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

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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) $\geq$ 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) $\geq$ 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.

Keywords: Preschool children, food, ingestion, TCP, 3-PBA, 2,4-D, urinary biomarkers

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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

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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-hour or 48-hour 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 measureable levels 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-hour 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 U.S. 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-hour 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 and 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-hour 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-hour 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-hour 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^{o}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-hour 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, and 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 (25^{th}, 75^{th}, and 95^{th})) 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 (≥ 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).

3. Results

3.1 Demographics

Table 1 presents the demographic characteristics for the 135 stay-at-home children from North Carolina and Ohio. The preschool children's ages ranged from 20 to 67 months and 75% of them were between the ages of 37 and 67 months. Of these children, 54% were female and 46% were male. The children's median body weight was 16.2 kg, and their weights ranged from 10.4 to 44.1 kg. The majority of the children were white (84%), resided in urban areas (84%), and came from middle/upper-income families (68%). A total of 18 households had a recent application of a pesticide occurring at their residence within seven days of field sampling. Of these residences, four homeowners reported using a product containing either chlorpyrifos, permethrin, or 2,4-D. In North Carolina, one homeowner sprayed permethrin using an aerosol can in their kitchen to control for cockroaches, and another owner applied chlorpyrifos granules with a broadcast spreader to their yard to control a variety of insect pests. In Ohio, one homeowner sprayed chlorpyrifos using an aerosol can in the garage to control ants, and a second one applied 2,4-D granules with a broadcast spreader to their landscaping and garden to control for weeds.

3.2 Weekly food consumption patterns

The weekly consumption of specific food items by the preschool children is presented in Supplemental Table S-1. In this cohort, apples, bananas, carrots, grapes, and potatoes were the most frequently eaten ($\geq$ 3 times a week) fresh fruits and vegetables. For meats, the top ones consumed three or more times a week by the children were chicken/turkey (baked, stewed, roasted, or broiled) and ham/lunch meats. Cheeses/cheese spreads, butter, margarine, and milk (2% and whole) were the most often consumed dairy products. Rarely or never eaten food items among the children were melons, grapefruits, oranges/tangerines, peaches/nectarines, pears, broccoli, cabbage, cauliflower, corn, greens/spinach, bacon, beef steak/roasts, beef stew/pot pie, fried chicken, fried fish/fish sandwich, other fish, hamburgers, hotdogs, liver, pork, sausage, frozen yogurt, cooked cereals, corn bread/muffins, rice, spaghetti/lasagna, canned soups, honey, pies, pizza, and sugar-free soft drinks.

3.3 Urinary biomarker concentrations

Table 2 presents the biomarker concentrations that were measured in the children's urine samples. TCP, 3-PBA (Ohio, only), and 2,4-D were detected in 99%, 64%, and 92% of the urine samples, respectively. The children's median urinary concentrations were 5.3 ng/mL for TCP, 0.24 ng/mL for 3-PBA, and 0.92 ng/mL for 2,4-D. The median level of TCP was at least 5.5 times greater than median levels of 3-PBA or 2,4-D in the children's urine samples. The two children that had a recent pesticide application of chlorpyrifos at their homes had mean urinary TCP concentrations of 1.5±1.1 ng/mL (range, <LOD-3.30 ng/mL) and 11.0±8.1 ng/mL (range, 4.3-25.3 ng/mL). For the latter child who had this insecticide applied in their garage, two of their spot urine samples had TCP concentrations (15.2 ng/mL and 25.4 ng/mL) that were greater

than the 95th percentile. In addition, the child that had a recent outdoor application of 2,4-D at their home had a mean urinary concentration of 0.66±0.38 ng/mL for this herbicide, and all of their spot urine measurements (range, 0.22-1.26 ng/mL) were below the 75th percentile.

3.4 Association between food habits and urinary biomarker concentrations

Tables 3, 4, and 5 present the association (i.e., differences between mean groups) between the consumption of a food item and urinary biomarker concentrations for TCP, 3-PBA, and 2,4-D, respectively, by intake group. 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) three or more times a week compared to less than three 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) ≥ 3 times a week compared to < 3 times a week; while mean urinary 3-PBA concentrations were marginally significant for those consuming butter (0.68±0.70 ng/mL versus 0.41±0.42, p=0.052) or fresh apples (0.65±0.71 versus 0.40±0.37, p=0.058). No association existed between the consumption of any food item and mean urinary 2,4-D concentrations by intake group. In our multivariate analyses after adjusting for gender, body weight, and recent residential pesticide application, all covariates were non-significant, except gender (p=0.034) in which fresh apple consumption was the main effect variable on the children's mean urinary TCP concentrations.

4. Discussion

4.1 Children's food consumption habits

Research has shown that children's diets can vary greatly by life stage (i.e., infant, toddlers, preschoolers, and adolescence) and among children of similar age bins (Hubal et al., 2000). Few studies have investigated the food consumption habits of preschool children, particularly over time in the US (Kranz et al., 2004; Reedy and Krebs-Smith, 2010; Ford et al., 2013). From 2003-2006, Reedy and Krebs-Smith (2010) reported that foods substantially contributing to the caloric intake of US children, ages 24-36 months, were whole and reduced fat milks, fruit juices (100%), pastas, and desserts (e.g., cakes, cookies, donuts, and pies). In our analyses, the top food items frequently eaten (≥ 5 times a week) by the preschool children between 2000-2001 were fruit juices (100%), 2% and whole milk, cereals, white breads/bagels/crackers, and cakes/doughnuts/cookies/pastries. In addition, Ford et al. (2013) recently reported that there has been a significant increase in the per capita intake of foods high in added sugars, salt, and solid fats (i.e., sweet and salty snacks, candy, pizzas, calzones, cheeses, and fruit juices) among children, ages 24-72 months of age, from 1989 to 2008 in the US. This is also supported by research conducted by Kranz et al. (2004) that showed that there was a marked increase in the dietary consumption of added sugars and fruit juices among US preschool children between 1977 and 1998. As dietary habits have changed over time in the US, more research is necessary to understand young children's food consumption patterns and how these patterns impact their exposures to pesticides and to other chemicals today.

4.2 Diet as a source of exposure to pesticides

The diet is a major source of children's exposure to a number of current-use pesticides including chlorpyrifos, permethrin, and 2,4-D (NRC, 1993; Morgan et al., 2004; Lu et al., 2006). It is suspected that only a portion of the foods eaten by children on a daily basis actually contain pesticide residues. However, little information exists on the pesticide levels in the actual foods and beverages consumed by children. In the US, only two studies have quantified the levels of some pesticide residues in individual food items consumed by children (Fenske et al., 2002; Lu et al., 2010). In 1998, Fenske et al. (2002) measured the levels of 15 different organophosphorus pesticides in a small number of foods and beverages consumed over a 24-hour period by 13 preschool children, ages 2-5 years old, in the state of Washington. Up to six different organophosphates were detected in fresh produce, beverage, and/or processed food samples, but none of them in dairy samples. Chlorpyrifos residues were found in one child's cherry tomato sample at 350 ng/g, which was the highest pesticide level measured in any food sample. Recently, Lu et al. (2010) quantified the levels of several different organophosphorus and pyrethroid pesticides in 24-hour duplicate diet samples consisting of all consumed fruits, vegetables, and fruit juices of 46 children, ages 3-11 years old, from the states of Georgia and Washington. A total of 11 different organophosphates (range, 1-387 ng/g) and three different pyrethroids (range, 2-1,133 ng/g) were found in the food samples (Lu et al., 2010). In individually analyzed food items, chlorpyrifos residues were measureable in a child's orange juice sample (1 ng/g) and fresh orange sample (3 ng/g). Permethrin residues were also present in a child's cooked broccoli sample (82 ng/g) and fresh apple sample (4 ng/g). In addition, Lu et al. (2010) conducted a market basket analysis of fresh fruits and vegetables frequently eaten by the study children at a local grocery store in Washington state. Chlorpyrifos residues were found in samples of grapes (22 ng/g), nectarines (1 ng/g), and strawberries (363 ng/g), however, no foods contained detectable levels of permethrin residues. This above research suggests that pesticide levels in agricultural foods are variable and are likely impacted by factors such as usage patterns, seasonality, weather conditions, geographical region, and peeling or prewashing fruits and vegetables prior to consumption (Kimata et al, 2009; Lu et al., 2010). As young children have a less varied diet, particularly consuming more fruits and vegetables based on their body size, they are likely exposed to higher amounts of pesticide residues in foods they eat than older children and adults (Hubal et al., 2000). More research is needed to quantify the levels of pesticide residues in food items that children eat as certain ones likely contribute to the majority of their dietary exposures to current-use pesticides.

4.3 Children's estimated intake doses of the pesticides

Assuming steady state absorption, the preschool children's estimated maximum aggregate intake doses to chlorpyrifos, permethrin, and 2,4-D were 2.32, 0.056, and 0.28 ug/kg/day, respectively. The children's intake doses were calculated by multiplying the highest urinary concentration of each metabolite in Table 2 by an estimated daily urine excretion rate (22.4 mL/kg body weight) for a child (Miller and Stapleton, 1989; Szabo and Fegyverneki, 1995). The children's exposure levels to permethrin and 2,4-D were 893 and 36 times lower than the established oral reference doses of 50 ug/kg/day and 10 ug/kg/day, respectively, in the US EPA's Integrated Risk Information System (IRIS) (IRIS, 2013). For chlorpyrifos, there is currently no oral reference dose available in IRIS. However as a protective measure in 2001, the US EPA phased out almost all residential uses of chlorpyrifos to greatly reduce children's potential exposures and possible health risks to this insecticide (US EPA, 2002).

4.4 Association between food consumption habits and urinary biomarker concentrations

A limited number of published studies have assessed the relationship between food consumption habits and urinary biomarker concentrations of pesticide in children or adults

(Kieszak et al., 2002; Riederer et al., 2008; Kimata et al., 2009; Munoz-Quezada et al., 2012; Fortes et al., 2013). Kieszak et al. (2002) reported that no association existed between the self-reported monthly consumption of fruits, vegetables, or bread products and urinary pesticide biomarker concentrations, including TCP, for 978 adults from the Third US National Health and Nutritional Examination Survey 1988-1994. However, the authors stated that the lack of association between food consumption habits and urinary biomarker levels in adults were likely due more to the limitations of the study data (i.e., frequency of intake). In a later Survey (1999-2002), Riederer et al. (2008) showed that diet was a significant predictor of urinary concentrations of 3-PBA in pre-adolescents (6-10 years old), adolescents (11-18 years old), and adults. In particular for 179 preadolescents, the consumption of cheese, cookies, ground beef, ice cream, tortilla chips, and white bread were statistically significantly ($p<0.05$) associated with urinary 3-PBA concentrations. The authors, however, reported that gender, body mass index, and residential pesticide-use were not significantly associated with the children's urinary levels of 3-PBA. Recently, Fortes et al. (2013) reported that a higher intake of fresh and cooked vegetables greater than five times a week were significantly ($p<0.05$) associated with higher mean urinary 3-PBA concentrations in 55 adults from Italy, and this association was not impacted by age, sex, body mass index, or household insecticide use. In our study, results showed that consumption of chicken/turkey meats and butter more than three times a week were significantly ($p<0.05$) associated with higher mean urinary 3-PBA concentrations in preschool children. We also showed similar results as Riederer et al. (2008) that gender and body weight of children and recent residential pesticide-use were not major factors influencing urinary 3-PBA concentrations. Lastly, Munoz-Quezada et al. (2012) reported that consumption of fruits containing chlorpyrifos residues was a significant dietary predictor of urinary (dialkylphosphate) metabolite concentrations in 190 children, ages 6-12 years old, from Chile. This is in agreement with our study that also showed that the frequent consumption of fruits (apples and juices) were significantly associated with higher urinary TCP concentrations in children which suggested that some of their consumed fruit items likely contain measureable levels of chlorpyrifos.

4.5 Limitations of the study data

The study data had several limitations. The food frequency consumption data were based on the children's normal weekly eating habits for 65 specific food items over the past year and may not reflect the actual food items they ate over the 48-hour monitoring period. In addition, no diary data were collected on the weight or volume of individual food items typically consumed by the children on a daily or weekly basis. The amount of solid or liquid food items consumed by a child over time would likely impact their exposure levels to pesticides and resulting urinary biomarker concentrations. Lastly as most of the children (87%) had one spot urine measurement, the temporal association between the children's food consumption habits and urinary pesticide biomarker concentrations could not be determined.

5. Conclusions

Urinary biomarker data showed that the majority of the preschool children were exposed to chlorpyrifos, permethrin, and 2,4-D at their homes during the 48-hour monitoring period. The frequency of consumption of fresh apples and fruit juices or chicken/turkey meats were confirmed to strongly predict the urinary concentrations of TCP or 3-PBA, respectively, in the children. However, no association was found between the consumption of any food item and children's urinary 2,4-D levels by intake group.

Conflict of Interest

The authors declare that there are no conflicts of interest.

Disclaimer

"This United States Environmental Protection Agency through its Office of Research and Development has provided administrative review of this article and approved it for publication."

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Table 1. Demographic characteristics of the preschool children

Characteristic	N[a]	%[b]
Age (months)		
< 24	3	2%
24 - 36	30	22%
> 36	102	76%
Gender		
Females	73	54%
Males	62	46%
Race		
Asian/Pacific Islander	1	1%
Black	15	11%
Hispanic	3	2%
White	114	84%
Other	1	1%
Unknown	1	1%
Urbanicity (county-level)		
Urban	114	84%
Rural	21	16%
Family income		
Low-income	34	25%
Middle/upper-income	91	68%
Unknown	10	7%
Recent residential pesticide-use[c]		
Yes	18	13%
No	117	87%

[a]Number of children
[b]Percentage of children
[c]Within seven days prior to field sampling at home

Table 2. Urinary biomarker concentrations in preschool children over a 48-hour monitoring period.[a]

Urine (ng/mL)	N[b]	%[c]	Mean[d]	SD[e]	GM[f]	Min.	*Percentiles* 25th	50th	75th	95th	Max.
TCP	135	99	7.1	9.9	5.04	<[g]	3.2	5.3	9.1	14.7	103.7
3-PBA[h]	69	64	0.50	0.54	0.33	<	<	0.24	0.63	1.9	2.5
2,4-D	135	92	1.2	1.7	0.77	<	0.40	0.92	1.4	3.0	12.5

[a]The children's urinary TCP, 3-PBA, and 2,4-D concentration data were calculated using data in Morgan et al. (2005, 2007, 2008)
[b]Number of children
[c]Percentage of samples with detectable concentrations of an analyte
[d]Arithmetic mean
[e]Standard deviation
[f]Geometric mean
[g]Below the limit of detection
[h]3-PBA was only quantified in the children's urine samples from Ohio

Table 3. Children's weekly consumption of food items and their mean urinary TCP concentrations (ng/mL) by intake group. [a]

Food Item	N[b]	%[c]	Mean±SD[d]	GM[e]	p-Value
Fresh fruits or vegetables					
Apples					
Low[f]	85	63	5.76±3.57	4.69	
High[g]	50	37	9.40±15.5	5.69	0.040
Bananas					
Low	84	62	7.34±11.3	5.20	
High	51	38	6.72±7.21	4.78	0.729
Grapes					
Low	98	73	6.84±10.5	4.93	
High	37	27	7.81±8.26	5.32	0.613
Strawberries					
Low	111	82	7.35±10.8	5.11	
High	24	18	5.99±4.14	4.69	0.545
Carrots					
Low	94	70	6.16±5.65	4.82	
High	41	30	9.27±15.8	5.56	0.094
String beans or green beans					
Low	115	85	7.01±9.88	5.05	
High	20	15	7.67±10.4	4.95	0.786
Tomatoes					
Low	112	83	7.41±10.8	5.14	
High	23	17	5.64±3.54	4.57	0.437
Potatoes					
Low	98	73	6.62±5.82	5.05	
High	37	27	8.41±16.5	4.99	0.351
Processed fruits or vegetables					
Canned or bottled fruit					
Low	102	76	6.53±5.82	4.97	
High	33	24	8.90±17.4	5.24	0.234
Canned or bottled vegetables					
Low	82	61	6.51±6.28	4.73	
High	53	39	8.03±13.8	5.55	0.385
Frozen vegetables					
Low	91	67	6.10±3.82	4.90	
High	44	33	9.18±16.4	5.32	0.091
French fries or fried potatoes					
Low	109	81	7.38±10.9	5.10	
High	26	19	5.97±3.72	4.76	0.518
Fortified fruit drinks					
Low	90	67	7.84±11.8	5.39	
High	45	33	5.63±3.72	4.40	0.223
Fruit juices					
Low	41	30	4.11±2.77	3.37	
High	94	70	8.41±11.5	6.00	0.020
Meats					
Chicken or turkey meats					

Low	112	83	7.22±10.8	4.98	
High	23	17	6.54±3.89	5.33	0.763
Ham or lunch meats					
Low	99	73	7.51±11.4	5.09	
High	36	27	5.99±3.74	4.88	0.435
Dairy					
Butter					
Low	88	65	6.91±11.1	4.77	
High	47	35	7.47±7.27	5.58	0.758
Cheeses and cheese spreads					
Low	43	32	7.61±15.4	4.50	
High	92	68	6.87±5.96	5.31	0.691
Eggs					
Low	114	84	7.56±10.7	5.33	
High	21	16	4.63±2.94	3.69	0.214
Ice cream					
Low	93	69	6.41±5.92	4.78	
High	42	31	8.65±15.5	5.65	0.225
Margarine					
Low	66	49	5.89±6.54	4.22	
High	69	51	8.27±12.3	5.96	0.165
Yogurt					
Low	92	68	7.64±11.8	5.10	
High	43	32	5.96±3.45	4.90	0.361
2% Milk or beverages with 2% milk					
Low	72	53	7.71±13.2	4.73	
High	63	47	6.41±3.47	5.41	0.450
Skim milk, 1% milk, or butter milk					
Low	104	77	7.25±11.1	5.01	
High	31	23	6.62±4.26	5.13	0.755
Whole milk or beverages with whole milk					
Low	81	60	6.98±6.08	5.39	
High	54	40	7.30±13.9	4.55	0.858
Grains					
Cereals					
Low	26	19	4.16±2.42	3.35	
High	109	81	7.81±10.9	5.55	0.093
Dark breads					
Low	79	59	7.09±11.6	4.97	
High	56	41	7.12±7.08	5.12	0.986
White breads, bagels, or crackers					
Low	52	39	6.33±7.21	4.30	
High	83	61	7.59±11.3	5.56	0.475
Other					
Cakes, doughnuts, cookies, or pastries					
Low	67	50	7.31±13.5	4.39	
High	68	50	6.91±4.05	5.77	0.816
Candies					
Low	68	50	6.26±6.50	4.57	
High	67	50	7.97±12.5	5.56	0.320

Jellies and jams					
Low	91	67	6.20±5.88	4.65	
High	44	33	8.98±15.1	5.94	0.128
Peanuts, peanut butter, or other nuts					
Low	81	60	7.40±12.4	4.84	
High	54	40	6.66±3.95	5.35	0.671
Salty snacks					
Low	59	44	6.73±6.98	4.83	
High	76	56	7.40±11.8	5.20	0.698
Soft drinks (with sugar)					
Low	101	75	6.56±5.79	4.98	
High	34	25	8.74±17.2	5.20	0.268

[a]TCP was quantified in the urine samples of 135 stay-at-home children in North Carolina and Ohio
[b]Number of children
[c]Percentage of children
[d]Arithmetic mean and standard deviation
[e]Geometric mean
[f]Low equals the group of children that typically consumed a food item < 3 times per week
[g]High equals the group of children that typically consumed a food item $\geq$ 3 times or greater per week

Table 4. Children's weekly consumption of food items and their mean urinary 3-PBA concentrations (ng/mL) by intake group.[a]

Food Item	N[b]	%[c]	Mean±SD[d]	GM[e]	p-Value
Fresh fruits or vegetables					
Apples					
Low[f]	42	61	0.40±0.37	0.29	
High[g]	27	39	0.65±0.71	0.40	0.058
Bananas					
Low	41	59	0.44±0.43	0.31	
High	28	41	0.59±0.67	0.37	0.276
Grapes					
Low	49	71	0.47±0.50	0.32	
High	20	29	0.57±0.63	0.36	0.515
Strawberries					
Low	56	81	0.50±0.54	0.33	
High	13	19	0.50±0.55	0.32	0.991
Carrots					
Low	43	62	0.51±0.55	0.33	
High	26	38	0.49±0.53	0.33	0.874
Potatoes					
Low	47	68	0.44±0.42	0.31	
High	22	32	0.63±0.73	0.38	0.164
String beans or green beans					
Low	58	84	0.48±0.51	0.32	
High	11	16	0.64±0.67	0.38	0.363
Tomatoes					
Low	59	86	0.47±0.52	0.32	
High	10	14	0.66±0.67	0.42	0.303
Processed fruits or vegetables					
Canned or bottled fruit					
Low	51	74	0.46±0.50	0.31	
High	18	26	0.62±0.64	0.37	0.275
Canned or bottled vegetables					
Low	41	59	0.45±0.49	0.30	
High	28	41	0.58±0.61	0.37	0.342
Frozen vegetables					
Low	46	67	0.45±0.47	0.31	
High	23	33	0.61±0.65	0.37	0.230
French fries and fried potatoes					
Low	56	81	0.52±0.58	0.34	
High	13	19	0.40±0.33	0.29	0.452
Fortified fruit drinks					
Low	45	65	0.58±0.59	0.38	
High	24	35	0.36±0.41	0.25	0.109
Fruit juices					
Low	22	32	0.64±0.64	0.41	
High	47	68	0.44±0.48	0.30	0.153
Meats					
Chicken or turkey meats					

Low	53	77	0.41±0.39	0.29	
High	16	23	0.79±0.81	0.48	0.013
Ham or lunch meats					
Low	51	74	0.44±0.48	0.30	
High	18	26	0.67±0.67	0.44	0.133
Dairy					
Butter					
Low	46	67	0.41±0.42	0.29	
High	23	33	0.68±0.70	0.42	0.052
Cheeses and cheese spreads					
Low	19	28	0.47±0.59	0.29	
High	50	72	0.51±0.52	0.34	0.775
Eggs					
Low	58	84	0.49±0.51	0.32	
High	11	16	0.55±0.69	0.35	0.743
Ice cream					
Low	45	65	0.46±0.53	0.30	
High	24	35	0.57±0.57	0.39	0.422
Margarine					
Low	36	52	0.62±0.67	0.38	
High	33	48	0.38±0.32	0.28	0.067
Yogurt					
Low	47	68	0.43±0.40	0.30	
High	22	32	0.66±0.75	0.40	0.092
2% Milk or beverages with 2% milk					
Low	34	49	0.55±0.60	0.36	
High	35	51	0.45±0.47	0.30	0.421
Skim milk, 1% milk, or butter milk					
Low	54	78	0.46±0.45	0.32	
High	15	22	0.66±0.79	0.38	0.195
Whole milk or beverages with whole milk					
Low	44	64	0.54±0.62	0.32	
High	25	36	0.44±0.37	0.33	0.485
Grains					
Cereals					
Low	10	14	0.36±0.30	0.27	
High	59	86	0.53±0.57	0.34	0.361
Dark breads					
Low	38	55	0.54±0.64	0.32	
High	31	45	0.45±0.39	0.33	0.463
White breads, bagels, or crackers					
Low	26	38	0.54±0.45	0.39	
High	43	62	0.47±0.59	0.30	0.622
Other					
Cakes, doughnuts, cookies, or pastries					
Low	33	48	0.50±0.56	0.33	
High	36	52	0.50±0.53	0.33	0.961
Candies					
Low	38	55	0.45±0.47	0.30	
High	31	45	0.56±0.62	0.36	0.412

Jellies and jams					
Low	45	65	0.52±0.60	0.34	
High	24	35	0.46±0.52	0.30	0.642
Peanuts, peanut butter, or other nuts					
Low	38	55	0.49±0.53	0.33	
High	31	45	0.51±0.56	0.33	0.858
Salty snacks					
Low	29	42	0.50±0.55	0.33	
High	40	58	0.50±0.54	0.33	0.955
Soft drinks (with sugar)					
Low	55	80	0.50±0.53	0.33	
High	14	20	0.52±0.58	0.33	0.907

[a]3-PBA was only quantified in the urine samples of 69 stay-at-home children in Ohio
[b]Number of children
[c]Percentage of children
[d]Arithmetic mean and standard deviation
[e]Geometric mean
[f]Low equals the group of children that typically consumed a food item < 3 times per week
[g]High equals the group of children that typically consumed a food item ≥ 3 times or greater per week

Table 5. Children's weekly consumption of food items and their mean urinary 2,4-D concentrations (ng/mL) by intake group.[a]

Food Item	N[b]	%[c]	Mean±SD[d]	GM[e]	p-Value
Fresh fruits or vegetables					
Apples					
Low[f]	85	63	1.21±1.79	0.76	
High[g]	50	37	1.17±1.52	0.77	0.894
Bananas					
Low	84	62	1.18±1.63	0.77	
High	51	38	1.23±1.79	0.76	0.884
Grapes					
Low	98	73	1.25±1.70	0.81	
High	37	27	1.06±1.67	0.66	0.567
Strawberries					
Low	111	82	1.26±1.84	0.78	
High	24	18	0.90±0.51	0.72	0.344
Carrots					
Low	94	70	1.23±1.96	0.73	
High	41	30	1.13±0.79	0.85	0.757
Potatoes					
Low	115	73	1.19±1.59	0.73	
High	20	27	1.22±1.95	0.86	0.913
String beans or green beans					
Low	112	85	1.18±1.80	0.72	
High	23	15	1.29±0.80	1.05	0.797
Tomatoes					
Low	98	83	1.23±1.81	0.77	
High	37	17	1.05±0.88	0.72	0.648
Processed fruits or vegetables					
Canned or bottled fruit					
Low	102	76	1.24±1.89	0.76	
High	33	24	1.06±0.81	0.79	0.587
Canned or bottled vegetables					
Low	82	61	1.04±1.23	0.73	
High	53	39	1.44±2.21	0.82	0.175
Frozen vegetables					
Low	91	67	1.31±1.99	0.79	
High	44	33	0.96±0.71	0.71	0.255
French fries or fried potatoes					
Low	109	81	1.27±1.86	0.78	
High	26	19	0.90±0.52	0.70	0.324
Fortified fruit drinks					
Low	90	67	1.23±1.60	0.80	
High	45	33	1.14±1.88	0.69	0.769
Fruit juices					
Low	41	30	1.16±1.75	0.68	
High	94	70	1.21±1.67	0.80	0.872
Meats					
Chicken or turkey meats					

Low	112	83	1.20±1.82	0.74	
High	23	17	1.18±0.81	0.92	0.954
Ham or lunch meats					
Low	99	73	1.22±1.68	0.78	
High	36	27	1.14±1.73	0.72	0.822
Dairy					
Butter					
Low	88	65	1.06±1.57	0.68	
High	47	35	1.46±1.88	0.95	0.186
Cheeses and cheese spreads					
Low	43	32	1.43±2.41	0.75	
High	92	68	1.09±1.22	0.77	0.273
Eggs					
Low	114	84	1.25±1.79	0.81	
High	21	16	0.90±1.00	0.55	0.375
Ice cream					
Low	93	69	1.05±1.39	0.71	
High	42	31	1.52±2.20	0.91	0.139
Margarine					
Low	66	49	1.39±2.03	0.79	
High	69	51	1.01±1.26	0.74	0.194
Yogurt					
Low	92	68	1.33±2.00	0.80	
High	43	32	0.92±0.59	0.69	0.187
2% Milk or beverages with 2% milk					
Low	72	53	1.26±1.92	0.74	
High	63	47	1.13±1.39	0.79	0.657
Skim milk, 1% milk, or butter milk					
Low	104	77	1.15±1.63	0.74	
High	31	23	1.36±1.88	0.84	0.541
Whole milk or beverages with whole milk					
Low	81	60	1.21±1.65	0.77	
High	54	40	1.19±1.76	0.75	0.947
Grains					
Cereals					
Low	26	19	0.71±0.83	0.50	
High	109	81	1.31±1.82	0.85	0.104
Dark breads					
Low	79	59	1.08±1.47	0.74	
High	56	41	1.36±1.96	0.81	0.349
White breads, bagels, or crackers					
Low	52	39	1.48±2.23	0.85	
High	83	61	1.02±1.22	0.72	0.128
Other					
Cakes, doughnuts, cookies, or pastries					
Low	67	50	1.19±1.96	0.69	
High	68	50	1.21±1.39	0.85	0.939
Candies					
Low	68	50	1.08±1.53	0.73	
High	67	50	1.32±1.84	0.80	0.409

Jellies and jams					
Low	91	67	1.29±2.00	0.76	
High	44	33	1.02±0.71	0.78	0.391
Peanuts, peanut butter, or other nuts					
Low	81	60	1.33±2.10	0.76	
High	54	40	1.00±0.71	0.77	0.268
Salty snacks					
Low	59	44	1.14±1.66	0.73	
High	76	56	1.24±1.73	0.80	0.747
Soft drinks (with sugar)					
Low	101	75	1.30±1.88	0.82	
High	34	25	0.90±0.85	0.63	0.230

[a]2,4-D was quantified in the urine samples of 135 stay-at-home children in North Carolina and Ohio
[b]Number of children
[c]Percentage of children
Arithmetic mean and standard deviation
eGeometric mean
[f]Low equals the group of children that typically consumed a food item < 3 times per week
[g]High equals the group of children that typically consumed a food item ≥ 3 times or greater per week

Table S-1. Weekly consumption of food items by the preschool children.

Food item	Number of children[a]				
	Never	Rarely (<1 time)	Low (1-2 times)	Moderate (3-5 times)	High ($\geq$ 5 times)
Fresh Fruit					
Apples	19	22	44	44	7
Bananas	10	16	59	42	9
Cantaloupes, honeydews, or watermelons	38	44	37	14	3
Grapefruits	120	9	5	0	2
Grapes	13	39	47	26	11
Oranges or tangerines	39	41	41	9	6
Peaches or nectarines	61	31	36	6	2
Pears	80	26	19	9	2
Strawberries	28	40	43	19	6
Fresh Vegetables					
Broccoli	58	34	34	10	0
Cabbage	96	34	5	0	1
Carrots	25	30	40	28	13
Cauliflower	102	23	10	0	1
Corn	21	35	63	17	0
Mustard greens, turnip greens, collards, or spinach	105	21	7	3	0
Potatoes	19	23	57	34	3
String beans or green beans	51	33	32	18	2
Tomatoes	65	20	27	20	4
Other Fruits or Vegetables					
Canned or bottled fruits	33	33	36	24	10
Canned or bottled vegetables	29	14	39	37	17
Frozen vegetables	20	24	48	30	14
Fortified fruit drinks (e.g., Hi-C)	51	18	21	18	28
French fries or fried potatoes	11	26	73	21	5
Fruit juices (100%)	15	7	20	20	74
Meats					
Bacon	52	44	36	4	0
Beef steak or roasts	39	48	42	7	0
Beef stew or pot pie	77	43	14	2	0
Chicken or turkey (baked, roasted, stewed, broiled)	15	24	74	16	7
Fried chicken	56	51	21	8	0
Fried fish or fish sandwich	80	38	18	0	0

Ham or lunch meats	27	18	55	30	6
Hamburgers, cheeseburgers, or meatloaf	58	40	33	5	0
Hot dogs	24	45	52	14	1
Liver (including chicken liver)	129	7	0	0	0
Other fish (broiled or baked)	95	31	9	1	0
Pork (including chops and roast)	38	61	35	2	0
Sausage	71	37	22	3	3
Dairy					
Butter	59	9	21	17	30
Cheeses and cheese spreads	13	5	25	55	38
Eggs	27	20	68	20	1
Frozen yogurt	109	18	4	3	2
Ice cream	9	31	54	31	11
Margarine	45	4	17	35	35
Yogurt	39	19	35	29	14
2% milk or beverages with 2% milk (including on cereal)	67	3	3	3	60
Skim milk, 1% milk, or butter milk (including on cereal)	93	4	8	3	28
Whole milk or beverages with whole milk (including on cereal)	79	2	1	7	47
Grains					
Cereals	5	5	16	56	54
Cooked cereals	65	24	30	14	3
Dark bread (e.g., whole wheat, rye or pumpernickel)	57	10	13	22	34
Corn bread, corn muffins, or corn tortillas	58	51	24	2	1
Rice	25	39	60	10	2
Spaghetti, lasagna, or other pasta (with tomato sauce)	10	31	83	11	1
White breads, bagels, or crackers	16	9	27	21	63
Other					
Cakes, doughnuts, cookies, or pastries	13	5	41	37	40
Candies	11	11	46	42	26
Canned soups	49	34	45	7	1

Honey	101	19	9	4	3
Jellies or jams	33	15	44	26	18
Peanuts, peanut butter, or other nuts	19	16	47	28	26
Pies	114	19	3	0	0
Pizza	4	56	72	3	1
Salty snacks (e.g., chips, popcorn, or pretzels)	9	7	44	44	32
Soft drinks (with sugar)	53	16	33	15	19
Soft drinks (sugar-free)	114	2	14	3	3

[a]One child was excluded from the data analyses because they did not have a valid urine sample.