

Comparison of Organochlorine Pesticide Levels in the U.S. Population from NHANES II (1976), NHANES 1999-2000 and NHANES 2001-2002

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Introduction

The *Third National Report on Human Exposure to Environmental Chemicals (3rd Report)*¹ is one of a series of reports of an ongoing biomonitoring exposure assessment of the U.S. population to environmental chemicals. Data in the *3rd Report* is from the 2001-2002 National Health and Nutrition Examination Survey (NHANES) conducted by the National Center for Health Statistics within the Center for Disease Control and Prevention (CDC). The sampling plan for NHANES is a complex, stratified, multistage, probability-cluster design to select a representative sample of the civilian, noninstitutionalized U.S. population. Presented here are selected serum organochlorine pesticides levels measured in a random one-third sample of people aged 12 years and older. The *3rd Report* presents pesticide data in both whole weight (ppb, ng/g serum) and lipid adjusted (ppb, ng/g lipid) units and includes data from NHANES 1999-2000. Pesticide data from this series of surveys can be used to establish reference ranges and track, over time, trends in levels of exposure.

Materials and Methods

Sample Preparation

Serum specimens (1-1.5 mL) were prepared by a modification of the procedure in Burse *et al.*² Samples were spiked with ¹³C₁₂-labeled internal standards followed by C₁₈ solid phase extraction (SPE) and extraction through neutral silica and Florisil SPE columns. After the addition of 2 µL of dodecane "keeper", excess solvent was evaporated. An analytical run consisted of nine unknown specimens, one method blank, and two quality control samples.

Mass Spectrometry

Before quantification, the extracts were reconstituted with 10µL external standard (25 pg/µL

¹³C₆-1,2,3,4 TCDD). Using a GC Pal (Leap Technology) auto sampler, 1µL of reconstituted extract is injected into an Agilent Technologies 6890 Gas Chromatograph operated in the splitless injection mode with a flow of 1 mL/min helium through a DB-5ms capillary column (30m x 0.25 mm x 0.25 µm film thickness). The pesticides were quantified by a Thermo Finnigan MAT95 XP (5kV) magnetic sector mass spectrometer operated in EI mode at 40 eV, using selected ion monitoring (SIM) at 10,000 resolving power (10% valley).³

The total lipid content of each specimen was estimated from its total cholesterol and triglycerides values using a "summation" method.⁴

Results and Discussion

Because the National Health and Examination Survey (NHANES) sample design is complex, statistical sample weights must be used to account for the unequal probability of selection into the survey. Statistical methods are also used to adjust for possible bias resulting from non-response and are post-stratified to U.S. Census Bureau estimates of the U.S. population. The *3rd Report* presents pesticide data for the total population and by age group, gender, and race/ethnicity. For these analyses, race/ethnicity is categorized as Mexican American (MA), non-Hispanic black

(NHB), and non-Hispanic white (NHW). Concentrations less than the limit of detection (LOD) were assigned a value equal to the LOD/ $\sqrt{2}$ for calculation of geometric means.

Since 1973, the US EPA has either banned the use of many persistent pesticides or highly regulated their applications. Some of those pesticides are still being applied for approved uses in other countries and may be present in food imported from those countries. In NHANES II, conducted from 1976-1980, selected organochlorine pesticides were measured in serum samples collected on a national probability half sample of persons 12-74 years of age from 64 locations across the United States. At that time, p,p' DDT, and mirex were banned for use, but other organochlorine pesticides were still being applied. In NHANES 2001-2002, many of the same organochlorine pesticides were measured in a probability one-third sample of persons 12-85+ years of age across the United States. All of the organochlorine pesticides reported here were banned before 1990 or their usage is highly regulated by US EPA. By comparing the NHANES 2001-2002 data to the NHANES II data, we can observe the changes in the serum pesticide levels in the United States populations over the past 25 years. We can also see how newer instrumentation has allowed us to detect lower concentrations of pesticides in human serum.

From Table 1 it is apparent that p,p'-DDE (a metabolite of DDT) is detected in virtually all of the specimens in NHANES II, NHANES 1999-2000 and NHANES 2001-2002. The analytical data for NHANES II is uncorrected for recovery whereas the latter data are corrected. Nonetheless, the median concentration in 2001-2002 is about a factor of 12 lower than the median concentration in 1976-1980. Currently, even the highest concentration of p,p'-DDE measured in 2001-2002 is less than 40% of the highest concentration measured in NHANES II. In NHANES 2001-2002, geometric mean levels of serum p,p'-DDE in the demographic groups were compared after adjusting for the covariates of race/ethnicity, age, gender, and lipid levels. The 12-19 year age group had more than a two-fold lower level of p,p'-DDE than the group aged 20 and older. This age trend can be seen in Figure 1. The same trend was observed in NHANES II participants and in NHANES 1999-2000.⁵ In addition to the age trend, the lipid adjusted geometric mean level of p,p'-DDE in Mexican Americans was approximately three-fold higher than in non-Hispanic whites and two-fold higher than in non-Hispanic blacks; this was also seen in NHANES 1999-2000.⁵

Table 1. Distribution of Pesticide Levels (ppb=ng/g whole weight) in Serum from

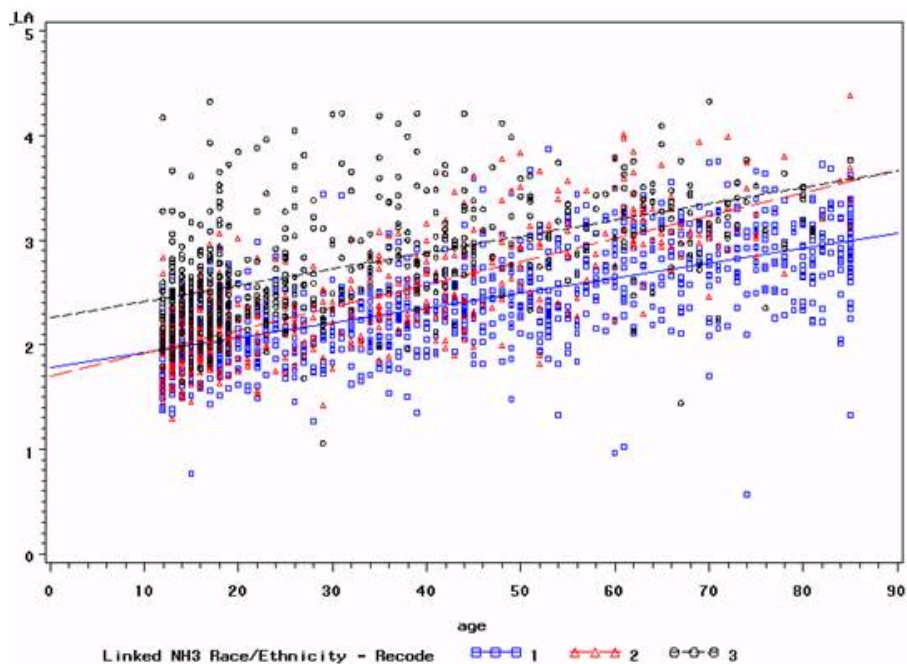
NHANES II (1976-1980)^{6,7}, NHANES 1999-2000⁵ and NHANES 2001-2002

Analyte (years)	Sample Size	Percent detected	50 th percentile (ppb)	95 th percentile (ppb)	Range (ppb)
p,p'-DDE (76-80)	4089	99.5	12.6	52.9	<1.0-378.6
(99-00)	1964	99.9	1.31	11.6	<0.11-173.3
(01-02)	2298	99.9	1.57	15.4	<0.05-142.4
p,p'-DDT (76-80)	3983	35.7	< 2	2.7	<2.0-57.8
(99-00)	1679	29.7	<0.12	0.172	<0.12-30.68
(01-02)	2305	28.9	< 0.10	0.184	< 0.10-15.46
o,p'- DDT (76-80)	3936	0.4	< 2	< 2	<2.0-42.1
(99-00)	1669	0.9	<0.12	<0.12	<0.12-0.701
(01-02)	2279	1.7	< 0. 10	< 0. 10	< 0.10-0.976
Analyte (years)	Sample Size	Percent detected	50 th percentile (ppb)	95 th percentile (ppb)	Range (ppb)
Heptachlor Epoxide (76-80)	3958	4.3	< 1	< 0.05	<1.0-22.4
(99-00)	1589	32.1	<0.09	0.177	<0.09-6.00
(01-02)	2259	43.3	< 0.06	0.153	<0.06-2.08
Oyxlchlordane(76-80)	0	-----	-----	-----	-----
(99-00)	1661	53.6	<0.08	0.310	<0.08-1.43

(01-02)	2249	62.1	< 0.06	0.352	<0.06-3.206
t-Nonachlor (76-80)	3941	7.1	< 1	1.2	<1.0-17.0
(99-00)	1933	70	0.105	0.544	<0.08-3.10
(01-02)	2286	73.7	0.112	0.589	<0.06-9.95
Dieldrin (76-80)	3742	10.6	< 1	1.4	<1.0-16.1
(99-00)	0	----	----	----	----
(01-02)	2159	49.9	< 0.06	0.146	<0.06-4.13
Aldrin (76-80)	4085	0	< 2	< 2	< 2
(99-00)	0	----	----	----	----
(01-02)	2275	0.2	< 0.03	< 0.03	<0.03-0.054
Endrin (76-80)	0	----	----	----	----
(99-00)	0	----	----	----	----
(01-02)	2187	0.02	< 0.03	< 0.03	<0.03-0.075
HCB (76-80)	2948	4.9	< 1	< 1	<1.0-16.9
(99-00)	1702	0.6	<0.7	<0.7	<0.7-1.98
(01-02)	2277	5.3	< 0.18	< 0.18	<0.18-1.64
β -HCCH (76-80)	3846	17.2	< 1	2.4	<1-27.8
(99-00)	1893	57	<0.05	0.447	<0.05-9.33
(01-02)	2291	53.5	< 0.04	0.296	<0.04-24.61
γ -HCCH (76-80)	4074	0.2	< 1	< 1	<1.0-6.2
(99-00)	1799	2.8	<0.08	<0.08	<0.08-1.31
(01-02)	2280	0.9	< 0.06	< 0.06	<0.06-3.84
Mirex (76-80)	4038	0	< 10	< 10	< 10
(99-00)	1853	7.7	<0.09	<0.09	<0.09-1.96
(01-02)	2257	23.6	< 0.06	0.414	<0.06-19.43

The 95th percentiles in NHANES 2001-2002 for all analytes in Table 1 were 3 to 10 fold lower than the 95th percentile in NHANES II. Despite this significant drop in serum pesticide levels in the US population, the effect of reducing the detection limits on the number of samples with measurable concentrations can also be seen in Table 1. For example, in NHANES 2001-2002, t-nonachlor, oxychlorane, dieldrin, and β -HCCH were reported in over half of the specimens. In NHANES II their median concentrations were below the detection limits. The increased number of detects can be explained by the lower detection limits that are now attained with newer instrumentation.

Figure 1. Log₁₀ of p,p'-DDE (ng/g lipid) vs. Age by Race: 1=NHW, 2=NHB and 3=MA



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