

## 6.0 INDIRECT INGESTION MEASUREMENTS

Children's ingestion of pesticide residues is not limited to residues in food and beverages acquired during cultivation, food production, and in-home preparation. Indirect ingestion refers to the ingestion of residues from hands or objects that enter the mouth, as well as to the ingestion of residues transferred to food items by contact with the floor or other contaminated surfaces during consumption. Indirect ingestion is believed to be an important route of exposure for children because of their frequent mouthing activities and their unique handling of foods while eating. Indirect ingestion may be the result of hand-to-mouth, object-to-mouth, or hand-to-object-to-mouth activity. Indirect ingestion may be estimated using an approach that lumps some of the exposure factors and activity patterns associated with indirect ingestion. This simplified approach allows for assessment of indirect ingestion exposure based on measurement data collected in the field and on factors that characterize the activities that lead to indirect ingestion. In this approach, objects (including food) that are commonly handled, mouthed, and/or ingested are identified in the field. The residue loadings on these objects are measured directly or estimated from surface loading measurements combined with transfer efficiencies measured in the laboratory. General information relating to the frequency and nature of these mouthing and ingestion activities is also collected. Data on the fraction of residues that may be removed from an object during mouthing that has been collected in the laboratory is then required to complete the assessment. In addition, the items identified as most often mouthed and/or eaten are assumed to represent the most significant sources of indirect ingestion exposure. This section presents summary data for studies addressing the indirect ingestion route of exposure (Table 6.1). Highlights of the data are presented below.

### 6.1 Characterizing Hand- and Object-to-Mouth Activities

Exposure models are based on two factors: how much pesticide residue is available for human uptake and what human activities occur that would result in contact with and uptake of residues. Hand-to-mouth and object-to-mouth activities are believed to directly impact ingestion of pesticides among children through the indirect ingestion exposure route, but the relative importance of these activities has not been established. In fact, the lack of empirical data showing that either hand- or object-to-mouth activities appreciably affect exposure makes it a hypothesis that has not yet been adequately addressed. The frequency of hand-to-mouth, object-to-mouth, and/or combo-to-mouth contacts were quantified for children in the MNCPEs and CPPAES studies using a computer software system (Table 6.2). These studies used Virtual Timing Device (VTD) software (Zartarian *et al.*, 1997) to quantify the children's normal daily activities captured on videotape. The following are highlights of the data from these studies.

- Assigning contact as either a hand-to-mouth or an object-to-mouth contact can cause the hand-to-mouth and/or object-to-mouth contacts per hour to be underestimated. A combo-to-mouth category that accounts for both simultaneous types of contacts may provide a more accurate estimate of the indirect ingestion route of exposure.
- An average frequency of 9 hand-to-mouth contacts per hour among 2 to 5 year olds is recommended for regulatory risk assessments (US EPA, 2002). The CPPAES results suggest that a higher value may be appropriate (Table 6.3).

- Figure 6.1 presents the average frequency of hand- and object-to-mouth contacts during all eating and non-eating events. The highest hand-to-mouth frequency was observed in CPPAES.
- Factors affecting hand-to-mouth contact frequencies may include inclusion of eating events, amount of time on tape, types of activities, number of children, and age range.
- An analysis of hand-to-mouth activities in MNCPES has been published by Freeman *et al.* (2001). They reported that hand-to-mouth activities were significantly more frequent (t test,  $P < 0.05$ ) among girls than among boys.
- The MNCPES data also showed that hand-to-mouth and object-to-mouth activities were more frequent (Mann–Whitney,  $p < 0.05$ ) indoors than outdoors (Freeman *et al.*, 2001).
- Published studies have quantified the hand- and object-to-mouth activities of young children (Zartarian *et al.*, 1998; Reed *et al.*, 1999; Tulve *et al.*, 2002; Freeman *et al.*, 2005). These studies suggest that young children may exhibit higher hand-to-mouth and/or object-to-mouth contacts than older children and adults.
- Standardized approaches for quantifying the activity patterns of children are needed in order to compare results among different studies.

## 6.2 Residue Loadings on Mouthed Objects and Removal by Mouthing

For indirect ingestion estimates, objects that are commonly mouthed are identified in the field and the residue loadings on these objects are measured. Objects commonly mouthed by preschoolers were identified in CTEPP. Pesticide loadings on toy surfaces were measured in the CHAMACOS and CPPAES studies. Data on the fraction of residues that may be removed by mouthing of fingers was collected in the laboratory-based Transfer studies using non-toxic fluorescent surrogates.

- Objects commonly mouthed by preschoolers were identified in CTEPP. These items were typically toys and food-related items (Table 6.4).
- Chlorpyrifos loadings on toy surfaces were much higher following recent applications, as evidenced by the higher values in CPPAES than in CHAMACOS (Table 6.5). Loading on toy surfaces in CPPAES (Table 6.5) were greater than surface loadings as measured by deposition coupons (Table 4.4).
- Measurements from CPPAES (data not presented) suggest that surface wiping of plush toys yields only a small fraction of the total amount of chlorpyrifos absorbed into the toys (as measured by extraction). Indirect ingestion among children who regularly mouth soft toys may thus be underestimated by toy surface wipes.
- In “transfer off” experiments conducted with a fluorescent tracer (riboflavin) as part of the Transfer studies, removal from skin via the mouthing of 4 fingers was measured. Eight replicates were performed with each of three participants (data not presented), with 0 to 26% of the tracer removed per replicate (loss was significantly different from zero in only one-half of the replicates).

Table 6.1 Collection methods for the transfer of pesticide surface residues to food or objects.

Study	Study Type	Age Range	Sampling Details	Collected After Application	Sample Handling	Composite Sample	Insecticides Measured	Comments
Food (Surfaces to Foods)	Laboratory	n/a	1, 10, & 60 min contact between food and contaminated surfaces	Yes-1 hr following applications	Foods extracted immediately following sampling	No	Chlorpyrifos Diazinon Heptachlor Isofenphos Malathion Permethrin	Surface wipes were collected. The influence from contact force and duration were evaluated
Food (Tile to Foods)	Laboratory	n/a	10 min contact between food and contaminated tile surface	Yes-1 hr following applications	Foods extracted immediately following sampling	No	Chlorpyrifos Cyfluthrin Cypermethrin Deltamethrin Fipronil Malathion Permethrin	Surface wipes and deposition on foil coupons collected
DIYC	Field	1-3 yr	Handled leftover food, untouched leftover food, food press	Yes	Collected in individual zip closure bags	Yes	Diazinon	Foods leftover from meal were combined into two types of samples; <i>i.e.</i> , all handled foods combined, all untouched foods combined
CHAMACOS	Field	0.5-2.5 yr	Teething ring or small ball provided 1.5 days before sampling	No	Stored at -20 C until analysis	No	Chlorpyrifos Diazinon Permethrin	Surface of toys wiped
CPPAES	Field	<5 yr	Plush toy given to child to handle for 11 days	Yes	No information	No	Chlorpyrifos	Surface of toys wiped; whole toys extracted
Transfer	Laboratory	Adult	Mouthing removal of fluorescent tracer	n/a	Video-fluorescence imaging	No	Surrogate (Riboflavin)	Many measurements at detection level of technique

n/a, Not applicable

Table 6.2 Videotaped children’s hand- and object-to-mouth activity details.

Study	N	Age (years)	Sampling Location	Time Period	Method of Analysis	Activity of Interest	Availability
MNCPES	19	3 to 12	Homes (inside and/or outside)	4 consecutive hours in normal daily activities	Methods of Reed <i>et al.</i> , 1999	Hand-to-mouth Object-to-mouth	Freeman <i>et al.</i> , 2001.
CPPAES	10	2 to 5	Homes (inside or outside)	4 hours on Day 2 following crack and crevice application of chlorpyrifos	Computer software (Virtual Timing Device) Quantified 4 hours of videotape for both hands	Hand-to-mouth Object-to-mouth	Freeman <i>et al.</i> , 2004.

Table 6.3 Videotaped hand-to-mouth and object-to-mouth counts.

Study	Hand-to-Mouth		Object-to-Mouth		Eating Events
	Mean	Median	Mean	Median	
CPPAES (2 to 5 yrs)	19.8	16	8.4	6.4	Unspecified
Tulve <sup>a</sup> ≤ 24 month old	18	12	45	39	Excluded
Tulve >24 month old	16	9	17	9	Excluded
MNCPES (3 to 12 yrs)	5.7	2.5	1.8	0	Unspecified
MNCPES boys indoor	4.7	NR	1.0	NR	Unspecified
MNCPES girls indoor	8.1	NR	2.6	NR	Unspecified

NR, Not Reported

<sup>a</sup> Tulve data (Tulve *et al.*, 2002) included for comparison.

Table 6.4 Objects commonly mouthed by preschoolers in CTEPP.

Category	Items
Toys	Plastic rings/bracelets, stuffed animals, balls, walkie talkie, building blocks, doll, bubble blower
Food-Related Items	Ice pops, candy wrapper, water bottle, utensils, napkins, drinks
Miscellaneous	Plastic blow-up chair, pens, greeting cards, clothing, CDs, towels, blanket, pets

Table 6.5 Median and 95<sup>th</sup> percentile pesticide loadings (ng/cm<sup>2</sup>) measured on toy surfaces.

Study	Chlorpyrifos		Diazinon		<i>cis</i> -Permethrin		<i>trans</i> -Permethrin		cyfluthrin	
	P50	P95	P50	P95	P50	P95	P50	P95	P50	P95
CHAMACOS	BDL <sup>a</sup>	0.15	0.034	0.27	BDL	0.053	BDL	0.072	BDL	BDL
CPPAES	3.0	21	-- <sup>b</sup>	--	--	--	--	--	--	--

<sup>a</sup> BDL, Below minimum detection limit

<sup>b</sup> Blank cells (--) indicate the pesticide was not measured in the study

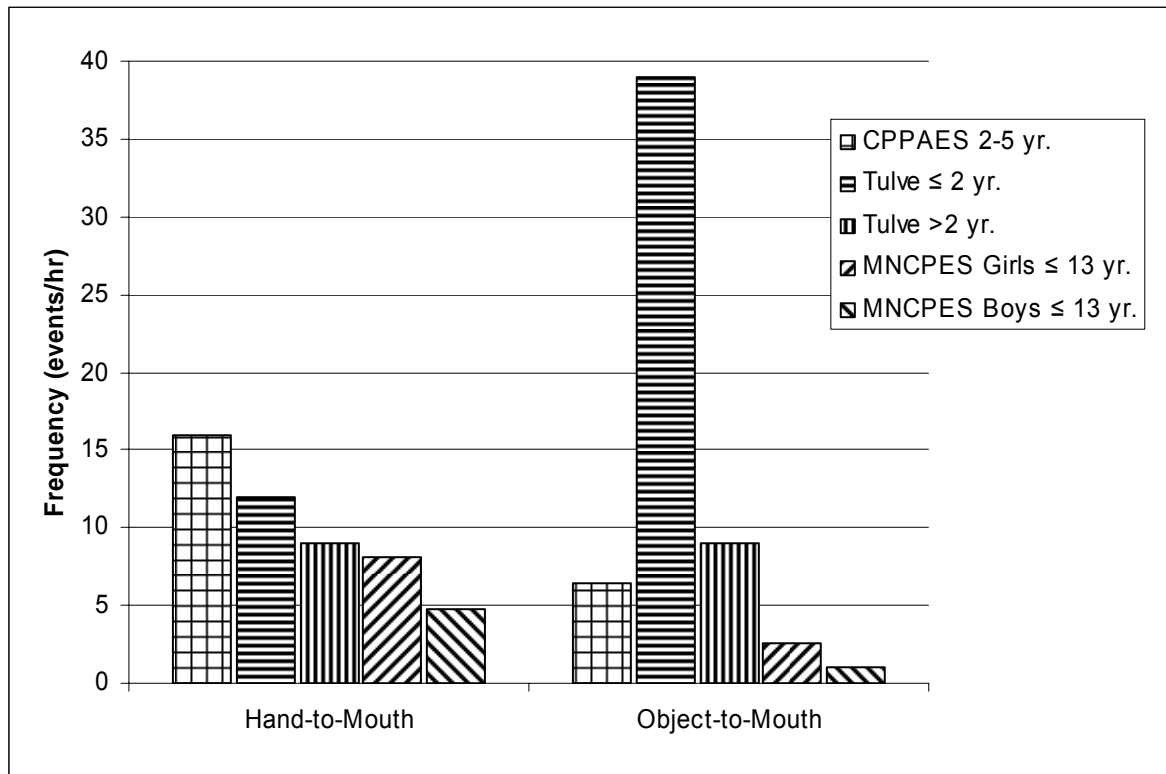


Figure 6.1 Comparison of the median hand-to-mouth and object-to-mouth contacts per hour among CPPAES and MNCPEs children. MNCPEs values are means instead of medians. Tolve data (Tolve *et al.*, 2002) included for comparison.

### 6.3 Transfer of Pesticide Residues to Food

- The experiments reported here (Appendix B, Food Transfer Studies) used loadings that were near to or greater than the 95<sup>th</sup> percentile for loadings in most of the recent field studies (See Table 4.4).
- Higher pesticide transfer to food occurred from hard, smooth surfaces, such as hardwood flooring; lower transfer occurred from carpet. For example, 33% of chlorpyrifos was transferred from wood flooring to an apple, whereas the amount transferred from carpet was not enough to be reliably quantified (Table 6.6).
- Bologna, a moist and fatty food, removed a higher percentage of pesticides from a hard surface than did fruit leather, a low-fat and low-water content food (Table 6.7).
- Comparison (Table 6.8, Figure 6.2) of measured dietary intake of diazinon (incorporating excess contamination due to handling) with estimates predicted by the Children's Dietary Intake Model (CDIM) suggests that use of fixed values for transfer efficiencies and for activity factors in the model may result in inaccurate estimates of daily dietary intake. Model-predicted estimates generally under-predicted intake.
- Diazinon concentrations in untouched leftover food were compared with those in handled leftover food in DIYC. Daily dietary intake estimates accounting for contamination due to handling by children were often double the intake estimates based on untouched food (Total Measured Dietary Intake vs. Duplicate Diet Intake, Table 6.8), indicating that duplicate diets may significantly underestimate actual intake in homes that have high surface pesticide residue loadings.
- Food transfer studies have provided evidence that transfer of pesticide residues from surfaces to foods is dependent on such factors as pesticide class, food type, contact duration, and contact force (data not presented).
- Applied force produced a considerable increase in transfer efficiency (data not presented). Moreover, the effect of applied force was even more dramatic as contact duration increased.

Table 6.6 The transfer efficiency (percent transfer, mean  $\pm$  sd) of pesticide residues from treated surfaces to foods (relative to transfer to IPA wipes), after a 10-min contact duration (Food Transfer Studies).

Pesticide	Sampling Media	N	Treated Surface		
			Ceramic Tile	Wood Flooring	Carpet
Chlorpyrifos (21-38 ng/cm <sup>2</sup> )	Bologna	2	36 $\pm$ 20	15 $\pm$ 4	BQL <sup>a</sup>
	Apple	2	18 $\pm$ 5	33 $\pm$ 8	BQL
	Cheese	2	7 $\pm$ 0	26 $\pm$ 1	BQL
Diazinon (20-30 ng/cm <sup>2</sup> )	Bologna	2	41 $\pm$ 5	29 $\pm$ 0	BQL
	Apple	2	35 $\pm$ 8	50 $\pm$ 5	BQL
	Cheese	2	20 $\pm$ 7	103 $\pm$ 18	BQL
Malathion (33-45 ng/cm <sup>2</sup> )	Bologna	2	60 $\pm$ 21	31 $\pm$ 1	BQL
	Apple	2	132 $\pm$ 74	18 $\pm$ 1	212 $\pm$ 60
	Cheese	2	94 $\pm$ 33	52 $\pm$ 37	400 $\pm$ 173
<i>cis</i> -Permethrin (40-53 ng/cm <sup>2</sup> )	Bologna	2	19 $\pm$ 15	70 $\pm$ 86	BQL
	Apple	2	26 $\pm$ 13	3 $\pm$ 1	BQL
	Cheese	2	BQL	BQL	BQL
<i>trans</i> -Permethrin (43-55 ng/cm <sup>2</sup> )	Bologna	2	23 $\pm$ 20	10 $\pm$ 1	BQL
	Apple	2	29 $\pm$ 14	5 $\pm$ 0	BQL
	Cheese	2	BQL	BQL	BQL

<sup>a</sup> BQL = Below Quantitation Limit

Table 6.7 The transfer efficiency (percent transfer, mean  $\pm$  sd) of pesticide residues from a treated ceramic tile surface to various foods and to an IPA Wipe (Food Transfer Studies).

Pesticide Class	Pesticide	Sampling Media	N	% Transfer
Organophosphate	Chlorpyrifos (123 ng/cm <sup>2</sup> )	Bologna	3	64.7 $\pm$ 15.0
		Apple	3	27.5 $\pm$ 8.0
		Fruit Leather	3	13.5 $\pm$ 2.0
		20-mL IPA Wipe	3	99.8 $\pm$ 10.8
	Malathion (193 ng/cm <sup>2</sup> )	Bologna	3	74.9 $\pm$ 17.7
		Apple	3	29.7 $\pm$ 8.4
		Fruit Leather	3	8.7 $\pm$ 2.7
		20-mL IPA Wipe	3	104.6 $\pm$ 10.9
Pyrethroid	Cyfluthrin (143 ng/cm <sup>2</sup> )	Bologna	3	47.8 $\pm$ 13.4
		Apple	3	24.0 $\pm$ 3.4
		Fruit Leather	3	0.7 $\pm$ 0
		20-mL IPA Wipe	3	108.5 $\pm$ 12.1
	Cypermethrin (185 ng/cm <sup>2</sup> )	Bologna	3	45.0 $\pm$ 10.7
		Apple	3	21.5 $\pm$ 6.9
		Fruit Leather	3	0.6 $\pm$ 0
		20-mL IPA Wipe	3	101.5 $\pm$ 7.0
	Deltamethrin (211 ng/cm <sup>2</sup> )	Bologna	3	39.2 $\pm$ 6.1
		Apple	3	22.2 $\pm$ 5.1
		Fruit Leather	3	2.4 $\pm$ 0.2
		20-mL IPA Wipe	3	83.7 $\pm$ 4.3
	Permethrin (147 ng/cm <sup>2</sup> )	Bologna	3	44.0 $\pm$ 11.5
		Apple	3	19.8 $\pm$ 7.1
		Fruit Leather	3	1.3 $\pm$ 0.1
		20-mL IPA Wipe	3	100.8 $\pm$ 4.8
Phenylpyrazole	Fipronil (203 ng/cm <sup>2</sup> )	Bologna	3	43.3 $\pm$ 1.6
		Apple	3	30.9 $\pm$ 14.8
		Fruit Leather	3	2.0 $\pm$ 1.7
		20-mL IPA Wipe	3	103.8 $\pm$ 10.4



Table 6.8 The measured and predicted ingestion (ng/day) of diazinon from the DIYC.

Child	Sampling Day	Duplicate Diet Intake	Excess Dietary Intake <sup>a</sup>	Total Measured Dietary Intake <sup>b</sup>	CDIM Predicted Dietary Intake <sup>c</sup>	Percent Difference <sup>d</sup>
		ng/d	ng/d	ng/d	ng/d	%
1	Pre	197	384	581	357	-39
	1	1063	1270	2333	1271	-46
	4	280	220	500	281	-44
	5	270	501	771	333	-57
	6	140	322	462	142	-69
	7	563	536	1099	702	-36
	8	253	160	413	397	-4
2	1	455	156	611	663	9
	2	233	95	328	402	23
	3	212	373	585	392	-33
	4	260	414	674	612	-9
	5	188	189	377	278	-26
3	2	95	90	185	509	175
	8	412	344	756	940	24

<sup>a</sup> Measured surface-to-food and hand-to-food transfer due to handling of foods, concentration in handled but uneaten portion extrapolated to eaten portion.

<sup>b</sup> Duplicate Diet intake plus Excess Dietary intake.

<sup>c</sup> Estimated by deterministic model using fixed transfer efficiency and activity values.

<sup>d</sup> Percent Difference =  $100 * [(CDIM \text{ Predicted Intake} - Total \text{ Measured Intake}) / (Total \text{ Measured Intake})]$ .

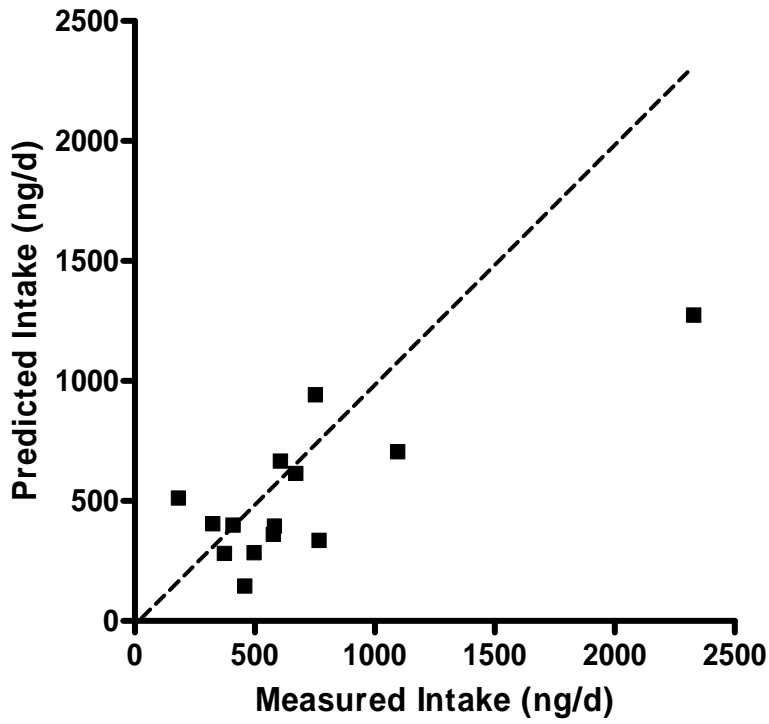


Figure 6.2 Comparison of measured and predicted ingestion of diazinon (ng/day) from the DIYC. Dashed line represents a hypothetical slope of 1. Measured intake generally exceeds predicted intake, as indicated by the majority of points lying to the right of the dashed line.

## 6.4 Indirect Ingestion of Dust and Soil

The potential indirect ingestion exposure (ng/day) can be estimated using indoor floor dust (ng/g) and outdoor soil sample concentrations (ng/g) together with the child's body weight (kg), estimated daily dust ingestion rate (g/day), estimated daily soil ingestion rate (g/day), and the estimated oral bioavailability. In CTEPP, the daily dust ingestion rates were calculated based on questionnaire responses related to specific activities of each child in the month prior to field sampling. These activities included pacifier use, teething, mouthing body parts, licking floors, and placing toys or other objects into the mouth. The daily soil ingestion rates were estimated based on how often a child played with sand/dirt and ate dirt, sand, or snow. Many of these parameters have very high uncertainty associated with them. The daily dust and soil ingestion rates were each estimated as 0.025, 0.050, or 0.100 g/day. The indirect exposure estimates, presented in Table 6.9, showed the following:

- Indirect ingestion estimates for the permethrin isomers were much higher than for chlorpyrifos or diazinon, largely because permethrin was measured at much higher concentrations in floor dust (Figures 4.6 and 4.7).
- The differences between NC and OH in mean permethrin concentrations in dust suggest potential regional differences in indirect ingestion.

Table 6.9 The estimated exposures (ng/day) of NC and OH preschool children in the CTEPP study to chlorpyrifos, diazinon, and permethrin through indirect ingestion.

Pesticide	State	N	Mean	SD	GM	GSD	Min	P25	P50	P75	P95	Max
Chlorpyrifos	NC	117	15.5	29.0	6.2	1.3	0.3	2.8	5.2	14.8	80.4	233
	OH	116	27.8	164	3.0	1.5	0.2	1.1	2.7	6.2	33.5	1570
Diazinon	NC	118	21.7	81.9	1.6	2.0	<MDL	0.4	1.0	4.3	150	622
	OH	116	49.1	367	1.5	1.9	<MDL	0.4	1.0	3.4	45.3	3800
<i>cis</i> -Permethrin	NC	120	220	670	48.4	1.6	1.7	17.1	48.1	113	718	4540
	OH	116	61.5	139	21.3	1.4	1.9	7.8	17.9	52.7	327	1210
<i>trans</i> -Permethrin	NC	120	222	698	42.7	1.7	1.1	11.9	35.4	119	680	4800
	OH	102	61.2	153	16.6	1.5	1.2	5.3	11.7	45.9	210	1190

<MDL, less than method detection limit

## 6.5 Indirect Ingestion: Summary

As shown in the bulleted lists of observations from these laboratory and observational studies, progress has been achieved in identifying and quantifying a number of factors that are believed to potentially impact indirect ingestion among children.

- Videotape analysis of children's hand- and object-to-mouth contacts has provided evidence that hand-to-mouth activities were more frequent: among infants and toddlers than among older children, among girls than among boys, and at indoor locations than at outdoor locations.
- Objects most commonly mouthed by preschoolers were identified as typically being toys and food-related items.
- High chlorpyrifos loadings were measured on toy surfaces following routine residential application.
- Fluorescent tracer experiments found that removal from skin (at very high tracer loadings) by mouthing was highly variable. Additional information is still needed on the fraction of residue transferred from the hands to mouth during typical mouthing events at dermal loading levels observed in field studies.
- At high surface loadings, pesticide transfer to food was greater from hard, smooth surfaces than from carpet.
- In homes with high surface pesticide residue loadings, residue concentrations in foods handled by children were often twice as high as concentrations in leftover unhandled foods.
- The transfer of pesticide residues from surfaces to foods appears to be dependent on such factors as pesticide class, food type, contact duration, and applied force.
- Indirect ingestion estimates for permethrin were much higher than for chlorpyrifos or diazinon, largely because permethrin was measured at much higher concentrations in floor dust.