Comparison of Pesticides and Other Compounds in Carpet Dust Samples Collected from Used Vacuum Cleaner Bags and from a High-Volume Surface Sampler

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Epidemiologic studies of the association between residential pesticide use and cancer risk require an assessment of past pesticide exposures. Pesticide levels in carpet dust are believed to reflect long-term pesticide use. Recent epidemiologic studies have found collection of dust samples using the high-volume surface sampler (HVS3) to be expensive and cumbersome. We compared the levels of pesticides and other compounds in dust obtained from subjects' personal used vacuum cleaner bags to that collected by the HVS3 to see if this simpler method could replace the HVS3 in epidemiologic research. We visited the homes of 15 subjects, took the used bags from their vacuums, and collected carpet dust samples with the HVS3. The samples were analyzed for 42 target compounds: 26 pesticides, 10 polycyclic aromatic hydrocarbons (PAHs), and six polychlorinated biphenyl (PCB) congeners using GC/MS in selected ion monitoring mode. The two methods agreed in detecting the presence of the target compounds between 80% and 100% of the time. Neither sampling method was consistently more sensitive. The median target compound concentrations were similar, and a paired *t*-test showed no significant differences. For many compounds, the concentrations of compounds in the HVS3 samples were higher than those in the used bag samples at the upper end of the concentration ranges. However, the Spearman rank correlation coefficients were 0.85 or higher for most compounds, indicating that homes would be ranked similarly using both methods. Overall, there appears to be no clear difference in the quality of the pesticide, PAH, or PCB concentration data for the two dust collection methods. *Key words*: carpet dust, high-volume surface sampler, pesticides, polychlorinated biphenyls, polycyclic aromatic hydrocarbons. *Environ Health Perspect* 106:721–724 (1998). [Online 14 October 1998]
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Exposure to pesticides has been linked to cancer in farmers and other occupational groups (*1*). The general population is exposed to pesticides as well, principally from indoor use and tracking or drifting indoors of pesticides applied outdoors (*1,2*). Recently, epidemiologic studies have begun to examine whether residential pesticide exposures can increase the risk of cancer (*1*).

Because of the potentially long latency period between pesticide exposure and cancer diagnosis, the relevant exposure may have occurred decades before diagnosis. Assessing past pesticide exposures in residential situations poses several challenges. The value of questionnaires is limited by the difficulty most respondents have in identifying pesticide products they have used. Air monitoring offers little promise because air concentrations of most pesticides dissipate quickly. On the other hand, pesticides entering the home are known to persist in carpet dust for years, where they are protected from degradation by sunlight, moisture, temperature extremes, and most microbial action (*3*). Thus, concentrations of pesticides in carpet dust may well reflect a person's pattern of pesticide use over the lifetime of the carpet.

In several recent studies, investigators have collected carpet dust samples for pesticides analysis using a modified commercial vacuum cleaner called the high-volume surface sampler (HVS3) (*4*). The dust sample is sent to a laboratory, where it is sieved to remove fibers and other large particles, solvent-extracted, cleaned, and analyzed using gas chromatography/mass spectrometry (GC/MS) (*5*). This procedure provides estimates of pesticide concentrations in the dust. By design, the HVS3 achieves a constant removal efficiency of surface dust across different types of carpets and can therefore be used to estimate a standardized dust "loading," the relative amount of surface dust present per unit surface area of carpet (*4,6*).

Recent experience with the HVS3 has shown that it can be expensive ($3,000 for the vacuum, $51 per teflon catch bottle, plus miscellaneous supplies), labor intensive (>1 hr for preparation and collection of samples, 0.5 hr for cleaning equipment between samples), and difficult to use (Carol Haines, personal communication, Westat, Inc., 1998). We postulated that if pesticide concentration is the main parameter of interest in a study (i.e., if dust loading is not critical), dust samples could be obtained from subjects' used vacuum cleaner bags. However, we questioned whether the quality of the pesticide concentration data would be compromised by variations in subjects' vacuuming equipment and practices and by the repeated passage of air through the dust bag during multiple uses of the vacuum. We therefore conducted a pilot study to determine whether the "used bag" dust sampling method would provide pesticide concentration data comparable to that of the HVS3.

Methods

To recruit subjects for the study, an announcement was sent to approximately 150 employees of the National Cancer Institute, all living in the Washington, DC, metropolitan area. The 21 people who responded to this announcement were asked to fill out a short questionnaire eliciting information about duration of residence in the current home, use of insecticides or herbicides, employment of a professional exterminator or gardener, and pet ownership. From these respondents, 15 were selected to participate in the study. These subjects represented a range of possible pesticide exposures, based on the questionnaire responses, and all had lived in their current residence for at least 5 years.

We visited the subjects' homes in the summer of 1997, collected a carpet dust sample with the HVS3, and took the used bag from their vacuum cleaners. We gave each subject $5.00 to purchase a new vacuum cleaner bag.

When the HVS3 is used to estimate dust loadings, a sample is taken from one or more fixed, carefully measured area(s) of carpet in accordance with American Society for Testing Materials (ASTM) Standard Practice D 5483-93 (*4*). Because loadings were unimportant in the pilot study, we collected the HVS3 sample from all rooms that the subject typically cleaned with their vacuum. The proportion of each room vacuumed was based on the frequency with which the subject typically vacuumed that room, so that the HVS3 and used bag sample were composed

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of similar proportions of dust from each room. Procedures for handling the collected HVS3 samples were in accordance with ASTM Standard Practice D 5483-93 (*4*).

For the used bag collection, we placed the subject's used vacuum cleaner bag in a sealable plastic bag. The sealed plastic bag and the HVS3 catch bottle containing the dust sample were shipped via overnight mail in styrofoam containers with ice packs to Southwest Research Institute (SRI). The ice packs prevented overheating of the samples during shipping, which could have led to volatilization of the pesticides from the dust.

At SRI, the vacuum cleaner bag was split open and its entire contents were processed through a hand-held 100 mesh sieve in approximately 50-ml aliquots to collect the fine fraction (<150 μm). Each aliquot was placed on the screen, the closed sieve was shaken and tapped several times, the collection pan was emptied, and the process was repeated until no more dust passed into the pan. The fine fractions derived from these aliquots were then combined and split into two aliquots. One aliquot was soxhlet extracted for 16 hr with 200 ml diethyl ether:*n*-hexane (6:94), and the extracts were cleaned through a florisil column (*5*). These extracts were analyzed for 39 "neutral extractable" target compounds including 23 pesticides, 10 polycyclic aromatic hydrocarbons (PAHs), and six polychlorinated biphenyl (PCB) congeners. The second aliquot was extracted with ethanol:water (4:1) and chloroform and the ethyl acetate extract was derivatized using Regisil MTBSTFA [*N*-methyl-*N*-(*t*-butyldimethylsilyl)trifluoroacetamide]. These derivatized extracts were analyzed for three "acid extractable" target compounds: 2,4-D, dicamba, and pentachlorophenol.

Chemical analyses were performed using a Fisons VG-MD800 GC/MS instrument (Danvers, MA) in selected ion monitoring mode. A J&W DB-5.625 30 m × 0.25 mm ID column (J & W Scientific, Folsom, CA) was used for the analysis. Confirmation analysis for selected samples was done using the same column under full-scan mass spectral analysis on a second Fisons GC/MS instrument. Quantitation was based on five-point calibration curves. Continuing calibration was performed using the mid-level standard.

Comparison of the two dust collection methods was performed on a compound-by-compound basis. We calculated the percent agreement in detection of each compound between the two types of dust samples. We compared the median concentration of each compound among the HVS3 samples with its median in the used bag samples (values that were below the limit of detection by GC/MS, or nondetects, were excluded), and used Microsoft Excel version 4.0 (Microsoft, Redmond, WA) to perform a paired *t*-test for each compound. Finally, we calculated a Spearman rank correlation coefficient (nondetects included) for each compound that was detected in more than two pairs of samples. Nondetects were included in the correlation analysis because the dust sampling methods were being evaluated for use in epidemiologic studies, for which subjects' exposures would be ranked or categorized over the entire range of observations, including nondetects.

Results

The two methods performed similarly in detecting the presence of the target pesticides (Table 1), PAHs and PCBs (Table 2) in the dust. Between 80% and 100% of the time, the two methods agreed in detecting the presence of the target compound. For the compounds with less than perfect agreement, neither sampling method was consistently more sensitive than the other. Chlordane, dicofol, propoxur, carbaryl, and 2,4-D were detected more frequently

Table 1. Ability to detect pesticides: comparison of the high volume surface sampler (HVS3) and used bag methods

Class	Pesticide	No. of homes in which detected		Percent agreement
		HVS3	Used bag	
Organochlorine insecticides	α + γ-Chlordane[a]	14	13	80
	p,p'-DDE + *p,p'*-DDT[b]	8	8	100
	Methoxychlor	5	5	87
	Aldrin	1	1	100
	Dieldrin	1	1	100
	Heptachlor	1	1	100
	Dicofol	1	0	93
	Lindane	0	0	100
Organophosphate insecticides	Chlorpyrifos	7	9	87
	Diazinon	0	0	100
	Dichlorvos	0	0	100
	Malathion	0	0	100
Carbamate insecticides	Propoxur	10	9	93
	Carbaryl	8	5	80
	Bendiocarb	2	2	100
Pyrethroid insecticides	*cis-* + *trans*-Permethrin[c]	9	9	100
Fungicides	*o*-Phenylphenol	15	15	100
	Pentachlorophenol	15	15	100
Herbicides	Alachlor	0	0	100
	Atrazine	0	0	100
	2,4-D	9	8	80
	Dacthal	0	0	100
	Dicamba	1	1	87

[a]Summed concentration of α- and γ-chlordane, which were measured separately.
[b]Summed concentrations of *p,p'*-DDE and *p,p'*-DDT, which were measured separately.
[c]Summed concentrations of *cis-* and *trans*-permethrin, which were measured separately.

Table 2. Ability to detect polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs): comparison of the high volume surface sampler (HVS3) and used bag methods

Class	Compound	No. of homes in which detected		Percent agreement
		HVS3	Used bag	
PAHs	Benz[*a*]anthracene	15	15	100
	Benzo[*b*]fluoranthene	15	15	100
	Benzo[*k*]fluoranthene	15	15	100
	Benzo[*ghi*]perylene	15	15	100
	Benzo[*a*]pyrene	15	15	100
	Chrysene	15	15	100
	Coronene	15	15	100
	Indeno[1,2,3-*cd*]pyrene	15	15	100
	Dibenz[*a,h*]anthracene	14	15	93
	Dibenzo[*a,e*]pyrene	14	15	93
	Total PAHs[a]	15	15	100
PCBs	PCB 153	6	6	100
	PCB 180	6	6	100
	PCB 170	3	4	80
	PCB 138	2	2	100
	PCB 105	1	2	93
	PCB 126	0	0	100
	Total PCBs[b]	6	6	100

[a]Summed concentrations of above-listed PAHs.
[b]Summed concentrations of above-listed PCBs.

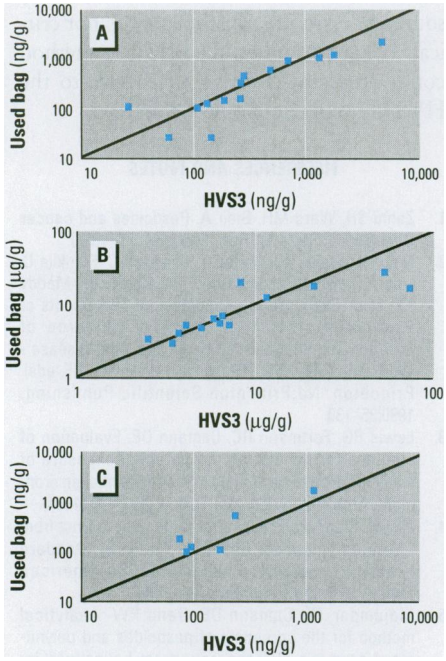

Figure 1. Pesticide concentrations measured in dust: high volume surface sampler (HVS3) versus used bag method. (A) α+γ-Chlordane. (B) Total polycylic aromatic hydrocarbons. (C) Total polychlorinated biphenyls.

in the HVS3 samples; chlorpyrifos, PCB 105, and PCB 170 were detected more frequently in the used bag samples. Seven pesticides and one PCB were not detected in any of the samples, indicating that these compounds had not been used or tracked into the subjects' homes or were present at levels below the analytical detection limit of the instrumentation.

Among the samples with detectable levels of pesticides, the median concentrations (not shown) were similar in the HVS3 and used bag samples, with two exceptions: methoxychlor (2.5 times higher in the HVS3 sample) and propoxur (2.1 times higher in the used bag sample). The medians for total PAHs and total PCBs were similar. A paired *t*-test showed no significant differences between the two methods for any of the compounds.

Figure 1A shows the concentrations of one of the pesticides (chlordane) in each pair of dust samples (the $y = x$ or "equal concentration" line is shown to aid interpretation). The chlordane levels in the two dust samples were remarkably similar throughout the range of concentrations encountered in this study. For chlordane and many other pesticides (chlorpyrifos, propoxur, carbaryl, *cis-* + *trans*-permethrin, *o*-phenylphenol, and 2,4-D), and for total PAHs (Fig. 1B), the two or three homes with the highest HVS3 concentrations fell below the "equal concentration" line, indicating that the concentrations of these compounds in the used bag sample were lower than in the HVS3 sample. This was not observed for DDE + DDT, methoxychlor, pentachlorphenol, or total PCBs (Fig. 1C).

Table 3. Spearman rank correlation: high volume surface sampler (HVS3) and used bag methods

Class	Compound	Spearman rank correlation coefficient
Organochlorine insecticides	α + γ-Chlordane[a]	0.93
	p,p′-DDE + *p,p′*-DDT[b]	0.97
	Methoxychlor	0.67
Organophosphate insecticides	Chlorpyrifos	0.87
Carbamate insecticides	Propoxur	0.95
	Carbaryl	0.74
Pyrethroid insecticides	*cis-* + *trans*-Permethrin[c]	0.95
Fungicides	*o*-Phenylphenol	0.46
	Pentachlorophenol	0.68
Herbicides	2,4-D	0.37
PAHs	Benz[*a*]anthracene	0.88
	Benzo[*b*]fluoranthene	0.89
	Benzo[*k*]fluoranthene	0.86
	Benzo[*ghi*]perylene	0.95
	Benzo[*a*]pyrene	0.85
	Chrysene	0.86
	Coronene	0.92
	Indeno(1,2,3-*cd*]pyrene	0.91
	Dibenz[*a,h*]anthracene	0.94
	Dibenzo[*a,e*]pyrene	0.89
	Total PAHs[d]	0.90
PCBs	PCB 153	0.96
	PCB 180	0.99
	PCB 170	0.64
	Total PCBs[e]	0.97

Abbreviations: PAHs, polycyclic aromatic hydrocarbons; PCBs, polychlorinated biphenyls.
[a]Summed concentrations of α- and γ-chlordane, which were measured separately.
[b]Summed concentrations of *p,p′*-DDE and *p,p′*-DDT, which were measured separately.
[c]Summed concentrations of *cis-* and *trans*-permethrin, which were measured separately.
[d]Summed concentrations of above-listed PAHs.
[e]Summed concentrations of all PCB congeners analyzed.

Despite the distributional differences noted above, when homes were placed in order of increasing concentrations of a target compound, the ordering was similar for most compounds regardless of whether it was based on levels in the HVS3 samples or used bag samples. Table 3 shows the Spearman rank correlation coefficient for all compounds that were detected in more than one pair of samples (nondetects included). The correlation coefficients were 0.85 or higher for all pesticides except carbaryl (0.74), pentachlorophenol (0.68), methoxychlor (0.67), *o*-phenylphenol (0.46), and 2,4-D (0.37), for all PAHs, and for all PCBs except PCB 170 (0.64).

Discussion

Based on the results of this pilot study, there appears to be no clear difference in the quality of the pesticide, PAH, and PCB concentration data for the two dust collection methods. The methods were remarkably close in their ability to detect these compounds in carpet dust, and neither method was consistently more sensitive than the other. The median levels in the detected samples were generally similar, and a paired *t*-test showed no significant differences. The Spearman rank correlation coefficients were high for most compounds, indicating that homes would be ranked in a similar order using both methods, making them equally suitable for assessing relative exposures in the context of an epidemiologic study. Excluding the nondetects from the data set reduced the correlation coefficients for some compounds, indicating that the methods should be reevaluated for use in other types of studies. Although the correlation coefficients were low for 2,4-D (a widely used herbicide) and *o*-phenylphenol (a widely used fungicide), there were limited ranges of detected levels for these compounds (only a fivefold difference between the highest and lowest values for 2,4-D and a sevenfold difference for *o*-phenylphenol). The rankings within these narrow ranges are likely unimportant from an etiologic perspective; a meaningful comparison of the dust collection methods would require a wider range of dust concentrations.

For many (but not all) of the pesticides and for total PAHs, concentrations in the HVS3 sample exceeded those of the used bag sample at the upper end of the concentration ranges. Given the small number of samples taken, this could have been due to chance. It is also possible that "hot spots" of these compounds in some homes were oversampled by the HVS3. Several other factors could have affected the relative concentrations of these compounds in the sample pairs. The HVS3 is fundamentally different from a typical household vacuum in that the collected dust is immediately removed from the air stream and diverted to a catch bottle. In a typical household vacuum, the collected dust remains in the bag, where air passes through it repeatedly during subsequent uses of the vacuum, possibly resulting in partial volatilization of the chemicals from the dust. This phenomenon could explain the observed data if it 1) operates more effectively when concentrations of chemicals in the dust are higher and 2) affects only certain types of compounds. Levels of target compounds in the used vacuum cleaner bag could also have been affected by the different designs of the household vacuums and the frequencies with which the bags were changed.

The most important limitation of this pilot study is its small sample size. It is difficult to determine whether the findings would hold across the wider range of concentrations likely to be encountered in a full-scale epidemiologic study.

In choosing between the two sampling methods, it is important to consider two points. First, some people do not own a vacuum and would be unable to provide a sample with the used bag method. Second, the used bag method is not appropriate for studies in which dust loading (i.e., the absolute amount of dust present) is an important parameter. Dust loading is important in studies looking at carpet dust as a direct source of exposure to pesticides; all else being equal, one would expect subjects living in dustier homes to have higher dust-related pesticide exposures than subjects living in cleaner homes. Because dust loading is important in some studies, additional research should be performed on the ability of certain types of standard vacuum cleaners to collect carpet dust samples in such a way that loadings can be estimated. On the other hand, if the carpet dust is being viewed as an indictator of the extent to which pesticides have been used in and around the home in the past, and not as a source of exposure, dust loading is not critical. For such studies, the used bag method could be a cost-effective alternative to the HVS3 for epidemiologic research.

References and Notes

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