, wipes, socks, food) following a reported pesticide application in numerous rooms in the home, including the kitchen. Lu et al. (2006b) suggest that for pyrethroids, the residential concentrations are most important for estimating exposure. His results, combined with the observations in this paper, would suggest that residential pesticide usage where pyrethroids are the active ingredients, and not just dietary intake, is important for estimating a child's exposure. However, due to the small sample size of this study, further research is needed to evaluate the impact of residential pesticide usage on the factors influencing a child's exposure.

Table 7. Spearman rank sum correlation matrix for the most frequently detected analytes across sampling media.

	Outdoor Air	Indoor Air	Wipe Appl	Wipe Play	Sock	Food
Cypermethrin						
Outdoor Air	1					
Indoor Air	0.97**	1				
Wipe Appl	0.73*	0.71*	1			
Wipe Play	0.74*	0.72*	0.81**	1		
Sock	0.72*	0.74*	0.88**	0.57	1	
Food	0.32	0.30	0.27	0.24	0.12	1
3-PBA	0.71*	0.73*	0.75*	0.64	0.81**	0.34
cis-DCCA	0.71*	0.73*	0.75*	0.66	0.78*	0.25
trans-DCCA	0.71*	0.73*	0.68*	0.61	0.75*	0.41
tot_DCCA	0.71*	0.73*	0.72*	0.68*	0.71*	0.41
cre_3-PBA	0.59	0.64	0.68*	0.44	0.80*	0.41
cre_*cis*-DCCA	0.71*	0.73*	0.73*	0.53	0.73*	0.22
cre_*trans*-DCCA	0.71*	0.73*	0.73*	0.46	0.83**	0.39
tot_cre_DCCA	0.71*	0.73*	0.68*	0.44	0.76*	0.34
cis-Permethrin						
Outdoor air	1					
Indoor air	−0.35	1				
Wipe appl	−0.11	0.94**	1			
Wipe play	−0.10	0.81**	0.92**	1		
Sock ($N=5$)	0.10	0.90*	0.97**	0.67	1	
Food	−0.29	−0.08	−0.21	−0.20	−0.31	1
3-PBA	0.57	−0.03	0.23	0.25	0.50	−0.86**
cis-DCCA	0.60	−0.15	0.07	0.07	0.20	−0.90**
cre_3-PBA	0.52	0.23	0.51	0.49	0.70	−0.63
cre_*cis*-DCCA	0.47	0.18	0.36	0.41	0.30	−0.43
trans-Permethrin						
Outdoor air	1					
Indoor air	0.23	1				
Wipe appl	0.08	0.95**	1			
Wipe play	0.10	0.73*	0.85**	1		
Sock ($N=5$)	0.12	0.73*	0.85**	0.82**	1	
Food	−0.01	−0.03	−0.13	−0.29	−0.44	1
3-PBA	0.47	0.05	0.14	0.33	0.32	−0.68*
trans-DCCA	0.42	0.10	0.22	0.38	0.33	−0.63
cre_3-PBA	0.33	0.28	0.39	0.53	0.42	−0.53
cre_*trans*-DCCA	0.38	0.35	0.41	0.60	0.37	−0.46
Chlorpyrifos						
Outdoor air	1					
Indoor air	0.37	1				
Wipe appl	0.07	0.32	1			
Wipe play	0.33	0.00	0.17	1		
Sock	0.07	0.13	0.91**	0.12	1	
Food	0.09	0.33	0.53	−0.27	0.40	1
TCPy	0.01	0.33	0.58	0.36	0.38	0.52

Abbreviation: cre, creatinine-corrected.
*Statistically significant Spearman ρ ($P<0.05$).
**Statistically significant Spearman ρ ($P<0.01$).

Few data are available to compare the concentrations of active ingredients measured in this study with other relevant residential environments, but where comparable, the data collected in this study have higher concentrations (Table 5). The median indoor air concentrations for *cis*-permethrin, *trans*-permethrin, and chlorpyrifos in this study are higher than both the Bradman et al. (2007) and CTEPP studies. Cypermethrin was not measured in the CTEPP study, and the method detection limits between this study and the Bradman et al. (2007) study make it impossible to compare. Interestingly enough, the median concentrations of the play area wipes collected in this study and the wipes collected in

the Bradman et al. (2007) and the CTEPP studies suggest that these wipes were all collected from similar type locations. The median concentrations of *cis*-permethrin and *trans*-permethrin in food for the CTEPP and Bradman et al. (2007) studies were less than the method detection limit, so comparisons to this study are difficult. The median concentration of chlorpyrifos in the food measured in this study is twice the median concentration measured in the CTEPP study (Table 5).

The most frequently detected pesticides in this study were cypermethrin, *cis*-permethrin, *trans*-permethrin, and chlorpyrifos. Bradman et al. (2007) also reported the concentrations of these pesticides from a small pilot observational study conducted in the Salinas Valley, CA, but the median concentrations reported by these researchers were lower than what was reported in this study (Table 5). *Cis*-permethrin and *trans*-permethrin were measured in the CTEPP study, but the median concentrations were lower than our reported results (Table 5). Our results support the observation made by Bradman et al. (2007) that pyrethroids are increasingly found in the home environment. Seifert et al. (2000a, b) also report the widespread use of permethrin in the indoor environment in Germany. The German Environmental Survey is a large-scale, representative population study conducted approximately every 7 years that measures the concentrations of chemicals to which German people are exposed. In the second environmental survey, permethrin was measured in 90.6% of the dust samples collected from vacuum cleaner bags (Seifert et al., 2000a, b). As we did not collect bulk dust in these homes, it is impossible to compare the concentrations reported by Seifert et al. (2000a, b) and the concentrations measured on the wipes and sock samples. The German Environmental Survey IV pilot study (GerES IV) reports on pesticide concentrations in dust and pesticide metabolite concentrations in urine for 500 children. In GerES IV, permethrin was detected in 79% of the dust samples with a median concentration of 0.09 mg/kg. Five pyrethroid metabolites were measured in the children's urine samples with 3-PBA measured in 90% of the samples with a median concentration of 0.29 μg/l; *trans*-DCCA measured in 74% of the samples with a median concentration of 0.19 μg/l; *cis*-DCCA measured in 56% of the samples with a median concentration of 0.11 μg/l; and 4F-3-PBA measured in less than 1% of the samples with a median concentration <0.1 μg/l (Becker et al., 2006).

In addition, the First National Environmental Health Survey of Child Care Centers (CCC) (Tulve et al., 2006a) and CTEPP (Morgan et al., 2004) studies reported median pesticide residue concentrations in day care centers at much lower concentrations than those reported in this study for the same active ingredients (compare to Table 5) (CCC study: floor wipes: *cis*-permethrin: median $= 0.03$ ng/cm^2; *trans*-permethrin: median $= 0.03$ ng/cm^2; cypermethrin: median $<$ detection limit; chlorpyrifos: median $= 0.02$ ng/cm^2; CTEPP study: hard floor surface wipes: NC: *cis*-permethrin: median $= 0.09$ ng/cm^2; *trans*-permethrin: median $= 0.07$ ng/cm^2; chlorpyrifos: median $= 0.01$ ng/cm^2; OH: *cis*-permethrin: median $= 0.006$ ng/cm^2; *trans*-permethrin: median $= 0.005$ ng/cm^2; chlorpyrifos: median $<$ detection limit). Agricultural OPs on our target analyte list were not measured in any environmental media.

Comparisons of the pyrethroid metabolites, 3-PBA, *cis*-DCCA, and *trans*-DCCA, in urine between this study and other available studies suggest that the median concentrations for these children are higher than any other published studies involving children. In the GerES IV study, the median 3-PBA concentration was 0.29 μg/l (Becker et al., 2006) and in NHANES the median 3-PBA concentration was 0.3 μg/l (95% confidence interval $= 0.2$–0.41 μg/l) for the age group 6–11 years (CDC, 2005); in contrast, the median 3-PBA concentration for the nine children in this pilot observational study was 2.2 μg/l. In the GerES IV study, the median concentrations of *cis*-DCCA and *trans*-DCCA were 0.11 and 0.19 μg/l, respectively, and in NHANES the median concentrations of *cis*-DCCA and *trans*-DCCA were <0.1 and <0.4 μg/l, respectively, (95% confidence interval for *cis*-DCCA <0.1 μg/l, 95% confidence interval for *trans*-DCCA <0.4 μg/l) for the age group 6–11 years (CDC, 2005); in contrast, the median *cis*-DCCA and *trans*-DCCA concentrations for the nine children in this pilot observational study were 0.62 and 1.1 μg/l, respectively. In the GerES IV study the median 4F-3-PBA concentration was <0.1 μg/l (Becker et al., 2006) and in NHANES the median 4F-3-PBA concentration was <0.2 μg/l (95% confidence interval <0.2 μg/l) for the age group 6–11 years (CDC, 2005). The median 4F-3-PBA concentration reported in this study is similar (0.09 μg/l), suggesting that diet may be a potential source of this metabolite. However, more research is needed to evaluate this hypothesis. The metabolite values reported here are a small subset of the results for the 201 children in the main biomonitoring study which will be published in an upcoming manuscript.

The results reported in this study show a wide variety of pesticide active ingredients found in the residential environment. Furthermore, when the results from this study are compared to other studies, our reported median results are almost always higher. Yet, with such a small sample size, it is not possible to do more than show trends in the data. Further research with a larger cohort is needed to better understand how pesticide concentrations change during routine pesticide use in an indoor environment and what factors most likely influence a child's potential exposure to pesticides.

Although there are limitations to this study, there are also significant facts that can be reported from these results. While chlorpyrifos was not reported used, it was measured in all media. A better understanding of the fate of chlorpyrifos and other pesticides indoors is needed to estimate how long the pesticide will be present in the indoor residential

environment. To predict the spatial and temporal distributions of pesticides in the indoor environment and how this may impact a child's exposure, we are developing a fugacity model that incorporates the physico-chemical properties of the pesticides. This model will need to be evaluated with real-world data.

Wipe samples showed a wide variety of pesticide active ingredients, suggesting that many different pyrethroid products were used in this area at the time of the study. With pyrethroid concentrations higher in the wipes and socks than the OP concentrations, this would suggest that pyrethroids are more likely to stick to surfaces and dust particles than to volatilize into the air. Socks showed potential usefulness for measuring pesticide residue concentrations for locations where children spent time. Additional methods development is needed for methods to estimate dermal exposure. The location of the pesticide and the child's activity patterns will influence the child's potential exposures to indoor chemicals.

The median urinary metabolite concentrations measured in these children's urine samples highlight the need for further research to understand the factors that result in these urinary levels. Pesticide metabolites have been measured in environmental media and diet samples, suggesting that individuals may be exposed to the pesticide metabolites during their normal day-to-day activities. In CTEPP, chlorpyrifos and its degradation product TCPy were measured in environmental media and diet samples, but the corresponding urinary TCPy concentrations could not be adequately explained by the amount of chlorpyrifos and TCPy measured in the multimedia samples (Morgan et al., 2005). Further research is also needed to understand whether urinary output is a better measure of a child's potential exposure than urine concentration.

Comparison of the application area floor wipe and the play area floor wipe concentrations show a large variation in measured concentration that can occur depending on the sampling location. This indicates that the sampling locations need to relate to the objective or hypothesis being evaluated.

Multimedia samples were collected in this study. In combination with time–activity profiles, aggregate and cumulative exposure assessments can be undertaken. Multimedia sampling increases our ability to accurately understand the routes and pathways contributing to a child's potential exposure.

Conclusions

Pyrethroid pesticides (e. g., *cis*-permethrin, *trans*-permethrin, cypermethrin) were frequently detected in multimedia samples (e.g., air, dust, food) at median concentrations that were often higher than reported previously in similar studies. In addition, the pyrethroid metabolite concentrations in the urine were higher than the concentrations reported by either NHANES or GerES studies. Results of this small pilot study suggest the need for larger observational exposure measurement studies to measure exposure and to determine the factors affecting children's exposures in homes where pesticides are used.

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