Profiles of Great Lakes Critical Pollutants: A Sentinel Analysis of Human Blood and Urine

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To determine the contaminants that should be studied further in the subsequent population-based study, a profile of Great Lakes (GL) sport fish contaminant residues were studied in human blood and urine specimens from 32 sport fish consumers from three Great Lakes: Lake Michigan (n = 10), Lake Huron (n = 11), and Lake Erie (n = 11). Serum was analyzed for 8 polychlorinated dioxin congeners, 10 polychlorinated furan congeners, 4 coplanar and 32 other polychlorinated biphenyl (PCB) congeners, and 11 persistent chlorinated pesticides. Whole blood was analyzed for mercury and lead. Urine samples were analyzed for 10 nonpersistent pesticides (or their metabolites) and 5 metals. One individual was excluded from statistical analysis because of an unusual exposure to selected analytes. Overall, the sample (n = 31) consumed, on average, 49 GL sport fish meals per year for a mean of 33 years. On average, the general population in the GL basin consume 6 meals of GL sport fish per year. The mean tissue levels of most persistent, bioaccumulative compounds also found in GL sport fish ranged from less than a twofold increase to that of PCB 126, which was eight times the selected background levels found in the general population. The overall mean total toxic equivalent for dioxins, furans, and coplanar PCBs were greater than selected background levels in the general population (dioxins, 1.8 times; furans, 2.4 times; and coplanar PCBs, 9.6 times). The nonpersistent pesticides and most metals were not identified in unusual concentrations. A contaminant pattern among lake subgroups was evident. Lake Erie sport fish consumers had consistently lower contaminant concentrations than consumers of sport fish from Lakes Michigan and Huron. These interlake differences are consistent with contaminant patterns seen in sport fish tissue from the respective lakes. GL sport fish consumption was the most likely explanation for observed contaminant levels among this sample. Frequent consumers of sport fish proved to be effective sentinels for identifying sport fish contaminants of concern. In the larger study to follow, serum samples will be tested for PCBs (congener specific and coplanar), DDE, dioxin, and furans. Key words: dioxin, fish consumption, furan, Great Lakes, metals, organochlorines, pesticides, polychlorinated biphenyls, serum, urine, whole blood. Environ Health Perspect 106:279–289 (1998). [Online 7 April 1998] http://ehpnet1.niehs.nih.gov/docs/1998/106p279-289anderson/abstract.html

The International Joint Commission (IJC) has identified approximately 1,000 substances of potential concern in the Great Lakes (GL) ecosystem. The IJC Water Quality Board identified 11 of these substances as priority contaminants. They are total polychlorinated biphenyl, mirex, hexachlorobenzene (HCB), dieldrin, DDT and metabolites of DDT, 2,3,7,8-TCDD, 2,3,7,8-tetrachlorodibenzo-furan (TCDF), benzo[a]pyrene, alkylated lead, toxaphene, and mercury. These contaminants were selected based on the following criteria: persistence in the environment, potential to bioaccumulate, and toxicity (1). Only polychlorinated biphenyls (PCBs), DDT, metabolites of DDT, and mercury have been systematically assessed in basin residents who consume sport fish from the Great Lakes. Few studies have been conducted to assess the body burden levels of other organochlorine compounds (dioxin, furans, and coplanar PCBs) (2–4). Among Canadian residents who reside within and outside of the Great Lakes basin, two studies found body burden levels to be higher among fish consumers than nonconsumers; however, neither of the studies included residents who consumed sport fish from Lakes Michigan, Huron, and Erie.

The presence of high concentrations of DDT and its metabolites in GL sport fish has been known since the 1960s (5). To minimize human exposure, public health sport fish consumption advisories were initiated by many states. In addition, the use of PCBs, DDT, and other chlorinated pesticides were banned in the United States in the 1970s. Stringent industrial discharge regulations, imposed since the contamination was identified, have resulted in substantial declines in the concentration of these chlorinated compounds in GL sport fish. Despite initial success in decreasing contaminant concentrations, most of the GL states continue to issue sport fish consumption advisories.

The dietary pathway dominates population exposure routes to lipophilic, bioconcentrating contaminants (estimated at 40–100% of exposure) (6). For GL anglers, consumption of fish from basins waters is the largest contributor to their dietary exposure component. Sport anglers who consume large quantities of such fish represent a subpopulation with the potential for significant exposure to contaminants known to be present in fish (7,8). Approximately 11% of basin residents hold sport fishing licenses. In 1991, an estimated 2.55 million licensed anglers fished on one or more of the Great Lakes (9). The sport fishing industry has been estimated to contribute over $1.3 billion annually to the basin economy (10). GL basin residents included in EPA National Human Adipose Tissue Survey (NHATS) ranked third among nine regions in tissue burdens of the substances surveyed (11). In addition, multiple human epidemiologic studies have correlated sport fish consumption with increased tissue concentrations, substantiating that consumption of sport fish is an important exposure pathway to critical pollutants (7,8,12,13).

The Agency for Toxic Substances and Disease Registry (ATSDR), under the auspices of the 1990 Great Lakes Initiative, established cooperative agreements to coordinate basin-wide human health effects research. This provided the opportunity to establish a consortium of five state health departments (Wisconsin, Illinois, Indiana, Michigan, Ohio) from states bordering Lakes Michigan, Huron, and Erie. The initial consortium activity, in collaboration with the National Center for Environmental Health (NCEH), Division of Environmental Health Laboratory Sciences, Centers for Disease Control and Prevention (CDC), was to measure serum and urine levels of a spectrum of Great Lakes critical pollutants among frequent consumers of sport fish from Lakes Michigan, Huron, and Erie. It

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was feasible to study a spectrum of contaminants in biological specimens because of improvements made in laboratory analytic methods. Sample preparation techniques refinements developed for use in the Third National Health and Nutrition Examination Survey (NHANES-III) now provide the means to efficiently identify human tissue accumulation of several different chemicals using small quantities of serum. In addition, methodological changes have increased sensitivity and specificity. The goal was to determine whether IJC priority substances, not previously assessed in basin residents, warranted further consideration in subsequent population-based studies of anglers.

### Table 1. Elements measured in Great Lakes study participants

| Metal  | Matrix | Method Description | Reference |
|--------|--------|--------------------|-----------|
| Lead   | Blood  | Graphite furnace AA| (19)      |
| Mercury| Blood  | Cold vapor AA      | (20)      |
| Arsenic| Urine  | Zeeman graphite furnace AA| (21) |
| Cadmium| Urine  | Zeeman graphite furnace AA| (22) |
| Chromium| Urine | Zeeman graphite furnace | (23)   |
| Mercury| Urine  | Cold vapor AA      | (24)      |
| Nickel | Urine  | Zeeman graphite furnace AA| (23) |

AA, atomic absorption.

### Table 2. Nonpersistent pesticide residues measured in urine

| Residue                  | Possible parent pesticide          |
|-------------------------|-----------------------------------|
| 2-Isopropoxyphenol       | Propoxur                          |
| Carbarylphenol           | Carboxuran                         |
| 2,4-Dichlorophenoxyacetic acid | Chlorpyrifos                  |
| 3,5,6-Trichloro-2-pyridinol | Naphthalene, carbyl             |
| 1-Naphthol               | Naphthalene                        |
| 2-Naphthol               | Naphthalene                        |
| 4-Nitrophenol            | Parathion, nitrobenzene           |
| Pentachlorophenol        | Pentachlorophenol                  |
| 2,4,5-Trichlorophenol    | Lindane, 1,2,4-trichlorobenzene   |
| 2,4,6-Trichlorophenol    | Lindane, 1,2,4-trichlorobenzene   |

### Table 3. Demographic and fish consumption characteristics

| Characteristic | All  | Lake Michigan (n = 31) | Lake Huron (n = 11) | Lake Erie (n = 10) | All men (n = 21) | All women (n = 10) |
|---------------|------|------------------------|---------------------|-------------------|-----------------|-------------------|
| Age (years)²,³ | 52 (36-76) | 52 (47-55) | 56 (39-76) | 48 (36-54) | 52 (41-76) | 49 (36-71) |
| BMI (kg/m²)³,⁴,⁵ | 27 (18-39) | 29 (23-37) | 29 (18-39) | 24 (20-30) | 28 (23-39) | 23 (18-27) |
| Education (%) | Elementary | 3.3 | 0.0 | 9.1 | 0.0 | 4.8 | 0.0 |
|              | Some high school | 6.7 | 0.0 | 9.9 | 10.0 | 4.8 | 11.1 |
|              | High school graduate | 33.3 | 44.4 | 27.3 | 30.0 | 33.3 | 33.3 |
|              | Some college | 40.0 | 33.3 | 36.4 | 50.0 | 38.1 | 44.4 |
|              | College graduate | 16.7 | 22.2 | 18.2 | 10.0 | 19.0 | 11.1 |
| Fish consumption⁶ | All fish | Mean (median) | 77 (52) | 88 (52) | 72 (52) | 73 (52) | 77 (52) |
|                | Range | 12-208 | 24-208 | 12-152 | 24-156 | 12-156 | 12-208 |
|                | GL sport | Mean (median) | 49 (41) | 43 (27) | 48 (46) | 55 (46) | 55 (45) | 34 (34) |
|                | Range | 10-172 | 10-172 | 14-122 | 24-121 | 12-121 | 10-52 |
| Years consumed sport fish⁷ | 36 (15-58) | 31 (15-54) | 40 (20-58) | 36 (20-53) | 37 (15-58) | 32 (20-58) |
| Years consumed GL sport fish | 33 (11-58) | 27 (15-45) | 39 (20-58) | 32 (11-45) | 34 (15-58) | 29 (11-58) |

Abbreviations: BMI, body mass index; GL, Great Lakes.

²Values shown are mean (range).
³All men are statistically different from all women (p<0.01).
⁴The three GL subgroups are statistically different (p<0.01).
⁵Meals consumed in the last year.
⁶Number of all types of fish consumed, purchased or sport caught.
⁷Number of different types of GL sport caught fish consumed.

### Methods

#### Subject Selection

Sea Grant Institutions of three states, Wisconsin, Ohio, and Michigan, sought participants from angling enthusiasts including sport fishing charter boat captains, their spouses, and other individuals involved in the sport fishing industry. To be eligible for participation, each subject must have self-reported that they consumed at least one GL sport fish meal per week. In fall 1993, Sea Grant Institutions recruited 32 subjects, approximately 10 who consumed sport fish from each of three Great Lakes, Lake Michigan (n = 10), Lake Huron (n = 11), and Lake Erie (n = 11). This convenience sample of 32 subjects completed a telephone survey and gave informed consent prior to donating blood and urine samples. The study protocol was approved by the Committee for the Protection of Human Subjects with the University of Wisconsin-Madison.

#### Data Collection

**Telephone survey.** Information about fish consumption habits and demographics were obtained via a telephone survey administered by the University of Wisconsin-Extension Survey Research Laboratory (Madison, WI). Information about the number of all types of fish meals consumed in the last 12 months was obtained using the following question: "About how many meals of fish did you eat per week or per month in the last 12 months, including all types of freshwater and saltwater fish, whether fresh, canned, smoked, or frozen?" If any of the meals they had consumed in the last 12 months included GL sport caught fish, each subject was asked how many meals of different types of GL sport fish they had consumed in that time period. The types of GL sport fish meals were 1) lake trout; 2) brown trout; 3) rainbow trout or chinook or coho salmon; 4) carp or catfish; and 5) perch, smelt, or wall-eye. Information obtained included the number of years they consumed sport fish and GL sport fish and if the amount they consumed had changed since they first began eating GL sport fish. Demographic information included date of birth, height and weight, education level attained, and whether they recalled any occupational or unusual environmental exposures to the study compounds. After completion of the telephone survey, each subject was invited to donate blood and urine samples.

**Blood sample collection.** The protocol used by all research staff for specimen collection, preparation, and shipment was prepared by the CDC. Approximately 120 ml whole blood was collected in vacutainer
tubes (no anticoagulant or serum separator). Whole blood was allowed to clot for at least 20 min and then centrifuged for 15 min at 2,500 rpm. The serum was transferred with solvent-rinsed glass pipettes into solvent-rinsed glass vials supplied to the research team by the CDC. The serum was placed at -20°C until laboratory analysis. In addition, 6 ml whole blood was collected in EDTA vacutainer tubes. These tubes were stored at 4°C until laboratory analysis was initiated.

**Urine sample collection.** Prior to the blood collection appointment, each participant received materials along with instructions needed to collect a urine specimen. Each participant collected the first morning void of the day of the appointment and transported the specimen to the clinic at the time of the scheduled appointment.

**Laboratory Analyses**

Copies of a summary of the methods, quality control protocols, or limits of detection are available upon request. The methods used for laboratory analysis have been described in detail (14). Briefly, 75 ml serum was tested for 8 polychlorinated dibenzo-p-dioxins (PCDDs), 10 polychlorinated dibenzofurans (PCDFs), and 4 coplanar PCB congeners, according to the high resolution mass spectrometric method of Patterson et al. (15). The results were summarized on a lipid adjusted (ppt) basis (16). Three milliliters of serum was analyzed for 31 individual PCB congeners and polybrominated biphenyls (PBBs) utilizing the capillary column gas chromatography with electron capture detection method of Burse et al. (17), which was modified to include analyses on two different gas chromatographic columns. Residues of 11 persistent pesticides were measured in the same serum extracts as the PCBs (18). These were HCB, \( p,p' \)-dichlorodiphenylchloroethylene (\( p,p' \)-DDE), \( \gamma \)-hexachlorocyclohexane (\( \gamma \)-HCH), \( \beta \)-HCH, oxychlordane, heptachlor epoxide, \( \alpha \)- and \( \beta \)-endosulfan, \( \alpha \)- and \( \beta \)- endrin, \( \alpha \)- and \( \beta \)-dieldrin. The PCB and persistent pesticide serum data were summarized on a nanograms per milliliter (ng/mL) basis. Whole blood and urine were analyzed for selected elements [Table 1 (19–25)]. Results were reported on a whole blood basis. In addition, 10 urinary pesticides or their metabolites were measured in 10 ml urine by the tandem quadrupole mass spectrometric method of Hill et al. (26). These analytes along with their potential parent compounds are listed in Table 2.

**Data Analysis**

**Demographics.** The age of each subject at the time of interview was calculated by subtracting the date of birth from the date of interview. Body mass index (BMI) was calculated by dividing each subject’s weight (kilograms) by the square of their height (meters\(^2\)). To describe fish and Great Lakes sport fish consumption habits of this sample, two variables were created: All Fish and GL Sport (Table 3). All fish is the number of all types of fish purchased or sport caught, consumed in the last year. GL sport is the sum of the number of different types of GL sport fish consumed in the last year. The GL sport variable is the mean of the sum for all subjects.

**Laboratory data.** The dioxin total (Table 4), furan total (Table 5), and coplanar PCB total (Table 6) were calculated to

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**Table 4. Mean and range (ppt) of serum chlorinated dibenzo-dioxins (CDD), lipid adjusted**

| Dioxin congener | All subjects \( n = 31 \) | Lake Michigan \( n = 9 \) | Lake Huron \( n = 11 \) | Lake Erie \( n = 11 \) | Comparison group* |
|-----------------|---------------------------|--------------------------|--------------------------|--------------------------|------------------|
| 2,3,7,8-TCDD\( b \) | 6.6 (ND-17.2) | 4.7 (ND-7.9) | 10.6 (4.4-17.2) | 4.3 (ND-9.5) | 2.8 (0.3-8.9) |
| 1,2,3,7,8-PentaCDD\( b \) | 10.6 (ND-31.5) | 9.8 (ND-23.7) | 16 (ND-31.5) | 12.3 (ND-26.3) | 6.6 (0.6-14.1) |
| 1,2,3,4,7,8-HexaCDD | 8.4 (ND-22.1) | 11.4 (ND-16.3) | 11.6 (2.1-22.7) | 11.6 (ND-16.6) | 9.0 (0.9-21.1) |
| 1,2,3,6,7,8-HexaCDD | 126 (71.9-228) | 120 (71.9-190) | 142 (88.7-226) | 115 (65.1-150) | 70.8 (24.8-160) |
| 1,2,3,7,8,9-HexaCDD | 7.0 (ND-22.8) | 8.7 (ND-22.8) | 6.7 (ND-16.1) | 11.6 (ND-13) | 9.4 (0.9-25.8) |
| 1,2,3,4,6,7,8-HeptaCDD\( b \) | 134 (ND-25.9) | 144 (92.5-254) | 163 (98.7-314) | 35.9 (ND-159) | 124 (29.1-358) |
| 1,2,3,4,6,7,9-HeptaCDD\( c \) | ND | ND | 4 (6.6-17) | 4 (4.4-17.2) | 4.4 (1.1-29.1) |
| OctaCDD | 777 (297-1869) | 793 (409-1587) | 919 (371-1869) | 632 (297-981) | 971 (286-2710) |
| Dioxin total (ppt) | 1,062 (450-2,410) | 1,087 (615-2,017) | 1,299 (729-2,410) | 944 (453-1,286) | 1,158* |
| Dioxin total (toxic equivalent)\( b \) | 27.5 (8.2-58.7) | 25.9 (13.8-38.3) | 36 (18.5-58.7) | 20.7 (8.2-31.0) | 15.5* |

*ND, none detected.

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**Table 5. Mean and range (ppt) of serum chlorinated dibenzofurans (CDF), lipid adjusted**

| Congener | All subjects \( n = 31 \) | Lake Michigan \( n = 9 \) | Lake Huron \( n = 11 \) | Lake Erie \( n = 11 \) | Comparison group* |
|----------|---------------------------|--------------------------|--------------------------|--------------------------|------------------|
| 2,3,7,8-TetraCDF | 2.2 (ND-6.9) | 2.1 (ND-4.2) | 2.1 (ND-4.1) | 3.1 (ND-8.9) | 3.1 (0.4-66.0) |
| 1,2,3,7,8-PentaCDF\( c \) | 2.0 (ND-4.1) | ND | 1.7 (ND-3.8) | ND | 1.6 (0.5-5.0) |
| 2,3,4,7,8-PentaCDF\( d \) | 17.7 (ND-40.4) | 20.4 (8.7-40.4) | 22.8 (10.1-36.5) | 10.4 (ND-16.0) | 5.5 (0.2-19.8) |
| 1,2,3,4,7,8-HexaCDF | 12.7 (ND-36.0) | 11.6 (ND-20.8) | 16.0 (6.6-36.0) | 10.2 (ND-15.9) | 8.0 (1.7-17.6) |
| 1,2,3,6,7,8-HexaCDF | 11.6 (ND-23.7) | ND | 18.8 (ND-23.7) | ND | 7.7 (1.0-12.7) |
| 1,2,3,7,8,9-HexaCDF | ND | ND | ND | ND | 1.8 (0.2-8.9) |
| 2,3,4,6,7,8-HexaCDF | 5.1 (ND-11.8) | 6.0 (ND-11.8) | 4.5 (ND-7.1) | 5.0 (ND-6.9) | 3.8 (0.7-8.8) |
| 1,2,3,4,6,7,8-HeptaCDF\( d \) | 20.0 (ND-39.8) | 22.1 (12.4-38.8) | 22.9 (14.4-35.8) | 15.2 (ND-23.9) | 21.3 (10.1-53.5) |
| 1,2,3,4,7,8,9-HeptaCDF | ND \( b \) | ND | ND \( b \) | ND | NA |
| OctaCDF\( d \) | ND | ND | ND | ND | 6.9 |
| Furan total (ppt)\( d \) | 86.2 (23.5-139) | 70.8 (39.3-110) | 79.3 (46.2-139) | 49.3 (23.5-127) | 57.3 (NA) |
| Furan total | 11.9 (3.4-25.3) | 13.2 (6.8-22.4) | 14.9 (7.4-25.3) | 7.8 (3.4-11.5) | 4.9 |

*ND, none detected.

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**Abbreviations:** ND, none detected; NA, not available.

*Unexposed sample residing in Jacksonville, Arkansas \( n = 70 \).

*One observation detected.

*Range not available.

*Three Great Lakes subgroups are statistically different \( p<0.05 \).

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describe the total concentration of all dioxin, furan, and coplanar PCB congeners. For each subject, the following variables were created: 1) dioxin sum, the sum of the eight dioxin congener values; 2) furan sum, the sum of the 10 furan congener values; and 3) coplanar PCB sum, the sum of the four coplanar PCB congener values. The dioxin, furan, and coplanar PCB totals are the sample means of the three respective summed variables. This method was used for the concentration (ppt) and the 2,3,7,8-TCDD equivalents (TEq).

The variable PCB total (Table 7) was calculated to describe the total concentration of all PCB congeners. For each subject, all PCB congener concentrations were summed. PCB total is the mean of the sum for all subjects.

Statistical estimation of nondetectable values. The methods described by Hornung and Reed (27) were used to estimate the average for the dioxins, furans, coplanar and congener-specific PCB residues, serum pesticides, and urine nonpersistent congeners (all compounds except metals). For the dioxins, furans, coplanar PCBs, congener-specific PCBs, and PBBs, the following guidelines were used: if <50% of the sample had nondetectable values for a compound, the concentration for the nondetectable was imputed by dividing the limit of detection by the square root of 2; if >50% of the sample had nondetectable values for a compound, the concentration was assigned half the limit of detection. Next, each imputed value was compared to the median; if it was greater than the median, the median was assigned as the concentration. If it was less than the median, the imputed value was assigned. For the serum pesticides and urine nonpersistent pesticides, the following guidelines were used: the concentration level for the nondetectable values were imputed by dividing the limit of detection by 2; the nondetectable values for the metals were set to zero.

Toxic equivalents. The total TEQ for dioxins, furans, and coplanar PCBs can be found in Tables 4–6 and has been described above. The TEQ for each congener is the product of the congener’s toxic equivalent factor (TEF) and serum concentration (ppt). The TEFs for the dioxin and furan congeners used in this study are described by the EPA (28). The TEFs for the coplanar PCBs are described by Alborg et al. (29). Because comprehensive data, including long-term carcinogenicity studies and multigenerational reproductive studies only exist for 2,3,7,8-TCDD and it is considered the most toxic congener, assignment of TEFs for the other 7 dioxins, 10 furans, and 4 coplanar PCB congeners included in this study are relative to 2,3,7,8-TCDD.

The percent TEQ for dioxins, furans, and coplanar PCBs were calculated. The total TEQ was calculated by summing the dioxin total TEQ, furan total TEQ, and coplanar PCB total TEQ. The proportion of TEQ due to dioxin was calculated by dividing the dioxin total TEQ (Table 4) by the total TEQ. The proportion of TEQ due to furans was calculated by dividing the furan total TEQ (Table 5) by the total TEQ. The proportion of TEQ due to coplanar PCBs was calculated by dividing the coplanar PCB total TEQ (Table 6) by the total TEQ. All proportions were multiplied by 100 to obtain the percent TEQ.

General population comparison laboratory values. A concurrent referent group was not identified in this study. Comparison data were chosen from previous studies to represent the general population with no apparent exposure to the analytes being studied. These groups were chosen because they were part of studies that were conducted outside of the Great Lakes basin, so there is little chance these individuals frequently consumed GL sport fish. Table 8 summarizes the comparison groups selected for each analyte. Except for the congener-specific PCBs and nickel and arsenic in urine, comparison data were chosen from studies in which laboratory analyses were conducted by the NCEH/CDC Laboratory [unpublished data; (30–32)]. This ensured consistency in the laboratory methods used for both the study and comparison samples and minimized potential interlaboratory differences.

Statistical analysis. Laboratory data were transferred electronically to the Wisconsin Bureau of Public Health and uploaded into SAS (SAS Institute, Cary, NC). All data sets were merged into a permanent SAS data set. Univariate analyses were conducted to generate ranges and measures of central tendency. The range of an analyte is the minimum and maximum value of the sample. The intent of this study was to develop descriptive profiles of environmental contaminants in human tissues of GL sport fish consumers. Kruskal-Wallis (3 stratum) and Wilcoxon (2 stratum) nonparametric tests of significant difference between lake subgroups and gender were conducted. Lake-specific group numbers are small; convenience volunteers were selected to meet minimum GL fish consumption rates and not intended to be representative of any specific regional or angling group. Therefore, we did not develop multivariate models to determine the association between fish consumption habits and serum contaminant levels.

Results

Demographic and fish consumption characteristics. Review of the interviews with participants identified one individual who reported unusual occupational/environmental exposures. This subject had been a herbicide worker, pesticide worker, farmer, fire fighter, and plasticizer maker. The subject also consumed a large amount of GL sport fish in the last 12 months (96 meals) and had the highest serum total PCB level in the group (57 ppb), nearly four times the highest level in the remainder of the group. Because of these previous exposures and this elevated total PCB level, this subject was excluded from the analysis. The remaining 31 individuals, who did not report any unusual personal, occupational, or environmental sources of exposure to the compounds being evaluated, were included in the study sample.

Table 3 contains the distribution of both demographic and fish consumption characteristics of the sample group. Of the

| Congener               | All subjects | Lake Michigan | Lake Huron | Lake Erie | Comparison group |
|------------------------|--------------|---------------|------------|-----------|-----------------|
|                        | (n = 31)     | (n = 9)       | (n = 11)   | (n = 11)  |                 |
| 3,3',4,4'-TetraCB (PCB 77) | 14.6         | 16.5          | 14.2       | 13.3      | 12.6            |
| (ND-48.8)              | (ND-48.8)    | (ND-34.9)     | (ND-24)    | (2.3-176) |
| 3,4,4',5-TetraCB (PCB 81)b | 13.5         | 17.4          | 13.2       | 13.2      | 8.5             |
| (ND-38.1)              | (ND-38.1)    | (ND-32.5)     | (ND-24)    | (2.4-33.1)|                |
| 3,3',4,4',5-PentaCB (PCB 126)b | 148          | 261           | 187        | 28        | 18.4            |
| (ND-435)               | (32.3-435)   | (38.9-427)    | (10.3-49.8)| (4.8-57.9)|                |
| 3,3',4,4',5,5'-HexaCB (PCB 169)b | 80.8         | 113           | 94.2       | 48.4      | 17.9            |
| (28.3-259)             | (43.8-259)   | (28.9-184)    | (28.5-106)| (4.4-55.2)|                |
| Coplanar PCB total (ppt)b | 229          | 340           | 282        | 75.4      | 57.4            |
| (ND-709)               | (55.5-709)   | (90.8-333)    | (ND-124)   | (NA)      |                 |
| Coplanar PCB total     | 17.4         | 28            | 23         | 1.8c      |                 |
| toxic equivalentb      | (ND-56)      | (2.8-56)      | (6.0-47.8) | (ND-8.3)  | (NA)            |

Abbreviations: ND = none detected; NA, not available.

aExposed sample residing in Jacksonville, Arkansas (n = 30).

bThree Great Lakes subgroups are statistically different (p<0.04).

cOne observation detected.

dRange not available.

Table 8. Mean and range (ppt) of coplanar polychlorinated biphenyls (PCBs) in serum, lipid adjusted.
31 subjects, 21 were men and 10 were women. The mean age of the sample was 52 years (range 36–76) and the average age was 52 years for the men and 49 years for the women. The Lake Erie fish consumers were somewhat younger with a mean age of 48 years (range 36–54). Overall mean BMI was 27 (range 18–39) with men (BMI = 28) having a higher BMI than women (BMI = 23). The Lake Erie fish consumers had the lowest BMI with a mean of 24 (range 20–30). All subjects consumed on average 77 fish meals (range 12–208) of all types in the last year, and the number of meals consumed by men and women were similar. Lake Michigan fish consumers ate on average 88 fish meals (range 24–208) of all types in the previous 12 months, while Lake Huron fish consumers ate 72 fish meals (range 12–152). The overall average number of GL sport fish meals consumed in the last year was 49 (range 10–172), the men (55 meals) consumed more than the women (34 meals), and Lake Erie fish consumers ate more than consumers from the other two Lakes. All subjects reported they had been consuming sport fish for at least 36 years, the men (37 years) in the sample reported they consumed sport fish longer than the women (32 years). The fish consumers from Lake Huron have been consuming sport fish the longest (40 years). The average number of years all subjects consumed GL sport fish was 33 years, with the men (34 years) consuming GL sport fish longer than the women (29 years). The fish consumers from Lake Huron have been consuming GL sport fish the longest (39 years). The majority of the sample reported that since they first began eating GL sport fish, the amount they ate had either remained about the same or increased.

### Nonpersistent pesticides

Ten compounds, metabolites associated with nonpersistent pesticides, were assessed in urinary samples [Table 9 (34)]. The compounds detected most often were 1-naphthol (29 detect), 3,5,6-trichloro-2-pyridinol (28 detect), 2-naphthol (27 detect), and 4-nitrophenol (25 detect). The metabolites associated with Lindane, 1,3,5-trichlorobenzene, and Propoxur were not detected in any samples. All levels of the nonpersistent pesticides were not above the established reference range.

### Metals

Tables 10 and 11 summarize the urinary and blood metal levels. All 31 subjects had detectable levels of mercury and lead in their blood. In the urine samples, 30 individuals had detectable levels of cadmium and mercury, 10 had detectable levels of

| Table 7. Mean and range (ppb) of serum levels of congener-specific PCBs |
|--------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Congener (IUPAC no.)    | All subjects   | Lake Michigan | Lake Huron     | Lake Erie       | Comparison group |
|                         | (n = 30)       | (n = 8)        | (n = 11)       | (n = 11)        | (n = 41)        |
| 28                      | 0.08 (0.03-0.2) | 0.08 (0.04-0.1) | 0.1 (0.03-0.2) | 0.06 (0.04-0.09) | ND              |
| 52                      | 0.01 (ND-0.08) | 0.06 (ND-0.08) | ND             | 0.01 (ND-0.08)  | ND              |
| 56                      | 0.02 (ND-0.02) | 0.06 (ND-0.02) | ND             | 0.04 (ND-0.02)  | ND              |
| 74                      | 0.04 (ND-0.02) | 0.06 (ND-0.02) | ND             | 0.04 (ND-0.02)  | ND              |
| 99                      | 0.06 (ND-0.02) | 0.06 (ND-0.02) | ND             | 0.04 (ND-0.02)  | ND              |
| 101                     | 0.02 (ND-0.02) | 0.06 (ND-0.02) | ND             | 0.04 (ND-0.02)  | ND              |
| 105                     | 0.03 (ND-0.02) | 0.06 (ND-0.02) | ND             | 0.04 (ND-0.02)  | ND              |
| 118                     | 0.04 (ND-0.02) | 0.06 (ND-0.02) | ND             | 0.04 (ND-0.02)  | ND              |
| 130                     | 0.05 (ND-0.02) | 0.06 (ND-0.02) | ND             | 0.05 (ND-0.02)  | ND              |
| 138                     | 0.06 (ND-0.02) | 0.06 (ND-0.02) | ND             | 0.06 (ND-0.02)  | ND              |
| 146                     | 0.07 (ND-0.02) | 0.07 (ND-0.02) | ND             | 0.07 (ND-0.02)  | ND              |
| 153                     | 0.08 (ND-0.02) | 0.08 (ND-0.02) | ND             | 0.08 (ND-0.02)  | ND              |
| 156                     | 0.09 (ND-0.02) | 0.09 (ND-0.02) | ND             | 0.09 (ND-0.02)  | ND              |
| 170                     | 0.10 (ND-0.02) | 0.10 (ND-0.02) | ND             | 0.10 (ND-0.02)  | ND              |
| 172                     | 0.11 (ND-0.02) | 0.11 (ND-0.02) | ND             | 0.11 (ND-0.02)  | ND              |
| 176                     | 0.12 (ND-0.02) | 0.12 (ND-0.02) | ND             | 0.12 (ND-0.02)  | ND              |
| 178                     | 0.13 (ND-0.02) | 0.13 (ND-0.02) | ND             | 0.13 (ND-0.02)  | ND              |
| 180                     | 0.14 (ND-0.02) | 0.14 (ND-0.02) | ND             | 0.14 (ND-0.02)  | ND              |
| 185                     | 0.15 (ND-0.02) | 0.15 (ND-0.02) | ND             | 0.15 (ND-0.02)  | ND              |
| 187                     | 0.16 (ND-0.02) | 0.16 (ND-0.02) | ND             | 0.16 (ND-0.02)  | ND              |
| 189                     | 0.17 (ND-0.02) | 0.17 (ND-0.02) | ND             | 0.17 (ND-0.02)  | ND              |
| 193                     | 0.18 (ND-0.02) | 0.18 (ND-0.02) | ND             | 0.18 (ND-0.02)  | ND              |
| 194                     | 0.19 (ND-0.02) | 0.19 (ND-0.02) | ND             | 0.19 (ND-0.02)  | ND              |
| 201                     | 0.20 (ND-0.02) | 0.20 (ND-0.02) | ND             | 0.20 (ND-0.02)  | ND              |
| 203/196                  | 0.21 (ND-0.02) | 0.21 (ND-0.02) | ND             | 0.21 (ND-0.02)  | ND              |
| 205                     | 0.22 (ND-0.02) | 0.22 (ND-0.02) | ND             | 0.22 (ND-0.02)  | ND              |
| 209                     | 0.23 (ND-0.02) | 0.23 (ND-0.02) | ND             | 0.23 (ND-0.02)  | ND              |
| PCB total                | 5.2 (1.2-15.2) | 5.2 (3.6-15.2) | ND             | 5.2 (1.2-15.2)  | ND              |

Abbreviations: ND, none detected; NA, not available.

*Sample of non-Great Lakes sport fish consumers that are part of the full study control group.

**Concentration includes congeners 28/31.

†Three Great Lakes subgroups are statistically different (p<0.04).

‡Concentration includes congeners 56/80.

§Concentration includes congeners 66/95.

An only observation.

¶Concentration includes congeners 132/153/105.

‖Concentration includes congeners 138/163.

*Concentration includes congeners 170/190.

**Concentration includes congeners 172/197.

††Concentration includes congeners 182/187.

‡‡Concentration includes congeners 190/208.
nickel and/or chromium, and only 6 subjects had detectable levels of arsenic. Overall, urine metal levels among this sample were not above the selected comparison groups.

Chlorinated persistent pesticides. Table 12 summarizes the results of the analyses of chlorinated pesticides in urine. All subjects had detectable levels of HCB, with median concentrations comparable to the comparison group. Serum from the study subjects was analyzed for two hexachlorocyclohexanes (HCH), β-HCH and γ-HCH; only β-HCH was detected in every sample at concentrations slightly greater than the comparison group except for the Michigan subjects, which were considerably higher. γ-HCH was not detected in the subjects or in the comparison group, but p,p'-DDE and p,p'- and o,p'-DDT were detected in all subjects. In the comparison group, only DDE was detected. DDE was the predominant chlorinated pesticide derivative (median 5.2 ppb) in the total group as well as in each lake subgroup. Median DDE levels were highest among the Lake Michigan fish consumers (12.2 ppb), and along with the Lake Huron group (median 5.56 ppb), levels more than twice that of the comparison group (median 2.8 ppb). Lake Erie anglers had DDE levels (median 2.1 ppb) similar to those of the comparison group, but p,p'-DDT levels were above those of the comparison group. All subjects had detectable levels of chlordane constituents or metabolites, with median concentrations the highest for trans-nonachlor, followed by oxychlordane, heptachlor epoxide, and cis-nonachlor. For the comparison group, median concentrations for trans-nonachlor and oxychlordane were the same. cis-Nonachlor was not determined in the comparison group. Dieldrin was detected in all subjects, with equal median concentrations found in Lake Michigan and Lake Huron fish consumers, concentrations that were an order of magnitude higher than those found in Lake Erie fish consumers. Dieldrin was not detected in the comparison group.

Polychlorinated dibenzoepoxides. A summary of the distribution of dioxin is provided in Table 4. Dioxin congeners detected most often were 1,2,3,4,6,7,8-heptaCDF (31 subjects), octaCDD (31 subjects), 1,2,3,6,7,8-hexaCDF (30 subjects), 2,3,7,8-TCDD (25 subjects), and 1,2,3,7,8-pentaCDF (20 subjects). The overall mean concentration for the congener considered to be the most toxic, 2,3,7,8-TCDD, was 6.6 ppb. Total dioxin was highest for the Lake Huron fish consumers and lowest for the Lake Erie fish consumers.

Polychlorinated dibenzofurans. Table 5 summarizes the results of furan congener analyses. Furan congeners that were detected most often were 1,2,3,4,6,7,8-heptaCDF (30 subjects), 2,3,4,7,8-pentaCDF (29 subjects), 1,2,3,4,7,8-hexaCDF (28 subjects), 1,2,3,6,7,8-hexaCDF (27 subjects), and 2,3,4,6,7,8-hexaCDF (23 subjects). Total furan was similar for both Lake Huron and Lake Michigan fish consumers and lowest for Lake Erie fish consumers.

Coplanar PCBs. Serum was analyzed for four coplanar PCB congeners, and summary statistics are displayed in Table 6. The most frequently detected congeners were 3,3',4,4',5-pentaCB (PCB 126; 28 subjects) and 3,3',4,4',5'-hexaCB (PCB 126; 30 subjects). Less than half of the study subjects had detectable levels of congeners 3,4,4',5'-pentaCB (PCB 81) and 3,3',4,4',5'-hexaCB (PCB 77). The congener with the highest overall mean concentration was PCB 126 (148 ppt).

Congener-specific PCBs. Congener-specific PCBs are summarized in Table 7. The congeners detected most often (30 subjects)
were PCB 28, PCB 99, PCB 138, PCB 146, PCB 153, and PCB 180. Except for PCB 101 and PCB 180, PCB levels among all subjects were above the comparison group levels for those congeners that could be directly compared. The average serum level of PCB 180 in Lake Michigan fish consumers was 0.6 ppb; the comparison group had 0.4 ppb. The mean total PCB level for all subjects was 5.2 ppb compared to 1.2 ppb for the comparison group. Lake Michigan fish consumers had the highest total PCBs (0.6 ppb) and Lake Erie fish consumers had the lowest (0.2 ppb). The data of PBBs (0.6 ppb) and Lake Erie fish consumers had the lowest highest PCB level (0.7 ppb) and residents of Wisconsin had the lowest level (0.05 ppb).

**Polybrominated biphenyls.** The identification of PBBs in the serum samples is summarized in Table 13. When the data are stratified by lake, on average, the Lake Huron fish consumers had the highest level of PBBs (0.6 ppb) and Lake Erie fish consumers had the lowest (0.2 ppb). The data were then stratified by state of residence; on average, GL sport fish consumers who live in Michigan had the highest PBB level (0.7 ppb) and residents of Wisconsin had the lowest level (0.05 ppb).

**Discussion**

Our nonrandom sample represents a subset of sport anglers whose lifestyle routinely includes frequent consumption of their catch and who did not report unusual occupational or environmental exposures to these chemicals. Our group averaged 49 GL sport fish meals a year for a mean of 33 years. A recent random telephone survey of adult residents in all eight Great Lakes states found that approximately 8% had consumed GL fish in the previous year (34). Among the GL fish consumers in that survey, the median annual consumption was 6.5 meals per year. Clearly the amount and duration of GL fish consumption of our group places them among the most highly exposed. Thus, they should represent sentinel accumulations of contaminants from GL sport fish. Our findings indicate that contaminant concentrations from the select comparison groups were lower than concentrations among the convenience sample. These findings suggest that GL sport fish consumption contributes to the body burden of select contaminants among high consumers. However, some of the comparison groups were subpopulations who reside outside the Great Lakes basin region; higher residue values in our group compared to those could possibly be influenced by regional differences in generic food and air and water exposures.

This study provides two independent means of qualitatively assessing the contribution of sport fish consumption to the measured biomarkers. First, we compare

**Table 10. Mean and range (ppb) of metals in urine**

| Analyte       | All subjects (n = 31) | Lake Michigan (n = 9) | Lake Huron (n = 11) | Lake Erie (n = 11) | Comparison group |
|---------------|-----------------------|-----------------------|---------------------|---------------------|------------------|
| Cadmium       | 0.4 (ND-1.5)          | 0.4 (ND-1.0)          | 0.3 (0.1-1.0)       | 0.4 (0.2-1.5)       | 0.7*              |
| Arsenic b     | 2.3 (0.4-6.7)         | 2.8 (1.5-5.5)         | 5.2 (0.05-10.0)     | ND                  | 7.3d             |
| Mercury       | 2.2 (ND-6.2)          | 2.1 (ND-6.2)          | 2.4 (0.4-4.6)       | 2.0 (0.9-3.2)       | 3.4f             |
| Nickel        | 1.2 (ND-15.3)         | 1.1 (ND-3.7)          | 0.2f (ND-1.5)       | 2.2 (ND-15.3)       | 0.7-5.2          |
| Chromium      | 0.2 (ND-1.6)          | 0.1 (ND-0.5)          | 0.4 (ND-1.6)        | 0.2 (ND-0.5)        | 0.19f            |

Abbreviations: ND, none detected; NHANES, National Health and Nutrition Examination Survey; CDC, Centers for Disease Control and Prevention.

**Table 11. Mean and range of metal concentrations in whole blood**

| Compound                  | All subjects (n = 31) | Lake Michigan (n = 9) | Lake Huron (n = 11) | Lake Erie (n = 11) | Comparison group |
|---------------------------|-----------------------|------------------------|---------------------|---------------------|------------------|
| Lead a (µg/dl)            | 4.0 (0.9-13.0)        | 4.2 (0.9-12.0)         | 3.2 (1.1-6.8)       | 4.8 (1.0-13.0)      | 2.8f             |
| Mercury (µg/l)            | 3.8 (1.6-7.4)         | 4.7 (1.6-7.1)          | 3.8 (2.2-7.4)       | 3.8 (1.8-5.4)       | 0.6-8.5          |

**Table 12. Median and range (ppb) of serum concentrations of chlorinated pesticides**

| Compound                  | All subjects (n = 30) | Lake Michigan (n = 8) | Lake Huron (n = 11) | Lake Erie (n = 11) | Comparison group |
|---------------------------|-----------------------|------------------------|---------------------|---------------------|------------------|
| Hexachlorobenzene         | 0.1 (0.02-0.2)        | 0.1 (0.09-0.2)         | 0.1 (0.04-0.2)      | 0.09 (0.02-0.2)     | 0.1 (ND-0.3)     |
| γ-Hexachlorocyclohexane   | ND                    | ND                     | ND                  | ND                  | ND (ND-0.2)      |
| β-Hexachlorocyclohexane   | 0.05 (ND-1.2)         | 0.05 (0.04-0.8)        | 0.05 (0.04-1.2)     | ND (0.05-0.4)       | ND (ND-1.8)      |
| Oxychlordane b            | 0.3 (0.04-0.8)        | 0.5 (0.2-0.7)          | 0.2 (0.04-0.8)      | 0.3 (0.04-0.8)      | 0.2f             |
| Heptachlor epoxide b      | 0.1 (0.04-0.5)        | 0.2 (0.05-0.5)         | 0.2 (0.04-0.8)      | 0.2 (0.05-0.5)      | 0.05 (ND-0.2)    |
| trans-Nonachlor b         | 0.6 (0.01-0.8)        | 0.9 (0.6-1.8)          | 0.3 (0.04-1.7)      | 0.2 (0.2-6.0)       | 0.2 (ND-1.8)     |
| Dieldrin b                | 0.2 (0.02-1.1)        | 0.5 (0.02-1.0)         | 0.5 (0.02-1.1)      | 0.3 (0.03-0.5)      | 0.03 (ND-2.5)    |
| cis-Nonachlor             | 0.05 (0.04-0.5)       | 0.07 (0.04-0.5)        | 0.05 (0.04-0.5)     | 0.07 (0.05-0.5)     | 0.05 (ND-0.7)    |
| p,p′-DDE f                | 0.3 (0.05-0.5)        | 0.4 (0.05-0.5)         | 0.3 (0.05-0.5)      | 0.3 (0.05-0.5)      | 0.05 (ND-0.5)    |
| p,p′-DDT f                | 0.1 (0.03-0.3)        | 0.07 (0.03-0.3)        | 0.06 (0.03-0.3)     | 0.04 (0.04-0.3)     | 0.04 (ND-0.5)    |

Abbreviations: ND, none detected; NA, not available.

*Referent values (n = 180), Arkansas population (unpublished data).
*Three Great Lakes subgroups statistically different (p<0.06).
*Comparison group values, (n = 50) follow-up sample, Arkansas population, obtained from National Center for Environmental Health/Centers for Disease Control and Prevention laboratory.
the observed levels to those of selected comparison populations. Because a concurrent referent group could not be established at the onset of the study, we used several different groups from the available published literature and unpublished data sets. For all but three analytes (urine arsenic, nickel, and congener-specific PCBs), we used comparative data developed by the NCEH/CDC laboratory. This minimized interlaboratory differences that can further add to uncertainties in our comparative evaluation. We found that few data exist which can be used to represent the general nonspor fish consuming population, and most general population comparison groups do not provide information on sport fish consumption. Despite this weakness in our study, many of the levels were higher than the comparison groups.

Second, we stratified our participants by the Great Lake where they predominantly fished. If sport fish consumption was a contributor to observed biomarker levels, the different lake-specific levels of fish contamination would be reflected by differing body burdens. Even though our lake-specific groups had similar fish consumption rates, the levels of contaminants differed: Lake Michigan and Lake Huron have similar levels, but Lake Erie has the lower body burden levels. Of the three lakes targeted by our anglers, Lake Michigan and Huron fish have quite similar levels of contamination and Lake Erie has lower fish tissue concentrations. Fish monitoring data from the EPA indicate that fish contaminant levels differ among the lakes (35). The levels are lower among Lake Erie fish and higher among fish from Lake Michigan and Huron.

Schmitt et al. (36) provided information about contaminant levels in U.S. freshwater fish. For most chemicals, the levels were higher in the fish from Lake Michigan than levels in fish from Lakes Huron and Erie. The average p,p'-DDE level in fish from Lakes Michigan, Huron, and Erie were 0.65 ppb, 0.12 ppb, and 0.04 ppb, respectively. For PCBs (Aroclor 1254), the average levels in fish from Lake Michigan were higher (1.2 ppb) than in those from Lakes Huron and Erie (0.7 ppb and 0.3 ppb, respectively). This pattern is consistent with the other chemicals tested.

Comparisons to General Population Values and Among Lakes

Nonpersistent pesticides. None of the 10 analytes associated with nonpersistent pesticides in urine had unusual mean levels compared to the selected comparison group; however, the data suggested that some interlake differences may exist for 1-naphthol and 2-naphthol. For both compounds, Lake Michigan and Erie subgroups had similar mean levels, and the Lake Huron subgroup was the lowest. There is little lake-specific environmental fish tissue data for these analytes, which makes the assessment of the contribution from fish consumption difficult. The differences among the lakes for these two compounds, although within NCEH/CDC laboratory reference range, may reflect regional or residential differences in air and water pollution or local dietary exposures. The lack of interlake differences found among the other nonpersistent pesticides may be due to the small sample size.

Metals. There are many occupational, environmental and dietary sources of exposure to metals. Elevated arsenic, organic mercury, and lead excretion have previously been associated with consumption of fish and shellfish (13,37,38).

Blood mercury is an indicator of exposure to organic mercury compounds, such as methylmercury present in fish tissue. Urinary mercury best reflects inorganic mercury exposure, primarily from nonfish-related sources. Mean blood mercury concentrations in our study group were consistently higher, both overall and by lake subgroup, than the mean from the comparison group level, but still within the NCEH/CDC laboratory reference range (<30 μg/l). Even though none of the individual values approached a level of clinical concern, the fact that mercury levels among the sample were higher than background values suggests that methylmercury from fish consumption is the explanation for this slight increase. The finding that urinary mercury levels, which reflect inorganic mercury exposure, were similar or lower than the comparison group supports our conclusion that this sample may not have been exposed to mercury from nonfish sources. The mean urine mercury levels in the study sample were lower than mean comparison group levels, which confirms the self-reports of our group that they had no unusual occupational or environmental exposure to elemental mercury.

The overall mean blood lead level of the sample was somewhat higher than the mean level of the comparison group, but well below levels of clinical concern or levels commonly seen in lead-exposed occupational groups. Our finding is consistent with a prospective study of Lake Michigan sport fish consumers (37). For those with only incidental environmental exposure to lead, sport fish consumption may be a discernable source of lead exposure.

Mean urine concentrations of arsenic, cadmium, nickel, and chromium were not unusual. Except for cadmium (30 of 31 detect), the other metals were detected in less than one-third of urine samples. Although studies of fish and shellfish consumers suggest that arsenic excretion might have been increased, we did not observe such a pattern. In our group, only six subjects (19%) had arsenic above the limit of detection (4 μg/l) and five of the six consumed fish from Lake Huron. Also of interest was the mean chromium level in the Lake Huron subgroup, which was two times that of the comparison group. Several individual nickel and chromium values fell outside the range of the comparison group, suggesting an occupational, environmental, or dietary source of exposure. Although not a high priority for our study, additional investigation of the Lake Huron area for metal contamination may be warranted.

Chlorinated pesticides. With the exceptions of HCB, heptachlor epoxide, and γ-HCH, the study group had overall median residue concentrations above the median values of the comparison groups. The elevated levels of compounds (except for p,p'-DDT) among lake comparisons followed a similar pattern, with Lake Michigan median values being the highest, followed closely by Lake Huron and then Lake Erie, which had considerably lower levels. This parallels the pattern seen in the available EPA fish tissue-monitoring data (35).

For most of the chlorinated pesticide compounds measured, the Lake Erie subgroup had the lowest median level observed. However, the median level of p,p'-DDT for Lake Erie fish consumers was higher than for Lake Huron consumers but lower than Lake Michigan fish consumers. All three lake groups had similarly low median α,p'-DDT residue levels. No α,p'-DDT was detected in the comparison group.
Among all subjects, \textit{trans}-nonachlor had the highest median level (0.6 ppb) among the eight non-DDT related pesticide residues assessed and had the second highest level after DDE (5.2 ppb). \textit{trans}-Nonachlor was followed by oxychlorodane (median 0.3 ppb) and dieldrin (median 0.2 ppb). Because the participants did not report occupational exposure to these compounds, GL sport fish consumption is likely a prominent source of exposure.

Of interest was \textit{\beta}-HCH, which was noticeably higher in the Lake Michigan group (median 0.5 ppb) than in either the Lake Huron (median 0.05 ppb), the Lake Erie (median 0.05 ppb), or the comparison groups (the median was not detectable). This may represent a contaminant of special interest and may warrant further investigation in Lake Michigan.

Our results are consistent with other studies that found DDE to be the predominant DDT metabolite present in human serum. However, our data show that other organochlorine pesticide residues, especially \textit{trans}-nonachlor, oxychlorodane, and dieldrin are also commonly detected in frequent fish consumers and should be considered when evaluating the health hazards of fish consumption.

**Polychlorinated dibenzodioxins.** Of the eight dioxin congeners evaluated, the overall mean values for four (2,3,7,8-TCDD, 1,2,3,7,8-pentaCDF, 1,2,3,4,7,8-heptaCDF; 1,2,3,4,6,7,8-heptaCDF) were clearly above the comparison group. For all four congeners, the Lake Huron subgroup had the highest mean levels, followed by the Lake Michigan subgroup, with the Lake Erie subgroup having the lowest mean levels. This order (Lake Huron > Lake Michigan > Lake Erie) is the same as for PCDFs, but different from the mean levels of chlorinated pesticides and PCBs (coplanar and other congeners). For dioxin total TEq, the same lake subgroup order persists (Lake Huron, 36.8 TEq; Lake Michigan, 25.9 TEq; Lake Erie, 20.7 TEq). All three subgroups exceed the mean TEq for the comparison group (15.5 TEq). The differences between lake subgroups observed in this sample parallel the pattern seen in the available EPA fish tissue monitoring data (35).

Our data suggest that consumption of GL fish contributes enough to the body burden of PCDDs to allow sport fish consumers to be differentiated from the general population. Statistical differences in TCDD levels between lake subgroups were found to be significantly different ($p < 0.03$). The mean of the Lake Huron subgroup is twice that of Lakes Michigan and Erie and nearly four times the background mean level. In addition, the mean PCDD levels for Lake Michigan and Lake Erie subgroups are quite comparable, and the mean for the Lake Huron subgroup was the highest. Only four PCDD congeners in our study seem to be increased in our frequent sport fish consumption group: TCDD appears the most likely to have a substantial contribution from sport fish consumption. Further investigation of PCDD congener residues in the general population is needed before the proportional contribution of GL sport fish consumption can be fully understood.

**Polychlorinated dibenzofurans.** Of the 10 PCDF congeners evaluated, four (2,3,4,7,8-pentaCDF; 1,2,3,4,7,8-hexaCDF; 1,2,3,6,7,8-hexaCDF; and 2,3,4,6,7,8-hexaCDF) had overall mean values clearly above the comparison group. For these, the rank order of the lake subgroup was not as noticeable as that seen for the PCDD congeners. For the PCDFs, mean levels for Lake Huron and Lake Michigan subgroups were comparable, and Lake Erie was the lowest.

For the PCDDs, the greatest ratios between the overall means, the lake subgroup means, and the comparison group means were seen for TCDD. The mean levels of 2,3,4,7,8-pentaCDF were more than twofold higher in the Lake Michigan and Lake Huron subgroups than in the Lake Erie subgroup. For this same congener, mean levels were four times higher in the Lake Michigan and Lake Huron subgroups than in the comparison group.

Using TEq sums to derive a composite of PCDF residues, the overall furan total TEq is twice that of the comparison group, and the furan total TEq for all three lake subgroups exceed the comparison group. While GL fish consumption appears to contribute to PCDF tissue residues, the relative contribution appears to be less than for the PCBs and chlorinated pesticides.

**Coplanar PCBs.** Of the four coplanar PCBs evaluated, the overall mean levels for PCB 77 and PCB 81 were similar or slightly higher than the comparison group. However, lake subgroup differences followed the rank order observed for the chlorinated pesticides and the other PCB congeners (Lake Michigan > Lake Huron > Lake Erie). PCB 126 and PCB 169 exhibited the greatest increase in the mean levels for all subjects, the comparison group, and lake subgroups. The mean PCB 126 level for the Lake Michigan subgroup was nearly 10 times that of the Lake Erie subgroup and almost 15 times that of the comparison group. Among all the compounds evaluated, the means for two coplanar PCB congeners—PCB 126 and PCB 169, show the greatest increase between the overall mean and the comparison group as well as among lake subgroups.

When evaluating the total TEqs, these differences are even more apparent.

The percent TEq of the coplanar PCB total TEq was found to be considerably higher than the dioxin total TEq and the furan total TEq and was distinctly different among lakes. The percent TEq due to coplanar PCBs was 45% among the Lake Michigan subgroup, 31% among the Lake Huron subgroup, 15% among the Lake Erie subgroup, and only 8% among the comparison group. In contrast, if the coplanar PCB concentrations are added to the other measured PCB congeners, they add a constant 4–5% to each subgroup and to the comparison group. Even though coplanar PCBs have similar chemical properties as the dioxin and furan congeners, they are more closely correlated with the congener-specific PCBs.

**Congener-specific PCBs.** Consistent with past studies of GL sport fish consumers, total PCBs were considerably higher in our total group, as well as in each lake subgroup, than in the comparison group. However, the mean of total PCBs is low compared to those of Hovinga et al. (7). These differences can be explained by the decrease in contaminant levels in sport fish over 20 years, as well as differences in laboratory methodology used (Humphrey (7) used packed column gas chromatography and we used capillary column gas chromatography), but the ratio of the Lake Michigan subgroup to the comparison group was quite similar. Our Lake Michigan subgroup mean was seven times the comparison group. Among lake subgroups in this study, the same congeners were the most prevalent in all groups. As seen with other residues, the mean congener-specific PCP interlake differences follow the same order as seen in the EPA fish tissue monitoring (Lake Michigan > Lake Huron > Lake Erie). Of the all comparison groups used in this study, the PCB comparison group is most representative of individuals from the GL basin. These individuals were randomly selected from GL basin cities and reported eating very little or no GL fish.

**Polybrominated diphenyls.** In 1976, dairy cattle feed in Michigan was accidentally contaminated with a fire retardant containing PBBS. Before the contamination was recognized and efforts could be made to eliminate spread through the human food chain, most residents of Michigan were exposed to PBBS through consumption of contaminated dairy and meat products (39,40). Table 13 shows that the PBBS residues may be more associated with the state of residence than the lake from which fish were consumed. This is consistent with the identified Michigan dairy product route of exposure (39). The
levels among the Wisconsin subgroup may be low because, at this time, Wisconsin exported many dairy products, which impeded the spread of PBB-contaminated Michigan dairy products into that state. However, Ohio imported most dairy products and did receive contaminated dairy products from Michigan. Therefore, the Ohio subgroup mean levels are higher than the Wisconsin subgroup but lower than the Michigan subgroup.

Mean and median tissue residue concentrations of chlorinated pesticides, PCBs (especially coplanar PCBs), dioxin and furan TEs and several specific PCDD and PCDF congeners were higher than background levels among GL sport fish consumers. Lake subgroup differences are analogous to available fish tissue monitoring data.

Our data support the conclusion that GL anglers who are life-long frequent consumers of sport fish represent a subpopulation with the potential for significant exposure to contaminants present in fish. Our sample of life-long frequent consumers of sport fish from three different Great Lakes proved to include effective sentinels for identifying sport fish contaminants of concern. Not unexpectedly, the persistent bioaccumulative compounds found in sport fish and human tissue residues had levels above those seen in the general population. The nonpersistent pesticides and most metals were not identified in unusual concentrations. A contaminant pattern among the three Great Lakes was evident. Lake Erie sport fish consumers had consistently lower contaminant concentrations than consumers of Lake Michigan and Lake Huron sport fish. These differences are consistent with the patterns seen in sport fish tissue monitoring from these lakes. Levels of most compounds were similar in Lake Michigan and Lake Huron fish consumers; however, Lake Michigan anglers had higher PCB and chlorinated pesticide residues than the Lake Huron subgroup, and Lake Huron fish consumers had higher levels of dioxin and furan congeners.

Monitoring body burdens of contaminants in a sentinel group, such as frequent sport fish consumers, can assist in evaluating the success of source reduction of contaminants and in determining whether public health advisory programs have successfully disrupted transfer through the food chain. Environmental source control has led to an initial decrease in the level of contaminants in fish (35). Acceptance of, and compliance with, public health sport fish consumption advisories should further contribute to a decrease in human exposure and accumulation of contaminant body burdens (41).

Time-trend data that assess body burdens of contaminants can be extremely valuable. Our sample of life-long frequent sport fish consumers was useful in identifying parameters to monitor. However, because of the long biologic half-life for most of the compounds, serial study of such a group may not be best suited for detecting changes over time. The impact of exposure reduction should first become evident in cross-sectional age cohort surveillance, especially in younger individuals whose body burdens reflect more recent exposures and are less confounded by exposures decades ago. Few, if any, general comparison population values exist that allow characterization of nonsport fish consumer contaminant tissue residues. Development of such comparison data sets should be a priority.

Beyond the usual total PCBs, DDT, and metabolites of DDT measured in most exposure assessment studies, we recommend two coplanar PCBs—PCB 126 and PCB 169—as priorities for future assessment and consideration in health studies. While some of the dioxin and furan congeners were increased, the ratio to the comparison group was smaller than the PCBs and chlorinated pesticides, suggesting a proportionally smaller contribution of sport fish to the tissue burdens measured. To be useful, biomonitoring surveillance needs to control for regional sport fish consumption. In addition, within the GL basin, it is important to differentiate the source of sport fish consumed.

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