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Comparison of pesticide levels in carpet dust and self-reported pest treatment practices in four US sites

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Epidemiologic studies have used both questionnaires and carpet dust sampling to assess residential exposure to pesticides. The consistency of the information provided by these two approaches has not been explored. In a population-based case–control study of non-Hodgkin's lymphoma, carpet dust samples were collected from the homes of 513 control subjects in Detroit, Iowa, Los Angeles, and Seattle. The samples were taken from used vacuum cleaner bags and analyzed for 30 pesticides. Interviewers queried subjects about the types of pests treated in their home using a detailed questionnaire accompanied by visual aids. Geographic variations in pesticide levels were generally consistent with geographic differences in pest treatment practices. Los Angeles residents reported the most treatment for crawling insects, fleas/ticks, and termites, and Los Angeles dust samples had the highest levels of propoxur, chlorpyrifos, diazinon, permethrin, and chlordane. Iowa had the most treatment for lawn/garden weeds, and also the highest levels of 2,4-dichlorophenoxyacetic acid and dicamba. Although Seattle had the highest proportion of subjects treating for lawn/garden insects, the lawn/garden insecticides were higher in other sites. Multivariate linear regression revealed several significant associations between the type of pest treated and dust levels of specific pesticides. The strongest associations were between termite treatment and chlordane, and flea/tick treatment and permethrin. Most of the significant associations were consistent with known uses of the pesticides; few expected associations were absent. The consistency between the questionnaire data and pesticide residues measured in dust lends credibility to both methods for assessing residential exposure to pesticides. The combined techniques appear promising for epidemiologic studies. Interviewing is the only way to assess pesticide exposures before current carpets were in place. Dust sampling provides an objective measure of specific compounds to which a person may have been exposed through personal use of a pesticide or by drift-in or track-in from outside, and avoids recall bias.
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Introduction

Exposure to pesticides has been linked to cancer in farmers and other occupational groups (Zahm et al., 1997). The general population is also exposed to pesticides, principally in the home (Nigg et al., 1990). Pesticides enter the home from indoor use, track-in or drift-in from outdoors, intrusion of vapors from foundation treatments, or take-home contamination from occupational use (Bradman et al., 1997; Lewis et al., 1999, 2001). Pesticides may persist for months or years inside the home, where they are protected from degradation by sunlight, rain, temperature extremes, and most microbial action (Lewis et al., 1994).

Carpets and cushioned furniture are repositories for pesticides (Camann et al., 1991; Roberts et al., 1992; Simcox et al., 1995). Once in the carpet, pesticides migrate from the fibers into the underlying polyurethane foam pad. The fibers and pad appear to act as long-term reservoirs that continuously transfer pesticides to carpet dust (Camann, 1994; Fortune et al., 2000). Numerous studies have documented the presence of pesticides in carpet dust, including pesticides that had been commercially unavailable for many years (Starr et al., 1974; Roinestad et al., 1993;

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Whitmore et al., 1994; Lewis et al., 1994; Simcox et al., 1995; Bradman et al., 1997; Lioy et al., 2000; Pang et al., 2002).

In epidemiologic studies of the health effects of pesticides, exposure assessment has traditionally been based on self-reported information. However, detailed recall of past pesticide use can be difficult, and in case–control studies differential recall between cases and controls can lead to biased risk estimates. Carpet dust sampling provides a more objective basis for exposure assessment. Because carpets act as long-term pesticide repositories, pesticide concentrations in carpet dust may reflect integrated pesticide exposure over the lifetime of the carpet, potentially more relevant to disease risk than recent or current exposure. Carpet dust sampling can also reveal the specific active ingredients to which a person may have been exposed, which is difficult to collect by interview.

Several recent studies have used carpet dust sampling for pesticide exposure assessment, but the consistency of the information provided by self-report vs. carpet dust sampling is largely unexplored. In a population-based case–control study of adult non-Hodgkin's lymphoma (NHL), we are using both methods to assess exposure to pesticides and other compounds. This provided the opportunity to examine whether levels of pesticides measured in carpet dust parallel self-reported information on pesticide use.

Methods

Study Population and Data Collection

The NHL case–control study was conducted in four areas covered by the Surveillance, Epidemiology, and End Results (SEER) Program of the National Cancer Institute: the Detroit, Michigan metropolitan area (Wayne State University); the state of Iowa (University of Iowa); Los Angeles County, California (University of California); and the Seattle, Washington metropolitan area (Fred Hutchinson Cancer Research Center). Controls were 1057 residents of the four SEER registry areas between the ages of 20 and 74 years. They were frequency matched to NHL cases on age, gender, race, and area. Controls under age 65 years were identified from households contacted via a random-digit-dial procedure. Controls aged 65–74 years were identified from Center for Medicare and Medicaid Services files. In Los Angeles and Detroit, we oversampled for African-American subjects with the goal of interviewing 100 African-American cases and an equal number of controls in each of these centers. The study was approved by the human subjects review boards at all participating institutions and informed consent was obtained from each participant before interview.

Computer-assisted personal interviews were conducted in the homes of study subjects. Each subject completed a lifetime residential history calendar prior to the home visit. Starting with the current home, interviewers asked whether pesticides were used to treat each of several types of pests (e.g., flying insects, lawn weeds). As the interviewer asked about each pest type, the subject was shown a showcard with examples (words and drawings) of specific pests. The interviewer then asked who applied the pesticide(s), the application frequency, and the form of the product(s). This was repeated for each home in which the subject lived for at least 2 years during the past 30 years. We did not ask subjects to name the specific product used because people typically have trouble recalling this information (Bradman et al., 1997). The paper version of the questionnaire (programmed into a computer and administered verbally) and the showcards can be found in http://dceg2.cancer.gov/modules/PesticideHist.pdf.

We asked each interviewed subject for permission to collect a dust sample, and 95% agreed. The 1004 controls who consented were then screened for eligibility for dust sampling. Subjects were eligible if they had used their vacuum cleaner within the past year and had owned at least half of their carpets or rugs for 5 years or more. Those who had (521) gave their used vacuum bags to the interviewer, who placed them in insulated shipping boxes. The boxes were mailed overnight to Southwest Research Institute (SwRI) (San Antonio, TX, USA) and placed in freezers. Samples were collected between February 1999 and May 2001. We collected the samples from used vacuum cleaner bags rather than using a specially designed dust collection apparatus such as the High Volume Surface Sampler (HVS3), because a previous study found no clear difference in the quality of the pesticide concentration data between these two dust collection methods (Colt et al., 1998).

Laboratory Analysis

Portions of dust from each thawed vacuum cleaner bag were passed through a 100-mesh sieve to obtain the fine ($<150\,\mu$m) dust. The fine fraction was split into aliquots and placed in the freezer. The samples were grouped into batches of 13–15 for extraction and analysis by assigning at least four case and four control samples to each extraction batch. Laboratory personnel were blinded to case–control status. Extraction and analysis was performed on 513 samples; eight samples had insufficient dust or were not analyzed for other reasons.

For the neutral extractions, 2 g of sieved dust were spiked with *p*-terphenyl-d_{14}, octachloronaphthalene, and decachlorobiphenyl as surrogates. The spiked dust samples were Soxhlet extracted using 6% ether in hexanes for 16 h, and the extracts were cleaned through a Florisil column. Neutral-extractable analytes included 25 pesticides (18 insecticides, six herbicides, and ortho-phenylphenol), seven PAHs, and five PCB congeners. For the acid extractions, 2 g of sieved dust were spiked with 3,5-dichlorobenzoic acid and 2,3-dichlorophenoxyacetic acid (2,3-D) as surrogates. The spiked dust samples were sonication extracted with acidified acetonitrile:phosphate buffer. Base hydrolysis was performed to convert the ester forms of the analytes to the sodium salt form. The extract was cleaned by passage through a C18 solid-phase extraction cartridge and derivatized by silylation. Acid-extractable analytes included four herbicides and pentachlorophenol. The neutral and acid extracts were analyzed using gas chromatography/mass spectrometry (GC/MS) in selected ion monitoring (SIM) mode; analyte amounts were quantified using the internal standard method.

Lab spikes of 27 sieved dust samples showed that all target analytes were efficiently extracted, with recovery means ranging from 85% to 122% and recovery standard deviations from 9% to 34%, except for pentachlorophenol (74±20%), dichlorodiphenyltrichloroethane (DDT) (130±26%), *trans*-permethrin (135±31%), methoxychlor (140±27%), and carbaryl (144±54%). Reported levels in dust were not adjusted for spike recoveries. Analysis of lab splits of 27 dust samples showed close agreement between the regular sample and the lab split. The measurements for 89% of the 452 detection pairs agreed within 20%, and 98.5% agreed within 40%. Confirmation analyses performed by full-scan GC/MS on 55 samples generally verified the large results from the selected ion monitoring mode, indicating that the analytes had been properly identified despite the interfering organic compounds often present in the dust extracts.

Data Imputation and Statistical Analysis

The laboratory measurements for the 30 analytes contained various types of "missing data," primarily when the concentration was below the minimum level that could be detected by the GC/MS technique ("nondetect"). We also encountered missing data when interfering compounds that coeluted with the target analyte were present. To create a complete data set, we used an imputation procedure related to the "fill-in" approach described by Helsel (1990) and applied by Moschandreas et al. (2001), which assigns a value for each missing measurement by selecting a value from the assumed distribution using maximum likelihood parameter estimates (Appendix A). Overall, 34% of the values were imputed because the analyte concentration was below the usual detection limit of the GC/MS. An additional 8% of the values were imputed due to the presence of interfering compounds. Pesticides with relatively high occurrences of interferences were methoxychlor (32% of the samples), carbaryl (30%), and DDT (16%).

The pesticide concentrations were distributed log-normally; therefore, the dependent variable in all statistical models was the natural logarithm of the pesticide level. We used analysis of variance (ANOVA) (Stata Statistical Software: Release 6.0, College Station, TX, USA) to determine whether pesticide levels differed significantly by characteristics of the home and season the home was sampled. The independent variables were type of home (single family, townhouse/duplex/apartment, other), year the home was built (<1940, 1940–1959, 1960–1979, ⩾1980), season sampled, whether any oriental rugs were present, study center, sex, age (<45 years, 45–64 years, ⩾65 years), race (African American, Caucasian, other), and education (<12 years, 12–15 years, ⩾16 years).

We used multivariate linear regression models to estimate the effect of the types of pests treated on the pesticide levels in dust (Stata Statistical Software: Release 6.0, College Station, TX, USA). Only information pertaining to the subject's current home was used. In addition to the variables listed above, models for insecticides included five pest treatment variables: ever/never treated for crawling insects, flying insects, fleas/ticks, termites, and lawn/garden insects in the current home. Models for herbicides included two such variables: ever/never treated for lawn weeds and garden weeds. Type of home, age of the home, and the presence of oriental rugs were deleted from the models if they did not predict dust levels. The regressions for insecticides were based on 482 participants, who provided information for all of the insect treatment variables; the corresponding number for herbicides was 479. To investigate whether pesticide levels correlated with the total number of pesticide treatments, similar models were run in which the ever/never pest treatment variables were replaced with estimates of the total number of times each pest was treated.

To account for the additional random variation induced by imputed values, we repeated the imputation procedure five times for each analyte, fitted the regression model five times, and combined the results according to Rubin (1987), using the MIANALYZE procedure in the SAS analytic package (SAS Institute, Cary, NC, USA). Since this approach correctly accounts for the variance from the imputation, it results in wider confidence intervals than would be obtained from a single-imputation approach (i.e., one set of "fill-in" values).

To assess whether the associations in our data between pest type treated and pesticide levels in dust were consistent with known uses of the pesticides, we used data from the *National Home and Garden Pesticide Use Survey* (Whitmore et al., 1992) (the "EPA Survey") to identify pesticides that are typically used for each type of pest. This nationwide survey was conducted for the Environmental Protection Agency in 1990 and involved home visits and interviews with over 2000 households. Interviewers inventoried pesticide products present in the household, recorded the active ingredients listed on the label, and asked respondents to identify the pests on which the product had been used during the past year. The EPA Survey does not cover products used by professional applicators, but in our study most of the pesticides were applied by the subject or another household member; only termiticides were applied more frequently by professionals.

Table 1. Characteristics of study participants: 513 control subjects with dust samples.

	Detroit	Iowa	LA	Seattle	Total
Age (median)	65	66	61	62	64
Gender					
Male	44 (56.4%)	77 (52.4%)	73 (57.5%)	79 (49.1%)	273 (53.2%)
Female	34 (43.6%)	70 (47.6%)	54 (42.5%)	82 (50.9%)	240 (46.8%)
Race					
Caucasian	59 (75.6%)	146 (99.3%)	89 (70.1%)	149 (92.6%)	443 (86.4%)
African-American	18 (23.1%)	0 (0.0%)	26 (20.5%)	2 (1.2%)	46 (9.0%)
Other	1 (1.3%)	1 (0.7%)	12 (9.5%)	10 (6.2%)	24 (4.7%)
Education					
<12 years	9 (11.5%)	14 (9.5%)	17 (13.4%)	8 (5.0%)	48 (9.4%)
12–15 years	46 (59.0%)	110 (74.8%)	72 (56.7%)	85 (52.8%)	313 (61.0%)
$\geqslant 16$ years	23 (29.5%)	23 (15.7%)	38 (29.9%)	68 (42.2%)	152 (29.6%)
Years in current home (Median)	19	23	18	15	19
Type of home					
Single Family	64 (82.1%)	132 (89.9%)	93 (73.2%)	125 (77.6%)	414 (80.7%)
Duplex or townhouse	5 (6.4%)	5 (3.4%)	11 (8.7%)	15 (9.3%)	36 (7.0%)
Apartment	6 (7.7%)	5 (3.4%)	18 (14.2%)	12 (7.5%)	41 (8.0%)
Other	2 (2.6%)	5 (3.4%)	3 (2.4%)	8 (5.0%)	18 (3.5%)
Unknown	1 (1.3%)	0 (0.0%)	2 (1.6%)	1 (0.6%)	4 (0.8%)
Season sampled					
Winter	18 (23.1%)	19 (12.9%)	28 (22.1%)	28 (17.4%)	93 (18.1%)
Spring	32 (41.0%)	37 (25.2%)	33 (26.0%)	42 (26.1%)	144 (28.1%)
Summer	23 (29.5%)	54 (36.7%)	35 (27.6%)	43 (26.7%)	155 (30.2%)
Fall	5 (6.4%)	37 (25.2%)	31 (24.4%)	48 (29.8%)	121 (23.6%)
Total	78 (15.2%)	147 (28.7%)	127 (24.8%)	161 (31.4%)	513 (100%)

Results

The demographics of the 513 control subjects with analyzed dust samples (Table 1) reflect those of the larger NHL study population, disproportionately elderly, predominantly Caucasian, and typically living in single-family homes at the time of the dust collection. The proportion of African Americans was higher in Detroit and Los Angeles than the other sites, and the sites differed somewhat in terms of the season of dust collection. Subjects had lived in their current home for a median of 19 years.

Most subjects (94.3%) reported that they had treated in or around their current home for insects. Only 22 people (4.3%) never used insecticides. The remaining seven (1.4%) did not provide enough information to determine whether insecticides had been used. A total of 250 subjects (48.7%) treated their lawns or gardens for weeds, 234 (45.6%) never did, and 29 (5.6%) provided insufficient information.

Pest treatment practices varied among the four sites (Table 2). For crawling insects, for fleas and ticks, and for termites, Los Angeles residents reported the highest prevalence of treatment. Indeed, termites were treated in a substantially higher proportion of Los Angeles homes (49%) than the other sites (1% to 11%). Seattle had the highest percent of homes treated for lawn insects (30%) and garden insects (57%), while the percent treated for lawn weeds and garden weeds was highest in Iowa (66% and 8%, respectively).

The pesticides detected most frequently in the dust samples were ortho-phenylphenol (a fungicide and component of some disinfectant products); pentachlorophenol (a wood preservative no longer used in residential pesticides); 2,4-dichlorophenoxyacetic acid (2,4-D) (an herbicide commonly used on residential lawns and agricultural crops); the household insecticides propoxur, chlorpyrifos, and *cis*- and *trans*-permethrin; and the banned insecticide DDT (Table 3). In all, 15 pesticides were detected in fewer than 10% of the dust samples and are not considered further (acetochlor, alachlor, aldrin, atrazine, bendiocarb, cyanazine, dacthal, dicofol, dieldrin, heptachlor, lindane, malathion, MCPA, metolachlor, and 2,4,5-trichlorophenoxyacetic acid (2,4,5-T)). The pesticides with the highest concentrations (based on the geometric mean) were *trans*-, *cis*-permethrin, 2,4-D, and pentachlorphenol.

The concentrations of some pesticides were correlated. This occurred for three pairs of chemicals that coexist in pesticide formulations (*cis*- and *trans*-permethrin, Pearson's correlation coefficient = 0.93; α- and γ-chlordane, 0.73; 2,4-D and dicamba, 0.36). DDT and its degradation product dichlorodiphenyldichloroethylene (DDE) were also highly

Table 2. Self-reported pesticide use, by site: percent of participants treating for each pest type.

	Detroit (%)	Iowa (%)	LA (%)	Seattle (%)	Total (%)
Crawling insects	70	81	86	45	69
Flying insects	40	59	43	41	47
Fleas or ticks	32	50	62	55	52
Termites	1	11	49	6	18
Lawn insects	27	19	23	30	24
Garden insects	18	42	37	57	42
Lawn weeds	45	66	19	59	49
Garden weeds	6	8	2	7	6

Table 3. Pesticide levels in carpet dust: percent with any detected, arithmetic mean, geometric mean, and geometric standard deviation for all sites combined.

	UDL[a] (ng/g)	% >UDL[a]	Arithmetic mean[b] (ng/g)	Geometric mean[b] (ng/g)	Geometric standard deviation[b]
Herbicides					
2,4-D	84.3	78%	2422	419	6.9
Dicamba	85.3	19%	80[c]	19[c]	5.7
Organochlorine insecticides					
α-Chlordane	20.8	38%	85	11[c]	7.9
γ-Chlordane	20.8	48%	126	19[c]	6.7
4,4′-DDE	20.8	46%	43	18[c]	3.6
4,4′-DDT	20.8	70%	343	72	6.5
Methoxychlor	62.4	42%	497	40[c]	8.9
Pentachlorophenol	83.5	87%	1021	345	3.8
Carbamate insecticides					
Carbaryl	125	35%	1415	62[c]	11.7
Propoxur	21.2	77%	436	68	5.7
Organophosphate insecticides					
Chlorpyrifos	41.7	68%	888	113	7.8
Diazinon	46.3	39%	620	25[c]	9.0
Pyrethroid insecticides					
cis-Permethrin	76.2	72%	4339	337	13.2
trans-Permethrin	123	74%	7499	517	17.1
Other					
o-Phenylphenol	20.8	99%	442	248	2.6

[a]UDL = Usual detection limit.
[b]Based on measured and imputed values.
[c]Level is below the usual detection limit.

correlated (0.64). Correlations of 0.30 or higher were found among some of the commonly used household insecticides: chlorpyrifos and diazinon (0.34), chlorpyrifos and propoxur (0.30), and both *cis*- and *trans*-permethrin (0.32 for both). Correlations also exceeded 0.30 for some of the organochlorine insecticides, including γ-chlordane and DDT (0.31), γ-chlordane and DDE (0.29), and DDT and methoxychlor (0.35). Concentrations of *o*-phenylphenol and pentachlorophenol were also correlated (0.35).

The currently used insecticides were detected less frequently in the homes of 22 people who reported no insecticide use than in homes of users, but they were still detected in a substantial fraction of the nonuser dust samples (propoxur 73%, permethrin 59%, chlorpyrifos 55%, carbaryl 27%, diazinon14%) (data not shown). Similarly, 2,4-D and dicamba were detected frequently in the homes of the 234 people who did not treat for weeds (69% and 13%, respectively).

Consistent with the relatively high proportion of Los Angeles residents treating their homes for household insects and termites, dust sampled in Los Angeles had the highest mean levels of many household insecticides (propoxur, chlorpyrifos, diazinon, and *cis*- and *trans*-permethrin) and the banned termiticides α- and γ-chlordane (Table 4). Iowa, the site in which lawn/garden weed treatment was the most prevalent, had the highest levels of the herbicides 2,4-D and dicamba. Although Seattle had the highest proportion of residents treating for lawn

Table 4. Geometric mean of pesticide concentration in carpet dust, by site (ng/g dust).

	Detroit	Iowa	LA	Seattle
Herbicides				
2,4-D	606	1512	87	374
Dicamba	17	37	12	17
Organochlorine insecticides				
α-Chlordane	9	9	29	6
γ-Chlordane	15	16	50	11
4,4′-DDE	25	21	22	11
4,4′-DDT	81	111	75	44
Methoxychlor	36	58	32	36
Pentachlorophenol	332	453	193	431
Carbamate insecticides				
Carbaryl	38	83	73	53
Propoxur	47	43	203	52
Organophosphate insecticides				
Chlorpyrifos	128	83	255	73
Diazinon	33	13	66	20
Pyrethroid insecticides				
cis-Permethrin	111	163	1123	434
trans-Permethrin	169	228	1948	661
Other				
o-Phenylphenol	329	218	213	274

and garden insects, the lawn/garden insecticide levels were higher in other sites (e.g., carbaryl and methoxychlor were highest in Iowa); this could reflect degradation of the insecticides in Seattle's relatively moist climate.

ANOVA incorporating age and type of home, season, site, and demographic factors showed that age of the home was a significant factor for the organochlorines (chlordane, DDT, DDE, methoxychlor, and pentachlorophenol) and carbamates (propoxur and carbaryl); all were higher in older homes (data not shown). There were statistically significant differences by type of home only for carbaryl, pentachlorophenol, and 2,4-D, which were highest in single-family homes. Seasonal variation marked propoxur (highest in winter) and DDE (highest in fall). Higher levels of α-chlordane, DDT, and DDE were found in homes with oriental rugs.

Significant positive associations between treatment for crawling insects and dust levels of chlorpyrifos and propoxur were found in multivariate linear regression models (Table 5). Chlorpyrifos was 70% higher in dust sampled from the treated homes, and levels were significantly associated with the total number of crawling insect treatments. Propoxur was 40% higher in the treated homes. These findings are consistent with the EPA Survey, which found that these two active ingredients were among those used most frequently to treat cockroaches, ants, or spiders in 1990 (Table 6). Diazinon, also used frequently for crawling insects in the EPA survey, was 70% higher among the crawling insect treaters in our study ($P = 0.06$). Other active ingredients commonly used for crawling insects (e.g., pyrethrins) were not measured.

According to the EPA Survey, flying insects (bees, mosquitoes, and flies) were treated most frequently with pyrethrins, pyrethroids (allethrin, tetramethrin, resmethrin, and sumithrin), propoxur, and various repellents. Propoxur was not elevated in dust sampled from homes that had been treated for flying insects, and the other pesticides were not measured in the dust samples. Carbaryl was significantly lower in homes treated for flying insects, possibly by chance.

If a respondent reported treating the home or a pet for fleas/ticks, dust levels of carbaryl and permethrin were higher, and the levels increased as the number of treatments increased. According to the EPA Survey, carbaryl (80% higher in dust sampled from treated homes) was widely used to treat fleas/ticks in 1990. Permethrin was also used for flea/tick treatment, although less frequently than the other chemicals listed in Table 6; its levels were more than three times higher in dust sampled from treated homes. Chlorpyrifos, a common flea/tick pesticide in the EPA Survey, was 40% higher in the treated homes ($P = 0.08$). There was no association for diazinon.

Concentrations of α- and γ-chlordane were significantly higher (three- to four-fold) in homes treated for termites. Chlordane was a commonly used termiticide before being banned in the late 1980s. Most subjects who treated for termites in our study used professional exterminators. Chlorpyrifos, the active ingredient used most often by exterminators to treat termites in the early 1990s (Kline,

Table 5. Parameter estimates and 95% confidence intervals (exponentiated) from multiple linear regressions of the natural logarithm of pesticide level in dust on ever/never treated for pest type in current home[a].

	Crawling insects	Flying insects	Fleas/ ticks	Termites	Lawn/garden insects	Lawn weeds	Garden weeds	Intercept	R^2
Herbicides									
2,4-D[b]	—	—	—	—	—	1.7 (1.3, 2.3)*	0.7 (0.4, 1.3)	369	0.39
Dicamba	—	—	—	—	—	1.1 (0.6, 1.9)	1.5 (0.7, 3.2)	6	0.09
Organochlorine insecticides									
α-Chlordane[c,d]	0.9 (0.6, 1.5)	0.9 (0.6, 1.3)	0.8 (0.5, 1.3)	3.3 (1.9, 5.0)*	1.2 (0.7, 2.1)	—	—	7	0.23
γ-Chlordane[c,d]	0.8 (0.6, 1.3)	1.0 (0.7, 1.4)	0.8 (0.6, 1.2)	3.7 (2.2, 5.9)*	1.1 (0.8, 1.6)	—	—	15	0.29
4,4′-DDT[c,d]	0.8 (0.6, 1.2)	1.2 (0.9, 1.7)	1.0 (0.7, 1.3)	1.0 (0.6, 1.6)	1.4 (1.0, 1.9)	—	—	127	0.32
4,4′-DDE[c,d]	1.0 (0.7, 1.3)	1.2 (0.9, 1.6)	0.9 (0.6, 1.3)	1.0 (0.8, 1.5)	1.2 (0.8, 1.6)	—	—	51	0.27
Methoxychlor[b,d]	1.3 (0.7, 2.2)	0.9 (0.6, 1.3)	1.0 (0.6, 1.5)	1.1 (0.6, 2.1)	1.8 (1.1, 2.8)*	—	—	22	0.10
Pentachlorophenol[b,d]	1.0 (0.8, 1.3)	1.1 (0.9, 1.3)	0.9 (0.7, 1.1)	1.1 (0.8, 1.5)	1.0 (0.8, 1.2)	—	—	463	0.23
Carbamate insecticides									
Carbaryl[b,d]	1.0 (0.6, 1.7)	0.6 (0.4, 1.0)*	1.8 (1.0, 3.2)*	1.3 (0.7, 2.6)	1.8 (1.0, 3.3)*	—	—	15	0.15
Propoxur[d]	1.4 (1.0, 2.0)*	1.0 (0.7, 1.3)	1.3 (0.9, 1.8)	1.1 (0.7, 1.8)	0.7 (0.5, 1.0)	—	—	54	0.22
Organophophate insecticides									
Chlorpyrifos	1.7 (1.1, 2.6)*	0.7 (0.5, 1.1)	1.4 (1.0, 2.1)	1.7 (1.0, 2.9)	0.8 (0.5, 1.2)	—	—	252	0.14
Diazinon	1.7 (1.0, 2.8)	1.0 (0.7, 1.5)	0.9 (0.6, 1.4)	1.4 (0.7, 2.8)	1.3 (0.9, 1.9)	—	—	85	0.11
Pyrethroid insecticides									
cis-Permethrin[c]	1.2 (0.7, 2.0)	1.1 (0.7, 1.8)	3.5 (2.2, 5.4)*	1.2 (0.6, 2.3)	1.0 (0.6, 1.6)	—	—	172	0.21
trans-Permethrin[c]	1.2 (0.7, 2.2)	1.2 (0.7, 2.0)	3.6 (2.1, 6.0)*	1.0 (0.5, 2.1)	1.0 (0.6, 1.8)	—	—	393	0.20
Other									
o-Phenylphenol	0.8 (0.7, 1.0)	1.0 (0.9, 1.2)	1.0 (0.9, 1.2)	1.3 (1.0, 1.7)*	0.9 (0.8, 1.1)	—	—	337	0.09

* = Significant at the 0.05 α level.
— = Not included in the model.
[a] Models for herbicides included ever/never use of lawn herbicides, ever/never use of garden herbicides. Models for all other pesticides included ever/never use of insecticides for: crawling insects, flying insects, fleas/ticks, termites, lawn/garden insects. All models adjusted for gender, age, race, education, study center, and season.
[b] Model included the type of home.
[c] Model included the presence of oriental rugs in the home.
[d] Model included the year the home was built.

1993), was 70% higher in termite-treated homes ($P = 0.06$). Ortho-phenylphenol was significantly elevated in homes treated for termites; this chemical is not used as a termiticide, and the association is likely by chance.

History of lawn/garden insect use, and number of treatments, correlated with dust levels of carbaryl, malathion, and methoxychlor. Carbaryl and methoxychlor were 80% higher, and malathion was 20% higher (data not shown), in dust sampled from treated homes. Carbaryl and malathion were widely used for lawn/garden insect treatment in the EPA Survey; methoxychlor played a minor role. Diazinon, another common lawn/garden insecticide, was 30% higher in treated homes, but the association was not significant ($P = 0.23$).

In the EPA survey, the four active ingredients found most frequently in lawn and garden herbicide formulations were 2,4-D, dicamba, glyphosate, and mecoprop (MCPP). Only the first two were measured in our study. 2,4-D was significantly associated with ever/never use of lawn herbicides and with the total number of lawn herbicide treatments; dicamba was elevated, but not significantly, among garden herbicide users ($P = 0.29$).

The coefficients of determination (R^2) for the regression models ranged from 0.09 to 0.39, indicating that the factors included in the models explain only a fraction of the variability in pesticide levels in dust.

Discussion

Questionnaires and carpet dust samples provide independent measures of exposure to pesticides in the home environment. Different aspects of pesticide exposure are measured by the two techniques, but many of the pesticide exposure indicators for the current home ought to agree if both techniques work well. This study suggests that carefully designed questionnaire assessment of residential pesticide use and measured pesticide residues in carpet dust tend to agree quite well. This appears to hold for differences among geographic areas and for differences among individuals. The consistency lends credibility to both methods for assessing residential exposure to pesticides.

Most associations between the type of pest treated and pesticide levels in dust were consistent with information from

Table 6. EPA Survey: pesticides applied by homeowners to treat specific types of pests in 1990 (top five pesticides for each category)[a,b]

Crawling insects[c]	**Flying insects**[d]
*Propoxur***	Allethrin
Pyrethrins	*Propoxur*
*Chlorpyrifos***	Pyrethrins
Allethrin	Tetramethrin
*Diazinon**	Resmethrin
Fleas and ticks	**Lawn and garden insects**[e]
Pyrethrins	*Carbaryl***
*Carbaryl***	*Diazinon*
*Chlorpyrifos**	Pyrethrins
Tetrachlorvinphos	*Malathion***
Diazinon	Resmethrin
Lawn and garden weeds[f]	
Glyphosate	
*2,4-D***	
MCPP	
Dicamba	
Sodium acifluorfen	

Compounds were analyzed in the dust samples.
[a]Calculated from information presented in Table F of Whitmore et al. (1992).
[b]We have excluded repellents (DEET, Repellent R-11), synergists (piperonyl butoxide and MGK-264), and aliphatic and aromatic petroleum hydrocarbons.
[c]Cockroaches, ants, spiders.
[d]Bees, mosquitoes, flies.
[e]Soil insects, plant chewing insects, plant sucking insects, slugs.
[f]Brush, grass, broadleaf weeds.
** = Significant at the 0.05 level in multiple linear regression analysis.
* = Significant at the 0.10 level in multiple linear regression analysis.

a national survey of pesticide use in 1990. Positive, statistically significant associations were found between ever having treated for crawling insects and dust levels of propoxur and chlorpyrifos; fleas/ticks and carbaryl and permethrin; termites and chlordane; lawn/garden insects and carbaryl, methoxychlor, and malathion; and lawn/garden weeds and 2,4-D. For many of these, the pesticide levels also correlated with the total number of pesticide applications. In addition, there were positive but nonsignificant associations between crawling insects and diazinon, fleas/ticks and chlorpyrifos, and termites and chlorpyrifos. Two associations were unexpected and were likely due to chance: termites and *o*-phenylphenol, and a negative association between flying insects and carbaryl.

We did not observe expected associations between propoxur and flying insects, diazinon and flea/tick treatment and lawn/garden insect treatment, and dicamba and lawn/garden weed treatment. There are several possible reasons for this. Personal recall about the types of pests treated is imperfect. The EPA Survey data used to identify "expected" associations between pest treatment and specific active ingredients in dust is fixed in time (1990) and does not account for changes over time in the pesticides available for home and garden use. The EPA survey also does not account for regional differences in pesticides sold for residential use. Carpets accumulate pesticides from sources unrelated to personal pesticide use, such as drift-in from use in neighboring homes or farms, take-home contamination from occupational use, and pesticide treatment of rugs by manufacturers. In addition, for some participants there were discrepancies in time and/or place between the questionnaire data and dust sampling data. This could arise if the carpets sampled had been present in a previous home, if the carpets were replaced after the pest treatment occurred, or if the carpets were in place while a previous resident lived in the home. The fact that we detected many pesticides in dust from study participants who claimed not to have used them illustrates some of these phenomena. Finally, we do not fully understand the physical and chemical mechanisms that affect entry of pesticides into carpets and their persistence in carpets over time.

The chemical found at the highest concentration was *trans*-permethrin. Permethrins are widely used by consumers and pest control operators and are frequently added to carpets at the time of manufacture. Although permethrin was not among the chemicals most commonly used for flea/tick treatment in the EPA Survey, it had a strong (3.5-fold) and significant association with treatment for fleas/ticks in our study. This may reflect its presence in many types of flea bombs. 2,4-D was also found in high concentrations in the dust samples, particularly in Iowa. This is likely due to both the high prevalence of lawn weed treatment and the agricultural use of 2,4-D in that state.

There was a strong and significant association between termite treatment and chlordane levels. Although chlordane was banned in 1988, 330 (64%) of the study participants moved into their homes prior to that date. The magnitude of the association (3.5-fold) is evidence of chlordane's persistence in carpet dust, as is the presence of DDT in 70% of the dust samples despite its ban in the early 1970s. Elevated DDT levels in homes with oriental rugs is consistent with an earlier study in which DDT was found at over 100,000 ng/g in a 90-year-old oriental rug (Camann, 1994).

Sexton et al. (2003) conducted a conceptually similar study in Minnesota. Questionnaire data on pesticide use were compared to concentrations/loadings of pesticides (chlorpyrifos, diazinon, malathion, and atrazine) in environmental samples (air, soil, and surface wipe samples) from 102 homes. Unlike our study, Sexton found no significant associations between the questionnaire items and the detectability or levels of the pesticides. There are several possible reasons why the study findings differ. First, as the authors point out, the Minnesota study questions were generalized; combining different types of pests into one question (e.g., "fleas, roaches, ants, and other insects") likely limited the predictive capability of the questionnaire. Sexton further explains that the questions targeted the past several months of pesticide use, while the time frame reflected by the samples was days (somewhat longer for surface wipes). Our study had a longer-term focus for both exposure assessment techniques; the questionnaire covered all of the years in the current home, and the carpet dust sampling is believed to reflect pesticide use over the lifetime of the carpet. Finally, many more pesticides were measured in our study, expanding our ability to identify relevant associations.

A limitation of our study is that the participants are not representative of the general population of the four study areas. Control subjects in the NHL study came from a stratified random sample of the population of the four areas, with older people and African Americans purposely over-represented. Over half of the controls selected for the study did not participate. We collected dust samples only from study participants who owned most of their carpets for at least 5 years. Another limitation is that we did not analyze the dust samples for some of the pesticides commonly used in homes and gardens, limiting our ability to identify some associations between pest treatment practices and pesticide levels in dust. For example, we found no association between treatment for flying insects and pesticide levels in dust; however, we analyzed the dust for only one of the top five insecticides commonly found in flying insect products. It is also important to point out that this analysis was based on control responses only. In the future, we will analyze the consistency between questionnaire data and pesticide residues for cases, and examine whether there is evidence of bias in the responses of cases compared to controls.

In sum, this study suggests that home-specific questionnaire data on the types of pests treated are generally consistent with pesticide levels measured in the dust. This lends support to the validity of both exposure assessment approaches but does not eliminate the need for either. For example, interviewing is the only way to assess earlier pesticide exposures, before current carpets were in place. In this study, we attempted to improve interview data quality by querying home by home and, within each home, pest by pest. This approach was well tolerated and elicited detailed, plausible responses, but we do not know if another approach would be superior. We believe that respondents would find it very difficult to name specific products. In that respect, dust sampling offers a valuable supplement to questionnaires. Dust sampling identifies specific active ingredients to which a person may have been exposed, either through personal use of a pesticide or from drift-in or track-in from outside sources. It is also unaffected by recall bias, which could compromise case–control interview studies. Both techniques involve substantial expense and respondent burden, but in epidemiologic studies that require high accuracy, the combined techniques appear promising.

References

Bradman M.A., Harnly M.E., Draper W., Seidel S., Teran S., Wakeham D., and Neutra R. Pesticide exposures to children from California's Central Valley: results of a pilot study. *J Expo Anal Env Epidemol* 1997: 7(2): 217–234.

Camann D.E., Hsu, J.P., Fortmann R.C., Roberts J.W., and Lewis R.G. Association between measured pesticide levels in indoor air and carpet dust in the home. Presented May, 1991. In: Measurement of Toxic and Related Air Pollutants, Publication VIP-21, Air and Waste Management Association, Durham, NC. 1991: pp. 1113–1127.

Camann D.E. Investigating the effects of pesticide exposure in the home. Technol Today, June 1994: 2–7.

Colt J.S., Zahm S.H., Camann D.E., and Hartge P. Comparison of pesticides and other compounds in carpet dust samples collected from used vacuum cleaner bags and from the high volume surface sampler. *Environ Health Perspect* 1998: 106(11): 721–724.

Efron B. Missing data, imputation, and the bootstrap. *J Am Stat Assoc* 1994: 89: 364–475.

Fortune D.R., Blanchard F.T., Ellenson W.D., and Lewis R.G. Analysis of aged in-home carpeting to determine the distribution of pesticide residues between dust, carpet, and pad compartments. EPA 600/R-00/030. 2000. http://www.epa.gov/ORD/WebPubs/carpet/600r00030.pdf.

Helsel D.R. Less than obvious — statistical treatment of data below the detection limit. *Environ Sci Technol* 1990: 24(12): 1766–1774.

Kline & Company, Inc. *Professional Markets for Pesticides and Fertilizers. Year Two: 1993. c.1994*. Kline & Company, Inc., Little Falls, NJ, 1994.

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: 37–46.

Lewis R.G., Fortune C.R., Willis R.D., Camann D.E., and Antley J.T. Distribution of pesticides and polycyclic aromatic hydrocarbons in house dust as a function of particle size. *Environ Health Perspec* 1999: 107(9): 721–726.

Lewis R.G., Fortune C.R., Blanchard F.T., and Camann D.E. Movement and deposition of two organophosphorous pesticides within a residence after interior and exterior applications. *J Air Waste Manage Assoc* 2001: 51: 339–351.

Lioy P.J., Edwards R.D., Freeman N., Gurunathan S., Pellizzari E., Adgate J.L., Quackenboss J., and Sexton K. House dust levels of selected insecticides and a herbicide measured by the EL and LWW samplers and comparisons to hand rinses and urine metabolites. *J Expo Anal Environ Epidemiol* 2000: 10: 327–340.

Moschandreas D.J., Karuchit S., Kim Y., Ari H., Lebowitz M.D., O'Rourke M.K., Gordon S., and Robertson G. On predicting multi-route and multimedia residential exposure to chlorpyrifos and diazinon. *J Expo Anal Environ Epidemiol* 2001: 11(1): 56–65.

Nigg N.H., Beier R.C., Carter O., Chaisson C., Franklin C., Lavy T., Lewis R.G., Lombardo P., McCarthy J.F., and Maddy K.T. Exposure to pesticides. In: Mehlman M.A. (Ed.). *Advances in Modern Environmental Toxicology, Vol XVIII: The Effects of Pesticides on Human Health* (Baker SR, Wilkinson CF, Eds.). Princeton, NJ: Princeton Scientific, 1990: pp. 35–130.

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

Roberts J.W., Budd W.T., Camann D.E., Fortmann R.D., Lewis R.G., Ruby M.G., and Spittler T.M. Human exposure to pollutants

in the floor dust of homes and offices. *J Expo Anal Environ Epidemiol* 1992: 2(Suppl. 1): 127–146.
Roinestad K.S., Louis J.B., and Rosen J.D. Determination of pesticides in indoor air and dust. *J Assoc Off Anal Chem Intl* 1993: 76: 1121–1126.
Rubin D.B. *Multiple Imputation for Nonresponse in Surveys*. John Wiley & Sons, New York, 1987.
Sexton K., Adgate J.L., Elberly L.E., Clayton C.A., Whitmore R.W., Pellizzari E.D., Lioy P.J., and Quackenboss J.J. Predicting children's short-term exposure to pesticides: results of a questionnaire screening approach. *Environ Health Perspect* 2003: 111(1): 123–128.
Simcox J.J., Fenske R.A., Wolz S.A., Lee I.C., and Kalman D. Pesticides in household dust and soil: exposure pathways for children of agricultural families. *Environ Health Perspect* 1995: 103: 1126–1134.
Starr H.G., Aldrich F.D., McDougall W.D., and Mounce L.M. Contribution of household dust to the human exposure to pesticides. *Pesticides Monit J* 1974: 8: 209–212.
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 U.S. cities. *Arch Environ Contam Toxicol* 1994: 26: 47–59.
Whitmore R.W., Kelly J.E., and Reading P.L. National home and garden use survey, final report, Volume I: Executive summary, results, and recommendations. Prepared by Research Triangle Institute for the US. Environmental Protection Agency under Contract No. 68-WO-0032. Research Triangle Park, NC, 1992.
Zahm S.H., Ward M.H., and Blair A. Pesticides and cancer. *Occup Med: State of the Art Rev* 1997: 12(2): 269–289.

Appendix A. Imputation of missing data

Imputation procedures were used to fill in "missing" pesticide concentrations, which occurred when the analyte concentration was below the analytic detection limit of the GC/MS, or the presence of interfering compounds in the dust sample made it difficult for the GC/MS analyst to determine whether the target analyte was present and, if so, at what level. In this study, detection limits for individual analytes varied across samples. After about half of the samples had been analyzed, we began monitoring additional ions for some neutral analytes to clarify their identification at low levels; this resulted in raised detection limits for 14 pesticides. Deviations in detection limits also occurred when less than 2 g of dust was available.

When an interfering compound was present in the dust sample, the GC/MS analyst made a judgment as to whether the target compound was also present. If he/she judged that an interfering compound and the target analyte were both present, the result was reported as an "elevated detection" at a concentration equal to the entire peak of the coeluting compounds. If the analyst had insufficient evidence that the target analyte was present, the result was reported as a nondetect with a detection limit equal to the entire peak. In both cases, the result was flagged to indicate that the concentration or detection limit had been raised to account for the uncertainty posed by the presence of an interfering compound.

To account for the missing data, we first set a lower and upper bound for each missing data point based on information reported by the GC/MS analyst. If the result was a nondetect and no interferences were present, we set the lower bound at zero and the upper bound at the detection limit reported for that sample. If the result was a nondetect with a raised detection limit due an interfering compound, we bounded the concentrations between zero and 20% of the raised detection limit. If the result was an "elevated detection" because of an interference, we set the bounds at 20% and 90% of the reported concentration.

We then assigned a value for each missing measurement by selecting a value from the assumed distribution using maximum likelihood parameter estimates. We created a likelihood function as the product of the normal density function at the log-transformed value for the known measurements and the cumulative normal distribution function at the log-transformed bounds for the missing measurements. We then determined the maximum likelihood estimates of μ and σ, denoted $\hat{\mu}$ and $\hat{\sigma}$, respectively. We imputed a value for each missing measurement by randomly sampling from a log-normal distribution with parameters $\hat{\mu}$ and $\hat{\sigma}$, conditional on being bounded by the values discussed above. To account for uncertainty in the estimation of parameters, we did not use $\hat{\mu}$ and $\hat{\sigma}$ for the imputation, but instead used $\hat{\mu}^*$ and $\hat{\sigma}^*$, which were selected from a bootstrap sample of 500 realizations of $\hat{\mu}$ and $\hat{\sigma}$ (Efron, 1994). The bootstrap sample was generated by estimating $\hat{\mu}$ and $\hat{\sigma}$ from 500 data sets created by repeatedly sampling with replacement from the original data set to create 500 data sets of equal size. There is an additional source of variation which was not taken into account in this approach, since we implicitly assume that the bounds were fixed. For the cases of interfering compounds, this assumption cannot be justified. However, we believe that this additional random variation is not likely to affect inference.