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Biomonitoring of mercury and persistent organic pollutants in Michigan urban anglers and association with fish consumption

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ABSTRACT

The 32-mile Detroit River and surrounding tributaries have been designated as a Great Lakes Area of Concern due to pollution from decades of municipal and industrial discharges, sewer overflows and urban development. Key pollutants in fish samples from the Detroit River include mercury, polychlorinated biphenyls (PCBs), dichlorodiphenyldichloroethylene (DDE), dioxins and furans. A biomonitoring study was conducted to assess exposures to these persistent toxic substances in Detroit urban shoreline anglers who may be at high exposure risk due to consumption of locally caught fish. Using a modified venue-based sampling approach, 287 adult shoreline anglers along the Detroit River were recruited and participated in the program. Study participants provided blood and urine specimens and completed a questionnaire following informed consent. We examined percentile estimates for total blood mercury, PCBs, DDE, and dioxin-like total toxic equivalency (TEQ) concentrations among study participants. Multiple linear regression was used to identify important predictors of contaminant concentrations. Participants consumed a median of 64 Detroit River caught fish meals in the past year. The Detroit urban anglers' median total blood mercury concentrations was 3.2 times higher than that for the general adult U.S. population. PCB concentrations among the Detroit anglers aged 18–39 years were higher than the U.S. population of the same race/ethnicity. Elevated levels of DDE and total TEQ concentrations were not observed in the cohort. Eating more locally caught fish was associated with higher total blood mercury and serum PCB concentrations. The biomonitoring data served to inform public health officials and help guide environmental public health actions to reduce harmful exposures.

1. Introduction

Environmental contaminants have long been a problem in the Great Lakes basin. Over the past century, discharge of toxic chemicals from industrial, municipal, and non-point sources resulted in high concentrations of contaminants that persist in the sediments of lakes, streams, rivers and harbors (Adriaens et al., 2002). Despite efforts to reduce toxic discharges, legacy contaminants pose potential risks to aquatic organisms, wildlife, and humans. As a result, legacy pollutants, including mercury, polychlorinated biphenyls (PCBs), dichloro-diphenyl-trichloroethane (DDT), dioxins, and furans, are routinely monitored in fish and wildlife throughout the Great Lakes and surrounding areas (Gewurtz et al., 2011; MDEQ, 2014; EPA, 2017a; NOAA, 2017).

Great Lakes waterways have many beneficial uses, such as catching and eating local fish and wildlife and recreational water activities. The U.S.-Canada Great Lakes Water Quality Agreement defines Areas of Concern (AOC) as environmentally degraded geographic areas with restricted beneficial uses due to contamination. (EPA, 2012). The Detroit River, a 32-mile connecting channel linking Lake St. Clair and the upper Great Lakes to Lake Erie, is a binational AOC. The land use in Detroit is primarily industrial and urban development. The city of Detroit's water and sewer system, as well as 700 square miles of land in Michigan and Ontario, drain into the Detroit River. PCBs, mercury, and dioxins are the primary cause of fish consumption advisories for the Detroit River and connecting waters (MDEQ, 2014). While the Detroit River is contaminated, it also supports a large population of urban anglers, many of whom are low-income and fish for sustenance as well as recreation (Kalkirtz et al., 2008; MDEQ, 2014). Fish advisories are issued by the state to warn anglers that some species of fish from the Detroit River are highly contaminated and should not be eaten in any amount (catfish, carp), and others should be eaten only occasionally (bass, perch). Despite fish advisories, including signs posted at shoreline venues, previous studies found that shoreline anglers in this area frequently eat fish species containing high contaminant levels (Groetsch and Manente, 2010; Kalkirtz et al., 2008). As such, shoreline anglers in Detroit are at high risk of exposure to contaminants in the waterways.

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The U.S. Environmental Protection Agency (EPA) and other federal and state agencies have worked extensively to restore the AOCs on the U.S. side of the Great Lakes basin. The Great Lakes Restoration Initiative (GLRI), established in 2010 and led by the U.S. EPA, is providing resources to accelerate efforts to remediate and restore the Great Lakes and surrounding areas. GLRI activities include accelerating the cleanup of AOCs, combating invasive species, and protecting watersheds from polluted run-off (EPA, 2017b). As part of GLRI, the Agency for Toxic Substances and Disease Registry (ATSDR) established the Biomonitoring of Great Lakes Populations (BGLP) program to assess human exposure to toxic chemicals among susceptible populations (Wattigney et al., 2017). As part of the BGLP program, the Michigan Department of Health and Human Services (MDHHS) conducted a biomonitoring study targeting urban anglers. The study aimed to: (1) evaluate body burden levels of persistent toxic substances in Detroit urban (shoreline) anglers who are at high risk of exposure to persistent toxic contaminants due to regular consumption of fish from the Detroit River; and (2) assess potential exposure sources, particularly consumption of locally caught fish. The MDHHS used the biomonitoring information to guide public health actions to protect this community within their jurisdiction. This report presents total blood mercury, predominant PCBs, dichlorodiphenyldichloroethylene (DDE, a metabolite of DDT), and dioxin-like total toxic equivalency (TEQ) concentrations in Detroit urban anglers, and an assessment of potential exposure sources with a focus on locally caught fish consumption.

2. Material and methods

2.1. Participant recruitment and clinic visit

All study activities were approved by the federal Office of Management and Budget (Control Number 0923–0044) and the MDHHS Institutional Review Board. The program development and design, including sampling method, are described in more detail elsewhere (Wattigney et al., 2019). In brief, participants were recruited using a modified venue-based sampling method (Mackellar et al., 1996). Following a predetermined venue-day-time (VDT) schedule, anglers at shoreline fishing locations (venues) along the Detroit River were administered screening interviews to determine eligibility and willingness to participate in this study. Eligible anglers included respondents aged 18 years and older who ate at least two meals per month of fish from the Detroit River. MDHHS program staff conducted follow-up phone calls with randomly selected eligible VDT respondents to further confirm eligibility and schedule clinic appointments for the study.

Study clinics were held from June through December 2013 at church facilities and community centers located near the shoreline fishing locations. At the clinic visit, study staff obtained written informed consent, administered a questionnaire, measured participants' height, weight and blood pressure, and collected blood and urine samples. At the end of the clinic visit, participants received remuneration for their time and “Eat Safe Fish” guidelines (MDHHS, 2018a; MDHHS, 2018b). An interview questionnaire was administered to collect information on personal behaviors (e.g., consumption of locally caught fish, occupation, and hobbies) that could be a source of exposure to the target analytes and characteristics (e.g., gender, age, race/ethnicity) that could affect levels of the target analytes in participants' blood. In addition to eating contaminated fish, other potential sources of exposure to mercury or PCBs include some occupations or hobbies such as working with commercial electrical equipment or electronic assembly, certain recycling materials, automotive manufacturers, and chemical manufacturers (ATSDR, 2014; NIOSH, 2018). The core questionnaire domains were demographics, residential history, job history, hobbies, smoking history, recreational activities in local waterways, women's reproductive history, and dietary intake with a focus on fish consumption. Fish consumption questions included the number of years over their lifetime the respondent has eaten fish from local waters. Participants were then asked to identify species of caught fish eaten at least 5 times in their lifetime, and to recall the location and number of times each location/species was eaten in the past year. Similar questions were asked for wild birds and animals, and home-raised or home-grown food. Visual aids were used to assist in referral to specific waterways, fish species and animals. The interview also collected information on the frequency of commercially purchased fish eaten in the past year for specific species or species groupings.

2.2. Laboratory analysis

The MDHHS Analytical Chemistry Section performed laboratory analysis of mercury, DDT/DDE, and 97 PCB congeners (1, 3, 8, 11, 16, 17, 18, 22, 25, 26, 27, 28, 31, 32, 33, 37, 40, 42, 44, 45, 47, 48, 49, 52, 56, 60, 63, 64, 66/95, 70, 71, 74, 77, 81, 82, 83, 84, 87, 90, 91, 92, 95, 97, 99, 100, 101, 105, 110, 114, 118, 123, 126, 128, 130, 132, 134, 135, 136, 137, 138/163, 141, 144, 146, 149, 151, 153, 156, 157, 158, 160, 167, 170, 171, 172, 174, 175, 177, 178, 179, 180, 182, 183, 185, 187, 189, 190, 193, 194, 195, 196/203, 198, 199, 200, 201, 205, 206, 207). Mercury was measured in whole blood by inductively coupled plasma-mass spectrometry (ICP-MS) (Rodushkin et al., 1999; NCEH, 2004). PCBs and DDT/DDE were measured using gas chromatography with electron capture detection (GC-ECD) using an in-house method adapted from previous publications (Burse et al., 1990; Najam et al., 1999). Briefly, lipid separation was performed using Florisil column chromatography with 60–100 mesh pesticide residue grade packing. Further fractionation was accomplished by 3% deactivated Silica Gel. All fractions were concentrated to 1.0 mL and analyzed by GC-ECD (Burse et al., 1990; Najam et al., 1999). In accordance with program requirements, the MDHHS laboratory provided documentation of Clinical Laboratory Improvement Amendments (CLIA) certification, participated in appropriate external proficiency testing programs, provided proficiency testing results, and provided standard operating procedures for all analytical methods including detailed quality control and quality assurance procedures.

Dioxins, furans, coplanar PCBs (congeners 77, 81, 126, 169), total cholesterol and triglycerides were measured by the Centers for Disease Control and Prevention (CDC) National Center for Environmental Health (NCEH) Laboratories. Polychlorinated dibenzo-p-dioxins (PCDDs), dibenzofurans (PCDFs), and non-ortho-substituted or coplanar polychlorinated biphenyls (co-PCBs) were measured in serum by high-resolution gas chromatography/isotope-dilution high-resolution mass spectrometry (HRGC/ID-HRMS) using a previously published method (Sjodin et al., 2004). PCBs, DDE, and total TEQ serum concentrations are lipid-adjusted and presented as per gram of total lipid. These compounds are lipophilic and concentrate in the body's lipid stores, including the lipid in serum. Serum total lipids was provided by the NCEH laboratories using the enzymatic summation formula: total lipids = (2.27*total cholesterol) + triglycerides + 62.31 (Phillips et al., 1989; Bernert et al., 2007; Patterson et al., 2009). A summary of the analytes and limits of detection are provided in Supplemental Table 1.

2.3. Data analysis

Data were analyzed using the statistical software package SAS Version 9.3 (SAS Institute Inc., 2011). Descriptive statistics are presented to describe characteristics among all study participants. Percentile estimates and distribution-free 95% confidence interval (CI) estimates for chemical concentrations were calculated using the SAS® Univariate procedure. Percentile estimates and 95% CIs for all study participants are presented for total blood mercury, DDE, total TEQ, PCB 153 and 180 (the two most dominant PCB congeners in this study with 92% or more detects), and the sum of PCB congeners (ΣPCBs) with a 60% or more detection rate (PCBs 138/163, 153, 170, 180, 187, 194 and 199). The median and 95% CI for the ΣPCBs are also presented by age group and for non-Hispanic blacks (80% of study participants). TEQ values were calculated based on WHO 2005 toxic equivalency factors (Van den Berg, 2006) for PCDDs, PCDFs, co-PCBs, and mono-ortho-substituted PCBs as described elsewhere (Patterson et al., 2008, 2009).

Percentile estimates are compared to the National Health and Nutrition Examination Survey (NHANES) survey results that represent the background exposure of the U.S. population to environmental chemicals (NCHS, 2017). National-level biomonitoring data provide reference values to be able to identify comparatively high levels of exposure (Calafat et al., 2012; Saravanabhavan et al., 2017). Consistent with NHANES analysis, chemical measurements for the angler specimens below the limit of detection (LOD) were assigned a value equal to the LOD divided by the square root of 2 (NCHS, 2010). We used the most recent NHANES data available for adults aged 20 years and older for comparison. Total blood mercury in this study was compared to the NHANES 2013–2014 survey (NCEH, 2017). For POPs, individual results were only available for NHANES 2003–2004, while a weighted pooled-sample design was implemented in NHANES 2009–2010 with a published pooled-sample methodology to estimate point estimates with CIs (Caudill, 2012; Caudill, 2015). We used NHANES 2009–2010 pooled sample estimates for PCB and DDE comparisons (unpublished data). Total TEQ estimates were compared to NHANES 2003–2004 percentiles reported by Patterson et al. (2009).

In addition to descriptive analyses, biomarker data were analyzed to determine the relationship between contaminant levels and potential exposure sources. The main exposure of interest was locally caught fish consumption. For modeling potential confounders and covariates, random values from a uniform distribution between zero and the LOD were substituted for biomarker concentrations less than the LOD (Rocque and Winker, 2004). In a preliminary set of analyses, we used multivariate analysis of variance and regression models to identify participant characteristics other than fish consumption associated with each biomarker. Questionnaire items related to history of selected occupations (yes/no) and history of selected hobbies (yes/no) were explored as other potential exposure sources to the contaminants of interest.

Potential covariates included age, gender, race/ethnicity (non-Hispanic black, non-Hispanic white, other), years residing in the Detroit area, body mass index (BMI) ($< 25\,kg/m^2$-normal, 25 to < 30- overweight, and ≥30 - obese), educational attainment (high school graduate or more, less than high school graduate), annual family income (less than $50 K, $50 K or more), use of fish oil supplements (yes/no), use of herbal supplements (yes/no), recreational activities in local waterways in past year (yes/no), current smoker (yes/no), history of selected occupations (yes/no), history of selected hobbies (yes/no), consumption of fish or shellfish within the past week, number of store/restaurant fish meals in the past year, and wild game consumption. Study variables that were associated with the biomarker ($p < 0.05$) in the preliminary analysis were included in the multiple linear regression model to evaluate the association between each biomarker and locally caught fish meals. Race was included as two "dummy" variables using non-Hispanic whites as the referent (non-Hispanic Blacks and "Other"). For locally caught fish, species and location-specific data were summed into a single variable to represent the number of meals in the past year. Likewise, species-specific data were summed for purchased fish. Serum-based biomarkers were lipid standardized and covariate-adjusted using logarithm to base 10 ($\log_{10}$) total lipids as a covariate in the regression models (O'Brien et al., 2017). In addition, we $\log_{10}$-transformed biomarkers and number of fish meals to help normalize extreme values. Tests for normality and regression model diagnostics did not indicate a need for non-linear analysis. "The multiple imputation (MI) procedure in SAS® was used to impute 5 iterations of binary values for the 32 missing values for annual family income (less than $50 K, $50 K or more). The Markov Chain Monte Carlo algorithm produced imputed values that were set to 0 or 1 based on the distribution of non-missing values. The three missing values for race/ethnicity were coded as non-Hispanic black.

Table 1
Selected characteristics of Detroit Urban Anglers Study participants.

Characteristic (number excluded)*	Number (%)	Characteristic (number of missing)	Number (%)
Gender by age		**BMI - kg/m² (2)**	
Female		Normal < 25	49 (17.2%)
18–39 years	14 (25.5%)	Overweight 25–29.9	98 (34.4%)
40–59 years	25 (45.5%)	Obese 30+	138 (48.4%)
60–79 years	16 (29.1%)	**Smoking status**	
Male		Never smoked	78 (27.2)%
18–39 years	38 (16.4%)	Former smoker	65 (22.6)%
40–59 years	141 (60.8%)	Current smoker	144 (50.2)%
60–79 years	53 (22.8%)	**Education** (2)	
Race/Ethnicity (3)		Less than high school graduate	52 (18.3%)
White, non-Hispanic	27 (9.5%)	High school graduate	101 (35.4%)
Black or African American, non-Hispanic	226 (79.6%)	Some college	82 (28.8%)
Other (includes multi-racial)	31 (10.9%)	College degree	50 (17.5%)
Years residing in Detroit area (2)		**Annual Family Income** (32)	
2 years or less	< 4%**	Less than 25 K	111 (43.5%)
3–10 years	< 4%**	$25 K - less than 50 K	77 (30.2%)
11–20 years	23 (8.1%)	$50 K or more	67 (26.3%)
21 years or more	246 (86.3%)		

*A response of "don't know" or refusal and missing values.
** Percentages that represent 10 or less individuals are suppressed to protect privacy.

3. Results

Detailed recruitment results are presented elsewhere (Wattigney et al., 2019). In brief, during 439 shoreline venue-day-time visits, interview teams successfully administered eligibility questionnaires to 2,660 anglers. Of the 817 anglers initially determined to be eligible, the program recruited 287 participants. Reasons for non-participation included unable to contact, found to be ineligible via recontact phone interview, and refusals. The participants were primarily male (80.8%), 40–59 years old (57.8%) and reported being Black or African American of non-Hispanic origin (79.6%). The majority of participants had been living in the Detroit area for 21 years or longer (86.3%), tended to have a BMI of $30\,kg/m^2$ or more (48.4%), were current cigarette smokers (50.2%), achieved a high school diploma or higher level of education (81.7%), and indicated an income of less than $25,000 (43.5%) (Table 1).

Table 2 presents a summary of questionnaire interview responses to exposure related items. Participants were asked about specific jobs they had. The targeted jobs included pesticide application, recycling, lead paint removal, chemical manufacturing, and battery manufacturing. The most frequent jobs performed in the past are "worked for an automobile manufacturing company" (44.9%) which overlaps with participants who "worked in a foundry, a smelter, a welding facility or steel mill" (36.1%). Only a small percentage of Detroit urban anglers reported having performed the other specified job categories, ranging from 4.2% for "worked for a battery manufacturing or recycling company" to 19.4% who had "been a maintenance worker in any type of heavy industry." Participants were also asked about specific hobbies with potential for chemical exposure that they or someone in the home may do. The hobbies more frequently reported by participants are gardening or farming (54.9%), painting or glazing (37.3%), woodworking (20.7%), and electronic assembly (15.7%). A fish oil or omega 3 oil supplement was taken by 15% of participants. Swimming-related activities in Detroit area waterways in the past year was reported by

Table 2
Fish Consumption, work history of selected jobs, selected hobbies, and nutritional supplements with a potential of chemical exposure among Detroit Urban Anglers Study participants (n = 287).

Response (number excluded)*	Percentage, Yes
Ate fish or shell fish within the past week (1)	45.5%
As part of a job, have you ever ….	
applied pesticides that kill insects, fungus, or weeds? (1)	16.8%
worked for a trash or recycling company? (4)	15%
worked in a foundry, a smelter, a welding facility or steel mill? (2)	36.1%
removed lead paint? (14)	11.7%
worked with commercial electrical equipment such as transformers, or capacitors? (3)	10.2%
been a maintenance worker in any type of heavy industry? (4)	19.4%
worked for a battery manufacturing or recycling company? (4)	4.2%
worked for a chemical manufacturing company? (2)	9.8%
worked for an automobile manufacturing company? (2)	44.9%
In the past 12 months, hobbies and activities done by either yourself or someone inside your home.	
Dyeing material (1)	4.9%
Electronic assembly (1)	15.7%
Gardening or farming (1)	54.9%
Glass crafting (3)	< 4%**
Leather crafting (1)	< 4%**
Metal working (1)	8.7%
Painting or glazing (3)	37.3%
Printmaking (2)	4.6%
Woodworking (2)	20.7%
Other Questionnaire item	
Take fish oil or omega 3 oil supplement (2)	14.7%
Take a store-bought herbal supplement (3)	9.5%
In the past 12 months swim in the Detroit River (1)	13.6%
Ate locally caught wild game or birds in the past 12 months	11.5%
	Median (25th and 75th percentile)
Detroit River caught fish, meals in past 12 months (7)	64 (26, 155)
Years eating fish from the Detroit AOC	30 (15, 44)
Fish purchased in a restaurant or store, meals in past 12 months (20 people reported none)	28 (12, 65)

*A response of "don't know" or refusal and missing values.
** Percentages that represent 10 or less individuals are suppressed to protect privacy.

13.6% of participants.

In the past year, study participants consumed a median of 64 meals of fish caught from the Detroit River with an interquartile range of 26–155 meals. The median number of purchased fish meals in the past year was 28 with an interquartile range of 12–65 meals. Few Detroit urban anglers reported eating hunted wild birds or animals. Eating wild game such as deer, raccoon, rabbit, squirrel, or porcupine from the Detroit River AOC in the last 12 months was reported by 11.5% of participants.

Information on eating locally caught fish was gathered through a series of questions that identified species-specific and area-specific data (Detroit River and other local areas). Of the 281 participants who provided detailed frequency of caught fish consumption, 255 (90.7%) reported eating white bass caught from the Detroit River with a median of 20 meals in the past year (Supplemental Table 2). Other frequently consumed species included yellow perch (60.1%), smallmouth bass (55.2%), white perch (55.2%), rock bass (53.7%), walleye (51.6%), largemouth bass (49.1%), and catfish (45.2%). Data collected on fish purchased from a grocery store or restaurant included species and species groupings (Supplemental Table 3). In total, 267 study participants reported eating purchased fish or shellfish with a median of 31 meals in the past year. Canned tuna and "Group A" (cod, haddock, herring, freshwater perch, ocean perch, pollock, scallops, shrimp, or tilapia), were the most frequently consumed fish/shellfish (84.3% of participants). Fifty-one percent of participants ate catfish from a store or restaurant with a median of 6 meals in the past year (Supplemental Table 3). These frequently eaten store bought fish are in the lowest mercury advisory grouping with a guideline of a combined 8 meals per month (MDHHS, 2018b).

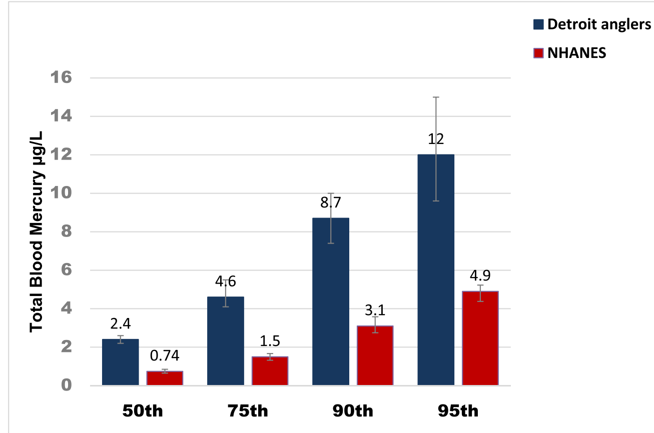

Fig. 1. Selected percentiles for total blood mercury concentration with 95% confidence intervals among Detroit urban anglers, 2013 and NHANES 2013/14.

Of the 287 study participants, 275 blood specimens were collected with 273 aliquots sufficient for total blood mercury analysis. The demographic characteristics of study participants with blood specimens are similar to all participants. Total blood mercury was detected in 93% of the 273 samples. Based on comparisons to NHANES percentiles, the Detroit urban anglers' total blood mercury concentrations were 2.4–3.2 times higher than the general U.S. adult population (Fig. 1). Twenty-three percent of Detroit urban anglers had a total blood mercury concentration above the NHANES 95th percentile (~5 µg/L). Less than 10 Detroit urban anglers had blood mercury levels above the MDHHS 'action level' of 15 µg/L for early reporting and follow-up.

DDE and 97 PCB congeners were analyzed in 275 specimens by the MDHHS laboratory. DDE was detected in 96% of the Detroit urban angler samples, and levels were about 25% and 47% lower than the NHANES 2009/10 50th and 95th percentile, respectively (Table 3). PCB concentrations in the Detroit urban angler cohort are higher than NHANES percentiles, as exemplified by ΣPCB and the two most abundant PCB congeners in the study, i.e., PCBs 153 and 180 (Table 3). Overall, the ΣPCB concentrations in Detroit urban anglers is 3.0 and 1.8 times higher than NHANES 2009/10 levels at the 50th and 95th percentiles, respectively (Fig. 2). Since 80% of the study participants were non-Hispanic blacks, we also compared our study with NHANES by race. As shown in Fig. 2, when median ΣPCB comparisons were restricted to non-Hispanic blacks, Detroit urban anglers and NHANES had similar concentrations for the age group 60 years or older, while angler's median ΣPCB concentrations were 2.5 times and 1.6 times those of the U.S. non-Hispanic black population for 20–39 years and 40–59 years age groups, respectively. Sample sizes were too small within age groups to support 95th percentile estimates – i.e., by definition, only three measures are above the 95th percentiles for age groups 18–39 years and 60 years or older. Twenty-seven percent of Detroit urban anglers aged 20–39 years, 30% of anglers aged 40–59 years and 16% of anglers aged 60 years or older had ΣPCB concentrations above the NHANES 95th percentile for similar aged non-Hispanic blacks.

The sample size for statistical analysis of dioxins, furans, and coplanar PCBs is 248, as 25 specimen results were not reported due to quality control issues. Total TEQ concentrations in the Detroit urban angler cohort are shown in Table 4. For TEQ estimates, we report the 90th and 95th percentile, because the detection frequency was less than

Table 3
p,p'-DDE and select PCB concentrations for Detroit Urban Anglers, 2013 (n = 275) and the adult U.S. population from NHANES 2009/10.

	Detroit Urban Anglers			NHANES		
Biomarker (ng/g lipid)	**50th Percentile** (95% CI)	**75th Percentile** (95% CI)	**95th Percentile** (95% CI)	**50th Percentile** (95% CI)	**75th Percentile** (95% CI)	**95th Percentile** (95% CI)
p,p'-DDE	127 (111–145)	247 (203–286)	816 (692–1394)	169 (148–192)	404 (357–459)	1531 (1194–1962)
PCB 153	48.1 (43.4–55.5)	90.8 (82.1–107)	212 (157–368)	17.1 (15.4–19.0)	37.7 (33.6–42.3)	111 (93.3–131)
PCB 180	40.5 (35.8–46.2)	69.8 (59.7–84.3)	148 (130–318)	12.7 (11.0–14.7)	30.2 (26.3–34.7)	80.9 (70.1–93.3)
Sum of PCBs*	170 (150–193)	317 (272–375)	639 (544–1333)	56.7 (49.1–65.4)	130 (114–148)	357 (303–421)

*Criteria for PCBs included is 60% or more detects in the Detroit urban angler study (PCBs 138/163, 153, 170, 180, 187, 194 and 199. The NHANES sum includes PCB 138/**158** (not PCB 138/163).

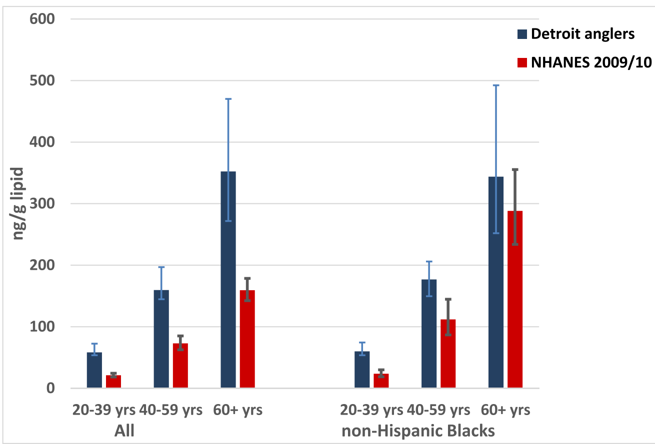

Fig. 2. Median (95% CI) for the Sum of 7 PCB concentrations for Detroit urban anglers, 2013 and NHANES 2009/10.

40% for some congeners included in the calculation. The 50th percentile is likely to be biased when the percentage of results below the detection limit is near or above 50% (Caudill et al., 2007). Total TEQ concentrations in the Detroit urban angler cohort were similar to the NHANES comparisons. The individual congeners contributing to total TEQ and their potency value (toxic equivalency factor, TEF), detection rate, 50th percentile, and 95th percentile are presented in Supplemental Table 4.

Table 5 presents the multivariate regression analyses results for mercury, DDE and ΣPCB. For the regression analysis, we combined species and location-specific fish consumption data into a single variable to represent the number of meals in the past year for locally caught fish and purchased fish, respectively. Because 91% of the study participants reported eating white bass with a median of 20 meals in the past year, and white bass is one of the most highly contaminated fish from the Detroit River, separate predictor models for individual species or fish species grouping was not plausible. Age, race/ethnicity, annual income of at least $50 k, eaten fish or shellfish in the past week, and number of Detroit River fish eaten in the past year were identified as predictors ($p < 0.05$) for total blood mercury levels. Total blood mercury increased with age, the number of Detroit River fish meals consumed in the past year, and having eaten fish or shellfish in the past week. Total blood mercury was also higher among the small number of participants whose race/ethnicity was classified as 'Other' than non-Hispanic black or non-Hispanic white. The total R^2 for these factors in explaining total blood mercury levels was 0.12 with fish consumption contributing to about half of the explained variation. Lipid-adjusted serum DDE was mostly predicted by increasing age (28% explained variation) and higher in those with race/ethnicity classified as 'Other.' The association between DDE and eating locally caught fish was not statistically significant ($p = 0.13$, < 1% explained variation). Age was the most important predictor of lipid-adjusted serum PCB concentrations, accounting for 42% of the variance in the ΣPCBs. Participants whose race/ethnicity was classified as non-Hispanic black and "Other" had higher ΣPCB concentrations than non-Hispanics whites. Eating more meals of locally caught fish was significantly associated with PCB concentrations ($p = 0.0002$, 2.9% explained variation). Age was the most important predictor of lipid adjusted total TEQ (32% explained variation) and higher in those with race/ethnicity classified as 'Other.' The association between total TEQ and eating locally caught fish was not statistically significant ($p = 0.07$, < 1% explained variation).

4. Discussion

The Great Lakes and their tributaries serve as a major navigation system for Canada and the United States to the Atlantic Ocean. Since the Industrial Revolution, it was a common practice to dispose of municipal and industrial waste into these vast waters. The Detroit River is one of many Great Lakes waterways to have persistent toxic substances from industrialization and human activities become concentrated in biota and sediments. These substances progressively bioaccumulate up the aquatic food chain and finally in humans (ATSDR, 1999; EPA, 2016). Urban anglers who fish along the shores of the Detroit River may have a disproportionately high risk of exposure to contaminants from eating their catch. Urban shoreline anglers tend to be a low-income minority subset of the Great Lakes angler population, and in many cases rely on the fish in the river as a food resource. Previous studies to assess contaminant levels in vulnerable Great Lakes populations have focused on Native American communities and sport anglers, who fish from boats and tend to be more affluent than shoreline anglers (Anderson et al., 1996; He et al., 2001; Bloom et al., 2008; DeCaprio et al., 2005; Schaeffer et al., 2006; Fitzgerald et al., 2004; Christensen et al., 2016a,b). Our study focuses on low-income minority urban shoreline anglers who eat fish from the Detroit River to assess human exposure to contaminants. These results add to our knowledge on exposure to environmental contaminants among potentially high-risk populations in the Great Lakes Basin. The MDCH used this information to improve the

Table 4
Total toxic equivalency (TEQ) values for Detroit Urban Anglers, 2013 (n = 248) and the adult U.S. population from NHANES 2003/04.

Total toxic equivalency (TEQ) values using 2005 TEFs, in pg/g lipid					
Detroit Urban Anglers		Detroit Urban Anglers	NHANES	Detroit Urban Anglers	NHANES
	Sample size	90th Percentile (95% CI)		95th Percentile (95% CI)	
TOTAL	248	31.3 (26.2–38.6)	32.5 (29.2–35.7)	40.5 (32.9–66.1)	39.9 (36.6–45.7)

Table 5
Results of multivariate regression models built with covariates correlated in preliminary analyses ($p < 0.1$) with the logarithm of blood mercury, DDE and the ΣPCB concentrations, Detroit urban anglers.

Biomarker	Parameter	10^{β}	P-value	Partial R^2
Total blood mercury*	Age at interview (years)	1.0143	0.005	0.0254
	Non-Hispanic Blacks**	1.4880	0.052	0.0022
	'Other' than non-Hispanic White or Black **	1.7342	0.029	0.0139
	Annual family income 50 K or more	1.3017	0.035	0.0155
	Ate fish or shellfish in past week (yes vs no)	1.4539	0.002	0.0442
	Number of Detroit River fish meals in the past year *	1.2407	0.011	0.0217
	Total R^2			0.1228
Total DDE, lipid adjusted*	Age at interview (years)	1.0433	< 0.0001	0.2785
	Females	1.3254	0.033	0.0123
	Non-Hispanic Blacks**	1.3534	0.092	0.0000
	'Other' than non-Hispanic White or Black**	1.7123	0.018	0.0131
	Total lipids**	1.2403	0.670	0.0007
	Number of Detroit River fish meals in the past year*	1.138	0.133	0.0060
			Total R^2	0.3107
Sum of 7 PCBs, lipid adjusted**, ***	Age at interview (years)	1.0536	< 0.0001	0.4241
	Non-Hispanic Blacks*	1.6797	0.001	0.0008
	'Other' than non-Hispanic White or Black*	2.3173	0.0001	0.0314
	Total lipids**	0.7073	0.424	0.0014
	Number of Detroit River fish meals in the past year*	1.3425	0.0002	0.0288
			Total R^2	0.4863
Total TEQ, lipid adjusted**	Age at interview (years)	1.0683	< 0.0001	0.3197
	Non-Hispanic Blacks	1.2791	0.327	0.0067
	'Other' than non-Hispanic White or Black	2.7096	0.002	0.0234
	Total lipids**	1.1003	0.893	0.0002
	Number of Detroit River fish meals in the past year**	1.2548	0.073	0.0087
			Total R^2	0.3586

* Log10 transformed.
** Referent is non-Hispanic Whites.
***Criteria for PCBs included is 60% or more detects in the Detroit urban angler study (PCBs 138/163, 153, 170, 180, 187, 194 and 199).

Michigan Fish Advisory program and to develop fish advisory outreach materials designed specifically for this subpopulation (Friends of the Detroit River, 2017; Detroit Free Press, 2017).

The Detroit urban anglers in this study had total blood mercury concentrations are 3.2 and 2.4 times higher than the NHANES 50th and 95th percentile, respectively. In addition, 23% of the study participants had a total blood mercury concentration above the 95th percentile of the adult U.S. population (5 μg/L), indicating higher mercury exposure. The NHANES 95% percentile does not imply a health-based reference level of exposure. Total blood mercury is mainly a measure of methylmercury exposure (ATSDR, 1999). Methylmercury exposures in the U.S. occur primarily through eating contaminated fish and shellfish (ATSDR, 1999; Mahaffey, 2004). Higher consumption of locally caught fish was associated with higher levels of total blood mercury among the Detroit angler cohort. Long-term methylmercury exposure has been associated with harmful neurological effects, which are of particular concern for developing fetuses, infants, and children (ATSDR, 1999). Recent studies have also shown that methylmercury exposure can increase the risk of cardiovascular disease (Genchi et al., 2017).

PCBs are industrial compounds developed in the late 1920s for an array of commercial uses including electrical applications. Manufacturing of PCBs was banned in the U.S. in 1979, however, PCBs are extremely resistant to degradation, persist in sediment and soils, and are regularly found in our waterways. PCBs have been associated with increased cancer risk, hormone disruption, and diabetes (ATSDR, 2000; ATSDR, 2011; Karmus and Zhu, 2004; Carpenter, 2006; Turyk et al., 2009). African-American ethnicity and increasing age have been shown to be related to higher PCB congener levels (McGraw and Waller 2009; Aminov et al., 2014; Pavuk et al., 2014). We found higher PCB concentrations in this study than NHANES 2009/10. Participants in this study are primarily non-Hispanic blacks, therefore, we conducted comparisons by age groups restricted to non-Hispanic blacks. Median PCB concentrations in this study were higher than NHANES 2009/10 for those aged 18–39 years and 40–59 years but similar among those aged 60 years and older (Fig. 1). NHANES 2-year survey data since 2003/04 have been showing a continuous decline in PCB serum concentrations among adult age groups (Sjodin et al., 2014). As such, our comparisons likely underestimate potential elevated PCB concentrations among the Detroit urban angler study participants.

The detection of high mercury, DDT, and PCB levels in Great Lakes fish in the 1970s evoked interest on human exposure assessment and potential human health effects (Hicks and De Rosa, 2002). Over decades, studies conducted in the Great Lakes basin have documented elevated mercury, DDT or its metabolite DDE, and PCB levels among angler cohorts in areas of contamination in relation to fish consumption (DeCaprio et al., 2005; Humphrey et al., 2000; Johnson et al., 1998; Schaeffer et al., 2006). The Michigan Fisheaters Cohort studies initially conducted in the 1970s found highly elevated total blood mercury (36.4 μg/L) among eaters of fish caught in areas of contamination; and, significantly greater DDT and PCBs, an emerging fish contaminant, among fisheaters (Humphrey and Budd, 1996). Following the ban of DDT and PCBs in the 1970s and 1980s, researchers have found lower body burdens of these chemicals in both frequent and infrequent sport fish eaters (Knobeloch et al., 2009). Over a 10 year follow-up of Great Lakes charter boat captains from 1994 to 1995 and 2001–2005, annual declines in serum DDE and PCB concentrations averaged 4.6% and 3.5%, respectively (Knobeloch et al., 2009). Despite these declines, studies of frequent sport fish consumers in the Great Lakes areas continue to find higher body burden levels of mercury and PCBs in relation to eating locally caught fish (Christensen et al., 2016a, 2016b).

The Detroit urban anglers consumed a median of 1.2 meals per week of locally caught fish which is less than the 3 or more meals per week used to define high-frequency fish consumers based on results of a national survey (Von Stackelberg et al., 2017). Contaminants vary greatly, however, among fish species and waterways. White bass, one of the most contaminated fish species in the Detroit River, was eaten by 91% of the study anglers with a median of 20 meals in the past year. Fish advisories in the Detroit area have a guideline of 1–2 servings per year for white bass (MDHHS, 2018d). Our findings of higher body burden levels of mercury and PCBs among Detroit urban anglers along with an association with eating locally caught fish demonstrate higher exposure in an area and a susceptible population that had not been studied before and call for more effective and targeted outreach on safe fish eating practices among susceptible populations in the Great Lakes basin.

Fish is known to contain high levels of nutrients, such as omega-3 fatty acid, that are beneficial to human health. Further, sport and subsistence anglers benefit nutritionally and recreationally from fishing in local waters. Therefore, our results and potential implications on fish consumption should be interpreted cautiously. Exposure profiles differ by fishing location and types of fish consumed. Prudent species-location-specific fish consumption choices can help balance the risk & benefits of eating locally caught fish (Turyk et al., 2012). Fish consumption advisories in Michigan are issued by the MDHHS based on levels of contaminants in fish tissue sampled by the Michigan Department of Environmental Quality's (MDEQ) Fish Contaminant Monitoring Program (MDEQ, 2014). The MDHHS *Michigan Eat Safe Fish Guide* includes local water/area specific consumption advisories for fish species (MDHHS, 2018c). For the Detroit River and Lake St. Claire area, white bass, catfish, smallmouth bass, and largemouth bass are the most contaminated species with a 1 to 2 servings per year guideline in 'healthy' adults and a "do not consume" guideline for women who are pregnant or plan to become pregnant. In this study, white bass was consumed by 90.7% of the Detroit urban anglers with a median of 20 meals in the last year. Our study findings indicate the need for more effective and targeted health communications on eating less contaminated caught species and store bought fish options among this population. Other frequently consumed species (45%–60% of anglers) included yellow perch, smallmouth bass, white perch, rock bass, walleye, largemouth bass, and catfish.

Low-income minority anglers in urban areas have been reported to exhibit a general lack of awareness or adherence to fish consumption guidelines (Beehler et al., 2001). Recent studies of urban anglers in the Great Lakes region have sought to characterize fish consumption patterns, awareness of consumption advisories, and factors that influence fish consumption and response to advisories. Shoreline urban angles tended to rely on perception and experience to direct their fishing practices. Water quality was perceived to be of moderate to high quality, much cleaner than it used to be, based on looks. Culture played a role in the types of fish preferred for meals despite knowledge of potential contaminant levels (Beehler et al., 2001; Kalkirtz et al., 2008). These studies highlight the importance of using community-based programs to communicate fish consumption advice for targeted subpopulations of urban anglers. Based on the findings of this study, the MDHHS was able to leverage community resources to help develop and deliver more targeted risk communication on eating fish from the Detroit River. The MDHHS staff developed and distributed brochures, easy to understand 1-page guidelines, and television 'spots' through local organizations such as the Detroit River Keepers and the Detroit Free Press (Friends of the Detroit River, 2017; Detroit Free Press, 2017).

There were some limitations to this study. First, the large number of anglers interviewed at fishing venues who were not successfully recruited introduced a possibility of bias. Reasons for non-participation include: (1) the angler reported eating less than the eligibility criteria of two or more locally caught fish meals per month; and, (2) the angler refused further participation (Wattigney et al., 2019). Second, data on fish consumption may be under or over estimated due to recall bias. Thirdly, PCB levels are consistently higher in older cohorts of both frequent and non-frequent fish consumers. The PCB levels in Detroit urban anglers aged 60 years and older is influenced by lifetime fish consumption and other exposures during time periods of peak emissions in conjunction with human metabolic rates (Quinn et al., 2012). Future studies to monitor trends in the exposure impact due to eating fish from the Great Lakes and surrounding waterways would likely benefit by focusing on younger age groups, e.g. 18–50 years (Xue et al., 2014). Another limitation is that the general population data for DDE, PCBs and total TEQ were collected in an earlier time frame than the Detroit urban angler study. We used NHANES 2009/10 as a referent for DDE, PCBs and NHANES 2003/04 for total TEQ. The comparisons with the reference levels from past years likely underestimated the extent of exposure in this study due to evidence of decreasing temporal trends for these legacy persistent organic pollutants. Average PCB 153 concentrations decreased by 36% in 12–19 year olds and 14% in people aged 60 years or older from NHANES 2003/04 through 2007/08 (Sjodin et al., 2014). Atmospheric deposition and fish tissue surveillance studies in the Great Lakes have also shown decreasing trends from 1992 to 2010 for PCB concentrations (halving times of 14 ± 2 years) and DDT (halving time of 8.7 ± 0.4 years) (Salamova et al., 2013). Lastly, it is difficult to assess whether a 1.6, 2 or 3 times higher contaminant concentration compared to NHANES data is meaningful in the context of health effects. These measurements are an indication of exposure and are not associated with disease or adverse health outcomes.

In conclusion, urban anglers in Detroit, Michigan had higher total blood mercury and serum PCB concentrations than the general U.S. population. This subpopulation of mainly low-income minority anglers had an exposure profile associated with eating locally caught fish. Fish from the Detroit River have been found to be contaminated with a multitude of chemicals including heavy metals, PCBs, dioxins, and pesticides. The fish consumption information collected from this study indicate that there may be a need for more effective and targeted health communications on fish advisories among this population. The MDHHS used the results of this biomonitoring study to develop and distribute educational materials specific to the Detroit area to promote safe fish eating practices (MDHHS, 2018c; MDHHS, 2018d). The biomonitoring data informed public health officials and helped guide environmental public health action to reduce harmful exposures.

Disclaimer

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijheh.2019.06.007.

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