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An observational study of 127 preschool children at their homes and daycare centers in Ohio: Environmental pathways to *cis*- and *trans*-permethrin exposure

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Abstract

The potential exposures of 127 preschool children to the pyrethroid insecticides, *cis*- and *trans*-permethrin, in their everyday environments were examined. Participants were recruited randomly from 127 homes and 16 daycare centers in six Ohio (OH) counties. Monitoring was performed over a 48-h period at the children's homes and/or daycare centers. Samples collected included soil, carpet dust, indoor air, outdoor air, diet, hand wipes, surface wipes, transferable residues, and urine. The environmental samples were analyzed for the *cis* and *trans* isomers of permethrin, and the urine samples were analyzed for the pyrethroid urinary metabolite, 3-phenoxybenzoic acid (3-PBA), by gas chromatography/mass spectrometry. The isomers were detected most often in the dust (100%) and hand wipe (>78%) samples collected at both homes and daycare centers. The median levels of *cis*-permethrin (470 and 1010 ng/g) were higher than the median levels of *trans*-permethrin (344 and 544 ng/g) in the dust samples at both the children's homes and daycare centers, respectively. In the children's hand wipe samples, the median levels of *cis*- and *trans*-permethrin were similar, ranging from 0.03 to 0.04 ng/cm², at both locations. The urinary metabolite 3-PBA was detected in 67% of the children's urine samples. The median urinary 3-PBA concentration for the children was 0.3 ng/mL, and the maximum value for one child was 33.8 ng/mL. The primary route of the children's exposure to the combined isomers was through dietary ingestion, followed by indirect ingestion. In addition, our calculated aggregate absorbed doses of permethrin accounted for about 60% of the excreted amounts of 3-PBA found in the children's urine. In conclusion, these children were potentially exposed to low levels of permethrin from several sources, and through several pathways and routes

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1. Introduction

Permethrin is a synthetic pyrethroid insecticide that is used worldwide to control a broad range of insect pests such as ticks, fleas, roaches, chiggers, and ants (Heudorf and Angerer, 2001; Kolaczinski and Curtis, 2004; Tomlin, 1994). The pesticide has been evaluated by the Agency and approved for use in agriculture, forestry, and in residential settings, and on pets to control for insect pests (USEPA, 2005a,b; Kuhn et al., 1999). Permethrin is also commonly

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incorporated into wool carpets and other textiles at manufacturing facilities to prevent insect damage (Leng et al., 1997; Lewis et al., 1999; Schettgen et al., 2002). Recent research has indicated that permethrin is usually persistent indoors, has low volatility, and tends to adsorb highly onto materials such as carpets, fabrics, and dust in dwellings (Kolaczinski and Curtis, 2004). Therefore, there is the potential for children to be exposed to low levels of permethrin in their daily environments (homes, daycare centers, and schools).

Permethrin is a lipophilic insecticide that has low mammalian toxicity (Anadon et al., 1991; Angerer and Ritter, 1997; Kuhn et al., 1999). As a neurotoxin, its mode of action is to alter the sodium ion channels in the peripheral and central nervous system of exposed mammals and insects (Angerer and Ritter, 1997; Schettgen et al., 2002). In humans, the absorption of permethrin through dermal and inhalation exposure is considered low. Ingestion of permethrin residues, particularly in food, appears to be the major route of human exposure (Heudorf and Angerer, 2001). Once absorbed into the body, permethrin (cis/trans) is quickly metabolized through ester cleavage and oxidation to more polar metabolites including cisand trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-1-carboxylic acid (cis-Cl₂CA and trans-Cl₂CA) and 3phenoxybenzoic acid (3-PBA) (Hardt and Angerer, 2003; Leng et al., 1999; Schettgen et al., 2002). Renal elimination is the primary pathway for removal of these metabolites in the body (Heudorf and Angerer, 2001; Kuhn et al., 1999).

A number of pyrethroids can be metabolized into 3-PBA in humans. Therefore, this metabolite is commonly used as a nonspecific urinary biomarker of exposure to pyrethroid pesticides. These include pyrethroids such as cypermethrin, deltamethrin, fenvalerate, fluvalinate, permethrin, and sumithrin (Baker et al., 2000; Berkowitz et al., 2003; Heudorf and Angerer, 2001; Hu et al., 2004; Leng et al., 1997; Schettgen et al., 2002). Recently, a few studies have detected measurable levels of 3-PBA in the urine of nonoccupationally exposed adults and children in the United States (US; Baker et al., 2000, 2004; Berkowitz et al., 2003; CDC, 2005).

In the US, little research has investigated the potential exposures of children to permethrin in their environments. A few studies have reported low levels of cis- and transpermethrin in carpet dust, floor wipe, and air samples at homes in several states (Clayton et al., 2003; Colt et al., 2004; Quandt et al., 2004; Whyatt et al., 2002). These studies imply that children could be exposed to permethrin from several sources and through several pathways and routes in their everyday environments. In the work reported here, the objectives were to quantify the distributions of cis- and trans-permethrin in several media at a large set of preschool children's homes and daycare centers in Ohio (OH); quantify the urinary 3-PBA concentrations for the children; estimate the children's potential absorbed doses to cis- and trans-permethrin compared to their excreted amounts of urinary 3-PBA; and identify the important sources and routes of the children's exposures to these isomers.

2. Materials and methods

The Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP) study investigated the aggregate exposures of 257 preschool children to chemicals commonly found in their everyday environments in North Carolina (phase I, 2000) and OH (phase II, 2001). As part of this study, the potential exposures and absorbed doses of the preschool children to *cis*- and *trans*-permethrin at their homes and daycare centers in OH were examined. Only the OH results are presented here, because we did not have a successful quantification method for 3-PBA in the children's urine samples until phase II of the study.

The study design for the CTEPP study has been previously described by Wilson et al. (2004a). In the OH phase, preschool children, ages 2–5 years, were randomly recruited from homes and daycare centers in six OH counties, between January and November 2001. A total of 127 children were recruited successfully: 69 and 58 children in the home and daycare groups, respectively. The median age of the children was 48 months, and their ages ranged from 20 to 67 months.

This was an "observational research" study, as defined in 40 CFR Part 26.402. The study protocol and procedures to obtain the assent of the children and informed consent of their parents or guardians were reviewed and approved by an independent institutional review board (IRB) and complied with all applicable requirements of the Common Rule regarding additional protections for children.

2.1. Field sampling

An in-depth description of the sampling methodology in the CTEPP study can be found elsewhere (Wilson et al., 2004a). Briefly, field sampling at the homes and daycare centers occurred from April 2001 through November 2001. The children from the home group had multimedia samples collected over a 48-h period at their residences. The daycare group participants had these samples collected simultaneously over the 48-h period at both their homes and daycare centers. Samples collected included soil, indoor air, outdoor air, carpet dust, drinking water, solid food, liquid food, hand wipes, and spot urine. Up to six spot urine samples (i.e., morning, after lunch, and before bedtime) were collected from each child over the 48-h sampling period. Additional samples were collected if a pesticide had been applied at a home or daycare center within seven days prior to field monitoring. These additional samples consisted of transferable residues, hard floor surface wipes, and food preparation surface wipes. Thirteen families reported that they had applied a neutral pesticide (i.e., pyrethroid, organophosphate) at their home within seven days prior to field monitoring. No daycare centers reported applying a pesticide during this time period. Having recent pesticide applications was coincidental to the scheduled visits to the homes. The researchers/field team had no prior knowledge about the use of pesticides or other chemicals in the homes at the time of scheduling, and use of pesticides was not a selection criterion for participation in the study. Other data that were collected included household observations, pre- and post-monitoring questionnaires, children's activity diaries, and food diaries. Multimedia samples were collected at 127 homes and 16 daycare centers.

2.2. Chemical analysis

Cis- and trans-permethrin were analyzed in all the collected environmental media, except urine. The metabolite 3-PBA was measured in the urine samples. The detailed extraction and analysis methods for cis- and trans-permethrin and 3-PBA in media can be found in Morgan et al. (2004).

Matrix spikes were used for the two isomers in all environmental media and for 3-PBA in urine. The surrogate recovery standard (SRS) for permethrin was p,p'-DDE-d₄, and the SRS for 3-PBA was 2,4-D-C₁₃.

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