A Pilot Study of Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP)

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Foreword

The mission of the National Exposure Research Laboratory (NERL) is to provide scientific understanding, information and assessment tools that will quantify and reduce the uncertainty in EPA's exposure and risk assessments for environmental stressors. These stressors include chemicals, biologicals, radiation, and changes in climate, land use, and water use. The Laboratory's primary function is to measure, characterize, and predict human and ecological exposure to pollutants. Exposure assessments are integral elements in the risk assessment process used to identify populations and ecological resources at risk. The EPA relies increasingly on the results of quantitative risk assessments to support regulations, particularly of chemicals in the environment. In addition, decisions on research priorities are influenced increasingly by comparative risk assessment analysis. The utility of the risk-based approach, however, depends on accurate exposure information. Thus, the mission of NERL is to enhance the Agency's capability for evaluating exposure of both humans and ecosystems from a holistic perspective.

The National Exposure Research Laboratory focuses on four major research areas: predictive exposure modeling, exposure assessment, monitoring methods, and environmental characterization. Underlying the entire research and technical support program of the NERL is its continuing development of state-of-the-art modeling, monitoring, and quality assurance methods to assure the conduct of defensible exposure assessments with known certainty. The research program supports its traditional clients -- Regional Offices, Regulatory Program Offices, ORD Offices, and Research Committees -- as well as ORD's Core Research Program in the areas of health and ecological exposure analysis and assessment.

Human exposure to multimedia contaminants, including persistent organic pollutants is an area of concern to EPA because of the possible adverse health effects of these compounds. These compounds may originate from industrial processes and combustion and are present in a variety of microenvironments. The efforts described in this report provide an important contribution to our ability to measure and evaluate human exposure to pollutants.

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Abstract

The Pilot Study of Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP) investigated the aggregate exposures of 257 preschool children and their primary adult caregivers to pollutants commonly detected in their everyday environments. The target compounds include organophosphate (OP) pesticides, OP metabolites, organochlorine (OC) pesticides, pyrethroid pesticides and metabolites, acid herbicides, polycyclic aromatic hydrocarbons (PAH), phthalates, phenols, polychlorinated biphenyls (PCB), PAH metabolites, and atrazine. Some of the target compounds are persistent indoors and sometimes outdoors, so that very low levels may exist in the children's surroundings and provide a source of non-acute exposure. The primary purposes of the research were to increase the understanding of children's exposures to persistent and non-persistent organic pollutants, and to gain information on the various activities, environmental media, and pollutant characteristics that may influence children's exposures. The overall objectives were to measure the aggregate exposures of approximately 260 preschool children and their adult caregivers to low levels of a suite of pesticides and other organic pollutants that the children may encounter in their everyday environments and to apportion the routes of exposure and estimate the relative contributions of each route. Within these objectives, four major, specific goals for the CTEPP study were accomplished in this report. These goals were: (1) to measure the concentrations of the target pollutants in multimedia samples collected at the homes and at day care centers of 257 preschool children in six North Carolina (NC) counties and six Ohio (OH) counties, (2) to determine the distributions of child characteristics, activities, and locations that contributed to their exposures, (3) to estimate the aggregate exposures of the preschool children to these pollutants that they may encounter in their everyday environments, and (4) to apportion the routes of exposure. Results will also be used to identify important hypotheses to be tested in future research.

A two-state sampling plan was used to select and recruit study participants. In each state, a total of four urban and two rural counties were randomly selected. The counties were located in three distinct geographical regions of each state. These regions were the mountains, the Piedmont, and the coastal plain of NC, and the northern, central, and southern regions of OH. Dual sampling frames (the day care and the telephone components) were used in each state. To recruit participants in households whose children attended child day care centers, 13 centers in the six NC counties and 16 centers in the six OH counties were selected using probability sampling. Children were then selected randomly from classrooms having children in the eligible age group of two to five years, and their participation was recruited through their parents. To recruit participants in households whose children did not attend child day care centers, list-assisted, random digit dialing telephone sampling in the selected counties was used.

The calculated response rates in NC were 53% for day care centers and 50% for day care parents. In OH these response rates were 57% for OH day care centers and 31% for OH day care parents. The calculated response rate for the telephone sample was 58% in NC and 57% in OH. In NC, children and their caregivers in 130 households participated in the study; in OH, 127 households participated. Approximately half of the children in each state attended child day care centers (63 in NC and 58 in OH). About 84% of the NC participants and 87% of the OH participants lived in urban locations. Low-income households, classified according to federal guidelines for the Women, Infants, and Children (WIC) program (185% of the federal poverty level), comprised 46% of the sampled households in NC and 38% of those in OH.

More than 5,000 discrete personal and environmental samples, including quality control samples, were collected in each state and analyzed. Additionally, house/building characteristics observation surveys, pre- and post-monitoring questionnaires, day care food menus, and detailed child/adult time-activity and food diaries provided ancillary information necessary to estimate aggregate exposures and to aid in interpretation of the CTEPP data.

Field sampling for the day care component took place over a 48-h period at each child's day care center and simultaneously at his/her home. Field sampling for the telephone component took place over a 48-h period at each participant's home. Environmental samples included indoor and outdoor air, outdoor play area soil, indoor floor dust (carpet dust) or if no carpet, hard floor surface wipes, and household/day care drinking water. Personal samples included duplicate diet, hand wipes, and urine. If a pesticide had been applied in the seven days prior to or during sampling, transferable residues, hard floor surface wipes and food preparation surface wipes were also collected. Approximately 10% of the children were videotaped for about 2 h at their homes in OH during sampling to supplement and validate the activity diaries and observations.

All samples, including quality control samples, were extracted, and then analyzed by gas chromatography/mass spectrometry for over 50 target compounds. These compounds included two organophosphorus (OP) pesticides, two OP metabolites, ten organochlorine (OC) pesticides, three pyrethroid pesticides, one pyrethroid metabolite, three acid herbicides, nine polycyclic aromatic hydrocarbons (PAHs), six PAH metabolites, two phthalates, three phenols, 17 polychlorinated biphenyls (PCBs), and atrazine. These compounds, with the exception of atrazine, PAH metabolites and pyrethroid metabolites, were analyzed in the environmental and personal samples. Atrazine was analyzed only in drinking water samples. Only one OP metabolite, 3,5,6-trichloro-2-pyridinol (3,5,6-TCP), was analyzed in the NC environmental and personal samples; both 2-isopropyl-6-methyl-4-pyrimidinol (IMP) and 3,5,6-TCP were measured in the OH samples. In the NC urine samples, two OP metabolites; IMP and 3,5,6-TCP; 2,4-dichlorophenoxyacetic acid (2,4-D), two hydroxy PAHs: 1-hydroxybenz[*a*]anthracene and 3-hydroxychrysene; and pentachlorophenol were analyzed. In the OH urine samples, these same metabolites and/or parent compounds were analyzed, in addition to five hydroxy PAHs (1-hydroxypyrene, 3-hydroxybenz[*a*]anthracene, 3-hydroxybenzo[*a*]pyrene, 6-hydroxychrysene, and 6-hydroxyindeno[1,2,3-*cd*]pyrene) and 3-phenoxybenzoic acid (3-PBA).

Two similarly formatted CTEPP databases were developed, one for the NC study and one for the OH study. Each database contained questionnaire data, analytical data, and metadata, and provided sufficient documentation to allow the data to be understood by a diverse set of users. Descriptive statistics were calculated for sample size, mean, standard deviation, percentage detected, minimum and maximum reported values, and selected percentiles (25^{th}, 50^{th}, 75^{th}, and 95^{th}). The distributions of participant characteristics, activities, and locations that are important for exposure were quantified, based on the questionnaire data. Potential exposures and potential absorbed doses were estimated for selected target compounds, based on the percentage of the samples that had detectable levels of these compounds, the measured concentrations, the participants' activity patterns, and assumed physiological parameters. Statistical analyses to meet the four goals of the study were performed on log-transformed data, using analysis of variance (ANOVA) models. The data summaries presented in this report represent only the children and their primary caregivers in NC and OH who participated in this study.

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Acknowledgments

In a large study such as CTEPP, many people contribute to its success. The CTEPP study was conceptualized by EPA's National Exposure Research Laboratory (NERL) on the basis of three small studies of preschool children's exposures conducted earlier by NERL. NERL staff, with support from Battelle, developed the study design. Battelle performed the recruitment, field sampling, sample analysis, and statistical and data analysis of the two (NC and OH) field exposure studies.

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Executive Summary

The Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP) study is one of the largest aggregate exposure studies of preschool children (i.e., 2 to 5 years of age) performed in the United States. These young children are suspected of having greater exposures to pesticides and other pollutants in their everyday environments compared to older children and adults. These greater exposures may result from what preschool children drink or eat, where they spend their time, and what they do in these locations. The primary goals of this landmark study were:

1. to measure the concentration of chemical pollutants in multimedia samples collected at the homes and day care centers of preschool children,
2. to determine the distribution of child characteristics, activities, and locations that contributed to their exposures,
3. to estimate the aggregate exposures to the pollutants they may come in contact with in their everyday environments, and
4. to evaluate the contribution of each route of exposure.

This report presents the results of statistical analyses conducted to address these primary study goals. Data analysis will continue over the next year to more fully characterize those factors that are responsible for preschool children's exposure and to evaluate the relationship between environmental concentrations, exposure factors, and biomarkers of exposure. The entire CTEPP study database will be made available to scientists in EPA program and regional offices, to researchers in industry and academia, and to the general public to allow the data to be used in additional analysis, as input to exposure models, and in developing risk assessments for preschool children.

The CTEPP study was conducted in six counties in North Carolina (NC) and six counties in Ohio (OH). These two states were selected to provide exposure information in two different geographical regions of the United States (i.e., the Southeast and Midwest). Overall, 257 preschool children and their adult caregivers took part in the study. Participants were recruited from eligible homes and child day care centers in the twelve counties. Participants were selected from several categories to allow for comparisons between home vs. day care settings, urban vs. rural locations, and low income vs. middle/high income environments. Although, the study focused on preschool children, information was also collected on the adult caregivers for comparison purposes. The results presented in this report apply only to the study participants; they have not been generalized to preschool children living in either state or to children in general.

Monitoring was performed over a 48-h period at the children's homes and/or day care centers. Environmental (air, dust, and soil) and personal (hand wipe, diet, water, and urine)

samples were collected. Surface wipe samples were collected from homes with recent pesticide applications. Questionnaires and diaries were used to collect information on housing characteristics, products used in the home, and activities of the participants. Multimedia samples were analyzed for over 50 pollutants belonging to such classes as the organophosphate (OP) pesticides, OP metabolites, organochlorine (OC) pesticides, pyrethroid pesticides, pyrethroid metabolites, acid herbicides, polycyclic aromatic hydrocarbons (PAHs), PAH metabolites, phthalates, phenols, and the polychlorinated biphenyls (PCBs). These pollutants were selected because they have been commonly detected in indoor and outdoor environments and/or because they are potentially carcinogenic, mutagenic, or endocrine-disrupting chemicals in humans.

Results of the study showed there were low levels of many pollutants in both the homes and day care centers where preschool children spend their time. Children can become exposed to these pollutants when they breathe the air, ingest food and water, ingest soil and dust, and touch contaminated surfaces. An absorbed dose occurs when pollutants are taken into the body though such routes as the lungs, intestines, and skin. Exposure and absorption into the body has been confirmed by measuring the same pollutants or metabolites of these pollutants in urine samples collected from children in the study.

The most frequently detected pollutants in environmental media were those commonly used in the home, those found in products used throughout the home, or those formed as a result of common processes. These pollutants included chlorpyrifos, diazinon, *cis*- and *trans*-permethrin, *alpha*- and *gamma*-chlordane, and pentachlorophenol, which are pesticides used in households. CTEPP was the first study to measure the metabolites of chlorpyrifos (3,5,6-trichloro-2-pyridinol [TCP]) and diazinon (2-isopropyl-6-methyl-4-pyrimidinol [IMP]) in environmental samples. These two compounds were detected at a very high rate in most sample types. Benzybutylphthalate, di-*n*-butylphthalate, and bisphenol-A, are commonly used plasticizers that were frequently detected. The PAHs were also frequently detected in most environmental samples. PAHs are formed during processes which involve burning of specific substances, with indoor sources including smoking and cooking, and outdoor sources including motor vehicles, incinerators, fires, and power plants. Target pollutants were detected most often in dust and indoor air samples. Only the PAHs were detected at a high rate in soil samples. Very few pollutants were detected in liquid food samples.

Median values of measured concentrations for selected pollutants are shown in Table ES-1 by state. The highest concentrations in most samples were found for the two phthalates, benzylbutylphthalate and di-*n*-butylphthalate. For the other pollutants, concentration rankings depended upon the media and the properties of the chemicals.

Table ES-1. Median Concentrations of Selected Pollutants Measured in Multiple Media.

	Indoor Air, ng/m^3		Dust, ng/g		Outdoor Air, ng/m^3		Dermal Wipe, ng/m^2		Solid Food, ng/g	
Pollutants/Metabolite	**NC**	**OH**	**NC**	**OH**	**NC**	**OH**	**NC**	**OH**	**NC**	**OH**
Chlorpyrifos	6.1	1.8	140	62	0.28	0.20	160	60	0.17	0.18
3,5,6-TCP	1.8	0.65	92	42	0.23	0.21	130	78	2.6	1.9
cis-Permethrin	0.41	<[a]	800	500	<	<	530	240	<	<
trans-Permethrin	0.27	<	730	390	<	<	300	190	<	<
Benzo[*a*]pyrene	0.08	<	200	930	0.09	<	<	40	<	<
Benzylbutylphthalate	<	<	19,000	19,000	<	<	7,900	<	<	11
Di-*n*-butylphthalate	240	260	6,800	6,400	<	<	9,000	<	<	<
Bisphenol-A	1.6	0.98	<	28	<	<	5,900	4,600	4.1	3.5

[a] "<" indicates that the median value falls below the MDL for the pollutant within the specified sample medium.

Comparisons of environmental measurements between home and day care settings, urban and rural locations, and low-income and middle/high-income environments showed few instances where the geometric mean concentration in one setting differed by a factor of three or more (when rounded) from the other setting, and where this difference was statistically significant. Incidences where such differences were observed included the following:

- ***Day Care vs. Home Environments.*** In both NC and OH, floor dust loadings (ng/m^2) averaged higher in day care centers than in homes, and this difference was statistically significant, for a number of current use pesticides, PAHs, and phthalates. This was likely a result of more dust being found in the day care centers, rather than higher concentrations of pollutants in the dust.

- ***Urban vs. Rural Environments.*** In OH, concentrations of the PAHs in dust samples, diazinon and IMP in outdoor air samples, and TCP in soil samples averaged higher in urban compared to rural settings, and this difference was statistically significant. In NC, the concentration of 2,4-D in floor dust samples tended to be higher in urban compared to rural settings.

- ***Low Income vs. Middle/High Income Environments.*** In NC, indoor air concentrations of diazinon and the permethrins averaged higher in low-income compared to middle/high-income environments, with the difference being statistically significant. The same was true for selected PAHs in soil. In both OH and NC, 2,4-D concentrations in dust were higher in middle/high-income compared to low-income homes. Finally in both states, floor dust loadings (ng/m^2) for pesticides were higher in low-income compared to middle/high-income homes. Again, this is likely a result of more dust found in low-income homes rather than to higher pesticide concentrations in the dust.

For 27 target pollutants, information on environmental and personal sample concentrations was combined with activity data to estimate potential exposure (ng/day) for each study participant by the inhalation, dietary ingestion, and indirect ingestion exposure routes. For each of these three exposure routes, potential absorbed dose (ng/kg/day) was also calculated by assuming a 50% absorption rate and dividing potential exposure by body weight. Results through the dermal route were not reported due to uncertainties in the assumptions required for the calculations. However, absorbed doses of these pollutants through the dermal route of exposure were assumed to be low.

For eight of the target pollutants (chlorpyrifos, diazinon, 3,5,6-TCP, *cis*-permethrin, *trans*-permethrin, 2,4-D, di-*n*-butylphthalate, and bisphenol-A), aggregate potential exposure and absorbed dose estimates were calculated by summing over all three routes. In both states, aggregate exposure and dose estimates were highest for di-*n*-butylphthalate, bisphenol-A, and 3,5,6-TCP. The NC and OH children had the highest median aggregate potential exposure levels to di-*n*-butylphthalate (42,900 and 8,310 ng/day), bisphenol-A (2,560 and 1,880 ng/day), and 3,5,6-TCP (1,230 and 930 ng/day). Median aggregate potential absorbed dose was highest among the NC and OH children for these same three pollutants (1,250 and 262 ng/kg/day for di-*n*-butylphthalate, 71.4 and 60.8 ng/kg/day for bisphenol-A, and 37.7 and 25.4 ng/kg/day for 3,5,6-TCP for NC and OH children, respectively). The median aggregate potential absorbed doses of di-*n*-butylphthalate was over four times greater in NC children compared to OH children. For di-*n*-butylphthalate, bisphenol-A, and 3,5,6-TCP, the relative importance of the exposure routes was dietary ingestion, followed by inhalation and indirect ingestion. In addition in both states, the children had the highest estimated aggregate exposures and absorbed doses to di-*n*-butylphthalate.

In several cases, there were significant differences in the calculated exposure and dose estimates between different groups of children. Those differences for which the geometric mean estimate was at least three times higher (when rounded) in one category than another included the following:

- ***Day Care vs. Stay-at-Home Children.*** In OH, exposure and dose estimates for diazinon, the PAHs, and benzylbutylphthalate via the indirect ingestion route were higher for day care children than stay-at-home children. Likewise, dietary exposure and dose estimates for benzylbutylphthalate and the permethrins were higher for the same group of children.

- ***Urban vs. Rural Children.*** In NC, exposure and dose estimates for 2,4-D by the indirect ingestion route were higher for children in urban compared to rural locations. In OH, PAHs showed higher estimates via the indirect ingestion route for urban children.

- ***Low Income vs. Middle/High Income Children.*** In NC, exposure and dose estimates for 2,4-D via the indirect ingestion route were higher for children in middle/high-income compared to low-income environments.

Because the indirect ingestion route was most frequently associated with sizable (and statistically significant) differences in exposure and dose estimates between groups of children, but yet accounted for a relatively small amount of the total or aggregate exposure for each child, it is not surprising that similar differences were not observed for aggregate exposure.

Some pollutants or metabolites were frequently detected and measurable in the children's urine samples, including 3,5,6-TCP, 2,4-D, and pentachlorophenol. Median urinary concentrations of 3,5,6-TCP, 2,4-D, and pentachlorophenol were 5.3, 0.7, and 0.4 ng/mL, respectively, for NC children. For OH children, median urinary concentrations of 3,5,6-TCP, 2,4-D, and pentachlorophenol were 5.1, 1.0, and 0.8 ng/mL, respectively. On average, levels of 3,5,6-TCP in urine samples for both NC and OH children were at least five times greater than those for 2,4-D or pentachlorophenol. As with estimates of aggregate potential exposure and absorbed dose, there were no incidences where differences in urinary concentrations were highly significant between various groups of children.

Finally, comparisons between children and their adult caregivers showed that children were generally exposed to higher levels of pollutants than adults in the same household, with the difference being statistically significant. Much of these differences was likely attributable to differences in physiological factors (i.e., ventilation rates and body weights) and activity patterns (i.e., daily soil and dust ingestion rates) between children and adults.