5.0 DIETARY EXPOSURE MEASUREMENTS

5.1 Introduction and Data Availability

Diet can be a significant pathway of exposure to humans. Infants and young children may be particularly vulnerable to exposure by dietary ingestion because they eat more than adults do relative to their body weights. Foods may contain residues of pesticides because of intentional agricultural applications or they may become contaminated during processing, distribution, storage, preparation, and even consumption. The ingestion of residues on foods resulting from contact with hands and surfaces during consumption as well as the ingestion of pesticide residues while mouthing contaminated hands and objects are considered "indirect ingestion" pathways and are the subject of the next chapter (Chapter 6.0). This chapter provides a comparative summary of measurements of pesticides in duplicate diet samples and of estimated dietary intakes. The sample collection methods for the studies that included duplicate diet measurements are summarized in Table 5.1.

Among the large observational studies, duplicate diet samples were collected in NHEXAS-AZ, MNCPES, and CTEPP. In CTEPP, food and beverage samples were collected at both homes and daycares. Duplicate diet samples were also collected in three pilot-scale studies, CHAMACOS (20 participants), DIYC (three participants), and JAX (nine participants).

- The most common measure of dietary exposure was by composited duplicate diet analyses (Table 5.1). This approach reduces study costs compared to analyzing individual foods, but it increases the complexity of the sample analysis and produces higher method detection limits.
- Duplicate diet samples measure the pesticide residues in the children's foods after processing and preparation by the caregiver. The samples, therefore, may include residues from contaminated food handling surfaces in addition to the residues contained in the food products. However, duplicate diets fail to capture the additional intake of pesticides resulting from the child's activities before and during consumption, as discussed in Chapter 6.
- Duplicate plate samples were used for dietary measurements at the daycares in CTEPP. The distinction between a duplicate plate and a duplicate diet (with the latter accounting for uneaten foods) is typically more important for children than adults because significant quantities of food may be left uneaten.

Table 5.1 Dietary exposure sample collection methods for pesticides.

Study	Children Ages (years)	Sample Type	Collection after Indoor Pesticide Use	Mass Recorded	Collection Period	Sample Handling	Composite	Relevant Analytes
NHEXAS-AZ	6 - 12	Duplicate diet	No	No	24 hr	Liquid and solid food collected separately in polyethylene containers	Yes	Chlorpyrifos, diazinon
MNCPES	3 - 12	Duplicate diet	No	Yes	4 d	Liquid and solid food collected separately; solid food split into potentially "high pesticide" foods and "remaining" foods	Yes	Chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin
CTEPP	2 - 5	Duplicate diet (homes), and duplicate servings (at daycare centers)	No	Home samples only	48 hr	Liquid and solid food collected separately in glass jars	Yes	Chlorpyrifos, TCPy, diazinon, IMP (Ohio only)
JAX	4 - 6	Duplicate diet	Yes	Yes	24 hr	Solid and liquid food stored in polyethylene containers	Yes	Chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin, cyfluthrin
CHAMACOS	0.5 - 2	Duplicate Diet	No	Yes	24 hr	Liquid collected in polycarbonate bottles and solid food in polyethylene zip closure bags	Yes	Chlorpyrifos, diazinon, *cis*-permethrin, *trans*-permethrin, cyfluthrin
DIYC	1 - 3	Duplicate diet, each food collected individually	Yes	Yes	24 hr	Each food stored in individual zip-loc bags	No	Diazinon

5.2 Pesticide Presence

Table 5.2 presents the detection limits for the studies. The frequency of detection for the selected pesticides is presented in Figure 5.1. The median and 95th percentile concentrations are presented in Table 5.3. Data are presented in lognormal probability plots (Figures 5.2 and 5.3) for the large observational field studies and box-and-whisker plots (Figures 5.4 and 5.5) for all of the studies. Where food mass measurements are available (Table 5.1), both concentration and intake (mass of compound ingested) are presented. Intake is defined as µg/day in keeping with the dietary exposure algorithm of the *Draft Protocol* (Berry *et al.*, 2001) rather than as µg/kg-bw/day which would be more consistent with the reference dose (RfD) paradigm.

- Reported method detection limits for chlorpyrifos ranged from 0.04 µg/kg in JAX up to 1.7 µg/kg in CHAMACOS (Table 5.2).
- Chlorpyrifos was detected in over 50% of the duplicate diet samples in MNCPES, CTEPP, and JAX (Figure 5.1). The median chlorpyrifos concentrations in the MNCPES and JAX diet samples were at least twice as high as in the CTEPP samples (Table 5.3).
- Diazinon was not frequently detected in any of the studies except DIYC, a study in which there had been prior indoor applications. The data from DIYC suggest that contamination of food due to handling and surface contact is important in homes with recent applications (see Section 6).
- While detection of diazinon in food samples was typically below 30% (Figure 5.1), detection immediately following crack and crevice application in DIYC was 100%.
- The logplots (Figures 5.2 and 5.3) show that in the upper half of the distribution (between the 50th and the 95th percentiles), higher concentrations of *cis*- and *trans*-permethrin were measured in solid food in North Carolina homes than in North Carolina daycares or Ohio homes or daycares.
- Model simulations using DIYC data (results not presented) revealed that pesticides transferred to food during contact with surfaces and handling by a child may increase dietary intake significantly (over 60% under the modeled scenario).
- Published results from the MNCPES (Clayton *et al.,* 2003) showed that extant residue databases can successfully be used to select samples for analysis, potentially reducing costs by avoiding analyses of foods not likely to contain measurable levels. Care must be taken, however, to avoid neglecting those residues that are transferred during handling.
- Measurable levels of these particular pesticides were rarely detected in beverages in any of these studies. Future studies with other such pesticides that are not expected to be found in drinking water may consider eliminating this costly measurement.
- Infants and children consume far fewer types of foods than do adults (while consuming much more of certain foods) (NRC, 1993). Thus, the number of days of collection may be less important for children than for adults.
- The large potential for enzymatic degradation of pesticides (especially chlorpyrifos) during food sample storage and during homogenation prior to analysis has not been directly addressed by any studies under this program.

Table 5.2 Limits of detection (µg/kg) for pesticides measured in duplicate diets.

Study	Compounds				
	Chlorpyrifos	Diazinon	*cis*-Permethrin	*trans*-Permethrin	Cyfluthrin
NHEXAS-AZ	1.0	0.7	--[a]	--	--
MNCPES	0.26	0.3	0.2	0.2	--
CTEPP	0.08	0.08	0.08	0.08	0.83
JAX	0.04	0.04	0.02	0.02	0.4
CHAMACOS	1.4	1.2	4.5	2.9	--
DIYC	--	0.36 – 1.25	--	--	--

[a] Blank cells (--) indicate that the pesticide was not measured in the study.

Table 5.3 Median and 95th percentile pesticide concentrations (µg/kg) measured in duplicate diet food samples.

Study	Chlorpyrifos		Diazinon		*cis*-Permethrin		*trans*-Permethrin		Cyfluthrin	
	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
NHEXAS-AZ	BDL[a]	5.7	1.8	1.9	--[b]	--	--	--	--	--
MNCPES	0.53	2.4	BDL	0.38	--	--	--	--	--	--
CTEPP-NC Home	0.2	2.1	BDL	0.4	BDL	15.6	BDL	8.7	BDL	0.9
CTEPP-NC Daycare	0.1	0.9	BDL	0.2	BDL	5.2	BDL	3.0	BDL	BDL
CTEPP-OH Home	0.2	1.6	BDL	0.2	BDL	8.8	BDL	8.0	BDL	BDL
CTEPP-OH Daycare	0.1	0.6	BDL	0.2	BDL	2.2	BDL	1.4	BDL	BDL
JAX	0.38	7.4	BDL	1.0	0.29	13	0.22	22	BDL	3.6
CHAMACOS	BDL	1.4	BDL	BDL	BDL	BDL	BDL	BDL	--	--
DIYC	--	--	0.17	0.78	--	--	--	--	--	--

[a] BDL, Below minimum detection limit
[b] Blank cells (--) indicate the pesticide was not measured in the study

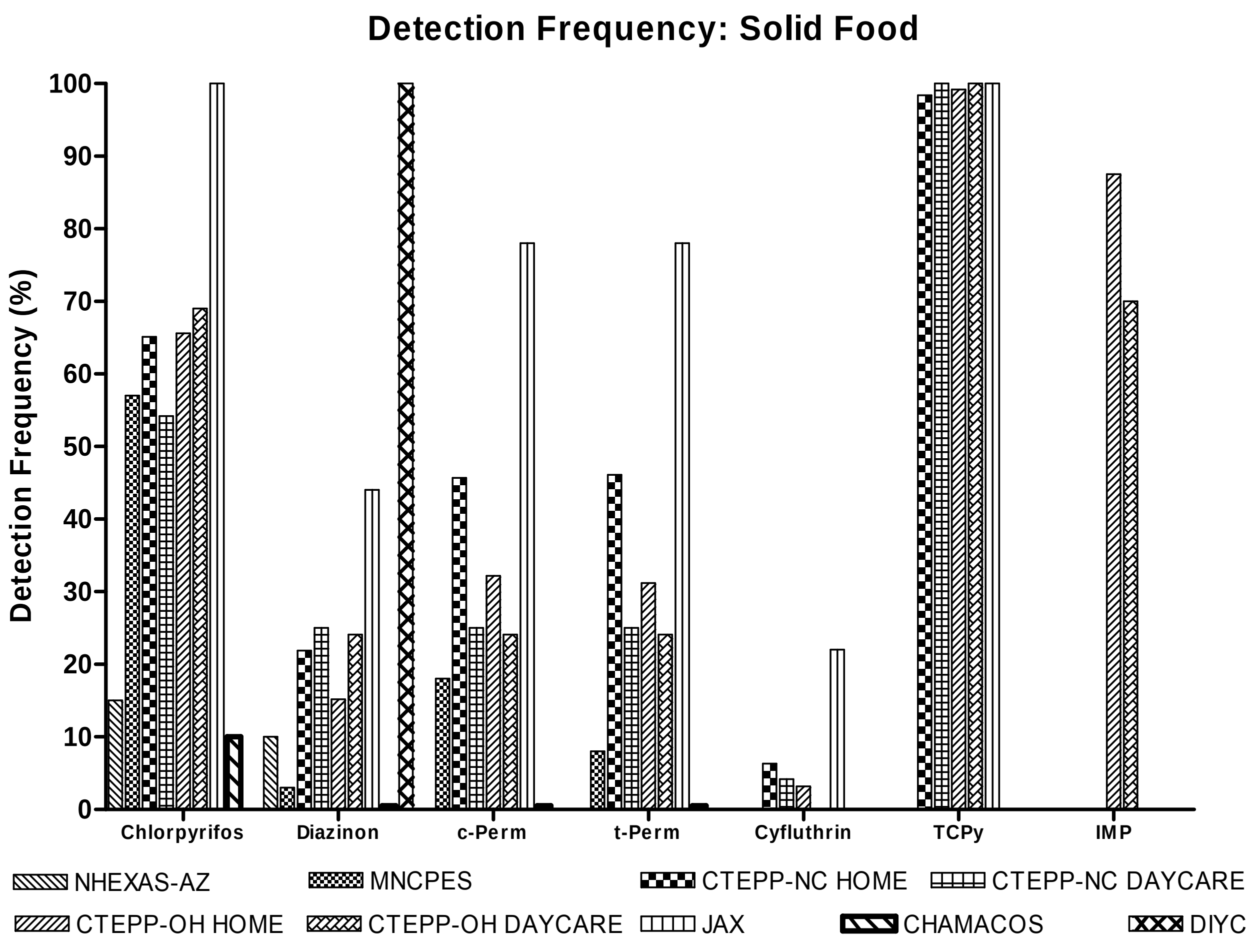

Figure 5.1 The detection frequency of pesticides measured in duplicate diet food samples.

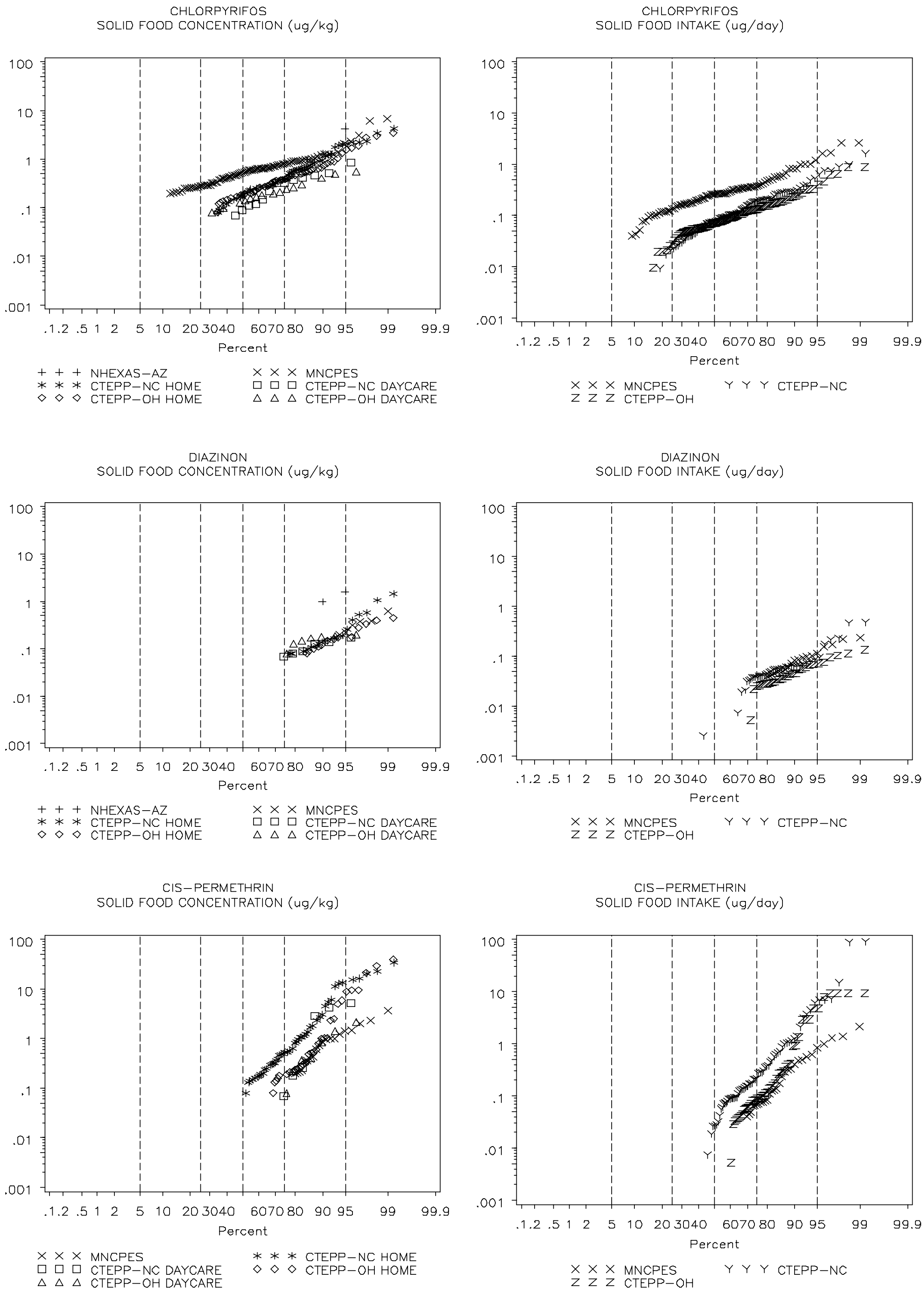

Figure 5.2 Lognormal probability plots of solid food concentrations (µg/kg) and intakes (µg/day) for chlorpyrifos, diazinon, and *cis*-permethrin from large observational field studies.

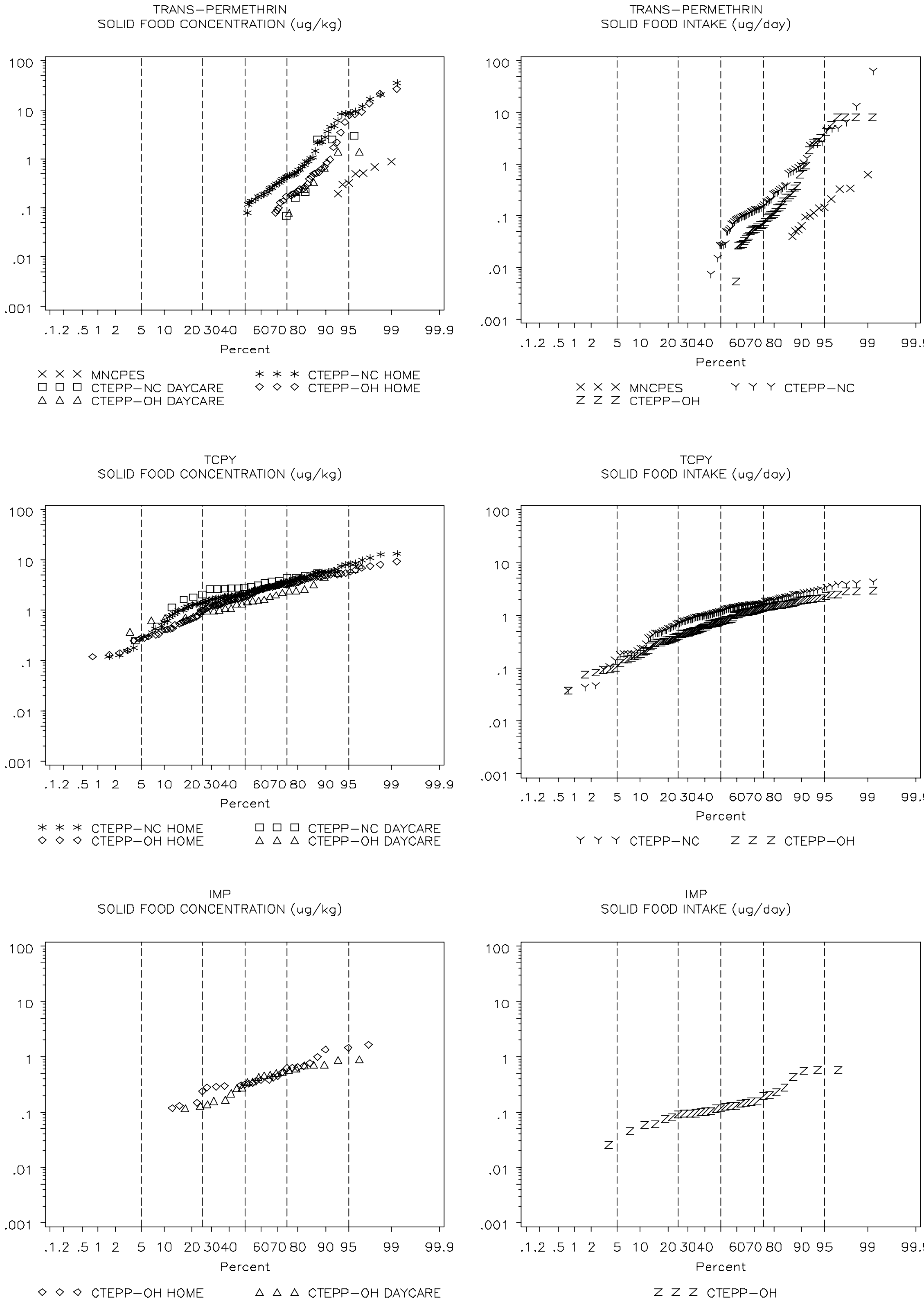

Figure 5.3 Lognormal probability plots of solid food concentrations (µg/kg) and intakes (µg/day) for *trans*-permethrin, TCPy, and IMP from large observational field studies.

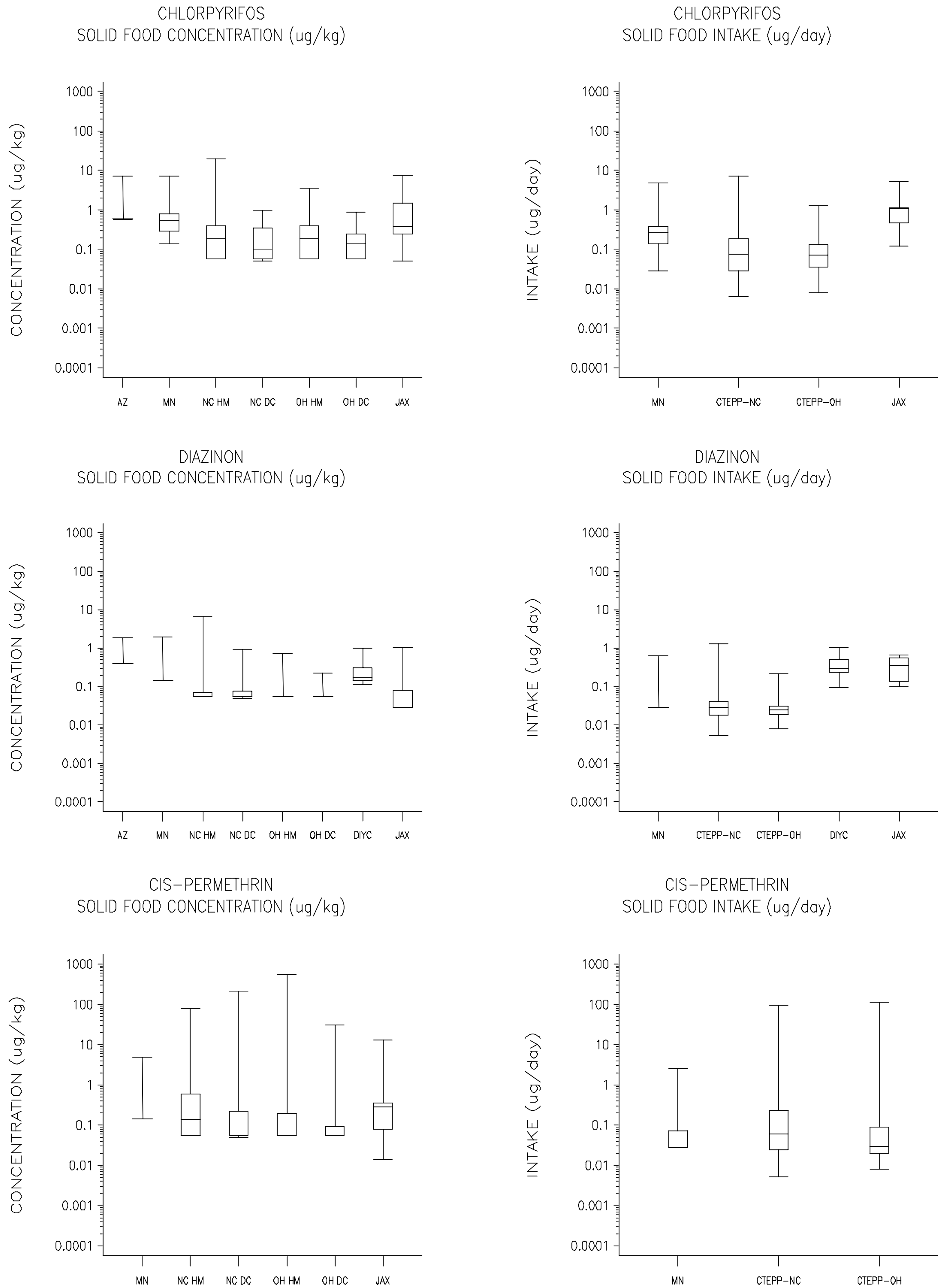

Figure 5.4 Box-and-whisker plots of solid food concentrations (µg/kg) and intakes (µg/day) for chlorpyrifos, diazinon, and *cis*-permethrin across all studies.

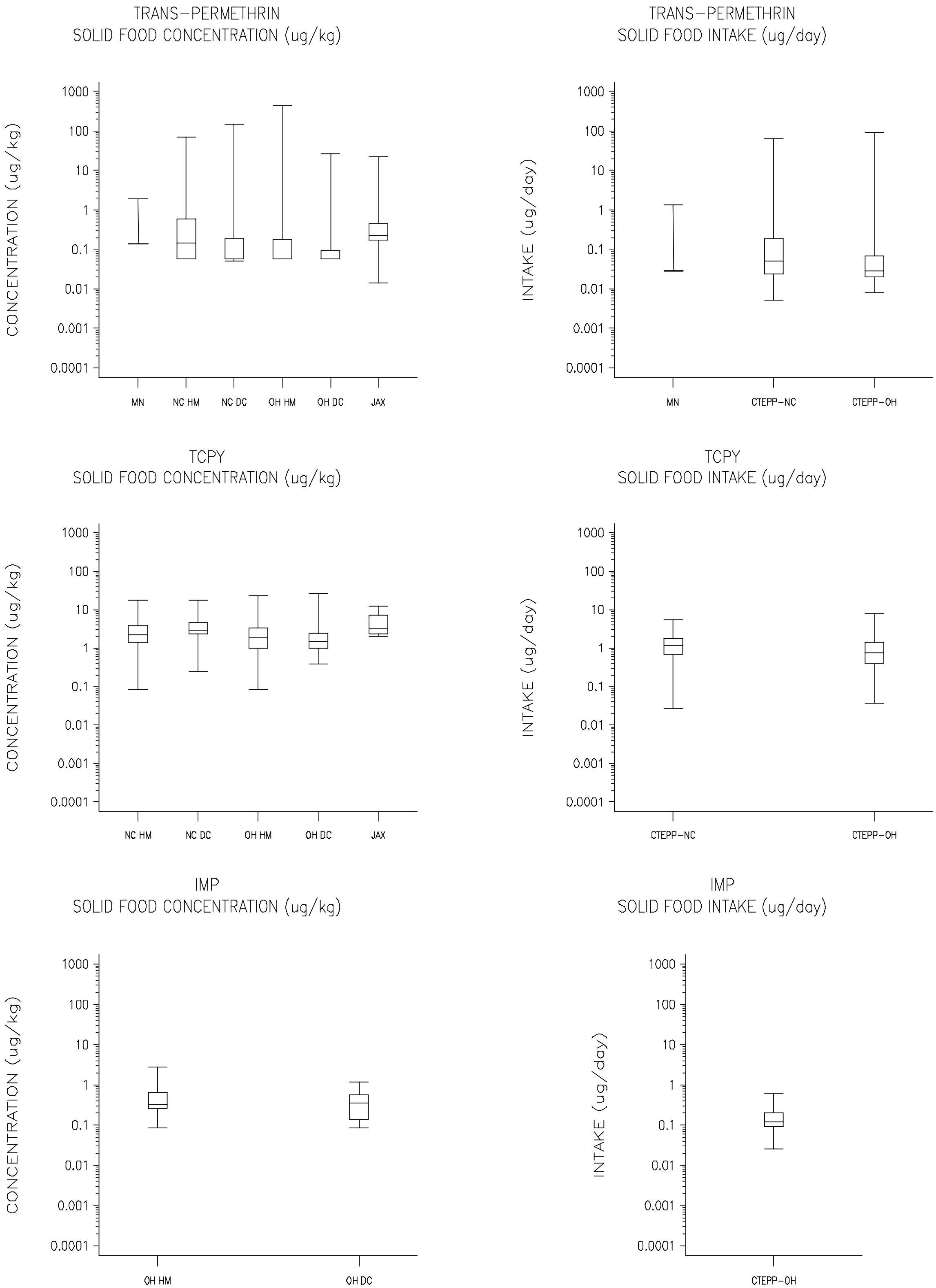

Figure 5.5 Box-and-whisker plots of solid food concentrations (µg/kg) and intakes (µg/day) for *trans*-permethrin, TCPy, and IMP across all studies.

5.3 Relative Importance of the Ingestion Route

The Stochastic Human Exposure and Dose Simulation (SHEDS) model (Zartarian *et al.*, 2000) prediction for dietary intake of *cis*-permethrin is compared to CTEPP measurements in Figure 5.6. The estimated proportion of aggregate exposure represented by dietary intake for CTEPP-NC and CTEPP-OH children is from the CTEPP Report (Morgan *et al.*, 2004) and is presented in Figures 5.6 and 5.7, respectively.

- An example of use of the SHEDS model to predict dietary intake of *cis*-permethrin in a study population is shown in Figure 5.6. The dietary intake estimates may then be compared to SHEDS model estimates of intake by other relevant routes to determine the relative importance of the ingestion route.
- Based on route-specific estimates (Figures 5.7 and 5.8), dietary ingestion represents the dominant route of exposure for chlorpyrifos, diazinon, and permethrin in the CTEPP study. Indirect ingestion, estimated based on dust and soil measurements, is a far greater concern for the permethrin than for chlorpyrifos and diazinon in the CTEPP study.
- The route that represents the dominant route of exposure (dietary ingestion) is also the route with the lowest detection frequencies (approximately 2/3 of the values for permethrin in CTEPP are nondetects), which increases the uncertainty in the estimates. Substituting a fraction of the detection limit for values below the limit of detection may have a disproportionate impact on the outcome.

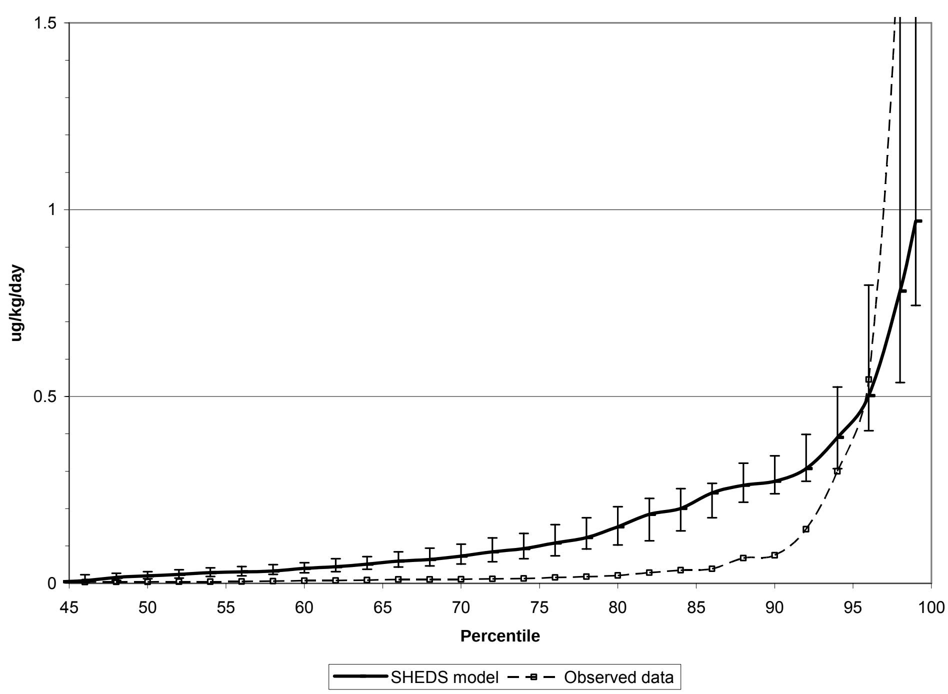

Figure 5.6 Comparison of SHEDS model prediction for dietary intake of *cis*-permethrin (μg/kg/day) and CTEPP measurement data.

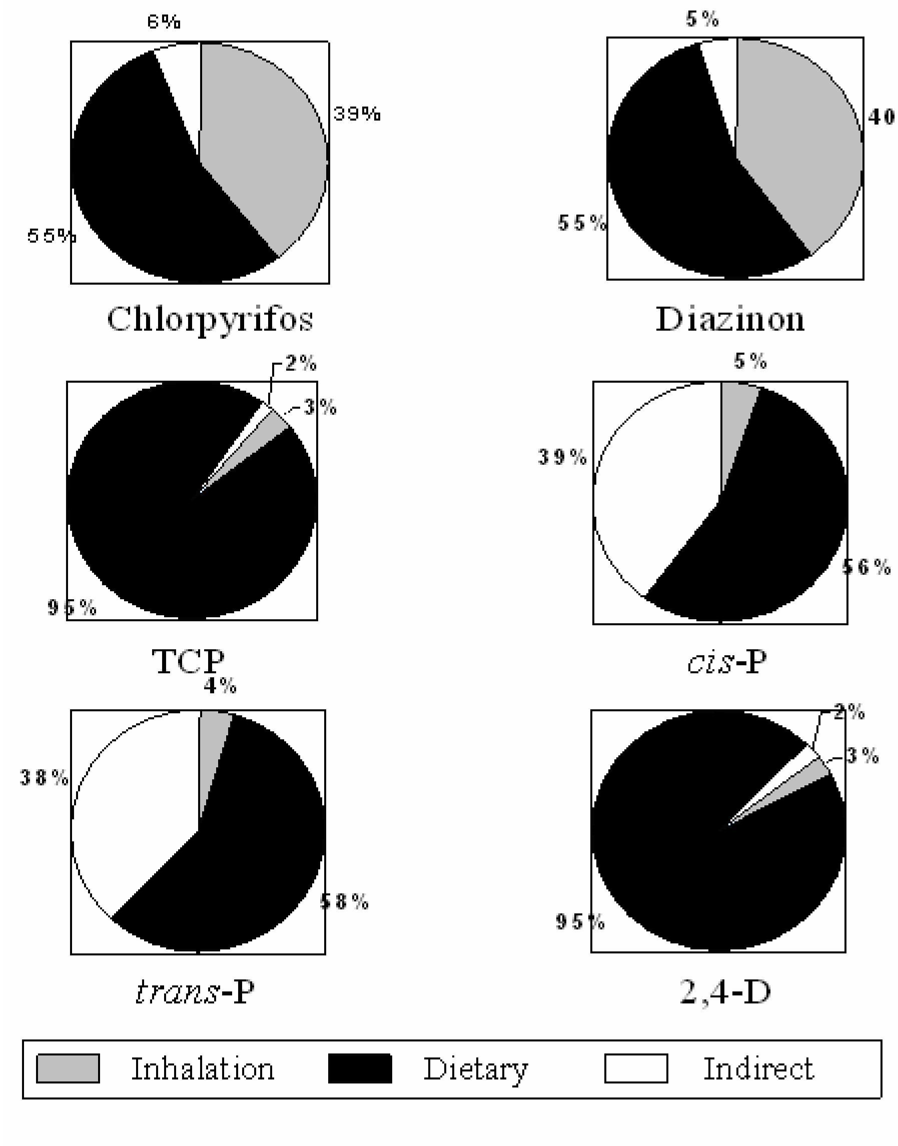

Figure 5.7 Estimated mean proportion of aggregate potential exposure for CTEPP-NC children by exposure route. (TCP = 3,5,6-Trichloro-2-pyridinol; *cis*-P and *trans*-P = *cis*- and *trans*-Permethrin; 2,4-D = 2,4-Dichlorophenoxyacetic acid.) From Morgan *et al*., 2004.

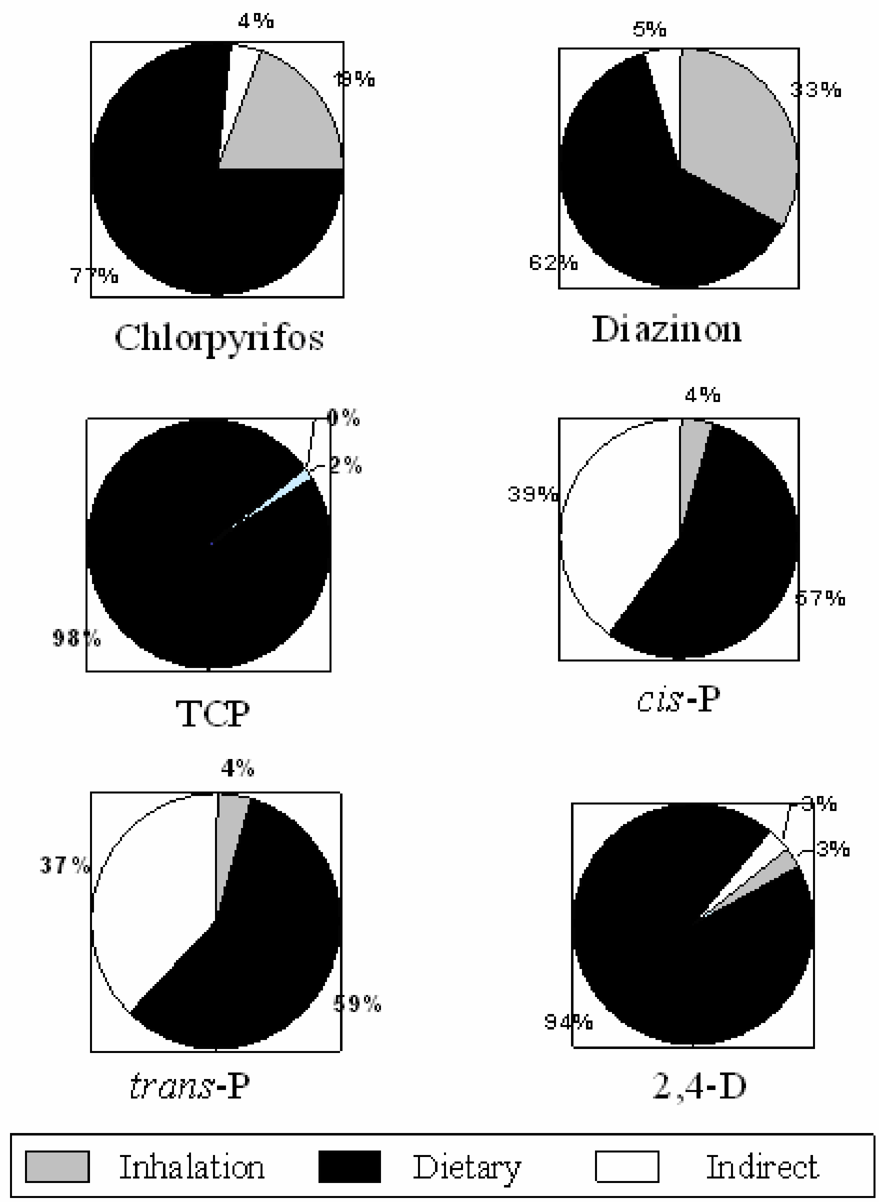

Figure 5.8 Estimated mean proportion of aggregated potential exposure for CTEPP-OH children by exposure route. (TCP = 3,5,6-Trichloro-2-pyridinol; *cis*-P and *trans*-P = *cis*- and *trans*-Permethrin; 2,4-D = 2,4-Dichlorophenoxyacetic acid.) From Morgan *et al.*, 2004.