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Article

Urinary Concentrations of Dialkylphosphate Metabolites of Organophosphorus Pesticides: National Health and Nutrition Examination Survey 1999–2004

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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

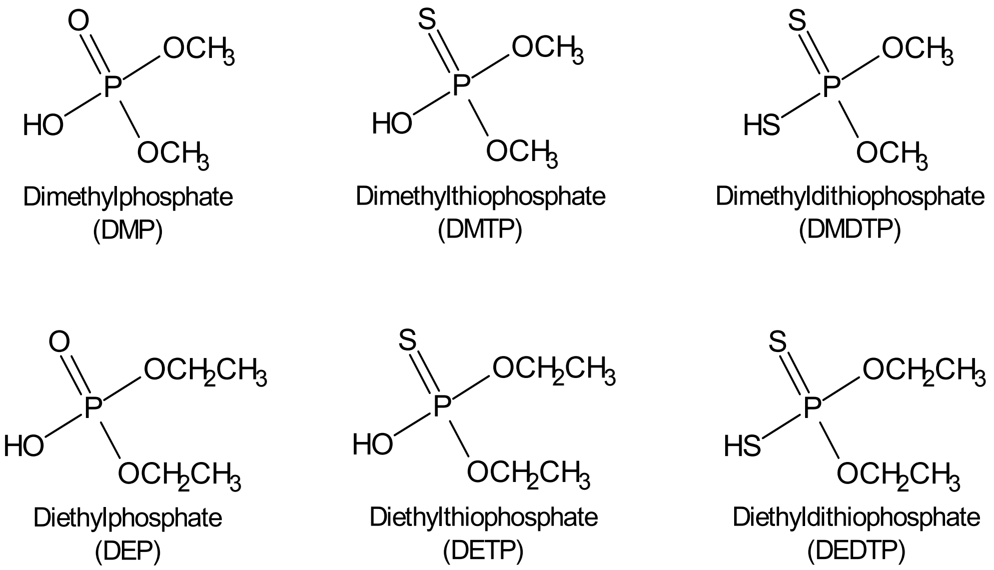
In 1999, an estimated 830 million pounds of pesticides were used in the United States [1]. Organophosphorus (OP) pesticides are among the most common in the United States and are applied in both agricultural and residential settings. Currently, 34 OP pesticides are registered with the U.S. Environmental 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), dimethylthiophosphate (DMTP), diethylthiophosphate (DETP), dimethyldithiophosphate (DMDTP), and diethyldithio-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.



2. Experimental

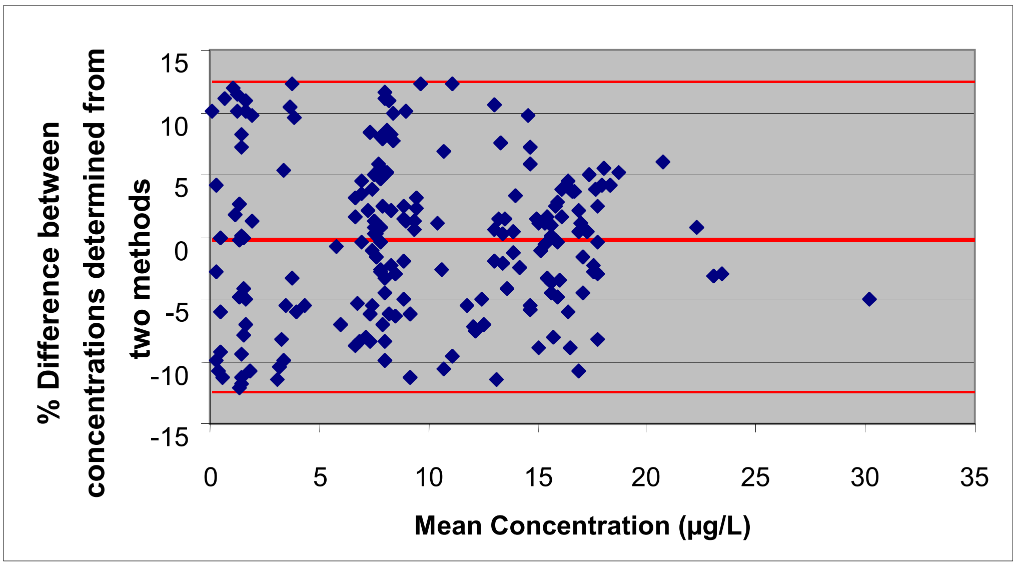
The National Center for Health Statistics of the Centers for Disease Control and Prevention's (NCHS/CDC) National Health and Nutrition Examination Survey (NHANES) is designed to measure the health and nutrition status of the civilian, noninstitutionalized U.S. population [31]. NHANES participants were 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 dimethylthiophosphate (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 β coefficients of the statistically significant main effects. If adding one of these excluded variables caused a change in a β coefficient by ≥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 analyte 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.

	Survey years	Geometric mean	(95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
Total	1999–2000	*		**0.740**	(<LOD–1.40)	**2.90**	(2.10–4.00)	**7.90**	(6.20–8.90)	**14.0**	(10.0–19.0)	1949	52.2
	2001–2002	*		**<LOD**		**3.25**	(2.77–3.67)	**8.22**	(6.95–9.27)	**13.4**	(10.9–15.6)	2519	47.7
	2003–2004	*		**<LOD**		**3.99**	(3.29–4.96)	**9.17**	(7.33–11.9)	**14.8**	(12.4–17.8)	1965	48.7
	1999–2000	*		**0.810**	(<LOD–1.15)	**2.93**	(2.11–3.92)	**8.50**	(6.96–10.4)	**16.1**	(13.3–17.6)	1949	52.2
	2001–2002	*		**<LOD**		**3.00**	(2.69–3.59)	**7.83**	(7.06–9.28)	**12.7**	(11.5–15.0)	2518	47.7
	2003–2004	*		**<LOD**		**3.86**	(3.50–5.18)	**9.54**	(8.78–11.2)	**14.6**	(13.0–18.1)	1962	48.7
Age group													
6–11 years	1999–2000	**1.58**	(1.15–2.18)	**1.10**	(0.590–2.20)	**4.40**	(2.80–6.80)	**10.0**	(9.40–17.0)	**22.0**	(16.0–33.0)	471	61.8
	2001–2002	*		**0.970**	(<LOD–2.00)	**5.04**	(3.31–7.66)	**12.2**	(9.10–15.0)	**18.3**	(12.6–41.7)	576	55
	2003–2004	*		**<LOD**		**4.53**	(3.34–5.96)	**11.0**	(5.62–17.9)	**16.2**	(7.46–28.3)	310	46.5
	1999–2000	**1.71**	(1.29–2.27)	**1.38**	(0.890–2.38)	**4.48**	(2.88–8.20)	**16.7**	(8.21–21.2)	**22.1**	(19.6–30.1)	471	61.8
	2001–2002	*		**1.93**	(<LOD–2.97)	**5.99**	(4.32–8.28)	**12.9**	(9.09–18.8)	**20.6**	(13.2–34.9)	576	55
	2003–2004	*		**<LOD**		**6.87**	(2.33–8.01)	**13.9**	(8.23–17.0)	**19.6**	(13.4–24.9)	310	46.5

Table 1. ***Cont.***

	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
12–19 years	1999–2000	*		**0.670**	(<LOD–1.80)	**3.80**	(2.50–5.00)	**9.90**	(6.10–18.0)	**22.0**	(12.0–29.0)	664	51
	2001–2002	*		**0.670**	(<LOD–1.31)	**4.27**	(3.41–5.35)	**9.27**	(7.80–12.3)	**14.7**	(11.8–21.3)	822	50
	2003–2004	*		**<LOD**		**4.61**	(3.47–6.72)	**10.9**	(7.90–15.0)	**20.9**	(12.5–26.8)	717	51.3
	1999–2000	*		**0.590**	(<LOD–0.950)	**2.28**	(1.72–2.80)	**7.78**	(4.16–14.4)	**16.0**	(8.70–35.3)	664	51
	2001–2002	*		**0.910**	(<LOD–1.25)	**3.29**	(2.77–3.78)	**6.29**	(5.51–7.31)	**9.83**	(7.94–14.2)	821	50
	2003–2004	*		**1.00**	(<LOD–1.68)	**4.16**	(3.37–5.07)	**8.81**	(7.78–10.2)	**12.7**	(10.3–17.7)	715	51.3
20–59 years	1999–2000	*		**0.680**	(<LOD–1.30)	**2.70**	(1.80–3.60)	**6.50**	(5.70–8.10)	**9.60**	(8.80–14.0)	814	50.9
	2001–2002	*		**<LOD**		**2.95**	(2.35–3.41)	**6.95**	(5.80–8.84)	**11.5**	(9.80–14.1)	1121	45.7
	2003–2004	*		**<LOD**		**3.75**	(2.84–4.88)	**8.52**	(8.14–13.5)	**14.1**	(10.8–17.5)	938	46.1
	1999–2000	*		**0.760**	(<LOD–1.12)	**2.88**	(1.89–3.99)	**8.11**	(5.89–10.3)	**14.6**	(10.4–16.4)	814	50.9
	2001–2002	*		**<LOD**		**2.55**	(2.05–3.02)	**6.92**	(5.85–8.00)	**11.5**	(9.38–13.6)	1121	45.7
	2003–2004	*		**<LOD**		**3.77**	(2.72–4.55)	**8.78**	(6.68–10.5)	**13.5**	(12.5–18.1)	937	46.1
60 years and older	2001–2002	*		**0.700**	(<LOD–1.50)	**3.68**	(2.91–4.56)	**8.93**	(6.79–10.7)	**14.4**	(9.63–19.1)	498	48.8
	2003–2004	*		**1.51**	(<LOD–2.35)	**4.16**	(3.52–4.93)	**9.81**	(6.65–12.4)	**15.0**	(11.9–18.6)	529	57
	2001–2002	*		**1.45**	(<LOD–2.00)	**4.30**	(3.44–5.38)	**10.6**	(7.92–12.0)	**14.3**	(11.7–18.5)	498	48.8
	2003–2004	*		**1.67**	(<LOD–2.33)	**5.13**	(3.84–6.79)	**11.7**	(9.43–14.4)	**16.7**	(12.3–23.4)	529	57

Table 1. ***Cont.***

	Survey years	Geometric mean (95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
Sex												
Males	1999–2000	*	**0.670**	(<LOD–1.30)	**2.90**	(2.20–4.00)	**7.50**	(6.00–9.30)	**18.0**	(10.0–23.0)	952	51.5
	2001–2002	*	**<LOD**		**3.40**	(2.49–4.30)	**8.22**	(6.67–10.3)	**12.6**	(11.3–14.7)	1187	48.1
	2003–2004	*	**<LOD**		**3.89**	(2.97–4.89)	**8.14**	(6.34–10.4)	**15.1**	(11.3–20.4)	946	46.5
	1999–2000	*	**0.620**	(<LOD–0.940)	**2.38**	(1.83–3.18)	**7.58**	(4.64–11.6)	**15.2**	(9.84–19.5)	952	51.5
	2001–2002	*	**<LOD**		**2.61**	(2.35–3.18)	**6.25**	(5.51–8.54)	**10.2**	(9.71–12.7)	1187	48.1
	2003–2004	*	**<LOD**		**2.95**	(2.58–4.02)	**7.93**	(6.80–10.4)	**12.5**	(10.5–15.4)	945	46.5
Females	1999–2000	*	**0.790**	(<LOD–1.60)	**2.90**	(1.90–4.20)	**7.80**	(5.70–9.00)	**11.0**	(9.00–15.0)	997	53
	2001–2002	*	**<LOD**		**3.06**	(2.63–3.63)	**8.34**	(6.56–9.63)	**13.7**	(10.6–17.6)	1332	47.3
	2003–2004	*	**<LOD**		**4.18**	(3.29–5.39)	**10.3**	(7.60–12.9)	**14.8**	(13.3–19.0)	1019	50.9
	1999–2000	*	**1.00**	(<LOD–1.71)	**3.63**	(2.30–5.19)	**9.12**	(7.82–11.7)	**16.4**	(11.7–19.7)	997	53
	2001–2002	*	**<LOD**		**3.43**	(2.86–4.50)	**9.00**	(7.83–10.6)	**15.0**	(12.7–17.8)	1331	47.3
	2003–2004	*	**<LOD**		**5.00**	(4.03–6.48)	**10.1**	(9.65–13.8)	**16.0**	(14.8–20.6)	1017	50.9
Race/ethnicity												
Non–Hispanic whites	1999–2000	*	**<LOD**		**2.90**	(1.80–4.30)	**7.90**	(5.90–9.00)	**11.0**	(9.00–18.0)	595	48.7
	2001–2002	*	**<LOD**		**3.01**	(2.49–3.57)	**7.39**	(6.52–9.22)	**12.3**	(10.3–14.4)	948	46.5
	2003–2004	*	**<LOD**		**3.75**	(3.85–6.22)	**8.35**	(8.18–14.8)	**14.6**	(14.0–23.2)	757	49.7

Table 1. ***Cont.***

	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
	1999–2000	*		**<LOD**		**3.15**	(1.97–4.30)	**8.73**	(5.89–13.3)	**15.8**	(10.0–21.6)	595	48.7
	2001–2002	*		**<LOD**		**2.77**	(2.50–3.66)	**8.00**	(6.71–9.94)	**12.9**	(11.3–16.9)	948	46.5
	2003–2004	*		**<LOD**		**3.89**	(3.98–6.74)	**9.67**	(9.75–12.7)	**14.6**	(13.0–19.6)	756	49.7
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
	1999–2000	*		**1.06**	(<LOD–1.55)	**3.89**	(2.54–5.45)	**9.41**	(7.69–11.5)	**16.7**	(11.7–24.3)	672	57.5
	2001–2002	*		**0.920**	(<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**		**4.14**	(2.44–6.30)	**12.3**	(7.39–17.6)	**19.5**	(15.3–21.8)	497	46.4
Non–Hispanic blacks	1999–2000	**1.42**	(1.16–1.74)	**1.00**	(0.620–1.50)	**3.60**	(2.50–5.50)	**8.90**	(6.90–15.0)	**21.0**	(15.0–24.0)	509	59.9
	2001–2002	*		**0.910**	(<LOD–2.29)	**5.45**	(3.81–6.62)	**11.5**	(8.77–14.1)	**19.4**	(14.7–22.8)	695	52.2
	2003–2004	*		**<LOD**		**5.12**	(4.11–6.37)	**10.5**	(8.31–12.0)	**14.3**	(11.7–19.6)	579	49.6
	1999–2000	**0.973**	(0.780–1.21)	**0.690**	(0.530–1.06)	**2.67**	(1.89–3.77)	**7.07**	(5.09–10.9)	**14.0**	(10.6–19.1)	509	59.9
	2001–2002	*		**0.850**	(<LOD–1.34)	**3.36**	(2.74–4.22)	**7.63**	(6.35–9.45)	**13.2**	(10.8–15.0)	694	52.2
	2003–2004	*		**<LOD**		**3.38**	(2.75–3.95)	**6.94**	(4.86–7.43)	**10.8**	(7.46–15.3)	578	49.6

<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.

	Survey years	Geometric mean	(95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
Total	1999–2000	**1.03**	(0.670–1.58)	**1.20**	(0.750–1.70)	**3.20**	(2.30–4.80)	**7.60**	(5.00–12.0)	**13.0**	(7.90–23.0)	1949	69.2
	2001–2002	*		**<LOD**		**2.76**	(2.50–3.26)	**6.33**	(5.71–7.52)	**11.4**	(9.14–11.9)	2520	48.2
	2003–2004	*		**<LOD**		**4.54**	(3.57–5.83)	**10.2**	(9.00–11.5)	**15.7**	(13.5–16.9)	1931	48.3
	1999–2000	**0.924**	(0.608–1.41)	**0.920**	(0.570–1.40)	**2.73**	(1.68–4.60)	**7.94**	(4.40–12.2)	**12.2**	(8.00–19.6)	1949	69.2
	2001–2002	*		**<LOD**		**2.39**	(2.17–2.86)	**5.23**	(4.75–6.06)	**8.53**	(7.25–10.2)	3017	48.2
	2003–2004	*		**<LOD**		**4.42**	(3.82–5.79)	**9.02**	(7.85–10.8)	**13.2**	(10.5–16.1)	2450	48.3
Age group													
6–11 years	1999–2000	**1.32**	(0.757–2.29)	**1.50**	(0.940–2.50)	**4.50**	(2.00–7.80)	**11.0**	(5.80–20.0)	**16.0**	(8.50–35.0)	471	72.9
	2001–2002	*		**0.290**	(<LOD–1.04)	**3.45**	(2.41–4.47)	**9.56**	(6.44–17.8)	**20.0**	(9.44–38.2)	576	50.4
	2003–2004	*		**<LOD**		**5.13**	(1.84–6.64)	**10.9**	(7.50–13.8)	**16.1**	(10.4–18.5)	308	45.8
	1999–2000	**1.43**	(0.870–2.34)	**1.47**	(1.02–2.43)	**3.94**	(2.20–8.57)	**10.5**	(4.50–20.8)	**16.6**	(10.5–32.7)	471	72.9
	2001–2002	*		**0.890**	(<LOD–1.76)	**4.02**	(2.87–5.25)	**8.85**	(6.88–15.6)	**18.4**	(9.40–28.9)	576	50.4
	2003–2004	*		**<LOD**		**6.10**	(2.93–8.45)	**11.9**	(8.45–13.3)	**16.1**	(11.6–21.7)	308	45.8

Table 2. *Cont.*

	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
12–19 years	1999–2000	**1.21**	(0.758–1.94)	**1.40**	(1.00–2.00)	**3.70**	(2.30–5.40)	**7.90**	(4.70–20.0)	**20.0**	(8.00–27.0)	664	73.7
	2001–2002	*		**<LOD**		**2.86**	(1.98–3.95)	**7.58**	(5.71–9.15)	**11.0**	(9.45–12.4)	822	44.6
	2003–2004	*		**0.530**	(<LOD–2.32)	**5.80**	(4.34–7.67)	**14.8**	(9.12–19.8)	**20.8**	(14.8–32.7)	701	50.3
	1999–2000	**0.818**	(0.533–1.26)	**0.790**	(0.560–1.25)	**2.35**	(1.37–3.78)	**5.44**	(2.82–14.4)	**12.4**	(4.66–34.2)	664	73.7
	2001–2002	*		**<LOD**		**2.05**	(1.55–2.67)	**4.40**	(3.38–5.28)	**7.28**	(5.28–9.75)	821	44.6
	2003–2004	*		**0.440**	(<LOD–2.05)	**4.47**	(3.90–5.43)	**10.1**	(7.10–13.5)	**14.7**	(7.31–18.2)	699	50.3
20–59 years	1999–2000	**0.955**	(0.623–1.47)	**1.10**	(0.690–1.60)	**3.00**	(1.80–4.70)	**7.30**	(4.70–11.0)	**11.0**	(6.80–22.0)	814	67.6
	2001–2002	*		**<LOD**		**2.71**	(2.34–3.12)	**5.79**	(5.05–7.12)	**10.4**	(7.52–12.4)	1122	48.1
	2003–2004	*		**<LOD**		**4.37**	(3.86–7.20)	**9.74**	(8.35–11.3)	**14.2**	(11.5–16.2)	922	46.2
	1999–2000	**0.883**	(0.574–1.36)	**0.860**	(0.500–1.35)	**2.66**	(1.53–4.95)	**7.37**	(4.32–12.1)	**12.1**	(8.00–16.7)	814	67.6
	2001–2002	*		**<LOD**		**2.28**	(2.02–2.56)	**4.75**	(3.92–5.83)	**7.37**	(5.93–9.72)	1122	48.1
	2003–2004	*		**<LOD**		**4.29**	(3.80–6.32)	**8.34**	(7.14–11.9)	**11.9**	(10.1–16.6)	921	46.2
60 years and older	2001–2002	*		**0.440**	(<LOD–1.63)	**3.30**	(2.65–4.51)	**7.04**	(5.17–8.69)	**9.41**	(8.31–10.6)	498	50.2
	2003–2004	*		**1.04**	(<LOD–2.79)	**4.51**	(3.78–5.34)	**8.94**	(7.46–10.7)	**12.1**	(10.0–15.5)	522	55.8
	2001–2002	*		**1.00**	(<LOD–1.88)	**3.28**	(2.74–4.14)	**6.50**	(4.92–8.21)	**9.61**	(7.05–14.4)	498	50.2
	2003–2004	*		**1.56**	(<LOD–2.86)	**4.93**	(4.03–6.77)	**9.12**	(8.04–9.90)	**12.5**	(9.90–17.0)	522	55.8

Table 2. *Cont.*

	Survey years	Geometric mean	(95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
Sex													
Males	1999–2000	**1.11**	(0.717–1.73)	**1.20**	(0.830–1.70)	**3.80**	(2.50–5.00)	**8.00**	(5.00–19.0)	**19.0**	(7.20–30.0)	952	70.8
	2001–2002	*		**<LOD**		**3.13**	(2.67–3.56)	**6.99**	(5.83–7.71)	**11.5**	(9.31–12.1)	1187	47.4
	2003–2004	*		**<LOD**		**4.85**	(3.97–6.56)	**11.1**	(10.1–12.7)	**17.2**	(14.2–18.8)	928	46.5
	1999–2000	**0.855**	(0.566–1.29)	**0.820**	(0.510–1.34)	**2.61**	(1.76–4.03)	**7.69**	(4.41–12.1)	**12.2**	(6.94–23.8)	952	70.8
	2001–2002	*		**<LOD**		**2.04**	(1.88–2.64)	**4.31**	(3.94–5.29)	**6.88**	(5.79–9.42)	1420	47.4
	2003–2004	*		**<LOD**		**4.03**	(3.35–5.30)	**8.34**	(7.10–10.1)	**12.1**	(9.31–14.7)	1197	46.5
Females	1999–2000	**0.954**	(0.599–1.52)	**1.20**	(0.630–1.70)	**2.90**	(1.80–4.80)	**7.50**	(4.60–11.0)	**11.0**	(7.60–16.0)	997	67.6
	2001–2002	*		**0.290**	(<LOD–0.780)	**2.58**	(2.22–3.07)	**5.93**	(4.59–8.38)	**10.4**	(7.52–14.5)	1333	49
	2003–2004	*		**<LOD**		**4.39**	(3.17–5.78)	**9.39**	(8.17–10.8)	**13.5**	(11.3–15.9)	1003	50.1
	1999–2000	**0.996**	(0.620–1.60)	**0.960**	(0.540–1.62)	**2.81**	(1.45–5.85)	**8.00**	(4.05–13.6)	**12.1**	(6.49–19.6)	997	67.6
	2001–2002	*		**0.780**	(<LOD–1.29)	**2.66**	(2.44–3.33)	**6.28**	(4.95–7.76)	**9.57**	(7.12–13.6)	1597	49
	2003–2004	*		**<LOD**		**4.87**	(3.98–6.82)	**9.83**	(8.25–11.8)	**13.8**	(10.9–21.1)	1253	50.1
Race/ethnicity													.
Non–Hispanic whites	1999–2000	**0.981**	(0.579–1.66)	**1.10**	(0.490–2.10)	**3.30**	(2.20–4.90)	**7.60**	(4.70–14.0)	**14.0**	(7.60–25.0)	595	66.7
	2001–2002	*		**<LOD**		**2.44**	(2.20–3.15)	**5.56**	(5.05–7.36)	**10.2**	(8.38–11.6)	948	47
	2003–2004	*		**<LOD**		**4.16**	(3.98–7.11)	**9.74**	(8.40–11.6)	**14.8**	(12.5–17.2)	752	47.4

Table 2. *Cont.*

	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval)								Sample size	Weighted Detection Percent
				50th		75th		90th		95th			
	1999–2000	**0.932**	(0.549–1.58)	**0.900**	(0.430–1.68)	**2.87**	(1.51–5.88)	**8.57**	(4.40–14.4)	**13.0**	(8.21–23.8)	595	66.7
	2001–2002	*		**<LOD**		**2.30**	(2.04–2.95)	**4.95**	(4.53–5.93)	**7.80**	(6.61–10.7)	1265	47
	2003–2004	*		**<LOD**		**4.47**	(4.27–6.90)	**9.03**	(8.34–11.6)	**12.1**	(10.9–17.0)	1071	47.4
Mexican Americans	1999–2000	**1.22**	(0.740–2.01)	**1.20**	(0.840–1.70)	**4.10**	(2.20–7.00)	**11.0**	(6.40–18.0)	**18.0**	(12.0–23.0)	672	72.6
	2001–2002	*		**0.600**	(<LOD–1.49)	**3.10**	(2.27–3.72)	**6.26**	(5.00–7.80)	**11.2**	(7.80–12.4)	678	54.7
	2003–2004	*		**1.08**	(<LOD–2.81)	**5.58**	(2.08–4.85)	**10.8**	(8.35–16.1)	**17.7**	(12.0–28.4)	473	54.7
	1999–2000	**1.09**	(0.633–1.89)	**1.05**	(0.640–1.98)	**3.78**	(2.13–6.46)	**9.84**	(5.66–15.7)	**15.7**	(8.61–29.0)	672	72.6
	2001–2002	*		**0.890**	(<LOD–1.42)	**2.38**	(1.77–3.19)	**5.00**	(4.10–6.53)	**7.66**	(6.40–10.2)	763	54.7
	2003–2004	*		**0.960**	(<LOD–1.38)	**4.43**	(3.03–6.69)	**9.80**	(7.56–13.3)	**16.6**	(8.18–20.2)	580	54.7
Non–Hispanic blacks	1999–2000	**1.56**	(1.13–2.14)	**1.60**	(1.30–1.80)	**4.30**	(2.90–5.80)	**10.0**	(5.60–18.0)	**18.0**	(8.00–27.0)	509	79.8
	2001–2002	*		**0.890**	(<LOD–2.31)	**4.61**	(3.48–6.52)	**10.2**	(7.40–13.9)	**15.4**	(10.1–23.4)	696	51.9
	2003–2004	*		**0.830**	(<LOD–3.28)	**6.83**	(5.26–8.80)	**12.2**	(9.86–15.6)	**16.2**	(14.1–23.2)	578	50.1
	1999–2000	**1.07**	(0.773–1.47)	**1.18**	(0.830–1.54)	**2.61**	(1.89–3.47)	**5.98**	(3.96–9.67)	**11.9**	(5.98–19.7)	509	79.8
	2001–2002	*		**0.780**	(<LOD–1.60)	**2.80**	(2.57–3.61)	**7.02**	(5.41–8.72)	**9.75**	(7.84–12.1)	769	51.9
	2003–2004	*		**0.710**	(<LOD–1.98)	**4.14**	(3.46–5.09)	**7.77**	(6.57–10.5))	**11.7**	(10.5–13.4)	644	50.1

<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.

	Survey years	**Geometric mean** (95% confidence limit)		**Selected percentile** (95% confidence interval) **50th**		**75th**		**90th**		**95th**		**Sample size**	**Weighted Detection Percent**
Total	1999–2000	**1.82**	(1.36–2.44)	**2.70**	(1.40–4.10)	**11.0**	(8.40–16.0)	**38.0**	(25.0–41.0)	**48.0**	(38.0–62.0)	1948	62.9
	2001–2002	*		**0.470**	(<LOD–1.41)	**4.02**	(2.92–5.70)	**16.2**	(12.4–22.9)	**32.6**	(26.6–45.3)	2518	50.3
	2003–2004	**2.10**	(1.83–2.40)	**1.90**	(1.61–2.26)	**5.65**	(4.63–6.80)	**17.3**	(14.5–20.1)	**31.1**	(26.5–40.0)	1965	76.8
	1999–2000	**1.64**	(1.22–2.20)	**2.12**	(1.22–3.35)	**9.57**	(6.59–15.8)	**32.0**	(23.9–41.1)	**51.0**	(39.0–70.1)	1948	62.9
	2001–2002	*		**0.860**	(<LOD–1.33)	**3.79**	(2.50–5.19)	**13.3**	(10.9–18.8)	**27.2**	(21.7–37.7)	2517	50.3
	2003–2004	**1.97**	(1.71–2.27)	**1.75**	(1.54–2.06)	**5.21**	(4.46–5.95)	**15.7**	(11.7–19.7)	**30.4**	(25.4–34.2)	1962	76.8
Age group													
6–11 years	1999–2000	**2.72**	(1.93–3.85)	**4.20**	(2.50–7.20)	**20.0**	(13.0–29.0)	**40.0**	(38.0–52.0)	**62.0**	(40.0–92.0)	471	67.8
	2001–2002	*		**1.46**	(0.600–2.69)	**8.33**	(5.75–14.0)	**28.4**	(19.7–41.4)	**45.7**	(28.5–74.5)	575	58.9
	2003–2004	**2.79**	(2.25–3.45)	**2.67**	(1.81–3.88)	**6.95**	(5.64–8.58)	**19.4**	(10.5–27.4)	**30.9**	(19.4–76.5)	310	83.7
	1999–2000	**2.95**	(2.25–3.86)	**5.32**	(3.75–6.33)	**19.1**	(12.2–28.0)	**47.0**	(32.1–60.3)	**66.1**	(50.9–95.0)	471	67.8
	2001–2002	*		**2.16**	(1.32–3.12)	**10.6**	(7.84–13.6)	**28.7**	(18.8–45.0)	**48.1**	(33.4–71.1)	575	58.9
	2003–2004	**3.40**	(2.70–4.28)	**3.41**	(2.40–4.17)	**7.91**	(6.43–12.2)	**25.2**	(16.8–34.2)	**36.1**	(25.4–67.7)	310	83.7

Table 3. *Cont.*

	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
12–19 years	1999–2000	**2.53**	(1.64–3.92)	**3.70**	(1.70–6.80)	**16.0**	(11.0–31.0)	**38.0**	(33.0–58.0)	**69.0**	(38.0–260)	664	66.2
	2001–2002	*		**1.04**	(<LOD–2.12)	**4.83**	(3.35–6.48)	**20.8**	(12.2–27.9)	**34.9**	(23.6–54.7)	822	55.9
	2003–2004	**2.21**	(1.81–2.70)	**1.83**	(1.46–2.16)	**5.91**	(4.04–8.78)	**18.7**	(12.2–33.9)	**47.1**	(22.2–80.8)	717	77.4
	1999–2000	**1.71**	(1.07–2.75)	**2.14**	(0.890–4.83)	**13.5**	(6.46–22.6)	**36.0**	(25.9–51.4)	**61.5**	(41.7–109)	664	66.2
	2001–2002	*		**0.930**	(<LOD–1.56)	**3.56**	(2.38–5.57)	**12.2**	(8.95–16.0)	**22.5**	(13.2–34.7)	821	55.9
	2003–2004	**1.66**	(1.37–2.03)	**1.52**	(1.18–1.82)	**4.38**	(3.29–5.66)	**13.3**	(9.94–20.5)	**26.5**	(15.5–36.0)	715	77.4
20–59 years	1999–2000	**1.59**	(1.17–2.16)	**2.30**	(0.830–3.80)	**9.10**	(7.10–13.0)	**38.0**	(19.0–39.0)	**39.0**	(38.0–58.0)	813	61.3
	2001–2002	*		**<LOD**		**3.32**	(2.29–4.96)	**13.6**	(9.50–20.0)	**30.0**	(20.5–45.3)	1121	46.9
	2003–2004	**1.98**	(1.71–2.30)	**1.78**	(1.48–2.18)	**5.11**	(4.31–6.53)	**16.7**	(12.1–20.8)	**28.5**	(24.1–40.0)	938	73.5
	1999–2000	**1.47**	(1.07–2.02)	**1.90**	(0.870–3.11)	**8.09**	(5.19–14.6)	**27.0**	(19.8–37.6)	**47.5**	(34.2–70.1)	813	61.3
	2001–2002	*		**<LOD**		**3.16**	(1.99–4.62)	**11.9**	(7.79–17.2)	**25.2**	(15.9–37.0)	1121	46.9
	2003–2004	**1.88**	(1.61–2.19)	**1.67**	(1.45–1.94)	**4.88**	(4.20–5.68)	**13.9**	(10.3–19.7)	**30.4**	(19.7–38.2)	937	73.5
60 years and older	2001–2002	*		**0.860**	(<LOD–2.16)	**5.82**	(3.33–9.56)	**19.2**	(13.0–27.9)	**31.6**	(22.4–51.0)	498	53.7
	2003–2004	**3.18**	(2.47–4.08)	**2.43**	(1.95–3.24)	**7.91**	(5.07–12.8)	**21.2**	(12.8–44.8)	**44.8**	(19.6–119)	529	84.5
	2001–2002	*		**1.39**	(<LOD–2.40)	**6.38**	(3.68–9.97)	**24.0**	(15.5–35.2)	**45.5**	(26.5–69.7)	498	53.7
	2003–2004	**3.86**	(2.99–4.97)	**2.92**	(2.21–4.65)	**11.2**	(6.92–18.6)	**26.0**	(17.5–46.7)	**47.9**	(23.7–92.9)	529	84.5

Table 3. *Cont.*

	Survey years	**Geometric mean** (95% confidence limit)		**Selected percentile** (95% confidence interval) **50th**		**75th**		**90th**		**95th**		**Sample size**	**Weighted Detection Percent**
Sex													
Males	1999–2000	**2.10**	(1.48–2.98)	**3.50**	(2.20–4.80)	**14.0**	(8.00–24.0)	**38.0**	(21.0–49.0)	**42.0**	(38.0–53.0)	952	65.2
	2001–2002	*		**0.610**	(<LOD–1.42)	**4.21**	(3.07–5.97)	**18.3**	(12.2–27.2)	**31.1**	(25.0–43.3)	1187	52.2
	2003–2004	**2.13**	(1.80–2.53)	**1.94**	(1.49–2.44)	**6.09**	(4.44–7.23)	**16.1**	(10.9–21.1)	**26.8**	(22.0–41.6)	946	78.2
	1999–2000	**1.61**	(1.11–2.34)	**2.39**	(1.27–3.51)	**9.27**	(6.00–16.9)	**28.9**	(19.0–40.2)	**40.4**	(34.9–52.9)	952	65.2
	2001–2002	*		**0.780**	(<LOD–1.17)	**3.82**	(2.65–5.02)	**13.6**	(10.0–17.3)	**25.2**	(17.1–37.6)	1187	52.2
	2003–2004	**1.70**	(1.43–2.02)	**1.67**	(1.38–1.96)	**4.47**	(3.63–5.36)	**12.3**	(8.47–17.7)	**24.5**	(17.6–32.5)	945	78.2
Females	1999–2000	**1.59**	(1.23–2.06)	**2.00**	(0.690–3.60)	**9.70**	(7.30–14.0)	**38.0**	(26.0–39.0)	**52.0**	(38.0–110)	996	60.6
	2001–2002	*			<LOD	**3.76**	(2.50–5.71)	**15.9**	(10.6–22.0)	**34.3**	(23.2–47.3)	1331	48.5
	2003–2004	**2.06**	(1.74–2.44)	**1.86**	(1.58–2.18)	**5.21**	(4.27–6.77)	**19.8**	(12.8–24.0)	**33.8**	(26.1–47.8)	1019	75.5
	1999–2000	**1.66**	(1.26–2.18)	**2.01**	(0.870–3.33)	**10.0**	(6.67–16.2)	**34.9**	(26.2–47.1)	**70.1**	(39.0–118)	996	60.6
	2001–2002	*			<LOD	**4.22**	(2.40–7.00)	**15.6**	(11.3–22.6)	**29.6**	(24.8–43.8)	1330	48.5
	2003–2004	**2.28**	(1.91–2.72)	**1.88**	(1.59–2.44)	**6.00**	(4.80–8.30)	**19.4**	(12.8–26.2)	**32.6**	(27.3–42.0)	1017	75.5
Race/ethnicity													
Non–Hispanic whites	1999–2000	**1.77**	(1.23–2.53)	**2.70**	(0.830–4.40)	**11.0**	(7.50–17.0)	**38.0**	(17.0–53.0)	**48.0**	(38.0–69.0)	595	62.3
	2001–2002	*			<LOD	**3.99**	(2.46–6.14)	**17.3**	(10.1–25.0)	**33.1**	(25.0–50.2)	947	49.7
	2003–2004	**2.10**	(1.79–2.45)	**1.90**	(1.57–2.30)	**5.71**	(4.43–7.10)	**17.3**	(12.8–21.1)	**31.3**	(24.0–47.8)	757	77.4

Table 3. *Cont.*

	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
	1999–2000	**1.68**	(1.16–2.43)	**2.22**	(0.870–3.51)	**9.40**	(5.58–17.0)	**33.3**	(20.6–49.4)	**52.9**	(39.0–71.1)	595	62.3
	2001–2002	*			<LOD	**3.82**	(2.19–6.38)	**14.3**	(10.1–22.1)	**27.4**	(21.7–43.8)	947	49.7
	2003–2004	**2.08**	(1.76–2.48)	**1.80**	(1.59–2.27)	**5.36**	(4.46–6.23)	**17.4**	(11.4–21.6)	**31.7**	(24.2–38.2)	756	77.4
Mexican Americans	1999–2000	**1.79**	(1.05–3.05)	**2.00**	(0.530–4.40)	**11.0**	(6.70–17.0)	**39.0**	(32.0–62.0)	**140**	(46.0–230)	671	61
	2001–2002	*		**<LOD**		**3.76**	(2.66–5.18)	**15.1**	(11.1–19.1)	**35.2**	(19.1–46.0)	678	46
	2003–2004	**2.41**	(1.86–3.11)	**2.54**	(1.41–4.04)	**6.52**	(4.90–8.13)	**18.6**	(11.7–22.3)	**28.9**	(19.2–39.8)	498	77.3
	1999–2000	**1.60**	(0.899–2.86)	**1.83**	(0.680–4.23)	**10.4**	(5.95–16.9)	**37.0**	(23.1–63.1)	**112**	(40.5–190)	671	61
	2001–2002	*		**<LOD**		**3.55**	(2.52–4.93)	**13.2**	(9.61–22.7)	**30.2**	(22.7–47.7)	678	46
	2003–2004	**2.16**	(1.58–2.94)	**2.24**	(1.67–3.00)	**5.88**	(4.14–8.71)	**11.8**	(10.7–20.1)	**23.7**	(18.9–36.9)	497	77.3
Non–Hispanic blacks	1999–2000	**2.13**	(1.57–2.88)	**3.60**	(2.10–4.70)	**12.0**	(8.80–18.0)	**38.0**	(20.0–41.0)	**41.0**	(37.0–110)	509	66.1
	2001–2002	**1.61**	(1.19–2.19)	**1.26**	(0.660–2.05)	**5.54**	(3.29–9.41)	**20.6**	(15.6–27.7)	**42.9**	(27.2–62.8)	695	59.9
	2003–2004	**2.10**	(1.83–2.41)	**1.85**	(1.50–2.29)	**5.12**	(3.87–7.90)	**20.4**	(15.2–26.1)	**32.0**	(24.9–51.9)	579	76.9
	1999–2000	**1.45**	(1.03–2.06)	**1.75**	(1.17–3.06)	**8.48**	(4.36–13.4)	**25.5**	(15.4–39.3)	**54.4**	(25.5–97.6)	509	66.1
	2001–2002	**1.15**	(0.888–1.48)	**1.02**	(0.670–1.35)	**3.62**	(2.33–5.18)	**13.4**	(9.69–18.8)	**23.0**	(17.5–43.8)	694	59.9
	2003–2004	**1.37**	(1.23–1.53)	**1.19**	(1.06–1.31)	**3.50**	(2.69–5.07)	**11.8**	(7.95–16.2)	**23.9**	(13.3–27.1)	578	76.9

<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.

	Survey years	Geometric mean	(95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
Total	1999–2000	*		**0.500**	(<LOD–0.690)	**0.760**	(0.620–1.10)	**1.40**	(1.10–1.80)	**2.20**	(1.70–2.80)	1949	52.6
	2001–2002	**0.457**	(0.353–0.592)	**0.570**	(0.390–0.880)	**1.53**	(1.25–1.79)	**2.48**	(2.22–3.04)	**3.94**	(3.17–4.95)	2519	69.5
	2003–2004	*		**<LOD**		**0.830**	(0.690–0.950)	**1.77**	(1.42–2.31)	**2.80**	(2.31–3.78)	1905	49
	1999–2000	*		**0.250**	(<LOD–0.480)	**0.710**	(0.460–1.07)	**1.72**	(1.17–2.32)	**2.64**	(2.08–3.06)	1949	52.6
	2001–2002	**0.453**	(0.348–0.590)	**0.520**	(0.330–0.760)	**1.33**	(1.04–1.66)	**2.84**	(2.22–3.76)	**4.61**	(3.42–6.65)	2518	69.5
	2003–2004	*		**<LOD**		**0.700**	(0.580–0.830)	**1.47**	(1.16–2.04)	**2.62**	(2.00–3.72)	1903	49
Age group													
6–11 years	1999–2000	*		**0.600**	(<LOD–0.810)	**0.910**	(0.720–1.30)	**1.70**	(1.20–3.20)	**3.20**	(1.70–7.30)	471	58.1
	2001–2002	**0.453**	(0.350–0.585)	**0.550**	(0.350–0.850)	**1.58**	(1.33–2.04)	**2.75**	(2.22–3.38)	**4.08**	(2.95–5.16)	575	68.1
	2003–2004	*		**<LOD**		**0.820**	(0.580–0.970)	**1.45**	(1.05–2.16)	**2.18**	(1.45–4.13)	296	48.9
	1999–2000	*		**0.470**	(<LOD–0.870)	**1.08**	(0.800–1.32)	**1.75**	(1.44–2.36)	**2.45**	(2.04–5.32)	471	58.1
	2001–2002	**0.591**	(0.471–0.742)	**0.640**	(0.400–1.05)	**1.63**	(1.31–1.94)	**3.22**	(2.72–4.16)	**5.70**	(3.84–6.80)	575	68.1
	2003–2004	*		**<LOD**		**0.870**	(0.590–1.09)	**1.57**	(1.08–2.67)	**2.67**	(1.57–4.05)	296	48.9

Table 4. *Cont.*

	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
12–19 years	1999–2000	*		**0.210**	(<LOD–0.710)	**0.780**	(0.600–1.20)	**1.50**	(1.20–2.30)	**2.30**	(1.60–4.30)	664	49.8
	2001–2002	**0.505**	(0.388–0.657)	**0.690**	(0.440–0.960)	**1.61**	(1.32–1.94)	**2.57**	(2.23–3.39)	**4.08**	(2.73–5.86)	822	71.8
	2003–2004	*		**0.260**	(LOD–0.670)	**0.930**	(0.750–1.13)	**2.14**	(1.75–2.89)	**3.27**	(2.69–4.83)	690	51.7
	1999–2000	*		**0.180**	(<LOD–0.400)	**0.510**	(0.320–0.820)	**1.07**	(0.720–1.61)	**1.97**	(1.07–3.92)	664	49.8
	2001–2002	**0.393**	(0.300–0.515)	**0.530**	(0.310–0.740)	**1.23**	(0.980–1.53)	**2.19**	(1.61–3.07)	**3.14**	(2.25–3.97)	821	71.8
	2003–2004	*		**0.300**	(<LOD–0.350)	**0.640**	(0.560–0.730)	**1.49**	(1.16–1.60)	**1.97**	(1.57–2.43)	689	51.7
20–59 years	1999–2000	*		**0.490**	(<LOD–0.670)	**0.740**	(0.600–0.930)	**1.30**	(0.990–1.80)	**2.00**	(1.50–2.60)	814	52.3
	2001–2002	**0.449**	(0.340–0.592)	**0.540**	(0.380–0.880)	**1.45**	(1.19–1.79)	**2.46**	(2.11–3.17)	**3.83**	(2.96–5.34)	1122	67.8
	2003–2004	*		**<LOD**		**0.800**	(0.650–0.960)	**1.76**	(1.37–2.32)	**3.65**	(2.31–3.89)	919	47.2
	1999–2000	*		**0.250**	(<LOD–0.460)	**0.680**	(0.440–1.08)	**1.79**	(1.08–2.39)	**2.75**	(2.02–3.22)	814	52.3
	2001–2002	**0.447**	(0.335–0.597)	**0.490**	(0.320–0.740)	**1.32**	(0.990–1.71)	**2.87**	(2.08–3.95)	**4.69**	(3.20–8.71)	1122	67.8
	2003–2004	*		**<LOD**		**0.700**	(0.550–0.880)	**1.47**	(1.11–2.23)	**3.82**	(2.02–3.80)	918	47.2
60 years and older	2001–2002	**0.662**	(0.571–0.768)	**1.00**	(0.740–1.19)	**1.76**	(1.50–2.10)	**2.81**	(2.37–3.46)	**4.45**	(2.95–8.31)	497	75.5
	2003–2004	*		**0.220**	(<LOD–0.430)	**0.810**	(0.670–0.950)	**1.93**	(1.33–2.37)	**3.27**	(2.06–4.91)	517	53.5
	2001–2002	**0.821**	(0.708–0.952)	**0.970**	(0.850–1.10)	**2.25**	(1.81–2.66)	**3.97**	(3.30–5.03)	**5.51**	(4.79–7.46)	497	75.5
	2003–2004	*		**0.460**	(<LOD–0.520)	**0.880**	(0.740–1.08)	**1.97**	(1.36–2.87)	**3.76**	(2.14–8.11)	517	53.5

Table 4. *Cont.*

	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
Sex													
Males	1999–2000	*		**0.510**	(<LOD–0.700)	**0.790**	(0.680–1.10)	**1.50**	(1.20–2.20)	**2.70**	(1.90–4.10)	952	56.4
	2001–2002	**0.495**	(0.359–0.587)	**0.570**	(0.390–0.860)	**1.50**	(1.30–1.78)	**2.54**	(2.16–3.34)	**3.83**	(3.07–5.85)	1187	70.8
	2003–2004	*		**<LOD**		**0.880**	(0.720–1.01)	**2.20**	(1.54–2.50)	**2.95**	(2.36–4.29)	907	51.5
	1999–2000	*		**0.270**	(<LOD–0.470)	**0.670**	(0.520–0.840)	**1.34**	(1.08–2.17)	**2.67**	(1.67–3.23)	952	56.4
	2001–2002	**0.372**	(0.285–0.485)	**0.460**	(0.270–0.690)	**1.11**	(0.940–1.33)	**2.05**	(1.55–3.11)	**3.38**	(2.47–4.71)	1187	70.8
	2003–2004	*		**<LOD**		**0.590**	(0.500–0.760)	**1.42**	(0.950–2.07)	**2.62**	(1.61–3.97)	906	51.5
Females	1999–2000	*		**<LOD**		**0.720**	(0.570–1.00)	**1.30**	(0.910–1.60)	**1.70**	(1.30–3.20)	997	48.9
	2001–2002	**0.455**	(0.336–0.618)	**0.550**	(0.380–0.940)	**1.48**	(1.14–1.89)	**2.45**	(2.11–3.35)	**3.94**	(2.68–5.49)	1332	68.4
	2003–2004	*		**<LOD**		**0.780**	(0.590–0.970)	**1.47**	(1.22–2.00)	**2.57**	(1.87–3.78)	998	46.6
	1999–2000	*		**<LOD**		**0.790**	(0.380–1.50)	**1.89**	(1.07–2.52)	**2.52**	(1.89–3.75)	997	48.9
	2001–2002	**0.552**	(0.412–0.739)	**0.580**	(0.370–0.910)	**1.60**	(1.18–2.42)	**3.70**	(2.77–4.99)	**6.57**	(3.92–8.82)	1331	68.4
	2003–2004	*		**<LOD**		**0.750**	(0.660–0.900)	**1.50**	(1.22–2.23)	**2.60**	(2.08–3.98)	997	46.6
Race/ethnicity													
Non–Hispanic whites	1999–2000	*		**0.160**	(<LOD–0.700)	**0.740**	(0.580–1.10)	**1.30**	(0.930–1.90)	**1.90**	(1.50–2.80)	595	50.1
	2001–2002	**0.425**	(0.303–0.597)	**0.510**	(0.280–0.930)	**1.46**	(1.10–1.83)	**2.41**	(2.05–3.17)	**3.73**	(2.59–6.15)	948	67.2
	2003–2004	*		**<LOD**		**0.710**	(0.560–0.910)	**1.73**	(1.29–2.31)	**2.64**	(1.96–3.89)	745	46.5

Table 4. *Cont.*

	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
	1999–2000	*		**0.230**	(<LOD–0.550)	**0.710**	(0.390–1.22)	**1.88**	(1.05–2.58)	**2.64**	(2.08–3.07)	595	50.1
	2001–2002	**0.448**	(0.318–0.630)	**0.510**	(0.270–0.800)	**1.38**	(1.00–1.88)	**3.08**	(2.29–4.23)	**5.77**	(3.42–8.44)	948	67.2
	2003–2004	*		**<LOD**		**0.700**	(0.560–0.840)	**1.45**	(1.06–2.08)	**3.27**	(1.73–3.97)	744	46.5
Mexican Americans	1999–2000	*		**0.560**	(<LOD–0.780)	**0.840**	(0.740–0.980)	**1.50**	(1.20–1.90)	**2.20**	(2.00–2.90)	672	56.7
	2001–2002	**0.549**	(0.398–0.759)	**0.710**	(0.460–0.960)	**1.40**	(1.01–1.98)	**2.63**	(1.98–3.47)	**3.98**	(2.74–5.21)	678	77.5
	2003–2004	*		**0.240**	(<LOD–0.380)	**0.960**	(0.680–1.54)	**2.22**	(1.46–3.83)	**3.97**	(2.22–8.80)	478	51.2
	1999–2000	*		**0.330**	(<LOD–0.790)	**0.830**	(0.550–1.20)	**1.69**	(1.20–2.43)	**2.71**	(1.75–3.78)	672	56.7
	2001–2002	**0.509**	(0.390–0.693)	**0.560**	(0.380–0.840)	**1.28**	(1.03–1.67)	**2.55**	(1.77–3.72)	**3.72**	(2.58–6.30)	678	77.5
	2003–2004	*		**0.310**	(<LOD–0.390)	**0.750**	(0.480–1.22)	**1.92**	(1.22–3.43)	**3.66**	(2.02–6.17)	478	51.2
Non–Hispanic blacks	1999–2000	**0.343**	(0.201–0.584)	**0.570**	(<LOD–0.750)	**0.820**	(0.690–1.20)	**1.80**	(1.30–3.20)	**3.60**	(2.00–4.80)	509	62
	2001–2002	**0.749**	(0.592–0.949)	**1.18**	(0.740–1.49)	**1.86**	(1.77–2.03)	**3.55**	(3.01–3.91)	**5.27**	(3.89–6.74)	695	78
	2003–2004	**0.467**	(0.382–0.570)	**0.450**	(<LOD–0.730)	**1.09**	(0.930–1.26)	**2.32**	(1.59–2.88)	**3.26**	(2.41–5.46)	553	60
	1999–2000	**0.234**	(0.136–0.403)	**0.310**	(<LOD–0.580)	**0.720**	(0.510–0.850)	**1.39**	(1.03–2.10)	**2.91**	(1.49–4.24)	509	62
	2001–2002	**0.535**	(0.444–0.645)	**0.710**	(0.550–0.920)	**1.43**	(1.32–1.60)	**2.73**	(2.30–2.98)	**4.00**	(3.05–4.99)	694	78
	2003–2004	**0.305**	(0.253–0.368)	**0.280**	(<LOD–0.380)	**0.640**	(0.540–0.820)	**1.41**	(0.930–1.79)	**2.13**	(1.38–3.90)	552	60

<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.

	Survey years	**Geometric mean**	**(95% confidence limit)**	**Selected percentile (95% confidence interval) 50th**		**75th**		**90th**		**95th**		**Sample size**	**Weighted Detection Percent**
Total	1999–2000	*		**<LOD**		**2.30**	(1.30–3.90)	**13.0**	(5.00–17.0)	**19.0**	(17.0–38.0)	1949	49.2
	2001–2002	*		**<LOD**		**0.890**	(0.210–1.30)	**2.49**	(1.88–3.40)	**5.10**	(3.55 –8.35)	2518	32.7
	2003–2004	*		**<LOD**		**0.640**	(0.480–0.800)	**1.99**	(1.38–3.30)	**5.05**	(3.30–7.16)	1930	39
	1999–2000	*		**<LOD**		**1.88**	(0.970–3.86)	**10.1**	(5.31–18.3)	**21.7**	(12.8–33.7)	1949	49.2
	2001–2002	*		**<LOD**		**0.670**	(0.330–1.08)	**2.60**	(1.85–3.69)	**5.83**	(4.23–7.75)	2517	32.7
	2003–2004	*		**<LOD**		**0.500**	(0.340–0.650)	**2.14**	(1.21–3.18)	**5.27**	(2.69–7.61)	1927	39
Age group													
6–11 years	1999–2000	**0.691**	(0.425–1.12)	**0.740**	(0.080–1.80)	**4.30**	(2.40–8.60)	**17.0**	(6.90–37.0)	**32.0**	(17.0–44.0)	471	60.8
	2001–2002	*		**<LOD**		**1.30**	(0.750–2.11)	**3.53**	(2.20–4.50)	**7.33**	(4.32–9.74)	575	38.9
	2003–2004	*		**<LOD**		**0.900**	(0.620–1.14)	**2.94**	(1.14–5.48)	**5.48**	(2.94–8.53)	306	48.5
	1999–2000	**0.748**	(0.474–1.18)	**0.790**	(0.190–1.60)	**4.07**	(2.31–7.18)	**16.2**	(8.22–27.0)	**30.8**	(20.2–38.9)	471	60.8
	2001–2002	*		**<LOD**		**1.36**	(0.800–2.31)	**4.10**	(2.67–6.24)	**6.98**	(4.40–12.8)	575	38.9
	2003–2004	*		**<LOD**		**0.960**	(0.580–1.57)	**3.38**	(2.28–6.15)	**7.12**	(4.38–7.55)	306	48.5

Table 5. *Cont.*

	Survey years	**Geometric mean** (95% confidence limit)	**Selected percentile** (95% confidence interval)							**Sample size**	**Weighted Detection Percent**
			50th	**75th**		**90th**		**95th**			
12–19 years	1999–2000	*	**<LOD**	**2.30**	(1.40–4.50)	**13.0**	(5.40–20.0)	**20.0**	(17.0–38.0)	664	48.8
	2001–2002	*	**<LOD**	**0.830**	(0.400–1.14)	**2.52**	(1.85–3.07)	**4.63**	(3.59–5.83)	821	31.3
	2003–2004	*	**<LOD**	**0.580**	(0.350–0.770)	**1.46**	(1.12–1.99)	**2.67**	(1.82–4.49)	699	36.7
	1999–2000	*	**<LOD**	**1.52**	(0.620–3.47)	**9.48**	(4.04–16.8)	**21.5**	(9.48–42.3)	664	48.8
	2001–2002	*	**<LOD**	**0.540**	(0.310–0.770)	**2.02**	(1.49–2.40)	**3.13**	(2.51–4.67)	820	31.3
	2003–2004	*	**<LOD**	**0.360**	(0.240–0.580)	**1.02**	(0.630–1.57)	**2.45**	(1.33–3.39)	697	36.7
20–59 years	1999–2000	*	**<LOD**	**2.10**	(0.840–3.60)	**11.0**	(4.00–17.0)	**17.0**	(7.70–50.0)	814	47.3
	2001–2002	*	**<LOD**	**0.840**	(<LOD–1.31)	**2.32**	(1.70–3.40)	**4.90**	(2.90–9.52)	1122	30.8
	2003–2004	*	**<LOD**	**0.610**	(0.350–0.800)	**2.00**	(1.36–3.63)	**5.07**	(3.62–8.62)	925	35.8
	1999–2000	*	**<LOD**	**1.71**	(0.850–3.56)	**8.50**	(4.00–19.1)	**20.5**	(8.57–40.7)	814	47.3
	2001–2002	*	**<LOD**	**0.600**	(<LOD–1.05)	**2.56**	(1.64–4.03)	**6.33**	(3.96–8.17)	1122	30.8
	2003–2004	*	**<LOD**	**0.450**	(0.340–0.580)	**2.17**	(1.10–3.64)	**5.71**	(2.47–10.1)	924	35.8
60 years and older	2001–2002	*	**<LOD**	**1.02**	(0.540–1.37)	**3.54**	(2.58–5.96)	**7.89**	(5.09–16.1)	497	37.3
	2003–2004	*	**<LOD**	**1.04**	(0.580–2.07)	**3.35**	(2.06–11.3)	**11.3**	(3.14–30.1)	528	46.6
	2001–2002	*	**<LOD**	**1.10**	(0.630–1.85)	**4.89**	(3.36–7.82)	**13.4**	(6.98–19.8)	497	37.3
	2003–2004	*	**<LOD**	**1.19**	(0.590–1.91)	**5.03**	(2.16–13.0)	**13.0**	(4.45–23.0)	528	46.6

Table 5. *Cont.*

	Survey years	Geometric mean (95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
Sex												
Males	1999–2000	*	**0.110**	(<LOD–0.610)	**2.30**	(1.20–4.90)	**16.0**	(5.70–17.0)	**19.0**	(17.0–38.0)	952	49.4
	2001–2002	*	**<LOD**		**0.840**	(0.190–1.28)	**2.40**	(1.83–3.28)	**5.13**	(3.53–7.86)	1187	33.4
	2003–2004	*	**<LOD**		**0.600**	(0.260–0.910)	**1.76**	(1.07–3.35)	**4.45**	(2.24–7.97)	935	37.5
	1999–2000	*	**0.150**	(<LOD–0.370)	**1.79**	(0.840–3.97)	**11.0**	(4.62–17.4)	**18.1**	(7.51–44.7)	952	49.4
	2001–2002	*	**<LOD**		**0.580**	(0.270–0.820)	**2.01**	(1.40–2.67)	**4.67**	(2.90–6.80)	1187	33.4
	2003–2004	*	**<LOD**		**0.370**	(0.260–0.590)	**1.57**	(0.730–3.32)	**3.74**	(2.14–6.53)	934	37.5
Females	1999–2000	*	**<LOD**		**2.20**	(1.10–3.90)	**11.0**	(4.20–17.0)	**20.0**	(13.0–40.0)	997	48.9
	2001–2002	*	**<LOD**		**0.960**	(0.170–1.39)	**2.52**	(1.94–3.68)	**5.10**	(3.31–10.6)	1331	32
	2003–2004	*	**<LOD**		**0.690**	(0.510–0.850)	**2.40**	(1.43–4.15)	**5.07**	(3.35–10.3)	995	40.5
	1999–2000	*	**<LOD**		**2.06**	(0.940–4.00)	**9.30**	(4.96–25.5)	**27.0**	(9.66–47.5)	997	48.9
	2001–2002	*	**<LOD**		**0.820**	(0.370–1.43)	**2.92**	(2.29–4.56)	**7.73**	(4.44–11.9)	1330	32
	2003–2004	*	**<LOD**		**0.560**	(0.390–0.790)	**2.62**	(1.33–4.41)	**5.88**	(2.82–11.6)	993	40.5
Race/ethnicity												
Non–Hispanic whites	1999–2000	*	**<LOD**		**2.00**	(0.800–4.00)	**13.0**	(3.90–20.0)	**20.0**	(16.0–40.0)	595	47.2
	2001–2002	*	**<LOD**		**0.960**	(LOD–1.42)	**2.49**	(1.83–3.65)	**5.74**	(3.28–9.87)	947	32.6
	2003–2004	*	**<LOD**		**0.640**	(0.380–0.880)	**1.96**	(1.21–3.87)	**5.05**	(2.29–10.6)	752	39.4

Table 5. *Cont.*

	Survey years	Geometric mean (95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
	1999–2000	*	**<LOD**		**1.77**	(0.780–4.02)	**11.4**	(4.07–21.5)	**21.5**	(11.4–34.8)	595	47.2
	2001–2002	*	**<LOD**		**0.710**	(<LOD–1.31)	**2.85**	(1.91–4.96)	**7.29**	(4.25–9.47)	947	32.6
	2003–2004	*	**<LOD**		**0.470**	(0.340–0.700)	**2.25**	(1.02–4.03)	**5.89**	(2.47–10.1)	751	39.4
Mexican Americans	1999–2000	*	**0.250**	(<LOD–0.870)	**1.90**	(1.10–3.00)	**5.80**	(4.10–9.70)	**12.0**	(5.90–28.0)	672	52.5
	2001–2002	*	**<LOD**		**1.03**	(0.750–1.37)	**2.67**	(2.07–3.42)	**4.47**	(3.70–7.01)	678	37.6
	2003–2004	*	**<LOD**		**0.730**	(0.360–1.21)	**2.07**	(1.36–3.15)	**5.26**	(2.30–6.99)	498	41.8
	1999–2000	*	**0.270**	(<LOD–0.660)	**1.35**	(0.860–2.53)	**6.55**	(3.83–11.8)	**16.7**	(6.25–38.8)	672	52.5
	2001–2002	*	**<LOD**		**0.830**	(0.540–1.11)	**2.59**	(1.88–3.22)	**4.86**	(3.22–6.37)	678	37.6
	2003–2004	*	**<LOD**		**0.650**	(0.320–1.10)	**2.04**	(1.09–3.84)	**4.50**	(2.11–5.88)	497	41.8
Non–Hispanic blacks	1999–2000	*	**0.330**	(<LOD–1.20)	**3.20**	(1.40–7.00)	**14.0**	(5.70–30.0)	**19.0**	(17.0–39.0)	509	53.6
	2001–2002	*	**<LOD**		**0.770**	(<LOD–1.67)	**2.11**	(1.55–4.18)	**4.39**	(2.51–8.66)	695	30.8
	2003–2004	*	**<LOD**		**0.590**	(0.370–0.720)	**1.61**	(1.05–3.11)	**4.51**	(2.23–6.97)	552	36.6
	1999–2000	*	**0.250**	(<LOD–0.700)	**2.40**	(0.690–5.44)	**9.41**	(4.81–17.8)	**17.9**	(11.5–40.7)	509	53.6
	2001–2002	*	**<LOD**		**0.430**	(<LOD–0.930)	**1.80**	(0.830–3.50)	**3.65**	(2.33–5.91)	694	30.8
	2003–2004	*	**<LOD**		**0.340**	(0.260–0.430)	**0.890**	(0.740–1.50)	**2.33**	(1.12–4.55)	551	36.6

<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.

	Survey years	Geometric mean (95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
Total	1999–2000	*	**0.090**	(<LOD–0.140)	**0.210**	(0.140–0.290)	**0.470**	(0.380–0.640)	**0.870**	(0.640–1.10)	1949	54.7
	2001–2002	*	**<LOD**		**<LOD**		**0.610**	(0.410–0.770)	**0.850**	(0.700–1.30)	2516	18.7
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.320**	(0.170–0.540)	1965	7.91
	1999–2000	*	**0.070**	(<LOD–0.110)	**0.200**	(0.140–0.290)	**0.550**	(0.390–0.700)	**0.860**	(0.670–1.14)	1949	54.7
	2001–2002	*	**<LOD**		**<LOD**		**0.580**	(0.390–0.750)	**1.01**	(0.710–1.43)	2515	18.7
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.410**	(0.330–0.510)	1962	7.91
Age group												
6–11 years	1999–2000	*	**0.090**	(<LOD–0.160)	**0.190**	(0.130–0.280)	**0.430**	(0.300–0.650)	**0.850**	(0.470–1.00)	471	58.5
	2001–2002	*	**<LOD**		**<LOD**		**0.630**	(0.380–0.870)	**0.940**	(0.690–1.42)	576	20.4
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.540**	(<LOD–0.650)	310	8.47
	1999–2000	*	**0.100**	(<LOD–0.140)	**0.190**	(0.150–0.270)	**0.570**	(0.410–0.760)	**1.03**	(0.570–1.58)	471	58.5
	2001–2002	*	**<LOD**		**<LOD**		**0.780**	(0.610–1.12)	**1.36**	(1.02–1.86)	576	20.4
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.470**	(<LOD–0.970)	310	8.47

Table 6. *Cont.*

	Survey years	Geometric mean (95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
12–19 years	1999–2000	*	**0.080**	(<LOD–0.180)	**0.260**	(0.120–0.350)	**0.640**	(0.420–0.840)	**0.930**	(0.720–1.30)	664	53.1
	2001–2002	*	**<LOD**		**<LOD**		**0.560**	(0.330–0.730)	**0.820**	(0.610–0.990)	822	17.3
	2003–2004	*	**<LOD**		**<LOD**		**0.150**	(<LOD–0.350)	**0.450**	(0.370–0.570)	717	11.1
	1999–2000	*	**0.050**	(<LOD–0.080)	**0.170**	(0.100–0.220)	**0.440**	(0.230–0.730)	**0.730**	(0.380–1.09)	664	53.1
	2001–2002	*	**<LOD**		**<LOD**		**0.360**	(0.250–0.540)	**0.670**	(0.380–0.990)	821	17.3
	2003–2004	*	**<LOD**		**<LOD**		**0.230**	(<LOD–0.260)	**0.330**	(0.240–0.600)	715	11.1
20–59 years	1999–2000	*	**0.090**	(<LOD–0.130)	**0.210**	(0.130–0.290)	**0.450**	(0.360–0.640)	**0.900**	(0.610–1.10)	814	54.4
	2001–2002	*	**<LOD**		**<LOD**		**0.620**	(0.430–0.770)	**0.830**	(0.700–1.32)	1118	18.8
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.220**	(<LOD–0.580)	938	7.51
	1999–2000	*	**0.080**	(<LOD–0.110)	**0.210**	(0.140–0.310)	**0.550**	(0.360–0.730)	**0.860**	(0.650–1.20)	814	54.4
	2001–2002	*	**<LOD**		**<LOD**		**0.580**	(0.380–0.740)	**1.03**	(0.700–1.60)	1118	18.8
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.400**	(<LOD–0.540)	937	7.51
60 years and older	2001–2002	*	**<LOD**		**<LOD**		**0.630**	(0.360–1.08)	**1.10**	(0.610–1.41)	497	18.5
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**<LOD**		529	6.72
	2001–2002	*	**<LOD**		**<LOD**		**0.810**	(0.370–1.28)	**1.44**	(0.830–2.61)	497	18.5
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**<LOD**		529	6.72

Table 6. *Cont.*

	Survey years	Geometric mean (95% confidence limit)	Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
Sex												
Males	1999–2000	*	**0.090**	(<LOD–0.150)	**0.220**	(0.140–0.310)	**0.490**	(0.380–0.680)	**0.870**	(0.680–1.10)	952	56
	2001–2002	*	**<LOD**		**<LOD**		**0.600**	(0.370–0.740)	**0.770**	(0.680–1.03)	1187	18.8
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.390**	(0.130–0.540)	946	7.99
	1999–2000	*	**0.070**	(<LOD–0.110)	**0.190**	(0.140–0.230)	**0.410**	(0.340–0.500)	**0.720**	(0.520–0.940)	952	56
	2001–2002	*	**<LOD**		**<LOD**		**0.380**	(0.300–0.650)	**0.740**	(0.580–1.03)	1187	18.8
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.330**	(0.260–0.410)	945	7.99
Females	1999–2000	*	**0.090**	(<LOD–0.130)	**0.190**	(0.110–0.310)	**0.460**	(0.320–0.840)	**0.870**	(0.440–1.40)	997	53.4
	2001–2002	*	**<LOD**		**<LOD**		**0.660**	(0.460–0.850)	**0.990**	(0.700–1.42)	1329	18.6
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.240**	(<LOD–0.700)	1019	7.84
	1999–2000	*	**0.090**	(<LOD–0.120)	**0.220**	(0.140–0.360)	**0.670**	(0.410–0.870)	**0.890**	(0.660–1.62)	997	53.4
	2001–2002	*	**<LOD**		**<LOD**		**0.700**	(0.490–1.00)	**1.24**	(0.800–1.86)	1328	18.6
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.500**	(<LOD–0.640)	1017	7.84
Race/ethnicity												
Non–Hispanic whites	1999–2000	*	**0.080**	(<LOD–0.160)	**0.190**	(0.120–0.290)	**0.450**	(0.310–0.720)	**0.870**	(0.510–1.30)	595	51.9
	2001–2002	*	**<LOD**		**<LOD**		**0.610**	(0.360–0.780)	**0.830**	(0.650–1.36)	947	17.4
	2003–2004	*	**<LOD**		**<LOD**		**<LOD**		**0.290**	(<LOD–0.540)	757	7.58

Table 6. *Cont.*

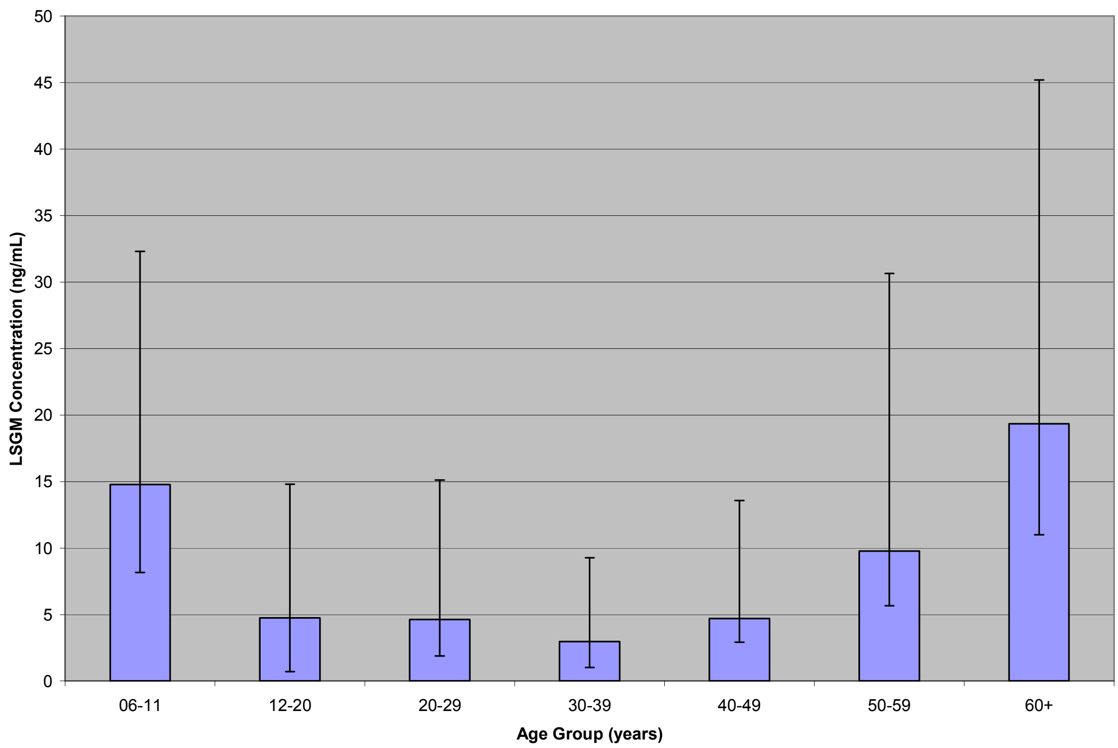
	Survey years	Geometric mean (95% confidence limit)		Selected percentile (95% confidence interval) 50th		75th		90th		95th		Sample size	Weighted Detection Percent
	1999–2000	*		**0.070**	(<LOD–0.120)	**0.200**	(0.140–0.310)	**0.560**	(0.380–0.730)	**0.880**	(0.600–1.38)	595	51.9
	2001–2002	*		**<LOD**		**<LOD**		**0.540**	(0.360–0.780)	**1.03**	(0.640–1.67)	947	17.4
	2003–2004	*		**<LOD**		**<LOD**		**<LOD**		**0.370**	(<LOD–0.520)	756	7.58
Mexican Americans	1999–2000	**0.130**	(<LOD–0.171)	**0.100**	(0.060–0.190)	**0.310**	(0.230–0.390)	**0.650**	(0.520–0.860)	**1.10**	(0.790–1.30)	672	63.4
	2001–2002	*		**<LOD**		**0.200**	(<LOD–0.520)	**0.700**	(0.410–1.12)	**1.11**	(0.700–1.58)	763	24.7
	2003–2004	*		**<LOD**		**<LOD**		**<LOD**		**0.170**	(<LOD–0.600)	610	11.6
	1999–2000	**0.116**	(0.084–0.161)	**0.090**	(0.060–0.170)	**0.300**	(0.190–0.410)	**0.810**	(0.570–0.990)	**1.19**	(0.860–2.66)	672	63.4
	2001–2002	*		**<LOD**		**<LOD**		**0.850**	(0.440–1.24)	**1.29**	(0.880–1.78)	678	24.7
	2003–2004	*		**<LOD**		**<LOD**		**0.320**	(<LOD–0.450)	**0.540**	(0.330–0.940)	497	11.6
Non–Hispanic blacks	1999–2000	*		**0.090**	(<LOD–0.170)	**0.270**	(0.140–0.400)	**0.560**	(0.400–0.830)	**0.870**	(0.660–1.20)	509	59.1
	2001–2002	*		**<LOD**		**<LOD**		**0.610**	(0.440–0.800)	**0.800**	(0.670–0.970)	767	20.9
	2003–2004	*		**<LOD**		**<LOD**		**<LOD**		**0.150**	(<LOD–0.210)	648	7.33
	1999–2000	**0.080**	(0.057–0.111)	**0.070**	(<LOD–0.110)	**0.170**	(0.110–0.280)	**0.450**	(0.300–0.580)	**0.700**	(0.500–1.02)	509	59.1
	2001–2002	*		**<LOD**		**<LOD**		**0.460**	(0.330–0.580)	**0.720**	(0.510–0.960)	693	20.9
	2003–2004	*		**<LOD**		**<LOD**		**<LOD**		**0.270**	(0.180–0.400)	578	7.33

<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 (β for linear = 0.008, β 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).

	DMP	DEP	DMTP	DETP	DMDTP	DEDTP
Dimethylphosphate (DMP)	1.000	0.325	0.391	0.221	0.282	0.029
		<.0001	<.0001	<.0001	<.0001	0.1518
	2,494	2,453	2,494	2,422	2,458	2,494
Diethylphosphate (DEP)		1.000	0.165	0.317	0.135	0.101
			<.0001	<.0001	<.0001	<.0001
		2,453	2,453	2,381	2,417	2,453
Dimethylthiophosphate (DMTP)			1.000	0.329	0.585	0.061
				<.0001	<.0001	0.0023
			2,494	2,422	2,458	2,494
Diethylthiophosphate (DETP)				1.000	0.237	0.171
					<.0001	<.0001
				2,422	2,386	2,422
Dimethyldithiophosphate (DMDTP)					1.000	0.129
						<.0001
					2,458	2,458
Diethyldithiophosphate (DEDTP)						1.000
						2,494

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.

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