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Dietary predictors of young children's exposure to current-use pesticides using urinary biomonitoring

Marsha K. Morgan *, Paul A. Jones

United States Environmental Protection Agency's National Exposure Research Laboratory, 109 T.W. Alexander Drive, Durham, NC 27711, USA

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ABSTRACT

Few data exist on the association between dietary habits and urinary biomarker concentrations of pesticides in children. The objective was to examined the association between the weekly intake frequency of 65 food items and urinary biomarkers of exposure to chlorpyrifos (3,5,6-trichloro-2-pyridinol [TCP]), permethrin (3-phenoxybenzoic acid [3-PBA]), and 2,4-dichlorophenoxyacetic acid [2,4-D] in 135 preschool-aged children. TCP and 3-PBA are nonspecific biomarkers as they are also urinary metabolites of other pesticides. TCP, 3-PBA, and 2,4-D were detected in 99%, 64%, and 92% of the urine samples, respectively. Mean urinary TCP concentrations were statistically significantly higher in children consuming fresh apples (9.40 ± 15.5 ng/mL versus 5.76 ± 3.57 ng/mL, $p = 0.040$) and fruit juices (8.41 ± 11.5 ng/mL versus 4.11 ± 2.77 ng/mL, $p = 0.020$) $\geqslant$3 times a week compared to <3 times a week. For 3-PBA, mean urinary metabolite concentrations were statistically significantly greater in children consuming chicken/turkey meats (0.79 ± 0.81 versus 0.41 ± 0.39, $p = 0.013$) $\geqslant$3 times a week compared to <3 times a week. No association occurred between the consumption of any food item and children's mean urinary 2,4-D concentrations by intake group. In conclusion, frequent consumption of fresh apples and fruit juices or chicken/turkey meats were significant dietary predictors of urinary levels of TCP or 3-PBA, respectively.

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1. Introduction

In the United States (US), chlorpyrifos and permethrin are insecticides that are frequently used to control insect pests on a variety of agricultural crops (US EPA, 2007; Dow AgroSciences, 2013). 2,4-Dichlorophenoxyacetic acid (2,4-D) is a phenoxy herbicide that is also commonly applied to farm fields for selective control of broadleaf weeds (Arbuckle and Ritter, 2005). It is estimated that approximately 10, 1, and 30 million pounds of chlorpyrifos, permethrin, and 2,4-D, respectively, are used in agricultural settings each year (US EPA, 2002, 2005, 2007). Chlorpyrifos and permethrin residues have been detected in a number of different types of fresh fruits and vegetables and other processed food products (e.g., fruit juices, grains, and meats) purchased at supermarkets and grocery stores across the US (Katz and Winter, 2009; FDA, 2013). For 2,4-D, residues have been mainly found in purchased cereal and bread products (FDA, 2013). As children typically consume these kinds of foods, they are likely being exposed to pesticides in their diets.

Once absorbed into the body, 2,4-D undergoes little metabolism and is mainly renally eliminated as the parent compound (Sauerhoff et al., 1977). For chlorpyrifos, it is quickly metabolized into diethylphosphate, diethylthiophosphate, and 3,5,6-trichloro-2-pyridinol (TCP), and these metabolites are primarily excreted in urine (CDC, 2009). Permethrin is also rapidly metabolized and is mainly eliminated in urine as *cis*-3-(2,2-dichlorovinyl)-2,2-dimethyl-cyclopropane carboxylic acid, *trans*-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane carboxylic acid, and 3-phenoxybenzoic acid (3-PBA) (CDC, 2009). These metabolites of chlorpyrifos and permethrin are nonspecific urinary biomarkers as other pesticides in consumed foods can also be broken down in the body to form these metabolites in urine (CDC, 2009).

In the past, several observational exposure measurements studies have shown that dietary ingestion is a major route of children's exposure to these three pesticides (Clayton et al., 2003; Wilson et al., 2003; Morgan et al., 2005; Wilson et al., 2010). These types of studies, however, typically measure the levels of pesticide residues in solid or liquid food samples that have been composited over a 24-h or 48-h period due to the high cost of chemical analyses (Morgan et al., 2005; Bradman et al., 2007; Tulve et al., 2008; Chuang and Wilson, 2011). This makes it difficult to identify the specific food item(s) that contribute to children's dietary exposures to pesticides. Recently, Lu et al. (2010) reported measurable levels

Abbreviations: 2,4-D, 2,4-dichlorophenoxyacetic acid; 3-PBA, 3-phenoxybenzoic acid; LOD, limit of detection; LOQ, limit of quantification; TCP, 3,5,6-trichloro-2-pyridinal; US, United States.

* Corresponding author. Tel.: +1 919 541 2598; fax: +1 919 541 0905.
E-mail addresses: morgan.marsha@epa.gov (M.K. Morgan), jones.paul-a@epa.gov (P.A. Jones).

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of chlorpyrifos (up to 3 ng/g) and permethrin (up to 82 ng/g) in some individual fruits, vegetables, and/or fruit juices that were consumed by children over a 24-h monitoring period in the states of Georgia and Washington. This information suggests that some foods (e.g., fruits and vegetables) commonly consumed by children may be contributing to the majority of their dietary exposures to these current-use pesticides.

Only a few studies have assessed the relationship between food consumption habits and urinary biomarker concentrations of pesticides in children (Riederer et al., 2008; Munoz-Quezada et al., 2012). Riederer et al. (2008) conducted a study of 179 US children (ages 6–10 years of age) showing that the consumption of cheese, cookies, ground beef, ice cream, tortilla chips, and white bread were significant dietary predictors of the children's urinary 3-PBA concentrations. Recently, Munoz-Quezada et al. (2012) reported that the consumption of fruits containing chlorpyrifos residues was a strong dietary predictor of dialkylphosphate metabolite concentrations in the urine of 190 Chilean children, ages 6–12 years of age. These studies provide evidence that the consumption of specific foods likely increases children's exposures to some pesticides.

In previous work, we quantified the aggregate exposures of preschool children to chlorpyrifos, permethrin, and 2,4-D in environmental and personal media over a 48-h monitoring period at their homes and daycare centers in 2000–2001 (Morgan et al., 2004, 2005, 2007, 2008). Dietary ingestion was found to be a major route of the children's exposure to all three pesticides. However, since the duplicate diet samples were composited over the monitoring period for each child, it remains unclear which of the children's consumed food(s) likely contributed to their dietary exposures to the pesticides. For this paper, the objective was to examine the association between the weekly intake frequency of 65 selected food items and urinary biomarkers of exposure to chlorpyrifos (TCP), permethrin (3-PBA), and 2,4-D (unchanged parent) in preschool-aged children.

2. Materials and methods

2.1. Study cohort

The Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants study examined the aggregate exposures of 256 preschool children, ages 2–5 years, and their primary adult caregivers to chemicals commonly found in their everyday environments. A detailed description of the study design has been described earlier in Wilson et al. (2004). Briefly, this study was conducted at 256 children's homes and 29 child daycare centers in six counties each in North Carolina and Ohio from 2000 to 2001. This study cohort consisted of a group of 135 children who stayed-at-home with their adult caregivers (usually a parent) during the day, and another group of 121 children who attended daycare during the day. Field staff collected soil, outdoor air, indoor air, and carpet dust samples at each location over a 48-h monitoring period. Adult caregivers collected duplicate diet (solid and liquid food samples), hand wipe, and urine samples from themselves and their children during this same time period. In addition, adult caregivers filled out several different types of questionnaires and diaries on their children's activities (i.e., children's dietary habits).

For this present work, we used a subset of the CTEPP data consisting of 135 children that stayed-at-home with their adult caregivers during the day. In our statistically analyses, we examined the children's weekly food consumption diary data and their 48-h urinary measurement data for TCP, 3-PBA, and 2,4-D. One additional child was excluded from this dataset as they had a missing urine sample.

2.2. Human subjects research

This was an observational research study, as defined in 40 Code of Federal Regulations, Part 26.402. The study protocol and procedures to obtain the assent of the preschool children and informed consent of their adult caregivers were reviewed and approved by an independent institutional review board and complied with all applicable requirements of the Common Rule regarding additional protections for children (Subpart D).

2.3. Food frequency consumption diary and urine sample collection

During the 48-h monitoring period, the adult caregivers filled out a food frequency consumption diary that recorded information on their children's usual eating habits (i.e., on a weekly basis) for 65 specific food items that were consumed over the past year. Food items in the diary included a number of different types of fruits, vegetables, meats, breads, snacks, and beverages (excluding water). In addition, up to six spot urine voids (i.e., morning, after lunch, and before bedtime) were collected from each child by their adult caregiver over this monitoring period. A spot urine void was collected from a child by placing a plastic collector (bonnet) under the toilet seat prior to urination. The adult caregiver transferred the urine void from the bonnet into a 120 mL plastic container with lid. Individual urine samples were stored in provided coolers with ice by adult caregivers at residences until they were picked up by field staff. Urine samples were stored in freezers (<−20 °C) at the Battelle laboratory in Columbus, Ohio until analyses.

2.4. Urine sample analysis

At the laboratory, spot urine samples were pooled over a 48-h monitoring period for each child, except for children that had a recent pesticide application(s) at home within seven days of field sampling. For these children, all of their spot urine samples (up to six) were analyzed separately. For chemical analyses, 1 mL of urine was used per sample. The urine samples were analyzed for biomarkers of exposure to chlorpyrifos (TCP), permethrin (3-PBA), and 2,4-D (parent compound). Detailed information on the extraction and analysis procedures used to quantify for TCP, 3-PBA (Ohio, only), and 2,4-D in the urine samples, including quality assurance and quality control information, can be found in Morgan et al. (2005, 2007, 2008). Briefly, urine samples were analyzed for the target analytes by a gas chromatography/mass selective detector (Hewlett–Packard 6890/5973A) in the selected ion-monitoring mode. The limit of quantification (LOQ) was defined "as the minimum concentration at which an analyte can be quantitatively measured in a sample medium with acceptable accuracy and precision" (Morgan et al., 2005). For the LOQs, they were derived from the lowest calibration standard for each analyte with a signal-to-noise ratio above 2. The lowest calibration standard was 2 ng/mL for TCP and 2,4-D and 1 ng/mL for 3-PBA. The estimated LOQ's for TCP, 3-PBA, and 2,4-D in urine were 2.0, 0.4, and 0.4 ng/mL, respectively. The estimated limit of detection (LOD) was defined "as the minimum analyte level detected in a sample and was estimated to be ½ of the LOQ" (Morgan et al., 2004).

2.5. Data analysis

All pooled and non-pooled urine measurement values less than the LOD were replaced by the LOD divided by the square root of two. For the non-pooled urine samples, the mean concentration value was used for each analyte. Descriptive statistics (sample size, frequency of detection, arithmetic mean and standard deviation, geometric mean, median, range, and percentiles (25th, 75th, and 95th)) were calculated as unadjusted urine values (ng/mL) by analyte. Unadjusted urine values were used in all statistical analyses. Creatinine-adjusted urine values were not used as this common correction method for variable dilutions in spot urine samples is probably not a reliable method for young children (O'Rourke et al., 2000; Barr et al., 2005).

Each child's consumption frequency for the 65 specific food items was computed on a weekly basis. Based on the children's weekly food habits, they were then placed into either a low food intake group (<3 times per week) or a high food intake group ($\geqslant$3 times per week) for each food item. A total of 31 food items were excluded because they were not commonly consumed by either intake group (<20 children for both states or <10 children for Ohio [3-PBA, only]).

The distribution of the metabolite concentrations of TCP, 3-PBA and 2,4-D were first examined for normality. Based on a normality plot and the Kolmogorv Smirnov test, the data from all three distributions were found to be non-normal. For this reason, both the arithmetic and geometric means were presented in the descriptive analysis for comparison. Although these data are non-normal, the standard *t*-test was used based on the central limit theorem (CLT), a well-established and commonly used statistical principle. For a large sample size *n*, the CLT indicates that the means of the samples from a population with finite variance approach a normal distribution even if the underlying distribution of the individual observations (x_i's) in the population is not normal. Therefore, a more robust two-sample *t*-test was used to determine whether statistically significant differences existed between the children's weekly food habits for 34 specific food items and their urinary TCP, 3-PBA, or 2,4-D concentrations by intake group. In addition, a multivariate analysis was used to examine the impact of selected covariates (gender, body weight, and recent residential pesticide-use) on the relationship between a food item found to be statistically significant and urinary biomarker concentrations. All statistical analyses were performed using SAS version 9.1 (SAS, Cary, North Carolina).

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