Journal of Exposure Science and Environmental Epidemiology (2007), 1–8

www.nature.com/jes

Pesticide loadings of select organophosphate and pyrethroid pesticides in urban public housing

RHONA JULIEN[a], GARY ADAMKIEWICZ[a], JONATHAN I. LEVY[a], DEBORAH BENNETT[a,b], MARCIA NISHIOKA[c] AND JOHN D. SPENGLER[a]

[a]*Harvard School of Public Health, Harvard University, Boston, Massachusetts, USA*
[b]*University of California, Davis, California, USA*
[c]*Battelle Memorial Institute, Columbus, Ohio, USA*

We investigated the magnitude and distribution of pyrethroid and organophosphate pesticide loadings within public housing dwellings in Boston, Massachusetts and compared the results using various sampling methods. We collected dust matrices from living room and kitchen in 42 apartments and analyzed for eleven pyrethoids (e.g., permethrin and cyfluthrin) and two organophosphates (chlorpyrifos and diazinon) in house dust using GC/MS. Agreement between sampling methods were evaluated using Spearman correlations and Kappa statistics. Permethrin and chlorpyrifos were detected in kitchen floor wipes in all homes, followed in frequency of detects by diazinon (98%), cypermethrin (90%) and cyfluthrin (71%). At least six pesticides were detected in kitchen floor wipes in the majority of the homes (range 3–8). Positive and statistically significant correlations among dust matrices were observed between kitchen floor wipes and living room vacuum dust, including for diazinon ($r = 0.62$) and cyfluthrin ($r = 0.69$). Detection of several pesticides including banned or restricted use products in some public housing units, underscore the need for alternative pest management strategies that embrace the safe and judicious use of pest control products.
Journal of Exposure Science and Environmental Epidemiology advance online publication, 9 May 2007; doi:10.1038/sj.jes.7500576

Keywords: *chlorpyrifos, house dust, pesticides, public housing, pyrethoids, urban.*

Introduction

In the US, people spend approximately 90% of their time indoors (Gurunathan et al., 1998) and about 74% of households use pesticides (US EPA, 2002), indicating that indoor residential exposures may be a significant exposure pathway for many individuals. In urban multiunit dwellings, pesticide usage is prevalent due to problems with pest infestation (Landrigan et al., 1999; Whyatt et al., 2002), especially in older, poorly maintained housing stock (Kitch et al., 2000). In these homes, pesticide usage is sometimes excessive (Landrigan et al., 1999) and oftentimes includes the use of prohibited or restricted-use pesticides (Adgate et al., 2000; Surgan et al., 2002).

Adverse health effects associated with pesticide exposure from residential use include altered fetal growth from prenatal exposure (Berkowitz et al., 2004; Whyatt et al., 2004), childhood cancer (Buckley et al., 2000; Daniels et al., 2001; Flower et al., 2004) and asthma (Salam et al., 2004). The association with asthma may be particularly concerning, given the fact that pest management may have the objective of reducing allergen exposures and related asthma development and exacerbations.

While it is known that non-dietary exposure to pesticides occurs mostly in the home (Lewis et al., 1994; Whitmore et al., 1994; Simcox et al., 1995), significant data gaps exist for residential pesticide exposure in urban households, as most studies have focused primarily on agricultural communities and their exposures to organophosphates, including diazinon and chlorpyrifos (Simcox et al., 1995; Loewenherz et al., 1997; Gordon et al., 1999; Fenske et al., 2000, 2002). More recently, studies have focused on urban settings and have highlighted the widespread use of pesticides indoors (Quackenboss et al., 2000; Pang et al. 2002; Whyatt et al. 2002; Berkowitz et al. 2003).

Two classes of pesticides, which have been widely used in residential settings, including urban multiunit dwellings, are organophosphate and pyrethroid pesticides (Landrigan et al., 1999). Based on the potential to cause adverse health effects to occupants, especially children, two organophosphates (chlorpyrifos and diazinon) were withdrawn from the indoor residential market in 2001 and 2002, respectively (US EPA, 2000a, b). Organophosphates exert their toxic effect by inhibiting the enzymatic degradation of the neurotransmitter acetylcholine; at extremely high doses, inhibition of acetylcholine esterase results in the continued firing of the neuron

1. Address all correspondence to: Dr. R. Julien, 64 Beaumont Street, Dorchester, MA 02124, USA.
Tel.: +1 617 918-1782. Fax: +1 617 918-0782.
E-mail: julien.rhona@epa.gov
Received 4 December 2006; accepted 4 February 2007

and subsequent paralysis or death of the affected organism (He, 1994).

With the withdrawal of the organophosphates, pyrethroids are being used increasingly to control pests indoors (Adgate et al., 2000). Synthetic pyrethroids have insecticidal properties similar to the botanical pesticides known as pyrethrins (i.e., neurotoxin with rapid paralysis or "quick knock-down effect" of target pest). However, pyrethroids are more persistent in the environment than the naturally occurring pyrethrins and are therefore used indoors as well as in agricultural applications (Todd et al., 2003). The toxic mode of action for pyrethroids is the disruption of the conductance of neuronal impulses, which results in the generation of multiple action potentials that leads to tremors and incoordination. These cellular level effects can lead to various health concerns (He, 1994).

In addition to the limited exposure data on pyrethroids, there is not a single standardized or consistent approach for analysis or comparison among pesticide exposure studies (Quandt et al., 2004), as previous studies have focused on various matrices including blood, urine, air and/or vacuum dust or floor wipes to quantify exposures (Lioy et al., 2002). Therefore, the aim of the current study was to quantify the distribution of common pesticides (e.g., select pyrethroids and diazinon and chlorpyrifos) within the homes of public housing residents in Boston and compare pesticide loadings from various collection media. We used house dust as our target medium, since our selected pesticides are semi-volatile (organophosphates) or nonvolatile (pyrethroids), and are therefore preferentially bound in the particle phase (Berger-preieß et al., 1997). Dust samples were collected from the kitchen, where pesticides are most frequently applied due to cockroach infestation (Brenner et al., 2003), and from the living room, where families tend to spend significant time while indoors. In addition, we examined the agreement between floor wipes and vacuum dust sampling methods when classifying pesticide exposure within these homes.

Methods

The current study is a component of the Healthy Public Housing Initiative (HPHI), a longitudinal intervention study targeting apartments of pediatric asthmatics (between 4 and 17 years of age) living in urban housing. Sixty households were recruited from three public housing developments in Boston, Massachusetts. Of the households who were recruited for the intervention study, 43 households participated in the Integrated Pest Management (IPM) program, which was one of the primary environmental interventions tested. Environmental measurements for the current study were obtained from 42 of these households. Additional information on HPHI and characteristics of the study participants can be found elsewhere (Clougherty et al., 2006; Levy et al., 2006).

Sampling Protocol

Home visits were scheduled for families who provided written consent for their participation in the IPM program. During these visits, environmental measurements as well as health and quality of life information related to asthma were obtained both before and after IPM interventions. During the first visit, vacuum dust and floor wipe samples were collected to provide baseline information about the prevalence and surface loadings of target pesticides. Environmental measurements were recorded between July 2002 and August 2003 from homes in all three developments. Additional information on household characteristics such as ethnicity and pesticide use were collected at the time of enrollment. Data discussed in this paper will only focus on the measurements taken before the IPM interventions, to establish baseline pesticide loadings.

Floor wipes were taken on vinyl floor surfaces in both living room and kitchen from standardized locations. Kitchen floor wipes were taken adjacent to the stove and living room floor wipes were taken adjacent to the sofa. If the latter location was not feasible (e.g., due to clutter), an alternate area adjacent to the linen closet in the hallway was designated. For the floor wipe samples, we used a sampling protocol that was adapted from the National Human Exposure Assessment Survey in Arizona (NHEXAS-AZ) (Gordon et al., 1999), which involved wiping a one-square-foot area ($0.0929\,m^2$) with a three-inch square ($58\,cm^2$; Johnson and Johnson™) sterile gauze wetted with 5 ml 99% isopropanol. Once collected, each wipe sample was placed in a labeled 60 ml amber glass jar and placed in a cooler.

Vacuum dust samples were collected only in the living room due to anticipated low dust loadings on bare kitchen floors (Nishioka et al., 1999). Living room samples were collected from all accessible surfaces including sofas and carpets since these fabric surfaces are primary reservoirs for dust borne pesticides (Lewis et al., 1994) and can serve as a source of continued exposure to occupants (Fenske et al., 1991; Simcox et al., 1995; Gurunathan et al., 1998; Landrigan et al., 1999). Vacuum dust was obtained using a sampling protocol that was adapted from the epidemiologic study conducted in Cape Cod, Massachusetts that looked at the associations between environmental exposures and breast cancer (Rudel et al., 2001). The sampling apparatus consisted of a 9 A Eureka Mighty-mite™ vacuum cleaner, adjusted to collect dust in a 19×90 mm cellulose extraction thimble which was placed into an extended arm. This arm was connected at one end to the vacuum cleaner and capped at the other end with a crevice tool. The samples were collected by slowly moving the crevice tool using back and forth motions over the designated areas for a total sampling time of five minutes. Once samples were collected, the

cellulose thimbles were removed from the extended arm using gloves and placed into a labeled zip loc™ bag and stored in a cooler. Both vacuum dust and floor wipe samples were later stored in a freezer at −22°C until shipment for analysis.

Chemical Analysis

To determine pesticide concentrations, each sample was analyzed for two organophosphates (chlorpyrifos and diazinon) and eleven pyrethroids [allethrin (*cis*- and *trans*-isomers) coelute, bifenthrin, isomers of cyfluthrin (three chromatographically resolved isomers), cyhalothrin, cypermethrin (three resolved isomers), deltamethrin, esfenvalerate, permethrin (*cis*- and *trans*- isomers), resmethrin, sumithrin and tetramethrin]. Target pesticides were selected because of potential adverse health effects, persistence indoors and/or widespread residential use.

Vacuum Dust Extraction The available dust for each sample, up to 0.50 g, was weighed and fortified with 250 ng of the compound class-specific surrogate recovery standards (SRSs), fenchlorphos for the organophosphates and $^{13}C_6$-labelled mix of *cis*/*trans*-permethrin for pyrethroids. The dust was extracted using ultrasonication in 12 ml of 1:1 hexane:acetone. After centrifugation, 10 ml of the extract was removed, concentrated and solvent exchanged into hexane. A C18 solid phase extraction (SPE) cartridge (1000 mg; Bakerbond) was conditioned in sequence with dichloromethane (DCM), 15% diethyl ether in hexane and hexane. The extract was added, the sample was eluted in reverse order with these solvents and the resulting eluant was concentrated to a final 1 ml volume. Dibromobiphenyl was added as the internal standard for quantification. A nine-point calibration curve, spanning the range of 0–750 ng/ml for analytes and 0–300 ng/ml for SRSs, was analyzed concurrently with each sample set. Linear regression analysis was used to establish the calibration curve for each analyte. Samples with analytes that exceeded the calibration curve range by more than 15% were diluted, respiked with internal standard and reanalyzed. After quantification, analyte concentrations were corrected by the recovery of the matched compound class SRS in that sample.

Floor Wipe Extraction Each wipe sample was fortified with 100 ng of fenchlorphos and $^{13}C_6$-*trans*-permethrin, and extracted using accelerated solvent extraction (ASE) technology (ASE 200; Dionex Corp) in an 11 ml cell using DCM at 2000 psi and 100°C through two cycles. The extract was concentrated, solvent exchanged, cleaned up and analyzed as described above for the dust extracts.

Extracts were analyzed using GC/MS in the multiple ion detection mode (6890 GC interfaced to a 5973 MSD; Agilent) using a DB-1701 GC column (30 m; 0.25 mm id; 0.15 μm film thickness) with the GC temperature programmed to 280°C. Two diagnostic ions were monitored for identification of each analyte.

Quality Assurance/Quality Control QA/QC samples were analyzed with each sample set and were used to assess method performance. For the 78 dust analyses, the QA/QC samples included eight solvent method blanks, three solvent method spikes (100 ng/analyte), three reference dust samples, six fortified reference dust samples and five duplicate analyses. Low-level dust fortification was 50 ng/analyte, except 62.5 ng/isomer for cyfluthrin and cypermethrin, and 250 ng/analyte for esfenvalerate and deltamethrin; high-level fortification was 5 × higher.

The average solvent spike recovery was 86%, with a range of 82 ± 12 to 93 ± 13% for chlorpyrifos and sumithrin, respectively. The average low-level spike recovery was 93%, with a range of 56 ± 7 to 120 ± 10% for resmethrin and cyhalothrin, respectively. The average high-level spike recovery was 107%, with a range of 84 ± 23–132 ± 48% for diazinon and tetramethrin, respectively. There was a consistent interference to allethrin in the method that prevented detection and quantification of this analyte in dust sample extracts. For the 31 pairs of analytes detected in the duplicate samples, the average relative percent difference for concentration was 25% (0–154%). The SRS recoveries in the dust samples showed very good method performance on a sample-by-sample basis: recovery of 93 ± 10% for fenchlorphos, 105 ± 16% for $^{13}C_6$-*cis*-permethrin and 95 ± 18% for $^{13}C_6$-*trans*-permethrin.

The QA/QC samples for the 192 wipe samples included 15 field matrix blanks and 15 matrix spikes, with the latter fortified with 150 ng/analyte (with scaling as described above for cyfluthrin, cypermethrin, esfenvalerate, deltamethrin) before extraction. Average recovery in the spiked wipes was 98%, with a range of 86 ± 9 to 112 ± 13% for cypermethrin isomers and chlorpyrifos, respectively. The SRS recoveries in the wipe samples showed good method performance on a sample-by-sample basis: recovery of 78 ± 15% for fenchlorphos and 91 ± 25% for $^{13}C_6$-*trans*-permethrin.

Data Analysis

The normality of each pesticide distribution was determined using the Shapiro–Wilks test. Where the data were skewed, we used non-parametric tests for analyses. In order to ensure unbiased estimates of the correlations, samples with concentrations below the limit of detection (LOD) were assigned random values between zero and the LOD according to an assumed uniform distribution.

Because there is no "gold standard" for sampling indoor concentrations of dust-borne pesticides, we examined the agreement between two sampling methods conducted in this study, vacuum dust and floor wipe sampling. We first evaluated the relationships within matrices (kitchen vs. living room floor wipes) and between matrices (living room floor

wipes vs. vacuum dust) using Spearman rank correlations. For floor wipes, we also tested whether concentrations significantly differed between the kitchen and living room, using the Wilcoxon rank-sum test.

We categorized measured concentrations as dichotomous variables and evaluated the agreement between the vacuum dust method and the kitchen floor wipe method. As there are no well-defined thresholds for health effects for the target pesticides, we considered three alternatives: above/below the LOD, the 50th percentile and the 75th percentile. Agreement was evaluated using the Kappa statistic, which captures the level of agreement between these two alternate measurement approaches (where 1 = perfect agreement, 0 = no agreement above that expected by chance, −1 = perfect disagreement). All statistical analyses were performed using SAS Version 9 (SAS, 2002).

Results

Table 1 provides study demographics and information on participants' choice of pest control methods, including traps (41%), non-volatile formulations (gels (25%)) and volatile formulations (sprays (34%), and smoke bombs (27%)). Although 84% of the families reported pesticide use within the past year, 92% indicated an interest in using pest control remedies that did not rely on pesticides.

Table 2i reports summary statistics for kitchen and living room floor wipes and vacuum dust in the baseline pre-intervention samples. Target analytes with isomers (e.g., permethrin, cypermethrin and cyfluthrin) are presented as the sum for each compound based on high Spearman rank correlations among specific isomers for each compound. For kitchen floor wipes, permethrin and chlorpyrifos were detected in every home. Diazinon and cypermethrin were detected at fairly high frequencies (≥90%). Cyfluthrin was detected in 71% of homes. For living room floor wipes, permethrin and chlorpyrifos were also detected at fairly high frequencies, followed by diazinon, which was detected in 80% of the homes. For vacuum dust, similarly high detection frequencies were also observed, with *cis*- and *trans*-permethrin detected in every home. In addition to detection frequency, the data in Table 2i indicate that most analytes have pesticide loadings that vary across several orders of magnitude. This variation in pesticide distribution is also illustrated in Figure 1, which presents the cumulative frequency distribution of the five most prevalent analytes in kitchen floor wipes. The distribution of cyfluthrin, which is a restricted-use pesticide in certain formulations such as 25% emulsified concentrate (US EPA, 2003), is somewhat broader and more skewed than the distribution for the other pyrethroids. At the upper end of the distribution (>90th percentile), cyfluthrin loadings were over 2 orders of magnitude higher than the loadings at the 50th percentile.

Table 1. Selected household characteristics of IPM participants ($N = 43$).

Ethnic composition:	
Hispanic	65%
Black	33%
Caucasian	2%
Pesticide use within past year:	
Ever used pesticides	84%
Use weekly or more often	53%
Use monthly or less often	29%
Pesticide type[a]:	
Traps	41%
Gels	25%
Sprays	34%
Smoke bombs	27%
Self-report cockroach infestation as severe	52%

[a]Total percentage will exceed 100% because families used multiple pesticide types at any given time in the home.

One way to examine the degree to which analytes occur simultaneously with one another is to consider the number of analytes detected in each home. All homes had at least two pesticides present in vacuum dust, and 17 homes (49%) had five or more present. Similarly, in examining kitchen floor wipes, all homes had at least three pesticides present and 27 homes (64%) had six or more present. For the living room floor wipes, 17 homes (56%) had five or more pesticides present.

We also considered the agreement between sampling sites and between sampling methods (Table 2ii). Spearman rank correlations were positive and statistically significant for the majority of prevalent analytes (diazinon, chlorpyrifos, permethrin and cyfluthrin), ranging from 0.38 to 0.64 between living room and kitchen floor wipes, from 0.41 to 0.69 between kitchen floor wipes and living room vacuum dust samples and from 0.44 to 0.61 between living room floor wipes and living room vacuum dust.

Based on the results for the Wilcoxon rank-sum test (not shown here), with the exception of cypermethrin, there were no statistically significant differences in pesticide loadings between living room and kitchen floor wipes for the prevalent analytes. Thus, given the similarity between these two matrices for the prevalent analytes, only the results from the kitchen floor wipes will be represented hereafter.

To test the agreement between the two forms of measurements (floor wipes and vacuum dust) from the perspective of categorization as high/low exposure based on specific exposure thresholds (Table 3), the magnitude of Kappa coefficients was considered for the following exposure

Table 2i. Summary statistics for pesticide prevalence (% above limit of detection) and pesticide loadings in kitchen and living room floor wipe samples ($\mu g/m^2$) and vacuum dust ($\mu g/g$)

Analyte	Average LOD	%>LOD	Minimum	Median	75th percentile	Maximum
Kitchen floor wipes (N=42)						
Chlorpyrifos	0.05	100	0.03	0.3	1.3	19.5
Permethrin	0.01	100	0.21	6.8	33.0	226.5
Diazinon	0.02	98	<LOD	0.4	2.6	556.2
Cypermethrin	0.08	90	<LOD	3.7	16.2	330.7
Cyfluthrin	0.12	71	<LOD	1.1	16.4	567.1
Esfenvalerate	0.27	67	<LOD	0.7	2.5	16.8
Cyhalothrin	0.11	26	<LOD	<LOD	0.4	4.1
Deltamethrin	2.70	16	<LOD	<LOD	<LOD	45.2
Tetramethrin	0.11	12	<LOD	<LOD	<LOD	5.9
Sumithrin	0.02	5	<LOD	<LOD	<LOD	2.3
Bifenthrin	0.02	5	<LOD	<LOD	<LOD	0.2
Resmethrin	0.05	2	<LOD	<LOD	<LOD	0.05
Living room wipes (N=30)						
Permethrin	0.01	93	0.74	5.97	24.68	74.6
Chlorpyrifos	0.05	93	0.06	0.49	1.63	7.7
Diazinon	0.02	80	0.06	0.35	0.92	16.3
Cypermethrin	0.08	63	0.58	3.80	9.35	63.2
Esfenvalerate	0.27	50	0.24	1.00	2.05	27.4
Cyfluthrin	0.12	43	0.59	3.70	15.98	56.9
Cyhalothrin	0.11	23	0.20	1.67	3.98	7.5
Tetramethrin	0.11	10	0.10	8.19	8.59	8.6
Deltamethrin	2.70	7	2.36	3.43	4.49	4.5
Bifenthrin	0.02	3	<LOD	<LOD	<LOD	0.1
Sumithrin	0.02	3	<LOD	<LOD	<LOD	0.4
Resmethrin	0.05	3	<LOD	<LOD	<LOD	0.05
Vacuum dust (N=35)						
Permethrin	0.002	100	0.13	0.92	1.3	13.1
Diazinon	0.004	94	<LOD	0.05	0.2	4.4
Chlorpyrifos	0.010	89	<LOD	0.06	0.2	3.0
Cypermethrin	0.015	60	<LOD	0.30	0.8	5.2
Cyfluthrin	0.022	43	<LOD	<LOD	1.2	48.1
Esfenvalerate	0.050	29	<LOD	<LOD	0.20	1.2
Deltamethrin	0.500	9	<LOD	<LOD	<LOD	7.0
Tetramethrin	0.020	6	<LOD	<LOD	<LOD	6.0
Cyhalothrin	0.020	3	<LOD	<LOD	<LOD	0.1
Sumithrin	0.004	3	<LOD	<LOD	<LOD	0.1
Bifenthrin	0.004	3	<LOD	<LOD	<LOD	0.01
Resmethrin	0.010	0	<LOD	<LOD	<LOD	<LOD

thresholds, LOD, the median percentile and the 75th percentile. Kappa coefficients were positive and strong (significantly >0) for cyfluthrin at all three thresholds (0.33, 0.47 and 0.30), permethrin (0.59 and 0.51) and diazinon (0.53 and 0.51) at the median and 75th percentile, respectively, and for chlorpyrifos (0.35), at the median percentile. We were unable to compute Kappa coefficients for the two most ubiquitous target pesticides (permethrin and chlorpyrifos) due to 100% detection in at least one medium (Sim and Wright 2005) and in general, the high detection rates for the analytes in question limit the statistical power of the Kappa coefficients.

Discussion

The findings of the high prevalence of pesticides, for example, permethrin and chlorpyrifos, in these urban housing units are consistent with other studies that obtained information on pesticide prevalence via surveys and questionnaires (Landrigan et al., 1999; Adgate et al., 2000; Kinney et al., 2002; Brenner et al., 2003).

While sampling occurred after diazinon and chlorpyrifos were removed from the residential market, the measured concentrations in house dust can most likely be attributed to the persistence of these pesticides indoors. However, the

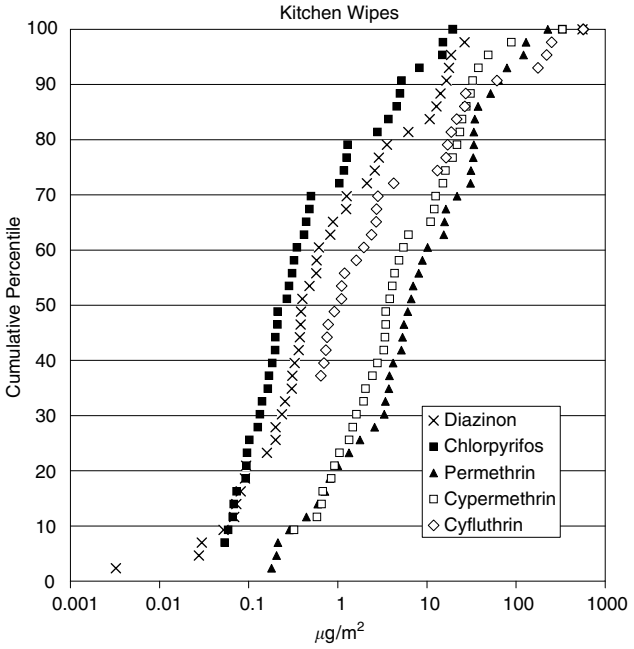

Figure 1. Cumulative frequencies of five target pesticides detected in kitchen floor wipes.

Table 2ii. Spearman correlations between sampling matrices and betwen sampling locations for the most prevalent pesticides (LR = living room, K = kitchen)

Prevalent pesticides	LR and K wipes ($N = 29$) r (P-value)	K wipes and LR dust ($N = 34$) r (P-value)	LR wipes and LR dust ($N = 21$) r (P-value)
Diazinon	0.48**	0.62**	0.48*
Chlorpyrifos	0.63**	0.41*	0.61**
Permethrin	0.64**	0.51**	0.44*
Cypermethrin	0.42*	0.01	0.22
Cyfluthrin	0.38*	0.69**	0.50*

*Statistically significant at the 0.05 level.
**Statistically significant at the 0.01 level.

presence of cyfluthrin was a cause for concern for several reasons. It is the active ingredient found primarily in a product known as Tempo. Based on information obtained from the Material Safety Data Sheet (MSDS) prepared by the manufacturer, Bayer (e.g., MSDS #R000023651 and MSDS #29752), this compound is available in a variety of formulations (e.g., wettable powders and suspension concentrate) and in certain formulations is licensed for pest management professionals and/or commercial use only.

Moreover, field staff were informed that Tempo was being applied by residents in its concentrated form and was not mixed with water as required per labeling instructions. This misuse of Tempo would result in substantially higher exposures than intended and should be investigated in further detail. In homes where cyfluthrin was detected, the concentrations above the 90th percentile exceeded the median concentrations by several orders of magnitude, which may indicate that a subset of residents is applying the product incorrectly.

Table 3. Test of equivalence between kitchen floor wipe and vacuum dust methods ($N = 34$), considering multiple exposed/unexposed thresholds

	Exposure threshold		
Analyte	Detected (> LOD)	Median	75th percentile
Chlorpyrifos	Not applicable	0.35 (0.16)	0.23 (0.19)
Permethrin	Not applicable	0.59 (0.14)	0.51 (0.17)
Diazinon	−0.04 (0.03)	0.53 (0.15)	0.51 (0.17)
Cypermethrin	0.05 (0.13)	0.12 (0.17)	−0.02 (0.17)
Cyfluthrin	0.33 (0.13)	0.47 (0.15)	0.30 (0.18)

Values represent Kappa statistics and standard errors.

Since few studies have measured concentrations of these analytes in house dust in urban settings, there are limited data to make a determination of whether residents in these public housing developments are disproportionately exposed to pesticides and may be at risk of adverse health effects. A study which looked at exposures to several pesticides including chlorpyrifos, diazinon and *cis*- and *trans*-permethrin in predominantly single-family homes measured geometric mean concentrations of approximately 113 ng/g for chlorpyrifos, 25 ng/g for diazinon, 337 ng/g for *cis*-permethrin and 517 ng/g for *trans*-permethrin (Colt et al., 2004). These measurements were similar to our median concentrations in vacuum dust, although it should be noted that the limit of detection in that particular study was an order of magnitude higher than the limit of detection in our study, limiting quantitative comparisons. Comparable measurements were also observed in the floor dust of a day care setting with preschoolers, with observed median chlorpyrifos concentrations of 135 ng/g (Morgan et al., 2005). However, these measurements were taken before the phase-out of organophosphates. We are also unaware of other studies, which measured levels of numerous analytes, especially pyrethroids, in low-income multiunit dwellings, which underscores the importance and uniqueness of our findings. And given the known neurological pathways affected by pyrethroids, the levels we have observed in this study suggest that the current substitution of organophosphates with these chemicals may incur their own health risks in exposed populations.

In terms of our methodological conclusions, there were some positive and statistically significant correlations between the vacuum dust and floor wipe samples. However, based on our findings of a modest agreement between these two matrices for some analytes, we are unable to conclude that floor wipe samples can serve as a universal proxy for vacuum dust samples. In instances where there was a strong

agreement between the analytes, for example, permethrin and diazinon, this may be due in part to the liquid formulation of these pesticides (e.g., smoke bombs and aerosol sprays) used in the home. Applications such as smoke bombs intentionally distribute the pesticide throughout the home. The aerosol spray is a more targeted application; however, there is potential for considerable overspray. Due to relatively high vapor pressures as well as movement of dust, many pesticides migrate from treated areas to non-target surfaces (Gurunathan et al., 1998; Lioy et al., 2002; Bennett and Furtaw 2004). The frequency of pesticide use and the close proximity of rooms in our study homes, which is quite typical in inner-city public housing, will undoubtedly facilitate this process. On the other hand, if Tempo (with cyfluthrin as its active ingredient) is applied in powdered form in the kitchen, barring any mechanical means of resuspension such as vacuuming, people and pet traffic, it is unlikely that significant deposition onto non-target surfaces will occur. For cypermethrin, we do not have information on whether the product formulation that was used in these homes came in liquid formulation (emulsifiable concentrates) or dry formulation (e.g., wettable powder and dust granules), making it difficult to determine the precise reason for non-agreement.

There are some potential limitations that influence the interpretation of our findings. In exposure studies where dermal exposure is the potential pathway under consideration, dislodgeable pesticides (i.e., pesticide residue on a surface that is removed by the skin) are regarded as the most appropriate measurement for human exposure (Fenske et al., 1991). Our choice of isopropanol as the wetting agent to collect floor wipe samples likely improved our collection efficiency, but may also result in removal of these residues from both the surface and sub-surface, impairing comparability with other exposure measures.

We did not obtain information about the approximate time of the most recent pesticide application in the home, a factor, that would clearly influence the concentration of detectable pesticide residues (Berger-preieß et al., 1997; Bennett and Furtaw 2004). Also, in obtaining floor wipe samples, we assumed that sampling occurred where pesticides were applied by the residents. It is believed that this assumption would not introduce much error since the movement of pesticide residues from the point of application to other areas in the home does occur (Matoba et al., 1998).

It is also possible that our findings are not generalizeable to all home environments; the modest yet significant agreement between kitchen and living room floor wipes could be attributed to the fact that in almost all of these homes, both rooms were adjacent to each other. However, the size of these units measuring 65–84 m^2 (Zota et al., 2005) is typical of inner-city urban dwellings, and our findings can likely be generalized to those settings.

To the best of our knowledge, this study is the first to report on concentrations of 10 pyrethroids and two organophosphates in an urban setting, and more specifically, in a multiunit residential setting. Our results show a wide range of pesticide levels with certain pyrethroids (permethrin) and organophosphates (chlorpyrifos) detected in every home, and restricted-use pesticides (cyfluthrin) detected in a majority of homes. In addition, the significant correlation between vacuum and floor wipe sampling methods for certain ubiquitous analytes indicates that kitchen floor wipe samples could potentially serve as a less expensive and non-intrusive proxy, which can be especially helpful in a challenging sampling environment. More broadly, our findings imply that interventions may be warranted in these developments, with efficient and practical methods needed to preferentially select homes for possible interventions, given the substantial variability in concentrations across units. Equally important are the policy implications of our findings, which suggest that efforts should be made to promote and institutionalize viable and safer pest control alternatives in these households.

Acknowledgements

Funding was provided by US Department of Housing and Urban Development (grant# MALHH0077-00), W.K. Kellogg Foundation, Boston Foundation and Jessie B. Cox Charitable Trust Melvin W. First Scholarship and Akira Yamaguchi Endowment. Special thanks to the families who participated in the study, as well as the community partners for their cooperation and commitment to the success of this project.

References

Adgate J.L., Kukowski A., Stroebel C., Shubat P.J., Morrell S., and Quackenboss J.J., et al. Pesticide storage and use patterns in Minnesota households with children. *J Expo Anal Environ Epidemiol* 2000: 10(2): 159–167.

Bennett D.H., and Furtaw Jr E.J. Fugacity-based indoor residential pesticide fate model. *Environ Sci Technol* 2004: 38(7): 2142–2152.

Berger-preieß E., Preieß A., Sielaff K., Raabe M., Ilgen B., and Levsen K. The behaviour of pyrethroids indoors: a model study. *Indoor Air* 1997: 7: 248–261.

Berkowitz G.S., Obel J., Deych E., Lapinski R., Godbold J., and Liu Z., et al. Exposure to indoor pesticides during pregnancy in a multiethnic, urban cohort. *Environ Health Perspect* 2003: 111(1): 79–84.

Berkowitz G.S., Wetmur J.G., Birman-Deych E., Obel J., Lapinski R.H., and Godbold J.H., et al. *In utero* pesticide exposure maternal paraoxonase activity, and head circumference. *Environ Health Perspect* 2004: 112(3): 388–391.

Brenner B.L., Markowitz S., Rivera M., Romero H., Weeks M., and Sanchez E., et al. Integrated pest management in an urban community: a successful partnership for prevention. *Environ Health Perspect* 2003: 111(13): 1649–1653.

Buckley J.D., Meadows A.T., Kadin M.E., Le Beau M.M., Siegel S., and Robison L.L. Pesticide exposures in children with non-Hodgkin lymphoma. *Cancer* 2000: 89(11): 2315–2321.

Clougherty J.E., Levy J.I., Hynes H.P., and Spengler J.D. A longitudinal analysis of the efficacy of environmental interventions on asthma-related quality of life

and symptoms among children in urban public housing. *J Asthma* 2006: 43(5): 335–343.

Colt J.S., Lubin J., Camann D., Davis S., Cerhan J., and Severson R.K., et al. Comparison of pesticide levels in carpet dust and self-reported pest treatment practices in four US sites. *J Expo Anal Environ Epidemiol* 2004: 14(1): 74–83.

Daniels J.L., Olshan A.F., Teschke K., Hertz-Picciotto I., Savitz D.A., and Blatt J., et al. Residential pesticide exposure and neuroblastoma. *Epidemiology* 2001: 12(1): 20–27.

Fenske R.A., Curry P.B., Wandelmaier F., and Ritter L. Development of dermal and respiratory sampling procedures for human exposure to pesticides in indoor environments. *J Expo Anal Environ Epidemiol* 1991: 1(1): 11–30.

Fenske R.A., Lu C., Barr D., and Needham L. Children's exposure to chlorpyrifos and parathion in an agricultural community in central Washington State. *Environ Health Perspect* 2002: 110(5): 549–553.

Fenske R.A., Lu C., Simcox N.J., Loewenherz C., Touchstone J., and Moate T.F., et al. Strategies for assessing children's organophosphorus pesticide exposures in agricultural communities. *J Expo Anal Environ Epidemiol* 2000: 10(6 Part 2): 662–671.

Flower K.B., Hoppin J.A., Lynch C.F., Blair A., Knott C., and Shore D.L., et al. Cancer risk and parental pesticide application in children of Agricultural Health Study participants. *Environ Health Perspect* 2004: 112(5): 631–635.

Gordon S.M., Callahan P.J., Nishioka M.G., Brinkman M.C., O'Rourke M.K., and Lebowitz M.D., et al. Residential environmental measurements in the national human exposure assessment survey (NHEXAS) pilot study in Arizona: preliminary results for pesticides and VOCs. *J Expo Anal Environ Epidemiol* 1999: 9(5): 456–470.

Gurunathan S., Robson M., Freeman N., Buckley B., Roy A., and Meyer R., et al. Accumulation of chlorpyrifos on residential surfaces and toys accessible to children. *Environ Health Perspect* 1998: 106(1): 9–16.

He F. Synthetic pyrethroids. *Toxicology* 1994: 91(1): 43–49.

Kinney P.L., Northridge M.E., Chew G.L., Gronning E., Joseph E., and Correa J.C., et al. On the front lines: an environmental asthma intervention in New York city. *Am J Public Health* 2002: 92(1): 24–26.

Kitch B.T., Chew G., Burge H.A., Muilenberg M.L., Weiss S.T., and Platts-Mills T.A., et al. Socioeconomic predictors of high allergen levels in homes in the greater Boston area. *Environ Health Perspect* 2000: 108(4): 301–307.

Landrigan P.J., Claudio L., Markowitz S.B., Berkowitz G.S., Brenner B.L., and Romero H., et al. Pesticides and inner-city children: exposures, risks, and prevention. *Environ Health Perspect* 1999: 107(Suppl 3): 431–437.

Levy J.I., Brugge D., Peters J.L., Clougherty J.E., and Saddler S.S. A community-based participatory research study of multifaceted in-home environmental interventions for pediatric asthmatics in public housing. *Soc Sci Med* 2006: 63(8): 2191–2203.

Lewis R.G., Fortmann R.C., and Camann D.E. Evaluation of methods for monitoring the potential exposure of small children to pesticides in the residential environment. *Arch Environ Contam Toxicol* 1994: 26(1): 37–46.

Lioy P.J., Freeman N.C., and Millette J.R. Dust: a metric for use in residential and building exposure assessment and source characterization. *Environ Health Perspect* 2002: 110(10): 969–983.

Loewenherz C., Fenske R.A., Simcox N.J., Bellamy G., and Kalman D. Biological monitoring of organophosphorus pesticide exposure among children of agricultural workers in central Washington State. *Environ Health Perspect* 1997: 105(12): 1344–1353.

Matoba Y., Takimoto Y., and Kato T. Indoor behavior and risk assessment following residual spraying of D-phenothrin and D-tetramethrin. *Am Ind Hyg Assoc J* 1998: 59(3): 191–199.

Morgan M.K., Sheldon L.S., Croghan C.W., Jones P.A., Robertson G.L., and Chuang J.C., et al. Exposures of preschool children to chlorpyrifos and its degradation product 3,5,6-trichloro-2-pyridinol in their everyday environments. *J Expo Anal Environ Epidemiol* 2005: 15(4): 297–309.

Nishioka M.G., Burkholder H.M., Brinkman M.C., and Lewis R.G. Distribution of 2,4-dichlorophenoxyacetic acid in floor dust throughout homes following homeowner and commercial lawn applications: quantitative effects of children, pets, and shoes. *Environ Sci Tech* 1999: 33(9): 1359–1365.

Pang Y., MacIntosh D.L., Camann D.E., and Ryan P.B. Analysis of aggregate exposure to chlorpyrifos in the NHEXAS-Maryland investigation. *Environ Health Perspect* 2002: 110(3): 235–240.

Quackenboss J.J., Pellizzari E.D., Shubat P., Whitmore R.W., Adgate J.L., and Thomas K.W., et al. Design strategy for assessing multi-pathway exposure for children: the Minnesota Children's Pesticide Exposure Study (MNCPES). *J Expo Anal Environ Epidemiol* 2000: 10(2): 145–158.

Quandt S.A., Arcury T.A., Rao P., Snively B.M., Camann D.E., and Doran A.M., et al. Agricultural and residential pesticides in wipe samples from farmworker family residences in North Carolina and Virginia. *Environ Health Perspect* 2004: 112(3): 382–387.

Rudel R.A., Brody J.G., Spengler J.D., Vallarino J., Geno P.W., and Sun G., et al. Identification of selected hormonally active agents and animal mammary carcinogens in commercial and residential air and dust samples. *J Air Waste Manag Assoc* 2001: 51(4): 499–513.

Salam M.T., Li Y.F., Langholz B., and Gilliland F.D. Early-life environmental risk factors for asthma: findings from the Children''s Health Study. *Environ Health Perspect* 2004: 112(6): 760–765.

SAS Institute Version 9 SAS Institute Inc: Cary, NC, 2002.

Sim J., and Wright C.C. The kappa statistic in reliability studies: use, interpretation, and sample size requirements. *Phys Ther* 2005: 85(3): 257–268.

Simcox N.J., Fenske R.A., Wolz S.A., Lee I.C., and Kalman D.A. Pesticides in household dust and soil: exposure pathways for children of agricultural families. *Environ Health Perspect* 1995: 103(12): 1126–1134.

Surgan M.H., Congdon T., Primi C., Lamster S., and Loiu-Jacques J. Pest control in public housing, schools and parks: urban children at risk. In: Bureau DoLE, (ed.). New York State Library, Albany, NY, 2002, 202–7643.

Todd G.D., Wohlers D., and Citra M. Toxicology profile for pyrethrins and pyrethroids, US Department of Health and Human Services. Agency for Toxic Substance and Disease Registry: Atlanta, GA, 2003 http: //www.atsdr.cdc.gov/toxprofiles/tp155.html.

US EPA. Chlorpyrifos. Revised Risk Assessment and Agreement with Registrants, Washington DC USA, 2000a.

US EPA. Diazinon. Revised HED Preliminary Human Health Risk Assessment for the Reregistration Eligibility Decision (RED). D262343, PC Code:057801. List A, Case No. 0238 Washington, DC: USA, 2000b.

US EPA. Pesticides industry Sales and Usage Report. 1998 and 1999 Market Estimates. Office of Prevention, Pesticides and Toxic Substances, Washington, DC: USA, 2002.

US EPA. Restricted Use Products Report. June 2003.

Whitmore R.W., Immerman F.W., Camann D.E., Bond A.E., Lewis R.G., and Schaum J.L. Non-occupational exposures to pesticides for residents of two US cities. *Arch Environ Contam Toxicol* 1994: 26(1): 47–59.

Whyatt R.M., Camann D.E., Kinney P.L., Reyes A., Ramirez J., and Dietrich J., et al. Residential pesticide use during pregnancy among a cohort of urban minority women. *Environ Health Perspect* 2002: 110(5): 507–514.

Whyatt R.M., Rauh V., Barr D.B., Camann D.E., Andrews H.F., and Garfinkel R., et al. Prenatal insecticide exposures and birth weight and length among an urban minority cohort. *Environ Health Perspect* 2004: 112(10): 1125–1132.

Zota A., Adamkiewicz G., Levy J.I., and Spengler J.D. Ventilation in public housing: implications for indoor nitrogen dioxide concentrations. *Indoor Air* 2005: 15(6): 393–401.