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American Healthy Homes Survey: A National Study of Residential Pesticides Measured from Floor Wipes

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The U.S. Department of Housing and Urban Development in collaboration with the United States Environmental Protection Agency conducted a survey measuring lead, allergens, and insecticides in a randomly selected nationally representative sample of residential homes. Multistage sampling with clustering was used to select the 1131 homes of which a subset of 500 randomly selected homes included the collection of hard surface floor wipes. Samples were collected by trained field technicians between June 2005 and March 2006 using isopropanol wetted wipes. Samples were analyzed for a suite of 24 compounds which included insecticides in the organochlorine, organophosphate, pyrethroid and phenylpyrazole classes, and the insecticide synergist piperonyl butoxide. The most commonly detected were permethrin (89%), chlorpyrifos (78%), chlordane (64%), piperonyl butoxide (52%), cypermethrin (46%), and fipronil (40%). Mean and geometric mean (GM) concentrations varied widely among compounds, but were highest for *trans*-permethrin (mean 2.22 ng/cm^2 and GM 0.14 ng/cm^2) and cypermethrin (mean 2.9 ng/cm^2 and GM 0.03 ng/cm^2). Results show that most floors in occupied homes in the U.S. have measurable levels of insecticides that may serve as sources of exposure to occupants.

Introduction

Insecticides are commonly applied in and around homes to control a variety of insect pests. In the United States, insecticides representing multiple chemical classes and different formulations are available for purchase by consumers or professionals. Estimates derived from the National Home and Garden Survey conducted in 2000 and 2001 indicate that about 78 million U.S. households (74%) use pesticides (*1*), spending nearly 1.3 billion dollars to purchase insecticides and applying 888 million pounds of active ingredient (*2*). One of the earliest large regional studies, the Non-Occupational Pesticide Exposure Study (NOPES) (*3*), showed that pesticide usage in and around homes resulted in elevated concentrations (*4*). Other large regional surveys have continued to support this observation and expanded on the relationships between measured household concentrations and pesticide applications (agricultural, home garden, professional), proximity to outdoor sources, and potential pathways for occupant exposure (*5*−*7*). Past residential studies have been limited in scope, with measurements derived from smaller geographical areas precluding any estimates of a national distribution of insecticide residues.

The popularity and availability of residential-use insecticides have transitioned over the last 30 years through the different classes of organochlorine, organophosphate, carbamate, and pyrethroid insecticides. These changes in availability and consumer preference contribute to an ongoing need to survey homes to obtain current and high quality measurement data to assess risk and evaluate mitigation efforts. The Food Quality Protection Act of 1996 requires the United States Environmental Protection Agency (U.S. EPA) to consider aggregate and cumulative risks and the need for additional safeguards to protect children during the reassessment of current-use pesticides and future pesticide registrations. Data are required to assess the potential for human exposure in homes and, in particular, to better understand children's exposures.

EPA collaborated with the U.S. Department of Housing and Urban Development (HUD) on a national survey of housing related hazards in U.S. residences. The resulting study, the American Healthy Homes Survey (AHHS), was designed to provide nationally representative estimates of demographic and environmental parameters related to American housing. The AHHS measured residential concentrations and loadings of lead, allergens, mold, pesticides, and arsenic to estimate the distributional parameters and to provide information that can be used to examine changes in levels of these indoor contaminants over time. Questionnaire and environmental data were collected from a stratified, nationally representative sample of 1131 U.S. residences. AHHS sample collection included surface wipes from the kitchen area in homes, homeowner vacuum bags, and soil samples from outside the home. Samples were analyzed to determine lead in dust, soil, and paint and the prevalence of lead hazards, the levels and patterns of various indoor allergens and molds from dust in homes, and the occurrence of pesticides in homes.

This study was designed to enhance the understanding of current levels of selected contaminants in U.S. homes, provide predictions of the ranges of concentrations to which the U.S population might be exposed in their living environments, and support the identification of the key household factors that influence those concentrations. The data presented in this paper are the insecticide loadings. These data provide a nationally representative distribution of indoor insecticide loadings measured on hard surface floors of residential housing. These findings represent a first step to providing baseline data for understanding the types of pesticides found in residences and temporal changes in

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chemical loadings. They are also useful for determining potential occupant exposure to insecticide residues.

Experimental Section

Sampling Strategy. A wide range of demographic and environmental measurements were collected from a nationally representative sample of 1131 public and private housing units across the U.S. The housing units were selected as a 3-stage cluster sample of residential housing. The first stage of sampling was the selection of 100 Primary Sampling Units (PSUs) that were counties or groups of contiguous counties having a minimum population of 15 000 based on the 2000 Census and an end-to-end distance generally not exceeding 100 miles. Most were Metropolitan Statistical Areas (MSAs) or natural parts thereof. The PSU samples were drawn with a probability proportional to the 2000 census population in households, after stratification based on census region, MSA status, population size, income and percent minority (Black and Hispanic). The second stage sample consisted of five Census block segments chosen in each PSU with a probability proportional to the number of housing units. The third and final stage was the selection of four to six candidate housing units in each segment from validated lists of households acquired from commercial sources. Floor wipe samples were collected from a random subsample of 500 housing units. In addition, the first pesticide housing unit in each PSU was designated for a quality control (QC) pesticide sample.

Human Subjects Review. This study required Institutional Review Board (IRB) approval, which was obtained as defined by the U.S. Department of Health and Human Services (DHHS) (see 45 CFR 46 as codified by HUD at 24 CFR 60), due to the involvement of human subjects in the research plan. The protocol was reviewed and approved by the DHHS Federalwide Assurance for the Protection of Human Subjects.

Sampling Procedure. The field study was performed between June 2005 and March 2006 by trained two-person field teams that included one interviewer and one licensed lead-based paint inspector/risk assessor technician. The team administered a survey questionnaire and collected a variety of environmental samples and observations at each successfully recruited housing unit.

The floor wipe media consisted of two Excilon brand (10 × 10 cm, 6-ply, Tyco Health Care Group, Mansfield, MA) surgical sponges (wipes) composed of bonded rayon microfibers. The wipe media were precleaned using Soxhlet extraction in dichloromethane followed by hexane for a minimum of 10 h to remove potential chemical interferences and reduce the total mass of microfiber binder present in the media. Following extraction, the media were dried in a vacuum oven at 40 °C until no solvent odors were detected (approximately 8 h), and placed into solvent-rinsed amber glass jars for storage until deployment to the field.

Floor wipe sample kits provided to the field teams consisted of an aluminum template with an internal sampling dimension of 929 cm^2 (144 in^2), one glass Petri dish (10 cm diameter), one roll PTFE tape (1 in wide) for sealing jars, one insulated shipping box (28 × 23 × 18 cm), one square piece of aluminum foil, one pair of Nitrile gloves, two vials containing 6 mL each of isopropanol (IPA), and one solvent cleaned jar containing two precleaned Excilon wipes.

The surface selection protocol directed pesticide wipe samples to be collected from hard surfaces with priority given to the kitchen area, although provisions were made to sample surfaces in other locations if sampling in the kitchen was not possible. The field sampling teams were directed to perform the wipe sampling in two separate locations within the selected room in areas of low foot traffic and away from cooking areas (i.e., the stove). Prior to sampling, the tools associated with the sample collection were cleaned by wiping with prepackaged, disposable isopropanol wipes. A clean work surface was prepared by unfolding an alcohol-wiped sheet of aluminum foil flat on the floor near the locations to be sampled and placing the cleaned Petri dish onto the foil surface. The two wipes were removed from the jar and placed onto the Petri dish. Each wipe was wetted with a 6 mL aliquot of analytical grade IPA. The precleaned aluminum template was positioned at the sampling location on the floor. The surface wiping was performed over the entire sampling location within the confines of the aluminum template (sampling area = 929 cm^2) using an overlapping side-to-side "S" or "Z" pattern. Upon completion of wiping within the template from one side of the template to the other, the wipe was folded in half, sample side folded inward, and a similar procedure was performed from the top to bottom of the template. The template was moved to a second location and a second wipe was collected. The two wipe samples were composited in the original sample jar and Teflon tape was used to seal the outside of the jar. The jars were stored on frozen blue ice in an insulated shipper. All sample jars were individually inserted into a tight fitting bubble pack bag followed by placement into a 1-quart plastic bag and placed into an insulated shipper. The samples were transported in insulated shipping boxes under darkened conditions at reduced temperatures (approximately 40 °C) to the local base of operations where they were transferred to a portable freezer for interim storage at 10 °C. At the termination of field activities for the PSU, all collected insecticide wipe samples were moved from the freezer and placed into insulated shippers with frozen blue ice for express shipment to the designated laboratory. Chain-of-custody (CoC) was maintained for each sample throughout the study.

Quality Control Procedures. Quality control (QC) media were prepared by the U.S EPA for field deployment as well as supplied to the analytical laboratory. The results are summarized in Supporting Information Tables S1−S4. Field QC consisted of (a) field blank media; (b) blind blank media and blind spiked media; (c) laboratory solvent blanks and spikes; and (d) laboratory media blanks and media spikes. The field blank media demonstrates the effect of shipping and handling on the sample integrity. They were shipped from the U.S. EPA directly to the field teams and consisted of 55 solvent cleaned media prepared in a manner consistent with the media used during field sampling. At the completion of sampling the media were shipped to the analytical laboratory. Field spikes were prepared to assess media and analyte stability following shipping and handling. The media were prepared by fortifying two wipes with a spiking solution that contained the insecticides allethrin, chlorpyrifos, cyfluthrin, λ-cyhalothrin, cypermethrin, diazinon, esfenvalerate, permethrin, sumithrin, and tetramethrin. Four field spikes were prepared by fortifying the sample (composed of two wipes) with 10 ng, and six field spikes were prepared by fortifying the media with 500 ng. These QC samples were shipped to the field, retained under darkened conditions at reduced temperatures until the completion of sampling at the PSU, then shipped to the analytical laboratory. In addition, nine blind spiked media (also composed of two wipes) were prepared by spiking with 750 ng of standard solution, but were shipped directly to the analytical laboratory. Blind blank media were precleaned and packaged by the U.S. EPA and shipped directly to the analytical laboratory as a performance check.

In order to ensure the integrity of laboratory processes during the chemical analysis, laboratory QC was implemented consisting of solvent blanks, solvent spikes, blank wipe media, and spiked wipe media. A total of 55 solvent blanks and 43 media blanks were analyzed during the chemical analysis of field collected samples. A total of 48 laboratory solvent spikes and 42 laboratory media spikes were prepared and analyzed.

TABLE 1. Weighted Summary Statistics for Insecticides Measured in Hard Surface Floor Wipe Samples in the AHHS

				ng/cm^2								
class[a]	insecticide name	N[b]	detection frequency (%)	MDL	mean[c]	SD	50th	75th	95th	maximum	GM	GSD
PP	fipronil	478	40	0.0007	0.16	1.06	<MDL	<MDL	0.40	20	0.002	9.7
OC	γ-chlordane	478	74	0.0004	0.11	0.72	<MDL	0.02	0.57	14.	0.01	10
OC	α-chlordane	478	69	0.0012	0.08	0.38	<MDL	0.02	0.37	6.5	0.01	7.3
OC	p,p′-DDT	422	41	0.0003	0.03	0.12	<MDL	0.01	0.11	2.1	0.001	10
OC	p,p′-DDE	478	33	0.0002	0.01	0.04	<MDL	<MDL	0.04	0.75	0.0006	7.9
OC	heptachlor	478	13	0.0024			<MDL	<MDL	0.06	2.1		
OP	chlorpyrifos	479	78	0.0009	0.50	6.3	0.01	0.06	0.7	135	0.01	10
OP	diazinon	480	35	0.0007	0.03	0.12	<MDL	0.01	0.15	1.1	0.002	8.3
OP	malathion	480	15	0.0015			<MDL	<MDL	0.05	4.1		
PY	*cis*-permethrin	459	89	0.0018	1.4	4.8	0.11	0.56	6.72	68	0.11	12
PY	*trans*-permethrin	448	88	0.0024	2.2	7.9	0.13	0.74	10.14	102	0.14	13
PY	cypermethrin	480	46	0.0016	2.9	19	<MDL	0.16	10.66	231	0.03	20
PY	bifenthrin	466	33	0.0002	0.18	2.1	<MDL	<MDL	0.12	43	0.0007	12
PY	deltamethrin	468	27	0.0097			<MDL	0.03	0.91	72		
PY	sumithrin	468	22	0.0007			<MDL	<MDL	0.36	4.8		
PY	λ-cyhalothrin	467	21	0.0006			<MDL	<MDL	0.18	61		
PY	cyfluthrin	480	17	0.0007			<MDL	<MDL	0.50	129		
PY	esfenvalerate	468	15	0.0086			<MDL	<MDL	0.43	17		
PY	tetramethrin	480	15	0.0003			<MDL	<MDL	0.18	24		
PY	allethrin	468	7	0.023			<MDL	<MDL	0.21	10		
PY	pyrethrin I	467	5	0.033			<MDL	<MDL	0.09	222		
PY	fenpropathrin	479	4	0.0016			<MDL	<MDL	<MDL	1.1		
PY	imiprothrin	468	3	0.0414			<MDL	<MDL	<MDL	3.7		
PY	pyrethrin II	468	2	0.12			<MDL	<MDL	<MDL	156		
PY	prallethrin	468	1	0.014			<MDL	<MDL	<MDL	0.71		
PY	resmethrin	467	0.4	0.0027			<MDL	<MDL	<MDL	14		
SYN	PBO[d]	475	52	0.0005	4.5	37	<MDL	0.10	5.26	747	0.01	30

[a] PP = phenylpyrazole, OC = organochlorine, OP = organophosphate, PY = pyrethroid, SYN = insecticide synergist. [b] The differences of the N values reported in the table are associated with the removal of compounds due to interferences or missing ancillary information. [c] Mean values are not reported for insecticides with <30% detection frequencies. [d] PBO = piperonyl butoxide.

Spike amounts varied by the insecticide, but ranged between 20 and 1250 ng.

Chemical Analysis Procedures. Samples were analyzed for a suite of 24 past and current residential-use insecticides from the organochlorine, organophosphate, pyrethroid, and phenylpyrazole classes as well as an insecticide synergist (Table 1). The surrogate recovery standards (SRSs) included $^{13}C_{12}$-p,p-DDT, ^{13}C-*trans*-chlordane, fenchlorphos, and $^{13}C^6$-*trans*-permethrin. The wipes were fortified with 250 ng of the SRS mix prior to extraction to approximate analyte levels expected from field samples. The wipe samples were prepared for extraction by packing two wipes into a 33 mL accelerated solvent extraction cell and extracting with dichloromethane at 2000 psi and 100 °C. The extracts were concentrated using Kuderna Danish with hexane additions to achieve an azeotropic distillation of the IPA from the extract. The extracts were further reduced to 1 mL and loaded onto a solid-phase extraction cartridge containing 0.5 g of aminopropyl (Discovery DSC-NH2, Supelco) conditioned with 6 mL of dichloromethane. The samples were eluted with two, 5-mL washings of dichloromethane. The eluant was reduced using nitrogen evaporation and brought up to 1 mL in hexane. The final extract was spiked with the internal standard (IS) dibromobiphenyl at 100 ng/mL. Sample extracts were analyzed using GC/MS for a suite of 24 insecticides. Two or three ions, a quantification ion and 1−2 qualifier ions, were monitored for each compound. The IS method of quantification was used. Pesticides were quantified using eight calibration curve solutions that typically ranged from 5−1000 ng/mL; the calibration solutions were interspersed among the samples throughout the run order. Linear regression analysis was used to generate the calibration curve for each analyte, and this curve was applied to quantify the detected analytes in samples. Solutions where an analyte concentration exceeded the upper calibration point by more than 15% were diluted, respiked with the IS, and reanalyzed. The data for all analytes in the nominal extract that did not exceed the upper calibration point were reported as such; only the analytes that exceeded the upper calibration point were reported from the diluted sample analysis data. GC conditions were as follows: Phenomenex ZB-35 column, 0.25 mm id × 0.25 μm film thickness × 30 m length; flow rate of 1 mL/min helium; injection port temperature of 300 °C; 2 μL injection volume; oven temperature program: 100 °C for 1 min; 100−130 °C @ 25 °C/min, 130−340 °C @ 6 °C/min, hold 340 °C for 5 min. Detection limits are shown in Table 1.

Method Detection Limits. The method detection limit (MDL) was determined for the analytes following the guidelines in 40 CFR Part 136, Appendix B (*8*). Specifically, seven wipe samples were spiked with surrogate recovery standards and extracted; the extract was cleaned using SPE, and concentrated to 1 mL. The extracts were combined and a 1 mL aliquot of the extract was removed as the matrix blank and analyzed seven times. Three additional 1 mL aliquots were removed to produce the spiked samples. These were labeled "A", "B", and "C" with each aliquot spiked with a 2.5, 5, and 10 μL volume of a multistandard solution composed of analytes ranging from 0.8 to 600 ng/mL and an internal standard (10 μL, 100 ng/sample). The solutions were analyzed seven times each.

The MDLs, were calculated separately for the A, B, and C solutions, as follows:

$$\mathrm{MDL}_{Xi} = t_{(n-1,1-\alpha=0.99)} S_i$$

where MDL_{Xi} = method detection limit determined for target analyte X from solution i, where i = A, B, or C, $t(n-1, 1-\alpha = 0.99) = 3.143$, the student's t value appropriate for a 99% confidence level and a standard deviation estimate with six degrees of freedom (seven replicate analyses), and S_i =

standard deviation of the seven replicate analyses for solution *i*, where i = A, B, or C.

The relative percent difference (RPD) between the calculated MDL and the solution's theoretical analyte concentration was calculated for each analyte in each solution. For a given analyte, the calculated MDL with the lowest RPD from the solution's theoretical concentration was reported.

Data Analysis. The data underwent corrections in preparation for finalization of the database. The final data were surrogate recovery corrected using the previously described groupings. Observations with levels below the MDL were replaced by values equal to the individual MDL divided by the square root of 2 (*9*), recognizing the potential for bias in the summary statistics among those compounds with low detection frequencies. Sample analysis results were not corrected for background or for recoveries that were measured in field and laboratory quality control samples. Values with reported interferences were individually evaluated and generally excluded from the final data set. The values reported for cyfluthrin, cypermethrin, and tetramethrin represent the summation of their individual isomers measured above the MDL. Sample weights were calculated to adjust for bias resulting from nonresponse and to provide national estimates. The means and standard deviations for values with detection frequencies less than 30% were not reported. A total of 495 samples were actually collected with a laboratory loss of five samples. Inadequate or missing ancillary information and analytical interferences in target or SRS analytes resulted in the removal of some samples from the final data set, thus the number of samples reported for each compound varies.

Results and Discussion

1. Quality Control Results. Quality control results are shown in Tables S1 through S4 of the Supporting Information, with the field blank results summarized in Table S1. Although 54 field blanks were submitted for analysis, only 37 are reported. The results showed some of the QC samples with slightly elevated background concentrations at 5−10 times the MDL for bifenthrin, chlorpyrifos, permethrin, cyfluthrin, λ-cyhalothrin, cypermethrin, malathion, and piperonyl butoxide. A close examination of the data revealed three QA samples that were actually field samples and were removed from the QA report. A lack of consistency in the co-occurrence and concentration of the analytes suggested that 14 field blank media might have been inadvertently used by field teams to wipe surfaces. Additional analysis of a randomly selected compound (α-chlordane) comparing blank media to the field samples collected from the same homes showed relatively similar results (Pearson rho = 0.96), further supporting the contention that field blank media were incorrectly used to wipe surfaces. As a result, any field blank containing >100 ng of any analyte was omitted from final reporting.

Blind media blanks (Supporting Information Table S1) showed slightly elevated backgrounds at 3−5 times the MDL for only five of the pesticides in the standard spiking solution, namely bifenthrin, permethrin, cypermethrin, malathion, and piperonyl butoxide. Supporting Information Table S2 describes the results of the field spiked media. Recovery for the 10 ng spike ranged from 0% (below the detection limit) to 137%. This range is not unexpected due to the wide ranging detection limits spanning from 1 to 230 ng across the various analytes. However, the fortifications at the 500 ng concentration also showed variable performance, with recoveries ranging from 46 to 99%. The results of the media spiked at 750 ng of standard solution gave improved mean recoveries ranging from 69 to 108% for all compounds. It is doubtful the varied recoveries are associated with losses from field handling and shipping, but are more likely linked to the difficulties associated with implementing a multi residue method while retaining optimization for all compounds.

Laboratory solvent blanks (Supporting Information Table S3) showed contamination levels similar to the laboratory media blanks, with the exception of λ-cyhalothrin and malathion, which were not detected in any of the 55 laboratory solvent blank samples. Laboratory blank media had contamination levels for the same pesticides detected in the field blanks, but at roughly half the levels, with the exception of malathion, which was detected at 160 ng (60 times the MDL) in one laboratory media blank sample.

Results for the laboratory solvent spikes and media spikes are shown in Supporting Information Table S4. Laboratory fortified media gave recoveries ranging from 70 to 94%, demonstrating adequate method performance for most compounds. Although deltamethrin, fipronil, and pyrethrin recoveries from spiked media fall outside this range, their recoveries from solvent spikes were good, ranging from 87−114%. This suggests that further method optimization for these compounds are needed; specifically more vigorous accelerated solvent extraction conditions may be needed to extract these compounds from the wipe media.

Laboratory QC results revealed no similar background issues and supported the determination that contamination likely occurred under field conditions, perhaps as a result of technician error while handling the QC samples. Furthermore, since some contaminants were not part of the spiking solution they could not have been introduced during laboratory procedures.

2. Pesticide Distributions. *Detection Frequency.* The residential use insecticides measured varied in their detection frequency both across and within chemical classes. Table 1 lists the insecticides studied by class. The phenylpyrazole, fipronil, for example, was detected in about 40% of all the homes sampled. Fipronil is a relatively new residential use insecticide formulated to be topically applied to pets for the control of ectoparasites, but it is also used as a termiticide and contained in baits to control ants and cockroaches. The organochlorine (OC) insecticides, which include chlordane, heptachlor, DDT and its degradate DDE, were popularly used to control a variety of urban insect pests but were voluntarily withdrawn from use or banned in the 1970s and 1980s. Nonetheless the frequencies of detection for chlordane, DDT and heptachlor were 74, 42, and 13%, respectively. Moreover, chlordane was among the three most frequently detected insecticides in the study. The organophosphate compounds, including chlorpyrifos and diazinon, replaced the OCs in the marketplace. They are broad spectrum insecticides that were formulated for the control of general pests both in the home and garden. Chlorpyrifos was deregistered by the EPA for most residential uses in 2000 (10), and diazinon in 2004 (11). Chlorpyrifos and diazinon were detected in 78 and 35%, respectively, of the homes. Currently, pyrethroid insecticides are marketed to consumers and applied by pest control professionals to control general insect pests. Among the suite of pyrethroid insecticides and botanically derived pyrethrins measured in this study, permethrin was observed most frequently (89%). Cypermethrin was ranked second, but was detected only about half as frequently (46%). All other compounds were measured in less than 35% of the homes, with the exception of piperonyl butoxide (PBO). PBO is an insecticide synergist typically coformulated with the botanical extract pyrethrum (represented here by pyrethrin I and pyrethrin II) and analogs of its active isomers (resmethrin, allethrin, tetramethrin). PBO was detected in 52% of the homes sampled. Resmethrin, allethrin and tetramethrin, however, were detected in 0.4, 6.6, and 15% of the homes, respectively. The unexpected disparity of the detection frequency of these compounds relative to PBO suggests rapid environmental degradation compared to PBO. In addition,

FIGURE 1. A comparison of detection frequencies of select insecticides from the AHHS and child care centers (Tulve et al., 2006).

the relative detection frequencies may be impacted, in part, by the different detection limits.

Concentrations. The surface loadings determined in this study are summarized in Table 1. The mean and geometric mean (GM) surface loadings for compounds with detection frequencies of less than 30% are not reported. A consideration of the GM by classes shows that the OCs, OPs are uniformly low, whereas the pyrethroids exhibit a broader range. The highest measured GM were *cis*- and *trans*-permethrin (0.11 and 0.14 ng/cm^2, respectively), whereas chlorpyrifos (0.01 ng/cm^2) and cypermethrin (0.03 ng/cm^2) were found at the next highest levels, but were an order of magnitude lower. The high surface loadings for permethrin are consistent with its current popularity for residential use. The results for chlorpyrifos, on the other hand, were notable since chlorpyrifos has not been available for consumer purchase since the beginning of 2002 (*10*). The presence of chlorypyrifos at these elevated levels might be associated with its limited remaining registrations for the control of termites, or more likely may be a result of the compound's persistence in the indoor residential environment.

Within the pyrethroid class there is a large difference between the detection frequency and GM of the natural pyrethrins and early analogs (allethrin and resmethrin), and the later generation pyrethroids (e.g., cyfluthrin, cypermethrin, deltamethrin, permethrin). The utility of the pyrethrins and early analogs were limited based on their rapid decomposition in air and light. The later compounds were intentionally synthesized to be more thermally and photolytically stable while maintaining excellent insect toxicity (*12*). Accumulation of the later generation pyrethroids over time may result in a higher GM concentration among homes.

In general, the observed variability for all compounds across classes was high (Table 1), with geometric standard deviations (GSD) greater than six. The variability among pyrethroids, as a class, was particularly high (GSD typically greater than 10). The higher GSD for pyrethroids relative to OPs and OCs translates into a higher fold-difference between the 95th percentile and the 50th percentile values. The relatively high 95th percentile values for the pyrethroids likely reflect market availability and recent use. The variability observed with PBO (GSD = 25.6) was much greater than that of the pyrethroids and might reflect its proportionality in formulations. Insecticides that contain PBO as a synergist are typically formulated with a ratio of PBO to pyrethrins ranging from 5:1 to 20:1, based on both economic and biological effectiveness considerations. In addition, the higher variability of PBO may be associated with the popularity of PBO formulations in the consumer market and the hunting mentality of consumers using point and spray application, contrasted against the more systematic applications performed by professional services (*13*).

TABLE 2. The Mean Surface Loadings of Select Insecticide Measures from Residential Homes and Child Care Centers

		insecticide name/mean concentration(ng/cm^2)										
study name	**N**	**%D**	**chlorpyrifos**	**%D**	**diazinon**	**%D**	**c-permethrin**	**%D**	**cypermethrin**	**%D**	**α -chlordane**	**%D**	**4,4′-DDT**
AHHS[b]	480	78	0.01(10.49)	35	0.00(8.25)	89	0.11(12.20)	45	0.03(19.95)	69	0.01(7.25)	42	0.00(10.36)
CTEPP North Carolina (Morgan et. al, 2007)[b]	28	89	0.0063(4.6)	68	0.002(8.4)	93	0.034(8.6)	NR[c]	NR	NR	NR	NR	NR
CTEPP Ohio (Morgan et. al, 2004)[b]	21	86	0.0043(8.8)	NR	NR	71	0.011(12)	NR	NR	NR	NR	NR	NR
CCC (Tulve et. al, 2006)[a]	168	89	0.03	67	0.01	72	0.03	23	0.01	NR	NR	NR	NR
Julien et al., 2007[d]	42	100	0.03	98	0.04	100	0.68[f]	90	0.37	NR	NR	NR	NR
Quandt et al., 2004[e]	41	78	0.89 ± 1.84	34	0.26 ± 0.31	66	3.06 ± 6.64	NR	NR	34	0.19 ± 0.24	5	1.00 ± 0.05
CHAMACOS (Bradman et. al, 2006)[d]	20	95	0.046	95	0.038	85	0.1	NR	NR	NR	NR	NR	NR

[a] The CCC was conducted in private and intitutional daycares. [b] Values in parenthesis are the geometric standard deviation. [c] NR = not reported. [d] Median values are reported. [e] Value represents mean ± standard deviation. [f] Value is arithmetic sum of cis and trans-permethrin.

In 2001, HUD and the EPA previously collaborated in performing a national survey, titled the First national Environmental Health Survey, which estimated surface loadings of insecticides in child care centers (CCC) in both commercial buildings and residential dwellings (*14*). A comparison of detection frequencies for select insecticides sampled from hard surfaces (the AHHS MDLs were set to the higher limits in the CCC study to eliminate inconsistencies in detection limits) is presented in Figure 1. Fipronil was detected four times more frequently in the AHHS. Similarly, the pyrethroids (including PBO) were consistently more frequently detected in the AHHS than in the child care centers. Interestingly, the individual compounds appear to exhibit similar proportionality in both studies. Diazinon and chlorpyrifos were detected at lower frequencies in the AHHS (35 and 66%, respectively) than in child care centers (70 and 90%, respectively). Comparatively the GMs for all compounds were similar to those from the AHHS except for chlorpyrifos and cypermethrin, which were 1 and 2 orders of magnitude lower in CCC than AHHS, respectively (Table 2).

The findings from this study stand in contrast to those of studies performed on a more regional level or targeted toward specific subpopulations expected to experience elevated exposures (i.e., farm worker families and residents of urban housing). Table 2 summarizes geometric mean concentrations for α-chlordane, chlorpyrifos, cypermethrin, 4,4′-DDT, diazinon, and *cis*-permethrin from select studies (*5, 6, 14*−*16*). The Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP) (*6*) study measured surface loadings from North Carolina and Ohio. The AHHS GMs were higher for two of three analytes common to both studies, namely, chlorpyrifos and permethrin, but was similar for the third, diazinon. Since the AHHS values are representative of national distributions, the differences observed between AHHS and CTEPP surface loadings may point to differences among different regions of the U.S. for these compounds or temporal changes due to regulations. Examination of the AHHS data for regional differences is needed.

Studies establishing indoor surface loadings from the homes of farm workers have been reported for agricultural areas in the Salinas Valley of California (*5*) and the mountains of North Carolina and Virginia (*16*). In both of these locations higher indoor surface loadings were expected due to proximity of homes to agricultural fields, spray drift, wind circulation of dust (*16*), or transport of pesticides on workers' clothing and track-in (*17*). Although the median surface concentrations for chlorpyrifos and *cis*-permethrin in AHHS homes differed little from the concentrations in homes of farm workers in the Salinas Valley (*5*), a comparison with the homes of farm workers in North Carolina and Virginia revealed that the geometric mean values for diazinon, *cis*-permethrin, cypermethrin, chlordane, and DDT were lower in AHHS.

Urban multiunit housing is considered to have a high prevalence of insecticide use due to higher rates of pest infestation (*18, 19*). Consequently it would be expected that surface loadings might be lower in AHHS than those measured from the urban housing. Indeed, the pyrethroid surface loadings at the 50th percentile in AHHS were much lower than the surface loadings reported by Julien et al. (*15*) from the kitchen floors of 42 apartments in urban public housing in Boston, Massachusetts. The values of 0.68, 0.37, and 0.11 ng/cm^2 reported for permethrin, cypermethrin, and cyfluthrin, respectively, were 3−40 times greater than corresponding AHHS values. Surprisingly, the median value of 0.03 ng/cm^2 reported for chlorpyrifos was not much different than the AHHS.

The AHHS survey establishes baseline estimates of pesticide loadings in homes across multiple classes of insecticides. These findings suggest that insecticides used in and around homes remain measurable from surfaces in homes. The high detection frequencies observed for chlordane, chlorpyrifos, and permethrin suggest these compounds are essentially ubiquitous in our living areas and that popular use, both past and present, has a major influence on their occurrence in homes. Clearly, insecticides removed from the U.S. market persist in homes and should continue to be considered in cumulative and aggregate risk assessments. The methods applied in the AHHS to examine a broad spectrum of insecticides will be of value to future study design. In addition, these high quality national estimates provide the best baseline measurements of surface loadings in homes currently available. Future analysis of these data will expand the utility of the insecticide estimates by correlating surface loadings with housing factors and occupant questionnaire data collected during the AHHS. The analysis of the residential surface loadings based on regional distributions will expand our understanding of potential exposure to residential use insecticides.

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Supporting Information Available

Quality assurance data for the study. This material is available free of charge via the Internet at http://pubs.acs.org.

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