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Factors influencing total dietary exposures of young children

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A deterministic model was developed to identify the critical input parameters needed to assess dietary intakes of young children. The model was used as a framework for understanding the important factors in data collection and data analysis. Factors incorporated into the model included transfer efficiencies of pesticide from surfaces to food, transfer efficiencies of pesticide from surfaces to hands to food, and more accurate microactivity data related to contact frequency for the three variables of interest — hands, surfaces, and food. Results from range-finding measurements of transfer efficiencies using an aqueous pesticide solution of a mixture of malathion, diazinon, and chlorpyrifos sprayed on the surfaces indicate that a higher pesticide transfer occurred from hard surfaces to food (hardwood, plastic), with low transfer from soft surfaces (carpet, cloth). Six children, all less than 4 years old, were videotaped to obtain realistic contact frequency and times for the interaction of hands, surfaces, and foods during eating meals and snacks while in their homes or day care centers. The time range of eating events varied from about 2 to 55 min, with an average of about 20 min. The average number of contact frequencies between food and hands was 19 times for each eating event, with a range of 10–40. Contacts between the surface and hand were about the same as the food and hands. Contacts between foods and surfaces ranged from 0 to 32, but only five or less of the contacts per eating event were associated with surfaces other than eating utensil. The children's microactivity data collected during the eating events, together with the laboratory results from the transfer studies, were provided as input into a Monte Carlo simulation of the dietary ingestion model. Simulation results indicate that children's handling of the food could contribute 20–80% of the total dietary intake of pesticides. Dietary exposure due to residues in the food before handling accounted for 16% and 47%, respectively, of the total mean intake from simulations for a child's consumption of an apple or banana. These results indicated that transfer efficiencies for foods on various surfaces typically found in homes as well as children's hand contacts with the food and surfaces are important as determinants of dietary exposure. *Journal of Exposure Analysis and Environmental Epidemiology* (2000) **10,** 710–722.

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Introduction

The dietary contribution to an aggregate exposure is potentially an important route of exposure, especially for young children, and for certain scenarios may potentially be the dominant route. Young children do not consume foods in a structured manner, which leads to several pathways for dietary exposure. Hands and surfaces that may be contaminated come in contact with foods while they are eating. Environmental contamination appearing in the child's diet can result from contamination in the food as purchased or from preparing, serving, handling, and/or eating food in the home. Food contamination while eating is of special concern for children 1–3 years old, since this age group has a high frequency of hand-to-food, hand-to-surface, and surface-to-food interactions. These pathways of food contamination may be very important in homes with high surface concentrations of chemical residue which may have resulted from outdoor air, track-in of particles, and/or indoor sources. One recent study designed to evaluate the sources and amount of dietary lead exposure for children living in lead-contaminated homes in the Newark, New Jersey area indicates that this pathway of exposure is significant (Melnyk et al., 1999) and that dietary exposures were substantially higher than predicted from food sources alone. Melnyk et al. used preliminary experimental techniques and surrogate foods to estimate the potential for excess dietary exposure caused by the child's handling of foods and established the foundation for further methods refinement.

There has been limited research into the interacting factors associated with children's dietary exposures, other than the above referenced study of the contribution of lead to the dietary ingestion of small children (Freeman et al., 1999). Research driven by the Food Quality Protection Act of 1996, which seeks to understand the potential dietary pesticide exposures to children, is being conducted. Interest in potential pesticide exposure of young children results from recent studies, including pilot studies of the Lower Rio

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Grande Valley (LRGV) in Texas (Akland et al., 1997; Berry et al., 1997) and the Agricultural Health Study (Melnyk et al., 1997). In addition to these efforts, one module of the Region V NHEXAS Study involved a study of pesticides and children's exposures (Pellizzari et al., 1995), but it did not focus on the potential contribution of contamination of foods on the total dietary exposure of small children caused by handling and surface transfers.

Three separate areas of research associated with dietary contamination of foods eaten by young children will be discussed in subsequent sections of this paper. These areas are model development, residue transfer, and studies of children's behavior that affect dietary exposure.

Methods

Model Development

This research began with a simple, deterministic model to serve as a framework for understanding the concept of dietary ingestion, and for identifying the important factors requiring subsequent laboratory studies and data collection. The sum of three major terms characterizes a young child's dietary intake of any chemical(s), not just pesticides. These terms are as follows:

- the original contaminant residue on or in the food after preparation, but before handling by the child;
- surface-to-food contamination as the food comes into contact with the contaminated surfaces while being handled by the child; and
- surface-to-hand-to-food contamination as the child touches contaminated surfaces and then handles and eats the food.

For each respective term described above, the intake of a pesticide associated with one food item and the number of specific eating activities resulting in that food item's contact with contaminated surfaces and the child's hands before it is eaten (N), are described by:

$$I = RW_{\mathrm{T}} + \{\underbrace{\left[C_{\mathrm{S}}F_{\mathrm{S}}T_{\mathrm{S/F}}A_{\mathrm{S/F}}\right]}_{\text{term 1}} + \underbrace{\left[\left(C_{\mathrm{S}}T_{\mathrm{S/H}}A_{\mathrm{S/H}}\right)\left(T_{\mathrm{H/F}}A_{\mathrm{H/F}}F_{\mathrm{H}}\right)\right]}_{\text{term 3}}\}_{\mathrm{N}} \quad (1)$$

where:

I=total dietary intake of the pesticide for one food eaten (μg);

R=pesticide residue concentration of food item after preparation for consumption (μg/g food);

W_{T}=total amount of the individual food consumed (g);

C_{S}=concentration (or loading) of the pesticide on a contacted surface (μg pesticide/cm^2);

F_{S}=total food surface area in contact with a contaminated surface (cm^2);

$T_{\mathrm{S/F}}$=surface-to-food pesticide transfer efficiency (dimensionless);

$A_{\mathrm{S/F}}$=surface-to-food child activity factor; a function of contact frequency (dimensionless);

$T_{\mathrm{S/H}}$=surface-to-hand mass transfer efficiency (dimensionless);

$A_{\mathrm{S/H}}$=surface-to-hand child activity factor (dimensionless);

$T_{\mathrm{H/F}}$=hand-to-food mass transfer efficiency (dimensionless);

$A_{\mathrm{H/F}}$=hand-to-food child activity factor (dimensionless);

F_{H}=total food surface area in contact with the portion of the hand that has contacted a contaminated surface (cm^2).

Using this model, a computer simulation of dietary exposures was undertaken to understand the impact of measurement errors in each parameter on the model prediction. The simulation and analysis followed the techniques discussed in the literature (Fiacco, 1983; Hamby, 1995). A Monte Carlo method was employed using a random number generator routine available in SAS (SAS Inc., Cary, NC) to generate 5000 values of the parameters. Input values for parameters of the model were based on published data on surface loadings, including NHEXAS (Gordon et al., 1999), and measured data for pesticide transfer efficiencies (surface to food) and children's contact frequencies of food and surfaces while eating. Measurement data were obtained from experimental studies related to transfer efficiencies and video analysis. Table 1 provides a summary of information used as input into the model.

To test parameter sensitivity, the parameter of interest was perturbed at both sides of its mean value, while letting the other parameters vary to produce two distributions — high and low. An index similar to the one proposed by Hoffman and Gardner (1983) was used to measure the sensitivity index (SI) defined as:

$$\mathrm{SI} = \left(D_{\mathrm{high}} - D_{\mathrm{low}}\right)/D_{\mathrm{high}} \quad (2)$$

where D_{high} is the median or 90th percentile of the output when the parameter of interest is set at its high value and D_{low} is the result for a low parameter value. To estimate the contribution for each term to the overall intake, the total intake value was calculated while letting the other terms vary within their assigned distribution. Terms 1, 2, and 3 in Equation 1 were calculated and compared with the reference output to determine the relative importance of each of the terms.

Table 1. Parameter definitions and ranges for model input.

Name	Description	Units	Distribution	Range of parameter
R	Contaminant residue	$\mu g/g$	Log normal	0.01–0.9 ppm apple, chlorpyrifos; 0.01–0.04 ppm banana, chlorpyrifos (Pennington, 1992)
W_T	Total amount of food item consumed	g	Degenerate	1/4 apple slice — 49 g; one peeled banana — 133 g (experimental measurements)
$T_{H/F}$	Hand-to-food mass transfer efficiency	NA[a]	Beta	0.5 with SD=0.2 (assumed to be the same as surface-to-hand transfer efficiency for hard surfaces)
$A_{H/F}$	Hand-to-food child activity factor	NA	Poisson	average 19; range 10–39 (videotapes)
F_S	Total surface area of food in contact with surface	cm^2	Degenerate	1/4 apple slice — 75 cm^2; one peeled banana — 30 cm^2 (experimental measurements)
C_S	Loading of the contaminant on the surface	$\mu g/m^2$	Log normal	median 0.14; 0.003–48.5 $\mu g/m^2$ (Gordon et al., 1999)
$T_{S/H}$	Surface-to-hand mass transfer efficiency	NA	Beta	0–1, with mean=0.5, SD=0.2 for hard surfaces; mean=0.1, SD=0.1 for cloth and carpet (experimental measurements)
$A_{S/H}$	Surface-to-hand child activity factor	NA	Poisson	range 1–39 (videotapes)
$T_{S/F}$	Surface-to-food mass transfer efficiency	NA	Beta	Same as $T_{S/H}$ (experimental measurements)
$A_{S/F}$	Surface-to-food child activity factor	NA	Poisson	range 0–32 (videotapes)

[a]Not applicable.

Contaminant Transfer Studies

Pathways of dietary ingestion (surface-to-food and surface-to-hand-to-food) are based on the physical transfer of a contaminant from one surface to another. In the process of contaminant transfer, the surface of a food item becomes contaminated and then the food item is consumed. The amount or ratio of contaminant transferred from a surface to the food during a contact, called the mass transfer efficiency, plays an important role in the magnitude of exposure from these dietary pathways. Early screening studies (Sheldon and Berry, 1996) measured transfer efficiency between some food and surface media indicating that significant transfer of contaminants can occur. However, these experiments were limited to a few foods, chemicals, and conditions. More comprehensive testing was needed to identify the foods, surfaces, and conditions with the greatest potential for increasing children's dietary exposures.

The pesticides selected for experimentation were based on several factors, including: (1) likely presence in home and day care environments; (2) potential toxicity; (3) ability to mix and apply in a water-based form; and (4) availability and ease of analysis. A list of the most commonly used pesticides in residential settings was derived from literature searches and interviews with agricultural extension agents and from lists of pesticides found in monitoring studies conducted by FDA or EPA. Table 2 provides the results of this effort. Using this information, the range-finding experiments concentrated on

Table 2. Candidate pesticides (insecticides) with evidence of current indoor usage.

Pesticide	Use	LD_{50}[a] (mg/kg)	Evidence of food contaminants[b]	NHANES IV (urine)	Ease of analysis[c]
Carbaryl	Lawn, pets	246–283	–	Yes	Separate method required
Chlorpyrifos	Pest control	96–270	Yes	Yes	Yes
Diazinon	Residential	1250	Yes	Yes	Yes
Isofenphos	Termite	28–38	–	–	Yes
Permethrin	Home garden	430–4000	Yes	Yes	Yes
Propoxur	Residential	50	Yes	Yes	Yes
Pyrethrins	Lice/cockroach	200	Yes	–	NA
Malathion	Flying insects	5500	Yes	Yes	Yes

[a]Oral LD_{50} in rats; Farm Chemicals Handbook 1997, Table E, Meister Publishing Co., Willoughby, OH.
[b]Among 10 highest concentrations found in duplicate diet samples collected in the Lower Rio Grande (Berry et al., 1997) and Agricultural Health Study (Melnyk et al., 1997) pilot studies.
[c]Amenable to extraction, cleanup and analysis using previously developed analytical methods (Sheldon et al., 1997).

three organophosphate pesticides (chlorpyrifos, malathion, and diazinon) with more limited experimentation conducted on the transfer efficiencies of heptachlor, permethrin (*cis* and *trans*) and isofenphos.

Surfaces were selected based on three factors: (1) surfaces likely to be contacted in home or day care environments; (2) surfaces likely to have food placed on prior to consumption; and (3) a range of different surface types, both hard and soft. The four surface types selected for the initial studies were hardwood (floors), carpet (medium plush), denim cloth (medium weight and grade), and plastic (such as that which might be found on a child's high chair; polypropylene, cut from a 121.9×243.8 cm^2 [4×8 in.^{2}] sheet). All of the test surfaces were prepared by cutting the surface material to a standard square size, 30.5 cm^2, which was referred to as a "coupon." Prior to testing, the surface materials were prepared as follows: hardwood and plastic were washed with soap and water, cloth was laundered and dried in a clothes dryer and then ironed, and carpet was vacuumed to remove loose particles.

The six commonly eaten foods selected for the range-finding studies were based on four factors: (1) high frequency of consumption (by children); (2) frequency of handling by children; (3) surface characteristics; and, if possible, (4) used in previous exposure studies. The food items selected were apple, luncheon meat, cheese, bread, tortilla, and chicken nuggets.

A pesticide deposition chamber (PDC) was developed to uniformly deposit liquid pesticides onto test surfaces. The overall inside dimensions were 167.6 cm (66 in.) long by 76.2 cm (30 in.) high by 55.9 cm (22 in.) wide. The walls and floors were constructed using polypropylene to ease cleanup between deposition experiments. A charcoal-filtered exhaust system was attached to prevent contamination of surrounding air during use. The PDC was configured for testing as follows: spray tank pressure of 30 psi; spray head transverse speed of 19.8 cm/s (0.65 ft/s); three passes of the spray tip; and distance from spray tip to deposition area of 53.3 cm (21 in.). Tests of deposition uniformity were conducted to determine Relative Standard Deviation (RSD=standard deviation/mean) across a 38.1×30.5 cm^2 (15×12 in.^{2}) spray area. Figure 1 illustrates a side view of the PDC.

The experiment was designed to evaluate the transfer efficiency of selected foods in contact with contaminated surfaces for a specified period of time, either 1 or 10 min. The experiments were performed on surfaces contaminated with a predetermined amount of aqueous solution containing the selected pesticides, primarily chlorpyrifos, malathion, and diazinon, prepared using a water-based solution of combined commercially available pesticide concentrates. This solution was prepared at concentrations that, when sprayed onto the surfaces and allowed to dry for at least 2 h, yielded surface concentrations in the 5–80 ng/cm^2 range. To achieve these surface concentrations, the spray solution was prepared by diluting small volumes (50–100 μl) of the concentrated pesticide solutions into 2000 ml of water. In addition to the food matrices, a press sampler (the EL sampler) developed by Edwards and Lioy (1999) was modified to utilize commercially available 90-mm filter disks. This sampler was used to determine the transfer efficiency from the surface to a C_{18} filter on the sampler that has been shown to closely mimic the transfer characteristics of human skin (Edwards and Lioy, 1999). In their paper, the authors note that the EL methodology was designed and tested on an adult hand due to limitations related to Institutional Review Board (IRB) approval for testing on children's hands. It is likely that characteristics such as wetness, stickiness, and skin characteristics may affect the amount of material adhering

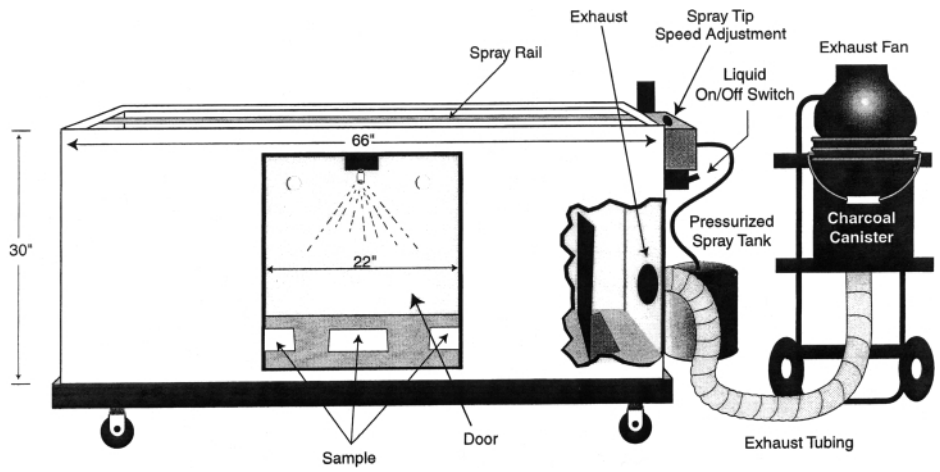

Figure 1. Pesticide spray deposition chamber.

to the skin of a child, relative to that of the adult. Once on the skin, however, the cutaneous permeability is likely to be similar (Liou and Janniger, 1997). For the purpose of performing mass transfer experiments, test surfaces were prepared in sets of three; one surface served as the control while the other two surfaces were used for food or press transfer measurements.

The spray liquid deposition rate was determined by weighing the liquid deposited onto aluminum weighing boats placed in the spray deposition chamber with the test coupons. The liquid deposition was monitored by weighing the boats following the spraying of multiple sets of coupons to insure a consistent spray deposition rate. The liquid deposition rate was also used to determine the theoretical pesticide concentration sprayed on the test surface coupons.

After the test surfaces were prepared, the surface concentration of each contaminated coupon and each food-exposed coupon was determined by either wiping the surface (hardwood or plastic) or extracting the entire surface coupon (cloth or carpet). For the hard surfaces, the entire surface was wiped with a set of two cotton sponges wet with isopropanol to remove the pesticides. The cotton sponges were then extracted with solvent (hexane/ethyl ether 95:5). For the soft surfaces, all the materials were extracted in solvent (hexane/ethyl ether 95:5). Typical extract concentrations ranged from 10 to 100 ng/ml.

Food samples were placed on the contaminated surface with no additional pressure added. After initial experimentation, the contact time selected was 10 min. For each test, duplicate food samples were contacted with a second surface. A control coupon was processed with each test batch to determine the surface loading concentration. Analysis of the amount of pesticide transferred into each food item was determined by difference between the amount of the pesticide analyzed from the coupon with the food item place on it subtracted from the amount determined to be on the control coupon. A small subset of the food samples was analyzed by gas chromatography/mass spectrometry (GC/MS) to verify the acceptability of the difference procedure.

A second set of experiments was conducted in an independent laboratory to confirm the results from the PDC experiments. This set of experiments employed the method of pipetting the aqueous solution onto the surface materials. An eight-channel multipipette was used to deposit 40 ml of the pesticide solution at 1-cm intervals across the test surface. Target concentrations were 15–55 ng/cm^2. Pesticides were applied to surfaces in a multihazard glove box, transferred to a laminar flow hood, and allowed to dry for a total of 2.5 h. Food samples were placed on the surfaces, with and without pressure (0.1 lb/cm^2) for 1, 10, and up to 30 min. The pesticides were then extracted from food samples by blending foods with 50/50 methylene chloride/acetone followed by a two-step clean up and analysis by GC/MS. The C_{18} filters were extracted with 1/1: methylene chloride/ethyl acetate and the isopropanol wipes with 100% methylene chloride.

Pesticide levels on surfaces available for transfer to the foods were determined in three ways: (1) compared to the transfer to the C_{18} filter material using the EL sampler; (2) compared to the applied concentrations of pesticides as determined by analysis of the aqueous solution; and (3) compared to the control coupon result as determined by surface wipes (hard surfaces) or direct extraction of the material (soft surfaces). It should be noted that the second laboratory used 100% methylene chloride as the solvent, whereas the first laboratory used a 95:5% mixture of hexane:ether. Each of the measurements served as a relative basis for calculating transfer efficiency to the foods contacted with the surfaces.

Children's Behaviors

Videotaping and analysis of children's behaviors while eating were undertaken to provide information about the child activity factors as they affect potential dietary contamination. The frequency and duration of hand and food contacts with different surfaces, types, and amounts of foods consumed and other factors were recorded on videotape and translated by the computerized software program described by Zartarian et al. (1998).

Videotapes of four farm children, which had been previously studied as part of a microactivity pattern database (Zartarian et al., 1995), were used to modify the software to analyze for hand, surface, and food contact activities. The videotapes of the four children were processed to provide preliminary data for the needed dietary activity factors and to provide videotaping protocols for taping during eating events.

Participants were recruited by word-of-mouth, contacts with local day care centers, and local community organizations. Incentives of US$10 per segment were paid, where a segment is defined as a meal, snack, or short non-dietary activity period. It should be noted that it was difficult to obtain consent from the day care centers to videotape a likely participant, even though we had prior approval from the parent. Six children (two 1 year, three 2 years, and one 3 years of age; two females and four males) were videotaped during the course of the study. Four children were taped at home during the day and two while attending private day care centers. Prior to videotaping, videotaping protocols were developed so that information could be compared across children. The camera was focused on the child's hands and food during taping. When food and hands could not be captured concurrently, the hand was considered the primary body part to be taped, with voiceover annotation describing the activities related to the food item.

For each of the children, two of the main meals (breakfast, lunch, dinner) and a snack event were captured. Children were videotaped over 2, usually non-consecutive, days. Before translation of the tapes could occur, each coder underwent training. Each trainee had to translate a "standard" file with less than 10% error, prior to being certified for the task. Translation of the information was performed for 20 possible object categories contacted by the food and 25 possible object categories contacted by the primary hand for each eating occasion.

Results and discussion

Total daily dietary intake for a contaminant was determined by summing intakes for all food items and associated contacts of each food with hands and contaminated surfaces. In measurable quantities, term 1 may be obtained by duplicate diet sampling procedures (Thomas et al., 1997) to provide total, daily intake of pesticides that are present on the foods themselves, plus those that have been introduced during preparation but prior to being handled by the child. Terms 2 and 3 are much more difficult to quantify and require measurements of specific factors (e.g., concentrations, areas, transfer efficiencies) in the eating environment, plus detailed microactivity analysis of eating activities and food handling characteristics of the child (e.g., child activity factors).

Using this model, a computer simulation of dietary exposures was undertaken to identify the parameters that have the greatest impact on dietary intake, to determine which variables must be measured with most accuracy, and to prioritize in the absence of data each component's (term 1, 2, or 3) contribution to the overall pesticide intake under realistic situations. Distributions of the input parameters were based on the statistical understanding of the underlying properties for the parameters in the model, e.g., the distribution was bounded by zero and 1.0, or it was known from the literature that the expected concentrations could be adequately characterized by a selected distribution, e.g., log normal.

Computer simulations were conducted using the dietary model to estimate the overall intake of a pesticide (chlorpyrifos) for single servings of different food items (apple and banana) and the relative importance of each model term to total pesticide intake. Term 1 of the total intake model was represented by chlorpyrifos levels reported (U.S. Food and Drug Administration, 1993) for apple, 0.01–0.9 ppm, and for banana, 0.01–0.14 ppm, and a maximum consumption of 49 g of apple or 133 g of banana. It was assumed that no additional pesticide was added during preparation in the home prior to handling by the child. Table 3 summarizes results of the computer simulations for each term of the model.

Table 3. Percent contribution of each term of Equation 1 of total estimated dietary intake.

Food item	Term 1	Term 2	Term 3	Total intake (μg)
Apple	0.16	0.19	0.65	3.99
Banana	0.47	0.13	0.40	5.71

Table 4. Sensitivity of model parameters.

Parameter	Median				90th Percentile			
	D_{low}	D_{high}	SI	Rank	D_{low}	D_{high}	SI	Rank
Loading	0.06	6.46	0.99	1	0.61	22.89	0.97	1
Residual	0.26	2.59	0.90	2	1.11	2.62	0.58	6
$T_{H/F}$	0.29	0.64	0.55	3	1.73	5.24	0.67	5
$T_{S/H}$	0.28	0.62	0.55	4	0.28	1.77	0.84	2
$T_{S/F}$	0.34	0.57	0.40	6	0.34	2.81	0.88	4
Food/hand[a]	0.42	0.49	0.14	7	2.82	3.75	0.25	7
Food/surface[b]	0.36	0.58	0.38	5	0.36	2.58	0.86	3

[a]Proportion of food area in contact with hand.
[b]Proportion of food area in contact with surface.

For both apple and banana, the simulations indicated that pesticide transfer to food caused by contact with surfaces and handling by the child (terms 2 and 3) would increase the pesticide intake significantly. In both instances, handling of the food by the child's contaminated hand (term 3) accounted for over 40% of the excess pesticide intake. This would occur when the frequency of hand-to-surface-to-food contacts is high, and the transfer from the hands to the food is also high, e.g., as from a hard surface. Children's handling of the food could contribute 20–80% of the total dietary intake of pesticides. These modeled results indicated that transfer efficiencies for foods on surfaces typically found in homes and accurate frequencies of children's hand contacts with the food and surfaces are important as determinants of dietary exposure.

Sensitivity Analysis

Results of the sensitivity index (SI) are shown in Table 4 for the median and 90th percentile. It can be seen that when the model input parameters are around the medians of the distribution, the most sensitive parameters are pesticide residue levels on foods and pesticide surface loading. However, when the actual measurements are at the higher end of the distribution, surface-to-food transfer efficiencies, surface-to-hand transfer efficiencies, and proportion of total food surface in contact with the surface become more important, in addition to surface loading. This result documents the need to more accurately measure surface loading and contact area with the surface, followed by measuring transfer efficiencies (surface-to-food and surface-to-hand). For high exposures, the importance of these factors increases and the contribution of food

handling by the child becomes the dominant factor in total dietary intake.

Transfer Efficiencies

Accurately measuring mass transfer efficiencies is difficult due to the large variability of the associated parameters encountered in everyday life. In order to accurately estimate exposure, estimates of contaminant transfer under very specific conditions must first be understood and characterized. In practice, it is unlikely that laboratory simulations of the real world can capture all the various conditions that are encountered by all children of all age groups and all

Table 5. Results of transfer experiments for two foods and two surfaces.

	Hardwood	Carpet	By difference	Food
Experimental conditions	Deposition: ~7.1 μl/cm^2	Deposition: ~7.1 μl/cm^2		
	Spiked gauze control (5.4 ng/cm^2)	Spiked carpet (10.7 ng/cm^2)		
	Diazinon: 5.4 (100%)	10.7 (100%)		
	Malathion 6.5 (120%)	8.5 (79%)		
	Chlorpyrifos 5.1 (94%)	9.4 (88%)		
	Spray solution (ng/cm^2)	Spray solution (ng/cm^2)		
	Diazinon 50.5	42.7		
	Malathion 51.4	31.3		
	Chlorpyrifos 62.9	51.5		
	Wipe results (ng/cm^2), n=3	Carpet results (n=3)		
	Diazinon 11.4[a] (16.5%)[b]	27.7 (19.7)		
	Malathion 30.3 (10.8%)	13.8 (7.6)		
	Chlorpyrifos 25.6 (7.5%)	37.5 (2.4)		
	Dermal press (ng/cm^2), n=2	Dermal press (n=2)		
	Diazinon 0.3 (10.1%)	26.2 (22.8)		
	Malathion 7.9 (32.5%)	14.8 (6.8)		
	Chlorpyrifos 2.7 (18.5%)	36.4 (2.9)		
Ham[c]	Transfer (%) coupon, (n=2)	Transfer (%) coupon		
	Diazinon 14 (0)	3 (833%)	3 (35)	11 (1)
	Malathion 39 (0)	−21 (86%)	33 (8)	23 (0)
	Chlorpyrifos 28 (25%)	−17 (94%)	10 (10)	13 (1)
	Transfer (%) applied, (n=2)	Transfer (%) applied		
	Diazinon 2 (0)	2 (900%)		
	Malathion 20 (0)	−10 (80%)		
	Chlorpyrifos 9 (22%)	−13 (92%)		
	Transfer (%) dermal press, (n=2)	Transfer (%) dermal press		
	Diazinon 333 (0)	64 (852%)		
	Malathion 128 (0)	−282 (82%)		
	Chlorpyrifos 207 (25%)	−179 (93%)		
Apple[c]	Transfer (%) coupon, (n=2)	Transfer (%) coupon		
	Diazinon 50 (0)	10 (17%)	24 (2)	0 (0)
	Malathion 23 (13%)	−4 (475%)	28 (14)	10 (5)
	Chlorpyrifos 42 (5%)	11 (73%)	18 (3)	12 (4)
	Transfer (%) applied, (n=2)	Transfer (%) applied		
	Diazinon 7 (0)	7 (557%)		
	Malathion 10 (10%)	−2 (450%)		
	Chlorpyrifos 12 (10%)	8 (75%)		
	Transfer (%) dermal press, (n=2)	Transfer (%) dermal press		
	Diazinon 1200 (0)	204 (582%)		
	Malathion 61 (0)	−55 (467%)		
	Chlorpyrifos 269 (5%)	113 (72%)		

[a]Mean (n=2, if not expressed).
[b]RSD (SD/mean×100%).
[c]Contact time is 10 min except for difference vs. food analysis which is 1 min.

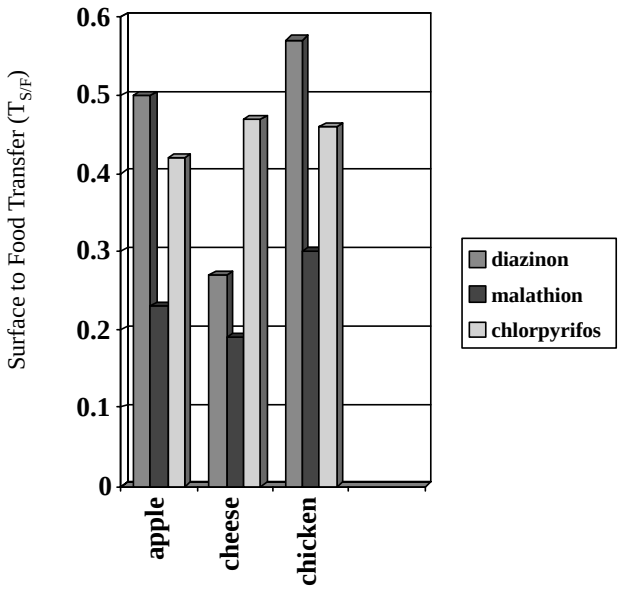

Figure 2. Transfer efficiencies from wood surfaces to food.

socioeconomic conditions. Therefore, information is needed about the relative importance of major conditions and the ranges in the values for the transfer efficiencies, and the sensitivity of the predicted dietary intake associated with changes in the various variables.

Prior to conducting the transfer studies, performance testing of the PDC was conducted. The spray liquid deposition rate was determined by weighing the liquid deposited onto aluminum weighing boats placed in the spray deposition chamber with the test coupons. In the first set of experiments conducted to test spray deposition variation, the liquid deposition was monitored by weighing the boats following the spraying of multiple sets of coupons with water at a loading rate of 11 mg/cm^2. The maximum variation within a coupon (n=3) was measured to be 11.3% RSD, with a median RSD of 8.9%. The maximum variation across coupons (n=3) was 4.4% RSD, with a median RSD of 3.9%. In the second set of experiments, Dursban Pro was

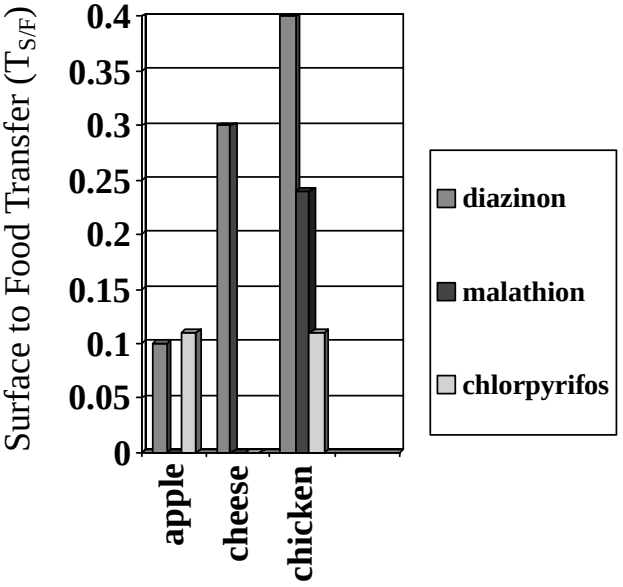

Figure 3. Transfer efficiencies from carpet surfaces to food.

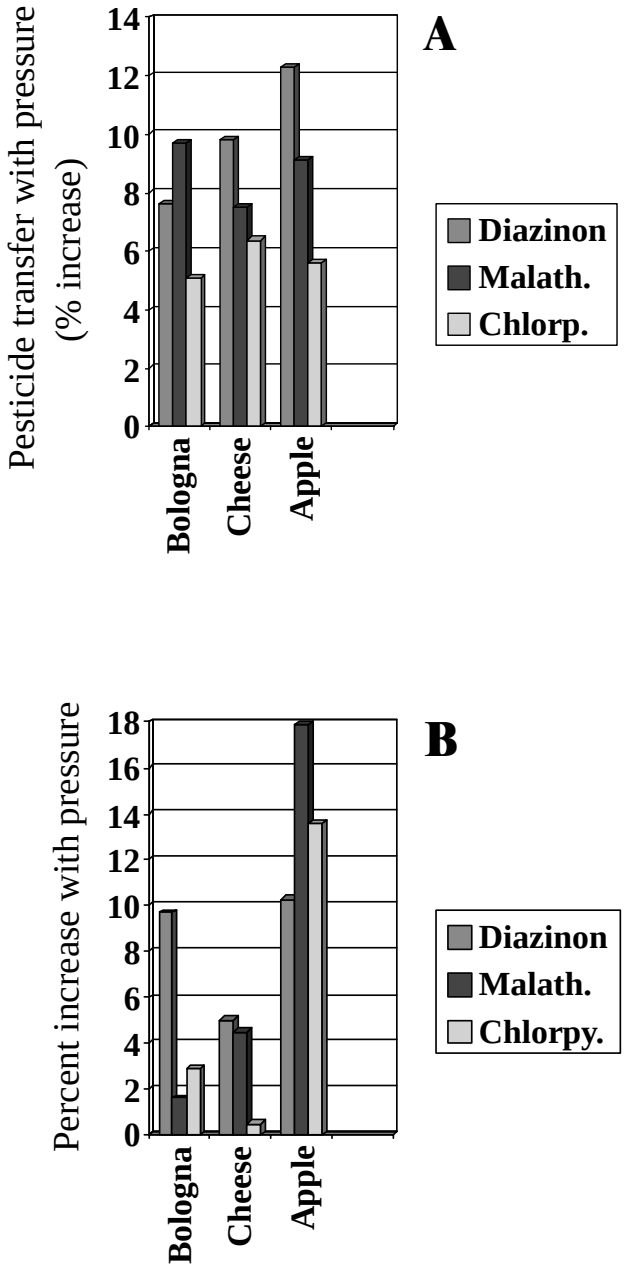

Figure 4. (A) Effect of pressure on tile. (B) Effect of pressure on transfer from hardwood.

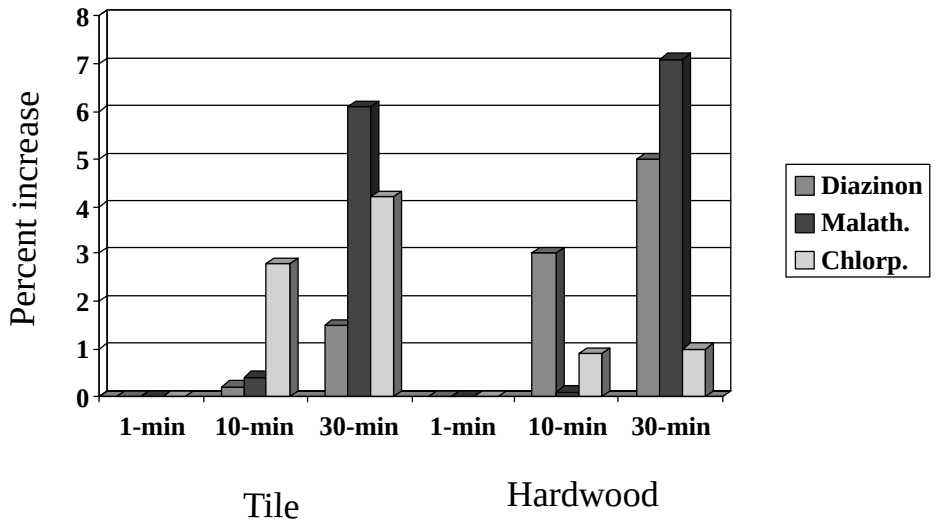

Figure 5. Effect of time on transfer efficiencies.

added to the water at a rate of 9.0 ml in 200 ml of water to determine if the emulsifier in the solution had a measurable effect. The loading rate for this experiment was 6.47 mg/cm^2. In this experiment, the maximum variation within a coupon (n=3) was measured to be 11.2% RSD, with a median RSD of 8.4%. The maximum variation across coupons (n=3) was 8.1% RSD, with a median RSD of 6.4%. These experiments indicated that the variation in spray pattern due to the PDC was about 10%. Using the method of pipetting, the pesticide solution onto the test surfaces indicated that the variation in the deposition pattern was 10–14%.

Table 5 presents the results of the range-finding experiments for two surfaces (hardwood and carpet) and two food items (ham and apple) and the experimental conditions related to the experiments. The last two columns provide comparison of results between the method of analysis of the food sample for the amount of pesticide transferred in comparison with the procedure of measuring the difference between the amount of residue remaining on the surface of one coupon subtracted from the amount measured on the control coupon. It should be noted that these comparisons were made from experiments conducted with 1-min contact times. Note that the transfer efficiencies are lower than those measured for the 10-min contact times.

Results of these *range-finding experiments* indicate that a higher pesticide transfer occurred from hard surfaces to food, with low transfer from soft surfaces, including carpet and cloth. Figures 2 and 3 illustrate how the transfer efficiencies can vary by food and pesticide on hardwood

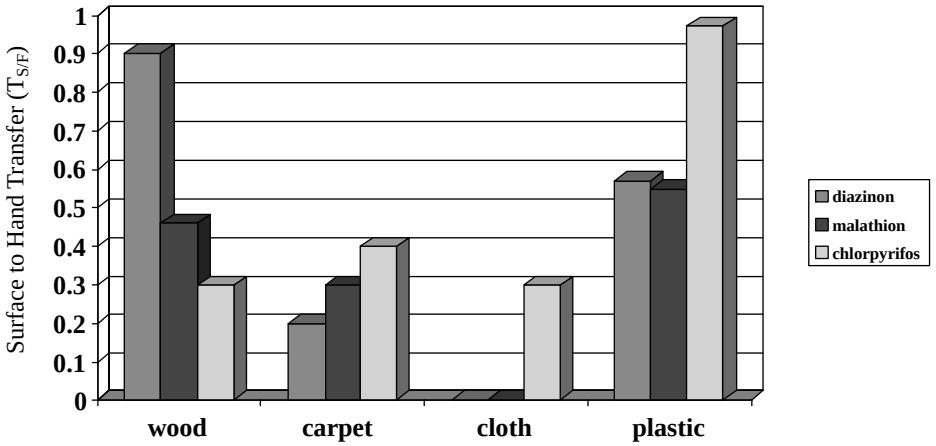

Figure 6. Surface-to-hand transfer.

(Figure 2) and carpets (Figure 3). A substantial increase in pesticide transfer is seen for both increased contact pressure (Figure 4A and B) and increased contact time (Figure 5). Transfer of all pesticides from hard surfaces generally increased by 5–15% when additional pressure was applied to the food. Similarly increased pesticide transfers were measured for contact times up to 30 min, as compared to a 1-min contact time.

Figure 6 illustrates how the EL sampler measurements — the surrogate measurement technique for dermal transfer — vary across surfaces and pesticide, with the hard materials yielding higher transfer efficiencies.

Quality control (QC) samples were analyzed during the transfer efficiency experiments. These QC samples consisted of sample blanks, food method controls, food matrix, and food matrix spikes. To monitor the extraction results for all surface samples collected from surface materials, a surrogate standard, 4,4-dichlorobiphenyl, was added to each sample, sample control, and sample blank prior to extraction. Blanks for all wipes were below the method detection limit of 5 ng/ml, and only the carpet blank indicated a finite value of 7.4 ng/ml for diazinon. Controls (spiked at 50 ng/ml) were within ±5% for wipes, ±15% for carpet, and ±25% for cloth. Recovery results from the surrogate ranged from 70% to 130%, which was considered acceptable.

Transfer efficiencies are highly variable depending on the method used to characterize pesticides available for transfer from the surfaces (wipes/dermal press/applied concentrations). It should be noted that the concentrations of the pesticides that were applied to the test surface generally were much greater than the measured surface concentration. The difference between the applied surface concentration and the measured surface concentration was dependent on the surface type and generally increased as a function of the pesticide solution drying and contact time with the surface. Several reasons for these differences are hypothesized to be due to pesticides binding with the test surface material, which make them more difficult to extract, and/or volatilization of the pesticide during the drying process. It also was possible to measure higher pesticide levels in foods (measured by either the difference method or directly by measuring what was in the food) than levels available for transfer, as measured by either the surface wipe or dermal press measurements, resulting in transfer efficiencies greater than 100%. A standardized surface measurement is needed to provide a meaningful baseline for quantifying food/surface and hand/surface transfer efficiencies. The method must be amenable to laboratory and residential surface measurements. Additional transfer efficiency measurements are focusing on hard surfaces and foods, in particular moist food types, that transfer pesticides most readily. An additional phase of laboratory experimentation will be the application of pesticide-laden particles onto test surfaces to determine how wet or dry deposition of the pesticide onto the surface affects the transfer efficiency.

Children's Behaviors

The time range of eating events (meals or snacks) varied from 129 to 3318 s (2–55 min), with an average of about 20 min per eating event. The average time the food was in contact with the child's hand was 134 s for each eating event, with food placed on a surface other than a plate on

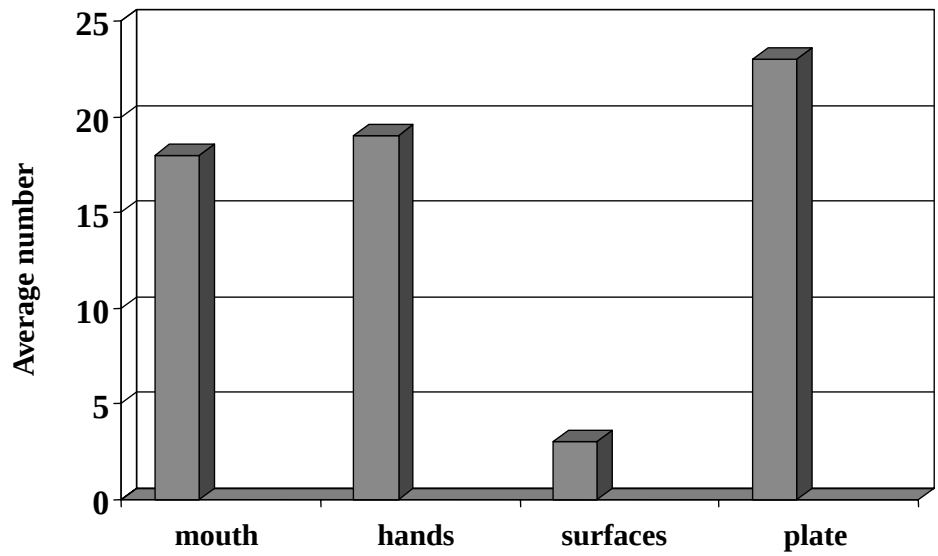

Figure 7. Average frequency of contact between food and objects.

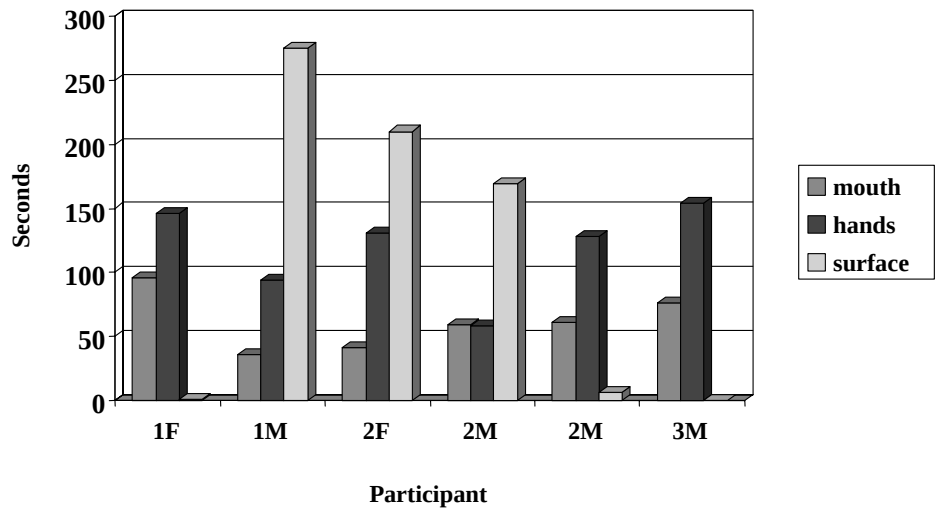

Figure 8. Average contact time between food and objects.

average 117 s. The average number of contact times between food and hands averaged 19 times, but ranged from 10 to 39 over the six children (used for the term $A_{H/F}$ in the model). A specific food item contacting the child's hands during an eating event depended on the type of food eaten and age (where older children may use an eating utensil). For example, a 2-year-old male handled a bologna slice for a total of about 100 s and a banana for about 500 s — a factor of 5. The children contacted 2–10 surfaces during their eating events, with each surface in contact with the hand from 1 to 39 times ($A_{S/H}$ in the model). The food item contacted one to three surfaces during a typical eating event, with contact of a specific surface ranging from 0 up to 32 times ($A_{S/F}$ in the model).

Figure 7 illustrates the average number of times, across the eating events, food came in contact with the mouth, hands, surfaces, and plate. It can be seen that food comes in contact with surfaces, other than the plate, less than five times on average per eating event. In Figure 8, the average time of food contact with mouth, hands, and surfaces (other than plate) by the six children is illustrated. Note that for some children, food can be in contact with surfaces (other than a plate) for over 200 s whereas for other children, food does not come in contact with surfaces other than the plate during the eating event. The average time food was in contact with a plate was 577 s, which is the basis for the 10-min contact time used in the transfer experiments.

One of the difficulties associated with analysis of video data is the complexity of multiple food items, hands, and to a lesser extent, surfaces that make videotaping and processing difficult. The percent of time that objects were not in view, on average, was 40.5% for food and object, and 10.25% for hand and object (which should be less because the hand is the primary target for the camera).

Prior to undertaking the videotaping, there was a general concern that the normal behavior of the child (or children in day care centers) would be modified by the presence of the camera and the addition of a "stranger" to the normal environment. However, after a brief period of adjustment (usually less than 15 min), the "curiosity factor" was overcome by hunger or the normal routine. It should be noted that an experienced child behavior specialist, who was trained in ways to establish rapport with the child and his/her family or the day care center children and staff, was utilized for this purpose. This probably made the transition easier.

Capturing the activity patterns for six children on 2 different days does not provide a representative sample of either the six children or any other child. However, the results capture a representation of events that can provide actual data for use in modeling exposures.

Conclusions

The dietary contribution to aggregate exposure is potentially an important route of exposure, especially for young children who may eat foods with contaminated hands and foods dropped or eaten from contaminated surfaces. This research is an attempt to ascertain the potential significance of children's eating behaviors on their total dietary exposure. One of the challenges of this research was to develop a method for estimating the contribution of the various children's behaviors that could contribute to this route of exposure. The research approach taken was to collect a snap shot of children's microactivity data during actual dietary events, both in the home and day care settings. These data, although limited

to a few children during only 2 days of their lives, served to provide information about what were the primary contact surfaces and food items to study, as well as getting detailed information about contact frequency and duration for various eating events. Results from this research suggest that children spend a significant portion of their awake hours engaged in eating activities, approximately 20% of their awake hours for some young children. During these eating events, their hands may become contaminated from contact with surfaces (measured to occur at a frequency of up to 32 times during an eating event). These hands then contact the food prior to entering the mouth (measured to occur at a frequency of 10–39 times during the eating event). These observations verify that there is a potential for contaminant transfer to the food which results from the child's eating activity.

In the laboratory, studies were conducted to determine, for a very limited number of surfaces and food types, the amount of transfer of several commonly used pesticides. Experiments, which would mimic the contact observed in the videotapes from surfaces to food and from surfaces to a surrogate material for hands, were conducted. The laboratory studies were examined with respect to a range of input parameters for input into a dietary ingestion model developed for this research. It was observed that up to 50% of the material measured as deposited onto hard surfaces could be transferred to food and up to 90% from hard surfaces to the surrogate used for hand transfer. However, the amount of transfer depended on type of surface, type of food item, the pesticide, amount of contact time, contact pressure, and probably other physical and chemical factors. These general observations were also confirmed by an independent laboratory using different procedures for depositing the pesticide solution onto the surfaces.

Computer simulations were conducted using the collected activity, the laboratory transfer data, and literature results as input factors to the dietary model to estimate the overall intake of a pesticide for single servings of different food items and the relative importance of each model term to total pesticide intake. These limited data indicate that under conditions of high surface loading of pesticide residue, handling of foods while eating by a young child can be a significant contribution to excess dietary exposure to pesticides. In addition, results from time and pressure studies indicate that the model should be enhanced to incorporate contact time (terms 2 and 3) and pressure. In field studies designed to actually measure the total contribution of dietary exposure, there is a need to capture the amount of time food and/or hand(s) are in contact with a surface and whether there is additional pressure placed on the food or with the hand. This information is not quantitative from observational techniques (videos), but qualitative information may be available for interpretation.

The conclusions in this paper are based on small numbers of children and laboratory experiments which should be viewed with caution until more data are collected. Further research is underway to evaluate the model results and laboratory experiments in a field experiment involving children which will measure environmental concentrations on surfaces and hands, as well as air, water, soil, and food; collect microactivity data related to dietary events; measure transfer of residue into the foods that are eaten or touched; and relate these data to internal dose measurements of actual exposure.

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