

# **Draft Protocol for Measuring Children's Non-Occupational Exposure to Pesticides by all Relevant Pathways**

# **Draft Protocol for Measuring Children's Non-Occupational Exposure to Pesticides by all Relevant Pathways**

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## Abstract

In support of the Food Quality Protection Act (FQPA) of 1996, research is being conducted by the U.S. EPA National Exposure Research Laboratory to develop methods, data, and models for evaluating children's aggregate exposure to pesticides by all relevant pathways. The FQPA requires the EPA to use exposure assessments in the pesticide tolerance setting process. The exposure assessments must consider the aggregate exposures of infants and children from all sources (food, water, soil, dust, and air) and routes (inhalation, dermal exposure, indirect ingestion, and dietary ingestion). FQPA requires that risk assessments must be based on exposure data that are of high quality and high quantity or exposure models using factors that are based on existing, reliable data. Currently, the data on children's exposures and exposure factors are limited and generally not adequate to assess residential exposures to consumer products and environmental contaminants. Several general areas of research are needed to improve the quality and quantity of data available for exposure assessments for children. Appropriate age and developmental benchmarks for categorizing children in exposure assessments must be identified. The activity pattern data for children (especially very young children) required to assess exposure by all routes need to be developed. Methods for measuring children's exposures need to be developed and improved. Finally, field studies are needed to develop distributions of exposure and associated exposure factors.

The goal of this document is to provide guidance for generating data that can be used to improve exposure assessments for young children, as required by FQPA. Currently, standard protocols for conducting exposure field studies that provide data for measurement-based exposure assessments do not exist. Likewise, protocols for developing exposure factor data to be used for modeling assessments are not available. Although research on children's exposure to pesticides and other toxic chemicals is being performed within EPA, academia, industry, and other research organizations, protocols that have been developed by individual researchers for specific studies do not always collect all of the data required for reliable exposure assessments, and the data collected cannot always be interpreted.

The draft protocol provides approaches and methods that can be used for conducting field studies to collect exposure measurement data and to develop exposure factors. The protocol first provides a framework for conducting measurement studies for aggregate exposure assessments then describes the algorithms developed to assess exposure by each route. The algorithms are used to determine *a priori* what data must be collected in field studies to quantify exposure; the protocol provides explicit data requirements for each route of exposure. The approaches for estimating exposure by each route are described and include discussions of the data requirements, general considerations related to data collection, measurement methods, collection of activity pattern information, and exposure factors. The use of activity diaries and questionnaires is discussed for each route of exposure. The use of biomonitoring data is also discussed.

This report covers the period from January 1999 to September 2001.

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## 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.

In evaluating environmental health risks to children, it is important to understand that children are not little adults. Children's exposures to environmental contaminants and consumer products are expected to be different and, in many cases, much higher than older individuals. These differences in exposure are due to differences in physiological function and surface to volume ratio. Children's behavior and the way that they interact with their environment may have a profound effect on the magnitude of their chemical exposures. Children crawl, roll, and climb over contaminated surfaces, resulting in higher dermal contact than would be experienced by adults in the same environment. Children's mouthing activities (hand-to-mouth and object-to-mouth) will result in indirect ingestion of chemicals if the hands or objects are contaminated. Increased indirect ingestion of contaminants also occurs when children handle and eat foods that have come in contact with the floor or other contaminated surfaces.

In order to articulate the problems and research needs associated with children's exposure to environmental pollutants, the EPA Office of Research and Development (ORD) developed the *Strategy for Research on Environmental Risks to Children* (U.S. EPA, 2000a). This strategy is centered on the child with the overall goal of improving risk assessments for children and reducing those risks. Within the Children's Risk Strategy three specific objectives have been formulated to (1) make use of existing information to develop improved risk assessment methods and models for children; (2) design and conduct research on exposure, effects, and dose-response that will answer questions about age-related differences in exposure and risks and that will lead to better risk assessments for children; and (3) explore opportunities for prevention and reduction of risks to children.

ORD also conducts research related to children's exposure in support of the Food Quality Protection Act (FQPA) of 1996. FQPA 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. Very importantly, FQPA requires that risk assessments must be based on exposure data that are of high quality and high quantity or exposure models using factors that are based on existing, reliable data.

Currently, the data on children's exposures and exposure factors are limited and generally not adequate to assess residential exposures to consumer products and environmental contaminants. Several general areas of research are needed to improve the quality and quantity of data available for exposure assessments for children. Appropriate age/developmental benchmarks for categorizing children in exposure assessments must be identified. The activity pattern data for children (especially young children) required to assess exposure by all routes need to be developed. Methods for measuring children's exposures need to be developed and improved. Finally, field studies are needed to develop distributions of exposure and associated exposure factors.

The Children's Exposure Research Program at the EPA National Exposure Research Laboratory (NERL) is designed to meet several of the above research needs. Research in support of FQPA has been conducted to: (1) identify those pathways and activities that represent the highest potential exposures; (2) determine the factors that influence exposures; (3) develop approaches and methods for measuring and assessing aggregate exposures that account for children's activities; (4) develop distributional data on aggregate exposures; and (5) generate data on multimedia pesticide concentrations, pesticide biomarkers, and exposure factors that can be used as inputs to aggregate exposure models for exposure assessment.

## **1.2 Purpose**

The overall goal of this document is to provide guidance for generating data that can be used to improve exposure assessments for young children, as required by FQPA. Typically, exposure assessments are conducted using either a measurement-based approach or a modeling-based approach. Data requirements for both types of assessments are addressed in this document.

Exposure assessments for FQPA must consider pesticide exposures of infants and children from all sources and all potential exposure media, including those from food, water, dust, soil, and air. The definition of a complete and reliable data set for pesticide exposures of children was provided in *Exposure Data Requirements for Assessing Risks from Pesticides Exposure of Children* (U.S. EPA, 1999a). As specified in that document, an exposure assessment should include the following four elements:

1. An initial screening-level exposure assessment to identify all important sources and pathways of exposure for the pesticide.
2. An initial assessment to identify the age groups that are at the greatest risk from aggregate pesticide exposures.
3. Protocols for measuring exposure for all relevant pathways and age groups. Protocols should include:
  - the algorithms for combining the environmental monitoring data with exposure factor data to estimate an exposure,
  - a description of the environmental media that should be measured,

- standard methods for measuring pesticides in those environmental media,
  - a description of the activity patterns and exposure factors required, and
  - methods for collecting data for all of the relevant activity pattern and exposure factors.
4. An aggregate exposure assessment using probabilistic multimedia, multipathway models to develop population exposure distributions.

Currently, standard protocols for conducting exposure field studies that provide data for measurement-based exposure assessments (element 3) do not exist. Likewise, protocols for developing exposure factor data to be used for modeling assessments are not available. Although research on children's exposure to pesticides and other toxic chemicals is being performed within EPA, academia, industry, and other research organizations, protocols that have been developed by individual researchers for specific studies do not always collect all of the data required for reliable exposure assessments, and the data collected cannot always be interpreted.

The purpose of this document is to address element 3, as described above. This document is a draft protocol that provides approaches and methods that can be used for: (1) conducting field studies to collect exposure data, (2) developing exposure factor data, and (3) interpreting data to estimate exposure.

The methods, measurements, and modeling research conducted by NERL in support of FQPA serves as the basis for this document. The focus of this document is to provide a draft protocol for measuring aggregate exposures for children from residential uses of pesticides and/or for collecting data on exposure factors. However, the document is also intended to provide basic insights into data requirements and approaches for assessing children's aggregate and cumulative exposure and may be generalized to many environmental pollutants.

### **1.3 Scope**

This document presents a draft protocol for measuring children's exposure to pesticides by all relevant pathways. It addresses approaches and methods for measurements of children's exposure that can be used as part of field monitoring studies. The protocol describes the algorithms for each route of exposure, specifies the data required to conduct the aggregate exposure assessment, and describes methods for collecting the data. The approach is provided for estimating exposure by each route. References are provided to assist the reader in obtaining detailed information on the utility of measurement methods, procurement of materials and supplies, and implementation in the field.

There are a number of elements of an exposure measurement study that are not addressed in this protocol because they are specific to the study objectives and study design and are beyond the scope of this document. For example, this document does not discuss sample selection and participant recruitment. The survey design is a critical, and very complex, element of any

exposure study, but discussion of this study element is beyond the scope of this document. The protocol also does not address screening methods that may be used to identify potentially highly exposed sub-populations or environments. Because the methods in the protocol should be applicable to a wide range of pesticides and to selected environmental contaminants with a variety of analytical requirements, analytical methods are not discussed in the protocol. The user of the protocol will need to identify and use the appropriate analytical methods to measure the compounds of interest after collection.

This is a draft protocol that does not specify the detailed methods to be used for data collection. A number of research studies are on-going or planned that will be used to further evaluate the protocol, data collection methods, and questionnaires to be used in future children's exposure measurement studies. Results of these studies will be used to refine the protocol and to develop detailed specifications for approaches and methods.

#### **1.4 Format of the Document**

The document is organized to provide general information on exposure and a modeling framework for addressing children's exposure. Information is given on the algorithms and methods for collecting data on exposure and exposure factors. Specific sections are as follows:

- Section 2 discusses the basic concepts of exposure including definitions. It also provides a framework for conducting measurement studies for aggregate exposure assessments.
- Section 3 gives proposed exposure algorithms along with explicit data requirements for each route of exposure.
- Section 4 describes the exposure scenario addressed by this draft protocol.
- Sections 5 through 8 describes approaches for measuring exposure by various routes and pathways.
- Section 9 discusses other data collection methods including questionnaires.
- Section 10 includes references cited in the document.

## 2.0 BASIC CONCEPTS OF AGGREGATE EXPOSURE

The purpose of this chapter is to first define the concepts of exposure. The reader is then introduced to the basic framework that NERL has been using to develop a protocol that defines both approaches and methods for measuring exposure and exposure factors in field studies.

### 2.1 Definitions

**Exposure** is defined as the contact (at visible external boundaries) of an individual with a pollutant for specific durations of time. For exposure to occur, environmental media must be contaminated with a pollutant, an individual must be in the same microenvironment with the contaminated media, s/he must come in contact with a contaminated medium, and the contact activity must cause a transfer of the contaminant from the media to the portal of entry of the individual.

Children's exposure to environmental contaminants is a complex process that may occur from several sources through a number of different pathways and routes. **Sources** include all uses of a chemical that could result in children's exposure. Within this document, only nonoccupational exposures to environmental contaminants are considered. **Route** of exposure (i.e., dermal, oral, inhalation) is defined as the portal of entry. There are three routes of exposure: the skin is the portal of entry for the dermal exposure route; the mouth is the portal of entry for the ingestion exposure route, and the lung is the portal of entry for the inhalation exposure route. **Pathway** is defined as the course that the contaminant takes from its source to the portal of entry. In some cases, we have simplified the pathways to only include the contaminated exposure media and route of exposure. Exposure pathways include those that occur indoors and outdoors at the home and at other institutional and non-residential settings (e.g., schools and daycare centers). **Aggregate exposure** is the combined exposures to a single chemical from all sources across all routes and pathways.

**Exposure Factors** are the factors related to human behavior and characteristics that determine an individual's exposure to a pesticide or contaminant. For example, an individual's exposure to a pesticide by the inhalation route is determined by factors that include the duration of time spent in different microenvironments during the day and the individual's inhalation rates during the period of exposure.

Other definitions used in this document that are pertinent to conducting aggregate exposure assessments are given in Table 2-1.

### 2.2 Measurement Methods Versus Approaches

Traditionally, exposure measurement studies have been based on using a set of methods to measure contaminants in environmental media. Questionnaires and diaries are then used to

Table 2-1. Definitions Related to Aggregate Exposure Assessments

Term	Definition
Acute exposure	An exposure period of less than one day.
Aggregate exposure	The combined exposures to a single chemical from all sources across all routes and pathways.
Approach	The process for combining data from single determinants to estimate exposure.
Biomarker of exposure	Exogenous chemicals, metabolites, or the products of interactions between a chemical and target molecules or cells that are measured within a compartment or within an organism. This includes internal dosimeters of a chemical or metabolite concentrations and markers of biologically effective doses.
Chronic exposure	An exposure presumed to occur over a substantial portion of the individual's lifetime.
Cumulative exposure	The total exposure to chemicals that cause a common toxic effect(s) to human health by the same, or similar, sequence of major biochemical events.
Exposure	The contact (at visible external boundaries) of an individual with a pollutant for specific durations of time.
Exposure algorithm	A mathematical expression of the approach. It expresses exposure as a function of pesticide concentration in the exposure medium, contact rate, rate of transfer from the exposure medium to the portal of entry, and exposure duration.
Exposure factors	The factors related to human behavior and characteristics that determine an individual's exposure to a pesticide or contaminant. For example, duration of exposure, inhalation rates, transfer coefficients.
Exposure pathway	The course that the chemical takes from its source to the receptor's portal of entry.
Exposure route	The portal of entry of a chemical into the body.
Exposure scenario	The combination of facts, assumptions, and inferences that define a discrete situation or activity where potential exposures may occur. These include the source, the exposed population, the time frame of exposure, microenvironment(s), and activities.



Term	Definition
Intermediate-term exposure	An exposure lasting from one week to several months.
Macroactivity	Aggregated series of contact events in the same microenvironment and the same activity level.
Method	A process for measuring a single determinant such as an environmental concentration of a pesticide or an activity frequency.
Microactivity	Individual skin-to-surface or object-to-mouth contact event.
Microenvironment	A space or location defined for dermal exposure on the basis of specific surface types that may be contacted (e.g., indoors at home on carpet). For inhalation exposure, it is defined as an air space with a homogenous concentration of the chemical.
Pathway	The course that the contaminant takes from its source to the portal of entry.
Short-term exposure	An exposure lasting from one to seven days.
Transfer coefficient	A measure of contaminant transfer resulting from contact of an object or skin with a contaminated microenvironmental surface while engaged in a specific macroactivity, expressed as surface contact area per unit time ( $\text{cm}^2/\text{h}$ ).
Transfer efficiency	The fraction of mass transferred from a contaminated surface to skin, food, or other object per unit contact (unitless).
Transferable surface residue	The mass of contaminant per unit area ( $\mu\text{g}/\text{cm}^2$ ) measured by a standard transfer method.
Total surface loading	The total mass of contaminant per unit area ( $\mu\text{g}/\text{cm}^2$ ).

collect information on activities and locations. Often a systematic selection of methods and questions is not developed and the resulting data cannot be used to estimate exposure by multiple routes and pathways. Within this document, the emphasis is on the use of approaches to estimate exposure rather than the application of a set of methods. Such a process first determines how exposure for each route will be estimated, then defines the data needed, and finally identifies specific methods for data collection.

Within this protocol, a **method** is defined as a process for measuring a single determinant such as an environmental concentration of a pesticide or an activity frequency. An **approach** defines the process for combining data from single determinates to estimate exposure. The **exposure algorithm** defines the approach. For each route, the algorithm mathematically expresses exposure as a function of pesticide concentration in the exposure medium, contact rate, rate of transfer from the exposure medium to the portal of entry, and exposure duration. Consequently, the exposure algorithm describes the specific data needs for estimating exposure and the process for combining the data. This protocol describes both methods for the single determinants and the algorithms for estimating exposure by each pathway and route.

## 2.3 Exposure Assessment

Typically, exposure assessments are conducted using either an individual measurement-based approach or a population modeling-based approach. For simplicity, these will be referred to as measurement and modeling assessments throughout this document. Data requirements and measurement study designs will vary for the two approaches. This document emphasizes data collection methods and approaches for measurement-based assessments.

**Measurement assessments** measure the contact of the individual with the chemical in the exposure media over an identified period of time. Direct assessments are made through field monitoring studies of children in their everyday environments. In such studies, data are collected on pollutant concentrations in a variety of exposure media (i.e., air, drinking water, food, house dust, surface residues), activities, and exposure factors so that exposure can be measured or estimated for each child in the study. Often pesticides or their metabolites are analyzed in biological media as a direct measure of exposure aggregated over all sources and pathways for a given time period. A comparison of exposure estimated from measurement assessments to exposure estimated with biomarkers often provides a evaluation of both approaches. For measurement assessments, it is imperative to collect all of the data on exposure media concentrations, activities, and exposure factors that are required to quantify exposure for an individual using the exposure algorithms for each route and pathway.

**Modeling assessments** use available information on concentrations of chemicals in exposure media along with information about when, where, and how individuals might contact the exposure media. The modeling approach then uses models and a series of exposure factors (i.e., contact duration, contact frequency, contaminant transfer) to estimate exposure. For modeling assessments, distributional data on exposure factors and environmental concentrations are used to estimate exposure distributions for a population. However, the data do not need to be collected on the same individuals. For the modeling approach, studies can be conducted to obtain data for only a single exposure factor or a combination of exposure factors. No attempt is made to actually measure or estimate exposure for the individual participants in the study with the modeling approach. Data on activities and exposure factors collected as part of a measurement assessment can also be applied to modeling assessments.

## 2.4 Framework for Exposure Assessment

Aggregate exposure includes exposure from all sources, routes and pathways for individual pesticides. Given this definition, a comprehensive approach is required to understand and adequately address all of the components of an aggregate exposure assessment. NERL has developed a framework to systematically identify the important sources, routes, and pathways for exposure (Cohen Hubal et al., 2000). This framework is based upon the development of a conceptual model for aggregate exposure and provides the basis for developing a protocol to measure and assess aggregate exposures, as well as for developing sophisticated stochastic models. This framework also allows us to systematically identify the most critical research needs and data gaps associated with children's exposures to pesticides. The steps of the framework are as follows:

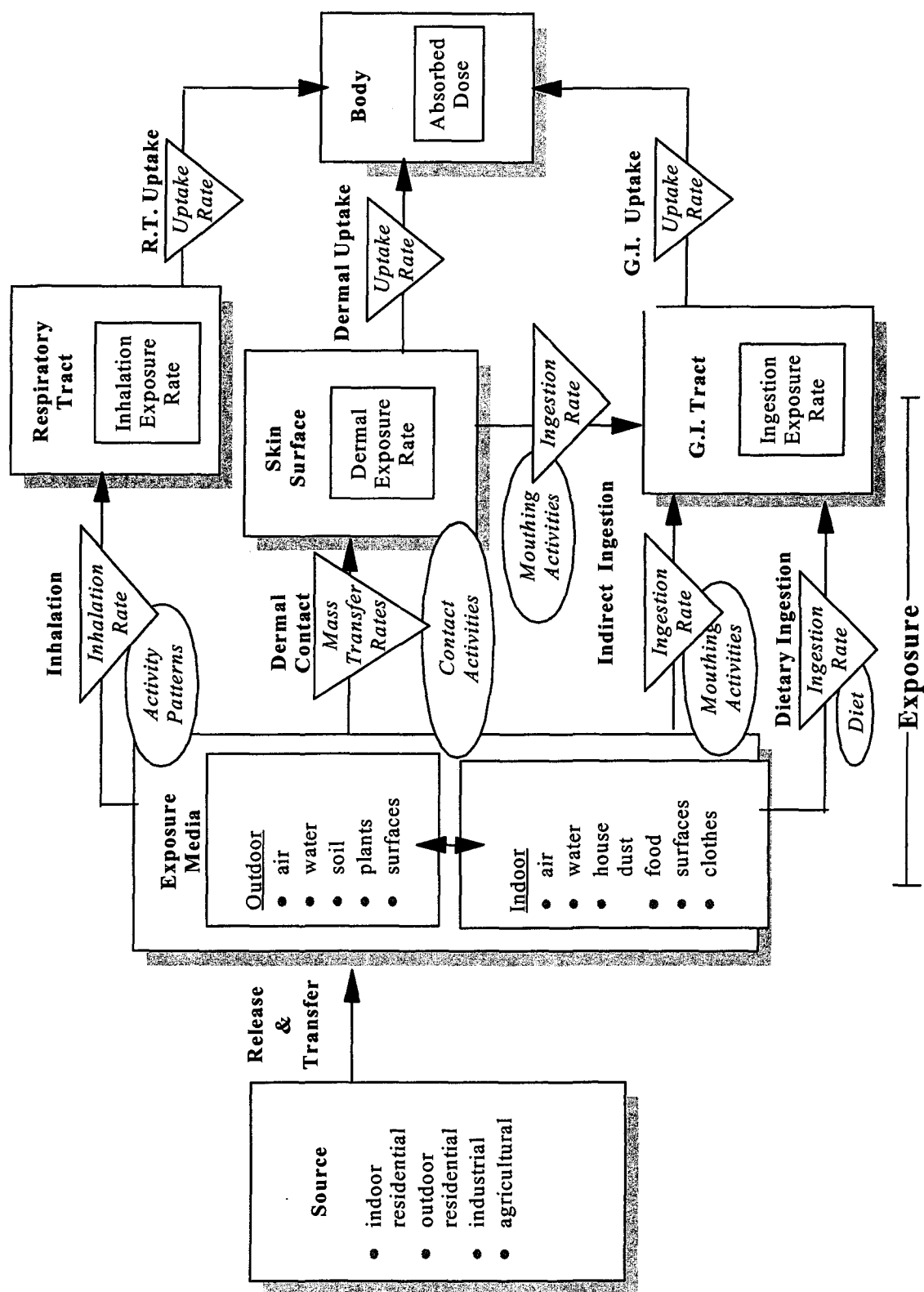
1. Develop a model that describes aggregate exposure,
2. Identify potential exposure pathways and scenarios,
3. Define algorithms, exposure factors, and data requirements for each route,
4. Develop a probabilistic model for assessing aggregate exposure,
5. Perform a screening assessment to evaluate the range of exposures for, and significance of, each pathway,
6. Identify critical data gaps in the assessment process, and
7. Conduct field studies to address data gaps and reduce uncertainty.

Steps 1 through 3 have been critical in the development of this protocol. Steps 4, 5, and 6 have been used to identify research needs. The protocol developed here will be applied to studies in step 7.

Although the emphasis of this protocol is on measuring and assessing residential pesticide exposure to infants and young children, this same framework could be adapted for other exposure scenarios.

**Model.** A conceptual model of children's residential exposure to pesticides was developed by NERL that was the initial focal point for the research strategy and protocol development. This conceptual model (Figure 2-1) shows the exposure process from source to absorbed dose for all routes of exposure. Pesticides may be released into the outdoor or indoor environment by residential, commercial, or agricultural use. Once released into the environment, pesticides can transfer from one medium to another (e.g., air to soil) and from one micro-environment to another (e.g., yard to house). Exposure occurs once a human contacts a contaminated exposure medium and the contaminant is transferred from the medium to the portal of entry. Exposure is a function of the time spent in the microenvironment of interest, contact rate, and the mass transfer of pesticide from the exposure medium to the portal of entry. Contacts rates and mass transfer are a function of human activity patterns (indicated by the shaded ovals). Finally, uptake of the pesticide through the respiratory tract, the skin, or the

Figure 2-1. Conceptual model of children's residential exposure to pesticides.



gastrointestinal tract will result in an absorbed dose.

**Exposure Pathways.** The conceptual model was used to systematically identify all potential exposure pathways. In general terms, a pathway is defined as the course that a pesticide takes from its source to the receptor's portal of entry. However to specifically evaluate potential for exposure, simplified pathways were defined by the exposure medium and the route of exposure. Essentially, the evaluation focused on exposure, without considering transport of the pesticide to the exposure medium. Using this simplified definition, the pathway crosses the activity with the exposure medium that leads to exposure. For example, inhalation (activity) of indoor air (exposure medium) is one pathway, and dermal contact (activity) with turf (exposure medium) is another pathway. A comprehensive list of potential pathways was developed and is presented in Table 2-2.

**Exposure Algorithms.** Algorithms were developed to assess exposure by each route. The algorithm mathematically expresses exposure as a function of pesticide concentration in the exposure medium and various exposure factors, including contact rate, rate of transfer from the exposure medium to the portal of entry, and exposure duration. As described in Section 3, exposure algorithms are also used to describe the data requirements for each route in field monitoring studies to assess exposure using the measurement-based approach.

**Time Frame for Exposure Measurements.** Risk assessments must take into account the frequency and duration of exposure, as well as its magnitude. In pesticide risk assessments, four exposure durations generally are considered. **Acute** exposure is defined as an exposure period of less than one day. Exposures through food and drinking water have been included in acute exposure assessments. **Short-term** exposure is defined as an exposure lasting from one to seven days. Possible short-term exposures to pesticides in and around the home could come from uses such as on lawns and home gardens, as a crack and crevice treatment for insects, a treatment for carpets or other surfaces, or a flea treatment for pets. Other short-term exposures could occur in public places such as parks, school playgrounds, and playing fields. Data indicate that post-application exposures from these uses typically last from a day to several weeks. **Intermediate-term** exposure is defined from one week to several months. Possible intermediate-term exposures to pesticides in and around the home could occur due to use of rodenticides as well as some of the exposure scenarios described above in the acute and short-term categories. **Chronic** exposure is presumed to occur over a substantial portion of the individual's lifetime. Although chronic exposure can occur via all routes and pathways, dietary is considered to be the largest component. Pesticides, such as those used as termite control, could also result in chronic exposures.

**Exposure Scenarios.** For any given pathway, a set of associated exposure scenarios can be described. An exposure scenario is defined by the combination of:

- Source or application method (e.g., crack and crevice application of pesticides, residential

Table 2-2: Pathways For Children's Non-occupational Exposure to Pesticides

Exposure Medium	Route
OUTDOOR PATHWAYS	
Pesticide pellets and granules	Ingestion (direct)
	Dermal contact
	Ingestion (hand-to-mouth)
Outdoor air	Inhalation
	Dermal contact
	Ingestion of particles
Outdoor water a) natural water body b) swimming pool	Dietary ingestion
	Dermal contact (e.g. ,while swimming)
	Ingestion (direct e.g., while swimming)
	Inhalation of vapors (e.g., while swimming)
Soil	Ingestion (direct)
	Indirect ingestion (object-to-mouth, hand-to-mouth)
	Dermal contact
Plants a) turf b) gardens c) fruit on trees	Ingestion (direct)
	Indirect ingestion (object-to-mouth)
	Dermal contact
	Indirect ingestion (hand-to-mouth)
Outdoor surfaces/objects a) paint chips b) concrete c) toys, furniture, tools, etc.	Indirect ingestion (object-to-mouth)
	Dermal contact
	Indirect ingestion (hand-to-mouth)
INDOOR PATHWAYS	
Indoor air	Inhalation
	Dermal contact
	Ingestion of particles
Indoor water	Dietary ingestion

Exposure Medium	Route
	Dermal contact (e.g., showering)
	Inhalation of vapors (e.g., showers, dishwashers, etc.)
Food	Dietary ingestion (food contaminated with agricultural residues)
	Indirect ingestion (food contaminated by contact with contaminated residential surfaces)
	Indirect ingestion (food contaminated by contact with contaminated hands)
Indoor objects/surfaces: a) carpeted surfaces b) hard surfaces c) upholstery and bedding d) toys	Indirect ingestion (object-to-mouth)
	Dermal contact
	Indirect ingestion (hand-to-mouth)
House dust (Includes tracked in soil)	Ingestion (direct)
	Indirect ingestion (object-to-mouth, hand-to-mouth)
	Dermal contact
OTHER PATHWAYS	
Pets	Dermal contact
	Indirect ingestion (hand-to-mouth)
Material impregnated with pesticides	Dermal contact
	Indirect ingestion (hand-to-mouth)
	Indirect ingestion (object-to-mouth)
	Inhalation of vapors
Clothes	Dermal contact
	Indirect ingestion (hand-to-mouth)
	Indirect ingestion (object-to-mouth)

- use of consumer product, lawn and garden applications, agricultural use),
- Exposed population (e.g., age group, geographical location),
- Time frame of exposure (acute, short term, chronic), Microenvironments for exposure, and
- Activity that results in exposure.

When exposure assessments are conducted by the modeling approach, specific exposure scenarios determine the values of the exposure factors that should be used in the algorithms to estimate exposures. For measurement assessments, field studies are conducted to assess exposure for individual participants. For these studies, the participants actually define the scenario based on their everyday activities. Field studies can be conducted on a general population to understand distributions of exposures and exposure factors and the relationship between various exposure factors. These studies can also provide information to determine what scenarios actually exist in the population and to aid in selecting the most appropriate scenarios for modeling assessments. Alternately, studies can be conducted to evaluate exposure and exposure factors for predefined scenarios. In either case, it is necessary to collect all of the data that are needed to adequately define the scenario and the exposure factors that are used in the algorithm for that scenario.

NERL has used the conceptual model discussed here to develop the Stochastic Human Exposure and Dose Simulation Model for Pesticides (SHEDS-Pesticides). SHEDS is a probabilistic multi-media, multi-pathway model (Zartarian et. al, 2000) that is designed to develop probability distributions of exposure and to also estimate inter-individual variability in the population and uncertainty in the estimated empirical exposure and dose distributions. The model is described in Appendix A of this document. Measurement data collected with the protocol described in this document will be used as inputs and for evaluation of the SHEDS-Pesticides model. Results of the sensitivity and uncertainty analyses conducted with SHEDS-Pesticides will be used to further refine the protocol for the exposure measurements. Hence, SHEDS-Pesticides will be used in an iterative fashion with the conceptual framework presented here to refine the protocol.



### 3.0 EXPOSURE ALGORITHMS AND DATA REQUIREMENTS

The purpose of this chapter is to present the exposure algorithms that have been developed for assessing exposure by each route and pathway. The data requirements associated with the algorithms are also given. Details of the methods to collect these data are presented in subsequent chapters.

#### 3.1 Exposure Algorithms

Exposure algorithms have been developed to assess exposure by each route. The algorithms are used here to determine *a priori* what data must be collected in field studies to quantify exposure. Thus, the algorithms provide a convenient framework for developing and using field monitoring methods.

Although it is convenient to identify pathways by first considering the exposure medium and then considering the route, the associated exposure algorithms are route specific. Aggregate assessments for children must include all three exposure routes: inhalation, dermal contact, and ingestion. In addition, ingestion can be divided into two important subroutes, dietary and indirect ingestion [i.e., ingesting pesticides from contaminated objects (including food) and hands placed in the mouth].

The exposure algorithm defines the measurement approach. For each route, the algorithm mathematically expresses exposure as a function of pesticide concentration in the exposure medium, contact rate, rate of transfer from the exposure medium to the portal of entry, and exposure duration. The basic components of the algorithm are used to define the monitoring, activity pattern, and source usage data that must be collected to estimate exposure.

Algorithms are applied separately to all of the microenvironments and activities that an individual experiences in a given time period. Within this document, microenvironment is referred to as the location where an individual spends time. For inhalation exposure, Duan (1982) defined a microenvironment as “a [portion] of air space with homogeneous pollutant concentration.” It has also been defined as a volume in space, for a specific time interval, during which the variance of concentration within the volume is significantly less than the variance between that microenvironment and surrounding microenvironments (Mage, 1985). For dermal exposure, microenvironment has been defined based upon the location and surface type. Homogeneity of the surface concentration has been considered within the algorithms.

Activities are defined as either macroactivities or microactivities. Macroactivity is a series of contact events in the same microenvironment and the same activity level that are aggregated for the purposes of estimating dermal exposure. The macroactivity approach has been used extensively for estimating worker exposures to pesticides. For children, an example of a macroactivity would be lying on a carpeted floor for one hour watching television in the family

room. Microactivities are defined as discrete, individual, skin-to-surface or object-to-mouth events, such as when a child puts a toy in his/her mouth.

Exposure models for assessments use one of two general approaches: a time-series approach that estimates microenvironmental exposures sequentially as individuals go through time, or a time-averaged approach that estimates microenvironmental exposures using average microenvironmental concentrations and the total time spent in each microenvironment. The time-series approach to modeling personal exposures provides the appropriate structure for accurately estimating personal exposures (Esmen and Hall, 2000; Mihlan et al., 2000). In addition, the time-varying dose profile of an exposed individual can be modeled only by using the time-series approach (McCurdy, 1997, 2000). However, a time-averaged approach is typically used since the input data needed to support a time-series model are usually not available or cannot be easily collected. Real-time monitoring techniques for measuring pesticide concentrations are very limited. Most environmental monitoring provides either an integrated 24-hour concentration (as in air or duplicate diet samples) or a single time-point concentration (as in transferrable residue samples). Thus, the algorithms presented here use a time-averaged approach over a 24-hour period. They could, however, be modified to provide time-series data, especially for activity patterns.

Approaches for aggregating exposure estimates across routes are not presented here. Since absorbed dose may be different depending upon the route, it is not appropriate to sum exposure across routes. Exposure for each route is estimated independently. These exposures can then be used as inputs to exposure/dose models to estimate dose. The algorithms presented here are similar to those used elsewhere in the literature (U.S. EPA, 1997a; U.S. EPA 1997b).

### 3.2 Inhalation Route

Inhalation exposure may result from pesticides applied indoors or due to infiltration of pesticides applied adjacent to buildings. Although current use pesticides, such as the pyrethroids, are generally less volatile than many of the pesticides previously used indoors (e.g., chlorpyrifos), they may be detected in the air following application.

**Exposure Algorithm.** Inhalation exposure is estimated for each of the micro-environments where a child spends time and each macroactivity that would result in a different inhalation rate while engaging in that activity. Exposure over the 24-hour period is then the sum of all of the microenvironmental/macroactivity (me/ma) exposures. This may be expressed mathematically as:

$$E_{i24} = \sum E_{ime/ma} \quad (1)$$

where

$E_{i24}$  = the total inhalation exposure over a 24-hour period ( $\mu\text{g/d}$ )

$E_{ime/ma}$  = the inhalation exposure for a given me/ma over a 24-hour period ( $\mu\text{g/d}$ )  
 For each me/ma, inhalation exposure over the 24-hour period ( $E_{ime/ma}$ ) is defined as:

$$E_{ime/ma} = C_{ame} \times T_{me/ma} \times IR_{ma} \quad (2)$$

where

$C_{ame}$  = the air concentration measured in the micro environment ( $\mu\text{g/m}^3$ )  
 $T_{me/ma}$  = the time spent in that me/ma over the 24 hour period (h/d)  
 $IR_{ma}$  = the child's inhalation rate representing his activity level for that macroactivity ( $\text{m}^3/\text{h}$ )

**Data Requirements.** In order to apply the above model, the following data are required:

- **Definition of the important microenvironments/macroactivities for inhalation exposure.** Four generalized microenvironments have been defined for very young children (4 years old and younger). These include indoors and outdoors at home and indoors and outdoors at daycare centers. If the air concentrations indoors are not homogenous, there may be more than one microenvironment indoors at home or indoors at daycare. There may also be *other indoor* and *other outdoor* microenvironments that are important if the child spends substantial amounts of time away from the home or daycare. Four macroactivities have been defined for children: sleeping/napping, active play, quiet play, and eating.
- **Air concentration in each microenvironment.** Ideally, an integrated air concentration should be measured only during the time that the subject is in each microenvironment. Alternatively, an integrated 24-hour measurement should be adequate if it is assumed that air concentrations do not vary substantially over time or space within any microenvironment. Since the air concentration for the *other indoor* and *other outdoor* categories will not be measured, an approach for developing a reasonable estimate must be made. This estimate becomes important for inhalation exposure if the subject spends substantial time in these other microenvironments.
- **Amount of time the child spends in each me/ma over 24-hours.** The amount of time a child spends in each microenvironment/macroactivity is collected with a time-activity diary for the period of monitoring. The diary, at a minimum, should record the child's time in each microenvironment and information on the child's activities that can be used to estimate inhalation rate while in that microenvironment.
- **Inhalation rate for each me/ma.** The rate of inhalation will be estimated based on age and weight of the child and activity in each microenvironment.

Table 3-1 summarizes the data requirements as they relate to equation (2) to estimate exposure by the inhalation route.

Table 3-1. Summary of Data Collection Requirements by Exposure Route

Parameter	Measurement	How Collected	Units
Inhalation Exposure $E_{ime/ma} = C_{ame} \times T_{me/ma} \times IR_{ma}$			
$C_{ame}$	Air concentration in me	Measured with active sorbent collection	$\mu\text{g}/\text{m}^3$
$T_{me/ma}$	Time spent in each me/ma	Time-activity diary, questionnaire	h/d
$IR_{ma}$	Inhalation rate	Estimated from size, age, and activity data collected with diaries and questionnaires using reference values	$\text{m}^3/\text{h}$
Dermal Exposure - Macroactivity Approach $E_{dme/ma} = C_{surf} \times TC_{me/ma} \times AD_{me/ma}$			
$C_{surf}$	Surface loading (total or transferrable) in each me	Measured by wipe, press, or roller methods	$\mu\text{g}/\text{cm}^2$
$TC_{me/ma}$	Transfer coefficient <sup>a</sup>	Empirically determined for each me/ma from laboratory experiments or field studies	$\text{cm}^2/\text{h}$
$AD_{me/ma}$	Activity duration for ma in a specific me	Time-activity diary, questionnaire	h/d
Dermal Exposure - Microactivity Approach $E_{dmi} = C_{surf} \times TE \times SA \times EF$			
$C_{surf}$	Surface loading (total or transferable) in me	Measured by wipe, press, or roller method	$\mu\text{g}/\text{cm}^2$
TE	Transfer efficiency <sup>a</sup>	Empirically determined from laboratory experiments	unitless
SA	Surface area contacted	Visual observation or videotape	$\text{cm}^2/\text{event}$
EF	Frequency of contact events	Visual observation or videotape	events/d
Dietary Ingestion Exposure $E_f = \Sigma C_f W_f$			
$C_f$	Concentration of pesticide in the food item (s)	Measurement in individual food items or composite duplicate diet samples	$\mu\text{g}/\text{kg}$

Parameter	Measurement	How Collected	Units
$W_f$	Weight of food item consumed	Measured in duplicate diet sample	kg/d
Indirect Ingestion Exposure - Microactivity Approach $E_{ingmi} = C_x \times TE_x \times SA_x \times EF$			
$C_{surf}$	Surface loading (total or transferable) on object x	Measure by a wipe or press method	$\mu\text{g}/\text{cm}^2$
$TE_x$	Transfer efficiency <sup>a</sup>	Empirically determined from laboratory experiments	unitless
$SA_x$	Surface area contacted	Visual observation or videotape	$\text{cm}^2/\text{event}$
EF	Frequency of mouthing events	Visual observation or videotape	events/d

<sup>a</sup> This parameter must be calculated using the same surface loading measurement method as used to measure  $C_{surf}$

### 3.3 Dermal Route

Two main approaches are currently used to assess dermal exposure. These assessment approaches provide different ways of integrating exposure over time and space. In the macroactivity approach, exposure is estimated individually for each of the microenvironments where a child spends time and each macroactivity that the child conducts within that microenvironment. To do this, exposure is estimated using empirically derived transfer coefficients to aggregate the mass transfer associated with a series of contacts with a contaminated medium.

In the microactivity approach, exposure is explicitly modeled as a series of discrete transfers resulting from each contact with a contaminated medium. To estimate dermal exposure with the microactivity approach, exposure must be estimated for all contacts made by child during a 24-hour period. To use the microactivity approach, a substantial amount of detail is needed to characterize children's dermal contact with chemical residues in all of their microenvironment/macroactivity combinations and to quantify subsequent dermal absorption. These data include: definitions of the microenvironments/macroactivities that are important for dermal exposure; surface loading measurements (total or transferable surface residue) for each microenvironment/macroactivity combination; amount of time a child spends in each microenvironment/macroactivity during a 24-h period; the transfer coefficient for each microenvironment/macroactivity; surface area of exposed skin; contact frequency of exposed skin in a given microenvironment; and transfer efficiency for each microactivity. Collection of

this level of detailed data is extremely resource intensive and not practical in most field measurement studies. The microactivity approach can be applied in small research studies, but has limited utility for exposure measurement studies. Therefore, although the algorithm is described below, methods for collection of the required data are not described in this protocol.

### 3.3.1 Macroactivity Approach

**Exposure Algorithm.** To estimate exposure using the macroactivity approach, microenvironments are defined by location and surface type. Macroactivities (i.e., active play, quiet play, sleeping/napping, and eating) are defined based on the expected magnitude and variability of the pesticide transfer coefficient. For any given microenvironment/macroactivity combination transfer coefficients are developed using carefully controlled laboratory or field studies. Exposure in field studies can then be estimated individually for each of the microenvironments where a child spends time and each macroactivity that the child conducts within that microenvironment using transfer coefficients and the surface loading in the microenvironment. The surface loading may be either the total residue concentration present on a surface or the amount of pesticide residue on the surface that is available for transfer to the skin (referred to as the transferable residue). Different methods are used to make the measurements of these two categories of residues, as discussed in Section 6.0.

Exposure over a 24-hour period is the sum of all the microenvironment/macroactivity exposures, expressed as:

$$E_{\text{derm24}} = \sum E_{\text{dme/ma}} \quad (3)$$

where

$$\begin{aligned} E_{\text{derm24}} &= \text{dermal exposure over a 24-h period for all microenvironments and macroactivities } (\mu\text{g/d}) \\ E_{\text{dme/ma}} &= \text{dermal exposure for a given microenvironment/macroactivity combination } (\mu\text{g/d}) \end{aligned}$$

For each microenvironment/macroactivity combination, dermal exposure is defined as :

$$E_{\text{dme/ma}} = (C_{\text{surf}})(TC_{\text{me/ma}})(AD_{\text{me/ma}}) \quad (4)$$

where

$$\begin{aligned} E_{\text{dme/ma}} &= \text{dermal exposure for a given microenvironment/macroactivity combination over a 24-h period } (\mu\text{g/d}) \\ C_{\text{surf}} &= \text{surface loading (total or transferable) measured in the microenvironment } (\mu\text{g/cm}^2) \\ TC_{\text{me/ma}} &= \text{transfer coefficient for the microenvironment/macroactivity } (\text{cm}^2/\text{h}) \\ AD_{\text{me/ma}} &= \text{activity duration that represents the time spent in each} \end{aligned}$$

microenvironment/macroactivity combination with a specific clothing pattern for the child that would affect the surface area available for transfer over a 24-h period (h/d)

The transfer coefficient,  $TC_{me/ma}$ , provides a measure of dermal exposure resulting from contact with a contaminated microenvironmental surface while engaged in a specific macroactivity. The transfer coefficient takes into account the fraction of the transferable surface residue that is transferred from a surface to skin, the character of the microenvironmental surface that is contacted, and the area of the microenvironmental surface that is contacted during a time increment for a given activity. Transfer coefficients are empirically derived in laboratory tests or controlled field experiments.  $TC_{der}$  can be defined as follows:

$$TC_{me/ma} = (E_{dme/ma}) / (C_{surf})(AD_{me/ma}) \quad (5)$$

**Data Requirements.** Table 3-1 shows the data requirements as they relate to equation (4) which is used to estimate dermal exposure by the macroactivity approach. The following data will be required for each microenvironment/macroactivity combination to estimate dermal exposure:

- **Definition of the microenvironments/macroactivities that are considered important for dermal exposure.** These microenvironments/macroactivities account for:
  - Various microenvironments with different residue concentrations,
  - Various types of surfaces that affect the transfer rate,
  - Child activities that affect the transfer coefficient. Macroactivities have been selected that should have fairly uniform transfer coefficients within a microenvironment.
- **Surface loading.** For each microenvironment/surface combination, measurements will be made on those surfaces for which the child is expected to have substantial contact. Measurements should provide a representative loading for the entire area of contact. The measurement may be of total residue or the transferable residue.
- **Amount of time the child spends in each microenvironment/macroactivity during a 24-h period.** These data can be collected using a time-activity diary or questionnaire.
- **Transfer coefficient for each microenvironment/macroactivity.** These are data that are currently not available. NERL is in the process of developing specific children's age related transfer coefficients in the laboratory and in controlled field experiments.
- **Clothing pattern for the child that would affect the surface area available for transfer.** The amount of clothing and exposed skin needs to be determined.

### 3.3.2 Microactivity Approach

**Exposure Algorithm.** To assess dermal exposure using the microactivity approach, exposure is estimated individually for all of the microactivities in a given microenvironment in

which dermal contact occurs. Exposure over a 24-h period is then the sum of all of the individual exposures:

$$E_{\text{derm24}} = \sum_i \sum_j E_{\text{dmi}} \quad (6)$$

where

$E_{\text{derm24}}$	=	dermal exposure over a 24-h period for all microactivities ( $\mu\text{g/d}$ )
$i$	=	sum of all microenvironments
$j$	=	sum of all microactivities in a given microenvironment
$E_{\text{dmi}}$	=	dermal exposure for each microactivity over a 24-h period ( $\mu\text{g/d}$ )

For each microenvironment/microactivity, dermal exposure over a 24-h period can be defined as:

$$E_{\text{dmi}} = (C_{\text{surf}})(\text{TE})(\text{SA})(\text{EF}) \quad (7)$$

where

$E_{\text{dmi}}$	=	dermal exposure for each microactivity over a 24-h period ( $\mu\text{g/d}$ )
$C_{\text{surf}}$	=	surface loading (total or transferable) measured in the microenvironment ( $\mu\text{g/cm}^2$ )
TE	=	transfer efficiency, fraction transferred from surface to skin (unitless)
SA	=	surface area contacted ( $\text{cm}^2/\text{event}$ )
EF	=	frequency of contact events during a 24-h period (events/d)

Transfer efficiency is defined as the fraction of mass transferred from a contaminated surface to skin per unit contact and can be represented as follows:

$$\text{TE} = (L_{\text{mi}}) / (C_{\text{surf}}) \quad (8)$$

where

$L_{\text{mi}}$	=	loading ( $\mu\text{g/cm}^2$ ) on the transfer medium (i.e., skin)
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**Data Requirements.** To use the microactivity approach, a greater level of detail is needed to characterize children's dermal contact with chemical residues in their environments. Given the greater level of detail that is required, the microactivity approach is not used for directly estimating exposure in field studies. Rather, it is applied to indirect modeling assessments. Data are usually only collected on exposure factors, most particularly in the form of videotaping children's activities. Data required to estimate dermal exposure include:

- **Definition of the microenvironments that are considered important for dermal exposure.** The microenvironments should account for:
  - Various microenvironments with different residue concentrations,
  - Various surfaces that affect the transfer rate, and



- Clothing worn by the child.
- **Contact frequency of exposed skin in a given microenvironment.** This is determined using a videography method or direct visual observations.
- **Surface area of skin exposed during contact.** Data are collected on a child specific basis.
- **Parameters describing the nature of the contact for each microactivity.** Information should be collected on a child specific basis on those parameters that influence transfer efficiency from the surface to the skin. Currently, this information is not available; however, research at NERL is underway to identify these parameters. Potentially important contact parameters include duration, pressure, motion, and skin surface (sticky, wet, dry).
- **Surface loading at the point of contact.** When surface loadings are not homogenous, data should be collected for every point on the surface where contact is made. Unfortunately, measurement data in this detail cannot be collected in a field study. Thus pesticide distributions within each microenvironment must be modeled using data generated during laboratory experiments or carefully controlled field experiments. Sufficient field measurement data should be available to verify the modeled distributions.
- **Transfer efficiency for each microenvironment and microactivity.** These are data that are currently not available and need to be generated experimentally in controlled laboratory experiments.

Table 3-1 shows the data requirements as they relate to equation (7).

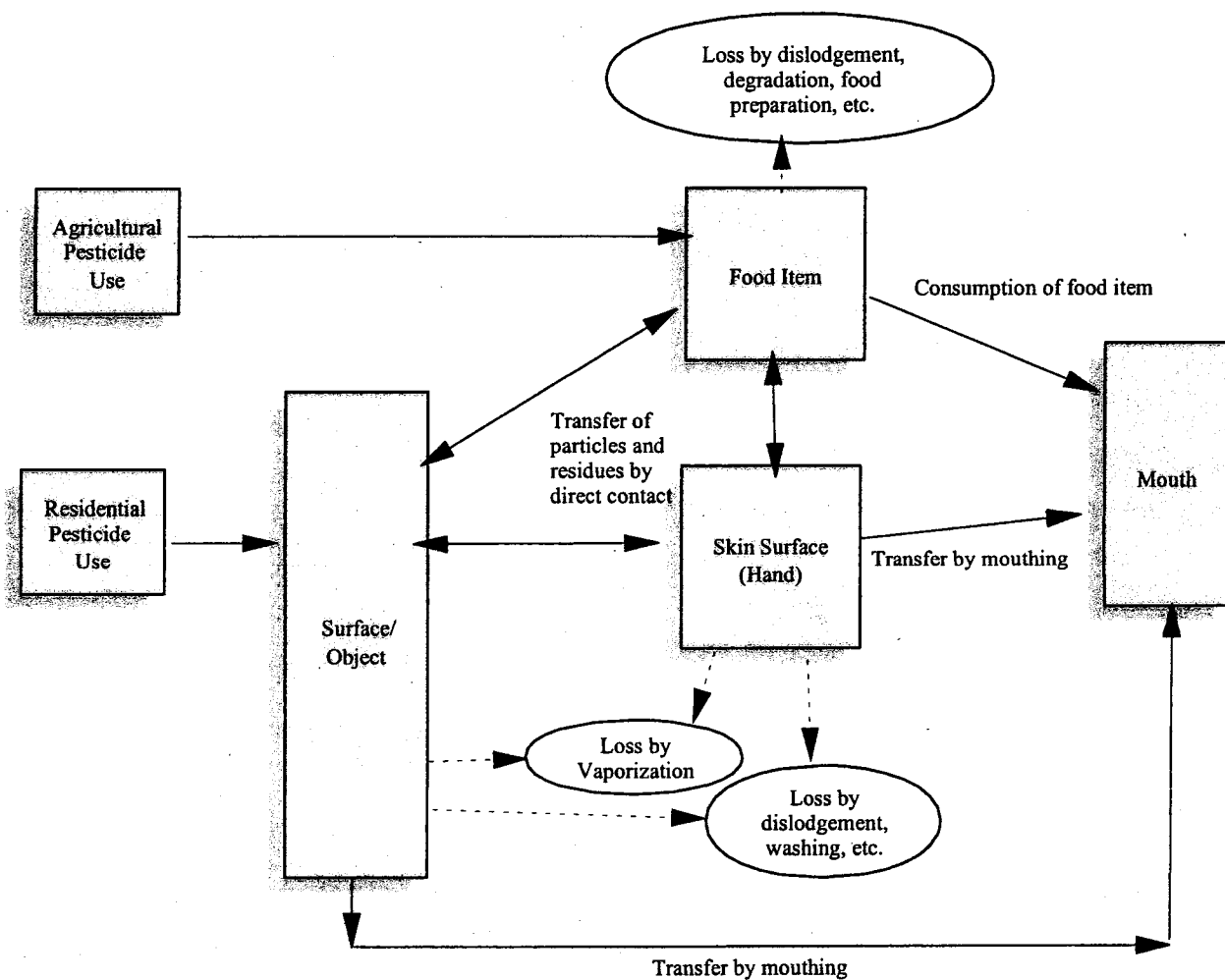
### 3.4 Ingestion Route

Characterizing ingestion of pesticides by children may involve several pathways:

- Direct ingestion of foods brought into the home or other eating places containing pesticide residues primarily from agricultural applications, but also from contamination during storage and preparation in the residential or other environments (i.e., dietary ingestion),
- Ingestion of foods that have been contaminated as the result of contact with contaminated hands and surfaces during preparation and consumption,
- Pesticide residues ingested while mouthing contaminated hands and objects, and
- Ingestion of contaminated soils or contaminated house dust found in the residential or other environments.

A conceptual model of the potential pathways for ingestion exposure is presented in Figure 3-1. Ingestion pathways 2 through 4 above are referred to here as indirect ingestion and may be the result of hand-to-mouth, object-to-mouth, or hand-to-object-to-mouth activity where the objects may be items such foods contaminated while being consumed or toys contaminated in the child's environment as a result of routine activities.

Figure 3-1. Conceptual model of pesticide exposure by the ingestion route.



Infants and young children may be particularly vulnerable to exposure by the ingestion route for several reasons. The specific foods comprising the diets children eat may result in higher dietary ingestion of contaminants and children eat more relative to their body weights than adults. Indirect ingestion of contaminants may also occur when children handle and eat foods that have come in contact with the floor or other contaminated surfaces. In many cases, indirect ingestion may occur after repeated contacts of the same object (food or any other object that enters the mouth) with multiple contaminated media, and from multiple contacts with the mouth. For example, a food item may contact several surfaces, including eating surfaces, hands, and utensils, before it is partially or completely ingested. Finally, children's mouthing activities will result in indirect ingestion of environmental contaminants if the hands or non-dietary objects entering the mouth are contaminated.

### **3.4.1 Dietary Ingestion**

To determine the ingestion of pesticides through the dietary pathway, duplicates of all foods consumed (i.e., duplicate diets) during the monitoring period are collected and analyzed. In duplicate diet studies for children, a caregiver generally provides second portions (e.g., a duplicate plate) of the foods given to the child for consumption. The portions are identical to what has been served to the child with respect to preparation, type of food, and amount of food. Following the eating activity by the child, portions are adjusted to account for foods not consumed (e.g., a duplicate diet). The distinction between a duplicate plate (consisting of all foods served) and a duplicate diet (consisting of all foods eaten) is typically more significant for children than adults because significant quantities of food may be left uneaten. During the brief monitoring period for the child, duplicate diet methodology should provide the most accurate measure of dietary exposure because it accounts for all foods consumed, even those from non-commercial sources (Thomas et al., 1997). It accounts for gains or losses of the contaminant that may occur during transport, storage, and preparation, and most importantly, when combined with methodology to assess indirect ingestion during consumption, measures the actual and total contaminant intake for the child during the exposure monitoring period.

Duplicate diets are collected over some specific period of time, with one day being the time increment most often used (i.e., for acute assessment). For longer periods (i.e., short-term, intermediate term or chronic assessments), multiple consecutive or non-consecutive days may be used to better describe the child's contaminant intake, since there is the potential for a large daily intake variability. In some cases, particularly for risk assessment, it may be necessary to collect duplicate diet samples at different times of the year to assess seasonal intake variability.

**Exposure Algorithm.** Assessing the dietary ingestion of a contaminant is estimated by the sum of the concentration of the contaminants multiplied by the amount consumed of all foods

eaten during the monitoring period.

$$E_f = \sum C_f W_f \quad (9)$$

where

$E_f$	=	the total dietary ingestion during the 24-hr period ( $\mu\text{g/d}$ )
$C_f$	=	concentration of pesticide in the food item ( $\mu\text{g/kg}$ )
$W_f$	=	weight of food item consumed ( $\text{kg/d}$ )

**Data Requirements.** To assess total contaminant intake, duplicate portions of all foods and beverages consumed by the child must be collected and analyzed. In selected cases, a study may focus on the contaminant intake from a single dietary source (e.g., fish or vegetables), and the duplicate diet methodology can be applied to the specific food or group of foods.

### 3.4.2 Indirect Ingestion

To date, indirect ingestion has been estimated using two approaches. One approach is similar to the microactivity approach described in Section 3.3.2 for assessing dermal exposure by which each contact with a contaminated medium is described (U.S. EPA 1997b, Melnyk et al., 2000, Akland et al., 2000). A second approach is to measure the concentration of pesticide or contaminant in soil or house dust and then assume a mass of soil/dust that is consumed by the child (U.S. EPA 1997b) in association with activities such as mouthing objects or eating foods. A third approach is proposed here that uses some additional assumptions to lump details associated with some of the exposure factors and activity patterns leading to indirect ingestion. This macroactivity-type approach allows for a simplified assessment of indirect ingestion exposure to an individual based on measurement data collected in the field and factors that characterize the activities that lead to indirect ingestion of contaminants.

**Exposure Algorithm for Microactivity Approach.** To assess indirect ingestion exposure using the microactivity approach, exposure is estimated individually for all of the microactivities (e.g., hand-to-mouth, object-to-mouth, food-to-mouth, hand-to-food-to-mouth contacts) in which indirect ingestion occurs. Exposure during the 24-h period is then the sum of all of the individual exposures:

$$E_{\text{ing/mi24}} = \sum E_{\text{ing/mi}} \quad (10)$$

where

$E_{\text{ing/mi24}}$	=	indirect ingestion exposure during the 24-h period for all microactivities ( $\mu\text{g/d}$ )
$E_{\text{ing/mi}}$	=	indirect ingestion exposure for each microactivity during the 24-h period ( $\mu\text{g/d}$ )

For each microactivity, indirect ingestion exposure during the 24-h period can be defined as:

$$E_{ing/mi} = (C_{surfx})(TE_x)(SA_{xm})(EF) \quad (11)$$

where

$E_{ing/mi}$	=	indirect ingestion exposure for each microactivity over a 24-h period ( $\mu\text{g/d}$ )
$x$	=	hand, object, food item or anything else that enters the mouth
$C_{surfx}$	=	surface loading (total or transferable) on $x$ ( $\mu\text{g/cm}^2$ )
$TE_x$	=	transfer efficiency of contaminant from $x$ to mouth (unitless)
$SA_{xm}$	=	area of $x$ contacted by mouth ( $\text{cm}^2/\text{event}$ )
$EF$	=	frequency of indirect ingestion events over a 24-h period (event/d)

For food or any other item that is ultimately consumed,  $TE_x$  is equal to unity. When transfer from  $x$  to the mouth is for items other than food,  $TE_x$  is a function of:

- Characteristics of  $x$  (hard, plush, porous, moisture, oil content, age, loading) and
- Contact mechanics (sucking, licking, duration, repetition)

In addition, the loading of pesticide on an object (e.g., toy or food) contaminated as a result of contact with a contaminated residential surface (e.g., hand or floor) can be defined as:

$$C_{surfx} = (C_y)(TE_y)[(SA_{yx}) / (SA_{xy})] \quad (12)$$

where

$C_{surfx}$	=	surface loading (total or transferable) on $x$ ( $\mu\text{g/cm}^2$ )
$x$	=	hand, object, food item or anything else that enters the mouth
$C_y$	=	surface loading (total or transferable) on surface $y$ ( $\mu\text{g/cm}^2$ )
$y$	=	contaminated residential surface
$TE_y$	=	transfer efficiency of contaminant from $y$ to $x$ (unitless)
$SA_{xy}$	=	area of the object ( $x$ ) contaminated as a result of contact with contaminated surface ( $y$ ) [ $\text{cm}^2$ ]
$SA_{yx}$	=	area of the surface ( $y$ ) contacted by the object ( $x$ ) [ $\text{cm}^2$ ]

Note that the surface to object transfer efficiency ( $TE_y$ ) is a function of:

- Form of the pesticide (residue, particle bound, formulation, age, physicochemical properties),
- Characteristics of surfaces (hard, plush, porous, loading, previous transfer),
- Characteristics of  $x$  (moisture, oil or fat content, age, loading, previous transfer),
- Contact mechanics (pressure, duration, smudge, repetition), and
- Environmental conditions (temperature, relative humidity, air exchange, redeposition)

rate).

**Data Requirements for Microactivity Approach.** To use the microactivity approach, a significant level of detail is needed to characterize the potential for children's indirect ingestion exposure to chemical residues and to quantify intake. Information and data required to estimate indirect exposures include the following.

*Common data needs for all events that lead to indirect ingestion exposure:*

- Information on microenvironments/macroactivities that lead to indirect ingestion,
- Surface loadings in the important microenvironments,
- Residue loadings on hands, if the child's hands are in contact with objects mouthed or ingested, and
- Information on an individual child's hand washing practices.

*Data needs for indirect ingestion exposure due to hand-to-mouth activities:*

- Fraction of residue transferred from the hands to mouth during a mouthing event,
- Number of mouthing events in a 24-h period, and
- Surface area of hand contacted by the mouth.

*Data needs for indirect ingestion exposure due to surface (including hand)- or object-to-mouth activities:*

- Information on what surfaces, body parts, toys, etc., are mouthed,
- Surface loadings for any objects or surfaces (including hands) mouthed by children,
- Transfer efficiency from the surface (including hands) to mouth during a mouthing event,
- Number of mouthing events during a 24-h period, and
- Surface area of object mouthed.

*Data needs for indirect ingestion exposure due to consumption of handled food:*

- Information on locations where an individual child consumes foods,
- Information on handled and consumed foods for an individual child,
- Area of surfaces and hands contacted by food,
- Transfer efficiency from surface or hand to food,
- Number and duration of food-to-hand and food-to-surface contact events, and
- Information on amount of specific foods that are consumed.

**Macroactivity Approach.** Because it would be too burdensome and costly to collect all the data required to apply the microactivity approach for the time-sequence of events that occurs on an individual basis, a macroactivity approach is proposed here to provide a simplified

assessment of indirect ingestion exposure to an individual based on measurement data collected in the field. In this approach, objects (including hands and 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 this approach, only equations 10 and 11 are used. Information on each of the individual contacts and transfer leading up to a surface loading on an important item is lumped into the one loading measurement taken from that item. In addition, the items identified as most often mouthed and/or eaten are assumed to represent the most significant sources of indirect ingestion exposure. Note that a macroactivity approach analogous to the one used for dermal exposure is not recommended here for indirect exposure. Currently, a method for developing empirically derived transfer coefficients that lump mouthing contact, surface area, and transfer for a series of mouthing events does not exist. No measure of indirect ingestion exposure analogous to a dermal dosimeter exists. It is possible that in the future, controlled studies could be conducted using a nontoxic tracer that could be tracked in biological samples such as urine. Such a tracer would need to be applied as a surrogate for the environmental contaminants of interest in a setting where children could interact with the items of interest and exposures could be limited to indirect ingestion pathways. For now, we propose an approach which attempts to directly link surface loadings and indirect ingestion activities to provide a very basic screening assessment of indirect ingestion exposure.

**Data Requirements for Macroactivity Approach.** To use this macroactivity approach to assess indirect ingestion exposure for an individual in a measurement study, information on residue concentrations and factors characterizing general contact with items that are mouthed or consumed is combined with transfer efficiencies that have been measured in the laboratory. Research is continuing on the parameters that characterize the most common eating and mouthing activities. The type of data that must be collected in the field include the following.

*Data needs for indirect ingestion exposure due to hand-to-mouth activities:*

- Residue loadings on the hands,
- General information on the frequency and nature (e.g., portion of hand that is mouthed) of hand-to-mouth activity, and
- Information on an individual child's hand washing practices.

*Data needs for indirect ingestion exposure due to surface- or object-to-mouth activities, other than hands:*

- Information on most commonly mouthed objects for an individual child,
- Surface residue loading measured from these objects, and

- General information on the frequency and nature (e.g., portion of object that is mouthed) of mouthing.

*Data needs for indirect ingestion exposure due to consumption of handled food:*

- Information on most commonly handled and consumed foods for an individual child,
- Information on contacts of foods with intermediate surfaces, including hands,
- Samples of these foods collected after handling, and
- General information on amount of these foods that are consumed.



## 4.0 EXPOSURE SCENARIO

While many of the methods and approaches presented in this protocol should be generally applicable or easily modified to address many children's exposure scenarios, this protocol focuses on exposure of infants and young children to pesticides. The exposure scenarios used in the development of this protocol are summarized in Table 4-1 and described below.

Table 4-1. Scenario for Protocol Development

Parameter	Description
Pesticide Source	Any residential or daycare pesticide application
Exposure Population	Children 4 years old or younger
Time Frame for Exposure	Short-term, 1 to 7 days following application
Microenvironments	Indoors at home, outdoors at home, indoors at daycare centers, and outdoors at daycare centers
Activities	Active play, quiet play, sleeping, and eating

**Sources.** This protocol focuses on sources of pesticides in the residential and daycare center environments. Indoor sources include: regularly scheduled professional crack and crevice applications; general residential use of off-the-shelf formulations; and outdoor sources of turf and garden pesticides. Following outdoor applications, exposure indoors may occur due to infiltration of outdoor air into the residence or daycare or track-in of residues or particle-bound pesticides.

For dietary exposures to occur, foods must contain pesticide residues, then the food must be consumed. Many different sources can contribute to dietary residues and subsequent exposure: foods containing pesticide residues are purchased from a commercial source and eaten; foods containing residues are obtained from a noncommercial source (i.e., home gardens) and eaten; and, foods from either commercial or noncommercial sources are obtained then subsequently contaminated during transport, storage, or preparation. Lastly, foods from all sources can be subsequently contaminated during consumption by a child (i.e., indirect ingestion exposure).

**Exposed population.** The protocol describes the approaches for estimating the exposures of children 4 years old or younger. Very young children may be particularly

susceptible to pesticide exposures as the result of the microenvironments in which they spend time (e.g., kitchen floor), and the activities in which they are involved (e.g., mouthing of hands and toys and handling foods). It is important to understand that physiological characteristics and behavioral patterns will result not only in different exposures for children and adults, but also for children of different developmental stages. Thus, exposure assessments are required for children in each age group, with age group being defined by developmental stage. Developing a classification scheme for children by age group has been the subject of significant debate. The Risk Assessment Forum (RAF) held a workshop on this topic in July of 2000 (U.S. EPA, 2000b). Some examples associated with relevant age-related developments for several exposure pathways are presented in Table 4-2. The age bins recommended by the RAF workshop for classifying children based on behavior are presented in Table 4-3.

Table 4-2. Relevant Age-Related Developments (From U.S. EPA, 2000b)

Exposure Pathway	Examples of Relevant Age-Related Developments
Breast Milk/Nursing	Nursing takes place roughly from 0 to 18 months of age, though this varies by culture.
Bottle Feeding	Bottle feeding takes place roughly from 0 to 12 or 24 months.
Food	Head control (2 months), sitting (6 months), finger feeding (8 to 9 months), use of utensils (10 to 12 months), and the final shift to adult patterns of eating. Solid food, served in a bottle as a slurry, is often consumed as early as 1 month of age, but 4 to 6 months is the typical age range for beginning solid foods by themselves.
Water	Use of cups (6 to 9 months).
Mouth-Hand Contact	Prevalence of hand-to-mouth behaviors, such as thumb-sucking. Gross motor skills determine access to areas where the hand can become contaminated. Succession of gross motor milestones: rolling (4 months), creeping (6 months), crawling (8 months), walking (12 months), and climbing (18 months).
Mouth-Object Contact	The ability to interact with objects is a major factor. The ability to grasp an object to one's mouth begins roughly at 3 to 5 months. A pincer grasp and moderate strength are achieved by 9 months. Children become aware that objects exist even when covered around 6 months but generally do not understand the meaning of the word "no" until 12 months.

Table 4-3. Behavioral Age Bins (From U.S. EPA, 2000b).

Age Bin	Characteristics Relevant to Oral and Dermal Exposure	Characteristics Relevant to Inhalation Exposure
0 to 2 months	Breast and bottle feeding. Hand-to-mouth activities. Rapid growth makes children particularly vulnerable to chemicals.	Children spend a great deal of their time asleep.
3 to 5 months.	Solid food is introduced. Contact with surfaces increases. Object-to-mouth activities increase.	Children may breathe close to floor level when placed in play pens or infant seats on the floor.
6 to 11 months	Food consumption expands. Children's floor mobility increases. Children are increasingly likely to mouth non-food items.	Development of personal dust clouds.
12 to 23 months	Children consume a full range of foods. They participate in increased play activities, are extremely curious, and exercise poor judgment. Breast and bottle feeding cease.	Children walk upright, run, and climb. They occupy a wider variety of breathing zones and engage in more vigorous activities.
2 to 5 years	Children begin wearing adult-style clothing. Hand-to-mouth activities begin to approximate adult patterns.	Occupancy of outdoor spaces increases.
6 to 10 years	There is decreased oral contact with hands and non-food items, as well as decreased dermal contact with surfaces.	Children spend time in school environments and begin playing sports.
11 to 15 years	Smoking may begin. There is an increased rate of food consumption.	Increased independence. Work outside of home begins.
16 to 20 years	High rate of food consumption continues.	Independent driving begins. Expanded work opportunities.

**Time frame of exposure.** This protocol focuses on high-level, short-term (one to seven days post-application) exposures resulting from recent pesticide applications. This time frame may result in relatively high exposures. Because the explicit focus of this research is exposure and not health risk, the relative health implications from a series of higher short-term exposures versus lower chronic exposures were not considered although this is an important question requiring a significant research effort. It is also assumed that for this time frame for indoor applications, pesticides are primarily present in the form of residues, rather than being particle-bound.

**Microenvironments of exposure.** The protocol addresses data collection in residential dwellings and daycare centers, which are considered the most important microenvironments for the exposure of infants and very young children. Both the indoor and outdoor microenvironments are considered.

**Activities that result in exposure.** Exposure associated with children's normal daily activities are considered here. These include sleeping, quiet play, active play, and eating. The activities most likely to result in significant exposures are likely to vary with the developmental stage of the child. Activities specifically of interest for the ingestion pathways include all eating and mouthing activities.

**Assumptions.** The most important assumptions made in applying this protocol for assessing exposure for this scenario are as follows:

- The most significant concentrations of pesticide in this exposure time frame are present as residues,
- The distribution of the pesticide residues on foods, objects, surfaces, and in the air in the residential environment is not homogeneous,
- Measurement of residues on hands, objects, and foods collected at specific time points can be used to estimate ingestion exposures over the time frame of interest, and
- In the time frame of concern for this scenario (short-term following an application), exposure resulting from ingestion of soil and house dust is less important than indirect ingestion of residues.

For dietary exposure, transport, storage, preparation, and consumption may have an affect on the pesticide levels in the foods. All but the consumption aspects of these activities are taken into consideration when duplicate diet samples are collected. The most important assumptions made for assessing dietary exposure by the duplicate diet methods are as follows:

- Sample collected represents the foods consumed by the child,
- The portion sizes are adjusted for actual amounts of foods eaten,
- The variability in pesticide levels in the foods collected and those eaten is very small, and
- Exposures are only representative of those incurred during the monitoring period.

## **5.0 APPROACH FOR ESTIMATING INHALATION EXPOSURE**

### **5.1 Introduction**

Inhalation is a potentially important route of exposure to pesticides for children in residences, daycares, schools, and other microenvironments. Inhalation exposure depends on many factors including the physical characteristics of the pesticides (e.g., vapor pressure), formulation, application method (e.g., crack and crevice application versus room fogger), location of application (e.g., indoors, outdoors, basement, living areas), and factors related to the macroenvironment (e.g., air exchange rate of the building, mixing between rooms, indoor temperature). Inhalation exposure may be more significant for very young children than for older children or adults. Infants and young children have a higher resting metabolic rate and rate of oxygen consumption per unit body weight than adults. They may also spend more time indoors and in closer proximity to pesticide sources (e.g., while playing or sitting on the floor). Young children who spend substantial amounts of time in residences may also have potentially higher inhalation exposure than children in daycares or schools due to lower air exchange rates in homes than in commercial buildings. However, the latter types of buildings may have higher pesticide usage than residences.

Inhalation exposure has been estimated for a wide range of volatile and semi-volatile organic compounds, including pesticides. There have been a number of studies involving measurements of pesticides in air (e.g., Lewis, et al., 1994; Whitmore et al., 1994; Gordon et al., 1999; Quackenboss et al., 2000). Much of the prior data on indoor air concentrations are for concentrations of organophosphorous, organochlorine, and other pesticides that are not currently used indoors. There are few data available on pesticides currently used indoors, such as the pyrethroids. Of all the potential routes of exposure to pesticides, inhalation has been studied the most. The protocols and methods for measurements of pesticides in air are the most well-developed of the aggregate exposure measurement methods.

### **5.2 Summary of Data Requirements**

As described in Section 3.2, inhalation exposure is estimated for each of the microenvironments where a child spends time and for each macroactivity that would result in a different inhalation rate while engaged in that activity. Exposure over the 24-hour period is then the sum of all of the microenvironmental/macroactivity (me/ma) exposures.

The data required to estimate inhalation exposure are summarized in Table 5-1.

### **5.3 General Considerations**

To estimate inhalation exposure for young children, it is necessary to use stationary samplers in selected microenvironments to collect air samples for pesticide analyses. Although

Table 5-1. Data Requirements for Estimating Inhalation Exposure Route

Parameter	Measurement	How Collected	Units
Inhalation Exposure $E_{ime/ma} = C_{ame} \times T_{me/ma} \times IR_{ma}$			
$C_{ame}$	Air concentration in me	Active sorbent collection	$\mu\text{g}/\text{m}^3$
$T_{me/ma}$	Time spent in each me/ma	Time-activity diary, questionnaire	h/d
$IR_{ma}$	Inhalation rate	Estimated from size, age, and activity data collected with diaries and questionnaires and using reference values	$\text{m}^3/\text{h}$

a preferred method for measuring an individual's exposure to air contaminants is to have the study participant wear a personal exposure monitor (PEM), this method is not suitable for young children less than 4 years old. Because it is not possible to measure the air concentrations in all microenvironments that a child may occupy, it is important to identify which microenvironments represent the highest potential exposures based on the amount of time spent in each micro-environment. For children age 0 through 4 years, the important microenvironments include the residence and daycares. For infants, measurement of air concentrations in the home, preferably in the room where the infant spends the most time during the day, will be representative. Because of the small amount of time spent outdoors and the low outdoor concentrations relative to indoor concentrations after pesticide applications, it may not be necessary to measure outdoor air concentrations to estimate an infant's inhalation exposure. As children age and spend more time outdoors, it becomes important to measure outdoor air concentrations, although the levels may be very low for most pesticides.

Microenvironment/macroactivity data need to be collected to identify all of the important microenvironments that a child may occupy. If there are important microenvironments other than the residence and daycare, it is necessary to estimate air concentrations in those micro-environments. To make these estimates, it is generally necessary to assume that there have been no recent applications of the pesticides of concern in that microenvironment and a reasonable concentration must be used for the exposure estimate. This reasonable concentration would be the background concentrations measured outdoors or in indoor microenvironments without recent applications of the target pesticide.

Measurements of indoor air concentrations of pesticides require active pumping systems to collect air samples on sorbent media. Placement of the sampling equipment indoors presents a

variety of sampling challenges because of presence of the occupants, including small children, and the restricted space in indoor environments. Because air monitoring is often intrusive, it is particularly important that field personnel be sensitive to the burden placed on study participants. Pump noise is a critical concern when sampling indoors, particularly in sleeping areas. Low-noise pumps must be used indoors. Noise may be minimized by placing pumps in a small ice chests or metal boxes containing sufficient acoustic insulation to baffle the sound of pump motors. Pumps may be located in closets or behind furnishings to further minimize noise and remove the apparatus from traffic zones.

Because the sampling equipment is left unattended at the sampling site, field teams must consider both the safety of the children in the location where sampling is performed and the potential for tampering with instrumentation or theft (outdoors). Extreme care must be taken so that the pumps, sampling trains, and sorbent tubes pose no safety concerns. Samplers can not be accessible to children or placed on stands that can be tipped over. There should be no small parts that could be removed by children that could cause potential choking hazards. If glass sampling cartridges are used, they must be protected with unbreakable shields that prevent breakage or that will contain all media if breakage occurs. Pumps and sampling cartridges should be placed out of the reach of small children. Appropriate security measures include placing pumps in locked boxes, tamper proof shielding over the pump controls, and the placement of sampling apparatus out of the reach of small children and pets.

Power sources may be unstable in some locations and may produce disruptions during monitoring activities. When possible, battery back-up or an un-interruptible power source should be used to decrease the impact of these occurrences on sample collection.

Selection of sampling locations within a room is important to obtain representative air concentrations. As discussed in the following section, samplers should be placed at an appropriate height and location in the room. They should not be placed near windows, air supply diffusers or returns, or other locations where the air flow may affect air concentrations.

## **5.4 Monitoring and Sampling Methods**

Measurements of pesticides in air for estimates of young children's inhalation exposure require collection of air samples with active sampling systems consisting of sorbent media and vacuum pumps. Concentrations of pesticides in air are obtained by collection of integrated air samples on the sorbent media, extraction of the sampling media, and analysis by an appropriate method, generally gas chromatography (GC) or high performance liquid chromatography (HPLC).

Pesticides are semi-volatile compounds with saturation vapor pressures of less than  $10^{-2}$  kPa. Many of the synthetic pyrethroids (e.g., cyfluthrin, cypermethrin, esfenvalerate) that are currently used for indoor applications, have saturation vapor pressures of less than  $10^{-8}$  kPa. As a

result, the air concentrations of these compounds are generally low and decrease rapidly following an application (Lewis et. al., 2001). Sampling and analysis methods for the current generation of pesticides applied indoors must address the low volatility and potentially low concentrations. The methods must have sufficiently low detection limits and good performance characteristics at low levels. As an example, the median concentrations of chlorpyrifos and diazinon, which are relatively volatile compounds compared to pyrethroids and other current use pesticides, were 8.0 and 4.6 ng/m<sup>3</sup> respectively, in the Arizona NHEXAS samples (Gordon et al., 1999).

The collection of airborne pesticide residues on sorbent media is generally performed using commercially available small, portable, low volume pumps that can be operated over a range of flow rates of 0.1 to 4 L/min. The pumps, which can be operated on batteries or with AC power, must be sufficiently quiet for use in occupied environments and suitable for collection of integrated samples over a 24 hour period. It should be noted that these monitoring pumps as purchased are typically powered by rechargeable NiCad battery packs and are generally designed for 8 to 16 hour occupational exposure monitoring. These pumps may not have sufficient battery life for a 24 hour monitoring, but can usually be modified by a qualified electronics technician or by the manufacturer to operate using disposable alkaline batteries to provide adequate run times. Such modifications will generally void warranties and void the intrinsic safety of the pump for use in hazardous locations. Operation of the pumps on AC power circumvents the need to modify the pumps but lack of easily accessible power outlets may add significant set-up time, create safety hazards by requiring the use of extension cords, or force the collection of samples in less desirable locations due to the difficulties of having to supply AC power. Pump failures may also be caused by unstable or interrupted AC power. If rechargeable batteries are used, care must be taken to insure that the batteries are discharged and charged properly to minimize failure due to charge memory effects. Likewise, if alkaline batteries are used, voltages of the batteries (especially partially used batteries) should be determined prior to beginning sample collection to insure that the batteries will provide sufficient power to operate the pumps for the desired time period.

Flow rates of sampling pumps are set to obtain a specified volume based on the duration of the monitoring period, retention efficiency of the target pesticides on the sorbent, and the sensitivity of the analytical method. High volume pumps are not appropriate for sampling indoors because of considerations of noise and the impact that collection of high air volumes indoors may have on air exchange rates and air movement in the rooms. High volume pumps may be used outdoors.

A variety of sampling media are available for collection of pesticides in air. Available sorbents include polyurethane foam (PUF), Amberlite® XAD-2, Amberlite® XAD-4, Chromosorb® 102, Tenax® GC or TA, and Porapak®-R. These absorbents have similar efficiencies for collection of most pesticides (Lewis, 2000). PUF has been used as the sorbent media in a number of field measurement studies (e.g., Whitmore et al., 1994; Gordon et al.,



1999) and its use is described in an ASTM standard practice (ASTM, 2000a). XAD resin has been used extensively for collection of semi-volatile organic compounds (U.S. EPA, 1999b) and can be used as an alternative to PUF.

Sorbent media may be used individually or in multi-bed combinations. The sample media may be constructed in series to collect both gas phase residues as well as levels of airborne particles. The sampler may consist of a combination of particle sizing devices (to collect only inhalable or respirable particles below 10 or 2.5  $\mu\text{m}$  diameter), membrane filters to collect particles, sorbent resins, and/or polyurethane foam. Selection of the sampling media should be based on the physio-chemical characteristics of the compound(s) of interest, sampling efficiency, retention efficiency, performance characteristics based on available literature or laboratory validation studies for all analytes of interest, cost, and ease of use.

The selection of the multi-residue sampling and analysis methods should consider the following factors:

- The suite of pesticides to be targeted for quantification and their physical and chemical properties,
- Range of air concentrations expected,
- Minimum detection and quantification limits required,
- Availability of validated methods for the pesticides of interest,
- Available performance data (accuracy, precision, detection limits) for the method,
- Required sampling volumes and sampling duration.

Information that can be used to select sampling and analysis methods is available in the scientific literature, ASTM (2000a), and in U.S. EPA (1999b) methods. Information on pesticide sampling methods for measurements in occupational settings is available in publications by NIOSH (1994), OSHA (2000), and manufacturers of samplers and sorbent media. However, researchers conducting exposure measurements in residential and daycare environments should recognize that the methods developed to measure occupational exposure may not be sufficiently sensitive to measure the lower concentrations often encountered in non-occupational environments.

The performance of the sampling and analysis method needs to be fully evaluated prior to use in field studies. As discussed previously, method detection limits must be sufficiently low for measurements in residences and daycares. Typically, detection limits for analysis of pesticides by GC/MS can be expected to be in the range of 5 to 50  $\text{ng}/\text{m}^3$ , which varies by compound and sample collection volume. Precision should be  $\pm 25\%$  and the accuracy, expressed as the percent recovery of spiked samples, should be in the range of 75 to 125%. Users of the methods also need to determine the sampling and retention efficiency of the sorbent media for the target pesticides. Sampling efficiency is the ability of the sampling medium to trap the pesticides of interest (ASTM, 2000a). Retention efficiency is the ability of the sampling

medium to retain the compound of interest. Methods for determining sampling and retention efficiencies are described in ASTM practice D4861 (ASTM, 2000a). This practice lists sampling and retention efficiencies for a number of organochlorine and organophosphorous pesticides and for a few pyrethroids collected on PUF. However, there are limited data on performance characteristics for many of the pyrethroids.

The selection of the indoor sampling locations is contingent on the study objectives. Measurements of pesticide concentrations in the rooms where pesticide applications have been performed recently may provide an estimate of the highest potential exposure. But, measurements in locations where children spend the majority of their time may provide more accurate exposure estimates. The concentrations of pesticides in the air of a residence or daycare may vary substantially in different parts of the building following pesticide applications. Lewis et al. (2001) observed concentrations of diazinon in a bedroom that were less than one-third the concentrations measured in the room of application on the day following application. The difference between the rooms was even greater on the following days. Spatial differences may require measurements in more than one location in a residence or daycare to obtain accurate estimates of inhalation exposure. This may be cost prohibitive in large field studies. Therefore, emphasis should be placed on identifying the location where the child has the highest potential for exposure due to proximity to a source, activities (crawling, playing with pet, eating, etc.) or time spent in a location (i.e. living room, play ground, bedroom etc.). Outdoor sampling locations should be selected that are representative of the areas where the child spends time outdoors. Samplers should not be placed immediately adjacent to buildings where pesticides may be used or stored.

## **5.5 Exposure Factor/Activity Pattern Information**

To estimate inhalation exposure, information must be collected to describe the (1) child, (2) microenvironments occupied by the child, and (3) the child's activity while in those microenvironments.

The minimum information required to characterize the child is the child's age, weight, and gender. These variables are used to estimate inhalation rates.

Information on the child's location (microenvironment) and activity need to be recorded throughout the measurement period. For individual measurement assessments, the total time spent at each level of activity in each microenvironment must be recorded. For studies currently being performed in the NERL Human Exposure Analysis Branch, four microenvironments have been defined: indoors at home, outdoors at home, indoors at daycare centers, and outdoors at daycare centers. These four microenvironments are assumed to provide a reasonable estimate for inhalation exposure for young children (under age 4). Four macroactivities have been defined for this age group: active play, quiet play, sleeping/napping, and eating. An activity diary (Figure 5-1) is used for recording the activities in these microenvironment/macroactivity combinations.

Figure 5-1. Example of one page of the Indoors at Home section of a 24-h time-activity diary for estimating inhalation and dermal exposure of young children

Microenvironment	Clothing Level <sup>1</sup>	Macroactivity	Surface	6:00am	6:15am	6:30am	6:45am	7:00am	7:15am	7:30am	7:45am	8:00am	8:15am	8:30am	8:45am	9:00am	9:15am	9:30am	9:45am	10:00am	10:15am	10:30am	10:45am	11:00am	11:15am	11:30am	11:45am	12:00pm
Indoors at Home		Active Play	Hard Surface																									
			Carpet																									
			Upholstered furniture/ Bedding																									
		Quiet Play	Hard Surface																									
			Carpet																									
			Upholstered furniture/ Bedding																									
		Eating	Hard Surface																									
			Carpet																									
			Upholstered furniture/ Bedding																									
		Sleeping/ Napping	Hard Surface																									
			Carpet																									
			Upholstered furniture/ Bedding																									

<sup>1</sup>LS=long-sleeves; SS=short-sleeves; P=pants; S=socks; SH=shorts; N=naked

The diary also collects information on clothing level and the type of surface in the microenvironment, information needed for dermal exposure estimates. The diary includes multiple pages, one each for indoors at home, outdoors at home, indoors at daycare, and outdoors at daycare and covers a 24-h monitoring period. Activities must be recorded by the parent or caregiver of the child. For the most accurate data collection, activity should be recorded on a continuous basis during the 24-hour measurement period. However, data collection by recall may be suitable if the parent or caregiver is provided with adequate instructions at the start of the measurement period and is aware of the need to record the data at a later period. Recall periods should be kept relatively short, generally no longer than 24 hours. Although, parents and caregivers can provide reasonably accurate data on the location of the child in the four microenvironments, they may have difficulty defining active versus quiet play. Therefore, it is important to provide training to the parent or caregiver on how to determine what to record for the child's activity. To improve estimates of inhalation rates, researchers may want to define additional levels of activity. However, if additional levels of activity are defined, additional training of the parent or caregiver will be required. A videotape of children's different activity levels may be used for that purpose.

## **5.6 Estimation of Inhalation Rates**

Inhalation rates for children are highly variable and are a function of the child's age, weight, and activity. The actual inhalation rates are not routinely measured in individual measurement studies. As an alternative, information on the child's activities is collected during field measurement studies and used to estimate inhalation rates.

Ranges of inhalation rates for children developed by NERL (McCurdy, 2001) using available measurement data are presented in Tables 5-2 and 5-3. The data presented in the tables should be used to calculate inhalation rates based on children's age and weight. Data used to compile the ranges of inhalation rates shown in the tables are the same as those used by the EPA in the *Exposure Factors Handbook* (U.S. EPA, 1997a) and the *Child-Specific Exposure Factors Handbook* (U.S. EPA 2000c).

Inhalation rates for short-term exposures of children under age 18 are presented in the *Child-Specific Exposure Factors Handbook*. But they are based only on the activity levels and do not account for the child's age or weight. The recommended rates from the handbook for children 18 years of age and under are:

- Rest:  $0.3 \text{ m}^3/\text{h}$ ,
- Sedentary Activities:  $0.4 \text{ m}^3/\text{h}$ ,
- Light Activities:  $1.0 \text{ m}^3/\text{h}$ ,
- Moderate Activities:  $1.2 \text{ m}^3/\text{h}$ , and
- Heavy Activities:  $1.9 \text{ m}^3/\text{h}$

Table 5-2. Ranges of Inhalation Rates ( $V_E$ ) for "Normal" Female Children and Adolescents on a per Body Mass Basis by Generalized Type of Activity ( $L \min^{-1} kg^{-1}$ )

Age	Sleep/Nap /Rest	Sedentary/ Sitting Quietly	Light Activity /Walking	Moderate Activity /Jogging	Vigorous Activity/ Running
1	0.17 - 0.20	0.21 - 0.26	0.27 - 0.69	0.70 - 1.05	1.06 - 1.25
2	0.16 - 0.19	0.20 - 0.25	0.26 - 0.68	0.69 - 1.04	1.05 - 1.26
3	0.15 - 0.18	0.19 - 0.23	0.24 - 0.67	0.68 - 1.03	1.04 - 1.27
4	0.14 - 0.17	0.18 - 0.22	0.23 - 0.66	0.67 - 1.02	1.03 - 1.28
5	0.14 - 0.16	0.17 - 0.21	0.22 - 0.65	0.66 - 1.01	1.02 - 1.29
6	0.13 - 0.15	0.16 - 0.20	0.21 - 0.64	0.65 - 1.00	1.01 - 1.30
7	0.12 - 0.14	0.15 - 0.18	0.19 - 0.63	0.64 - 0.96	0.97 - 1.32
8	0.11 - 0.13	0.14 - 0.17	0.18 - 0.60	0.61 - 0.93	0.94 - 1.33
9	0.10 - 0.12	0.13 - 0.16	0.17 - 0.59	0.60 - 0.92	0.93 - 1.34
10	0.10 - 0.12	0.13 - 0.15	0.16 - 0.57	0.58 - 0.91	0.92 - 1.36
11	0.09 - 0.11	0.12 - 0.14	0.15 - 0.53	0.54 - 0.87	0.88 - 1.35
12	0.09 - 0.10	0.11 - 0.13	0.14 - 0.50	0.51 - 0.83	0.84 - 1.35
13	0.08 - 0.10	0.11 - 0.13	0.14 - 0.49	0.50 - 0.80	0.81 - 1.36
14	0.08 - 0.09	0.10 - 0.12	0.13 - 0.48	0.49 - 0.78	0.79 - 1.37
15	0.08 - 0.09	0.10 - 0.11	0.12 - 0.47	0.48 - 0.76	0.77 - 1.38
16	0.07 - 0.08	0.09 - 0.10	0.11 - 0.44	0.45 - 0.73	0.74 - 1.38
17	0.07 - 0.08	0.09 - 0.10	0.11 - 0.43	0.44 - 0.71	0.72 - 1.38
18	0.06 - 0.07	0.08 - 0.09	0.10 - 0.42	0.43 - 0.69	0.70 - 1.38

Notes: 1. These data should only be used for "normal" children and adolescents. Different estimates are needed for obese, underweight/sickly kids, as well as children/adolescents who are very fit due to partaking in frequent and "heavy" exercise.

2. To obtain activity-specific  $V_E$  (in L), simply multiply the estimate shown above by time spent in each category (in minutes) and also by body weight (in kg) of the child/adolescent in question. These values can then be converted into  $m^3$  per whatever time period is of interest by multiplying by the appropriate unit conversions.

3. The values shown have been "smoothed" to minimize abrupt jumps by age. The data within a range for each activity level probably are distributed log-normally, but definitive information on this distribution is scanty. The upper bound of the "Vigorous" class is the same as  $V_{E\max}$ , and it cannot be maintained more than approximately 5 minutes before it declines over time; see Bink (1962) and Erb (1981).

Sources: U.S. EPA (2000c), U.S. EPA (1997a), and McCurdy (2001)

Table 5-3. Ranges of Inhalation Rates ( $V_E$ ) for "Normal" Male Children and Adolescents on a per Body Mass Basis by Generalized Type of Activity ( $L \min^{-1} kg^{-1}$ )

Age	Sleep/Nap /Rest	Sedentary /Sitting Quietly	Light Activity /Walking	Moderate Activity /Jogging	Vigorous Activity /Running
1	0.18 - 0.21	0.22 - 0.27	0.28 - 0.74	0.75 - 1.13	1.14 - 1.76
2	0.17 - 0.20	0.21 - 0.26	0.27 - 0.74	0.75 - 1.13	1.05 - 1.77
3	0.16 - 0.19	0.20 - 0.24	0.25 - 0.73	0.74 - 1.12	1.04 - 1.78
4	0.15 - 0.18	0.19 - 0.23	0.24 - 0.72	0.73 - 1.12	1.13 - 1.79
5	0.14 - 0.17	0.18 - 0.21	0.22 - 0.71	0.72 - 1.11	1.12 - 1.80
6	0.14 - 0.16	0.17 - 0.20	0.21 - 0.70	0.71 - 1.10	1.11 - 1.81
7	0.13 - 0.15	0.16 - 0.19	0.20 - 0.69	0.70 - 1.05	1.06 - 1.83
8	0.12 - 0.14	0.15 - 0.17	0.18 - 0.64	0.65 - 1.04	1.05 - 1.77
9	0.11 - 0.13	0.14 - 0.16	0.17 - 0.64	0.65 - 1.03	1.04 - 1.72
10	0.11 - 0.13	0.14 - 0.15	0.16 - 0.63	0.64 - 1.00	1.01 - 1.64
11	0.10 - 0.12	0.13 - 0.15	0.16 - 0.59	0.60 - 0.95	0.96 - 1.59
12	0.09 - 0.11	0.12 - 0.13	0.14 - 0.57	0.58 - 0.94	0.95 - 1.56
13	0.09 - 0.10	0.11 - 0.12	0.13 - 0.56	0.57 - 0.93	0.94 - 1.50
14	0.09 - 0.10	0.11 - 0.12	0.13 - 0.55	0.56 - 0.90	0.91 - 1.47
15	0.08 - 0.09	0.10 - 0.11	0.12 - 0.53	0.54 - 0.89	0.90 - 1.44
16	0.07 - 0.08	0.09 - 0.10	0.11 - 0.52	0.53 - 0.88	0.89 - 1.42
17	0.07 - 0.08	0.09 - 0.10	0.11 - 0.51	0.52 - 0.87	0.88 - 1.39
18	0.07 - 0.08	0.09 - 0.09	0.10 - 0.51	0.52 - 0.86	0.87 - 1.38

Notes: 1. These data should only be used for "normal" children and adolescents. Different estimates are needed for obese, underweight/sickly kids, as well as children/adolescents who are very fit due to partaking in frequent and "heavy" exercise.

2. To obtain activity-specific  $V_E$  (in L), simply multiply the estimate shown above by time spent in each category (in minutes) and also by body weight (in kg) of the child/adolescent in question. These values can then be converted into  $m^3$  per whatever time period is of interest by multiplying by the appropriate unit conversions.

3. The values shown have been "smoothed" to minimize abrupt jumps by age. The data within a range for each activity level probably are distributed log-normally, but definitive information on this distribution is scant. The upper bound of the "Vigorous" class is the same as  $V_{EMax}$ , and it cannot be maintained more than approximately 5 minutes before it declines over time; see Bink (1962) and Erb (1981).

Sources: U.S. EPA (2000c), U.S. EPA (1997a), and McCurdy (2001)

These inhalation rates are within the range of rates presented in Tables 5-2 and 5-3. But more accurate estimates of inhalation exposure can be made using the range of rates presented in the tables for calculations based on the child's age and weight.

## **6.0 MACROACTIVITY APPROACH FOR ESTIMATING DERMAL EXPOSURE**

### **6.1 Introduction**

Data on children's exposures and activities are currently very limited and insufficient to support quantitative assessments that do not rely heavily on major default assumptions. Results derived from an initial assessment of critical exposure pathways and factors for assessing children's residential exposures to pesticides indicate that dermal exposure and indirect nondietary ingestion exposure may result in high residential exposures for children (Cohen Hubal et al. 2000).

Two main approaches are currently used to assess dermal exposure. These assessment approaches provide different ways of integrating exposure over time and space. In the macroactivity approach, exposure is estimated individually for each of the microenvironments where a child spends time and each macroactivity that the child conducts within that microenvironment. To do this, exposure is modeled using empirically derived transfer coefficients to aggregate the mass transfer associated with a series of contacts with a contaminated medium. In the microactivity approach, exposure is explicitly modeled as a series of discrete transfers resulting from each contact with a contaminated medium. The algorithm and data requirements for the microactivity approach were described briefly in Section 3.3.2. However, as discussed in that section, implementation of the microactivity approach is not practical in large exposure field studies. Therefore, details of the approach are not discussed in this protocol. This section describes the macroactivity approach for estimating dermal exposure.

### **6.2 Summary of Data Requirements**

To estimate exposure using the macroactivity approach, microenvironments are defined by location and surface type. Activity- and microenvironment- specific transfer coefficients are developed in laboratory experiments or controlled field studies. Exposure can then be estimated individually for each of the microenvironments where a child spends time and each macroactivity that the child conducts within that microenvironment using information on surface loadings, the empirically-derived transfer coefficients, and information on the amount of time in that microenvironment/macroactivity. The dermal exposure algorithm and data requirements are presented in Table 6-1.

### **6.3 General Considerations**

Numerous data must be considered for each microenvironment/macroactivity combination when estimating dermal exposure. These data include: definitions of the microenvironments/macroactivities that are important for dermal exposure; surface loadings (total or transferable) of pesticides or chemicals for each microenvironment/macroactivity combination; amount of time a child spends in each microenvironment/macroactivity during a



Table 6-1. Data Requirements for Estimating Dermal Exposure With the Macroactivity Approach

Parameter	Measurement	How Collected	Units
Dermal Exposure - Macroactivity approach $E_{dme/ma} = (C_{surf})(TC_{me/ma})(AD_{me/ma})$			
$C_{surf}$	Surface loading (total or transferable) in the me	Measure with $C_{18}$ surface press sampler, PUF roller, surface wipe, or soil sample	$\mu\text{g}/\text{cm}^2$
$TC_{me/ma}$	Transfer coefficient	Empirically determined for each me/ma from laboratory or field studies	$\text{cm}^2/\text{h}$
$AD_{me/ma}$	Exposure duration based on location, activity level, clothing	Time-activity diary, questionnaire	h/d

24-h period; and, the transfer coefficient for each microenvironment/macroactivity. These data considerations affect the subsequent sampling considerations. Each consideration is discussed more fully in the following paragraphs. Table 6-2 lists the various microenvironment /macroactivity combinations that are applicable to the exposure scenario described in Section 4.

For each microenvironment/macroactivity combination, the surface loading of the chemical must be determined. For recent pesticide applications, the assumption, based on limited field data, is that the pesticide surface loading is not homogeneous in the residential or daycare center environment. In order for the measurements that are collected to be most applicable and representative of the child's environment, it is important to determine those locations where the child spends the most time in the residential or daycare center environment and sample accordingly. For each home or daycare center, measurements will be made for only those surfaces for which the child is expected to have substantial contact. This sample measurement can be collected using the  $C_{18}$  surface press sampler, a PUF roller, surface wipe, or for outdoor locations to collect a soil or turf sample.

For each microenvironment/macroactivity combination, the amount of time the child spends in each combination must be determined. These data are collected using time-activity diaries or questionnaires.

Table 6-2. Microenvironment/Macroactivity Combinations for Estimating Dermal Exposure

Location	Surface	Activity			
		Eating	Sleeping/ napping	Quiet play	Active play
Indoor at home	Carpet	x	x	x	x
	Hard surface	x	x	x	x
	Upholstered furniture/bedding	x	x	x	x
Outdoor at home	Grass	x	x	x	x
	Soil	x		x	x
	Pavement	x		x	x
Indoor at daycare	Carpet	x	x	x	x
	Hard surface	x	x	x	x
	Upholstered furniture/bedding	x	x	x	x
Outdoor at daycare	Grass	x	x	x	x
	Soil	x		x	x
	Pavement	x		x	x

The transfer coefficients for each microenvironment/macroactivity must also be determined. These are data that are currently not available and need to be generated experimentally in the laboratory or carefully controlled field experiments. This is discussed more fully in Section 6.6, Estimation of Transfer Coefficients.

Environmental monitoring methods for assessing dermal exposure have few equipment considerations, as compared to the inhalation route. However, the concentrations associated with each sample are dependent on the locations sampled and the sampling method used. Pesticide distributions in the residential environment are not homogeneous which may result in significant concentration differences in adjacently sampled areas potentially leading to an over or underestimation of exposures depending on the representativeness of the sampling locations. For this reason, the appropriate sampling locations, sampling methods that address various surface

types, and the number of samples required to provide representative information must all be considered. Outlined below are issues that should be considered when assessing the dermal route of exposure as a component of a large field study:

- Surface sampling methods should be matched to the sampling method that was used to generate the transfer coefficients. For example, transferable residue measurements should be collected using a C<sub>18</sub> surface press sampler method if the C<sub>18</sub> surface press sampler method was used to generate the transfer coefficient.
- Methods of collection should be appropriate for the types of surfaces being monitored. For example, surface wipe samples should be collected from hard surfaces and not from carpets or other fabric surfaces.
- When possible, multiple individual samples (at least three) should be collected from various areas in the microenvironment where the child is in contact with surfaces. Analyzed individually, these samples will provide information regarding the distribution of pesticides in the microenvironment; when the results are combined, they provide an average value to help minimize under or overestimation of surface loadings, and thus, exposure.
- Locations for monitoring and sample collection should be selected that are representative of where the child spends his/her time. Locations targeted for monitoring should be consistent with the study objective. Avoid monitoring locations that may bias the results such as points of application or near sources of potential contamination unless they are part of the study design.
- To reduce analytical costs, it may be possible to collect aggregate samples (e.g., surface press), or combine samples (e.g., surface wipes) prior to analysis to obtain “average” concentrations.
- Be cognizant of the potential damage that sampling methods may pose to personal property. For example, isopropanol used during the collection of surface wipe samples can cause damage to wood finishes or other sensitive surfaces; therefore, samples should not be collected from surfaces that would be damaged by the collection method.

#### **6.4 Monitoring Methods**

As discussed above in General Considerations, the loadings of pesticides on surfaces associated with each of the microenvironment/macroactivity combinations that a child comes into contact with are essential data for estimating dermal exposure. However, since it is impossible to measure the pesticide loadings for all surfaces in all situations, surface loadings are measured for only those areas for which children are expected to have substantial dermal contact. It is extremely important that the surfaces that are sampled are representative of the surfaces for which the child may have appreciable contact during his/her various activities. These areas would include indoor surfaces, either at home or in a daycare center, such as floors and furniture, as well as outdoor surfaces such as grass, soil, and pavement. Floors include both carpeted areas and hard surface areas such as vinyl, tile, or wood. Furniture can be categorized as upholstered

(fabric) or hard (metal, vinyl, wood) surfaces. These previously mentioned microenvironment locations are specific to the exposure scenario as defined in Section 4. It should be recognize that other locations may be of equal, or greater importance for other exposure scenarios. For example, the interior of motor vehicles may be an important microenvironment in agricultural areas due to spray drift from pesticide applications on crops. Exposure to pesticides bound to settled dust on the seats or other interior surfaces of a vehicle may be a source of exposure.

Measurements to determine surface loadings can be divided into two categories: those representing the total amount of pesticide residue present on a surface and those representing the amount of pesticide residue that is available for transfer (i.e., transferable residues). Methods for estimating total pesticide residues include the collection, extraction, and analysis of upholstery fabrics, carpet, and soil samples. Transferable residues can be estimated by collecting and analyzing surface wipe, surface press or roller samples. These latter methods provide an estimate of the amount of each pesticide that is available for transfer from one surface to another. The methods used must be appropriate to the type of surface to be sampled.

The transferable residue measurement methods do not necessarily simulate the contact that a child may make with the surface being sampled and should not be considered surrogate exposure methods. It is not feasible to simulate all of the skin surfaces of a child that may contact surfaces in the microenvironment. A child may contact a surface with the feet, knees, legs, bottom, arms, hands, or face. Skin surfaces may be dry, wet, or sticky. Rather than attempting to simulate these skin surfaces, the approach has been to use a method to measure transferable residues on the surfaces in conjunction with transfer coefficients to estimate dermal exposure. As discussed previously, the transfer coefficients must be developed in laboratory tests or under carefully controlled field experiments. The transfer coefficients must be developed using the same method for measuring surface loading as is used in the field measurement studies. Transfer coefficients can be developed for a wide variety of microenvironment/macroactivity combinations, hard and soft surfaces, as well as dry, wet, and/or sticky skin.

Brief descriptions of some of the more common methods developed for estimating total and transferable pesticide residues are discussed below.

PUF Roller. The PUF roller method is designed to estimate transferable residues from carpeted or hard flooring surfaces (Camann et al., 1996). It has also been used to measure transferable residues from outdoor surfaces such as turf (Nishioka, et al., 1999). It is described in ASTM Standard Practice D6333 (ASTM 2000b) and consists of a polyurethane foam sleeve roller attached to an aluminum frame of specified dimensions and weight. The foam sleeve is rolled across a specified area of the surface being monitored at a specified pressure (6900 to 8600 Pa). The foam sleeve is removed, solvent extracted and analyzed for pesticides by GC/MS, GC/ECD, or other suitable instrumental method.

Drag Sled. The Drag Sled sampler is designed to estimate transferable residues from

floor surfaces (Vaccaro and Cranston, 1990). The sampler consists of a 7.6 cm x 7.6 cm x 1.9 cm block of wood or other material that holds a 10 cm x 10 cm piece of denim cloth as the sampling media. With the denim cloth in contact with the flooring surface, a weight is applied to the block to provide a specified pressure (4500 Pa). The device is pulled at a specified rate across the floor area. The denim cloth is removed, extracted, and analyzed for pesticides by GC/MS, GC/ECD, or other suitable instrumental method. This method has not been used extensively.

California Roller. The California Roller, described by Ross et al. (1991), is designed to estimate transferable residues from indoor and outdoor surfaces. The sampler consists of a large weighted roller that uses polyester-cotton percale bedding material as the sampling media. The roller is a large cylinder (13 cm OD, 63 cm in length) constructed of polyvinyl chloride pipe, partially filled with steel shot to provide a specified pressure (2300 Pa), and is covered with a 1 inch foam cushion. The polyester-cotton percale material is placed directly in contact with the flooring surface to be sampled, a sheet of plastic is placed on top of the cloth and the roller is rolled over the plastic/cloth. The cloth is solvent extracted and analyzed for pesticides by GC/MS, GC/ECD, or other suitable instrumental method. The original method has been modified and its performance has been compared to other methods (Fortune, 1997). The method has been used most extensively by pesticide registrants.

Surface Press or Mechanical Press. The surface press or mechanical press method is used to estimate transferable residues from hard and soft surfaces. Currently, a sampler based on the design of the EL press sampler (Edwards and Liroy, 1999) is being evaluated in pilot studies by NERL. In general, this method consists of a block shaped device constructed of Delrin polymer and uses C<sub>18</sub> impregnated Teflon extraction disks (3M Empore® disks) as the sample collection media. Other collection media are being considered but have not been fully evaluated at this time. The surface press sampler holds two C<sub>18</sub> disks while providing a specific contact area (114 cm<sup>2</sup>) and contact pressure (~1200 Pa). Once the disks are loaded into the sampler the sampler is placed on the surface to be monitored and allowed to remain in contact for a specified period of time. The C<sub>18</sub> disks are then solvent extracted and analyzed for pesticides by GC/MS, GC/ECD, or other suitable instrumental method.

Surface Wipe. Surface wipe methods are used to estimate the surface pesticide residue loading on hard surfaces such as floors, furniture, window sills, counters, toys, and other surfaces and objects a child may contact. Surface areas being sampled must be non-porous and relatively smooth in texture. There are several methods that have been developed but all are similar in that a material, generally cotton gauze or some filter material, often wetted with a solvent (isopropanol or water), is used to wipe a specified surface area (Wright et al., 1993; Camann et al., 1996; Liroy et al., 1998; Lu and Fenske, 1999). The collection material is then extracted and analyzed for pesticides by GC/MS, GC/ECD, or other suitable instrumental method.

Selection of practical, representative surfaces for monitoring can be one of the most

difficult issues associated with the collection of environmental samples for the estimation of children's dermal exposures. Financial and physical resources generally limit the number of samples that can be collected and analyzed at any one location making those that are collected critical to the success of the study. Children contact a wide variety of surfaces during their activities and, as previously discussed, the level of activity, surface type, and the number of surface contacts are all important variables. While the study design and field sampling protocols can define the appropriate criteria, the general surface types, and the general sample collection locations, the ultimate decision for sample collection is the responsibility of the field sampling personnel and relies to a great extent on their experience and training. Each home or daycare center situation requires decisions to be made that are specific to that particular home or daycare center and the activities of the children in those locations. The following are some general guidelines for determining appropriate sampling locations. The area and surfaces sampled should be:

- representative of where the child spends the majority of his/her time while awake,
- representative of the surfaces that the child frequently comes in contact with,
- amenable to the designated methods of sampling, and
- surfaces for which empirically-derived transfer coefficients are available.

Information pertaining to the first two guidelines is generally obtained through discussions with the child's caretaker and/or by observation of the child's activities. Questionnaires can also be developed to provide a systematic approach to defining the areas and surfaces appropriate for sampling.

The preceding paragraphs provided a brief overview of the generally available methods for measuring pesticide residue surface loadings. In NERL's children's exposure measurement studies, the most commonly used methods to measure surface pesticide residue loadings are surface wipes and the C<sub>18</sub> surface press sampler. The following is a more detailed discussion of these methods.

The surface wipe method used in the recent NERL children's exposure studies uses 4-in x 4-in, 6-ply, cotton dressing sponges wetted with pesticide grade isopropanol as the collection media. The surface to be sampled is wiped with an isopropanol dampened dressing sponge in one direction while frequently exposing a fresh surface of the wipe. The surface is then wiped in a perpendicular direction with the same wipe. Once this is complete, the first wipe is placed in a storage container and a second wipe is prepared and the process is repeated with this second wipe. The second wipe is added to the container holding the first wipe. Samples are stored frozen (-20 °C) and analyzed by solvent extraction followed by GC/ECD, GC/MS, or other appropriate analytical method.

A total sampled area of 930 cm<sup>2</sup>, representing an area with dimensions of 30.5 x 30.5 cm (12 x 12 inches), is generally sampled when collecting samples from a flat surface. Other areas

may also be used to accommodate spaces available. The area sampled for irregular shaped surfaces may be measured and determined on an individual basis. Smaller surfaces may be used if high concentrations are suspected or if there is limited area available for sampling. The actual surface area sampled must be determined and recorded. This method should not be used on surfaces that may be damaged by alcohol.

The surface press method used in recent NERL children's exposure studies consists of a specially constructed sampling device and utilizes  $C_{18}$  extraction disks (3M Empore) as the transfer/collection medium. The sampler is based on the design of the sampler described by Edwards and Lioy (1999). Two 90 mm,  $C_{18}$  extraction disks are mounted in a specially constructed sampling device constructed of Delrin polymer and having a total mass of 1340 g. The two disks provide a net surface contact area of 114 cm<sup>2</sup> and a contact pressure of 11.8 g/cm<sup>2</sup>. The disks are secured in the surface press sampler by means of a clamping system and during use are placed in direct contact with the surface being tested. The press sampler is left in contact with the surface for a prescribed period (generally 2 or 5 minutes) after which time it is lifted from the surface and the disks carefully removed, folded, and placed in a pre-cleaned and labeled, glass storage container. Samples are stored frozen (-20 °C) and analyzed by solvent extraction followed by GC/ECD, GC/MS, or other appropriate analytical method. Testing is ongoing in NERL to finalize the procedures for use of this method for a wide range of current use pesticides, particularly for the pyrethroids.

It should be recognized that selection of the extraction and analysis methods for use with these sampling media is critical to successful measurements of surface loading. Because of the low surface loadings that may be encountered in residences and daycares, the extraction method must have high recovery of the target compounds and the method detection limits must be sufficiently low. Levels of pesticides measured in households are generally very low. In a recent study, transferable residue concentrations measured from carpet with a PUF roller were in the range of 0.03 to 0.61 µg/m<sup>2</sup> in a room with a recent application of diazinon (Lewis et al., 2001). There are many factors that ultimately guide the selection of sampling and analysis methods. Among them are the sensitivity of the analytical instrument that will be used for the analysis of the extract, the type of analytical detector, the final volume to which the sample extract is concentrated, loading or concentration of the target compounds on the original sampled surface, and the transfer efficiency of the target compounds from the surface to the sampling media. All of the above parameters can be optimized by conducting pilot or scoping studies to field test methods for the pesticides of interest before a large field study is conducted. Although previously collected samples can initially be screened and the extracts adjusted by concentrating or diluting specific samples to insure that they fall within the analytical range, this is not advised due to the additional handling and potential associated errors as well as the inefficiency of multiple analyses of the same sample. In general, liquid injections of extracts on a GC/MS system will be measurable in the low pg/µL (ng/mL) range while operating in the selective ion mode and, therefore, an injected sample must be in this range to be measurable. If an area of 930 cm<sup>2</sup> is wiped using the surface wipe method, the sample extract is concentrated to a final volume

of 1.00 mL, and the transfer efficiency is near 100%, the initial surface concentration must be approximately 11 pg/cm<sup>2</sup> (0.11 µg/m<sup>2</sup>) in order to be measurable. Increasing the sampled surface area, concentrating the extract to a smaller volume, or optimizing the instrument are all viable steps to decreasing the detection limit. It should be noted that simply increasing the size of the sampled area may not be sufficient to increase the measurable pesticides since detection of the pesticides may also be affected by interferences associated with the sampled area. Precision for the surface press and surface wipe samples should be ± 25% for duplicate samples and the accuracy, expressed as the percent recovery of spiked samples, should be in the range of 75 to 125%.

## **6.5 Exposure Factor/Questionnaire Information**

The numerous data parameters that need to be considered when estimating dermal exposure were described above. Exposure factors are used in conjunction with the environmental measurement data to estimate exposure. Collection of exposure factor information is often accomplished through the use of activity diaries and questionnaires. Questionnaires break the day into discrete time periods of interest. Activity diaries can provide information as a function of time over the entire day. At a minimum, the diaries and questionnaires must include a notation of the time period of interest, indoor versus outdoor activities during this time period, clothing levels, and microenvironment/macroactivity information. Table 6-3 shows the microenvironments/macroactivity combinations and surfaces for which information is collected. An example of a time-activity diary that can be used to collect the exposure factor information required when assessing dermal exposure using the macroactivity approach and information for estimating inhalation exposure was depicted in Figure 5-1 in Section 5.5.

## **6.6 Estimation of Transfer Coefficients**

The transfer coefficient for each microenvironment/macroactivity must be empirically determined from controlled laboratory experiments or field studies. In laboratory studies, experiments can be designed to develop transfer coefficients for a variety of micro-environment/macroactivity combinations. In field studies, transfer coefficients are determined for a discrete period of time for a single microenvironment/macroactivity combination. For children, it is necessary to record activity patterns (i.e., contact activities, activity level, amount of clothing, locations), collect dermal wipes (i.e., based on exposed skin and activity information), and measure transferable surface residue loadings (i.e., from the location where the child spends the majority of his/her time). As an alternative to the dermal wipes, cotton dosimeters may be worn in a specific microenvironment/macroactivity for a defined period of time. These collected parameters can then be substituted into the equation to calculate the transfer coefficient.

The transfer coefficient,  $TC_{me/ma}$ , provides a measure of dermal exposure resulting from



Table 6-3. Microenvironments/Macroactivity Combinations and Surfaces for Which Activity Data Are Collected

Microenvironment	Macroactivity			
	Active Play	Quiet Play	Sleeping	Eating
<b>Indoors at Home</b>				
-Carpet				
-Hard Floor				
-Upholstery/Bedding				
<b>Outdoors at Home</b>				
-Grass				
-Dirt/Soil				
-Paved Surfaces				
<b>Indoors at Daycare</b>				
-Carpet				
-Hard Floor				
-Upholstery/Bedding				
<b>Outdoors at Daycare</b>				
-Grass				
-Dirt/Soil				
-Paved Surfaces				

contact with a contaminated microenvironmental surface while engaged in a specific macroactivity. The transfer coefficient takes into account the fraction of the surface residue that is transferred from a surface to skin, the character of the microenvironmental surface that is contacted, and the area of the microenvironmental surface that is contacted during a time increment for a given activity.  $TC_{der}$  can be defined as follows:

$$TC_{me/ma} = (E_{dme/ma}) / [(C_{surf})(AD_{me/ma})] \quad (13)$$

where

$TC_{me/ma}$	=	transfer coefficient for the microenvironment/macroactivity ( $\text{cm}^2/\text{h}$ )
$E_{dme/ma}$	=	dermal exposure for a given microenvironment/macroactivity combination over a 24-h period ( $\mu\text{g}/\text{d}$ )
$C_{surf}$	=	surface loading (total or transferable) measured in the microenvironment ( $\mu\text{g}/\text{cm}^2$ )
$AD_{me/ma}$	=	activity duration that represents the time spent in each micro-environment/macroactivity combination with a specific clothing pattern for the child that would affect the surface area available for transfer over a 24-h period ( $\text{h}/\text{d}$ )

The transfer coefficient relates to the specific type of pesticide residue loading measured. For example, if a transferable pesticide residue loading is measured, then the transfer coefficient is related to the transferable residue. However, if a total pesticide residue loading is measured, then the transfer coefficient must be related to the total residue. It is important to keep this distinction in mind when performing environmental measurements. The same sampling methods must be used both for the field surface loading measurements and in the experiments to determine the transfer coefficients for the same microenvironment/macroactivity combination.

## **7.0 APPROACH FOR ESTIMATING DIETARY INGESTION EXPOSURE**

### **7.1 Introduction**

Total ingestion of pesticides by children from foods and beverages involves two major components: (1) directly ingested foods brought into the home or other eating places containing pesticide residues primarily from agricultural applications (i.e., dietary ingestion exposure, or simply dietary exposure), and (2) indirect ingestion exposures associated with additional contamination of foods during consumption by children. This section focuses on the first component and is termed dietary exposure because it is associated with pesticides that are inherent to the foods themselves, and not how they may be further contaminated by the activities of the child during consumption in the residential or other eating environment. Approaches for estimating indirect ingestion from foods, as well as other pathways of indirect ingestion exposure not associated with diet, are included in Section 8.0, Approach for Estimating Indirect Ingestion Exposure.

For certain exposure scenarios (e.g., chronic exposures) and in the absence of recent pesticide applications, the dietary component of ingestion exposure is likely the dominant exposure pathway to pesticides, and potentially the most significant of all pathways for aggregate exposure. Such scenarios occur in particular when handling of foods by the child is minimal and when additional pathways of contamination, such as from surface deposits of residues resulting from outdoor air, track-in of particles, and/or indoor sources of pesticides, are minimal. Then, agricultural sources are typically the most significant sources of dietary, and hence aggregate exposure.

Personal monitoring methodology for measuring dietary exposure of children is based on the duplicate diet method for collection of food samples from study subjects with subsequent measurement of pesticide residues in the collected food samples. These procedures provide the ability to measure the importance of diet relative to other pathways of personal exposure and they directly measure, with reasonable certainty, the exposure from all foods and beverages in the diets presented to the child during the monitoring period. Children's diets differ significantly from those of adults and children eat more than adults relative to their body weights. The diet of newborns is limited exclusively to breast milk or formula, both of which may expose infants to significant concentrations of environmental contaminants (Mukerjee, 1998 and Chance et al., 1998). Infants and young children eat more fruit and milk products in proportion to their body size and have a less varied diet than adults. In addition, there may be tremendous variability in diet among young children of similar ages and for a single child at different periods in time. Some infants and toddlers go through phases where only a few preferred foods are eaten for weeks and months at a time. Such a limited diet may potentially increase dietary exposure of young children to environmental contaminants such as pesticide residues in fruit (NAS, 1993 and Goldman, 1995). The numerous factors that influence the diets of young children and resulting health implications make it extremely important to accurately assess their dietary intake of

pesticides.

## 7.2 Summary of Data Requirements

Dietary exposure to a pesticide is defined as the amount of pesticide ingested in the foods and beverages consumed by a child over some reference exposure period, exclusive of any additional contamination occurring during the actual process of eating the foods, as discussed above. Thus, exposure to pesticides in drinking water may also be included in estimating dietary exposure. Depending on dietary collection objectives and economic considerations, foods may be collected and analyzed as independent items or collections of items that are combined for analysis. A summary of the data requirements for this approach is presented in Table 7-1.

Table 7-1. Data Requirements for Estimating Dietary Ingestion Exposure

Parameter	Measurement	How Collected	Units
$E_f = \sum C_f W_f$			
f	food item or collection of items		
$C_f$	concentration of contaminant in food f	Analyze food item or items	$\mu\text{g/kg}$
$W_f$	weight of food f	Weigh food item or items	$\text{kg/d}$

## 7.3 General Considerations

Dietary samples are specific to the subject being monitored and are typically collected over intervals of one day, although other sample collection intervals may be used. Dietary samples can be collected for several consecutive days, several non-consecutive days, or at widely spaced times over weeks, months, or seasons. In most studies, the caregiver of the child will prepare a duplicate plate by measuring or estimating duplicate portions of each food given to the child. The portions are identical to those served to the child in all aspects of preparation, type of food, and amount of food. Following the eating activity by the child, portions are adjusted to account for foods not consumed, thus providing a duplicate diet sample. The distinction between a duplicate plate and a duplicate diet is typically more significant for children than adults because significant quantities of food may be left uneaten by the child. To accurately measure dietary exposure, samples must be collected for all foods and beverages consumed, including those obtained away from home at restaurants, schools, day cares, etc. The caregiver will often be

responsible for recording a diary of the type and amount of each food the child consumes. Supplemental questions are answered by the caregiver that allow evaluation of both the completeness and representativeness of the collected sample, and hygiene and dietary habits of the child. Duplicate diet samples are often separated into solid foods and beverages during the monitoring period because of analytical and economical considerations. Drinking water can be included in the beverage samples or collected separately. Both samples (i.e., solids and beverages) are composited to create two samples for the monitoring period. This allows for an exposure estimate for the dietary pathway that is equivalent to other pathways being measured. In some studies, individual foods may be collected. This allows subsequent compositing in the laboratory which may allow for improved detection limits and/or selection of foods that are more likely to contain pesticide residues.

Sample collection containers should be sufficiently large for the largest sample that might be collected or multiple containers must be provided. Container materials should neither add the contaminant of interest (or analytical interferences) to the food sample nor cause losses of the contaminant of interest from the collected foods. Containers should be made of materials that will withstand transport, handling, and in some cases, freezing. Storage and transport of samples are normally important parameters to prevent sample spoilage during collection. Samples should be stored and shipped refrigerated or frozen.

#### **7.4 Monitoring Method**

To monitor dietary ingestion exposure of children, the duplicate diet methodology is used. This is a well established method that has been used in several monitoring studies (Berry, 1997, Thomas et al., 1997, and Melnyk et al., 1997). Since children are the targeted subjects, caregivers are trained to collect and record information on duplicates of foods consumed using visual estimation procedures. Specifics of the sample collection are determined in the study design prior to field activities. NERL has developed a dietary model and database system (DEPM, Tomerlin et al., 1997) to assist in designing dietary monitoring studies and interpreting the results of composite diet sampling. The DEPM includes both consumption and pesticide residue data from existing notional monitoring programs. Consumption information for children is included. DEPM uses historical food information to estimate total daily intake of pesticides, and food groups and items that are major contributing factors to dietary exposure. DEPM is available at <http://www.epa.gov/nerlcwww/depm.htm>.

Typically, analyzing composited food samples is an integral part of determining dietary exposure. All methods for collection, storage, mixing, extraction, and analysis should be selected and implemented in such a way that they provide residue concentration data that are of known and acceptable quality for meeting the overall study objectives. Composited food samples present unique analytical challenges because the samples are very diverse and may contain varying amounts of fats and other interfering substances. In most cases, relatively few of the individual food items in a composited sample contain residues. While these residues may or

may not be present at high levels in the individual food item, the compositing process dilutes them by combining the contaminated item or items with a large mass of food which does not contain any residues. Thus, sensitivity of the analytical method for foods is extremely important.

Diet is often the limiting pathway in multi-pathway assessments due to the relative sensitivity of food analytical methods versus those for other media. A typical diet (even for a child) may result in collection of 100 to 500 g/d for composite solid food or beverage samples. Typically, 25 g is the maximum practical aliquot for analysis. This limitation allows for the analysis of only 5 to 25% of the total daily solid food or beverage sample. Analysis of other media is not subject to such sample size limitations and it is possible to analyze the entire sample collected in a one day period, and in some cases, even longer. Even with equivalent analytical sensitivity among media, overall method detection limits are 10 - 100 times higher for food than for other media due to relative amount of the daily sample analyzed.

Analytical methods to obtain pesticide concentrations for composited food samples have been developed with the goal of 1 ng/g detection limit. Specific detection limits for several pesticides in daily composited solid food samples are listed in Table 7-2 with the recoveries from medium fat composite diet samples (U.S. EPA, 2001).

## **7.5 Exposure Factor Information**

Activities affecting dietary exposure are recorded in food diaries and supporting questionnaires. This information includes what foods were eaten, where the foods were eaten, and how much was eaten. Supporting questionnaires obtain other important information such as whether the diet was typical, activities that occurred that may influence exposure like recent application of pesticides, etc. The NERL Hygiene and Dietary Habit Survey and food diary are presented in Appendix B.

Table 7-2. Method Detection Limits and Pesticide Recoveries<sup>a</sup> from Medium Fat Composite Diet Samples Fortified at 1, 5 and 10 ng/g.

Pesticide	%Recovery			MDL <sup>b</sup>
	1 ng/g	5 ng/g	10 ng/g	
Trifluralin	93	104	108	0.4 (1)
Phorate	80	94	113	1.7 (5)
Hexachlorobenzene	68	82	92	0.4 (1)
Dicloran	41	86	115	1.7 (5)
Simazine	56	86	85	1.9 (5)
Atrazine	101	100	106	2 (5)
Terbufos	80	96	113	0.3 (1)
Fonofos	99	101	105	1.5 (5)
Diazinon	101	102	106	1.4 (5)
Chlorothalonil	77	75	124	0.5 (1)
Acetochlor	184	95	110	1.4 (5)
Alachlor	106	88	104	0.4 (1)
Aldrin	108	84	99	1 (5)
Malathion	109	106	115	1.7 (5)
Metolachlor	78	92	105	0.2 (1)
Chlorpyrifos	97	97	102	0.5 (1)
Parathion	101	119	117	1.8 (5)
Dacthal	91	94	105	0.3 (1)
Isofenphos	80	99	111	0.4 (1)
g-Chlordane	62	81	95	0.2 (1)
Endosulfan-I	96	82	96	0.4 (1)
a-Chlordane	69	82	97	0.2 (1)
Dieldrin	115	92	99	0.4 (1)
DDE	80	94	102	0.2 (1)
Endrin	0	89	73	2.4 (10)
DDD	111	145	183	5.3 (10)
cis-Permethrin	175	120	105	3.5 (10)
trans-Permethrin	175	134	102	3.2 (10)

<sup>a</sup> n = 7 for each concentration.

<sup>b</sup> Method detection limits were calculated using either 1, 5 or 10 ng/g fortification levels as indicated in parentheses. Method detection limits are not reported for analytes with recoveries less than 60%. (Rosenblum et al., 2001)

## **8.0 APPROACH FOR ESTIMATING INDIRECT INGESTION EXPOSURE**

### **8.1 Introduction**

Ingestion exposure by indirect pathways has been identified as a potentially significant route of pesticide exposure for infants and young children (Cohen Hubal et al., 2000). Indirect ingestion exposures occur when children place objects that have become contaminated with pesticides, including their hands, in their mouths. Pesticides are ingested as a result of transfer from the object, or, for foods, when they are consumed.

In addition to the dietary ingestion exposures associated with the foods that children eat (Section 7.0), the manner in which children handle food as they eat may also impact their exposure to environmental contaminants. Small children are less likely than adults to consume food in a structured environment. Small children may sit on the floor or lawn to eat and often pick up and eat foods that have fallen on the floor. Infants and young children also eat most of their food with their hands. Increased exposure occurs when children handle and eat foods that have come in contact with the floor or other contaminated residential surfaces (Melnik et al., 2000; Akland et al., 2000) and hands. Indirect ingestion associated with foods consumed, together with dietary ingestion associated with contaminants inherent to foods, constitutes total ingestion from the dietary pathway.

Children's mouthing behaviors also contribute to the potential for indirect ingestion resulting from contact with contaminated objects and surfaces in the environment. Sucking and mouthing hands and objects are natural behaviors in childhood development. Infants are born with a sucking reflex, providing them with both nutrition and a sense of comfort or security. If infants do not receive unrestricted breast feeding, they will suck on a pacifier, thumb (or other finger), or other object like a blanket or stuffed animal. As infants develop, they begin to explore their world through mouthing (Groot, 1998). During this stage of development, children put almost everything that they contact into their mouths for a few seconds. Young children may also begin to use the mouth as a third hand, placing some objects in the mouth in order to manage them.

Teething is another important stimulus for mouthing activities. Biting and chewing on fingers and objects to relieve the discomfort of teething may be extensive. Teething usually begins between 4 and 7 months of age, but may start several months earlier or later. As with all childhood behaviors, mouthing activities vary significantly from child to child and, therefore, the impact on exposure will also be highly variable.

### **8.2 Summary of Data Requirements**

Because it would be too burdensome and costly to collect all the data required to apply the microactivity approach as presented in Section 3.4, a macroactivity approach is presented



here to provide a simplified assessment of indirect ingestion exposure to an individual based on measurement data collected in the field. In this approach, objects (including hands and 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 concentration measurements. 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 obtained in the laboratory experiments is then required to complete the assessment. A summary of the data requirements for this approach is presented in Table 8-1.

Table 8-1. Data Requirements for Estimating Indirect Ingestion Exposure

Parameter	Measurement	How Collected	Units
Indirect Ingestion Exposure - Macroactivity Approach $E_{\text{ing/mi}} = (C_x)(TE_x)(SA_x)(EF)$			
x	Hand, object, food item or anything else that enters the mouth		
$C_x$	Contaminant loading on x	Wipes, washes or surface samplers, samples of handled food	$\mu\text{g}/\text{cm}^2$
$TE_x$	Transfer efficiency of contaminant from x to mouth	Measured in the laboratory, estimated from the literature	unitless
$SA_x$	Area of x contacted by mouth	Questionnaire	$\text{cm}^2/\text{event}$
EF	Frequency of indirect ingestion events over a 24-h period	Questionnaire	event/d

### 8.3 General Considerations

Ingestion exposure occurs by direct ingestion of foods containing pesticide residues. These residues are the result of agricultural use of pesticides and are in the food when the food is brought into the residential environment. Ingestion exposure may also occur by indirect ingestion of residues on objects, hands, and food that are placed in the mouth. These residues are transferred from surfaces and objects in the residential environment directly to hands, to food, or additionally from hands to food. Indirect ingestion exposures are difficult to quantify and assess because there are no methods for directly measuring contaminants that are ingested by these

pathways.

Instead, measurement of residues on hands, objects, and foods collected at specific time points are used to estimate ingestion exposures over the time frame of interest. Measurements of contaminant loading collected at a single point in time, however, may not reflect changes in loading which occur prior to, or subsequent to, sampling (e.g., evaporation or removal by hand washing or mouthing). Contaminant loading over time can vary significantly and is often the result of discrete events. Thus, current sampling techniques result in an integrated loading over extended time periods, and variations in time cannot be characterized. To facilitate interpretation of these data, measurements of residues on hands, objects, and handled foods need to be related to activities that occur in the same time interval. As much as is possible, exposure media concentrations need to be linked directly to contact activities.

To conduct a more detailed analysis of the time course of exposure, as is the goal of an exposure model like SHEDS, very detailed time-sequence activity data are required. To collect this type of data, a diary survey structure designed to collect sequential location/activity data for each discrete behavior of interest or videotaping is required. The burden of such a diary survey precludes inclusion in the type of children's exposure study covered by this protocol and should therefore be considered for a separate study.

One additional consideration: in the time frame of concern for this scenario (short term following an application), exposure resulting from ingestion of soil and house dust is assumed to be less important than indirect ingestion of residues. If during the preliminary screening assessment, soil and dust ingestion pathways are identified as potentially important, these can be easily addressed by the addition of dust and soil samples to the required field measurements.

#### **8.4 Monitoring Methods**

To estimate the contaminant loading on the objects which a child may contact by indirect ingestion pathways, the following field samples are required:

- Samples of residue loadings from any residential surfaces that are frequently mouthed (e.g., surface press on coffee table),
- Samples of residue loadings from the surfaces of objects that are frequently mouthed (e.g., surface wipe on toy),
- Samples of residue loadings from the child's hands or other body parts that are frequently mouthed (e.g., hand wipe),
- Samples of foods that have been handled in the child's normal eating environment (e.g., cheese handled by child prior to eating), and
- Samples of foods that have contacted surfaces during eating (e.g., cheese that has been placed on counter tops, floors, or high chair trays).

Information on the relevant surface sampling techniques has been included in Section 6 on dermal exposure. Removal techniques used to measure residues on smaller objects, hands, and foods are discussed below.

Surface Loading Measurements. Surface wipe, press, or rolling methods are used to estimate the total or potential amounts of pesticides available for transfer from surfaces such as floors, furniture, window sills, counters, and toys. There are several methods that have been developed, as described in Section 6.4, but all are similar in that a material, generally cotton gauze or some filter material, either wetted with a solvent (isopropanol, water, saliva simulant) or dry, is used to wipe, press, or roll on a specified surface area. Appropriate methods need to be selected based on the type of surface (e.g., wipes are applicable for hard surfaces, but not for carpet). The collection material is then extracted, and analyzed for pesticides by GC/MS, GC/ECD, or other suitable instrumental method.

Hand Wipe or Rinse. Hand wipe and rinse methods are used to estimate the total or potential amounts of pesticides available for mouthing. Handwash sampling procedures can be standardized to ensure that they are operator-independent (Davies, 1980). Skin wiping procedures are inherently operator dependent. Removal sampling techniques are limited in that these measure only what can be removed from skin at the time of sampling rather than the actual skin loading. Findings suggest that data collected using removal techniques are difficult to interpret and require appropriate laboratory removal efficiency studies for use. However, because hand wipes are being used here to assess indirect ingestion exposure and not dermal exposure, the removal is likely more representative of what is available for indirect ingestion than for dermal absorption. As such the major limitation is that current sampling techniques do not reflect loadings or losses which occur subsequent to sampling. NERL is addressing issues related to hand wipe sampling and is developing and evaluating standardized procedures.

Collection of Handled Food. Individual samples of foods that have been handled by the child can be collected to estimate the pesticide loading on the food items caused by contact with contaminated surfaces and hands. One approach used by NERL has been to identify food items that a child in the study was known to handle when eating. During the monitoring period, the caregiver for the child collects one set of the individual food items that were not touched by the child and a second set that were touched (handled) by the child. Analyses of the two sets of the individual food items provide the amount of pesticides transferred onto the foods. In other studies, samples of foods have been contacted with surfaces by the monitoring technician to directly measure the potential for transfer.

Soil and Dust Sampling. Although not included as part of this scenario, soil and dust samples may be collected if it is determined that those pathways are potentially important for indirect ingestion.

Selection of Objects for Sampling. The objects to be sampled should be:

- representative of the objects that the child frequently comes in contact with and
- amenable to the designated methods of sampling.

Information pertaining to these two points is generally obtained through discussions with the child's caregiver and/or by observation of the child's activities. Questionnaires can also be developed to provide a systematic approach to defining the objects appropriate for sampling. All samples should be collected such that the measurements can be related in time to the activity data in the questionnaires.

Sampling Considerations. Many of the sampling considerations for the indirect ingestion route of exposure are similar to those presented for the dermal route. However, there are several considerations specific to this route and the residue removal and food samples that must be collected. Outlined below are the additional issues that should be considered when designing and collecting samples associated with the indirect ingestion route in a measurement-based assessment.

- Physical surface characteristics, contaminant surface loading, sampling material, and wipe sampling procedures all influence accuracy and precision of measurements. (Fenske, 1993)
- Area dimensions of objects to be monitored should be based on a practical limit of detection (LOD) for the analytical method that will be used. Samples being analyzed by a laboratory method that is very sensitive (low LODs) will require a smaller sampled area than samples being analyzed by a method that is less sensitive (high LODs). Detection limits should be sufficiently low to insure that not detected values represent concentrations considered insignificant as defined by the study goals.
- Practical limits of detection should also be considered in choosing the solvent for the removal techniques. While a saliva simulant is likely to give a sample that is more representative of a transferable residue, isopropanol may provide a sample that is easier to analyze. Requirements for method detection limits and performance were discussed in Section 6.4.
- Hand wipe and hand wash techniques assess contamination adhering to an individual's skin at the time of sample collection. Measurements of skin loading do not reflect losses which occur subsequent to sampling; e.g. evaporation or removal by hand washing. Skin loading over time may vary significantly and may be the result of many discrete events. Current sampling integrates dermal loading over extended time periods. Therefore, variations in time cannot be characterized.
- Food items should be collected that are of sufficient quantity such that there will be leftovers for collection, both handled and not handled. Alternatively, use standardized food items that can be contacted with contaminated surfaces so that sources of contamination can be identified, both within and among exposure scenarios.

## 8.5 Exposure Factor Information

To estimate indirect ingestion exposure, information must be collected to describe the (1) characteristics of the child, (2) activity information associated with mouthing and ingestion of objects by the child, and (3) information on the transfer of residues from the objects to the mouth.

Characteristics of Child. The minimum information required to characterize the child is the child's age, weight, gender, and hand surface area. These variables are used to estimate ingestion rates and may be used to estimate mouthing and related behavior.

Activity information. An activity questionnaire is used to collect information on: (1) the objects that are mouthed or eaten most often by a child and (2) the characteristics of the activities that potentially result in indirect ingestion of the contaminant of interest. By collecting information on the important objects, field sampling can be directed to collect residue loading samples from these items. Information on mouthing characteristics (e.g., frequency, surface area mouthed), hand washing practices, eating environment, and the likelihood of the child handling food items should be linked to the sampled items to facilitate assessment of indirect ingestion exposure. Examples of the information to be collected and the types of questions required to collect the relevant activity information are presented in Figures 8-1 and 8-2. Figure 8-2 presents examples of questions from the NERL Hygiene and Dietary Habit Survey, a copy of which is included in its entirety in Appendix B of this document.

Object-to-Mouth Transfer Efficiencies. Object-to-mouth transfer efficiency may be a function of object surface characteristics (e.g., plush vs hard), and mouthing mechanics (e.g., sucking vs licking). The need to develop residue transfer data for mouthing activities was identified in the NERL Dermal and Non-dietary Exposure Workshop conducted in 1999. Laboratory studies are being conducted by NERL using a surrogate mouthing method to identify the important parameters for characterizing these transfer efficiencies and to develop a set of transfer efficiency data.

Soil and dust ingestion rates. Indirect ingestion of soil and dust has been monitored in fecal samples using tracer elements (Binder et al., 1986; Calabrese et al., 1989; Van Wijnen et al., 1990; Davis et al., 1990; Calabrese et al., 1997). These studies require collection of dietary data and concentrations of contaminants in residential soil and dust to link the tracers to ingested soil and then to estimate ingestion of contaminants. Results of the limited monitoring conducted using this technique are currently used to provide bounding estimates for soil ingestion which can then be combined with information on the concentration of pesticides in dust and soil to estimate indirect ingestion exposure by this pathway.

Figure 8-1. Examples of data required for assessing indirect ingestion exposure and sample questions.

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HOME QUESTIONNAIRE

1. Hand surface area
2. Is your child currently teething?
3. Does your child, put toys or other objects in his/her mouth?
  1. Frequently (greater than 10 time/hour)
  2. Sometimes (2 to 10 times/hour)
  3. Almost Never (less than 2 times/hour)

4. Please list the objects your child puts in his/her mouth most frequently

Object	Portion put in mouth	Number of times/day	Where handled
1.			
2.			
3.			
4.			
5.			

5. Does you child lick or mouth surfaces?
  1. Frequently (greater than 10 time/hour)
  2. Sometimes (2 to 10 times/hour)
  3. Almost Never (less than 2 times/hour)
6. What surfaces does you child lick or mouth most frequently? Please list surface and location
  - a.
  - b.
  - c.

7. How frequently does you child put his/her hands in his/her mouth?

	During active play?	During quiet play?
Frequently (greater than 20 time/hour)		
Sometimes (5 to 15 times/hour)		
Occasionally (2 to 5 times/hour)		
Almost Never (less than 2 times/hour)		

8. How much of your child's hand does s/he put into his/her mouth?

- a. thumb
- b. 2 fingers
- c. 4 fingers
- d. whole hand

9. Does your child suck on his/her fingers when they are in the mouth?

10. Does you child put, his/her toes or feet in his/her mouth?

- a. Frequently (greater than 10 time/hour)
- b. Sometimes (2 to 10 times/hour)
- c. Almost Never (less than 2 times/hour)

DAYCARE QUESTIONNAIRE (Ask for each participating child:)

1. Does s/he child, put toys or other objects in his/her mouth?

1. Frequently (greater than 10 time/hour)
2. Sometimes (2 to 10 times/hour)
3. Almost Never (less than 2 times/hour)

2. How frequently does s/he put his/her hands in his/her mouth?

1. Frequently (greater than 20 times/hour)
2. Sometimes (5 to 15 times/hour)
3. Occasionally (2 to 5 times/hour)
4. Almost never (less than 2 times/hour)

3. Please lists the 5 toys or objects that children put in their mouths most frequently and where they are handled.

4. How are toys washed and recycled?

Figure 8-2. Examples of questions included in the NERL Hygiene and Dietary Habit Survey (included in Appendix B).

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1. Does your child eat food with his/her fingers? What types?

1. Yes	1. Often	_____
2. No	2. Sometimes	_____
9. Unknown	3. Almost never	_____
	9. Unknown	_____
  
  2. Where does your child usually eat his/her meals when at home?

1. Kitchen	1. At table
2. Living room	2. High chair
3. Bedroom	3. Chair or couch
4. Dining room	4. Sitting on the floor
5. Other _____	5. Other _____
9. Unknown	9. Unknown
  
  3. Where does your child usually eat his/her snacks?

1. Kitchen	1. At table
2. Living room	2. High chair
3. Bedroom	3. Chair or couch
4. Dining room	4. Sitting on the floor
5. Other _____	5. Other _____
9. Unknown	9. Unknown
  
  4. What snacks does your child usually eat at home?

1. _____
2. _____
3. _____
4. _____
5. _____
  
  5. How frequently does your child eat food off of the floor?

1. Often	2. Sometimes	3. Almost never
9. Unknown		
-



- 
6. Does your child ever prepare or get his/her own food?  
(for instance peel a banana, get a bowl of cereal, finger foods)
- |            |                 |             |
|------------|-----------------|-------------|
|            |                 | What foods? |
| 1. Yes     | 1. Often        | _____       |
| 2. No      | 2. Sometimes    | _____       |
| 9. Unknown | 3. Almost never | _____       |
|            | 9. Unknown      | _____       |

7. Does an older brother or sister ever prepare or get your child's food?  
(For instance peel a banana, get a bowl of cereal, finger foods)
- |                   |                 |             |
|-------------------|-----------------|-------------|
|                   |                 | What foods? |
| 1. Yes            | 1. Often        | _____       |
| 2. No             | 2. Sometimes    | _____       |
| 9. Not applicable | 3. Almost never | _____       |
|                   | 9. Unknown      | _____       |

8. Does your child ever eat food after it has dropped on the floor?
- |                   |                 |             |
|-------------------|-----------------|-------------|
|                   |                 | What foods? |
| 1. Yes            | 1. Often        | _____       |
| 2. No             | 2. Sometimes    | _____       |
| 9. Not applicable | 3. Almost never | _____       |
|                   | 9. Unknown      | _____       |

9. Does your child drink from bottles?
- |                   |                 |       |
|-------------------|-----------------|-------|
|                   |                 |       |
| 1. Yes            | 1. Often        | _____ |
| 2. No             | 2. Sometimes    | _____ |
| 9. Not applicable | 3. Almost never | _____ |
|                   | 9. Unknown      | _____ |
-

## **9.0 OTHER DATA COLLECTION**

### **9.1. Questionnaire Data To Identify Sources and Usage of Pesticides in Residences and Daycares**

#### **9.1.1 Introduction**

The previous sections of this document have described the data requirements and approaches for estimating children's aggregate exposure to pesticides. For the measurement based approach described in the previous sections, time-activity diaries and questionnaires are used to collect data on the exposure factors that are needed to estimate exposure by the different routes and pathways. Well-designed questionnaires are also important to characterize sources, transport processes, and parameters that may affect spatial and temporal distribution of pesticides and environmental contaminants in human exposure studies. Information collected with questionnaires during exposure measurement field studies can aid in the interpretation of the data collected in measurement assessments and provide data for use in modeling assessments.

Exposures to pesticides and environmental contaminants may result from many different sources and in many different microenvironments. Pesticide sources may include, but are not limited, to applications for the control of agricultural pests, outdoor turf and landscape pests, termite control, indoor pest control, and control of pests on pets. Pesticides may move or translocate from their source and point of application. They move from one location to another following several pathways. Pesticide applications may generate particles that drift from their original source. In addition, depending on the physical nature of the pesticide active ingredient and the formulation, vapors and/or pollutants sorbed to particles may result in pesticides or contaminants moving from the source to deposit at other locations. Finally, the physical uptake of residues and particles on an individual's hands, feet, or clothing or by adhesion on the fur of pets such as cats and dogs may result in the physical translocation of contaminants or pesticides. Pesticides and environmental contaminants in the air may also infiltrate into the homes, daycares, schools and other buildings from the outdoors.

#### **9.1.2 Administering Questionnaires**

Site surveys and questionnaires are common methods to screen and characterize sources, and aid in identifying transport mechanisms and exposure pathways. Furthermore, they can be useful to gather general information regarding the study participants and their lifestyles and activities, the home or facility under study, and other parameters that may affect exposure or the interpretation of exposure measurement results.

Prior to the initiation of the study, the research team should concisely define the type of information required to fulfill their research needs and evaluate the design to insure that while effectively capturing the desired information an excessive burden is not placed on the study

participants.

Surveys may be completed by field scientists or adult study participants. The survey team should be available to address questions the participant may have regarding the survey. When administering survey questionnaires that collect information on pesticide use, it is especially important to have knowledgeable field team members who can provide assistance to study participants to complete the questionnaires. Assistance may be particularly important when querying the participants regarding specific products and pesticide applications because many occupants of homes have little familiarity with specific terms, chemical classes, or product groupings. To overcome some of the problems of obtaining accurate information about pesticide use in residences, the field team may request to view areas where cleaners, pesticides and other household products are stored in order to collect chemical names and registration numbers for future identification. The problem of collecting accurate usage information may be even greater when attempting to collect information in daycares or schools where pesticides are applied by commercial applicators. In these environments, it is generally necessary to work with building management and facility managers to obtain service and application records.

Following completion of questionnaires, a member of the survey team should review the forms to insure completeness and legibility. Similarly, forms completed by the field team should undergo a quality assurance check by the team lead to determine completeness, accuracy and legibility.

#### **9.1.3. Information on Sources to be Collected in Pesticide Exposure Measurement Studies**

To obtain accurate information on pesticide sources and usage in residences, a simple questionnaire should be designed that collects information on what pesticides were used, when they were applied, where applied, and how applied. Figure 9-1 presents examples of questions that can be used to collect this information. The example questions would be used to address the specific scenario described in Section 4 for short-term exposure during a period of one to seven days following application. The sample questions are not all inclusive. Additional questions will be required to address different exposure scenarios. A different set of questions would be required to assess long-term exposure to pesticides. Additional questions would also be required for population modeling-based approaches for exposure assessment. Researchers in NERL are currently developing a questionnaire that will be used with this protocol.

In addition to the questions that specifically address recent pesticide applications in the home, questions should be included to determine other potential sources of pesticides in the home. Pesticide residues may be physically transported on the clothing, shoes and the body of individuals from their workplace to home. Questions to determine occupational exposure as a source might include those in Figure 9-2.

Figure 9-1. Example questions use to collect information on pesticide usage in a residence.

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1. Did anyone apply pesticides within your home, in your garden or in your yard within the past 2 weeks?  
☐ Yes. (If yes, go to 1.b. through 1.h)  
☐ No  
☐ Don't Know
  - 1.b. Where and when was it applied?  
☐ in the home (if checked, screen gives detailed list of choices for kitchen, pantry, living room, bathrooms, bedrooms, under sinks, floors, at baseboards, cupboards, window sills, at a specific site of infestation, etc.)  
☐ in the basement  
☐ in the garage  
☐ in storage areas  
☐ outside along the walls  
☐ under the crawl space  
☐ in the yard  
☐ in the garden  
☐ on the pet (s)  
☐ on a deck or wood surface  
☐ on cement or patio surface  
☐ Don't know  
  
(next screen gives choices of when applied)  
☐ today ☐ within 24-48 hours ☐ 3-5 days ☐ 1 week ☐ 2 weeks ☐ last month
  - 1.c. For indoor applications, how was it applied?  
☐ crack and crevice spray (liquid) along walls  
☐ sprayed (liquid) in the room  
☐ fogger (aerosol in a can)  
☐ dust  
☐ bait in a container  
☐ bait not in a container  
☐ applied to pet as a liquid or shampoo  
☐ applied to pet as a powder
-

---

1.d. For non-indoor living area applications, including crawl space, how was it applied?

- ☐ foundation spray (liquid) along walls  
☐ dust or pellets on yard, garden or lawn  
☐ spray on yard, garden or lawn  
☐ bait in a container  
☐ bait not in a container  
☐ applied to pet as a liquid, shampoo, or powder  
☐ don't know

1.e Who applied the pesticide?

- ☐ applied by yourself  
☐ applied by another adult in the home  
☐ applied by a commercial applicator  
☐ don't know

1.f. For what purpose was the pesticide applied?

- ☐ ants      ☐ mosquitos      ☐ fungi/molds/bacteria      ☐ weed control  
☐ roaches      ☐ other flying insects      ☐ plant disease      ☐ other purpose  
☐ fleas      ☐ termites      ☐ other insects      ☐ don't know

1.g. Did you have to mix the chemical with water before applying?

- ☐ yes      ☐ no

1.h. Give the name and EPA number (if known) of the products that were applied during the past 2 weeks. The EPA registration number is located on the label of the product. (Photo to show example)

Pesticide #1: \_\_\_\_\_ EPA Reg. No: \_\_\_\_\_

Pesticide #2: \_\_\_\_\_ EPA Reg. No: \_\_\_\_\_

Pesticide #3: \_\_\_\_\_ EPA Reg. No: \_\_\_\_\_

Pesticide #4: \_\_\_\_\_ EPA Reg. No: \_\_\_\_\_

Pesticide #5: \_\_\_\_\_ EPA Reg. No: \_\_\_\_\_

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Figure 9-2. Questions on occupational exposure to pesticides.

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2. Does anyone in the home work in a manufacturing job that involves handling of pesticides or who works in a facility where pesticides are produced or handled?

\_\_\_ Yes \_\_\_ No

2a. If yes, what pesticides?

Pesticide #1: \_\_\_\_\_

Pesticide #2: \_\_\_\_\_

Pesticide #3: \_\_\_\_\_

2b. If yes, are his/her clothes and shoes changed before leaving the facility?

\_\_\_ Yes \_\_\_ No

3. Does anyone in the home work in on a farm or in an agricultural job that involves handling of pesticides or crops treated with pesticides? \_\_\_ Yes \_\_\_ No

3a. If yes, what pesticides?

Pesticide #1: \_\_\_\_\_

Pesticide #2: \_\_\_\_\_

Pesticide #3: \_\_\_\_\_

Pesticide #4: \_\_\_\_\_

Pesticide #5: \_\_\_\_\_

3b. If yes, are his/her clothes and shoes changed before entering your home?

\_\_\_ Yes \_\_\_ No

---

Additional questions with greater detail may be added for field studies involving measurements of pesticide by children of agricultural farm worker's families. These questions may include identification of local sources of agricultural pest control applications, proximity to residences and daycares, direction from these sources, and questions related to potential spray drift.

#### **9.1.4 Information on Microenvironment Surfaces, the Structure and the Occupants**

In order to obtain accurate estimates of dermal exposure, it is important to collect information on the surfaces that the child contacts in the residence or daycare. This information will be used to determine the appropriate transfer coefficients and efficiencies to develop in laboratory and field experiments and to use in the algorithms for estimating dermal and indirect ingestion exposure. During field data collection, the type of flooring material should be identified in each room where the child spends time. This may include hard surfaces such as vinyl flooring, ceramic flooring, wood, and other materials. Carpet type (short nap, plush, etc.) should also be recorded. This information should be recorded for all rooms that are occupied by the child. Development of the questions to collect this information is on-going in NERL.

For most field exposure measurement studies, the information on the structure can be limited to a simple diagram of the residence showing the locations of rooms in the structure and the sampling locations. Detailed information on the construction materials, size, age, heating and cooling systems, etc. are not required for the purpose of estimating exposure, although they may be useful for understanding the measurement data. Similarly, detailed information on occupant activities beyond that collected with the time-activity diaries described in the previous sections is generally not required, but it may be useful for interpreting the measurement data.

The type of information to be collected on the structure and the occupant activities will be determined by the study objectives. Superfluous information should not be collected as it increases participant burden and resources for performing the field studies. If the study data analysis plan does not include a purpose for collection of information about the structure or the occupants, it should not be collected.

#### **9.1.5 Additional Data Collection for SHEDS-Pesticide Model**

Additional data are needed for the SHEDS-Pesticides model (described in Appendix A) that are not required in exposure assessments that use the individual measurement-based approach for which this protocol was developed. Some of those additional requirements for pesticide usage include the following:

1. Number of Applications
2. Month First Applied
3. Time Interval Between Applications

4. Day of Week Used
5. Reentry Interval
6. Scenario-Specific Area (e.g., Lawn, Garden, House)
7. Scenario-Specific Area with a Chemical
8. Scenario-Specific Area with the Chemical of Interest
9. Scenario-Specific Area with the Chemical of Interest via the Formulation
10. Scenario-Specific Area with the Chemical of Interest via the Formulation and Application Method
11. Residences Treating Entire (vs. Spot Treatment of) Scenario-Specific Area with the Chemical of Interest via the Formulation and Application Method
12. Number of Applications Treating Entire (or Spot Treated) Scenario-Specific Area with the Chemical of Interest via the Formulation and Application Method
13. Area Treated for Total Area Application
14. Fraction of Total Area for Spot Treatment
15. Application Rate



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## **APPENDIX A**

### **Description of the ORD/NERL Stochastic Human Exposure and Dose Simulation Model for Pesticides (SHEDS-Pesticides)**

The Stochastic Human Exposure and Dose Simulation model for pesticides (SHEDS-Pesticides) developed by ORD/NERL is a probabilistic, physically-based model that simulates aggregate exposures for population cohorts and multi-media pollutants of interest. SHEDS simulates individuals from the user-specified population cohort by selecting daily sequential time/location/activity diaries from surveys contained in EPA's Consolidated Human Activity Database (e.g., the National Human Activity Pattern Survey). Depending on the type of pesticide usage information entered, SHEDS can be used to simulate one day post-application exposures from a single application event or daily, weekly, monthly, seasonal, or annual average exposures from repeated pesticide applications over a year. It can also yield results for user residences only or for the entire population of both user and non-user residences.

Exposure time profiles are the basis of the SHEDS exposure calculations. These are plots of instantaneous exposure (mass, concentration, or mass loading at the external human boundary) against time that preserve within-day peaks and variability as an individual moves throughout his or her day. These exposure profiles can yield toxicologically relevant dose profiles, and ultimately, improved risk estimates. They are constructed separately for each of the four exposure routes included in SHEDS -- inhalation, dietary ingestion, dermal contact, and non-dietary ingestion (from hand-to-mouth and object-to-mouth pathways)-- and the time step is the duration of the CHAD diary location-activity combinations. To generate a daily inhalation exposure profile, SHEDS samples from indoor or outdoor air concentration distributions corresponding to locations occupied by the sampled individual's diary. The air concentrations are then combined with sampled values from activity-specific energy expenditure distributions and basal metabolic rates for the diary-reported activities. Dermal exposure is modeled by combining dermal transfer coefficient information with surface residues and time spent at and near the applied surfaces. For bathing related locations and activities, a washing removal efficiency is applied to the profile to simulate the reduced dermal loading. Non-dietary ingestion exposure from hand-to-mouth and object-to-mouth transfer is simulated by combining dermal hand loading or object residues with fraction of hands or objects inserted into the mouth, frequency of mouthing activities, and saliva removal efficiency. Non-dietary ingestion via hand-to-mouth contact is subtracted from the dermal exposure profiles. The dietary module in SHEDS uses the latest USDA/EPA recipe files and 1994-1996, 1998 Continuing Survey of Food Intakes by Individuals (CSFII) consumption data, which includes about 10,000 food types and 21,660 person-days. CHAD individuals are matched with CSFII individuals, and for each CSFII person, the reported consumption data are combined with sampled residue values in foods as eaten to yield a modeled mass of residue ingested by meal. To obtain residue values in foods as eaten, SHEDS applies the recipe files to the CSFII food types to break the food into raw agricultural commodities (RACs), and then combines the RAC residues with use and processing factors.

To simulate one day post-application exposures for a population cohort, SHEDS samples a single diary and combines the sequential location-activity durations with sampled values from user-specified probability distributions for environmental media concentrations (either calculated from user-specified application rates or sampled from user-specified distributions of measured



values) and exposure factors (e.g., saliva and washing removal efficiency, skin surface area contacted, surface area of objects mouthed) into route-specific algorithms described above to construct daily exposure time profiles. These exposure profiles can be combined with pharmacokinetic models to yield route-specific dose profiles that can then be aggregated.

To simulate exposures from repeated applications over a year, SHEDS-Pesticides simulates, for each individual in an age-gender cohort, 365 days by sampling 8 CHAD diaries representing 1 person from each of 4 seasons and 1 person from each of 2 day categories (weekend and weekday); fixing 5 weekday dairies and 2 weekend diaries; and then repeating the 7 day activity patterns within each season. It then sets days and times of pesticide applications over the year based on user-specified probabilities for pesticide usage. Based on these application times, environmental media residues and concentrations, either calculated from application rates or sampled from user-specified distributions of measured values, are set every day of the year for that individual. SHEDS then combines activities and residues with sampled values from probability distributions for exposure factors to generate longitudinal 1-year exposure profiles that can be entered as inputs to pharmacokinetic models to simulate the corresponding route-specific dose profiles. Once the dose profiles are obtained, they can be summed across routes to yield an individual's aggregate dose profile for the chemical of interest.

Once the exposure and dose profiles are generated for each individual, the metrics of interest (e.g., peak, time-averaged, time-integrated) are extracted from the individual's profiles, and the process is repeated thousands of times to obtain population distributions. This approach allows identification of the relative importance of routes, pathways, and model inputs. Sensitivity analyses are conducted using stepwise regression and correlation methods to identify the relative importance of routes and model inputs. If the user enters uncertainty distributions associated with model inputs, SHEDS applies two-stage Monte-Carlo simulation to derive estimates of inter-individual variability in the population and uncertainty in estimated empirical exposure and dose distributions.

## **APPENDIX B**

### **Examples of a Food Diary and a Hygiene and Dietary Habit Survey Used in Recent NERL Pilot Studies**

## **HOW TO COLLECT FOODS AND BEVERAGES**

### **WHERE WE WANT YOU TO COLLECT FOOD**

- 1) Please collect only foods and beverages that are eaten at home, or are prepared at home but are eaten elsewhere.

### **WHAT WE WANT YOU TO COLLECT**

- 1) Please prepare and collect a second portion (as close as possible to the exact amount) of every food or beverage your child eats at every meal, snack, or any other time on the collection days.
- 2) This does not include vitamins, medicines, chewing gum, toothpaste, or any other non-edible item.
- 3) Please collect a sample of any foods that your child has dropped on the floor. You should collect only those foods from the floor that your child is likely to eat after they have dropped on the floor.
- 4) Please have your child eat the same foods he/she would have eaten if we were not here.

### **WHEN WE WANT YOU TO COLLECT THE FOOD**

- 1) Please collect the foods and beverages eaten from midnight to midnight on \_\_\_\_\_

### **HOW TO COLLECT THE DUPLICATE-DIET SAMPLE**

- 1) At every meal or snack, prepare a second plate with the same type and amounts of food you have prepared for your child. Include all spices, sauces, butter, salt, ketchup, etc. Prepare a second cup, glass, or other container with the same amount of beverage that the child will drink. If you can, please use the same kind of plates, cups, and glasses for the food collection as used for the meal.
- 2) If you give your child more servings of food or beverage during the meal, add the same amount to the second plate, cup, or glass. Use more plates, cups, or glasses if necessary.
- 3) At the end of the child's meal, remove from the second plate, cup or glass the same amount of food as was left on your child's plate, glass or cup. If you are able, remove any inedible portions, like bones or pits, from foods on the second plate. The food or beverage that is now on the second plate or in the second cup should be the same amount that your child ate or drank.
- 5) We have given you four zip-lock bags: one marked breakfast, one marked lunch, one marked dinner, and one marked snacks. Transfer the food on the second plate (not your

child's plate) to a zip-lock bag for the meal that was just eaten. Seal the bag. Place the bag in your refrigerator if it contains foods that could spoil.

- 6) Add all beverages from the second cup (not your child's cup) to the plastic bottle. Frozen items that could melt, like ice cream or popsicles, should also be put into the plastic bottle with the beverage samples not with the food samples.
- 7) Close the jar lid and put the jar in your refrigerator.

## **HOW TO USE THE 24-HOUR FOOD DIARY**

### **INSTRUCTIONS**

- (1) We want you to list all of the foods, beverages, or drinking water that your child eats or drinks from midnight to midnight.
- (2) Every time your child eats, write down the name of the meal (breakfast, lunch, dinner, snack).
- (3) Then write down on a separate line the name of every food or beverage that your child eats or drinks.
- (4) For food mixtures such as stews or potpies, please write down the major kinds of foods in the mixture. Use the lines immediately below the one on which the name of the mixture is entered.
- (5) For beverages (including water), write down how many cups or glasses that your child drink(s).
- (6) When we collect the food samples, we will ask you several questions about each food that your child ate. These will include:
  - (a) In the last month, how often did your child eat this food each week?
  - (b) Where was the food that you collected eaten?
  - (c) Did any of the food eaten have contact with your child's hands, the floor, or other surfaces?
  - (d) Were foods cooked in or prepared with tap water?
  - (e) Were beverages cooked in or prepared with tap water?

# FOOD DIARY

START DATE:		TIME:		FOR INTERVIEWER USE ONLY						
END DATE:		TIME:								
Meal	PLEASE LIST ALL FOODS, BEVERAGES, THAT YOUR CHILD EAT(S) OR DRINK(S) AND HOW MANY OF EACH ITEM	How Many	Portion Size	Frequency Eaten	Where Eaten	Contact With			Food	Beverages
						Fingers	Floors	Other Surfaces	Tap Water	Tap Water
Lunch	EXAMPLE: CHEESEBURGER	1								
	EXAMPLE: SALAD WITH LETTUCE AND TOMATOES	1								
	EXAMPLE: WATER	2 glasses								

# FOOD DIARY - SUPPLEMENTARY QUESTIONS

COMPLETE ON SAME DAYS FOOD IS RECORDED IN DIARY AND SAMPLES COLLECTED	DAY: DATE:	1 / /	2 / /	3 / /
1. Please think back, were there any foods or beverages that you could not or did not collect for use: (LIST IDENTITY, SOURCE, AND AMOUNT OF EACH MISSING FOOD AND THE DAY IT WAS NOT COLLECTED.)				
a. At Breakfast _____ _____ _____		Y N	Y N	Y N
_____		Y N	Y N	Y N
b. At Lunch _____ _____				
_____		Y N	Y N	Y N
_____				
c. At Dinner _____		Y N	Y N	Y N
_____				
_____				
d. For Snacks _____				
_____				
_____				

COMPLETE ON SAME DAYS FOOD IS RECORDED IN DIARY AND SAMPLES COLLECTED	DAY: DATE:	1 / /	2 / /	3 / /
2a. Did (your child), for any reason, eat more or less food than usual? (READ CHOICES AND ENTER a OR b). a. More food than usual → GO TO 2b. b. Less food than usual → GO TO 2b. c. Same as usual → GO TO 3.				
2b. Because of: (READ CHOICES AND CIRCLE ALL THAT APPLY.)  a. Travel or vacation ..... b. Weight control diet ..... c. Illness or medical condition ..... d. Work or school schedule ..... e. Entertainment or social occasion ..... f. Because of the food collection study ..... g. Other (Specify day): ..... .....	a b c d e f g	a b c d e f g	a b c d e f g	
3a. Did (your child), for any reason, eat different foods than (your/his/her) usual diet? (CIRCLE Y FOR YES AND N FOR NO.) .....	Y N	Y N	Y N	
3b. If yes, was that because: (READ CHOICES AND CIRCLE ALL THAT APPLY.)  a. Travel or vacation ..... b. Weight control diet ..... c. Illness or medical condition ..... d. Work or school schedule ..... e. Entertainment or social occasion ..... f. Because of the food collection study ..... g. Other (Specify day): ..... .....	a b c d e f g	a b c d e f g	a b c d e f g	

COMPLETE ON SAME DAYS FOOD IS RECORDED IN DIARY AND SAMPLES COLLECTED	DAY: DATE:	1 / /	2 / /	3 / /
4a. List all of the floor foods collected.				
4b. Where were floor foods collected from?				



## Hygiene and Dietary Habit Survey

Parent's name: \_\_\_\_\_ Child's name: \_\_\_\_\_

How old is your child? \_\_\_\_\_ DOB: \_\_\_\_\_

How much does your child weigh (use a scale ?) \_\_\_\_\_

We want to ask some questions about \_\_\_\_\_ (name of child). We are interested in the foods your child eats, and how the food is stored and prepared. We want to find out if these things influence your child's lead exposure.

### Part 1. We now have some questions about your child's eating habits.

1. Does your child eat food with his/her fingers? What types?

- |            |                 |       |
|------------|-----------------|-------|
| 1. Yes     | 1. Often        | _____ |
| 2. No      | 2. Sometimes    | _____ |
| 9. Unknown | 3. Almost never | _____ |
|            | 9. Unknown      | _____ |

2. Where does your child usually eat his/her meals when at home?

- |                |                         |
|----------------|-------------------------|
| 1. Kitchen     | 1. At table             |
| 2. Living room | 2. High chair           |
| 3. Bedroom     | 3. Chair or couch       |
| 4. Dining room | 4. Sitting on the floor |
| 5. Other _____ | 5. Other _____          |
| 9. Unknown     | 9. Unknown              |

3. Where does your child usually eat his/her snacks?

- |                |                         |
|----------------|-------------------------|
| 1. Kitchen     | 1. At table             |
| 2. Living room | 2. High chair           |
| 3. Bedroom     | 3. Chair or couch       |
| 4. Dining room | 4. Sitting on the floor |
| 5. Other _____ | 5. Other _____          |
| 9. Unknown     | 9. Unknown              |

4. What snacks does your child usually eat at home?

1. \_\_\_\_\_
2. \_\_\_\_\_
3. \_\_\_\_\_
4. \_\_\_\_\_
5. \_\_\_\_\_

5. How frequently does your child eat food off of the floor?

- |            |              |                 |
|------------|--------------|-----------------|
| 1. Often   | 2. Sometimes | 3. Almost never |
| 9. Unknown |              |                 |

6. Does your child ever prepare or get his/her own food?  
(for instance peel a banana, get a bowl of cereal, finger foods)

1. Yes
2. No
9. Unknown

1. Often
2. Sometimes
3. Almost never
9. Unknown

What foods?

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7. Does an older brother or sister ever prepare or get your child's food?  
(For instance peel a banana, get a bowl of cereal, finger foods)

1. Yes
2. No
9. Not applicable

1. Often
2. Sometimes
3. Almost never
9. Unknown

What foods?

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8. Does your child ever eat food after it has dropped on the floor?

1. Yes
2. No
9. Not applicable

1. Often
2. Sometimes
3. Almost never
9. Unknown

What foods?

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9. Does your child drink from bottles?

- |                   |                 |       |
|-------------------|-----------------|-------|
| 1. Yes            | 1. Often        | _____ |
| 2. No             | 2. Sometimes    | _____ |
| 9. Not applicable | 3. Almost never | _____ |
|                   | 9. Unknown      | _____ |

10. Do you have a cat in the home?

- |        |   |            |
|--------|---|------------|
| 1. Yes | a. Does your child play with it before meals? | 1. Yes     |
| 2. No  |   | 2. No      |
|        |   | 9. Unknown |
|        | b. Does your child play with it during meals? | 1. Yes     |
|        |   | 2. No      |
|        |   | 9. Unknown |

11. Do you have a dog in the home?

- |        |   |            |
|--------|---|------------|
| 1. Yes | a. Does your child play with it before meals? | 1. Yes     |
| 2. No  |   | 2. No      |
|        |   | 9. Unknown |
|        | b. Does your child play with it during meals? | 1. Yes     |
|        |   | 2. No      |
|        |   | 9. Unknown |

12. Has your child eaten outside in the past 3 months?

- |            |                           |                                  |
|------------|---------------------------|----------------------------------|
| 1. Yes     | how often?                | where? (all that apply)          |
| 2. No      | 1. >3 times/week          | 1. backyard at home              |
| 9. Unknown | 2. about 1/week(go to 13) | 2. yard at friend's or neighbors |
|            | 3. <1/week (go to 13)     | 3. playground or park            |
|            |                           | 4. vacant lot or field           |
|            |                           | 5. alley                         |
|            |                           | 6. street                        |
|            |                           | 7. other places _____            |

b. If yes, what sort of eating surface

type of ground

- |                   |                        |
|-------------------|------------------------|
| 1. table          | 1. grass               |
| 2. bench or chair | 2. concrete or asphalt |
| 3. steps          | 3. dirt or soil        |
| 4. on the ground  | 4. sandbox             |
| 5. sandbox        | 5. other _____         |

- 6. stroller
- 7. other \_\_\_\_\_
- 9. unknown

9. unknown

13. a. How many times does your child wash his/her hands each day? \_\_\_\_\_

b. When does your child wash his/her hands? (check all that apply)

- 1. before meals
- 2. after meals
- 3. before snacks
- 4. after snacks
- 5. after going to the bathroom
- 6. before going to bed
- 7. after coming in doors
- 8. other \_\_\_\_\_

Part 2. We would now like you to tell us about some of your activities.

14. Where do you keep:
- |                       |                       |
|-----------------------|-----------------------|
| a. fresh fruits       | b. fresh vegetables   |
| 1. on counter/table   | 1. on counter/table   |
| 2. in cabinet         | 2. in cabinet         |
| 3. in refrigerator    | 3. in refrigerator    |
| 4. other _____        | 4. other _____        |
| 5. don't usually have | 5. don't usually have |

15. In what containers do you store

<u>Raw fruits?</u>	<u>Covered?</u>
<u>Raw vegetables?</u>	<u>Covered?</u>

16. In what containers do you store

<u>Cereals?</u>	<u>Covered?</u>
<u>Pastas?</u>	<u>Covered?</u>

17. In what containers do you store meats? Covered?

\_\_\_\_\_  
\_\_\_\_\_

18. Where do you prepare foods? (check all that apply)

- |                    |     |    |
|--------------------|-----|----|
| 1. kitchen counter | yes | no |
| 2. kitchen table   | yes | no |
| 3. kitchen sink    | yes | no |
| 4. chopping board  | yes | no |
| 5. other _____     |     |    |

19. Do you wash your hands before preparing the food?

- |        |              |
|--------|--------------|
| 1. yes | 1. always    |
| 2. no  | 2. usually   |
|        | 3. sometimes |
|        | 4. seldom    |

20. Do you wash your hands before serving the food?

- |        |              |
|--------|--------------|
| 1. yes | 1. always    |
| 2. no  | 2. usually   |
|        | 3. sometimes |
|        | 4. seldom    |

21. Do you wash the food preparation surface....

- |                            |                           |
|----------------------------|---------------------------|
| a. before food preparation | b. after food preparation |
| 1. yes                     | 1. yes                    |
| 2. no                      | 2. no                     |

22. How do you wash plates and glasses?

1. by hand
2. dishwasher
3. use throw aways/paper plates
4. other \_\_\_\_\_

23. How do you dry plates and glasses?

1. air dry
2. cloth towel
3. paper towel
4. dishwasher
5. other \_\_\_\_\_

24. What type of cookware (pots and pans) do you use? (check all that apply)

1. stainless
2. aluminum
3. cast iron
4. glass
5. ceramic/pottery
6. plastic
7. other \_\_\_\_\_

25. How do you wash cookware and utensils?

1. by hand
2. dishwasher
3. other \_\_\_\_\_

26. How do you dry cookware and utensils?

1. air dry
2. cloth (dish) towel
3. paper towel
4. dishwasher
5. other \_\_\_\_\_

27. If you use cloth dish towels -----

How often are they washed

1. as needed
2. more than once a week
3. once a week
4. less than once a week
9. don't know

Are they also used as

1. hand towels
2. face towels
3. to dry counters
4. other \_\_\_\_\_
9. don't know

Answer by Observation

1. Cleanliness of house

1. clean
3. dirty

2. somewhat clean

2. Cleanliness of child

1. clean
3. dirty

2. somewhat clean

3. Does the child fist his/her food when handling/eating?

1. yes

2. no

Thank you for your help. Do you have any questions at this time?