THE EPA NATIONAL EXPOSURE RESEARCH LABORATORY CHILDREN'S PESTICIDE EXPOSURE MEASUREMENT PROGRAM

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ABSTRACT

The U.S. Environmental Protection Agency (EPA) National Exposure Research Laboratory (NERL) is performing research in support of the Food Quality Protection Act (FQPA) of 1996. This act requires that pesticide exposure assessments to be conducted for all potential sources, routes and pathways, not just dietary intake. The goal of the NERL program is to develop and evaluate protocols and methods for assessing children's aggregate and cumulative exposures to pesticides, and to collect data required to reduce the reliance on default assumptions in development of quantitative exposure assessments. This paper provides an overview of the framework and approach for the research program and descriptions of studies being performed to fill data gaps in the following areas:

- Spatial and temporal distribution of pesticide residues indoors,
- Pesticide use patterns indoors,
- Dermal exposure,
- Indirect ingestion exposure,
- Microenvironments and macroactivity patterns of children, and
- Children's exposure measurements protocols and methods.

INDEX TERMS

Pesticides, Children, Human Exposure, Food Quality Protection Act

INTRODUCTION AND BACKGROUND

The U.S. EPA has pledged to increase its efforts to provide a safe and healthy environment for children by ensuring that all EPA regulations, standards, policies, and risk assessments take into account special childhood vulnerabilities to environmental toxicants. Children's exposures to environmental contaminants are expected to be different and, in many cases, much higher than older individuals due to differences in behavioral patterns and physiological functions (Bearer, 1995). Children's activities and the way that they interact with their environment may have a profound effect on the magnitude of their chemical exposures. They may crawl, roll, and lie on contaminated surfaces. Children's mouthing activities (hand-to-mouth and object-to-mouth) may result in indirect ingestion of chemicals if the hands or objects are contaminated. 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 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, 2000). 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. The NERL children's exposure research program is designed to address these objectives.

The ORD is conducting 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.

A comprehensive approach is required to understand and adequately address all of the components of aggregate and cumulative exposure assessments. To develop NERL's research strategy and approach, factors influencing children's exposure to environmental contaminants were reviewed and the quality and quantity of available data associated with default assumptions for exposure factors were evaluated (Cohen Hubal et al., 2000a). A framework to systematically identify the important sources, routes, and pathways for exposure was developed (Cohen Hubal et al., 2000b). 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 researchers to systematically identify the most critical research needs and data gaps associated with children's exposures to pesticides.

A conceptual model of children's residential exposure to pesticides was developed that was the initial focal point for the children's pesticide exposure research strategy and development of measurement studies and protocols (Cohen Hubal et al., 2000b). NERL researchers used the model to identify important routes and pathways of exposure and to identify four priority research areas that included the following:

- Pesticide use patterns what, where, and how are pesticides used in children's microenvironments.
- Spatial and temporal distribution of pesticides in residential dwellings,
- Dermal and indirect (non-dietary) ingestion need to develop measurement approaches (microactivity or macroactivity approaches), and
- Dietary exposure assessments, to include indirect ingestion due to the handling of food by children.

STUDIES TO ADDRESS PRIORITY RESEARCH AREAS

To address these research needs, targeted studies, described below, were designed. They included laboratory studies, small pilot field studies, and two large studies in which the EPA collaborated with other Federal agencies, as described below.

Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP)

The CTEPP study is the largest children's exposure study undertaken to date. It examines the aggregate exposures of 257 preschool children between the ages of 18 months and 5 years to pollutants commonly found in their everyday environments. The major objectives of this three-vear pilot study were to quantify children's aggregate exposures, apportion the exposure pathways, identify the important exposure media, and formulate the important hypotheses for future testing. Participants were recruited from daycare centers and from the general population in twelve urban and rural counties in North Carolina and Ohio using a random digit dialing (RDD) method. Monitoring was performed during 48-hour sampling periods at both daycare centers and homes. Samples collected include food, beverages, drinking water, indoor air, outdoor air, hand wipes, indoor floor dust, play area soil, transferable pesticide residues, floor wipes, food preparation surface wipes, and urine. The samples are being analyzed for over 40 persistent pesticides and organic pollutants, including herbicides (atrazine), insecticides (organophosphorus, carbamate, pyrethrin and organochlorine), phthalate esters (butylbenzyl, di-n-butyl), phenols, polychlorinated biphenyls, and polycyclic aromatic hydrocarbons. Sample analysis will be completed and preliminary results will be reported this year.

Study to Identify Important Parameters for Characterizing Pesticide Residue Transfer Efficiencies

The objective of this laboratory study is to evaluate parameters that affect pesticide residue transfer from surface-to-skin, skin-to-objects, skin-to-mouth, and object-to-mouth. The approach is to use fluorescent tracers as surrogates for pesticide residues. Transfers of riboflavin, the tracer in the initial tests, were compared to transfers of chlorpyrifos and bioallethrin. Following application of the tracers to realistic surfaces (e.g., carpet, laminate), controlled transfer experiments were conducted to evaluate parameters that included the surface type, surface loading, type of contact (press versus smudge), contact duration, contact pressure, and skin condition (dry, moist, or sticky). The study involved tests to evaluate repetitive contacts with contaminated surfaces, measurements of transfers off the skin, and simulated mouthing removal using saliva moistened PUF. Surface loading, skin condition, and surface type were significant parameters in the initial tests. In the second phase of this study, additional tracers and pesticides with a range of physiochemical properties will be evaluated. Controlled transfer experiments will be performed to refine the understanding of significant parameters and to develop a more comprehensive set of transfer efficiency data that can be used to predict dermal and indirect ingestion exposure from field measurements.

Feasibility of Using the Macroactivity Approach to Assess Dermal Exposure

Two main approaches are currently used to assess dermal exposure. In the macroactivity approach, exposure is estimated for each macroactivity that the child conducts within each microenvironment (e.g. quiet play in the living room, active play in the kitchen). With this approach, 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. With this approach, exposure must be estimated for all contacts made by child during a 24-hour period. The objective of this study is to test the feasibility of the macroactivity approach to assess children's dermal exposure to pesticides in daycare centers. Screening measurements were performed in nine daycares where pesticides were applied as crack and crevice treatments by commercial

applicators. At one daycare, children were asked to wear full-body cotton dosimeters for short time periods while involved in selected macroactivities (e.g., storytime, playtime indoors). Surface sampling of pesticide residues and videotaping of activities were performed simultaneously. Sample analysis is ongoing. Results will be used to calculate transfer coefficients and to evaluate this approach for estimating dermal exposure.

Post-Application Exposure Studies

Researchers in NERL are involved in two post-application studies. In both studies, aggregate exposure measurements were made following a crack and crevice application by a professional applicator. Objectives of these studies include development of dermal transfer coefficients for young children engaged in specific activities and evaluation of the macroactivity approach. In a collaborative study with researchers at the Environmental and Occupational Health Sciences Institute (EOSHI), aggregate exposure measurements were made at nine residences following a professional chlorpyrifos crack and crevice application. Measurements included indoor air, air exchange rates, surface residue wipes, dust wipes, toy wipes, dermal wipes (hands and knees), activity diary, videotaping, cotton dosimeters, and urine. Surface loadings of chlorpyrifos ranged from 0.002 to 0.12 ng/cm² based on wipe measurements and transfer coefficients ranged from 300 to 84,000 cm²/hr. Results of the study showed that pesticide loadings in the cotton dosimeters were related to exposure duration, surface loading, activity level and type of surface, suggesting that the macroactivity approach may be useful for estimating children's dermal exposure. The second postapplication exposure study, being conducted in nine homes in North Carolina, complements the first study. The objectives and methods for this study are similar. This study, however, addresses potential exposures following residential crack and crevice applications of synthetic pyrethroid pesticides, the most commonly used indoor pesticides since the discontinuation of indoor uses of chlorpyrifos and diazinon. This study is ongoing.

First National Environmental Health Survey of Child Care Centers

EPA's objective in this collaborative study with the Department of Housing and Urban Development (HUD) and the Consumer Product Safety Commission (CPSC) is to collect information on pesticide use and pesticide distributions in child care centers. Licensed institutional childcare centers were randomly selected for participation in this national study. The study design used multi-stage sampling with clustering to select approximately 150 childcare centers in 30 primary sampling units in the United States. Samples were collected for pesticides, lead, and allergens at multiple indoor locations in each child care center. Soil samples were also collected near a play area at each center. Pesticide use practices and application information were obtained from the commercial pest control applicators serving the centers. In this study, the EPA will gain a better understanding of pesticide use and young children's (less than 6 years old) potential exposure to pesticides in institutional childcare centers. The field data collection has been completed; sample analysis is ongoing.

Characterizing Children's Pesticide Exposures in Jacksonville, Florida

The Duval County, Florida Health Department with collaboration by the Centers for Disease Control and Prevention (CDC) and the EPA is conducting a research study to measure pesticide exposure in a sample of children in Jacksonville, FL. The objectives of the project are to (1) assess organophosphate (OP) and pyrethroid pesticide exposures in a group of 4-6 year old children from Jacksonville by measuring the urine metabolite levels of OPs and pyrethroids, (2) identify possible sources of these pesticides in homes by performing screening measurements and pesticide inventories, and (3) examine the relationship between

environmental levels of OP and pyrethroid pesticides and biological levels. For this study, the Duval County Health Department (DCHD) collected urine samples from 200 children visiting six public health clinics in Jacksonville, Florida. The CDC will perform analyses of these samples for OP and pyrethroid pesticide metabolites. In a second component of the study, DCHD staff collected environmental screening samples at approximately 25% of these children's homes. The third component of the study, performed by the EPA with assistance from DCHD, involved a detailed aggregate exposure assessment at nine of the study homes. The assessment involved collection of surface wipes, transferable residues, air, duplicate diet, cotton dosimetry samples, and urine samples. A time activity diary of the children's activities was completed. The field study has been completed and sample analysis is ongoing. Results of the pesticide inventory showed that synthetic pyrethroids were the primary pesticides used in the residences in addition to gels and baits containing hydramethylnon and fipronil.

Study to Evaluate the Potential for Human Exposure to Pet-borne Diazinon Residues Following Residential Turf Applications

The objectives of this exposure measurement study are to investigate the potential for indoor/outdoor pet dogs to transport diazinon residues into homes following residential turf application. The study was performed at six homes in North Carolina where a homeowner made a single application of a granular diazinon formulation to turf. Samples were collected at pre-application and at 1, 2, 4, and 8 days post-application. Environmental samples were collected to determine the mass of diazinon applied to the lawn, the movement of residues into the structure, and the spatial and temporal distribution following the application. Residues were measured in samples from the fur, paws and blood of the family dog and from the hands and urine of a child in each home. The study also included videotaping of the child playing with the dog and collection of transferable residues by petting the dog with cotton gloves. The field measurements have been completed; sample analysis is ongoing. In the pre-test for this study, results from one residence showed that diazinon was transferable from treated turf through 15 days and intruded into the dwelling through the air and by track-in. Analysis of fur clippings, and fur and paw wipes from the family's dog suggested that it served as an important medium for the uptake of turf transferable residues.

The Distribution of Chlorpyrifos Following a Crack and Crevice Type Application in the U.S. EPA Indoor Air Quality Test House

The primary objective of this study was to examine the spatial and temporal distributions of the insecticide chlorpyrifos following a crack and crevice application in the EPA Indoor Air Quality Test House located in Cary, NC. Measurements during the 18-day study included air concentrations using both polyurethane foam (PUF) and OVS samplers, surface transferable residues using the PUF roller, surface concentrations (loadings) employing deposition coupons and extractable residues from carpet. Results of the study are in a paper (Stout and Mason, 2002) presented at this conference.

Other EPA Children's Exposure Measurement Studies

In addition to the studies in support of the FQPA described in this paper, there are a number of other recently completed or ongoing measurement studies involving NERL researchers that address children's exposure to environmental contaminants. The National Human Exposure Assessment Survey (NHEXAS) Region 5 study, for example, included the Minnesota Children's Pesticide Exposure Study (MNCPES). The study obtained pesticide (chlorpyrifos, diazinon, malathion, and atrazine) data for 102 children, including measurements of personal exposure (air, hand rinse, duplicate diet), activity patterns (questionnaire, diary, and

videotaping), environmental concentrations (indoor and outdoor air, surface residues, drinking water, soil), and metabolites in urine (Quackenboss et al., 2000; Adgate et al., 2001). The Arizona NHEXAS study also included multi-media measurements of pesticides in households with children (Robertson et al., 1999). Pesticide exposures in young children along the US/Mexico border are being addressed in a number of studies (referred to as the NAFTA border studies) that are described on the U.S.-Mexico Border XXI Environmental Health Workgroup web site (www.epa.gov/orsearth/projects.htm).

A number of other children's exposure studies are being funded, or co-funded, by EPA through cooperative agreements and grants. Information on many of the current studies is available through various EPA web sites, including the Children's Environmental Health and Safety Inventory of Research site (http://oaspub.epa.gov/chehsir/chehsir.page), the ORD National Center for Research (NCER) web site (www.epa.gov/ncer), and the ORD/NERL site (www.epa.gov/ncer)).

U.S. EPA Disclaimer

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