4.0 SURFACE MEASUREMENTS

4.1 Introduction and Data Availability

The objectives of measuring pesticide surface residue concentrations and loadings are to describe the extent and distribution of concentrations, identify possible sources of indoor contamination, evaluate factors that may impact concentrations, and identify elevated concentrations for the purposes of intervention. Surface measurements tell us what pesticide residues are present in an environment and at what concentrations. With appropriate transfer coefficients and activity data, these measurements can be used to estimate dermal and nondietary ingestion exposure.

Although exposure potential is highest during the first few days following an application, pesticide residues introduced into the indoor residential environment may persist for months or even years on surfaces or embedded in carpets, where these are protected from sunlight, rain, temperature extremes, and microbial action (Lewis *et al.,* 1994). Surface residues may contribute to the exposure of household occupants through multiple routes: dermal absorption, inhalation of resuspended particles, nondietary ingestion of residues adhering to mouthed objects and skin, and dietary ingestion resulting from children's unique handling of food (Butte and Heinzow, 2002). Oral ingestion and dermal absorption of surface residues may be major routes of exposure for infants and toddlers who spend much of their time on the floor, explore their world through mouthing, experience frequent hand-to-mouth and object-to-mouth contacts, and who may have pica tendencies (Butte and Heinzow, 2002; Cohen Hubal *et al.,* 2000a, b; Freeman *et al.,* 2004; Lewis *et al.,* 1994; Tulve *et al.,* 2002). Ingestion of soil is also a special concern for young children, who may ingest up to 10 times more soil than adults on a per kilogram body weight basis (LaGoy, 1987).

Several surface sampling methods exist including deposition coupons, Octadecyl (C18) surface press sampler (EL Sampler), Lioy-Weisel-Wainman (LWW) sampler, vacuum, drag bar, California-roller, PUF roller, and surface wipes. These methods are generally classified by the degree to which they remove residues from surfaces: total available residue, transferable residue, and dust (Lewis, 2001). T*otal available residue* methods attempt to measure the total amount of contaminant on a surface (often with the aid of isopropanol as a solvent), *transferable residue* methods are intended to represent the amount that is transferred as a result of contact with the contaminated surface, and *dust collection* methods use a vacuum to collect dust-borne residue on surfaces and from carpet. Transferable residues are also referred to as dislodgeable residues. All studies discussed in this chapter employed more than one sampling method for surface measurements. Table 4.1 lists the studies that collected surface measurements along with the type of measurement taken. Limits of detection for each chemical by study and method are listed in Table 4.2.

Several variables may influence measured dust concentrations or surface loadings of pesticide residues. These variables include the collection method itself, surface type, compound physico-chemical characteristics, application method, application frequency, sampling locations, participant activities, and analytical capabilities. This chapter examines how these factors may have affected the surface residue measurements in the children's exposure measurement program, the implications for interpreting the data, and the consequences for exposure estimates.

Table 4.1 Studies and sample collection methods for surface measurements.

Study	Dust (ng/g)	Dust Load (ng/cm^2)	Soil (ng/g)	Total Surface Load (ng/cm^2)	Transferable Residues (ng/cm^2)
NHEXAS-AZ	✓	✓	✓	--	Wipes (water)
MNCPES				LWW	C18 Press
CTEPP	✓	✓	✓	--	Wipes (2 mL IPA), PUF Roller
CCC	--	--	✓	Wipes (20 mL IPA)	C18 Press
JAX	--	--	--	Wipes (20 mL IPA)	C18 Press
CHAMACOS	✓	✓	✓	Wipes (20 mL IPA)	C18 Press
CPPAES	--	--	--	Deposition Coupons, LWW	--
Test House	--	--	--	Deposition Coupons, Wipes (10 mL IPA)	PUF Roller C18 Press
PET	✓	--	✓		PUF Roller
DIYC	--	--	--	Wipes (20 mL IPA)	PUF Roller
Daycare	--	--	--	Wipes (20 mL IPA)	PUF Roller, C18 Press

--, matrix not sampled
LWW, Lioy-Weisel-Wainman sampler
C18, 3M Empore™ Octadecyl (C18) filters
PUF, Polyurethane foam

Table 4.2 Limits of detection (ng/g or ng/cm²) for surface measurements by study, method, and compound.

Study	Method	Chlor-pyrifos	Diaz-inon	*c*-Per-methrin	*t*-Per-methrin	Cyflu-thrin	Cyper-methrin	Esfen-valerate	TCPy	IMP
Soil (ng/g)										
MNCPES	Soil	10	10	10	10	--	--	--	--	--
CTEPP	Soil	0.5	0.5	0.5	0.5	5	--	--	0.2	0.2
CCC	Soil	5	2	5	5	6	6	--	--	--
PET	Soil	--	60	--	--	--	--	--	--	--
Dust (ng/cm² or ng/g)										
NHEXAS-AZ	Dust (ng/cm²)	0.002	0.002	--	--	--	--	--	--	--
CTEPP	Dust (ng/cm²)	0.0003	0.0003	0.0003	0.0003	0.0030	--	--	0.0003	--
NHEXAS-AZ	Dust (ng/g)	4	18	--	--	--	--	--	--	--
CTEPP	Dust (ng/g)	2	2	2	2	10	--	--	2	2
CHAMACOS	Dust (ng/g)	1	1	1	1	100	--	--	--	--
PET	Dust (ng/g)	--	60	--	--	--	--	--	--	--
Total Available Residue (ng/cm²)										
NHEXAS-AZ	IPA Wipe	0.070	2.00	--	--	--	--	--	--	--
MNCPES	LWW	1.200	3.50	--	--	--	--	--	--	--
CCC	IPA Wipe	0.005	0.002	0.005	0.005	0.006	0.006	--	--	--
JAX	IPA Wipe	0.005	0.002	0.005	0.005	0.006	0.006	0.008	--	--
CHAMACOS	IPA Wipe	0.005	0.005	0.005	0. 002	--	--	--	--	--
CPPAES	IPA Wipe	0.001	--	--	--	--	--	--	--	--
CPPAES	LWW	0.030	--	--	--	--	--	--	--	--
CPPAES	Dep Coup	0.010	--	--	--	--	--	--	--	--
TESTHOUSE	IPA Wipe	0.001	--	--	--	--	--	--	--	--
TESTHOUSE	Dep Coup	0.010	--	--	--	--	--	--	--	--
DIYC	IPA Wipe	--	0.300	--	--	--	--	--	--	--
DAYCARE	IPA Wipe	--	--	--	--	--	--	0.400	--	--
Transferable Residue (ng/cm²)										
MNCPES	C18 Press	0.330	0.140	--	--	--	--	--	--	--
CTEPP	IPA Wipe	0.0007	0.0007	0.0007	0.0007	0.007	--	--	0.0007	0.0007
CTEPP	PUF	0.0004	0.0004	0.0004	0.0004	0.004	--	--	0.0004	0.0004
TESTHOUSE	C18 Press	0.030	--	--	--	--	--	--	--	--
TESTHOUSE	PUF	0.001	--	--	--	--	--	--	--	--
PET	PUF	--	0.030	--	--	--	--	--	--	--
DIYC	C18 Press	--	1.200	--	--	--	--	--	--	--

--, analyte not measured

4.2 Dust and Soil Measurements

Dust is considered a repository of environmental pollutants that have accumulated indoors from both internal and external sources. Dust collected by vacuum is usually sieved to retain a particular size fraction for analysis, which may have important implications since pesticide concentrations are inversely related to particle size (Lewis *et al.,* 1999). Measurements in dust may be reported as concentrations (mass residue per unit weight of dust, ng/g) or as loadings (mass residue per unit area sampled, ng/cm^2). There is a lack of consensus on which of these metrics is more relevant to human exposure to pesticides; however, lead studies have suggested that lead loading correlates better with children's blood lead levels than does lead concentration (Lanphear, 1995).

Pesticides were measured in dust samples from the NHEXAS-AZ, CTEPP, CHAMACOS and PET studies. The CTEPP, CHAMACOS, and PET studies used the High Volume Small Surface Sampler (HVS3), whereas NHEXAS-AZ used a modified commercially available vacuum for ease of sample collection. The HVS3 was developed for the EPA and efficiently collects carpet-embedded dust retaining the associated pesticides (Roberts *et al.,* 1991; Lewis *et al.,* 1994). The HVS3 is a high-powered vacuum cleaner equipped with a nozzle that can be adjusted to a specific static pressure and air flow rate. A cyclone removes particles >5 μm from the air stream for collection in a catch bottle. Use of this sampler is limited to floors or other large flat surfaces (Roberts *et al.,* 1991; Ness, 1994; Lewis *et al.,* 1994). The ASTM (American Society for Testing and Materials) method for the collection of carpet-embedded dust requires an apparatus with the specifications of the HVS3 (ASTM, 1993). Pesticide concentrations in soil were measured in the same studies and results have been included in this chapter to allow comparisons between indoor and outdoor exposure pathways for the same children.

Pesticide Presence in Dust and Soil

Detection limits are listed in Table 4.2. Detection frequencies are presented in Figure 4.1 for soil samples and Figure 4.2 for dust samples. Concentrations of pesticides in soil and dust samples at the median and 95th percentile are listed in Table 4.3 (complete summary statistics are listed in Tables A.8 through A.19 in Appendix A).

- With the exception of cyfluthrin (for which analytical difficulties produced a higher detection limit), dust samples had high detection frequencies (>95%) in CTEPP and CHAMACOS. Detection frequencies were lower in NHEXAS-AZ due to higher detection limits.
- The high detection frequencies of pesticides observed in dust across studies is consistent with dust being a repository of contaminants.
- Detection frequencies for soil samples, on the other hand, were generally low (Figure 4.1). The high detection frequency of diazinon in PET study soil was due to direct lawn applications of the pesticide prior to sample collection.
- Pesticide concentrations were much lower in soil samples than in dust samples. In general, soil levels at the 95th percentile were a factor of 10 to 100 times lower than dust levels at the same percentile. This result suggests that in the absence of outdoor turf treatments, ingestion of soil may not be an important exposure pathway for these

pesticides, with the possible exception of children exhibiting pica behavior.

Concentrations in Dust and Soil: Summary Findings

Lognormal probability plots that graphically depict pesticide concentrations in soil from large observational field studies are presented in Figure 4.3. Plots that depict pesticide concentrations and loadings in dust are given in Figures 4.4 and 4.5. Box-and-whisker plots comparing pesticide concentrations and loadings in dust across all studies are given in Figures 4.6 and 4.7.

- The upper tails of the soil concentration distributions tend to be in the same range as the lower tails of the dust concentration distributions (Figures 4.3-4.5). For example, the 95th percentile for both chlorpyrifos and diazinon in *soil* is approximately 10 ng/g, and the 5th percentile for both of these compounds in *dust* is also near 10 ng/g.
- Among the pesticides measured in soil, cyfluthrin stands out for its high values at the 95th percentile (Table 4.3). Due to the low detection frequencies, no additional analysis was conducted with the soil data.
- Comparisons of concentrations in dust across studies (Figures 4.4-4.5) show permethrin (a pyrethroid) to be about an order of magnitude higher than chlorpyrifos and diazinon (both organophosphates).
- Overall, diazinon concentrations are lower than all other pesticides reported in dust, as illustrated in the box-and-whisker plots (Figures 4.6-4.7).
- High loadings of diazinon in indoor house dust following the lawn treatment in the PET study suggest translocation into the house by the occupants and their pets.
- The concentration ranking among the compounds in dust is the opposite of that found in air where the more volatile pesticides showed the higher concentrations. The less volatile pyrethroid pesticides tend to partition to the dust and may degrade more slowly, allowing accumulation over time from repeated applications. These results point to the importance of dust as a primary residential exposure medium for the less volatile pesticides. In addition, the exposure factors that are important for other nonvolatile contaminants such as lead (Melnyk *et al.*, 2000) may also be important for the less volatile pesticides.
- In general, the lognormal plots (Figures 4.4-4.5) indicate that differences between study populations are more apparent with dust loadings than with dust concentrations.
- In CTEPP, pesticide loadings in surface dust (ng/cm^2) were higher in daycare centers (DC) than in homes (HM) (Figures 4.6-4.7). This appears to be a function of the amount of surface dust present, as the pesticide concentrations in the dust do not differ by much (Figures 4.6-4.7). Studies with lead have suggested that loading has a greater impact than concentration on intake, and the same may or may not be true for pesticides.
- Concentrations of chlorpyrifos in dust (ng/g) are similar across studies (Figure 4.4) suggesting that the usage of chlorpyrifos did not change significantly from the timeframe of the NHEXAS-AZ study (1995-1997) to the CTEPP study (2000-2001).
- As with the other surface measurement methods, *cis*- and *trans*-permethrin have similar concentration profiles in dust samples.

Table 4.3 Median and 95th percentile values for soil (ng/g) and dust (ng/cm² and ng/g) measurements by study.

	Units	Chlorpyrifos		Diazinon		*c*-Permethrin		*t*-Permethrin		Cyfluthrin		TCPy		IMP	
		P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
SOIL															
MNCPES	ng/g	<10.0	<10.0	<10.0	<10.0	--	--	--	--	--	--	--	--	--	--
CTEPP-NC h[a]	ng/g	<0.5	17.0	<0.5	4.2	<0.5	13.0	<0.5	18.0	<5.0	32.0	0.6	11.0	--	--
CTEPP-NC d	ng/g	<0.5	0.8	<0.5	<0.5	<0.5	2.6	<0.5	2.2	<5.0	42.0	<0.2	1.2	--	--
CTEPP-OH h	ng/g	<0.5	14.0	<0.5	4.7	<0.5	2.7	<0.5	2.1	<5.0	64.0	0.7	8.9	<0.2	2.1
CTEPP-OH d	ng/g	<0.5	6.2	<0.5	7.1	<0.5	<0.5	<0.5	<0.5	<5.0	42.0	0.6	6.3	<0.2	1.4
CCC	ng/g	<5.0	27.0	<2.0	22.0	<5.0	8.6	<5.0	12	<6.0	8.6	--	--	--	--
PET	ng/g	--	--	22000	50000	--	--	--	--	--	--	--	--	--	--
DUST (Loadings)															
NHEXAS-AZ	ng/cm²	0.007	2.80	0.002	0.18	--	--	--	--	--	--	--	--	--	--
CTEPP-NC h	ng/cm²	0.009	0.42	0.002	0.12	0.10	4.90	0.09	4.40	<0.003	0.16	0.008	0.37	--	--
CTEPP-NC d	ng/cm²	0.066	1.30	0.026	9.90	0.69	5.50	0.41	6.30	<0.003	0.60	0.020	0.37	--	--
CTEPP-OH h	ng/cm²	0.006	0.35	0.002	0.31	0.05	3.80	0.03	3.90	0.018	0.25	0.004	0.16	0.001	0.046
CTEPP-OH d	ng/cm²	0.046	0.89	0.022	0.39	0.27	4.80	0.31	4.70	0.140	1.10	0.024	0.40	0.004	0.072
PET	ng/cm²	--	--	0.350	68	--	--	--	--	--	--	--	--	--	--
DUST (Concentrations)															
NHEXAS-AZ	ng/g	140	120000	150	8000	--	--	--	--	--	--	--	--	--	--
CTEPP-NC h	ng/g	130	1200	18	390	800	21000	630	19000	47	1700	96	1100	--	--
CTEPP-NC d	ng/g	140	920	47	6900	890	10400	760	12000	79	1500	63	300	--	--
CTEPP-OH h	ng/g	52	1400	20	1700	470	7600	340	9200	200	1300	41	820	14	
CTEPP-OH d	ng/g	180	1100	38	1600	690	3800	480	3400	350	890	67	500	17	310
CHAMACOS	ng/g	49	1200	21	820	150	2900	40	15000	<50	303.6	--	--	--	--
PET	ng/g	--	--	3100	150000	--	--	--	--	--	--	--	--	--	--

[a] CTEPP: h = home, d = daycare
--, analyte not measured

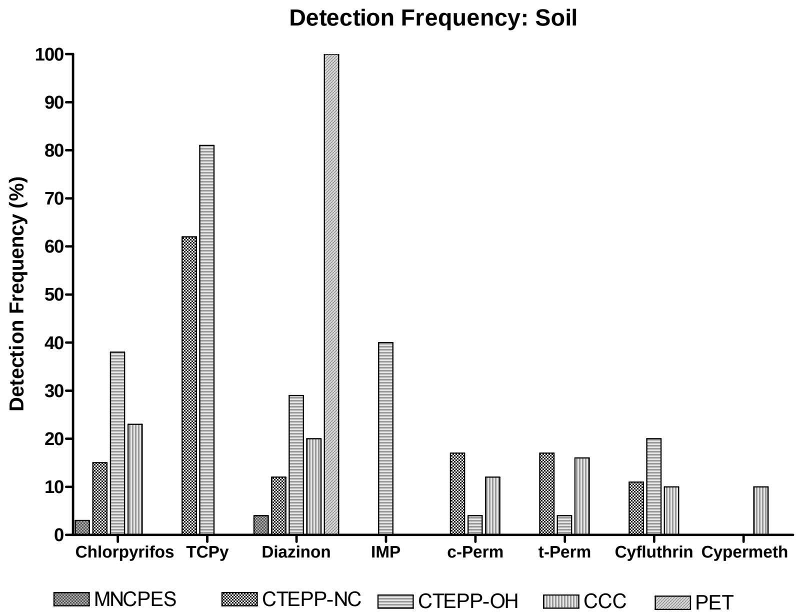

Figure 4.1 Detection frequencies of pesticides and degradates in soil.

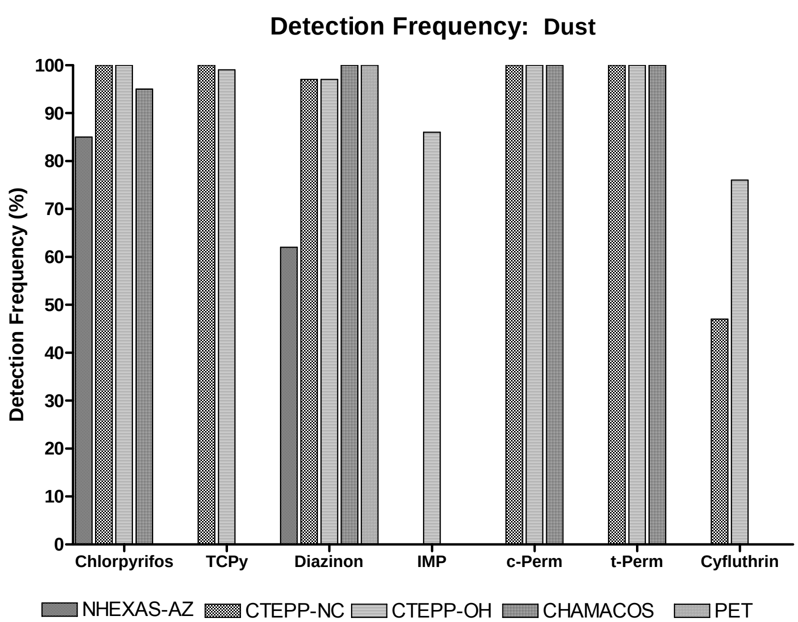

Figure 4.2 Detection frequencies of pesticides and degradates in dust.

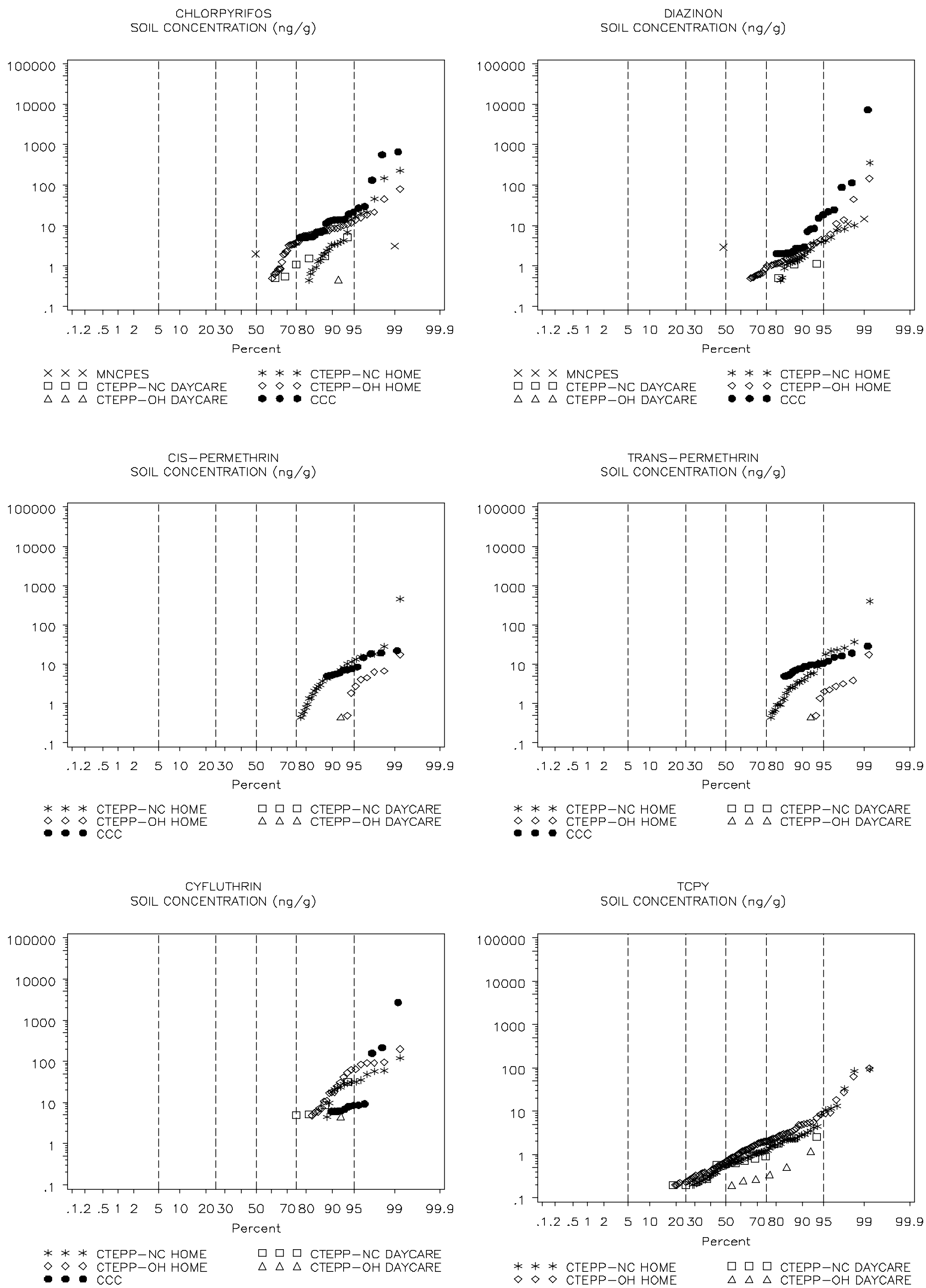

Figure 4.3 Lognormal probability plots of soil concentrations (ng/g) for chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin, cyfluthrin, and TCPy.

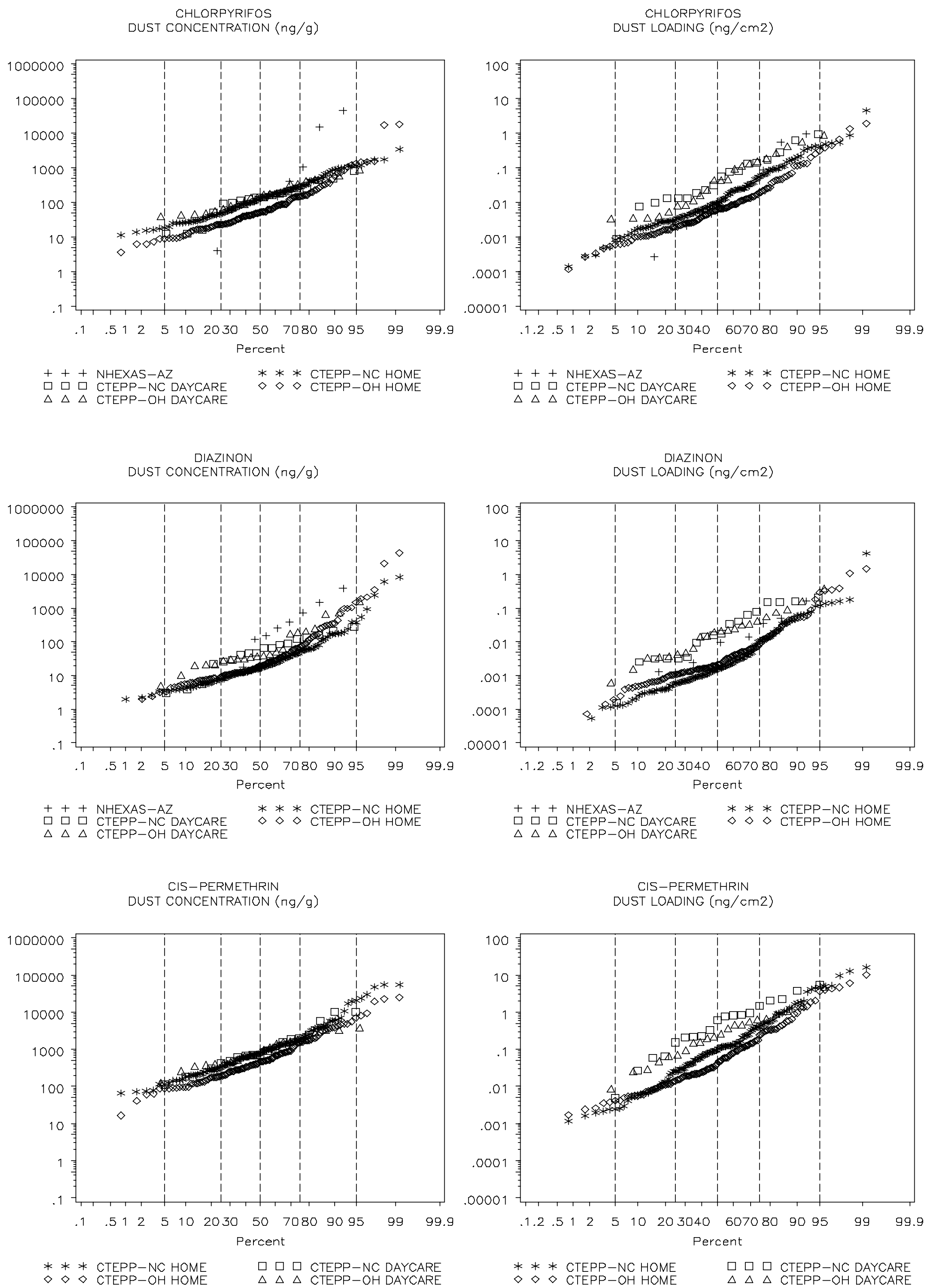

Figure 4.4 Lognormal probability plots of dust concentrations (ng/g) and loadings (ng/cm²) for chlorpyrifos, diazinon, and *cis*-permethrin.

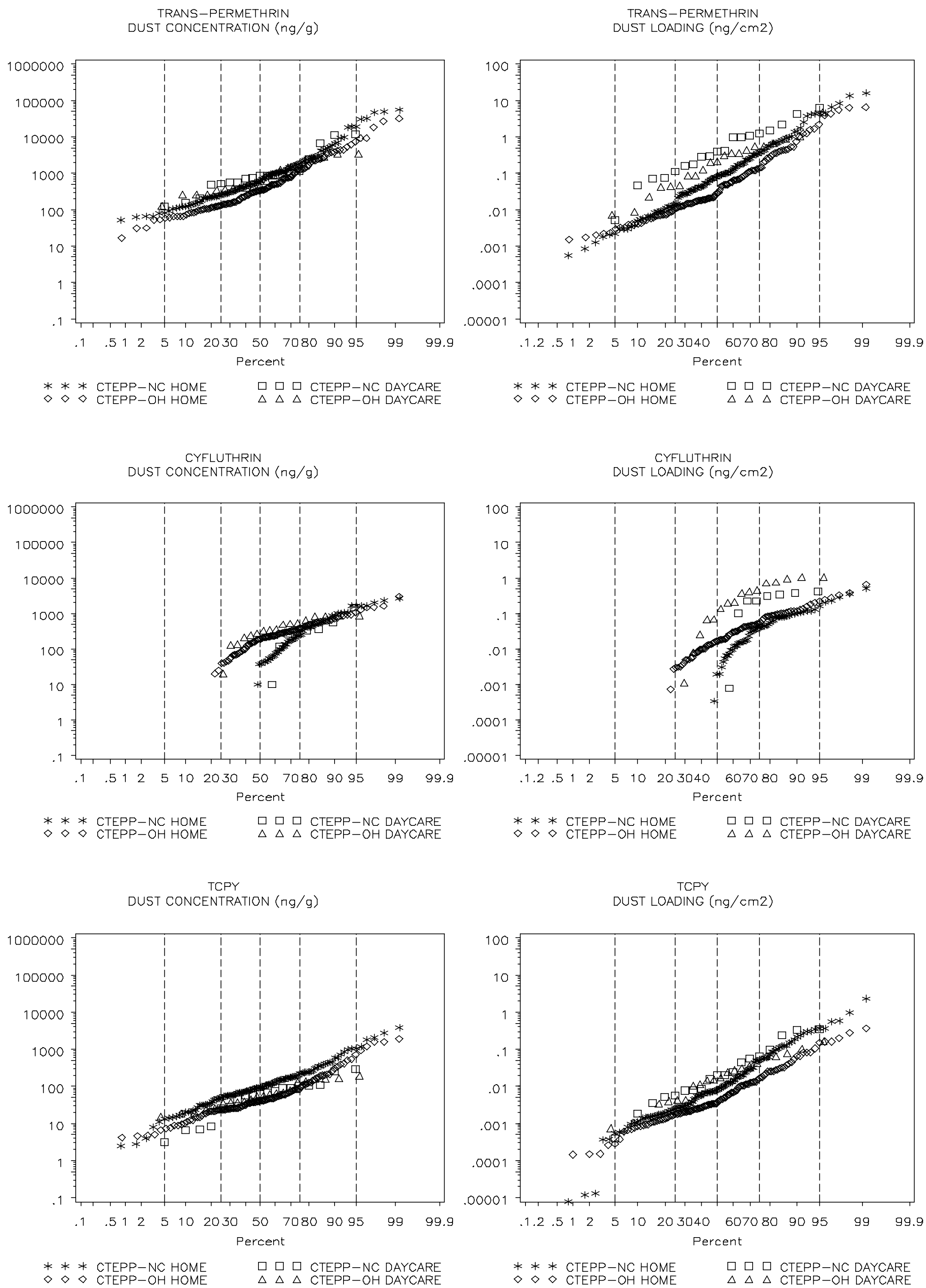

Figure 4.5 Lognormal probability plots of dust concentrations (ng/g) and loadings (ng/cm²) for *trans*-permethrin, cyfluthrin, and TCPy.

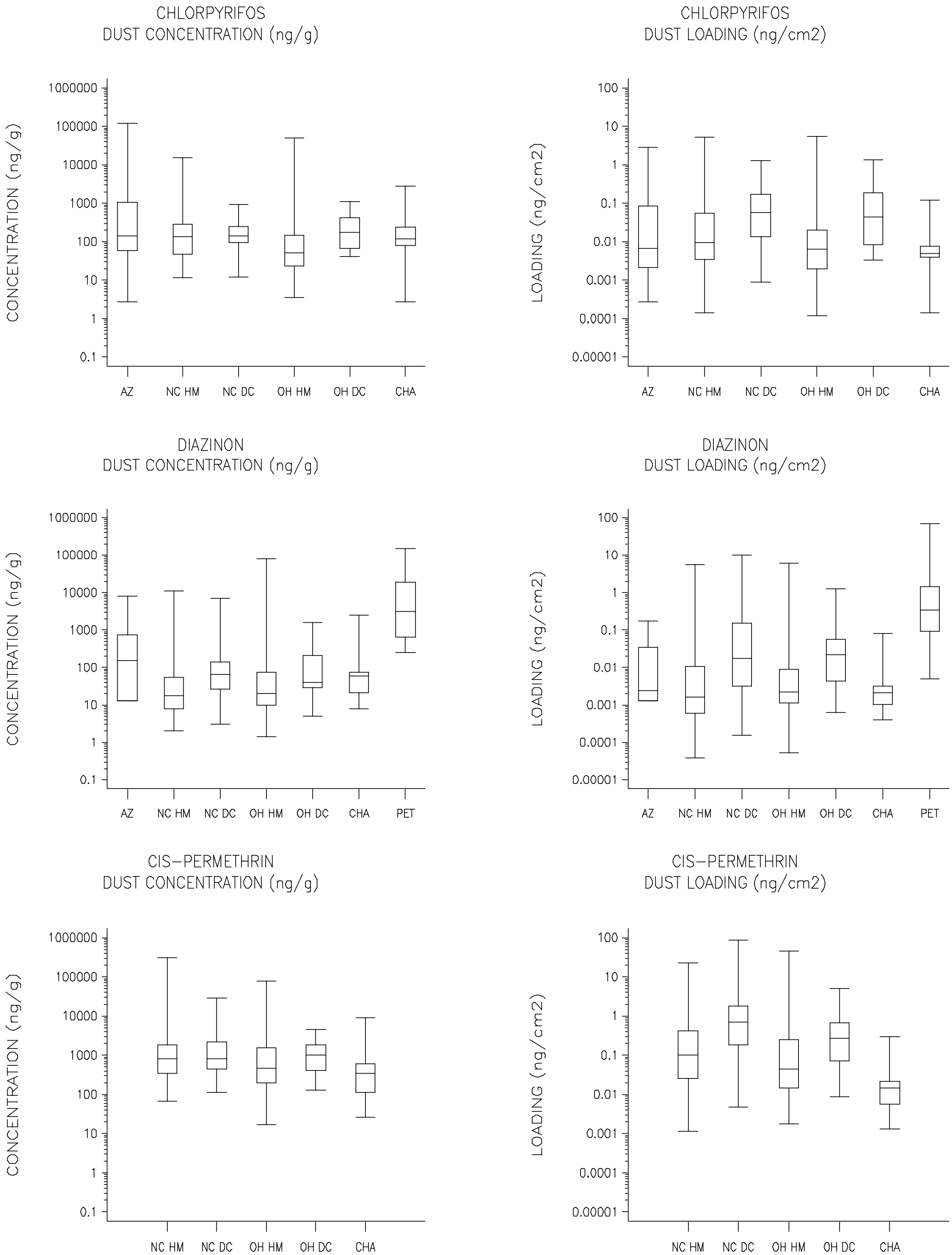

Figure 4.6 Box-and-whisker plots of dust concentrations (ng/g) and loadings (ng/cm²) for chlorpyrifos, diazinon, and *cis*-permethrin.

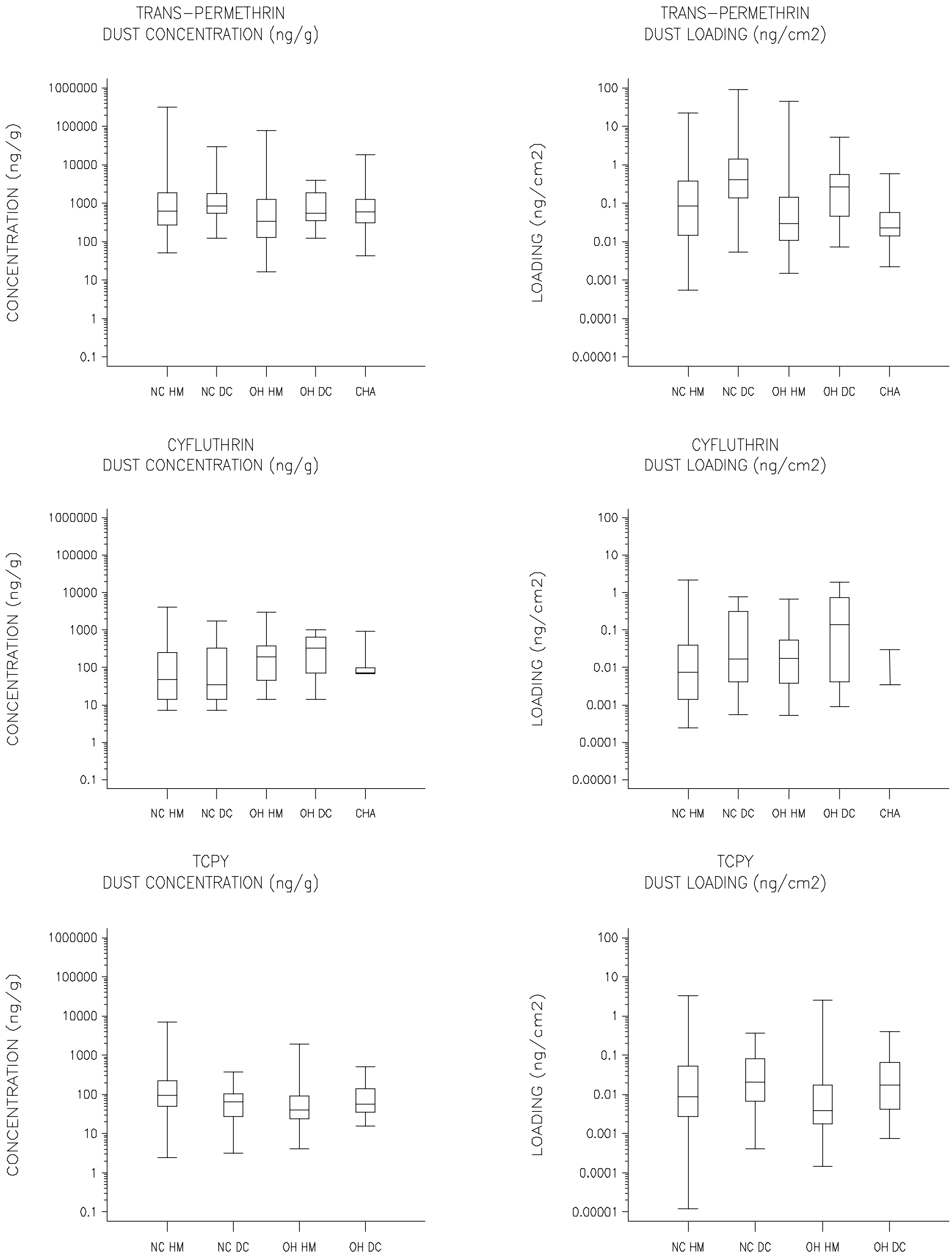

Figure 4.7 Box-and-whisker plots of dust concentrations (ng/g) and loadings (ng/cm²) for *trans*-permethrin, cyfluthrin, and TCPy.

4.3 Total Available Residue Measurements

Total available residue methods are intended to measure the total amount of contaminant on a surface. These methods involve either a solvent-assisted mechanical (wiping) action or the stationary capture of descending airborne droplets and particles. Total available residue loadings were measured in:

- NHEXAS-AZ using the LWW sampler,
- MNCPES using the LWW sampler,
- CCC from the floors and other surfaces (*e.g.*, counters, desktops) using surface wipes,
- JAX from the floor in the application area using surface wipes,
- CHAMOCOS using surface wipes,
- CPPAES using the LWW and deposition coupons,
- Test House using deposition coupons and surface wipes,
- DIYC using surface wipes, and
- Daycare using surface wipes.

The Lioy-Weisel-Wainman (LWW) sampler (Patent #RWJ-91-28) was developed to quantitatively measure dust on smooth surfaces and has been validated in laboratory and field tests (Lioy *et al.,* 1993; Freeman *et al.,* 1996). The LWW sampler achieves quantitative wipe collection using a movable constant pressure block within a template marking a specific area of 100 cm^2. Octadecyl-bonded (C18) disks that have been immersed in isopropyl alcohol are attached to a silicon rubber pad on the block. More details about this sampler can be found in Gurunathan *et al.* (1998) and Hore (2003).

Surface wipes are typically surgical dressing sponges wetted with isopropyl alcohol (IPA). The sponge is wiped multi-directionally through a defined area in an S-shaped configuration. Floor locations where young children may spend the most amount of time are usually selected. Residue loadings on irregularly shaped objects such as toys that are frequently handled by children (for estimating indirect ingestion exposures) are also measured using the wipe method.

Deposition coupons are used to estimate surface loadings of airborne and dust-bound residues that “settle out” of the air following an application (Ness, 1994). These consist of a sorptive material (*e.g.*, cotton, sponge, rayon) with a non-sorptive backing (aluminum foil) (Stout and Mason, 2003) and are placed in locations where the coupons will not be disturbed. Coupons may be repeatedly collected and replaced (interval) or collected only at the end of the sampling event (cumulative). Both interval and cumulative types were collected in CPPAES, whereas only interval deposition coupons were used in the Test House.

Pesticide Presence in Total Available Residues

Limits of detection for each chemical by study are given above in Table 4.2. Detection frequencies are given in Figure 4.8.

- The limits of detection varied widely among studies, but are similar within a study for both organophosphate and pyrethroid pesticides.
- Following dust methods, total available residue methods have the lowest limits for detection.
- Detection frequencies were slightly higher for the organophosphate pesticides in two of the three studies where both OP and pyrethroid pesticides were measured.
- Detection frequencies were higher in the smaller, focused studies than in the survey studies due to timing of the measurements with respect to recent applications.

Total Available Residues: Summary Findings

Surface loadings for the median and 95^{th} percentile are listed in Table 4.4 for all of the pesticides that were detected across studies (complete summary statistics are listed in Tables A.20 through A.24 in Appendix A). Lognormal probability plots are presented in Figure 4.9 for the most frequently detected pesticides which include chlorpyrifos, diazinon, *cis*- and *trans*-permethrin, cyfluthrin, and cypermethrin. The MNCPES data are not included because of the comparatively high detection limit and low detection frequencies. Box and whisker plots that graphically depict the total available residue loading results from all studies are given in Figure 4.10.

- In wipe samples, permethrin levels reported at the 95^{th} percentile were approximately an order of magnitude higher than chlorpyrifos and diazinon levels at the 95^{th} percentile (Table 4.4).
- Levels of diazinon and esfenvalerate reported at the 95^{th} percentile were at least an order of magnitude higher in studies with a known application (DIYC, Daycare) than in the survey studies (CCC, JAX-Screening).
- The lognormal probability plots (Figure 4.9) show that loadings of all frequently detected pesticides are substantially higher in the JAX screening wipe samples than in the CCC and CHAMACOS wipe samples.
- The total available residue distributions (Figure 4.9) of chlorpyrifos and *cis*- and *trans*-permethrin are relatively similar to each other within a specific large observational field study.
- Cypermethrin loadings tend to be the highest and diazinon loadings tend to be the lowest (Figure 4.9) of the pesticides of interest in the large observational field studies.
- The boxplots (Figure 4.10) reveal that chlorpyrifos, diazinon, and esfenvalerate loadings are substantially higher in those studies with a known application (CPPAES, Test House, DIYC, and Daycare).

- Low cyfluthrin loadings in wipe samples in Figure 4.9 (substantially lower than all other pesticide residues) suggest that cyfluthrin may not have been routinely used for pest treatment.

- MNCPES and CPPAES are the only studies that employed the LWW. The chlorpyrifos loadings measured in CPPAES were significantly higher (ANOVA, p=0.002, test results not presented) due to known pesticide applications coinciding with the sampling period.

- Although the MNCPES measurements did not coincide with a pesticide application, 62% of the LWW samples had detectable levels of chlorpyrifos, suggesting that chlorpyrifos remains on residential surfaces for a long period of time. It is unclear, however, how much of this is readily available for transfer and how much is freed from the pores and/or body material of the surfaces by the mechanical and solvent action of the LWW sampler.

- Mean post-application deposition coupon levels were significantly higher in the Test House than in CPPAES (ANOVA, p<0.0001, test results not presented). Factors responsible may include the following: three CPPAES homes received applications with only trace chlorpyrifos concentrations; the application performed in the Test House may have been more thorough than applications in the CPPAES homes; the Test House may have had a higher application of active ingredient per effective volume of the home (see Section 3.6), and some of the CPPAES occupants reported cleaning their homes and/or intentionally increasing ventilation after application, thereby reducing the amount of chlorpyrifos available for movement and capture on a deposition coupon.

- In studies (*e.g.*, CPPAES) where surface wipe samples were collected both pre- and post-application of a semi-volatile pesticide such as chlorpyrifos, the post-application pesticide loadings were higher than the pre-application values, including on surfaces that did not receive a direct application. This suggests that semi-volatile pesticides rapidly translocate from application surfaces to adjacent surfaces. We do not yet have information on the speed or extent of translocation for less volatile pesticides like pyrethroids.

- Two types of locations were sampled in JAX, the application area and a play area. In general, the surface residue loadings were higher at the application area than at the play area.

- The surface wipe samples collected in the CCC study were collected from two locations in each of the randomly selected rooms of the child care centers: a floor and desk top/table top surface. In general, the floor residue loadings were higher.

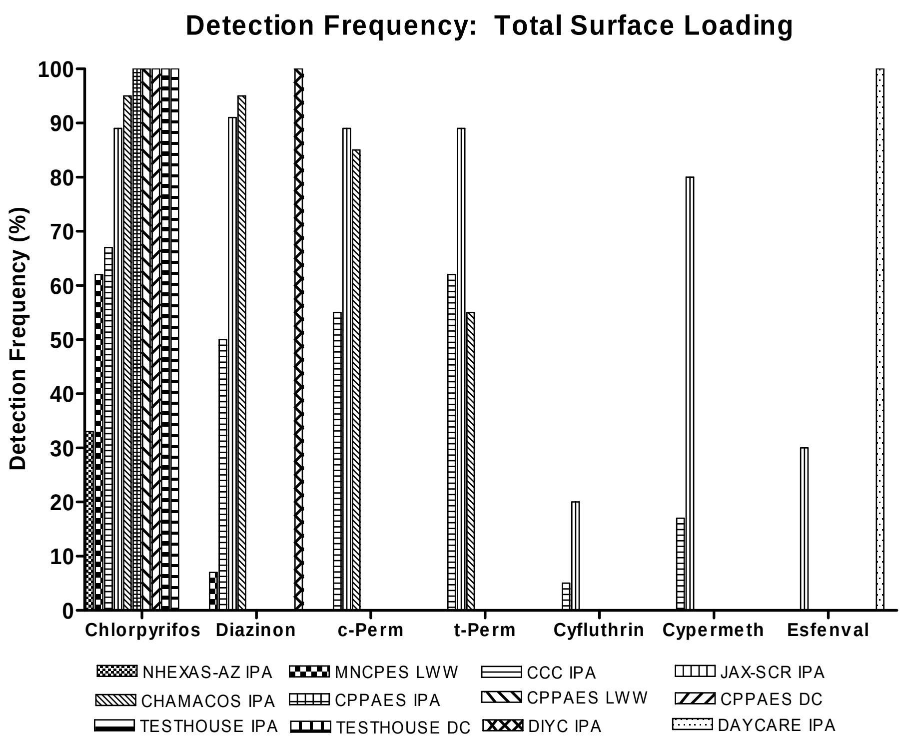

Figure 4.8 Detection frequencies for pesticides using total available residue collection methods.

Table 4.4 Median and 95th percentile values for total available residues (ng/cm²) by study.

Study	Method	Chlorpyrifos		Diazinon		*c*-Permethrin		*t*-Permethrin		Cyfluthrin		Cypermethrin		Esfenvalerate	
		P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
NHEXAS-AZ	IPA Wipe	<0.07	7.5	<2.000	<2.0	--	--	--	--	--	--	--	--	--	--
MNCPES	LWW	1.20	1.5	<3.500	3.5	--	--	--	--	--	--	--	--	--	--
CCC	IPA Wipe	0.03	0.9	0.002	0.5	0.009	0.67	0.02	1.1	<0.006	0.08	<0.006	0.8		
JAX-SCR	IPA Wipe	0.53	10.0	0.110	3.3	2.200	32.00	2.90	40.0	<0.006	4.30	2.600	750.0	<0.008	3.5
JAX-AGG	IPA Wipe	0.10	3.1	<0.002	4.0	0.210	42.00	0.26	67.0	<0.006	10.00	--	--	--	--
CHAMACOS	IPA Wipe	0.05	0.2	0.040	0.1	0.100	1.70	0.20	3.6	<0.050	0.40	--	--	--	--
CPPAES Pre	LWW	0.17	1.3	--	--	--	--	--	--	--	--	--	--	--	--
CPPAES	LWW	0.61	10.0	--	--	--	--	--	--	--	--	--	--	--	--
CPPAES	IPA Wipe	0.03	0.2	--	--	--	--	--	--	--	--	--	--	--	--
CPPAES	Dep Coup	1.40	9.6	--	--	--	--	--	--	--	--	--	--	--	--
TESTHOUSE Pre	IPA Wipe	4.70	9.1	--	--	--	--	--	--	--	--	--	--	--	--
TESTHOUSE	IPA Wipe	11.00	36.0	--	--	--	--	--	--	--	--	--	--	--	--
TESTHOUSE	Dep Coup	3.20	62.0	--	--	--	--	--	--	--	--	--	--	--	--
DIYC Pre	IPA Wipe	--	--	3.8	21.0	--	--	--	--	--	--	--	--	--	--
DIYC	IPA Wipe	--	--	5.5	72.0	--	--	--	--	--	--	--	--	--	--
DAYCARE	IPA Wipe	--	--	--	--	--	--	--	--	--	--	--	--	3.200	51.0

--, pesticide not measured

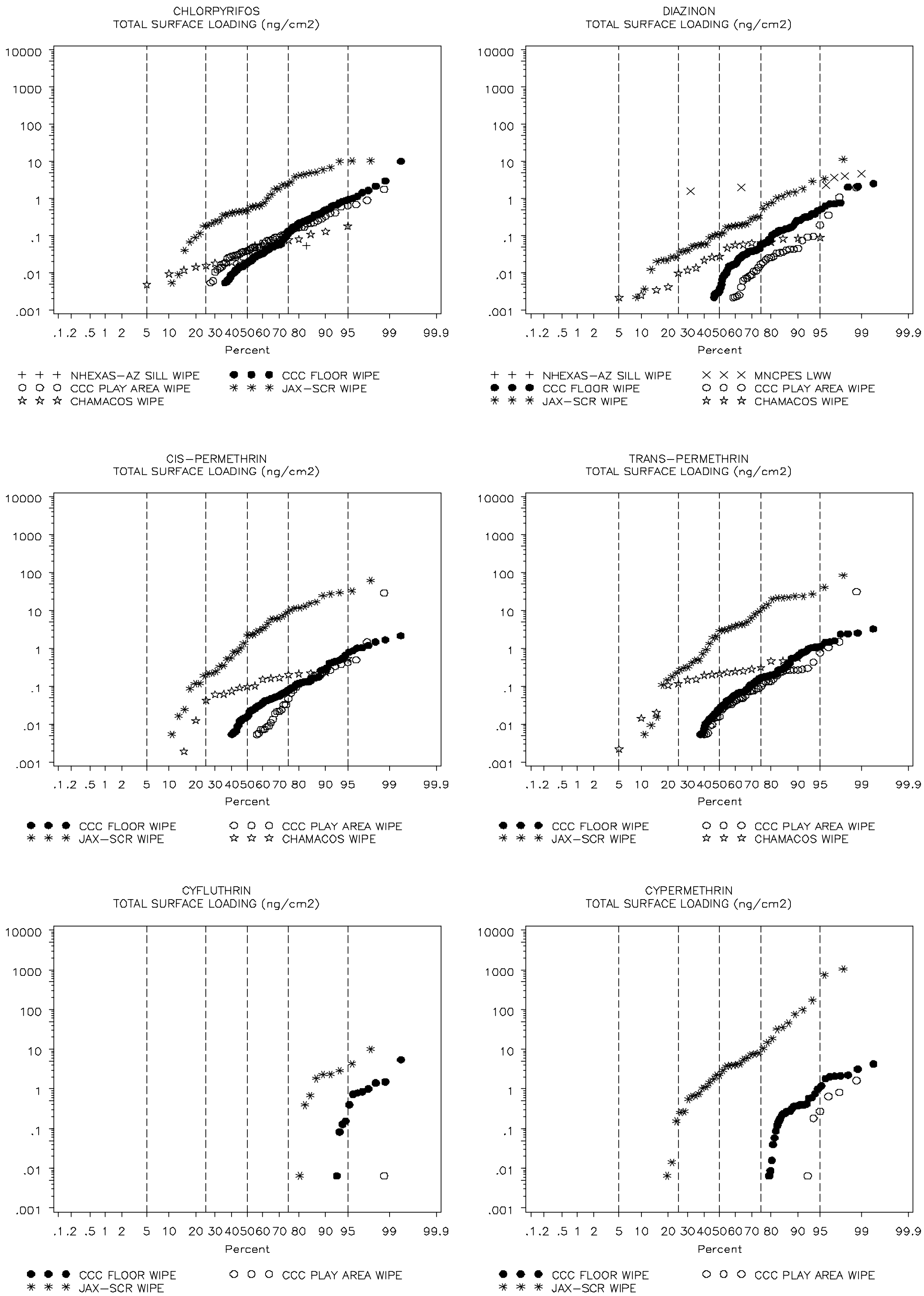

Figure 4.9 Lognormal probability plots for the most frequently detected pesticides which include chlorpyrifos, diazinon, *cis*- and *trans*-permethrin, cyfluthrin, and cypermethrin.

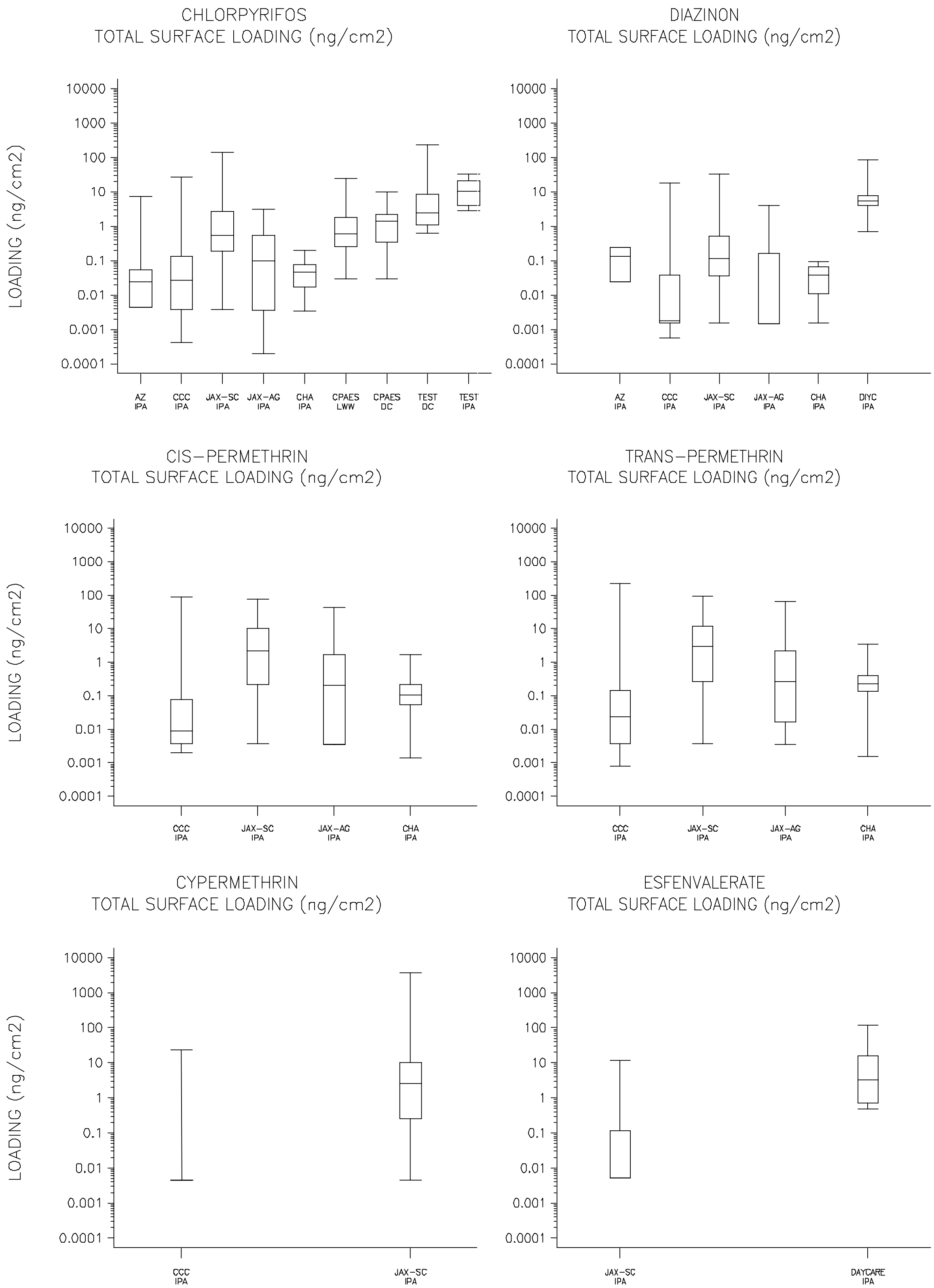

Figure 4.10 Box-and-whisker plots of total available residue surface loadings (ng/cm²) for chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin, cypermethrin, and esfenvalerate.

4.4 Transferable Residue Measurements

Transferable residue methods are intended to represent the surface loading that may be transferred as a result of contact with the contaminated surface; that is, instead of complete removal, they are typically intended to mimic transfer to skin during a single dermal contact with a surface, where transfer is aided by only saliva, sweat, or the sebum layer on the skin. Transferable residue loadings were measured in:

- MNCPES using the C18 press sampler on floors and non-floor surfaces,
- CTEPP using surface wipes with 2 mL 75% IPA on hard-surface floors and counters and a PUF roller on carpeted floors,
- CCC using the C18 press sampler on carpeted floors,
- JAX using the C18 press sampler on carpeted floors,
- CHAMACOS using the C18 press sampler on carpeted floors,
- Test House using the C18 press sampler and a PUF roller skin on carpeted floors,
- DIYC using the PUF roller on both hard-surface and carpeted floors, and
- Daycare using the C18 press sampler and the PUF roller on carpeted floors.

The Modified C18 Surface Press Sampler was based on the original EL Sampler designed by Edwards and Lioy to collect pesticides in house dust from carpeted floors (Edwards and Lioy, 1999; Hore, 2003). EPA modified the press sampler to use two 9-cm diameter sampling discs for a total sampling area of 114 cm^2 and eliminated the spring mechanism, henceforth it became known as the Modified C18 Surface Press Sampler. Unlike vacuum methods that collect household dust from all depths of the carpet pile and base, the surface press sampler is designed to only contact and remove residue from the surface. The developers maintain that the sampler replicates the collection efficiency of human skin and reflects transfer from single hand press (Edwards and Lioy, 1999; Lioy *et al.*, 2000), ignoring the inter- and intra-individual factors that may affect transfer.

The PUF roller transferable residue sampler was developed to simulate the pressure applied to a surface by a crawling child weighing 9 kg (7,300 Pa) (Hsu *et al.,* 1990). The PUF roller consists of a weighted roller fitted with a thick, moistened polyurethane foam (PUF) cover. Modifications include using either a dry PUF roller cover or a thinner PUF skin. More details can be found in the literature (Hsu *et al.,* 1990; Lewis *et al.*, 1994; Stout and Mason, 2003).

Discussion of the CTEPP surface wipe samples is included here rather than in Section 4.3 because of the small volume (only 2 mL) of isopropyl alcohol used. Also, it should be restated that in CTEPP transferable residue samples were only collected in those homes and daycare centers that reported recent pesticide use.

Limits of detection for each method and chemical are given by study above in Table 4.2. Detection frequencies are given in Figure 4.11. The C18 Press and PUF roller results from Daycare are not included (or further discussed) due to extremely poor detection frequencies, with only one C18 and two PUF samples above the limit of detection.

Pesticide Presence in Transferable Residues

- Overall, the detection frequencies for transferable residues were substantially lower than those for total available residues.
- Chlorpyrifos was detected in greater than 75% of transferable residues in all of the studies except MNCPES.
- *Cis*- and *trans*-permethrin were detected in greater than 50% of the transferable residue samples collected in CTEPP. These measurements were made in a subset of homes with recent indoor applications of unidentified pesticides.
- Transferable residues were rarely detected in field studies by the modified C18 surface press sampler. In CHAMACOS, the detection frequency for chlorpyrifos was zero. In MNCPES, the detection frequencies on the floor and on other surfaces were 8 and 5 percent, respectively. The only exception was the DIYC study, where the post-application detection frequency for diazinon was greater than 50%.
- The modified C18 press sampler was more successfully used in the laboratory studies (Test House and Food Transfer studies) where residues were measured on all surface types sampled.
- CTEPP used IPA wipes with only 2 mL isopropanol instead of the 10 to 20 mL often applied for total available residue measurements. It is likely that the amount of pesticide residue recovered from the sampled surfaces is influenced by the amount of IPA applied to the wipe. Other variables that should be considered include location sampled within the room and last known pesticide application.

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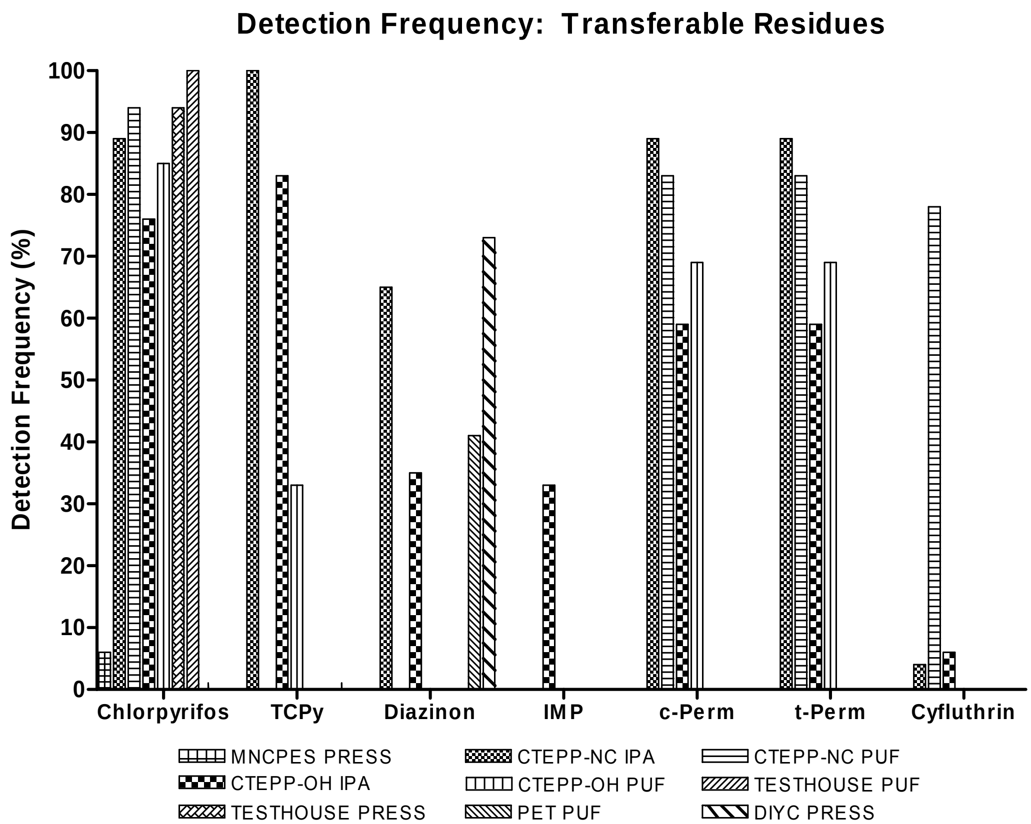

Figure 4.11 Detection frequencies for pesticides using transferable residue collection methods. All results from the C18 Press samplers used in CHAMACOS were below the limits of detection.

Transferable Residues: Summary Findings

Transferable residue loadings at the median and 95th percentile are given in Table 4.5 for all of the pesticides that were detected across studies (complete summary statistics are listed in Tables A.25 through A.29 in Appendix A). Transferable residue loadings of chlorpyrifos, diazinon, and permethrin are depicted in lognormal probability plots and box-and-whisker plots in Figures 4.12 and 4.13, respectively.

- The original C18 press sampler was designed to represent what adheres to the skin from a single hand press onto a carpeted surface. The uses for the modified C18 surface press sampler have expanded to include hard surfaces and longer contact times, contrary to its intended use. The data in Table 4.5 suggest that the sensitivity of the modified C18 surface press sampler is not adequate to measure typical residential pesticide residue levels due to its low collection efficiency (estimated as less than 1%).
- The mean transferable (2 mL IPA wipe) loadings were significantly different between CTEPP NC and OH for *cis*-permethrin ($p<0.01$), *trans*-permethrin ($p<0.05$), and diazinon ($p<0.01$). The mean loadings were not significantly different for either chlorpyrifos (ANOVA, $p=0.12$) or cyfluthrin (ANOVA, $p=0.17$).
- Wipe sampling methods varied in the volume of IPA used as a solvent (Table 4.1). The 2-mL IPA wipes used in CTEPP produced surface loading values that were very similar to those produced with the PUF roller (Figure 4.13). Since the PUF roller is a *transferable residue* method, it appears that the amount of IPA applied to the wipe determines the type of surface residue collected (*i.e.*, total or transferable residue). Interpretation of these results is complicated by other factors including recent application and sampling location with respect to application.

Table 4.5 Median and 95th percentile values for transferable residues (ng/cm²) by study.

Study	Method	Chlorpyrifos		Diazinon		*c*-Permethrin		*t*-Permethrin		Cyfluthrin		TCPy		IMP	
		P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
MNCPES	Press	<0.330	0.420	<0.140	1.13	--	--	--	--	--	--	--	--	--	--
CTEPP-NC h[a]	IPA Wipe	0.007	0.140	0.001	0.51	0.050	1.500	0.034	1.600	<0.007	<0.007	0.005	0.024		
CTEPP-OH h[a]	IPA Wipe	0.002	0.760	<0.001	0.05	0.005	0.780	0.005	0.790	<0.007	0.041	0.001	0.033	<0.001	0.007
TESTHOUSE	PUF	0.005	0.15	--	--	--	--	--	--	--	--	--	--	--	--
TESTHOUSE	Press	0.230	6.90	--	--	--	--	--	--	--	--	--	--	--	--
PET	PUF	--	--	<0.005		--	--	--	--	--	--	--	--	--	--
DIYC	Press	--	--	3.80	24.0	--	--	--	--	--	--	--	--	--	--

--, pesticide not measured
[a] Homes only (daycares excluded)

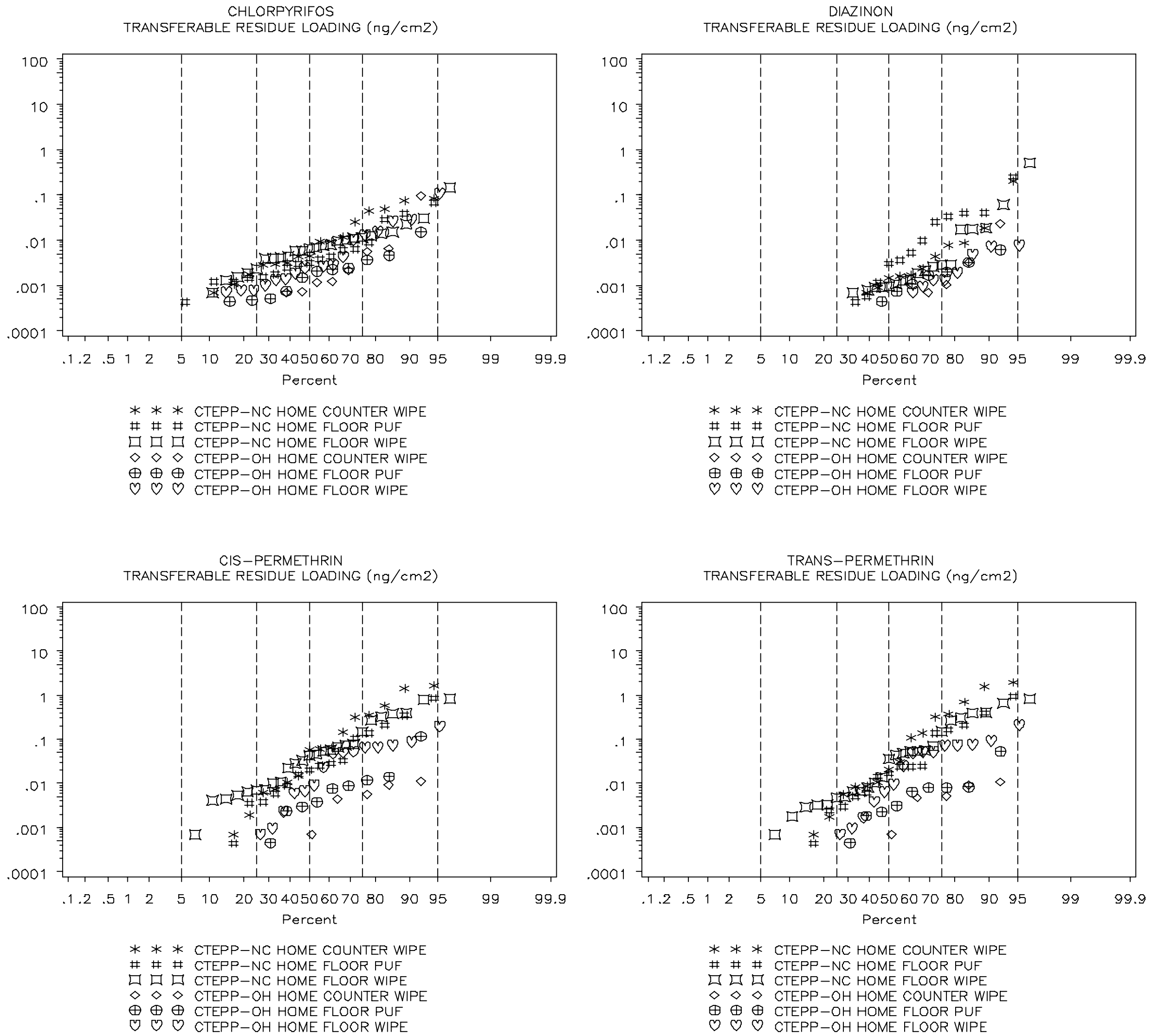

Figure 4.12 Lognormal probability plots for transferable residue loadings for the most frequently detected pesticides which include chlorpyrifos, diazinon, and *cis*- and *trans*-permethrin from CTEPP.

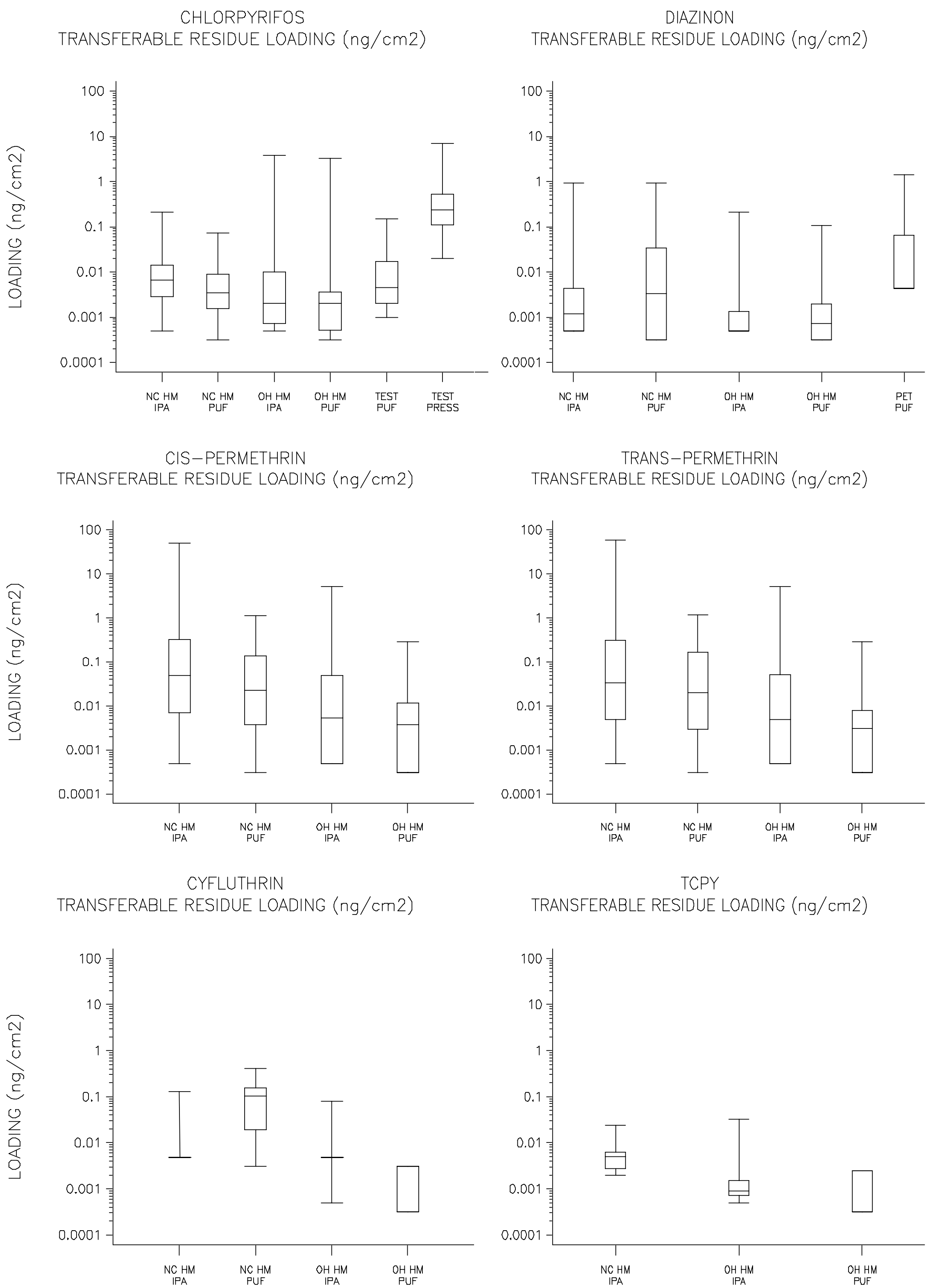

Figure 4.13 Box-and-whisker plots for transferable residue loadings for the most frequently detected pesticides which include chlorpyrifos, diazinon, *cis*- and *trans*-permethrin, cyfluthrin, and TCPy.

4.5 Spatial and Temporal Variability

Spatial and temporal variability were investigated in studies involving recent pesticide applications, including:

- Test House using IPA wipes, deposition coupons, C18 press sampler and PUF roller;
- CPPAES using IPA wipes, deposition coupons, and the LWW sampler;
- DIYC using IPA wipes and C18 press; and
- Daycare study using the IPA wipes.

In studies with a series of measurements over time, the interval of time between measurements ranged from one to three days. In CPPAES, multiple rooms in ten homes were monitored for two weeks post application. In DIYC, multiple surfaces in three homes were monitored for one week. In the Test House, multiple surfaces in multiple rooms of a single house were monitored for 21 days. The Daycare study included multiple applications, each separated by one to three months, in a single daycare facility. In addition to sampling main activity areas, some studies also sampled less frequently contacted areas.

Figure 4.14 presents total available surface residue loadings measured in multiple locations in multiple rooms over time in the Test House, in multiple rooms in ten homes in CPPAES, and on multiple surfaces in three homes in DIYC. Figure 4.15 presents transferable residue measurements over time in multiple rooms of the Test House and on multiple surfaces in three homes in DIYC. Figure 4.16 presents total available residue measurements from the Daycare study, collected immediately following applications on multiple surfaces in two rooms. Figure 4.17 presents spatial variability in deposition coupon loadings in the kitchen (application site) and den (adjoining room) of the Test House following pesticide application.

Spatial and Temporal Variability: Summary Findings

- Preliminary examination indicates that total available residue loadings decay at a slower rate than airborne concentrations (See Figures 4.14 and 3.8).
- In the Test House experiment, the transferable residue loadings appeared to decrease at a faster rate than the total available residues (Figures 4.14 and 4.15). This may have occurred because the pesticide residue became less available for transfer (for example, due to an interaction with the surface or because the dried residue was less available for transfer).
- The transferable residues on the counters in DIYC (Figure 4.15) are nearly as high as those on the floors immediately after application, suggesting translocation of the pesticide from the site of application (assuming counters were not application surfaces).
- Substantial variability within rooms (at times a 100-fold difference in loadings) is evident in the Daycare data (Figure 4.16). Exposure estimates using measurements at a single location based on an assumption of homogenous surface loadings may result in exposure misclassification. The spatial variability points to the need for sampling of multiple locations and perhaps for better resolution in the activity data that is gathered.

- Data from the Test House (Figure 4.17) show that surface loadings cannot be assumed to be homogenous within a room.
- In the CCC study, loadings on floors were generally higher than loadings on table tops.
- In a published analysis of the MNCPES LWW wipe data, Lioy and colleagues (2000) reported substantial variability in surface chlorpyrifos levels among different rooms.

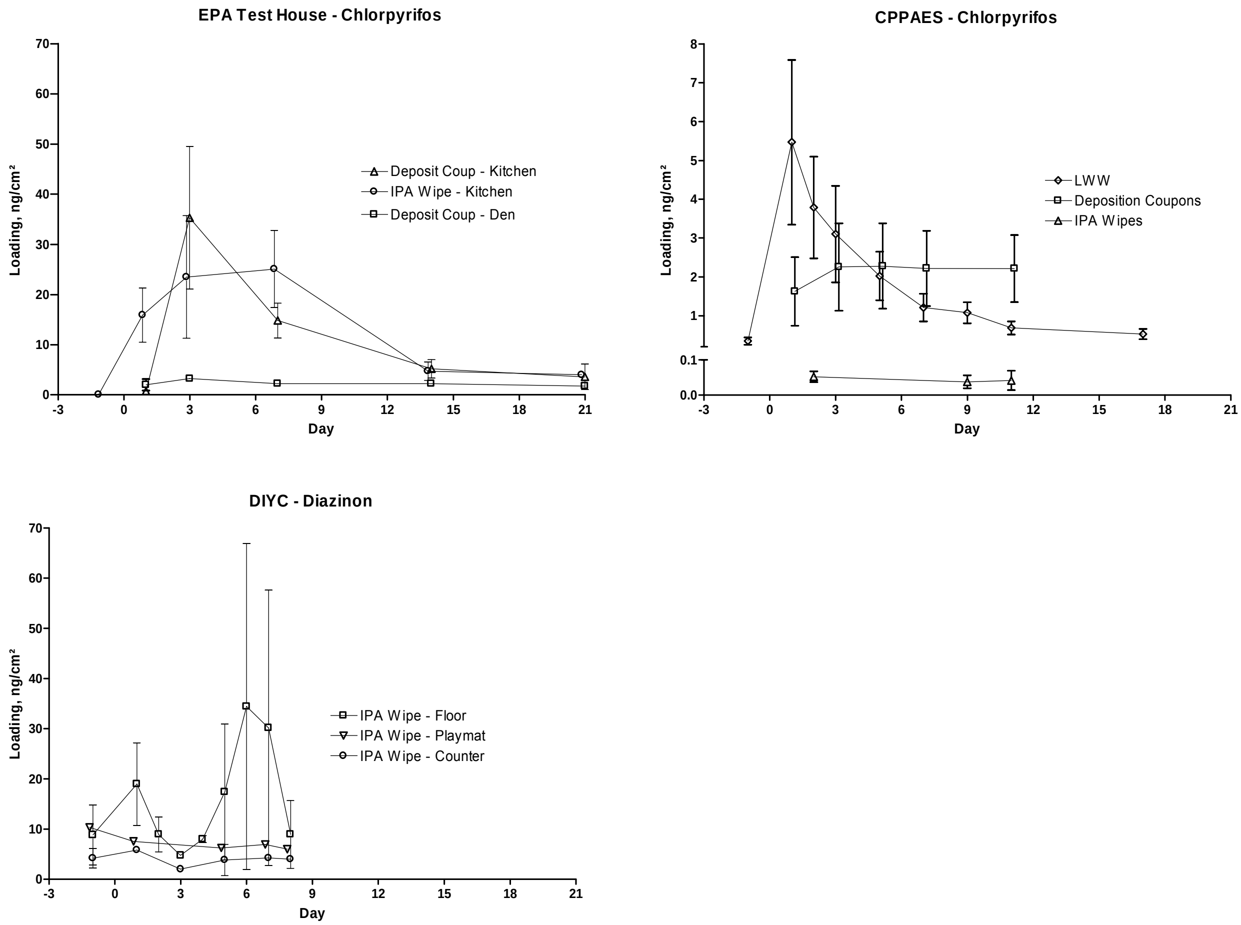

Figure 4.14 Total available surface residue loadings measured in multiple rooms over time in the Test House, in multiple rooms in ten homes in CPPAES, and on multiple surfaces in three homes in DIYC.

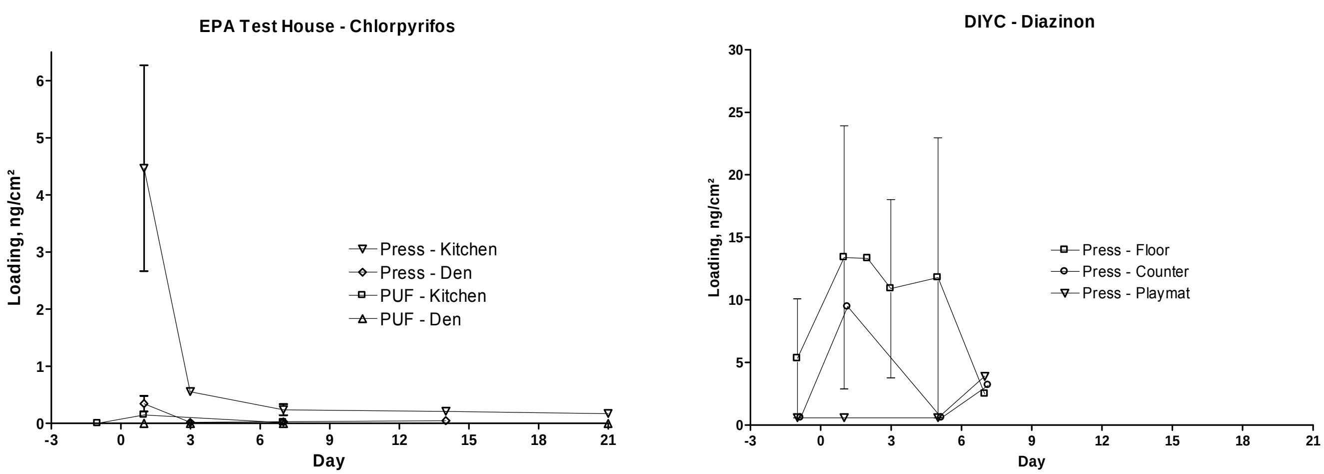

Figure 4.15 Transferable residue measurements over time following an application from multiple locations in multiple rooms of the Test House and multiple surfaces in three homes in DIYC.

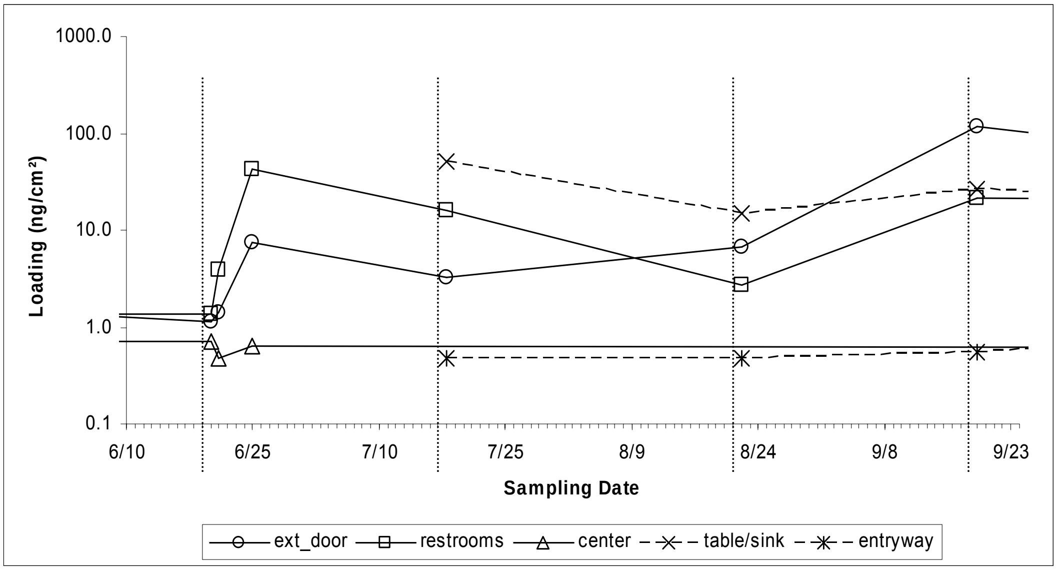

Figure 4.16 Total available residue measurements from the Daycare study, collected immediately following applications on multiple surfaces in two rooms in a single daycare facility. Solid Line represents the preschool room and dashed line represents infant room
Dotted vertical line represents application.

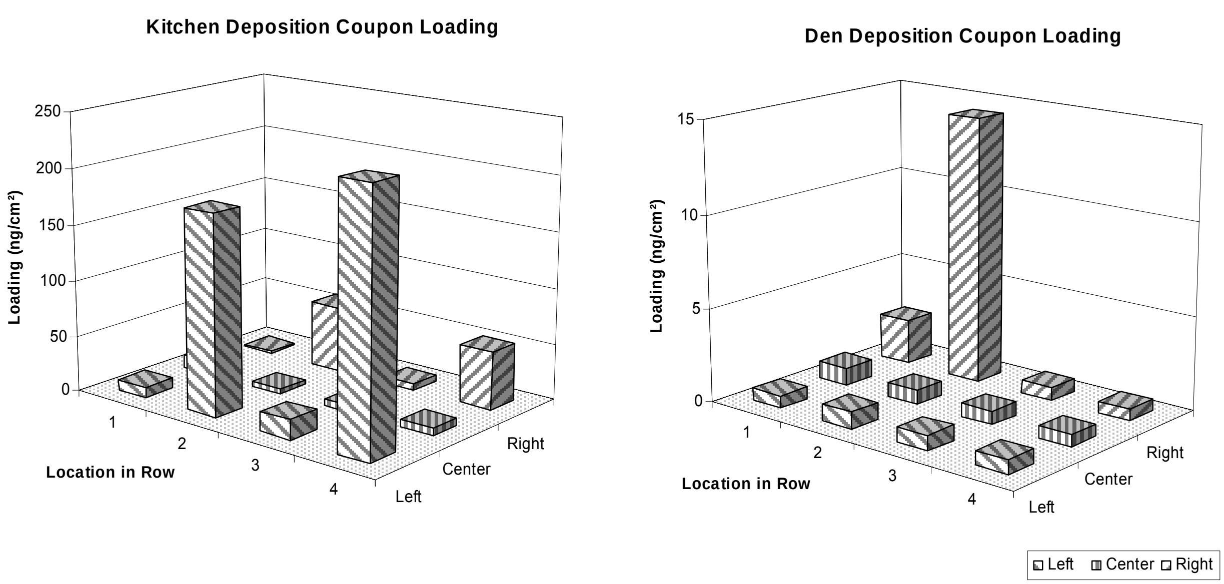

Figure 4.17 Spatial variability in deposition coupon loadings in the kitchen (application site) and den (adjoining room) of Test House following pesticide application.

4.6 Differences Related to Location

Regional Differences

Studies dating back to the Non-Occupational Pesticide Exposure Study (NOPES) from 1986 to 1988 (Whitmore *et al.,* 1994) have reported regional differences in environmental pesticide concentrations and loadings. Differences are thought to result from heavier use of insecticides in warm weather climates with higher year round insect control problems than in colder regions where hard winters help to curb insect populations.

- Median diazinon surface dust loadings (ng/cm^2) in home environments (daycares excluded) were very similar (about 0.002 ng/cm^2) across three states (NC, OH, and AZ, Table 4.3), and the 95th percentiles were also somewhat similar (0.12, 0.31, and 0.18, respectively). ANOVA analysis with Bonferroni adjustment for multiple comparisons found no significant differences among the three locations. These dust measurements do not provide evidence of the geographic variations consistent with geographic differences in pest treatment practices reported by Colt (1998).
- The overlapping distributions of pesticide concentrations in dust (ng/g) in the large observational field studies in Arizona, North Carolina, and Ohio (Figure 4.4) suggest that concentrations in dust may not be useful for determining region-specific pesticide use.
- For transferable residues obtained with 2-mL IPA surface wipes, the mean chlorpyrifos and cyfluthrin loadings were higher for CTEPP-NC compared to CTEPP-OH but not statistically different (Figures 4.12, 4.13). However, the mean loadings were significantly higher in NC for *cis*-permethrin (ANOVA; $p<0.01$) and *trans*-permethrin (ANOVA; $p<0.05$) and marginally significant for diazinon (ANOVA; $p<0.10$).
- Analysis of surface wipe samples from the national, probability-based Child Care Center study indicated no differences in the mean pesticide loadings among daycares in the four Census regions (data not shown, Tulve *et al.*, 2006).
- Differences in surface sampling methods, year of the study, and time of year when samples were collected make it difficult to examine any regional differences in surface pesticide loadings in homes. The transferable residue measurements suggest higher levels in NC than in OH, but no systematic differences are evident in dust concentrations or total surface residue loadings, although JAX had much higher surface loadings than any of the other studies without recent applications.

Urban vs. Rural

Lu and colleagues (2004) recently reported that at least one organophosphate pesticide was present in the house dust of 75% of agricultural area homes but only 7% of metropolitan area homes, suggesting different exposure pathways for children living in agricultural and nonagricultural regions. While concerns about pesticides may be more obvious in farming and other rural areas, widespread elevated pesticide residue levels have also been reported in highly urbanized minority communities of New York City (Whyatt *et al.,* 2002).

- Neither the median nor 95th percentile concentrations of chlorpyrifos measured in CHAMACOS dust was substantially higher than the median and 95th percentile in the other studies (Table 4.3). The assumption that children living in agricultural areas experience higher exposures than children in nonagricultural regions is not supported by these chlorpyrifos in dust measurements.
- Relatively high pre-application surface loadings in some of the CPPAES homes (data not presented) suggest possible contamination from pesticides applied in neighboring apartments in close proximity (Hore, 2003). Alternatively, the high loadings may suggest frequent treatments in those homes.

4.7 Influential Factors

As discussed above, the following factors appear to influence measured surface concentration or loading values:

Collection Methods

- The different types of collection methods are intended to have different collection efficiencies to serve different purposes. Efficiencies for various methods have been previously published.
- Total residue methods (which use both solvent and mechanical action to remove residues that may have penetrated into the surface) produce the highest values, followed by dust methods, and then by transferable residue methods.
- The low pesticide surface loadings obtained with 2 mL IPA wipes in both the NC and OH CTEPP studies (comparable to loadings obtained with the PUF roller) suggest that the amount of IPA applied to the wipe affects the amount of pesticide residue recovered.
- The C18 Press does not appear to be useful for determining typical surface pesticide residue loadings, for which it was never intended, because of its low collection efficiency and small size.

Surface Types

- Surface type has been shown to affect the collection efficiency of wipes. Recently published NERL data (Rohrer *et al.*, 2003) found that wiping from hard surfaces greatly exceeded carpet, and tile generally exceeded hardwood. As stated by Rohrer, “Highest pesticide recoveries were from tile with diazinon (59%), chlorpyrifos (80%), and permethrins (52% *cis*; 53% *trans*) being the only pesticides recovered by wiping at greater than 50% of the applied concentrations.”

Sampling Locations

- Despite evidence of translocation from direct application areas, the application area surface residue loadings were generally higher than the play area surface residue loadings in JAX.

- In the CCC study, floor residue loadings were typically higher than table top or desk top loadings.
- Experiments in the Test House showed high spatial variability in loadings in the room of application (kitchen) and transport of pesticide residues to the adjoining room.
- Results from the Daycare study showed substantial differences in surface loadings (up to two orders of magnitude) at different locations in a daycare center.

Occupant Activities

- Surface chlorpyrifos loadings were reportedly lower in the CPPAES homes in which the occupants performed cleaning activities and/or the homes that had high ventilation rates (Hore, 2003).
- Crack and crevice applications in the unoccupied Test House produced higher surface loadings and longer decay times than the same type of application (albeit with less active ingredient released) in the occupied CPPAES homes.

Pesticide Use Patterns

- On a regional level, surface loadings in Jacksonville, Florida, an area likely to have year-round pest control issues and high pesticide usage, were much higher than in any of the other observational studies.
- Within a given region, however, pesticide use information collected with questionnaires or inventories may not correlate with measured surface values. Published results from the MNCPES indicate that the residential pesticide use questions and overall screening approach used in the MNCPES were ineffective for identifying households with higher levels of individual target pesticides (Sexton *et al.*, 2003).

4.8 Correlations among Soil, Wipes, and Dust

- Analysis of CCC data (Tulve *et al.*, 2006) found little correlation between surface wipe loadings and soil concentrations for 16 common organophosphate and pyrethroid pesticides.
- In the CTEPP study, significant Spearman correlations between dust and soil concentrations were observed with diazinon ($r=0.26$, $p<0.01$) and TCPy ($r=0.21$, $p<0.05$) in NC homes and chlorpyrifos ($r=0.28$, $p<0.01$) and TCPy ($r=0.20$, $p<0.05$) in OH homes (data not presented).
- Identification of correlations is hindered by the low detection frequencies for many pesticides in soil.

4.9 Particle-Bound Pyrethroid Residues: Implications toward Exposure

The recent shift in commonly applied residential pesticides from organophosphate to pyrethroid compounds carries with it important implications for human exposure. The chemical and physical properties of a pesticide govern its behavior with respect to movement and fate. In general, pyrethroids have properties that favor the particulate phase, resulting in transport mechanisms preferentially involving dust rather than vapor. A tendency towards the particulate phase also suggests a decreased relative importance of the inhalation route and an increased relative importance of the dermal and indirect ingestion routes.

Pesticides applied in homes translocate from the point of application and deposit onto non-target surfaces. Because human contact with target surfaces (*e.g.*, cracks and crevices) is typically obstructed or otherwise hindered, it is largely the movement of residues from the point of application into the air and onto non-target surfaces that results in exposure. The movement of residentially applied insecticides follows a complex and poorly understood process of transformation and phase distribution and is influenced by several factors, namely: delivery system, application surface type, solvent, formulation, physicochemical properties of the active insecticide, and human and companion animal activity.

Overall, pyrethroids have similar physicochemical properties, and as a result, they display similar behavior in the residential environment (Laskowski, 2002; Oros and Werner, 2005). Pyrethroids generally have low vapor pressures and Henry's Law constants, thus they resist volatilization and exist almost entirely in the particulate phase at room temperature. They have high octanol/water partition coefficients (K_{ow}), which suggests they tend to partition into lipids, and very high water/organic carbon partition coefficients (K_{oc}), which suggests that they also tend to partition into organic matter. With these characteristics, pyrethroids can be expected to bind readily to the particulate matter that comprises house dust. Particles resuspended by human activity then act as the primary vector for pyrethroid transport and for human exposure.

Particle-phase contaminant transfer is strongly particle size dependent (Rodes *et al.*, 2001). Kissel *et al.* (1996) reported that dermal adherence of dry soil primarily involves particles in the <150 µm size fraction. Assuming that house dust behaves similarly with respect to transfer, the size fraction that preferentially adheres to skin not only comprises the bulk of house dust, but also contains the highest pesticide concentrations. Rodes *et al*. (2001) reported that the <150 µm size fraction comprises about 60% of house dust. Pesticide concentrations in house dust increase with decreasing particle size, and are highest in the <25 µm size fraction (Lewis *et al.,* 1999). Because the surface-to-volume ratio similarly increases with decreasing particle size, pesticides appear to be primarily attached to the surfaces of the particles (rather than trapped within).

Particle-bound movement and transfer of pyrethroids imply a decreased importance of the inhalation route and an increased importance of the indirect ingestion route. Exposure of young children, for whom indirect ingestion of residues from object- and hand-to-mouth activities is particularly important, may be most strongly affected. Particle-bound residues may also have a reduced potential for dermal absorption, as a consequence of being bound to the particle.