1.0 INTRODUCTION

1.1 Background

The U.S. Environmental Protection Agency (U.S. EPA) has pledged to increase its efforts to provide a safe and healthy environment for children by ensuring that all EPA regulations, standards, policies, and risk assessments take into account special childhood vulnerabilities to environmental toxicants. Children are behaviorally and physiologically different from adults. Their interaction with their environment, through activities such as playing on floors, and mouthing of hands and objects, and handling of food, may increase contact with contaminated surfaces. Proportionately higher breathing rates, relative surface area, and food intake requirements may increase exposure. Differences in absorption, metabolism, storage, and excretion may result in higher biologically effective doses to target tissues. Immature organ systems may be more susceptible to toxicological challenges. Windows of vulnerability, when specific toxicants may permanently alter the function of an organ system, are thought to exist at various stages of development.

Children are exposed to a wide variety of chemicals in their homes, schools, daycare centers, and other environments that they occupy. The chemicals to which they are exposed may originate from outdoor sources, such as ambient air contaminants, indoor sources such as building materials and furnishings, and from consumer products used indoors. One category of consumer products to which children may be exposed is pesticides that are used to control roaches, rats, termites, ants, and other vermin. Despite widespread residential and agricultural use of pesticides, only limited measurement data are available for pesticide levels in environments that children occupy and little is known about the factors that impact children's exposures to pesticides. The Food Quality Protection Act (FQPA) of 1996 requires EPA to upgrade the risk assessment procedures for setting pesticide residue tolerances in food by considering the potential susceptibility of infants and children to both aggregate and cumulative exposures to pesticides. Aggregate exposures include exposures from all sources, routes, and pathways for individual pesticides. Cumulative exposures include aggregate exposures to multiple pesticides with the same mode of action for toxicity. FQPA requires risk assessments to be based on exposure data that are of high quality and high quantity or on exposure models using factors that are based on existing, reliable data.

EPA's Office of Research and Development (ORD) is responsible for conducting research to provide the scientific foundation for risk assessment and risk management at EPA. In 2000, ORD released its *Strategy for Research on Environmental Risks to Children* addressing research needs and priorities associated with children's exposure to environmental pollutants and providing a framework for a core program of research in hazard identification, dose-response evaluation, exposure assessment, and risk management.

The National Exposure Research Laboratory (NERL) in ORD is working to achieve three specific objectives of the *Strategy* through its children's exposure research program: (1) develop improved exposure assessment methods and models for children using existing information; (2) design and conduct research on age-related differences in exposure, effects, and dose-response relationships to facilitate more accurate risk assessments for children; and (3) explore opportunities for reducing risks to children. After an exhaustive review of the volume and quality of the data upon which default assumptions for exposure factors are based (Cohen Hubal *et al.*, 2000a), a framework for systematically identifying the important sources, routes, and pathways for children's exposure was developed (Cohen Hubal *et al.*, 2000b).

This framework (Figure 1.1), based on a conceptual model for aggregate exposure, provides the foundation for a protocol for measuring aggregate exposures to pesticides (Berry *et al.*, 2001) and for developing sophisticated stochastic models (Zartarian *et al.*, 2000). Using the framework, four priority research areas, representing critical data gaps in our understanding of environmental risks to children, have been identified:

(1) Pesticide use patterns;
(2) Spatial and temporal distribution in residential dwellings;
(3) Dermal absorption and indirect (non-dietary) ingestion (including micro- and macro-activity approaches); and
(4) Direct ingestion.

Several targeted studies were designed and conducted to address these research needs. These include laboratory studies, small pilot field studies, and large collaborative observational studies. These studies aimed to: (1) evaluate methods and protocols for measuring children's exposure, (2) collect data on exposure factors to reduce the uncertainty in exposure estimates and risk assessments, and (3) collect data for use in exposure model development and evaluation.

1.2 Purpose of the Report and Intended Audience

This document is a comprehensive summary report of data collected under or otherwise related to the NERL children's exposure research program. Data are compared across studies and across compounds to identify or evaluate important factors influencing exposures along each relevant pathway. Summary statistics, comparative analyses, and spatial and temporal patterns are presented to address previously identified data gaps. The primary purpose of this document is to identify factors that are most important for children's exposures to pesticides. The objectives of this document are to:

- Compare results across studies in order to identify trends or similar observations that might provide a better understanding of the factors affecting children's exposures;
- Describe recent children's exposure studies conducted or funded by NERL, including descriptions of the parameters measured and the measurement methods;
- Provide concentration data and summary statistics for comparison of the studies; and
- Present highlights of the results of the studies.

The document was completed with input from staff in the EPA Program Offices, NERL researchers, and Science to Achieve Results (STAR) program grantees who gathered at the US EPA National Exposure Research Laboratory's Workshop on the Analysis of Children's Measurement Data (Tulve *et al.*, 2006) in September 2005 to discuss data presented in a draft summary report, assess the suitability of the data for testing key hypotheses, and propose additional analyses.

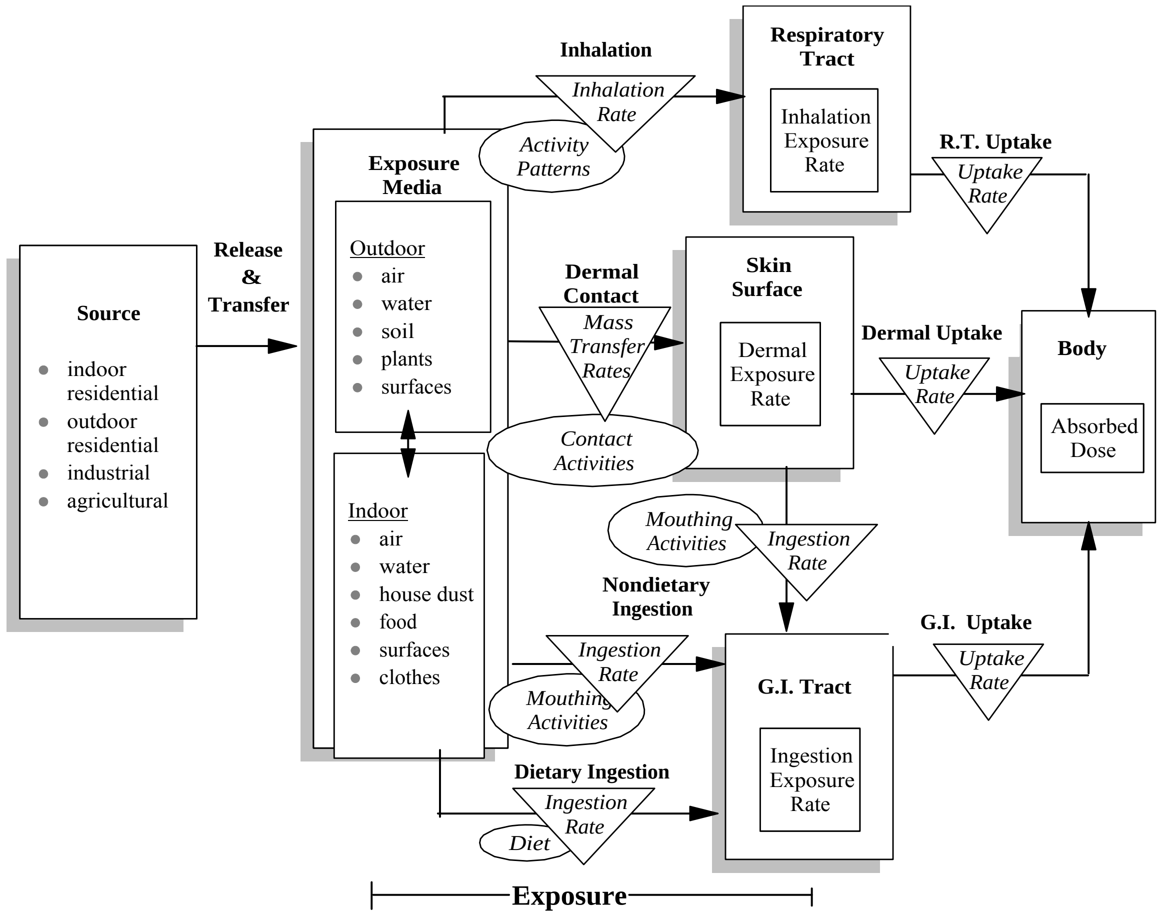

Figure 1.1 Modeling framework for children's pesticide exposure from Cohen Hubal *et al.* (2000b).

The document is intended for an audience of exposure scientists, exposure modelers, and risk assessors. Exposure scientists will find a useful evaluation of available sampling methods for all media relevant to children's exposures. Exposure modelers will be able to use the data to develop or improve probabilistic multimedia, multi-pathway human exposure models. Most significantly, the report may be used by EPA Program offices such as the Office of Pesticide Programs (OPP), the Office of Pollution Prevention and Toxics (OPPT), and the Office of Children's Health Protection (OCHP) to enhance the Agency's risk assessment activities by replacing default assumptions with high-quality, real-world data. Fewer default assumptions will lead to more accurate assessments of exposure and risk and will bolster ensuing risk reducing actions. Furthermore, by examining relationships among application patterns, exposures, and biomarkers for multiple compounds from different classes of pesticides, this report contributes to the development of more reliable approaches for assessing cumulative exposure. Some of the analyses and comparisons that are presented in this summary report include the following:

- Comparison of concentrations
- Spatial variability
- Temporal variability
- Regional comparisons
- Urban versus rural
- Home versus daycare
- Indoor versus outdoor
- Parent compound versus metabolite
- Effect of physical and chemical properties
- Impact of air exchange rate
- Effect of surface type
- Effect of surface concentration
- Effect of sampling method

Comparisons between studies may involve different numbers of measurements, different sampling strategies and methods, and different chemical analysis methods

1.3 Structure of the Report

This document presents data from studies to evaluate children's exposure to pesticides, spanning from pesticide use patterns, through concentrations in exposure media, to biological markers of exposure. The exposure media are listed in an order that roughly mirrors the complexity of the exposure mechanism; that is, beginning with inhalation exposure and ending with dermal exposure. At the beginning of each section, available data from the relevant studies are listed. Results are presented in tables and graphs to illustrate the available data and to facilitate comparisons both across studies and across pesticides.

Throughout the document, lognormal probability plots ("logplots") and box-and-whisker plots ("box plots") are used to graphically depict and compare distributions of concentrations or surface loadings. The logplots are used to compare results only from large observational field studies and the boxplots are used to compare results from the focused studies against each other and against the large observational field studies. In the lognormal probability plot, the ordered values of the measured concentration are plotted on a log-scale vertical axis, and the percentiles of the theoretical normal distribution are plotted on the horizontal axis. If the points in the plot form a nearly straight line, the data are approximately lognormal. The box-and-whisker plot is actually a group of side-by-side box-and-whisker plots along the x-axis, each representing a different study. The upper whisker extends to the maximum value, the upper edge of the box represents the 75th percentile, the line inside the box represents the median (50th percentile), the lower edge of the box represents the 25th percentile, and the lower whisker extends to the minimum value. Note that the vertical axis is log-scale.

1.4 Data Treatment

Values that are below the method detection limit (MDL) are common in environmental data sets. All values above the MDL are statistically different from zero; however, values near the MDL are generally less accurate than those much higher than the MDL. Laboratories often report a second limit, the Method Quantitation Limit (MQL), as the smallest amount that can be *reliably quantified* in a sample. Despite the higher relative uncertainty in values between the MDL and the MQL, all values above the MDL are retained for the purposes of this document. Values below the MDL are treated using simple substitution, wherein they are replaced with a fraction of the detection limit ($MDL/\sqrt{2}$), a common practice originally proposed by Hornung and Reed (1990). These substituted values are used in all statistical analyses performed specifically for this report and are presented in all data plots, except for lognormal probability plots, in which these substituted values were judged by the authors to be misleading. Detection frequencies (that is, the percent of measurements above the MDL) are presented for each compound by each relevant sampling method at the beginning of each chapter.

Sampling weights are available for all of the large-scale observational field studies, but, unless otherwise noted, only unweighted concentrations are presented in this report. Summary statistics based on unweighted observations may not provide as valid an estimate of true study population values as those based on weighted observations, but are used nonetheless to maintain consistency in comparisons with studies for which weights are not available. In all cases where a statistical test was done to assess differences, the name of the test and the resulting p-value are presented.

1.5 Description of the Studies and Data Collected

Data are included in this report from the following studies. (The acronyms in parentheses are used in the Tables and Figures of this report.)

- National Human Exposure Assessment Survey in Arizona (NHEXAS-AZ)
- Minnesota Children's Pesticide Exposure Study ("MNCPES")
- Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants ("CTEPP")
- First National Environmental Health Survey of Child Care Centers ("CCC")
- Biological and Environmental Monitoring for Organophosphate and Pyrethroid Pesticide Exposures in Children Living in Jacksonville, Florida ("JAX")
- Center for the Health Assessment of Mothers and Children of Salinas Quantitative Exposure Assessment Study ("CHAMACOS")

- Children’s Pesticide Post-Application Exposure Study (“CPPAES”)
- Distribution of Chlorpyrifos Following a Crack and Crevice Type Application in the US EPA Indoor Air Quality Research Test House (“Test House”)
- Pilot Study Examining Translocation Pathways Following a Granular Application of Diazinon to Residential Lawns (“PET”)
- Dietary Intake of Young Children (“DIYC”)
- Characterizing Pesticide Residue Transfer Efficiencies (“Transfer”)
- Food Transfer Studies (“Food”)
- Feasibility of Macroactivity Approach to Assess Dermal Exposure (“Daycare”)

All studies involving children were observational research studies, as defined in 40 CFR Part 26.402. All study protocols and procedures to obtain the assent of the children and informed consent of their parents or guardians were reviewed and approved by independent Institutional Review Boards (IRBs) and complied with all applicable requirements of the Common Rule (45 CFR 46) regarding additional protections for children (Subpart D). Further, all protocols regarding recruitment and treatment of participants were reviewed by the EPA Human Subjects Research Review Official (HSRRO) to assure compliance with the Federal Policy for the Protection of Human Subjects.

The studies discussed in the report included large observational studies, such as NHEXAS-AZ, MNCPES, CTEPP, and CCC, small pilot-scale observational studies (*e.g.*, JAX, CPPAES, DIYC, CHAMACOS, and Daycare), and laboratory studies (*e.g.*, Test House, Transfer, and Food).

- MNCPES, NHEXAS-AZ, CTEPP, and CCC were large observational exposure measurement studies with survey designs that involved random sampling. The CCC study was a nationwide survey and the others had a regional focus. Sampling weights are available for all of these studies, but, unless noted otherwise, only unweighted concentrations are presented in this report.

- The small pilot-scale observational studies are small-scale field studies, such as JAX and CHAMACOS, which were performed to evaluate methods for conducting aggregate exposure assessments for pesticides and to collect preliminary data that could be used to assist in the design of larger observational studies. Like the large observational studies, some of these smaller studies included measurements of multiple chemicals in multiple media.

- The laboratory studies consisted of experiments under controlled conditions to evaluate factors affecting transfer from surfaces (Transfer and Food studies). The Test House study investigated the fate and transport of chlorpyrifos following a crack and crevice application and provided valuable information on spatial and temporal variability of surface concentrations in the absence of human activity.

During these studies, the following types of measurements were collected (not all types of samples were collected in all studies):

- Air (indoor and outdoor)
- Soil
- House dust – Floors (carpet and hard surface)
- Surface wipes (including eating and food preparation surfaces)
- Transferable residues (*e.g.*, polyurethane foam roller, C18 press)
- Hand wipes
- Dermal surrogates (cotton garment and socks)
- Duplicate diet (solid food, beverages)
- Handled food
- Urine

Information was also typically collected by questionnaire on:

- Housing characteristics
- Participant characteristics
- Children's activities (timelines and logs)
- Recent pesticide use

The types of media sampled and questionnaires administered in each study are listed in Table 1.1. Other than the pesticide inventory and use questionnaires, questionnaire data are not the focus of this document.

1.6 Pesticides of Interest to this Report

The studies presented here were performed when a number of organophosphate and pyrethroid pesticides were in use; thus numerous pesticides from various chemical classes (including insecticides and herbicides) were measured. All measured insecticides (and insecticides synergists) are listed in Table 1.2, although not all of the studies collected data for all of the insecticides listed. To reduce complexity, this report focuses on the most commonly detected organophosphate and pyrethroid insecticides:

- Chlorpyrifos
- Diazinon
- Permethrin
- Cyfluthrin

Table 1.1 Available media, participant characteristics, and activities by study.

	NHEXAS-AZ	MNCPES	CTEPP	CCC	JAX SCREENING	JAX AGGREGATE	CHAMACOS	CPPAES	TESTHOUSE	PET	DIYC	DAYCARE
Air - Indoor	✓	✓	✓			✓	✓	✓	✓	✓	✓	
Air - Outdoor	✓	✓	✓			✓	✓				✓	
House Dust	✓		✓				✓					
Surface Residue Wipes	✓		✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
LWW Surface Sampler		✓						✓				
Transferable Residues	✓	✓	✓	✓		✓	✓		✓	✓	✓	✓
Hand Wipes	✓	✓	✓					✓		✓	✓	✓
Cotton Garments/Socks						✓	✓	✓		✓		✓
Soil		✓	✓	✓			✓			✓		
Duplicate Diet	✓	✓	✓			✓					✓	
Handled Foods											✓	
Urine	✓	✓	✓		✓	✓		✓		✓	✓	
Housing Characteristics	✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	
Participant Characteristics	✓	✓	✓	✓	✓	✓		✓		✓	✓	✓
Children's Activities	✓	✓	✓			✓		✓		✓	✓	✓
Recent Pesticide Use		✓	✓	✓	✓	✓						
Pesticide Inventory		✓			✓	✓						

Table 1.2 Pesticides and metabolites measured in the studies.

Pyrethroid	**Organophosphorus**		**Other**
Allethrin	Acephate	Ethyl parathion	Fipronil
Bifenthrin	Azinphos-methyl	Fonofos	Piperonyl butoxide
Cyfluthrin [a]	Chlorpyrifos [a]	Malathion	TCPy [a b]
Cyhalothrin	Chlorpyrifos-oxon	Malathion-oxon	IMP [a c]
Cypermethrin	Demeton-S	Methamidophos	3-PBA [a d]
Deltamethrin	Diazinon [a]	Methidathion	
Esfenvalerate	Diazinon-oxon	Methyl-parathion	
Permethrin [a]	Dichlorvos	Mevinphos	
Pyrethrins	Dimethoate	Naled	
Resmethrin	Disulfoton	Phosmet	
Sumithrin	Ethion		
Tetramethrin			
Tralomethrin			

[a] Pesticides and metabolites of primary interest in this document
[b] 3,5,6-Trichloro-2-pyridinol, a selective metabolite of chlorpyrifos
[c] 2-Isopropyl-4-methyl-6-hydroxypyrimidine, a specific metabolite of diazinon
[d] 3-phenoxybenzoic acid, a metabolite common to many pyrethroids

1.7 Summary Descriptions of the Studies

Individual study details are listed in Appendix B. Journal articles presenting results of these studies are listed in the Bibliography. The studies are summarized below.

The National Human Exposure Assessment Survey in Arizona (NHEXAS-AZ) was performed in collaboration with the University of Arizona, the Illinois Institute of Technology, and Battelle Memorial Institute. Probability-based samples were collected in each of Arizona's 15 counties from December 1995 to March 1997. Although 176 households participated, this report only includes data from 21 households with children ages 6-12 as primary participants. Environmental samples included indoor and outdoor air (3-day integrated samples), personal air (1-day), vacuumed surface dust, and window sill wipes. Personal samples included 24-hour duplicate diet and hand wipes. Biological samples consisted of urine samples (first morning void). Baseline and follow-up questionnaires and time-activity diaries captured activity patterns. Two pesticides (and their metabolites) were of primary interest, namely chlorpyrifos (TCPy) and diazinon, and two pesticides (and their metabolites) were of secondary interest, namely malathion (MDA) and carbaryl (1-naphthol).

The Minnesota Children's Pesticide Exposure Study (MNCPES) was an observational measurement study performed in collaboration with Research Triangle Institute (RTI), the Environmental and Occupational Health Sciences Institute (EOHSI), the Minnesota Department of Health, and the University of Minnesota. A telephone survey and in-home interviews were used to collect data on pesticide storage and use patterns from 308 households in both urban centers (Minneapolis/St. Paul) and rural counties (Goodhue and Rice) during the summer of 1997. Probability-based sampling weights were developed and intensive environmental and personal monitoring were performed for 102 children, ages 3-13. Households reporting more frequent pesticide use were oversampled. Environmental samples included personal, indoor, and outdoor air (6-day integrated), surface dust (wipe and press), surface soil, and tap water. Personal samples included solid food (4-day composite), beverages (4-day composite), hand rinse, and first morning void urine (days 3, 5, and 7). In addition to questionnaires and diaries, videotaping was performed in a subset of 20 homes. Four primary pesticides (and their metabolites), namely chlorpyrifos (TCPy), atrazine (atrazine mercapturate), malathion (malathion dicarboxylic acid), and diazinon, and 14 secondary pesticides were measured, along with 13 polycyclic aromatic hydrocarbons (PAHs).

The Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP) Study (Morgan *et al.,* 2004) was performed in collaboration with Battelle Memorial Institute as an observational study of preschool children's exposure to contaminants in their everyday environments (*i.e.*, homes and daycare centers). Monitoring was performed from July 2000 to March 2001 in North Carolina (spanning summer, fall, and winter) and from April 2001 to November 2001 in Ohio (spanning spring, summer, and fall). The study population consisted of 257 children, ages 18 months to five years, and their primary adult caregivers (130 children, 130 homes, and 13 daycare centers in North Carolina; 127 children, 127 homes, and 16 daycare centers in Ohio). Samples were collected over a 48-hr period at each home and daycare center, including indoor air, outdoor air, floor dust, soil, hand wipe, solid food, liquid food, and urine. Supplemental information included a recruitment survey, a house/building characteristics survey, pre- and post monitoring questionnaires, and activity and food diaries. In addition, 20% of the

OH participants were videotaped at home for about 2 hours. Additional samples (hard floor and food preparation surface wipes and transferable residues) were collected if the participant reported indoor or outdoor applications of pesticides within 7 days of the monitoring period.

The First National Environmental Health Survey of Child Care Centers (CCC) was performed in collaboration with HUD (US Department of Housing and Urban Development) and CPSC (US Consumer Product Safety Commission). Samples were collected from August through October (summer and fall) 2001, at 168 randomly-selected child care centers nationwide. Many facilities reported recent pesticide application (either by professionals or by employees). Samples included soil, surface wipes, and transferable residues (C18 Press). A multi-residue chemical analysis method was used to measure a large suite of current-use pesticides. The study aimed to collect data on pesticide use practices and to characterize the distributions of pesticide concentrations in a nationally-representative sample of child care centers in the U.S.

The study titled Biological and Environmental Monitoring for Organophosphate and Pyrethroid Pesticide Exposures in Children Living in Jacksonville, Florida (JAX) was performed in collaboration with CDC (Centers for Disease Control and Prevention) and DCHD (Duval County Health Department) in Jacksonville (Duval County), Florida, from August through October (summer and fall) 2001. The CDC performed a biomonitoring study to measure metabolites of organophosphate and pyrethroid pesticides in a sample of 200 children who were 4-6 years of age. The DCHD conducted a home screening survey in a subset of 42 of the homes. The screening phase employed a pesticide screening inventory, surface wipes, and urine collection. The EPA conducted an observational study in a subset of nine of the homes to evaluate sampling and analysis methods and protocols for conducting aggregate exposure estimates for children. The aggregate exposure study included the pesticide screening inventory, surface wipes, indoor and outdoor air, cotton garment, duplicate diet, and transferable residue measurements, a time activity diary, and a urine sample.

The Center for the Health Assessment of Mothers and Children of Salinas (CHAMACOS) Quantitative Exposure Assessment Study was a collaboration with the University of California at Berkeley. This observational study was performed in homes of agricultural workers living in Salinas, California. Twenty households with children ages 5 months to 3 years old (10 female and 10 male) were monitored during the period of June to October (summer and fall) 2002. Samples were collected over a 24-hour monitoring period and included indoor and outdoor air, house dust, transferable residues from floors (surface wipes and press samples), transferable residues from toys (surface wipes), urine, and cotton union suits and socks. A time/activity diary was also administered. The objective of the study was to evaluate sampling and analysis methods and study protocols that might be applied in larger studies such as the National Children's Study.

The Children's Pesticide Post-Application Exposure Study (CPPAES) was a collaborative field study with EOHSI (Environmental and Occupational Health Sciences Institute) in urban New Jersey over a two-year period stretching from April 1999 to March 2001. Ten homes with children 2-5 years of age participated. Each of the homes had a professional "crack and crevice"-type application of a chlorpyrifos-based formulation at the time of the study, but only trace amounts of chlorpyrifos were applied in three of the homes. The monitoring period typically lasted for two weeks with pre- and multiple post-application samples. Sampling was comprehensive with indoor air, deposition coupons, surface samples (LWW, Lioy-Weisel-Wainman sampler), toys, hand wipes, urine, air exchange rate, and time activity diary data collected throughout the study, and additional samples consisting of surface wipes, dermal wipes, cotton garments, and videotaped activities collected on the second day of the study.

A field laboratory study titled the Distribution of Chlorpyrifos Following a Crack and Crevice Type Application in the US EPA Indoor Air Quality Research Test House (Test House) was performed in collaboration with the National Risk Management Research Laboratory (NRMRL). The Test House is an unoccupied three-bedroom house in Cary, NC. The study investigated the translocation of chlorpyrifos and the spatial and temporal variability of chlorpyrifos levels in air and on surfaces following a professional "crack and crevice"-type application onto the floor and cabinetry of a kitchen. Samples included air, polyurethane foam (PUF) roller, carpet sections, C18 surface press, and surface wipes from multiple rooms. Samples were collected pre-application and on days 1, 3, 7, 14 and 21 post-application.

The Pilot Study Examining Translocation Pathways Following a Granular Application of Diazinon to Residential Lawns (PET) was performed during spring 2001 in six residential homes within a 50-mile radius of Durham, NC. Measurements were performed at homes where a homeowner applied a turf application of a granular formulation of diazinon. Sampling included indoor air (multiple rooms), PUF roller (outdoor and indoor), soil, doormat, high-volume small surface sampler (HVS3), dermal surrogate (cotton gloves), urine (adult and child), dog fur clippings, dog paw wipes, dog blood, and videotaping (15-min). Samples were collected pre-application and 1, 2, 4 and 8 days post-application. A feasibility study was also performed in a single home. The study focused on pesticide translocation and exposure pathways.

The Dietary Intake of Young Children (DIYC) study was a small observational field study in collaboration with RTI. It included three homes where diazinon had been applied (two homes with commercial crack and crevice applications and one home with non-professional application) and took place between November 1999 and January 2000 (fall and winter). Collected samples included indoor air, outdoor air, surface wipes, hand wipes, surface press, food press, food samples, PUF roller, entry wipe, and urine. A primary goal of the study was to evaluate the potential for exposure to pesticides due to food preparation and handling in the home.

The Feasibility of the Macroactivity Approach to Assess Dermal Exposure (Daycare) study was another collaboration with RTI (Cohen Hubal *et al.*, 2006). In this field study, nine daycare centers were identified that reported routine pesticide applications as part of the center's pest control program. In each daycare, screening sampling was conducted to evaluate the distribution of transferable pesticide residues on floor surfaces in the area where children spent the most time. One daycare was selected for more intensive monitoring during the summer of 2001, following a series of regularly scheduled (monthly) applications. Surface sampling and videotaping of activities were conducted simultaneously with dermal surrogate (cotton garment) sampling to calculate dermal transfer coefficients.

The Characterizing Pesticide Residue Transfer Efficiencies (Transfer) studies evaluated parameters that are believed to affect residue transfer from surface-to-skin, skin-to-object, skin-to-mouth, and object-to-mouth. The collaboration with Battelle was a series of controlled laboratory studies using fluorescent tracers as surrogates for pesticide residues. The protocol involved applying fluorescent tracers to surfaces of interest as a residue at levels typical of residential pesticide applications, and then conducting controlled transfer experiments varying six parameters in a systematic fashion. Repetitive contacts with contaminated surfaces were used to measure the following transfers: hand to clean surface, hand to washing solution, and hand to mouth. In the mouthing trials, mouthing was simulated using saliva-moistened PUF material to measure mass of tracer transferred. Laboratory evaluations were performed to relate transfer of tracer to transfer of pesticides (Ivancic *et al.*, 2004; Cohen Hubal *et al.*, 2005).

The Food Transfer Studies were controlled laboratory experiments investigating pesticide transfer from household surfaces to foods and evaluating factors that have been identified as important, including surface type, duration of contact, surface loading, and contact pressure (applied force). Organophosphate, pyrethroid, and pyrazole insecticides were applied onto various household surfaces using a customized spray chamber. Pesticide transfer efficiencies were measured for three different foods, with standardized surface contact areas. Amounts of pesticide residue transferred to foods were compared to the amounts removed using surface wipes. Transfer efficiency (TE) was defined as the amount of pesticide recovered from the food item divided by the pesticide concentration or loading level.

1.8 Exposure and Dose Models

It is neither within the scope nor the intention of this report to provide a detailed discussion of the exposure and dose models that have been developed using these data or applied to these data. However, since human exposure research progresses through an iterative series of models and measurements, it is often necessary to refer to these models. Models are constructed using current knowledge and are subsequently used to identify areas of greatest uncertainty. Modeled results are used to direct the focus of the measurement studies to address those identified uncertainties. As newly collected data yields new knowledge, models are refined and the entire process repeats. At each iteration, real-world data replace default assumptions to produce more accurate assessments of exposure and risk. Throughout this document models are mentioned. "Algorithms" are the set of deterministic mathematical expressions developed in the *Draft Protocol for Measuring Children's Non-Occupational Exposure to Pesticides by all Relevant Pathways* (Berry *et al.*, 2001) to assess exposure by each route as a function of concentration and various exposure factors. The Stochastic Human Exposure and Dose Simulation (SHEDS) model (Zartarian *et al.*, 2000) is a physically-based, probabilistic model that predicts multimedia/ multipathway exposures and doses incurred eating contaminated foods, inhaling contaminated air, touching contaminated surfaces, and ingesting residues from hand- or object-to-mouth activities. It combines information on pesticide usage, human activities, environmental concentrations, and exposure and dose factors using Monte Carlo methods. The Exposure Related Dose Estimating Model (ERDEM) (Blancato *et al.*, 2004) is a physiologically-based pharmacokinetic (PBPK) model used to make reliable estimates of the chemical dose to organs of animals or humans. It solves a system of differential equations that describes the organ system, directly addressing the uncertainties of making route-to-route, low-to-high exposure, and species-to-species extrapolations when there are exposures to one or to multiple chemicals. The Children's Dietary Intake Model (CDIM) (Hu *et al.,* 2004) estimates total dietary exposure of children to chemical contaminants by accounting for excess dietary exposures caused by chemical contaminant transfer from surfaces and/or hands to foods prior to consumption.

2.0 PESTICIDE USE PATTERNS

Very limited data are available to EPA researchers on what pesticides are currently being used in non-occupational environments, where they are being used, and the frequency of use. The EPA has not conducted a large scale survey to collect data on pesticide use patterns in the U.S. since 1990, but use patterns are believed to have substantially changed since that time. The children's observational studies described in this report collected information on household pesticide use as ancillary information that could be used to address this serious data gap. Despite the limited coverage of geographic regions, a relatively small number of study participants, and the general lack of knowledge about the active ingredients in brand name products on the part of consumers, valuable information was obtained. The NERL studies described in this section covered a period from 1997 to 2001. The indoor residential use of chlorpyrifos was cancelled while data collection was still ongoing in several studies (JAX, CCC, and CTEPP).

The pesticides available to consumers or professionals for use in residential settings have changed over time. By the late 1980s the use of most organochlorine pesticides (*e.g.*, DDT, chlordane, dieldrin, and heptachlor) was severely restricted in the U.S. The organophosphate (OP) insecticides (*e.g.*, malathion, chlorpyrifos, and diazinon), appealing for their high insect toxicity, low costs, and low likelihood of pest resistance, quickly filled the void and became the pesticides of choice for both consumers and professional pest control operators (Karalliedde *et al.*, 2001). The popularity of pyrethroid insecticides increased throughout the 1990s because of the following favorable properties: higher insecticidal toxicity, lower mammalian toxicity, and more rapid environmental degradation (Baker *et al.*, 2004). Passage of the Food Quality Protection Act of 1996 led the EPA to consider aggregate childhood pesticide exposure. The OPs were the first class of pesticides whose tolerances were reassessed, leading to withdrawal of the registrations for indoor applications of chlorpyrifos and diazinon in 2001 and 2002, respectively, because of concern regarding the risk to children. Consequently, pyrethroids have become the leading residential insecticides. While household use of diazinon and chlorpyrifos is now restricted, these and other OPs are still widely used in agriculture, and some structural uses for chlorpyrifos, including the treatment of house foundations, are still approved.

2.1 Sources of Information

Important sources of information on pesticide use patterns in non-occupational environments include Market Estimates from EPA's Office of Pesticide Programs (US EPA, 2004), national pesticide usage surveys, the Residential Exposure Joint Venture (REJV), the National Health and Nutrition Examination Survey (NHANES), and published scientific literature.

The Office of Pesticide Programs uses proprietary data sources in producing "Market Estimates" of pesticide sales and use in various market sectors. According to their estimates, the annual amount of insecticide active ingredients used in the home and garden sector declined from 24 million pounds in 1982, to less than 13 million pounds in 1988. Although the figure rose to 17 million pounds between 1998 and 2001, it still represents a significant decline from the early 1980s. In contrast, the amount of herbicides applied steadily increased over the same period, nearly doubling from 37 million pounds in 1982 to 71 million pounds in 2001 (US EPA, 2004)