



Journal of Toxicology and Environmental Health, Part A

Current Issues

ISSN: 1528-7394 (Print) 1087-2620 (Online) Journal homepage: http://www.tandfonline.com/loi/uteh20

Mercury concentrations in fish from three major lakes in north Mississippi: Spatial and temporal differences and human health risk assessment

Stacy Wolff, Garry Brown, Jingjing Chen, Keith Meals, Cammi Thornton, Steve Brewer, James V. Cizdziel & Kristine L. Willett

To cite this article: Stacy Wolff, Garry Brown, Jingjing Chen, Keith Meals, Cammi Thornton, Steve Brewer, James V. Cizdziel & Kristine L. Willett (2016) Mercury concentrations in fish from three major lakes in north Mississippi: Spatial and temporal differences and human health risk assessment, Journal of Toxicology and Environmental Health, Part A, 79:20, 894-904, DOI: 10.1080/15287394.2016.1194792

To link to this article: <u>http://dx.doi.org/10.1080/15287394.2016.1194792</u>

+	View supplementary material 🖸	Published online: 10 Aug 2016.
	Submit your article to this journal $arGamma$	Article views: 43
Q	View related articles 🖸	Uiew Crossmark data 🗹

Full Terms & Conditions of access and use can be found at http://www.tandfonline.com/action/journalInformation?journalCode=uteh20



Mercury concentrations in fish from three major lakes in north Mississippi: Spatial and temporal differences and human health risk assessment

Stacy Wolff^a*, Garry Brown^a*, Jingjing Chen^b, Keith Meals^c, Cammi Thornton^d, Steve Brewer^e, James V. Cizdziel^a, and Kristine L. Willett^d

^aDepartment of Chemistry and Biochemistry, University of Mississippi, University, Mississippi, USA; ^bCollege of Chemical Engineering, Zhejiang University of Technology, Hangzhou, Zhejiang, China; ^cMississippi Department of Wildlife, Fisheries, and Parks, University, Mississippi, USA; ^dDepartment of BioMolecular Sciences, Environmental Toxicology Research Program, University of Mississippi, University, Mississippi, USA; ^eDepartment of Biology, University of Mississippi, University, Mississippi, USA

ABSTRACT

The goal of this study was to compare total mercury (Hg) concentrations in fish muscle tissue and assess consumption health risks of fish collected from three north Mississippi lakes (Sardis, Enid, and Grenada) that are extensively used for fishing and recreation. Largemouth bass (LMB; n = 64), channel catfish (CC; n = 72), and white crappie (WC; n = 100), which represent a range of trophic levels, were collected during spring 2013 and 2014. Creel data estimated that anglers harvested approximately 370,000 kg of WC, 27,000 kg of CC, and 15,000 kg of LMB from the lakes annually. Median Hg wet weight concentrations were highest in LMB (443 ng/g), followed by CC (211 ng/g) and WC (192 ng/g). Fish-Hg concentrations were lower than those reported in fish >10 years ago. There were significant differences between lakes consistent across species. Grenada length-normalized fish-Hg concentrations were higher than those from Enid and Sardis. Because existing consumption advisories for CC are length based, the lack of relationship between length and Hg concentration indicated that the recommendations may not be sufficiently protective. Further, five different risk assessment paradigms yielded hazard quotient (HQ) values suggesting that existing fish consumption advisories may be insufficient to protect adults and especially children from exposure to Hq.

Mercury (Hg) is one of the most prevalent heavy metal contaminants in the human food chain, and consumption of fish is the primary vector for human exposure (Sweet and Zelikoff, 2001; Sunderland, 2007). Mercury from both natural and anthropogenic sources enters aquatic systems through wet and dry atmospheric deposition and through inputs from streams and rivers. Inorganic forms of Hg may be transformed to methylmercury (MeHg) by microorganisms in the sediment and elsewhere (Fitzgerald and Lamborg, 2014; Carneiro et al., 2014). Methylmercury bioaccumulates in organisms and biomagnifies in food webs, resulting in high concentrations in predatory fish, and levels generally increase with fish size and age (Gewurtz et al., 2011; Scudder Eikenberry et al., 2015; Wathen et al., 2015). Methylmercury, which

ARTICLE HISTORY

Received 30 March 2016 Accepted 24 May 2016

accounts for more than 95% of Hg found in fish muscle (Bloom 1992), is a neurotoxin that crosses the blood-brain barrier and affects neurological development and functions (Ginsberg and Toal, 2000; Sweet and Zelikoff, 2001; Karagas et al., 2012). Nearly 100% of MeHg consumed by humans is absorbed, and fetuses and children are particularly vulnerable (Ginsberg and Toal, 2000; Oken et al., 2008; Nunes et al., 2014a, 2014b).

As environmental Hg concentrations rise, so does concern for the risks of exposure to human health via fish consumption (Burger et al., 2011; Cladis et al., 2014; Shimshack et al., 2007; Vieira et al., 2015). The U.S. Food and Drug Administration (FDA) issued its first national fish consumption advisory in 2001 (U.S. FDA, 2001). The advisory was a direct response to reports released by the U.S. Environmental Protection

CONTACT Kristine L. Willett 🐼 kwillett@olemiss.edu 🝙 School of Pharmacy, Department of BioMolecular Sciences, Box 1848, 305 Faser Hall, University of Mississippi, University, MS 38677, USA; James V. Cizdziel 🐼 cizdziel@olemiss.edu 🗊 Department of Chemistry and Biochemistry, University of Mississippi, Box 1848, 380 Coulter Hall, University, MS 38677, USA.

^{*}Co-first authors.

Color versions of one or more of the figures in the article can be found online at www.tandfonline.com/uteh © 2016 Taylor & Francis

Agency (U.S. EPA, 1997) and National Academy of Science (National Research Council, 2000), which outlined the health risks associated with exposure to MeHg from contaminated fish. The FDA advisory targeted "at-risk" consumers, including women who were pregnant or nursing and young children, and advised them against eating the species of fish with the highest Hg concentrations, including shark, swordfish, king mackerel, and tilefish (Lando and Zhang, 2011; Burger et al., 2011). The problem is global, and similar directives have been issued by the World Health Organization (WHO/UNEP, 2008). Further, certain populations that eat more fish, such as sport anglers or subsistence-fishing Native Americans, are at higher risk than the general population (Tilden et al., 1997; Xue et al., 2015; Nunes et al., 2014a, 2014b).

To protect consumers from the harmful effects of MeHg exposure, state, national, and international agencies routinely measure Hg concentrations in air, water, and fish and issue recommendations and regulations concerning Hg levels (Vieira et al., 2015). Mercury is the leading cause of consumption advisories in the United States (U.S. EPA, 2010). In a national survey, 25.4% of the more than 51,000 river miles assessed exceeded the fish-tissue based water quality criterion for Hg (300 μ g/kg) (Wathen et al., 2015). The U.S. Geological Survey (USGS) maintains a national fish Hg model built from a calibrated national data set of Hg in fish tissue compiled from federal, state, and local databases (http://toxics.usgs. gov/highlights/mercury_model.html). As of 2010, all 50 states have Hg advisories in effect, with 81% of all fish consumption advisories related to Hg (U.S. EPA, 2010). Guidelines vary depending on issuing agency and fish type (Vieira et al., 2015). In the United States, the U.S. EPA uses a screening value of 300 ng/g, whereas the FDA has an action level of 1000 ng/g. The FDA jurisdiction is for commercial fish, and its action level was not intended for establishing local advisories. The European Union sets a maximum concentration for Hg in muscle meat of fish at 500 ng/g, with some exceptions, mostly for predatory fish such as sharks at 1000 ng/g.

The Mississippi Department of Environmental Quality (MDEQ) lists 11 water bodies under consumption advisories for Hg, including Enid and Grenada lakes (MDEQ, 2012). The Mississippi State Health Department recommends that individuals limit consumption of largemouth bass (LMB) and catfish exceeding approximately 69 cm in length from these waters. Mercury advisories in Mississippi are based upon 2 years or more of fish tissue data that demonstrated a large percentage of bass or large catfish with Hg tissue concentrations greater than 1000 ng/g. The agency recommends that children under 7 years and women of childbearing age should eat no more than 1 meal of these fish every 2 months, while adults should ingest no more than 1 meal of these fish every 2 weeks. Sardis Lake does not have a fish consumption advisory.

It was postulated that there may be differences in total-Hg concentrations in fish muscle (fillet) that were both species and lake dependent based upon trophic and historical data. Fish species routinely caught and consumed by anglers were targeted, as well as species representing a range of trophic levels. Fish-Hg concentrations were analyzed to (1) compare 2013–2014 concentrations to historical data (the last major study of Hg in fish from these lakes was in the 1990s) and (2) evaluate human health risk associated with consumption of the fish under different exposure scenarios.

Methods

Study Sites and Creel Data

Sardis, Grenada, and Enid lakes are U.S. Army Corps of Engineers flood control reservoirs in the upper Yazoo River Basin, the largest river basin in the state (Figure 1). At summer pool, Sardis (34°24'32" N, 89° 47'45" W), Enid (34°08'59" N, 89°54'18" W), and Grenada (33°49'48" N, 89°46'22" W) lakes are 130 km², 65 km², and 145 km², respectively. Their water levels are regulated to help control flooding in the agriculturally important Mississippi Delta. Lake levels follow a rule curve based upon monthly precipitation with a fall and winter drawdown to capture anticipated high spring rainfall and maintain a summer recreation pool. Sardis Lake has the largest watershed (approximately 3985 km²), followed by Grenada Lake (approximately 3406 km²) and Enid Lake (approximately 1153 km²).

The lakes are extensively used for recreation by the public, and are especially popular for crappie fishing, with Enid Lake holding the world record for white crappie (2.353 kg) (Straw, 2006, 2009). The lakes

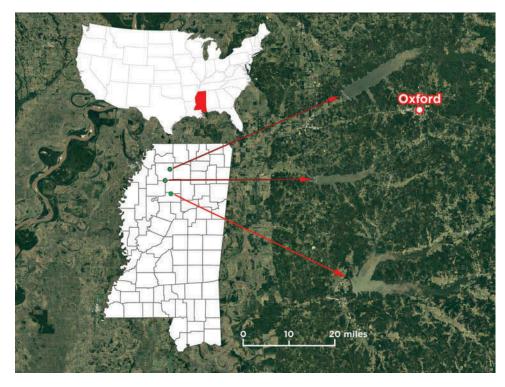


Figure 1. Aerial image and map showing Lake Sardis (top), Lake Enid (center), and Lake Grenada (bottom) in relation to the town of Oxford, state of Mississippi, and the United States (image from Google Earth).

receive hundreds of thousands of hours of fishing effort, but are also areas of concern for the Mississippi Department of Health due to MeHg. Angler roving creel data were collected by stratified random sampling (Malvestuto et al., 1978), dividing each of the lakes into sections and daily time blocks. Anglers were interviewed 8 d/mo (4 weekend days, 4 weekday days). Harvested fish were identified by species and aggregate of each species harvested and weighed. Effort and harvest data were expanded based upon proportions of anglers in each time/section for the day and month to obtain annual estimates. White crappie dominated angler harvests, with Sardis > Grenada > Enid. Anglers kept more LMB at Sardis and more CC at Enid (Table 1). Individual mean fish weights harvested by anglers (Table 1) were similar to

Table 1. Estimated annual angler harvest in kg and in parentheses mean individual fish weight kept by anglers (kg).

<u> </u>				
Sardis Lake	Grenada	Enid Lake		
(2013)	Lake (2011)	(2014)		
246,940 (0.55)	65,406 (0.64)	60,365 (0.58)		
8,032 (0.80)	5,737 (0.71)	1,102 (1.00)		
9,775 (0.58)	2,497 (0.48)	14,463 (0.52)		
	(2013) 246,940 (0.55) 8,032 (0.80)	(2013) Lake (2011) 246,940 (0.55) 65,406 (0.64) 8,032 (0.80) 5,737 (0.71)		

fish analyzed for Hg in this study (Table 2): WC, 0.32–0.89 kg; LMB, 0.35–1.86 kg; and CC, 0.2–0.92 kg.

Fish Collection

Fish from Grenada and Sardis lakes were collected by the Mississippi Department of Wildlife, Fisheries, and Parks (MDWFP), while fish from Enid Lake were collected by the USDA National Sedimentation Laboratory. In total, 236 fish from 3 different species, white crappie (Pomoxis annularis, WC), largemouth bass (Micropterus salmoides, LMB), and channel catfish (Ictalurus punctatus, CC), were caught by electro-shocking during spring 2013 and 2014. In 2014, fish were only collected from Grenada and Sardis lakes to confirm differences found in fish-Hg concentrations between these lakes in 2013. Fish were collected from Grenada Lake at Young's, Bryant, and Gums Crossing landings, from Sardis Lake at the Hurricane Creek and Teckville landings, and from Enid Lake at Cossar State Park. Fish were placed on ice in a cooler, and were taken to the University of Mississippi for analyses. At the lab, fillets were

Table 2. Summary of mercury concentrations in skeletal muscle of fish from Grenada, Enid and Sardis Lakes in 2013 and 2014. For each species, lakes not connected by the same letter are statistically significantly different (p < 0.05). Values in red represent Hg concentrations that exceed the EPA screening value (300 ng/g). The values in green exceed the FDA action level (1000 ng/g). na = not analyzed because ANOVA assumptions were met. LS-mean = least squares mean from ANCOVA analyses.

				Fish Length (cm)			Fish Weight (g)			Mercury (ng/g, wet mass)						
Year	Lake	Species	n	Median	Min	Max	Median	Min	Max	Mean	Median	Min	Max	LS-mean	Significance	
2013	Grenada	Largemouth Bass	8	40.2	36.6	48.5	904	673	1415	630	541	351	1066	462	Α	
	Enid	-	9	38.4	30.9	51.5	610	352	1862	386	344	189	954	360	A,B	
	Sardis		19	38.5	31.0	49.2	800	417	1810	334	279	102	723	267	В	
	Grenada	Channel Catfish	9	35.0	31.6	39.8	269	200	510	395	440	261	666	na	А	
	Enid		14	34.6	31.0	41.2	370	223	671	147	146	84	272	na	С	
	Sardis		18	41.0	31.0	49.0	498	217	919	222	190	142	432	na	В	
	Grenada	White Crappie	20	35.0	30.5	38.6	692	392	887	199	180	99	383	na	А	
	Enid		16	32.4	29.8	34.5	449	320	527	214	215	120	285	na	А	
	Sardis		20	32.8	30.5	36.0	559	380	749	147	138	109	237	na	В	
2014	Grenada	Largemouth Bass	12	40.1	23.0	50.5	760	155	1875	677	642	437	1154	676	А	
	Sardis		16	41.2	23.0	54.5	843	141	1841	445	496	95	838	446	В	
	Grenada	Channel Catfish	16	33.5	21.2	53.7	287	66	1226	382	241	138	1215	364	А	
	Sardis		15	33.5	14.5	64.0	248	119	2367	210	188	114	385	213	В	
	Grenada	White Crappie	24	33.8	11.2	41.2	584	106	1015	361	362	81	497	361	А	
	Sardis		20	32.0	24.9	43.0	465	33	949	196	176	97	518	196	В	

collected using a titanium knife, placed in labeled plastic bags, and frozen until analyzed.

Determination of Mercury (Hg) Concentrations

Total-Hg is commonly used as a proxy for MeHg because the latter accounts for more than 95% of Hg found in fish muscle (Bloom, 1992). In this study, total-Hg concentrations were determined in the fish muscle on a wet weight basis using a direct mercury analyzer (DMA-80; Milestone, Inc.) following U.S. EPA Method 7473 (U.S. EPA, 2007). Briefly, the DMA-80 is an automated system that is based upon sample combustion, isolation of Hg by amalgamation with gold, and detection using atomic absorption spectrometry. The DMA was calibrated using an Hg standard solution (SPEX Certiprep, Metuchen, NJ). Fish samples were allowed to thaw and approximately 0.2 g of tissue was placed in nickel boats and inserted into the instrument autosampler. The limit of detection (LOD) was 0.01 ng Hg based upon replicate measurements of a lowlevel Hg standard. Using 200 mg of tissue, the LOD determined in this study corresponds to a level of 0.05 ng/g. Certified reference material DOLT-2 (National Research Council of Canada) was run every 10th sample to check accuracy, and recoveries were between 80 and 120%. A second sample of axial muscle was collected and analyzed from every 10th fish, and reproducibility was less than 20% relative percent difference each time.

Exposure Assessment and Consumption Limits

Exposure to MeHg via fish consumption was estimated using methods outlined by the U.S. EPA (http://www2.epa.gov/region8/hh-exposure-assess ment). Risk assessment studies focused on WC, LMB, and CC, the most harvested species of fish in these lakes. Because fish were not collected from Enid in 2014, fish-Hg concentrations from 2013 were used for risk assessments. Calculations of intake rate (Eq. (1)), hazard quotient (HQ) (Eq. (2)), and monthly consumption limit (MCL) (Eq. (3)) were conducted using assumptions outlined in Table 3:

Intake Rate
$$(mgkg^{-1}day^{-1}) = \frac{CF \times IR \times EF \times ED}{BW \times AT}$$
(1)

where CF is the Hg concentration in fish (mg kg⁻¹), IR is ingestion rate (kg meal⁻¹), EF is exposure frequency (meals year⁻¹), ED is exposure duration (year), BW is body weight (kg), and AT is averaging time (ED x 365 days year⁻¹);

$$HQ = \frac{intake \ rate}{RfD}$$
(2)

where intake rate is calculated using Eq. (1) and RFD is the reference dose for MeHg (1×10^{-4} mg kg⁻¹ day⁻¹). The reference dose is an estimated exposure dose at which no adverse effects will be suffered, even if the exposure is long-lasting. The HQ is a ratio of an

Table 3. Values used for Risk Assessment Calculations.

Source	Intake rate, kg meal ^{–1} (oz)	Body weight (kg), adult (child)	Exposure frequency, meals year ⁻¹
Huggett (2001); U.S. EPA (1989)	0.227 (8)	70 (14.5)	48
U.S. EPA Exposure Factor Handbook	0.227 (8)	80 (16)	48
NOAA (2011) and U.S. EPA Exposure Factor Handbook (2011)	0.142 (5)	80 (16)	48
MDEQ Consumption Advisory (2002)	0.227 (8)	70 (14.5)	24
FDA (2001) and U.S. EPA Exposure Factor Handbook (2011)	0.170 (6)	80 (16)	96

individual's actual exposure over a time period (here, 30 years) to the reference dose established by the U.S. EPA. If HQ < 1, the expected potential for toxicity is low, and exposure is considered safe.

$$MCL = \frac{RfD \times BW}{C_m} \times \frac{30.44 \frac{days}{month}}{IR}$$
(3)

where RfD is the reference dose, BW is body weight, C_m is concentration in fish, and IR is ingestion rate. MCLs are often provided with consumption advisories released by the U.S. EPA, FDA, and state agencies. Each MCL calculation requires certain assumptions with respect to body weight, ingestion rate, consumption frequency, and mean meal size of the targeted population. Risk assessments using five different parameters (Table 3), including the 1989 EPA assumptions used by Huggett et al. (2001) for fish from these same lakes collected in the 1990s, were compared.

Data Analysis

Statistical analysis was conducted using Microsoft Excel, GraphPad Prism 4.0 and JMP v. 5.0.1 software. Linear regression and analysis of variance (ANOVA) was used to determine the relationship between Hg concentrations, fish weight, and fish length with p < .05 considered statistically significant. Linear regression analysis showed a strong direct relationship between fish length and weight for each species in each lake (Supplemental Figure 1). To compare Hg concentrations between lakes, data was log-transformed to reduce heterogeneity in variances among lakes to satisfy the assumptions of ANOVA/analysis of covariance (ANCOVA). For WC and CC, ANOVA was used because neither length nor weight was correlated with Hg concentration in 2013. For 2013 LMB and all three species in 2014, both weight and length were positively correlated with Hg concentration. Thus, in these cases, ANCOVA was employed using length as a covariate. Because length and weight were highly correlated with each other, only length was included in the ANCOVA to avoid multicollinearity effects. Adding the covariate did affect the interpretation of the analyses, as indicated by differences between least square means and unadjusted means. Tukey's tests were used to provide pairwise differences in means among lakes in 2013.

Results and Discussion

Fish-Hg Concentrations and Lake Comparisons

Summary statistics for fish size and Hg concentrations are presented in Table 2. Of the three fish species collected from each lake, LMB consistently displayed the highest mean Hg concentrations (in 2013, Grenada: 630 ± 104 ng/g, Sardis: 334 ± 40 ng/g, Enid: 386 ± 36 ng/g). The mean Hg tissue concentrations in LMB from each lake exceeded the U.S. EPA 300 ng/g human health criterion. The overall median Hg tissue level in LMB was 442 ng/g. In comparison, the median concentrations detected in CC and WC were 211 and 192 ng/g, respectively. Based on knowledge of the species, this pattern was consistent with each species' trophic level, namely, that LMB are carnivores, feeding mostly on shad, but also centrarchids and crayfish; CC are omnivores and bottom dwellers; and WC feed on insects, crustaceans, and smaller fish species. As bottom dwellers, CC may be exposed to Hg in sediment.

All three fish species collected from Grenada Lake had higher length-adjusted Hg tissue concentrations than those from Sardis Lake, but LMB and WC from Enid and Grenada were not markedly different (Table 2). Comparing across the lakes, white crappie, the species with the greatest angler harvest, displayed the highest Hg levels in Grenada, followed by Enid, and Sardis.

The source of Hg in these lakes has not been adequately studied, but likely includes atmospheric deposition and watershed runoff. Given similar watershed characteristics, land use (approximately 40% agricultural and 35% forested), and a lack of major point sources (with one possible exception), the difference in fish-Hg between Grenada and Sardis is intriguing. The point-source exception is the 514-MW Red Hills coal-fired power plant located approximately 65 km southeast of Grenada Lake, which released 186 kg Hg in 2010 (U.S. EPA 2010 Toxic Release Inventory). Sediment Hg concentrations were not measured in this current study; however, an earlier study found comparable concentrations between the lakes (Grenada 39-133 ng/g; Enid 34-154 ng/g; Sardis 31-112 ng/g) (Huggett et al., 2001). It is worth noting that Grenada Lake is shallower than the other lakes, resulting in more wind-prone resuspension of bottom sediments.

Compared to historical data, overall Hg concentrations in Enid Lake fish decreased. For fish collected from Enid Lake in 1999, mean concentrations were 1400 ng/g for LMB (n = 5), 820 ng/g for catfish (n = 4), and 1690 ng/g for black crappie (n = 3)(Huggett et al., 2001). A larger fish-Hg data set for Enid Lake was reported by MDEQ based on available data from 1994–1999 (MDEQ, 2002). In that investigation, 57% of LMB (n = 47) showed concentrations exceeding 1000 ng/g, whereas only a 25% incidence (16 out of 64) was noted here. This decline is in contrast to a 1969 to 2005 trend reported for increasing Hg levels in fish tissue collected in the southeastern United States associated with elevated wet deposition of Hg in the region (Chalmers et al., 2011). Whether Hg concentrations declined due to reduced deposition, anglers removing Hg through harvest, or some combination is unknown.

Fish Length Versus Hg Concentration

Length might reflect the age of the fish, and because older fish tend to possess higher Hg concentrations, length sometimes provides a general idea regarding the extent of Hg bioaccumulation in fish tissues (Gewurtz et al., 2011; Rypel, 2010; Scudder Eikenberry et al., 2015). Many risk assessments

and resulting consumption advisories assume a direct relationship between fish length and Hg concentration. Factors such as diet and metabolism may also affect Hg bioaccumulation. Although there was a trend toward a positive relationship between Hg concentration and length in 2013 Grenada Lake CC (Figure 2A), when combined with a lack of such an association in the other two lakes, the interaction between lake and length was not significant for any of the three angler-harvested species in 2013. Largemouth bass was the only species that showed a significant elevation in Hg concentration with fish length in all three lakes. The partial R^2 of .66 suggests that 66% of variation in LMB Hg tissue concentrations was associated with length. In contrast, partial R^2 values for CC and WC were .03 and .02, respectively. The lack of a consistently positive correlation between fish length and tissue Hg levels is problematic, considering the CC consumption advisory is length based.

Risk Assessment

Although maximal acceptable Hg concentrations in fish may vary between agencies due to different purviews, these are all generally designed to minimize risks associated with human or wildlife exposure to MeHg. Human health risk assessments may be conducted to assess hazard to adults and children associated with consumption of contaminated fish. The assessments serve as guides for issuing consumption advisories and consumption limits. Because risk assessments require making assumptions regarding body weight and consumption habits of the population, the U.S. EPA recommends selecting values that are most relevant to the specific audience, including type of fish sampled and age group targeted (U.S. EPA, "Intake of Fish and Shellfish," 2011; U.S. EPA/ FDA, "Fish: What Pregnant Women and Parents Should Know," 2014). Creel data indicate that despite regulatory advisories, anglers are harvesting significant quantities of fish from these water bodies for consumption (Table 1). For example, LMB collected from Grenada had an adult HQ > 1 by all 5 risk calculations. Similarly, MCLs calculated for children were ≤ 2 meals per month regardless of species or lake.

Five risk assessment calculations for each fish species and lake were undertaken varying at least one assumption value each time (Table 3). To

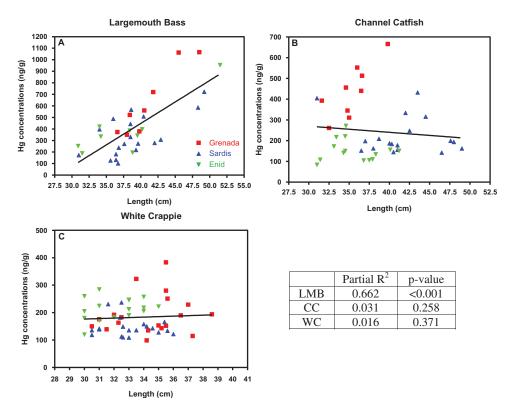


Figure 2. Covariate analyses of fish length vs. Hg concentration data from Grenada, Sardis, and Enid lakes. The table lists the partial R^2 values for each line and the *p* values for the lake effect (statistical comparisons were done on log transformed data). The LMB *p* values indicate a significant relationship between fish length and fish weight.

compare with previous studies, assumptions used by Huggett et al. (2001) served as a starting point for our calculations and analysis. Using an ingestion rate of $0.227 \text{ kg meal}^{-1}$ (8 oz), adult and child body weights of 70 kg and 14.5 kg, respectively, and an exposure frequency of 48 meals yr^{-1} (4 meals per month), ingestion of LMB from any of the lakes and CC from Grenada would represent increased risk of adverse effects in adults (e.g., HQ > 1, Table 4). The mean consumption limits using these assumptions suggest only one half to one meal per month is safe for children. The MDEQ has issued a fish consumption advisory warning the public not to consume more than 2 meals/mo (24 meals/yr) of catfish larger than 27 inches. When 24 meals/yr was used for the consumption frequency and 8 oz of fish/meal in the risk assessment calculations, only LMB from Grenada Lake had HQ > 1, but notably none of the catfish in this study were 69 cm long.

Recently, mean body weights for adults and children have increased. The 2011 *EPA Exposure Handbook* suggests employing a mean body weight of 80 kg for adults and 16 kg for children (U.S. EPA, 2011). While using the higher body weights did result in a lower calculated HQ and higher MCL, the overall conclusion for the risk assessment did not change. However, the National Oceanic and Atmospheric Administration (NOAA) estimates an mean consumption of 15 lb seafood per person per year (NOAA, 2011) and a 5-oz (vs. 8-oz) serving size. Utilizing higher body weights and smaller serving sizes (middle columns Table 4), only CC and LMB from Grenada Lake would result in HQ > 1 for adults, but all fish from all lakes would still exceed the HQ for children.

Worldwide, fish are an important source of protein and nutrients including amino acids, vitamins, and unsaturated fatty acids (Knuth et al., 2003; Nunes et al., 2014b; Carneiro et al., 2014; Vieira et al., 2015; Lunder and Sharp, 2014). For this reason, FDA (2001) suggested that U.S. residents include 6 oz of fish in their diet twice a week (8 meals/mo). Using this frequency, LMB and CC would exceed the acceptable HQ at every lake. Even WC from Grenada and Enid would also result in HQ > 1. In general, regardless of risk assumptions, most of the calculated MCLs for

Table 4. Comparison of mean HQ (Hazard Quotient) and MCL (Mean Consumption Limit) for three representative risk assessment assumptions. HQ values in red are above the EPA's current recommended threshold of HQ = 1, while green MCL values exceed the number of meals per month allowed given the assumptions used in the calculation. See Table 1 for complete listing of assumptions made foreach set of risk assessment calculations. MCL values are conservatively rounded down to the nearest 0.5 meals.

	Species	Huggett (2001); EPA (1989) 8 oz/meal, 4 meals/month, BW=70 or 14.5kg						(2011) 5 c 3W=80 or		FDA (2001) & EPA (2011) 6 oz/meal, 8 meals/month, BW=80 or 16kg			
		Mean HQ		MCL		Mean HQ		MCL		Mean HQ		MCL	
Lake		Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child	Adult	Child
Grenada	WC	0.9	4.1	5.0	1.0	0.5	2.3	9.5	1.5	1.1	5.6	8.0	1.5
	LMB	1.6	7.6	3.0	0.5	1.5	6.9	3.0	0.5	3.5	18	2.5	0.5
	cc	1.8	8.8	2.0	0.5	1.0	5.1	4.0	0.5	2.4	12	3.5	0.5
Sardis	WC	0.6	3.0	6.0	1.0	0.3	1.7	12.0	2.0	0.8	4.1	10	2.0
	LMB	1.4	6.9	3.0	0.5	0.8	3.9	6.5	1.0	1.9	9.3	5.5	1.0
	cc	1.0	4.6	4.0	1.0	0.5	2.6	8.5	1.5	1.2	6.2	7.0	1.5
Enid	WC	0.9	4.4	4.0	1.0	0.8	4.0	8.0	1.5	1.2	6.0	7.0	1.5
	LMB	1.7	8.0	3.0	0.5	0.9	4.5	5.0	1.0	2.2	11	4.5	1.0
	cc	0.7	3.1	7.0	1.0	0.4	1.8	12.0	2.0	0.9	4.3	10.5	2.0

Note. The assumption that was varied for each set of calculations is listed in red.

adults were well below 8 meals/mo. Advisories for children and pregnant women are more conservative to minimize exposure to Hg during critical stages of neurodevelopment. For children, the calculated MCL was 2.5 or less for all of the risk assessments completed. Ginsberg and Toal (2000) suggested that there should be a single-meal concentration cutoff during pregnancy of $\geq 2 \mu g/g$. The highest single fish concentration found in this study was 1.15 $\mu g/g$.

The current fish consumption advisories for Grenada and Enid lakes warn locals to minimize intake of LMB and to avoid eating catfish longer than 68.58 cm. The existing advisories do not place a length limit on WC. For WC population sustainability during the study period, the MDWFP enforced minimum length limits of 27.9 cm on WC from Sardis Lake and 30.5 cm on WC from Enid and Grenada lakes. The mean length for crappie collected and analyzed for this study was 33 cm. The smallest WC with HQ > 1 from each lake was Grenada, 33.6 cm; Sardis, 32.5 cm; and Enid, 33 cm.

Figure 3 shows length versus HQ (calculated using the Huggett/U.S. EPA assumptions) for all the 2013 fish. All CC collected in this study were well below the 69-cm guideline issued by the MDEQ for CC and thus would be considered safe to consume. However, the calculated adult HQ for many of the LMB and CC was greater than 1. Nearly all of the fish collected, regardless of species or lake, would have HQ > 1 for children. Therefore, the length-based consumption advisories issued by MDEQ for CC from Grenada and Enid lakes are not sufficiently conservative to protect the public from exposure to Hg. Further, Sardis Lake CC and LMB also need to be included in the advisory. As illustrated in Figure 2, WC and CC also did not exhibit a strong direct relationship between length and Hg concentration. This analysis suggests that a lengthbased consumption advisory for these species does not offer protection from MeHg.

There are several limitations of this study that need to be acknowledged. The study was not designed to track fate, transport, seasonality, or sources of Hg into these water bodies. Historical data from these specific waterways are limited such that it was not possible to statistically analyze differences from past data. Bearing this in mind, data indicated that keeping one water body (Sardis) out of the current MSDEQ advisory is not supported by the more recent findings. In addition, potential for children's risk overestimation is associated with using the same intake for children and adults. Lunder and Sharp (2014) suggested estimating children's fish intake to be 1 oz per 20 lb of body weight. The point estimate for children's body weight 14.5 kg (~32 lb) would suggest serving sizes of only 1.6 oz, compared to 5 to 8 oz sizes estimated for adults. Thus, our results are conservative with respect to protecting children if they ingest less. Future population-based studies would enable more accurate consideration of fish intake and population demographics such that risk assessments would be less point estimate dependent. Such studies were conducted with Portuguese cohorts (Nunes et al., 2014a, 2014b).

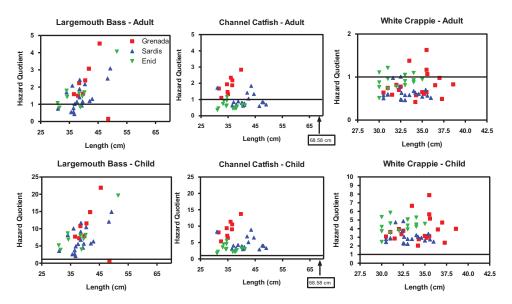


Figure 3. Fish length vs. hazard quotient HQ (calculated using assumptions in Huggett et al., 2001; U.S. EPA, 1989) for largemouth bass, channel catfish, and white crappie. The arrow below the CC *x*-axis shows where 27 inches (the fish length-based consumption advisory) falls compared to the fish collected.

Conclusions

Thousands of kilograms of LMB, CC, and especially WC were harvested by sport anglers for consumption from these three north Mississippi lakes. Largemouth bass tended to display the highest Hg concentrations, followed by CC and WC, which was consistent with each species' predicted diet and trophic level. Mean Hg concentrations in LMB from all three lakes exceeded the U.S. EPA threshold concentration of 300 ng/g. While fish Hg concentrations were lower than those reported a decade ago, based upon risk calculations, the existing length-based consumption advisory for CC issued by the MDEQ may not be sufficiently protective, and intake recommendations for fish may be too high to be safe for the general population. Fish Hg concentrations were higher in fish from Grenada Lake compared to Enid and Sardis lakes, and further research is necessary to identify potential point sources of Hg into these water bodies.

Funding

This research was partially funded by the U.S. Geological Survey and Mississippi Water Resources Research Institute: sub award number 440502-363465.01. Fish collections were provided by the Mississippi Department of Wildlife, Fisheries, and Parks and by Scott Knight at the USDA National Sedimentation Laboratory. Dr. Julia Gohlke (Virginia Tech) provided helpful suggestions during article preparation. We thank Sara Adams for help with the fish-Hg analyses. Jingjing Chen is co-advised by Dr. Ying Zhou at Zhejiang University of Technology.

References

- Bloom, N. S. 1992. On the chemical form of mercury in edible fish and marine invertebrate tissue. *Can. J. Fish. Aquat. Sci.* 49: 1010–1017.
- Burger, J., Jeitner, C., and Gochfeld, M. 2011. Locational differences in mercury and selenium levels in 19 species of saltwater fish from New Jersey. J. Toxicol. Environ. Health A 74: 863–874.
- Carneiro, M. F. H., Grotto, D., and Barbosa, F. 2014. Inorganic and methylmercury levels in plasma are differentially associated with age, gender, and oxidative stress markers in a population exposed to mercury through fish consumption. *J. Toxicol. Environ. Health A* 77: 69–79.
- Chalmers, A. T., Argue, D. M., Gay, D. A., Brigham, M. E., Schmitt, C. J., and Lorenz, D. L. 2011. Mercury trends in fish from rivers and lakes in the United States, 1969–2005. *Environ. Monit. Assess.* 175: 175–191.
- Cladis, D. P., Kleiner, A. C., and Santerre, C. R. 2014. Mercury content in commercially available finfish in the United States. J. Food Prot. 77: 1361–1366.
- Fitzgerald, W. F., and Lamborg, C. H. 2014. Geochemistry of mercury in the environment. In *Treatise on geochemistry*, 2nd ed., vol. 9, 1–47. Elsevier Science, Amsterdam, Netherlands
- Gewurtz, S. B., Bhavsar, S. P., and Fletcher, R. 2011. Influence of fish size and sex on mercury/PCB concentration: Importance for fish consumption advisories. *Environ. Int.* 37: 425–434.
- Ginsberg, G. L., and Toal, B. F. 2000. Development of a single-meal fish consumption advisory for methyl mercury. *Risk Anal.* 20: 41–47.

- Huggett, D. B., Steevens, J. A., Allgood, J. C., Lutken, C. B., Grace, C. A., and Benson, W. H. 2001. Mercury in sediment and fish from North Mississippi lakes. *Chemosphere* 42: 923–929.
- Karagas, M. R., Choi, A. L., Oken, E., Horvat, M., Schoeny, R., Kamai, E., Cowell, W., Grandjean, P., and Korrick, S. 2012. Evidence on the human health effects of low-level methylmercury exposure. *Environ. Health Perspect.* 120: 799–806.
- Knightes, C. D., Sunderland, E. M., Craig, B. M., Johnston, J. M., and Ambrose, R. B. 2009. Application of ecosystemscale fate and bioaccumulation models to predict fish mercury response times to changes in atmospheric deposition. *Environ. Toxicol. Chem.* 28: 881–893.
- Knuth, B. A., Connelly, A., Sheeshka, J., and Patterson, J. 2003. Weighing health benefit and health risk information when consuming sport-caught fish. *Risk Anal.* 23: 1185– 1197.
- Lando, A. M., and Zhang, Y. 2011. Awareness and knowledge of methylmercury in fish in the United States. *Environ. Res.* 111: 442–450.
- Lunder, S., and Sharp, R. C. 2014. US gives seafood eaters flawed advice on mercury contamination, healthy omega-3 fatty acids. Environmental Working Group, Washington, DC.
- Malvestuto, S. P., Davies, W. D., and Shelton, W. L. 1978. An evaluation of the roving creel survey with non-uniform probability sampling. *Trans. Am. Fish Soc.* 107: 255–262.
- Mississippi Department of Environmental Quality. 2002. Yocona River and Enid Reservoir Phase One Total Maximum Daily Load for Mercury. TMDL/WLA Section, Jackson, MS.
- Mississippi Department of Environmental Quality. 2007. Yazoo River Basin. Retrieved from http://www.deq.state. ms.us/MDEQ.nsf/page/WMB_Yazoo_River_Basin? OpenDocument
- Mississippi Department of Environmental Quality, Office of Pollution Control, Field Services Division. 2012. *State of Mississippi water quality assessment* 2012 *section* 305 *b report*. http://www.deq.state.ms.us/MDEQ.nsf/pdf/FS_MS_12_ Section_305b_WQA_report/\$File/2012_305b_report.pdf? OpenElement
- National Oceanic and Atmospheric Administration, National Marine Fisheries Service. 2011. *Per capita consumption*. http://www.st.nmfs.noaa.gov/st1/fus/fus11/08_perca pita2011.pdf
- National Research Council. 2000. *Toxicological effects of methylmercury*. Washington, DC: National Academy Press.
- Nunes, E., Cavaco, A., and Carvalho, C. 2014a. Exposure assessment of pregnant Portuguese women to methymercury through the ingestion of fish: Cross-sectional survey and biomarker validation. *J. Toxicol. Environ. Health A* 77: 133–142.
- Nunes, E., Cavaco, A., and Carvalho, C. 2014b. Children's health risk and benefits of fish consumption: Risk indices based on a diet diary follow-up of two weeks. *J. Toxicol. Environ. Health A* 77: 103–114.

- Oken, E., Radesky, J. S., Wright, R. O., Bellinger, D. C., Amarasiriwardena, C. J., Kleinman, K. P., Hu, H., and Gillman, M. W. 2008. Maternal fish intake during pregnancy, blood mercury levels, and child cognition at age 3 years in a US cohort. Am. J Epidemiol. 167: 1171–1181.
- Rypel, A. L. 2010. Mercury concentrations in lentic fish populations related to ecosystem and watershed characteristics. *AMBIO* 39: 14–19.
- Scudder Eikenberry, B. C., Riva-Murray, K., Knightes, C. D., Journey, C. A., Chasar, L. C., Brigham, M. E., and Bradley, P. M. 2015. Optimizing fish sampling for fish-mercury bioaccumulation factors. *Chemosphere* 135: 467–473.
- Selin, N. E., Sunderland, E. M., Knightes, C. D., and Mason, R. P. 2010. Sources of mercury exposure for U.S. seafood consumers: Implications for policy. *Environ. Health Perspect.* 118: 137–143.
- Shimshack, J. P., Ward, M. B., and Beatty, T. K. M. 2007. Mercury advisories: Information, education, and fish consumption. J. Environ. Econ. Manage. 53: 158–179.
- Straw, M. 2006. Where the slabs are. In-Fisherman 8:46-48.
- Straw, M. 2009. The arc of slabs: Revisited. *In-Fisherman* 2:59-62.
- Sunderland, E. M. 2007. Mercury exposure from domestic and imprted esuarine and marine fish in the U.S. seafood market. *Environ. Health Perspect.* 115: 235–242.
- Sweet, L. I. and Zelikoff, J. T. 2001. Toxicology and immunotoxicology of mercury: A compartive review in fish and humans. *J. Toxicol. Environ. Health B* 4: 161–205
- Tilden, J., Hanrahan, L. P., Anderson, H., Palit, C., Olson, J., and MacKenzie, W. 1997. The Great Lakes fish consortium, 1997: Health advisories for consumers of Great Lakes sport fish: Is the message being received? *Environ. Health Perspect.* 105: 1360–1365.
- United Nations Environment Programme, Chemical Branch, DTIE. 2008. The global atmospheric mercury assessment: Sources, emissions, and transport. Geneva: UNEP.
- U.S. Food and Drug Administration and U.S. Environmental Protection Agency 2014. Fish: What pregnant women and parents should know. http://www.fda.gov/Food/ FoodborneIllnessContaminants/Metals/ucm393070.htm
- U.S. Environmental Protection Agency 1989. Risk assessment guidance for Superfund: Volume 1, human health evaluation manual. EPA /540/1-89/002. https://www.epa.gov/ sites/production/files/2015-09/documents/rags_a.pdf
- U.S. Environmental Protection Agency. 1997. Mercury study report to Congress, EPA-452/R-97-003. http://www3.epa. gov/ttn/oarpg/t3/reports/volume1.pdf (accessed September 22, 2015).
- U.S. Environmental Protection Agency. 2007. Mercury in solids and solutions by thermal decomposition, amalgamation and atomic absorption spectrophotometry. Method 7473, SW-846. http://www.epa.gov/osw/hazard/test methods/sw846/pdfs/7473.pdf.
- U.S. Environmental Protection Agency 2010. Fish and Shellfish Advisories and Safe Eating Guidelines. https:// www.epa.gov/choose-fish-and-shellfish-wisely/fish-andshellfish-advisories-and-safe-eating-guidelines.

904 👄 S. WOLFF ET AL.

- U.S. Environmental Protection Agency. 2011a. Body weight studies. In *EPA exposure factors handbook*. http://www.epa. gov/ncea/efh/pdfs/efh-chapter08.pdf
- U.S. Environmental Protection Agency. 2011b. Intake of fish and shellfish. In *EPA exposure factors handbook*. http:// www.epa.gov/ncea/efh/pdfs/efh-chapter10.pdf
- U.S. Food and Drug Administration. 2001. FDA talk paper: FDA announces advisory on methylmercury in fish. http:// www.fda.gov/OHRMS/DOCKETS/ac/02/briefing/3872_ Advisory%205.pdf (accessed on September 22, 2015).
- Vieira, H. C., Morgado, F., Soares, A. M., and Abreu, S. N. 2015. Fish consumption recommendations to conform to current advice in regard to mercury intake. *Environ. Sci. Pollut. Res. Int.* 22: 9595–9602.
- Wathen, J. B., Lazorchak, J. M., Olsen, A. R., and Batt, A. 2015. A national statistical survey assessment of mercury concentrations in fillets of fish collected in the U.S. EPA national rivers and streams assessment of the continental USA. *Chemosphere* 122: 52–61.
- World Health Organization and United Nations Environment Programme. 2008. Guidance for identifying populations at risk from mercury exposure. Issued by UNEP DTIE Chemicals Branch and WHO Department of Food Safety, Zoonoses and Foodborne Diseases. Geneva, Switzerland.
- Xue, J., Zartarian, V., Mintz, B., Weber, M., Bailey, K., and Geller, A. 2015. Modeling tribal exposures to methyl mercury from fish consumption. *Sci. Total Environ.* 533: 102–109.