Abstract: Organophosphorus (OP) insecticides were among the first pesticides that EPA reevaluated as part of the Food Quality Protection Act of 1996. Our goal was to assess exposure to OP insecticides in the U.S. general population over a six-year period. We analyzed 7,456 urine samples collected as part of three two-year cycles of the National Health and Nutrition Examination Survey (NHANES) from 1999–2004. We measured six dialkylphosphate metabolites of OP pesticides to assess OP pesticide exposure. In NHANES 2003–2004, dimethylthiophosphate was detected most frequently with median and 95th percentile concentrations of 2.03 and 35.3 μg/L, respectively. Adolescents were two to three times more likely to have diethylphosphate concentrations above the 95th percentile estimate of 15.5 μg/L than adults and senior adults. Conversely, for
dimethyldithiophosphate, senior adults were 3.8 times and 1.8 times more likely to be above the 95th percentile than adults and adolescents, respectively, while adults were 2.1 times more likely to be above the 95th percentile than the adolescents. Our data indicate that the most vulnerable segments of our population—children and older adults—have higher exposures to OP pesticides than other population segments. However, according to DAP urinary metabolite data, exposures to OP pesticides have declined during the last six years at both the median and 95th percentile levels.

**Keywords:** NHANES; urine; organophosphorus; pesticide; dialkylphosphate

### 1. Introduction

In 1999, an estimated 830 million pounds of pesticides were used in the United States [1]. Organophosphorus (OP) pesticides are among the most common in the United States and are applied in both agricultural and residential settings. Currently, 34 OP pesticides are registered with the U.S. Environmental Protection Agency (EPA) for use in the United States [2]. According to the EPA, 60 million pounds of OP pesticides were used on corn, cotton, other field crops such as canola and alfalfa, fruits, nuts, and vegetables in 1999 [1]. Because of their widespread use on food crops, the EPA established food tolerance levels to prevent hazardous exposures in the diet as mandated by the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA) [2].

The Food Quality Protection Act of 1996 (FQPA) [3] amended FIFRA to include cumulative and aggregate exposure risk assessments in derivative food tolerance levels. In addition, special consideration was to be given to exposures among children. Because of their common mode of toxicity as potent acetyl cholinesterase inhibitors, the EPA selected OP insecticides as the first class of pesticides for reassessing food tolerances. The reassessment of OP pesticides was completed in August 2006. The EPA estimated that residential use of OP pesticides decreased by 20 million pounds annually, largely because of the voluntary cancellation of post-construction residential registrations on chlorpyrifos and diazinon in 2000 and 2002 [2]. Although the phase-out approach to eliminating residential uses of chlorpyrifos and diazinon occurred over a several month period, some reports suggest that use of OP insecticides declined shortly after the announcement of the cancellations [4].

Many biomonitoring studies evaluating occupational [5-9], para-occupational [8,10-13], and background exposures [4,14-25] have focused on OP pesticides. Because exposure typically occurs by multiple routes and dominant routes of exposure vary, assessing exposure to OP pesticides is not a trivial process. In many epidemiologic studies, exposure markers in biological samples have been measured to estimate the absorbed dose [10,26-30]. One of the most common measures of OP pesticide exposure is quantifying six common urinary dialkylphosphate (DAP) metabolites. This measure provides no specific information about the pesticide to which a person was exposed and it may represent exposure to both the pesticide itself and its environmental degradate. However, urinary DAP metabolite measurements may provide useful information about cumulative exposure to OP pesticides as a class because about 75% of the EPA-registered OP pesticides form one to three of these six DAP metabolites.
We reported urinary DAP metabolite concentrations among 7,456 persons aged 6–59 years old from 1999–2000 and among persons aged six years and older from 2001–2004. Specifically, we reported urinary concentrations of dimethylphosphate (DMP), diethylphosphate (DEP), dimethyldithiophosphate (DMTP), diethylthiophosphate (DETP), dimethylidithiophosphate (DMDTP), and diethylidithio-phosphate (DEDTP) (Figure 1). Our data were collected from NHANES 1999–2004 during three, two-year collection cycles and are representative of the civilian, non-institutionalized U.S. population, stratified by age, sex, and race/ethnicity.

**Figure 1.** Structures of dialkylphosphate metabolites of organophosphorus pesticides.

![Structures of dialkylphosphate metabolites](image)

**2. Experimental**

The National Center for Health Statistics of the Centers for Disease Control and Prevention’s (NCHS/CDC) National Health and Nutrition Examination Survey (NHANES) is designed to measure the health and nutrition status of the civilian, noninstitutionalized U.S. population [31]. NHANES participants were selected based on their age, sex, and racial/ethnic background through a complex statistical process using the most current census information. For this study, we analyzed urine samples from 7,456 people, an approximate one-third random subset of participants in NHANES 1999–2004. Urine specimens were collected from participants ages six years and older during one of the examination periods conducted three times daily. Sociodemographic information and medical histories of survey participants and their families were collected during the household interview.

The National Centers for Health Statistics Institutional Review Board reviewed and approved the study protocols from NHANES 1999–. Informed written consent was obtained from all participants; informed written consent for participants <18 years of age was obtained from parents or guardians.
2.1. Laboratory Methods

During the physical examinations, “spot” urine specimens were collected from participants, aliquoted, and stored cold (2 °C–4 °C) or frozen until shipment. To determine urinary creatinine concentrations, we used an automated colorimetric method based on a modified Jaffe reaction [32] on a Beckman Synchron AS/ASTRA clinical analyzer (Beckman Instruments, Inc., Brea, CA, USA) at the University of Minnesota’s Medical Center. Samples collected for OP pesticide measurements were shipped on dry ice to CDC’s National Center for Environmental Health. Urine samples were analyzed for DAP metabolites of OP pesticides using the methods of Bravo et al. [33,34]. The 2002 method was used to analyze samples collected in NHANES III and NHANES 1999–2000. The 2004 method was used for subsequent analyses. Both methods were shown to agree using a Pearson correlation analysis (r > 0.97, p < 0.001 for all analytes); a Bland-Altman plot (Figure 2) showed good agreement with no systematic bias between the two methods (mean percent difference <0.5% for all analytes). Briefly, 4 mL of urine were spiked with an isotopically-labeled internal standard mixture, then concentrated to dryness using an azeotropic codistillation with acetonitrile or lyophilization. The dried residue was dissolved in acetonitrile, and the DAPs were derivatized to their respective chloropropyl esters using 1-chloro-3-iodopropane and potassium carbonate. The solution containing the chloropropyl esters was concentrated and then analyzed using gas chromatography-positive chemical ionization-tandem mass spectrometry. The DAP metabolites were quantified using isotope-dilution calibration. The relative standard deviations ranged from 3–18% for these analyses. The analytic limits of detection (LODs) were 0.5–0.58 µg/L for dimethylphosphate (DMP), 0.18–0.5 µg/L for dimethyldithiophosphate (DMTP), 0.08–0.2 µg/L for dimethyldithiophosphate (DMDTP), 0.2 µg/L for diethylphosphate (DEP), 0.09–0.1 µg/L for diethylthiophosphate (DETP), and 0.05–0.1 µg/L for diethyldithiophosphate (DEDTP). The LOD differences were considered when making general observations regarding the frequencies of detection. Both laboratories and methods were certified according to the Clinical Laboratory Improvement Amendment [35] guidelines.

**Figure 2.** Bland-Altman plot of percent difference between two measurement methods for dimethylthiophosphate. The mean percent difference is −0.32%.
2.2. Statistical Analysis

SAS software (version 9.1.3, SAS Institute, Cary, NC, USA) and SUDAAN software (version 9.0.1, Research Triangle Institute, Research Triangle Park, NC, USA) produced estimates, regression coefficients, and related standard errors. SUDAAN uses sample weights to account for the unequal probability of selection. Geometric means were calculated for analytes detected in ≥60% of the samples. For a concentration below the LOD, we used a value equal to the LOD divided by the square root of 2 [36,37]. Spearman correlation coefficients were calculated in SAS.

For the race/ethnicity variable, we included every group in the calculation for total estimates, but only used Mexican Americans (MA), non-Hispanic blacks (NHB) and non-Hispanic whites (NHW) for descriptive analyses and for regression analyses. We stratified age by 6–11, 12–19, 20–39, 40–59 years old and 60 years and older for the geometric mean and the various percentiles.

We used analysis of covariance to examine the influence of demographic variables on the log-transformed urine concentrations for the analytes with ≥70% detection frequency (i.e., DMTP). The least square geometric mean (LSGM) was calculated from the regression model. For multiple regression analyses, the variables included in the initial model were age (continuous), sex (male or female), race/ethnicity (MA, NHB, and NHW) and log-transformed creatinine. We also included an age-squared term in the model for DMTP because the unadjusted geometric mean by age groups indicated a curvilinear relationship between age and the urinary DMTP concentration. We assessed all possible two-way interaction terms in the model. To evaluate the relationship between the log-transformed concentration of all analytes and age, we changed the continuous age to a categorical age-decades variable in the model to generate a bar chart of LSGM concentrations.

To reach the final model, we used backward elimination with a threshold of \( p < 0.05 \) for retaining the variable in the model, using Satterwaite-adjusted \( F \) statistics. We evaluated for potential confounding by adding each of the excluded variables back into the final model, one-by-one, and examining changes in the \( \beta \) coefficients of the statistically significant main effects. If adding one of these excluded variables caused a change in a \( \beta \) coefficient by \( \geq 10\% \), the variable was added back into the model. For NHANES 2003–2004 data, we conducted weighted univariate and multiple logistic regression to examine the chance of any demographic group (with sex (male, female), age (6–11, 12–19, 20–59, and 60 years and older), and race/ethnicity (MA, NHB, NHW) having a DAP concentration above the total population 95th percentile.

3. Results and Discussion

For 2003–2004, a total of 2,494 samples were available for statistical analysis. Geometric means could not be calculated for any analyte but DMTP because all the other analytes had a detection frequency of less than 60%. The geometric mean concentration and various percentiles stratified by age groups (6–11, 12–19, 20–39, 40–59, and ≥60 years), race/ethnicity (MA, NHB, NHW) and sex (male, female) for all three NHANES cycles are given in Table 1–Table 6. We analyzed 7,456 samples during the six-year period (1999–2004). No significant differences were noted in analytic concentrations from urine samples collected at different times during the day.
Table 1. Dimethylphosphate. Geometric mean and selected percentiles of urine concentrations for the U.S. population from the National Health and Nutrition Examination Surveys. Units are μg/L for unshaded values and μg/g creatinine for shaded values.

<table>
<thead>
<tr>
<th>Survey years</th>
<th>Geometric mean (95% confidence limit)</th>
<th>Sample size</th>
<th>Weighted Detection Percent</th>
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<tr>
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<td>Selected percentile (95% confidence interval)</td>
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<td>Geometric mean (95% confidence limit)</td>
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<td>50th</td>
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<td>Total</td>
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<tr>
<td>1999–2000</td>
<td>*</td>
<td>0.740 (&lt;LOD–1.40)</td>
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<td>2001–2002</td>
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<td>&lt;LOD</td>
<td>3.00 (2.69–3.59)</td>
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<tr>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>3.86 (3.50–5.18)</td>
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Age group

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<tr>
<td></td>
<td>50th</td>
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<td>90th</td>
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<td>6–11 years</td>
<td>1.58 (1.15–2.18)</td>
<td>1.10 (0.590–2.20)</td>
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Table 1. Cont.

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<td>75th: 9.90 (6.10–18.0)</td>
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<td>90th: 22.0 (12.0–29.0)</td>
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<td>20–59 years</td>
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<td>50th: 2.28 (1.72–2.80)</td>
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<td>75th: 7.78 (4.16–14.4)</td>
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<td>90th: 16.0 (8.70–35.3)</td>
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<td>95th: 20.9 (12.5–26.8)</td>
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<td>60 years and older</td>
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<td>2001–2002</td>
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<td>0.760 (LOD–1.12)</td>
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<td>90th: 14.6 (10.4–16.4)</td>
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<td>95th: 15.1 (10.8–17.5)</td>
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<td>2001–2002</td>
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<td>75th: 8.93 (6.79–10.7)</td>
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<td>90th: 14.4 (9.63–19.1)</td>
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<td>95th: 15.0 (11.9–18.6)</td>
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<td>2001–2002</td>
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<td>1.67 (LOD–2.33)</td>
<td>529</td>
<td>57</td>
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<tr>
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<td>1.45 (LOD–2.00)</td>
<td>498</td>
<td>48.8</td>
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<td>90th: 14.3 (11.7–18.5)</td>
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<td>95th: 16.7 (12.3–23.4)</td>
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<th>Weighted Detection Percent</th>
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<td>2.61 (2.35–3.18)</td>
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<td><strong>Race/ethnicity</strong></td>
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<tr>
<td>Non–Hispanic whites</td>
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<td>1.00 (&lt;LOD–1.71)</td>
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<td>9.12 (7.82–11.7)</td>
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<td>&lt;LOD</td>
<td>5.00 (4.03–6.48)</td>
<td>10.1 (9.65–13.8)</td>
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<td>Survey years</td>
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<td>Selected percentile (95% confidence interval)</td>
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<td>Weighted Detection Percent</td>
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<tr>
<td>1999–2000</td>
<td>&lt;LOD</td>
<td>3.15 (1.97–4.30) 8.73 (5.89–13.3) 15.8 (10.0–21.6)</td>
<td>595</td>
<td>48.7</td>
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<tr>
<td>2001–2002</td>
<td>&lt;LOD</td>
<td>2.77 (2.50–3.66) 8.00 (6.71–9.94) 12.9 (11.3–16.9)</td>
<td>948</td>
<td>46.5</td>
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<td>2003–2004</td>
<td>&lt;LOD</td>
<td>3.89 (3.98–6.74) 9.67 (9.75–12.7) 14.6 (13.0–19.6)</td>
<td>756</td>
<td>49.7</td>
</tr>
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</table>

| Mexican Americans 1999–2000 | 1.10 (<LOD–1.80) 3.80 (2.70–5.10) 9.60 (6.00–15.0) 16.0 (10.0–27.0) | 672         | 57.5                       |
| 2001–2002 | 0.670 (<LOD–1.52) 3.24 (2.45–4.42) 9.23 (7.14–10.7) 14.4 (11.0–20.6) | 678         | 50.7                       |
| 2003–2004 | <LOD                                | 4.37 (3.15–6.88) 10.3 (6.92–17.8) 23.3 (9.61–32.5) | 498         | 46.4                       |

| Non–Hispanic blacks 1999–2000 | 1.42 (1.16–1.74) 1.00 (0.620–1.50) 3.60 (2.54–5.45) 9.41 (7.69–11.5) 16.7 (11.7–24.3) | 672         | 57.5                       |
| 2001–2002 | 0.910 (<LOD–1.33) 3.03 (2.52–3.78) 8.03 (6.24–11.1) 14.6 (11.4–16.2) | 678         | 50.7                       |
| 2003–2004 | <LOD                                | 4.12 (3.44–6.30) 12.3 (7.39–17.6) 19.5 (15.3–21.8) | 497         | 46.4                       |

<LOD means less than the limit of detection.
* Not calculated. Proportion of results below limit of detection was too high to provide a valid result.
Table 2. Diethylphosphate. Geometric mean and selected percentiles of urine concentrations for the U.S. population from the National Health and Nutrition Examination Surveys. Units are μg/L for unshaded values and μg/g creatinine for shaded values.

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<td><strong>Total</strong></td>
<td>1999–2000 1.03 (0.670–1.58)</td>
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<td>2003–2004 *</td>
<td>&lt;LOD 4.54 (3.57–5.83) 10.2 (9.00–11.5) 15.7 (13.5–16.9)</td>
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<td>1999–2000 0.924 (0.608–1.41)</td>
<td>0.920 (0.570–1.40) 2.73 (1.68–4.60) 7.94 (4.40–12.2) 12.2 (8.00–19.6)</td>
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<td>6–11 years</td>
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<td>72.9</td>
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<td>0.290 (&lt;LOD–1.04) 3.45 (2.41–4.47) 9.56 (6.44–17.8) 20.0 (9.44–38.2)</td>
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<td>2003–2004 *</td>
<td>&lt;LOD 5.13 (1.84–6.64) 10.9 (7.50–13.8) 16.1 (10.4–18.5)</td>
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<td>1999–2000 1.43 (0.870–2.34)</td>
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<th>Selected percentile (95% confidence interval)</th>
<th>Sample size</th>
<th>Weighted Detection Percent</th>
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<td>12–19 years</td>
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<td>0.860 (0.500–1.35) 2.66 (1.53–4.95) 7.37 (4.32–12.1) 12.1 (8.00–16.7)</td>
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<td>1999–2000 1.11 (0.717–1.73)</td>
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<td>1999–2000 0.981 (0.579–1.66)</td>
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<LOD means less than the limit of detection.
* Not calculated. Proportion of results below limit of detection was too high to provide a valid result.
Table 3. Dimethylthiophosphate. Geometric mean and selected percentiles of urine concentrations for the U.S. population from the National Health and Nutrition Examination Surveys. Units are μg/L for unshaded values and μg/g creatinine for shaded values.

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<th>Selected percentile (95% confidence interval)</th>
<th>Sample size</th>
<th>Weighted Detection Percent</th>
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<td>95th</td>
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<tr>
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<td>1.64 (1.22–2.20)</td>
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<tr>
<td>Survey years</td>
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<td><strong>12–19 years</strong></td>
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<td>1.52 (1.18–1.82) 4.38 (3.29–5.66) 13.3 (9.94–20.5) 26.5 (15.5–36.0)</td>
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<td><strong>60 years and older</strong></td>
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Table 3. Cont.

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<th>Selected percentile (95% confidence interval)</th>
<th>Sample size</th>
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<td>17.3 (10.1–25.0)</td>
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<LOD means less than the limit of detection.

* Not calculated. Proportion of results below limit of detection was too high to provide a valid result.
Table 4. Diethylthiophosphate. Geometric mean and selected percentiles of urine concentrations for the U.S. population from the National Health and Nutrition Examination Surveys. Units are μg/L for unshaded values and μg/g creatinine for shaded values.

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<th>Survey years</th>
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<th>Selected percentile (95% confidence interval)</th>
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<th>Weighted Detection Percent</th>
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<td>0.760 (0.620–1.10)</td>
<td>1.40 (1.10–1.80)</td>
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<tr>
<td>2001–2002</td>
<td>0.457 (0.353–0.592)</td>
<td>0.570 (0.390–0.880)</td>
<td>1.53 (1.25–1.79)</td>
<td>2.48 (2.22–3.04)</td>
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<tr>
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<td>&lt;LOD</td>
<td>0.830 (0.690–0.950)</td>
<td>1.77 (1.42–2.31)</td>
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<td>1999–2000</td>
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<td>0.250 (&lt;LOD–0.480)</td>
<td>0.710 (0.460–1.07)</td>
<td>1.72 (1.47–2.31)</td>
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<td>2001–2002</td>
<td>0.453 (0.348–0.590)</td>
<td>0.520 (0.330–0.760)</td>
<td>1.33 (1.04–1.66)</td>
<td>2.84 (2.22–3.76)</td>
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<td>0.700 (0.580–0.830)</td>
<td>1.47 (1.16–2.04)</td>
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<td>1999–2000</td>
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<td>0.550 (0.350–0.850)</td>
<td>1.58 (1.33–2.04)</td>
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<td>&lt;LOD</td>
<td>0.820 (0.580–0.970)</td>
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Note: LOD = limit of detection.
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<th>Weighted Detection Percent</th>
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<td>0.550 (0.300–0.730)</td>
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<td>2001–2002</td>
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<td>0.425 (0.303–0.597)</td>
<td>0.740 (0.580–1.10)</td>
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<td>2001–2002</td>
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Table 4. Cont.
Table 4. Cont.

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<th>Selected percentile (95% confidence interval)</th>
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<th>Weighted detection Percent</th>
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<td>75th</td>
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<td>95th</td>
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<td>1999–2000</td>
<td>0.230 (&lt;LOD–0.550)</td>
<td>0.710 (0.390–1.22)</td>
<td>1.88 (1.05–2.58)</td>
<td>2.64 (2.08–3.07)</td>
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<td>2001–2002</td>
<td>0.448 (0.318–0.630)</td>
<td>0.510 (0.270–0.800)</td>
<td>1.38 (1.00–1.88)</td>
<td>3.08 (2.29–4.23)</td>
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<tr>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>0.700 (0.560–0.840)</td>
<td>1.45 (1.06–2.08)</td>
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<td>Mexican Americans</td>
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<td>1999–2000</td>
<td>*</td>
<td>0.560 (&lt;LOD–0.780)</td>
<td>0.840 (0.740–0.980)</td>
<td>1.50 (1.20–1.90)</td>
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<td>0.549 (0.398–0.759)</td>
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<td>1.40 (1.01–1.98)</td>
<td>2.63 (1.98–3.47)</td>
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<td>2003–2004</td>
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<td>0.960 (0.680–1.54)</td>
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<td>Non–Hispanic blacks</td>
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<td>1999–2000</td>
<td>0.343 (0.201–0.584)</td>
<td>0.570 (&lt;LOD–0.750)</td>
<td>0.820 (0.690–1.20)</td>
<td>1.80 (1.30–3.20)</td>
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<td>2001–2002</td>
<td>0.749 (0.592–0.949)</td>
<td>1.18 (0.740–1.49)</td>
<td>1.86 (1.77–2.03)</td>
<td>3.55 (3.01–3.91)</td>
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<td>2003–2004</td>
<td>0.467 (0.382–0.570)</td>
<td>0.450 (&lt;LOD–0.730)</td>
<td>1.09 (0.930–1.26)</td>
<td>2.32 (1.59–2.88)</td>
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<tr>
<td>1999–2000</td>
<td>0.234 (0.136–0.403)</td>
<td>0.310 (&lt;LOD–0.580)</td>
<td>0.720 (0.510–0.850)</td>
<td>1.39 (1.03–2.10)</td>
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<td>2001–2002</td>
<td>0.535 (0.444–0.645)</td>
<td>0.710 (0.550–0.920)</td>
<td>1.43 (1.32–1.60)</td>
<td>2.73 (2.30–2.98)</td>
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<td>2003–2004</td>
<td>0.305 (0.253–0.368)</td>
<td>0.280 (&lt;LOD–0.380)</td>
<td>0.640 (0.540–0.820)</td>
<td>1.41 (0.930–1.79)</td>
</tr>
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</table>

<LOD means less than the limit of detection.

* Not calculated. Proportion of results below limit of detection was too high to provide a valid result.
Table 5. Dimethyldithiophosphate. Geometric mean and selected percentiles of urine concentrations for the U.S. population from the National Health and Nutrition Examination Surveys. Units are μg/L for unshaded values and μg/g creatinine for shaded values.

<table>
<thead>
<tr>
<th>Survey years</th>
<th>Geometric mean (95% confidence limit)</th>
<th>Selected percentile (95% confidence interval)</th>
<th>Sample size</th>
<th>Weighted Detection Percent</th>
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<td></td>
<td>50th</td>
<td>75th</td>
<td>90th</td>
<td>95th</td>
</tr>
<tr>
<td>Total</td>
<td>&lt;LOD</td>
<td>2.30 (1.30–3.90)</td>
<td>13.0 (5.00–17.0)</td>
<td>19.0 (17.0–38.0)</td>
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<tr>
<td></td>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>0.890 (0.210–1.30)</td>
<td>2.49 (1.88–3.40)</td>
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<tr>
<td></td>
<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>0.640 (0.480–0.800)</td>
<td>1.99 (1.38–3.30)</td>
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<tr>
<td></td>
<td>1999–2000 *</td>
<td>&lt;LOD</td>
<td>1.88 (0.970–3.86)</td>
<td>10.1 (5.31–18.3)</td>
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<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>0.670 (0.330–1.08)</td>
<td>2.60 (1.85–3.69)</td>
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<td></td>
<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>0.500 (0.340–0.650)</td>
<td>2.14 (1.21–3.18)</td>
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</table>

Age group

<table>
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<th>6–11 years</th>
<th>Survey years</th>
<th>Geometric mean (95% confidence limit)</th>
<th>Selected percentile (95% confidence interval)</th>
<th>Sample size</th>
<th>Weighted Detection Percent</th>
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<tbody>
<tr>
<td></td>
<td>1999–2000</td>
<td>0.691 (0.425–1.12)</td>
<td>0.740 (0.080–1.80)</td>
<td>4.30 (2.40–8.60)</td>
<td>17.0 (6.90–37.0)</td>
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<td></td>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>1.30 (0.750–2.11)</td>
<td>3.53 (2.20–4.50)</td>
<td>7.33 (4.32–9.74)</td>
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<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>0.900 (0.620–1.14)</td>
<td>2.94 (1.14–5.48)</td>
<td>5.48 (2.94–8.53)</td>
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<td>1999–2000</td>
<td>0.748 (0.474–1.18)</td>
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<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>1.36 (0.800–2.31)</td>
<td>4.10 (2.67–6.24)</td>
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<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>0.960 (0.580–1.57)</td>
<td>3.38 (2.28–6.15)</td>
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Table 5. Cont.

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<th>Survey years</th>
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<th>Selected percentile (95% confidence interval)</th>
<th>Sample size</th>
<th>Weighted Detection Percent</th>
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<td>12–19 years</td>
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<td>(0.350–0.770)</td>
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<td>20–59 years</td>
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<td>(0.620–3.47)</td>
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<td>*</td>
<td>&lt;LOD</td>
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<td>(0.310–0.770)</td>
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<td>(0.240–0.580)</td>
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<td>60 years and older</td>
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<td>5.03</td>
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Table 5. Cont.

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<td>(95% confidence interval)</td>
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<td>2001–2002</td>
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<td>&lt;LOD</td>
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<td>2003–2004</td>
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<td>&lt;LOD</td>
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<td>Females</td>
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<td>2001–2002</td>
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<td>&lt;LOD</td>
<td>0.580 (0.270–0.820)</td>
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<tr>
<td>2003–2004</td>
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<td>0.370 (0.260–0.590)</td>
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<td>1999–2000</td>
<td>*</td>
<td>&lt;LOD</td>
<td>2.00 (0.800–4.00)</td>
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<tr>
<td>2001–2002</td>
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<td>0.820 (0.370–1.43)</td>
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<tr>
<td>2003–2004</td>
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<td>0.560 (0.390–0.790)</td>
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Table 5. Cont.

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<th>Survey years</th>
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<th>Selected percentile (95% confidence interval)</th>
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<th>Weighted Detection Percent</th>
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<td>2003–2004</td>
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<td>0.470</td>
<td>(0.340–0.700)</td>
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<tr>
<td>Mexican Americans</td>
<td>1999–2000 *</td>
<td>0.250 (&lt;LOD–0.870)</td>
<td>1.90</td>
<td>(1.10–3.00)</td>
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<td>2001–2002 *</td>
<td>&lt;LOD</td>
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<td>(0.750–1.37)</td>
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<td>0.730</td>
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<td>1999–2000 *</td>
<td>0.270 (&lt;LOD–0.660)</td>
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<td>Non–Hispanic blacks</td>
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<td>0.330 (&lt;LOD–1.20)</td>
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<td>(1.40–7.00)</td>
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<td>(0.370–0.720)</td>
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<td>0.430</td>
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<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>0.340</td>
<td>(0.260–0.430)</td>
</tr>
</tbody>
</table>

<LOD means less than the limit of detection.

* Not calculated. Proportion of results below limit of detection was too high to provide a valid result.
Table 6. Diethyldithiophosphate. Geometric mean and selected percentiles of urine concentrations for the U.S. population from the National Health and Nutrition Examination Surveys. Units are μg/L for unshaded values and μg/g creatinine for shaded values.

<table>
<thead>
<tr>
<th>Survey years (95% confidence limit)</th>
<th>Geometric mean</th>
<th>Selected percentile (95% confidence interval)</th>
<th>Sample size</th>
<th>Weighted Detection Percent</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Total</strong> 1999–2000 *</td>
<td>0.090 (&lt;LOD–0.140)</td>
<td>0.210 (0.140–0.290)</td>
<td>0.470 (0.380–0.640)</td>
<td>0.870 (0.640–1.10)</td>
</tr>
<tr>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.610 (0.410–0.770)</td>
<td>0.850 (0.700–1.30)</td>
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<tr>
<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.320 (0.170–0.540)</td>
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<td>1999–2000 *</td>
<td>0.070 (&lt;LOD–0.110)</td>
<td>0.200 (0.140–0.290)</td>
<td>0.550 (0.390–0.700)</td>
<td>0.860 (0.670–1.14)</td>
</tr>
<tr>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.580 (0.390–0.750)</td>
<td>1.01 (0.710–1.43)</td>
</tr>
<tr>
<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.410 (0.330–0.510)</td>
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<td><strong>Age group</strong></td>
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<tr>
<td>6–11 years 1999–2000 *</td>
<td>0.090 (&lt;LOD–0.160)</td>
<td>0.190 (0.130–0.280)</td>
<td>0.430 (0.300–0.650)</td>
<td>0.850 (0.470–1.00)</td>
</tr>
<tr>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.630 (0.380–0.870)</td>
<td>0.940 (0.690–1.42)</td>
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<td>2003–2004 *</td>
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<td>&lt;LOD</td>
<td>0.540 (&lt;LOD–0.650)</td>
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<tr>
<td>1999–2000 *</td>
<td>0.100 (&lt;LOD–0.140)</td>
<td>0.190 (0.150–0.270)</td>
<td>0.570 (0.410–0.760)</td>
<td>1.03 (0.570–1.58)</td>
</tr>
<tr>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.780 (0.610–1.12)</td>
<td>1.36 (1.02–1.86)</td>
</tr>
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<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.470 (&lt;LOD–0.970)</td>
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Table 6. Cont.

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<tr>
<th>Survey years</th>
<th>Geometric mean (95% confidence limit)</th>
<th>Selected percentile (95% confidence interval)</th>
<th>Sample size</th>
<th>Weighted Detection Percent</th>
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<tr>
<td>12–19 years</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1999–2000</td>
<td>*</td>
<td>0.080 (&lt;LOD–0.180)</td>
<td>664</td>
<td>53.1</td>
</tr>
<tr>
<td>2001–2002</td>
<td>*</td>
<td>&lt;LOD</td>
<td>822</td>
<td>17.3</td>
</tr>
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<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>717</td>
<td>11.1</td>
</tr>
<tr>
<td>1999–2000</td>
<td>*</td>
<td>0.050 (&lt;LOD–0.080)</td>
<td>664</td>
<td>53.1</td>
</tr>
<tr>
<td>2001–2002</td>
<td>*</td>
<td>&lt;LOD</td>
<td>821</td>
<td>17.3</td>
</tr>
<tr>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>715</td>
<td>11.1</td>
</tr>
<tr>
<td>20–59 years</td>
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<td></td>
</tr>
<tr>
<td>1999–2000</td>
<td>*</td>
<td>0.090 (&lt;LOD–0.130)</td>
<td>814</td>
<td>54.4</td>
</tr>
<tr>
<td>2001–2002</td>
<td>*</td>
<td>&lt;LOD</td>
<td>1118</td>
<td>18.8</td>
</tr>
<tr>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>938</td>
<td>7.51</td>
</tr>
<tr>
<td>1999–2000</td>
<td>*</td>
<td>0.080 (&lt;LOD–0.110)</td>
<td>814</td>
<td>54.4</td>
</tr>
<tr>
<td>2001–2002</td>
<td>*</td>
<td>&lt;LOD</td>
<td>1118</td>
<td>18.8</td>
</tr>
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<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>937</td>
<td>7.51</td>
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<td>60 years and older</td>
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<td>18.5</td>
</tr>
<tr>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>529</td>
<td>6.72</td>
</tr>
<tr>
<td>2001–2002</td>
<td>*</td>
<td>&lt;LOD</td>
<td>497</td>
<td>18.5</td>
</tr>
<tr>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>529</td>
<td>6.72</td>
</tr>
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<td>Survey years</td>
<td>Geometric mean (95% confidence limit)</td>
<td>Selected percentile (95% confidence interval)</td>
<td>Sample size</td>
<td>Weighted Detection Percent</td>
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<tr>
<td></td>
<td>50th</td>
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</tr>
<tr>
<td>Sex</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Males</td>
<td>1999–2000 *</td>
<td>0.090 (&lt;LOD–0.150)</td>
<td>0.220</td>
<td>0.490</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(0.140–0.310)</td>
<td>(0.380–0.680)</td>
<td>(0.680–1.10)</td>
</tr>
<tr>
<td></td>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.600</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.140)</td>
<td>(0.370–0.740)</td>
<td>(0.680–1.03)</td>
</tr>
<tr>
<td></td>
<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.390</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.140)</td>
<td>(&lt;LOD–0.150)</td>
<td>(0.130–0.540)</td>
</tr>
<tr>
<td></td>
<td>1999–2000 *</td>
<td>0.070 (&lt;LOD–0.110)</td>
<td>0.190</td>
<td>0.410</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(0.140–0.230)</td>
<td>(0.340–0.500)</td>
<td>(0.520–0.940)</td>
</tr>
<tr>
<td></td>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.380</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.300)</td>
<td>(0.300–0.650)</td>
<td>(0.580–1.03)</td>
</tr>
<tr>
<td></td>
<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.330</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.260)</td>
<td>(&lt;LOD–0.410)</td>
<td>(0.130–0.540)</td>
</tr>
<tr>
<td>Females</td>
<td>1999–2000 *</td>
<td>0.090 (&lt;LOD–0.130)</td>
<td>0.190</td>
<td>0.460</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(0.110–0.310)</td>
<td>(0.320–0.840)</td>
<td>(0.440–1.40)</td>
</tr>
<tr>
<td></td>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.660</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.460)</td>
<td>(0.460–0.850)</td>
<td>(0.700–1.42)</td>
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<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.240</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.540)</td>
<td>(&lt;LOD–0.700)</td>
<td>(0.130–0.540)</td>
</tr>
<tr>
<td>Race/ethnicity</td>
<td>1999–2000 *</td>
<td>0.090 (&lt;LOD–0.120)</td>
<td>0.220</td>
<td>0.670</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.360)</td>
<td>(0.410–0.870)</td>
<td>(0.660–1.62)</td>
</tr>
<tr>
<td></td>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.700</td>
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<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.490)</td>
<td>(0.490–1.00)</td>
<td>(0.800–1.86)</td>
</tr>
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<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.500</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.540)</td>
<td>(&lt;LOD–0.640)</td>
<td>(0.130–0.540)</td>
</tr>
<tr>
<td>Non–Hispanic whites</td>
<td>1999–2000 *</td>
<td>0.080 (&lt;LOD–0.160)</td>
<td>0.190</td>
<td>0.450</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.290)</td>
<td>(0.310–0.720)</td>
<td>(0.510–1.30)</td>
</tr>
<tr>
<td></td>
<td>2001–2002 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.610</td>
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<tr>
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<td></td>
<td>(&lt;LOD–0.360)</td>
<td>(0.360–0.780)</td>
<td>(0.650–1.36)</td>
</tr>
<tr>
<td></td>
<td>2003–2004 *</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>0.290</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(&lt;LOD–0.540)</td>
<td>(&lt;LOD–0.540)</td>
<td>(0.130–0.540)</td>
</tr>
<tr>
<td>Survey years</td>
<td>Geometric mean (95% confidence limit)</td>
<td>Selected percentile (95% confidence interval)</td>
<td>Sample size</td>
<td>Weighted Detection Percent</td>
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<td>---------------------------</td>
</tr>
<tr>
<td></td>
<td>50th</td>
<td>75th</td>
<td>90th</td>
<td>95th</td>
</tr>
<tr>
<td>1999–2000</td>
<td>*</td>
<td>&lt;LOD (0.070) (&lt;LOD–0.120)</td>
<td>0.200</td>
<td>0.560</td>
</tr>
<tr>
<td>2001–2002</td>
<td>*</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
<tr>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
<tr>
<td>Mexican Americans</td>
<td>1999–2000</td>
<td>0.130 (&lt;LOD–0.171)</td>
<td>0.100 (0.060–0.190)</td>
<td>0.310 (0.230–0.390)</td>
</tr>
<tr>
<td></td>
<td>2001–2002</td>
<td>*</td>
<td>&lt;LOD</td>
<td>0.200 (&lt;LOD–0.520)</td>
</tr>
<tr>
<td></td>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
<tr>
<td>Non–Hispanic blacks</td>
<td>1999–2000</td>
<td>0.116 (0.084–0.161)</td>
<td>0.090 (0.060–0.170)</td>
<td>0.300 (0.190–0.410)</td>
</tr>
<tr>
<td></td>
<td>2001–2002</td>
<td>*</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
<tr>
<td></td>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
<tr>
<td>1999–2000</td>
<td>0.090 (&lt;LOD–0.170)</td>
<td>0.270 (0.140–0.400)</td>
<td>0.560 (0.400–0.830)</td>
<td>0.870 (0.660–1.20)</td>
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<td>2001–2002</td>
<td>*</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
<tr>
<td>2003–2004</td>
<td>*</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
<td>&lt;LOD</td>
</tr>
</tbody>
</table>

<LOD means less than the limit of detection.

* Not calculated. Proportion of results below limit of detection was too high to provide a valid result.
When examining the 2003–2004 data by multiple regression, the final model for DMTP included sex ($p = 0.056$), age (continuous, $p < 0.001$), age-square ($p < 0.01$), race ($p = 0.058$), and log creatinine ($p < 0.0001$). We observed a curvilinear relationship between the log-transformed DMTP concentration and age (Figure 3). We found both a linear increase and positive quadratic trend with age ($\beta$ for linear $= 0.008$, $\beta$ for quadratic $= 0.0005$). In the model where age was treated categorically, the LSGM covariate-adjusted concentration of DMTP was highest among those ages 60 years and older (3.2 µg/L) while the second highest level was among children ages 6–11 years old (2.45 µg/L). NHB participants had the highest LSGM covariate-adjusted concentration of DMTP (3.57 µg/L), which was significantly higher than for NHW (1.97 µg/L, $p = 0.02$). Similarly, NHB had a higher LSGM covariate-adjusted concentration than MA (1.94. µg/L, $p = 0.02$). MA and NHW had similar LSGM covariate-adjusted concentrations for DMTP. The LSGM covariate-adjusted concentration of DMTP for females (2.46 µg/L) was marginally higher than the one for males ($p = 0.055$).

**Figure 3.** A curvilinear relationship was observed between age and the least squares geometric mean (LSGM) dimethylthiophosphate concentrations.

Regarding potential pathways of exposure, we noted that all analytes except DMP and DEDTP were significantly correlated with each other (Table 7). DMTP and DMDTP had the highest significant correlation ($r = 0.59$), whereas the significant correlations between other analytes ranged from 0.06 to 0.39 (all $p$-values $< 0.0001$).
Table 7. Spearman correlation coefficients between dialkylphosphate metabolites of organophosphorus pesticides (correlation, p value, number of observations).

<table>
<thead>
<tr>
<th></th>
<th>DMP</th>
<th>DEP</th>
<th>DMTP</th>
<th>DETP</th>
<th>DMDTP</th>
<th>DEDTP</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dimethylphosphate (DMP)</td>
<td>1.000</td>
<td>0.325</td>
<td>0.391</td>
<td>0.221</td>
<td>0.282</td>
<td>0.029</td>
</tr>
<tr>
<td>Diethylphosphate (DEP)</td>
<td></td>
<td></td>
<td>&lt;.0001</td>
<td>&lt;.0001</td>
<td>&lt;.0001</td>
<td>&lt;.0001</td>
</tr>
<tr>
<td>Dimethylthiophosphate (DMTP)</td>
<td>2,494</td>
<td>2,453</td>
<td>1.000</td>
<td>0.165</td>
<td>0.317</td>
<td>0.135</td>
</tr>
<tr>
<td>Diethylthiophosphate (DETP)</td>
<td></td>
<td></td>
<td>&lt;.0001</td>
<td>&lt;.0001</td>
<td>&lt;.0001</td>
<td>&lt;.0001</td>
</tr>
<tr>
<td>Dimethyldithiophosphate (DMDTP)</td>
<td></td>
<td></td>
<td>2,494</td>
<td>2,453</td>
<td>2,494</td>
<td></td>
</tr>
<tr>
<td>Diethyldithiophosphate (DEDTP)</td>
<td></td>
<td></td>
<td></td>
<td>1.000</td>
<td>0.237</td>
<td>0.171</td>
</tr>
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<td></td>
<td></td>
<td></td>
<td>&lt;.0001</td>
<td>&lt;.0001</td>
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</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2,422</td>
<td>2,386</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1.000</td>
<td>0.129</td>
</tr>
<tr>
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<td></td>
<td></td>
<td></td>
<td>&lt;.0001</td>
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</table>

In examining the likelihood for those participants whose concentrations were above the 95th percentile, urine concentrations of DEP was significantly associated with age (p = 0.045), but not sex and race/ethnicity. Adolescents were 2.96 and 2.4 times more likely to be above the total population 95th percentile than senior adults (60 and older) (odds ratio [OR] [95% CI]: 2.96 [1.7, 5.2]) and other adults (20–59 years old) (OR [95% CI]: 2.4 [1.1, 5.1]), respectively. Similarly, for DMDTP, age was significantly associated with the chance to be above the 95th percentile (p=0.05). Participants aged 60 years and older were 3.8 times more likely to be above the 95th percentile than that for adolescents aged 12–19 years old (OR [95% CI]:3.8 [1.2, 11.9]) and 1.8 times more likely than adults aged 20–59 years old (OR [95% CI]: 1.8 [1.02, 3.2]). Also, adults aged 20-59 years old were 2.1 times more likely than adolescents ages 12–19 years old to be above the 95th percentile (OR [95% CI]: 2.1 [1.0, 4.4]). No differences were found for the chance to be above the 95th percentile in any other age categories or for any other analytes.

During the three two-year NHANES cycles, the 75th, 90th, and 95th percentile estimates for DMDTP and DEDTP in 1999–2000 were 3 to 5 times higher than for the following two NHANES cycles in the total population. This observation was consistent across demographic groups. We also observed a reduction in DETP median concentrations in 2003–2004 from the 2001–2002 and 1999–2000 median estimates; however, upper distribution percentiles were similar with the ones in year 1999–2000 and appeared to be lower as compared to the ones in year 2001–2002.

DAP metabolites of OP pesticides have been measured in a random subset of three NHANES cycles since 1999. These data from 2001–2004 represent the first time reference DAP concentrations have been reported for adults >60 years old. Additionally, these are the first data reported on the sample sets...
collected before and after a voluntary withdrawal of registrations for chlorpyrifos and diazinon. Although the DAP metabolite measurements are not specific for one OP pesticide in particular and a sizeable proportion probably derives from exposure to the preformed DAP metabolite in the environment, these data may be useful. They may serve as an indicator for the maximum potential of OP pesticide exposure over time.

Interestingly, the most vulnerable age groups, children and the elderly, had the highest concentrations of DMTP in 2003–2004, while adolescents and other adults had much lower levels. These data suggest that older adults and children share common behaviors or activities. For example, time spent indoors may contribute to their high urinary DAP concentrations. However, adolescents were more than twice as likely to have concentrations of DEP above the 95th percentile estimate than senior adults and adults. For DMDTP, senior adults were 3.8 times more likely to be above the 95th percentile than adolescents and 1.8 times more likely than adults. Also, adults were marginally more likely than adolescents to be above the 95th percentile. Understanding the predictors of exposure of the upper tail of the distribution is particularly important for regulatory mitigation efforts, so adolescents may be a good subpopulation to target for reducing exposure.

The significant correlation of all of the DAP metabolites in urine suggests common pathways for both exposure and excretion. Because the use of diethyl OP pesticides in applications with dimethyl pesticides is unexpected, the correlation among these likely points to dietary exposures from produce where both groups of OP pesticides were used regularly. The strongest correlation was between DMTP and DMDTP, both potential metabolites of the common OP pesticide malathion.

These data sets collectively suggest that human exposure to OP pesticides has decreased during the last six years. For example, the median and 95th percentile estimates of DMTP concentrations from NHANES 2003–2004 appeared to be one-third less than that found in NHANES 1999–2000 even though the frequency of detection only decreased by about 20%. Urinary concentrations of DETP were detected significantly less in samples collected in 2003–2004 than in those collected in 2001–2002; however, DEP concentrations were largely unaffected. Because DETP can be derived from chlorpyrifos and diazinon, strategies targeting residential pesticide use to reduce exposure may have contributed to the decrease. DEP is common to all diethyl-substituted OP insecticides which perhaps is why we may not have observed a similar decrease.

As noted earlier, the biggest limitation of our study is the lack of specificity of DAP metabolites for a given OP pesticide. According to some estimates, 70% or more of urinary OP metabolite concentrations may be attributable to exposure to the preformed metabolite in the environment, which is not known to be toxic [38-41]. Thus, we caution that the concentrations reported here be used as a maximum possible estimate of OP pesticide exposures. Regardless, reducing the use or tolerance levels of OP pesticides would also result in reduced preformed metabolite concentrations in the environment so that changes in urinary DAP concentrations over time may parallel exposure mitigation strategies demonstrating their effectiveness.

4. Conclusions

We reported data collected from three NHANES survey periods. Our data indicated that the most vulnerable segments of our population—children and senior adults—have a greater risk of exposure to
OP pesticides than other population segments. However, for DEP, adolescents were more than twice as likely to have concentrations greater than the 95th percentile compared with adults. For DMDTP, senior adults were 3.8 times more likely to be above the 95th percentile than adolescents and 1.8 times more likely than adults. Also, adults were marginally more likely than adolescents to be above the 95th percentile. In addition, NHB have higher DMTP concentrations than NHW and MA. Based on the DAP urinary metabolite data, overall exposures to OP pesticides have declined during the last few decades, suggesting that regulatory efforts have been effective.

Acknowledgments

The findings and conclusions in this report are those of the authors and do not necessarily represent the views of the Centers for Disease Control and Prevention.

References and Notes


37. Techniques for imputing values below the LOD have been debated in the literature. We used the imputation technique consistent with CDC’s National Report on Human Exposure to Environmental Chemicals. Other imputation techniques may produce slightly different estimates.


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