DISTRIBUTIONS AND QUALITY OF PESTICIDE, PAH, AND PCB MEASUREMENTS IN BAG DUST FROM FOUR AREAS OF USA

DE Camann[1*], JS Colt[2] and MM Zuniga[1]

[1]Southwest Research Institute, San Antonio, TX, USA
[2]National Cancer Institute, Rockville, MD, USA

ABSTRACT
Distributions of the concentrations of prevalent pesticides, polycyclic aromatic hydrocarbons (PAH), and polychlorinated biphenyl (PCB) congeners were measured in vacuum bag dust samples obtained from the homes of case and control adults in four areas of the United States for a study of non-Hodgkin's lymphoma (NHL). All target analytes were efficiently extracted from the sieved fine dust. The distribution of detectable concentrations of prevalent pesticides and PAH spans more than two orders of magnitude. Discontinued pesticides such as DDT and chlordane are still common in house dust, and indicate continual exposure of the residents since discontinuation. Comparing the study sites, percentiles of the dust concentration distributions were generally larger for current-use insecticides and chlordane in Los Angeles County, ortho-phenylphenol in the Seattle area, agricultural herbicides in Iowa, and PAH and PCB congeners in the Detroit area.

INDEX TERMS
House dust, Pesticides, PAH, PCBs.

INTRODUCTION
House dust is a repository of semivolatile organic chemicals, such as pesticides, PAH, PCBs, and phthalates. Protected indoors from sunlight, moisture, temperature extremes, and most microbial action, semivolatile organics are more persistent in carpet dust than in soil (Simcox, 1995). Pesticide concentrations in dust have been found to correlate well with levels in indoor air and surface residues in the house (Camann, 1995) due to redistribution from the indoor source, via both resuspension and settling of particles and volatilization and condensation of the vapor phase. The concentration in the fine dust may thus correlate with biomarkers of the resident's exposure (Buckley, 1997). This paper presents the distributions of the concentrations of prevalent pesticides, PAH, and PCB congeners measured in vacuum bag dust samples obtained from the homes of case and control adults in four areas of the United States for a study to assess association with NHL.

METHODS
Vacuum cleaner bags were obtained between December 1998 and March 2000 from the first 616 homes analyzed of the over 1,200 homes sampled. 110 of these analyzed bags were from the Detroit area, Michigan, 204 from the entire state of Iowa, 143 from Los Angeles County, California, and 159 from the Seattle area, Washington. Bags in current use were collected from adults aged 20 to 74, both cases with recent NHL diagnosis and random-selected controls, when the subject had a working vacuum cleaner and owned half or more of the carpets at least five years. Bags were shipped overnight to the laboratory and stored frozen. Six representative portions of dust from the opened bag were passed though a 100-mesh sieve to obtain the fine (< 150 μm) dust. In batches of 15 samples, including at least 4 case and 4

* Contact author email: dcamann@swri.org

control dusts, up to 2.0 g of fine dust was soxhlet-extracted with 6% ether/hexanes, concentrated, cleaned through a florisil column, and analyzed by GC/MS selected ion monitoring for 25 pesticides, 7 PAH, and 5 PCB congeners. Analyte amounts were quantified using the internal standard method with deuterated PAH as internal standards.

All detections and co-eluting interferences from dust were identified, coded, and treated consistently. Sample detection limits in μg/g of fine dust were normally defined as equivalent to 1/3 of the lowest analytical standard, but were raised to 1/5 of a larger false interference peak when present. The largest non-detection among the clustered non-detects for an analyte was treated as its detection limit, and larger non-detections were treated as missing values. When the analyte and an interference co-eluted, the analyte amount was considered half of the co-elution peak.

RESULTS AND DISCUSSION

The quality of the bag dust measurements is excellent. Lab spikes of 13 home dusts showed that all target analytes were efficiently extracted, as shown in Table 1, but recovery of some pesticides was often elevated. Reported levels in dust have not been adjusted for the mean spike recoveries. Analysis of lab splits of 15 dust samples showed close agreement between the regular sample and the lab split. The measurements for 91% of the 239 detection pairs agreed within 20%, and 99% agreed within 40%. Confirmation analyses performed by full-scan GC/MS on 32 samples generally verified the large regular analysis results, indicating that the analytes had been properly identified despite the interferences often present in dust.

Table 1. Percent recovery of analytes spiked into aliquots of 13 sieved dust samples

Pesticides:	% Recovery, mean ± std dev			% Recovery, mean ± std dev
Acetochlor	113 ± 21		Methoxychlor	141 ± 26
Alachlor	112 ± 18		Metolachlor	113 ± 20
Aldrin	96 ± 8		cis-Permethrin	122 ± 30
Atrazine	107 ± 20		trans-Permethrin	139 ± 54
Bendiocarb	103 ± 28		ortho-Phenylphenol	96 ± 22
Carbaryl	129 ± 50		Propoxur	106 ± 23
alpha-Chlordane	96 ± 7		PAH and PCBs:	
gamma-Chlordane	95 ± 7		Benz(a)anthracene	106 ± 9
Chlorpyrifos	105 ± 17		Benzo(b)fluoranthene	98 ± 10
Cyanazine	84 ± 23		Benzo(k)fluoranthene	94 ± 7
Dacthal	98 ± 8		Benzo(a)pyrene	101 ± 12
4,4'-DDE	96 ± 7		Chrysene	97 ± 8
4,4'-DDT	137 ± 30		Dibenz(ah)anthracene	88 ± 12
Diazinon	101 ± 13		Indeno(123cd)pyrene	99 ± 14
Dicofol	133 ± 25		PCB 105	95 ± 6
Dieldrin	93 ± 7		PCB 138	97 ± 9
Heptachlor	104 ± 6		PCB 153	95 ± 8
Lindane	95 ± 7		PCB 170	105 ± 13
Malathion	113 ± 22		PCB 180	101 ± 10

Dust concentration percentiles are not presented for the 13 pesticides which were detected in 10% or less of the dust samples. These pesticides are lindane (detected in 10%), malathion

(8%), metolachlor (8%), bendiocarb (7%), dieldrin (7%), heptachlor (7%), atrazine (5%), dacthal (5%), acetochlor (3%), alachlor (3%), dicofol (3%), aldrin (1%), and cyanazine (< 1%).

Percentiles of the distributions of the concentrations in fine house dust (in μg/g) of these four areas of USA are presented for the 12 prevalent pesticides in Table 2, for the PAH in Table 3, and for the PCB congeners in Table 4. The concentration distributions in fine dust are very broad, ranging between two and three orders of magnitude in concentration from the 10th percentile to the maximum value. Among pesticides, the 95th concentration percentiles exceeded 20 μg/g for trans-permethrin and cis-permethrin, and exceeded 1.0 μg/g for chlorpyrifos, carbaryl, methoxychlor, 4,4'-DDT, propoxur, and ortho-phenylphenol. It is noteworthy that the dust concentrations of two discontinued insecticides, DDT (use discontinued in 1971) and chlordane (use discontinued in 1988), are still similar to those of the major organophosphate (chlorpyrifos and diazinon) and carbamate (carbaryl and propoxur) insecticides in use in the USA during the collection period. Pesticide concentrations were strongly correlated only within families: alpha-chlordane with gamma-chlordane, cis-permethrin with trans-permethrin, and DDE with DDT. The 95th concentration percentiles of six PAH exceeded 1.0 μg/g. Concentrations of the seven PAH are highly correlated, as expected, since all of the PAH are produced by the same combustion processes. Concentration percentiles of the PCB congeners are considerably lower than those of the PAH.

Table 2. Percentiles of concentration distributions of prevalent pesticides in dust (μg/g)

	n	10th	25th	50th	75th	90th	95th	Max
Carbaryl	609	< 0.027	< 0.027	0.050	0.31	1.48	3.01	223.
alpha-Chlordane	599	< 0.021	< 0.021	< 0.021	0.060	0.187	0.44	5.8
gamma-Chlordane	604	< 0.021	< 0.021	0.021	0.084	0.28	0.69	8.5
Chlorpyrifos	610	< 0.021	0.030	0.108	0.48	1.87	3.53	38.2
4,4'-DDE	600	< 0.021	< 0.021	0.023	0.053	0.111	0.20	2.45
4,4'-DDT	603	< 0.021	< 0.021	0.090	0.31	0.75	1.60	24.6
Diazinon	606	< 0.021	< 0.021	0.025	0.086	0.36	0.76	8.3
Methoxychlor	462	< 0.025	< 0.025	0.074	0.33	1.02	2.07	8.2
cis-Permethrin	609	< 0.055	< 0.055	0.33	2.16	9.90	20.9	240.
trans-Permethrin	595	< 0.059	< 0.059	0.70	4.15	16.4	38.7	328.
ortho-Phenylphenol	616	0.081	0.144	0.25	0.45	0.88	1.25	38.2
Propoxur	609	< 0.021	0.026	0.072	0.25	0.77	1.45	38.2

Table 3. Percentiles of concentration distributions of measured PAH in dust (μg/g)

	n	10th	25th	50th	75th	90th	95th	Max
Benz(a)anthracene	616	0.038	0.062	0.136	0.38	1.05	1.79	30.5
Benzo(b)fluoranthene	616	0.077	0.152	0.31	0.86	2.32	3.95	47.0
Benzo(k)fluoranthene	611	0.024	0.048	0.099	0.29	0.76	1.40	13.1
Benzo(a)pyrene	616	0.034	0.068	0.154	0.45	1.29	2.39	25.5
Chrysene/ iso-Chrysene	616	0.086	0.151	0.27	0.68	1.56	2.84	28.0
Dibenz(ah)anthracene	605	< 0.021	< 0.021	0.036	0.095	0.25	0.50	4.5
Indeno(123cd)pyrene	614	0.035	0.080	0.161	0.44	1.34	2.39	23.4

Table 4. Percentiles of concentration distributions of PCB congeners in dust (μg/g)

	n	50th	75th	90th	95th	Max
PCB 105	594	< 0.021	< 0.021	0.046	0.090	3.9
PCB 138	593	< 0.021	0.030	0.076	0.22	10.2
PCB 153	597	< 0.021	0.036	0.092	0.24	6.5
PCB 170	590	< 0.021	< 0.021	0.029	0.056	1.4
PCB 180	592	< 0.021	0.022	0.053	0.112	2.9

Separate concentration distributions in dust were also determined for the four study sites. The same chemicals were prevalent in each area. The 90th percentiles are compared by site in Table 5. Some geographic patterns are evident. The concentration percentiles of each of the insecticides in current use were generally larger in the dust from Los Angeles County than from the other sites. This may reflect the warmer climate in Los Angeles and the need for year-round insect control. The Seattle area had higher concentration percentiles of the disinfectant ortho-phenylphenol, consistent with its more humid climate. Upper concentration percentiles of agricultural herbicides (metolachlor, atrazine, alachlor, and acetochlor) were substantially higher in dust from Iowa than from the other sites (results not shown), reflecting their greater usage in Iowa. PAH percentile levels were markedly highest in the house dust in the Detroit area, and lowest in the Los Angeles dust; this may relate to the differential in the use of combustion for heating among the four sites. Dust levels of PCBs 138 and 153 were also higher in the Detroit area than the other study sites.

Table 5. 90th percentile of concentration in dust (μg/g) by chemical and site

	Detroit area	Iowa	Seattle area	Los Angeles Co.	High site*
Organochlorine pesticides					
4,4'-DDT	0.56	0.93	0.57	0.88	Iowa/
4,4'-DDE	0.13	0.13	0.07	0.14	LA
gamma-Chlordane	0.08	0.36	0.10	0.62	
alpha-Chlordane	0.07	0.22	0.08	0.38	LA
Methoxychlor	0.95	1.3	1.3	0.52	
Current use pesticides					
Chlorpyrifos	2.0	1.2	0.87	2.6	LA
Diazinon	0.39	0.11	0.32	0.64	LA
Carbaryl	0.32	1.7	1.3	2.5	LA
Propoxur	0.47	0.45	0.99	1.45	LA
cis-Permethrin	3.0	5.6	13.8	16.0	
trans-Permethrin	5.8	11.0	22.5	33.4	LA
ortho-Phenylphenol	0.98	0.70	1.01	0.87	Seattle
Polycyclic aromatic hydrocarbons (PAH)					
Benz(a)anthracene	3.2	0.73	0.68	0.16	
Benzo(b)fluoranthene	8.1	1.5	1.5	0.38	Detroit
Benzo(a)pyrene	4.8	0.87	0.88	0.21	
Polychlorinated biphenyl (PCB) congeners					
PCB 138	0.19	0.06	0.11	0.04	Detroit
PCB 153	0.17	0.07	0.15	0.04	Detroit
PCB 180	0.06	0.05	0.07	0.04	

* consistently higher site at most percentiles

CONCLUSIONS AND IMPLICATIONS

The method of dust collection and analysis described herein is feasible for epidemiologic studies of home environmental exposures. Detectable concentration distributions of prevalent pesticides and PAH in fine dust span more than two orders of magnitude, suggesting that residents have inhalation, dermal, and non-dietary ingestion exposures to these chemicals at home which span a similar broad range. Discontinued pesticides such as DDT and chlordane are still common in house dust, and indicate continual exposure of the residents at gradually declining levels since discontinuation. Among the study sites, upper percentiles of the dust concentration distributions for current-use insecticides and chlordane were generally larger in Los Angeles County, for ortho-phenylphenol in the Seattle area, for agricultural herbicides in Iowa, and for PAH and PCB congeners in the Detroit area.

ACKNOWLEDGEMENT

This study was sponsored by contract 263-MQ-821644 from the National Cancer Institute to Southwest Research Institute.

REFERENCES

Buckley TJ, Liddle J, Ashley DL, *et al.* 1997. Environmental and biomarker measurements in nine homes in the lower Rio Grande valley: multimedia results for pesticides, metals, PAHs, and VOCs. *Environment International.* Vol. 23 (5), pp 705-732.

Camann DE, Harding HJ, Clothier JM, *et al.* 1995. Dermal and in-home exposure of the farm family to agricultural pesticides, *Measurement of Toxic and Related Air Pollutants,* Pittsburgh PA: Air & Waste Mgmt Assn, pp 548-554.

Simcox NJ, Fenske RA, Wolz SA, *et al.* 1995. Pesticides in household dust and soil: exposure pathways for children of agricultural families. *Environmental Health Perspectives,* Vol. 103 (12), pp 1126-1134.