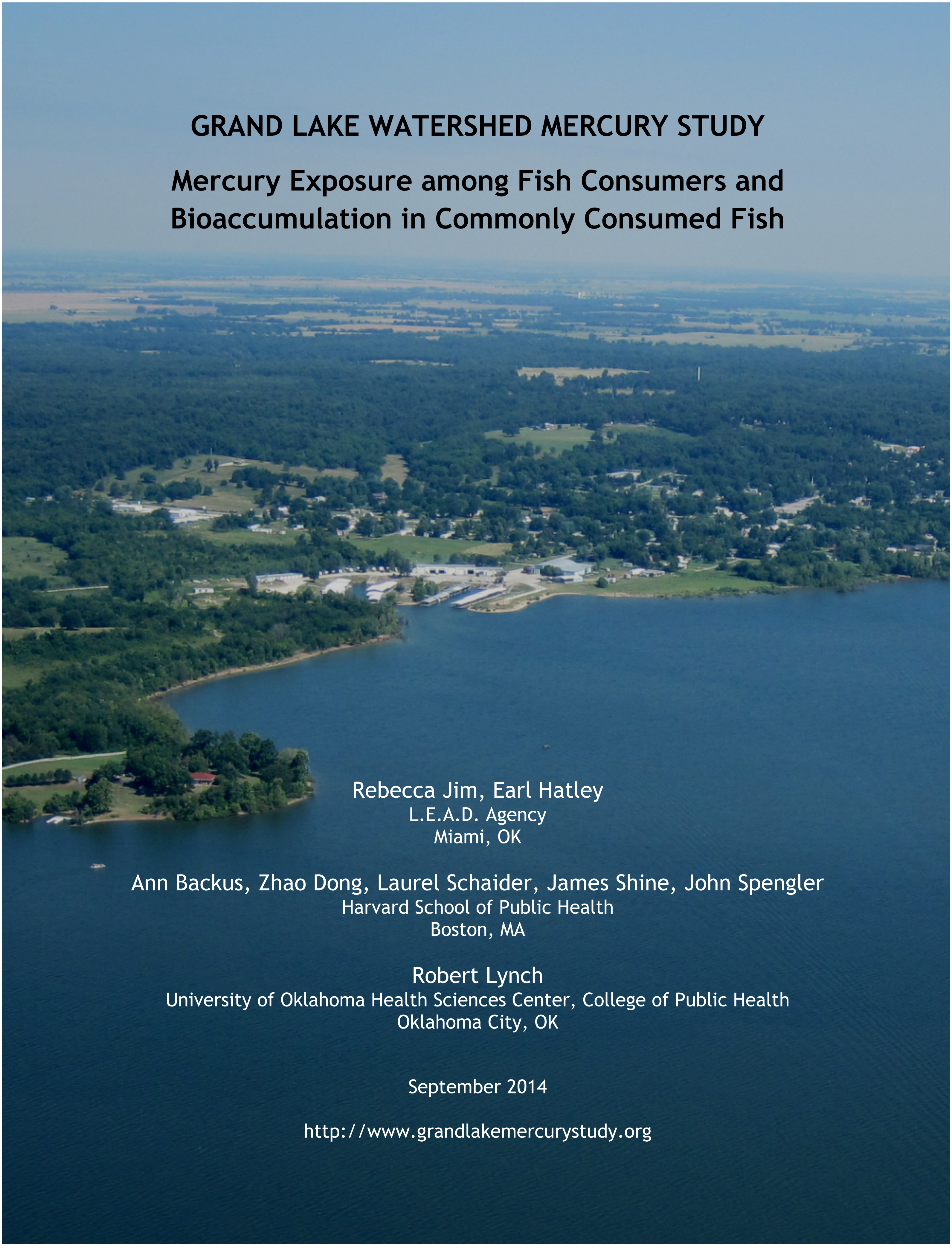

GRAND LAKE WATERSHED MERCURY STUDY

Mercury Exposure among Fish Consumers and Bioaccumulation in Commonly Consumed Fish

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http://www.grandlakemercurystudy.org

Executive Summary

Overview

Previous studies have identified mercury exposure through fish consumption as an important public health concern, and numerous fish consumption advisories have been developed to protect the health of fish consumers. Since fish can also be part of a healthy diet, it is important to balance the benefits of consumption with any health threats that may be caused by consumption of associated contaminants. Additional research is warranted to better understand linkages between exposure and health outcomes.

The Grand Lake Watershed Mercury Study (GLWMS) was designed as a community-based participatory research project to address community concerns in northeastern Oklahoma about mercury exposure and mercury levels in fish. Mercury exposure from consumption of local fish was examined in anglers and their families in northeastern Oklahoma, centered around Grand Lake O' the Cherokees (Grand Lake). Project participants recorded their fish consumption over a one-year period and provided quarterly hair samples to test for mercury exposure. In addition, mercury concentrations were assessed in samples of commonly-consumed fish species collected throughout the watershed.

Findings

Over 1500 fish collected throughout the watershed were tested for mercury during the study. The majority of fish analyzed contained mercury below guidelines established by the United States Environmental Protection Agency (US EPA) and Oklahoma Department of Environmental Quality (ODEQ). Flathead catfish, largemouth bass, and blue catfish had the highest levels of mercury, and in general, larger fish had higher levels of mercury.

Furthermore, 95% of study participants had hair mercury levels below established criteria for exposure, which are directed at women of child-bearing age and children. Taken together, these data indicate that mercury exposure risk from fish consumption in the Grand Lake area is generally low; however, excess consumption of particular fish could result in mercury exposure above recommended criteria. On average, project participants receive around 60% of their total mercury exposure from local freshwater fish. A significant portion (about 40%) comes from non-local (e.g., seafood) sources, so consumption guidelines should consider both sources of exposure in order to accurately reflect risk. Mercury exposure should be evaluated in other areas of Oklahoma with higher levels of mercury in fish.

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ACKNOWLEDGEMENTS

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1. Introduction and Background

Motivation and goals for this study

The research described in this report builds upon past collaborative efforts between researchers at Harvard School of Public Health (HSPH) and the University of Oklahoma Health Sciences Center (OUHSC) and a local environmental organization, LEAD Agency (Local Environmental Action Demanded), based in Miami, Oklahoma. These collaborations initially focused on heavy metal contamination associated with the Tar Creek Superfund Site and over time evolved to address other environmental topics. In 2009, the National Institute of Environmental Health Sciences, part of the National Institutes of Health, started a Research to Action program to explore topics of interest to local communities and encouraged community organizations and researchers at universities to submit community-based participatory research (CBPR) proposals that explored questions of interest to environmental groups with a firm research foundation provided by the universities. CBPR involves true partnerships between researchers and community members throughout the entire process, starting with the development of a research topic that addresses community concerns, through all stages of project development and implementation, while also providing new knowledge with broader applications (O'Fallon 2002).

The LEAD Agency, in collaboration with researchers at HSPH and OUHSC, submitted a proposal to explore the relationship between consumption of fish and mercury exposure in fishermen around the Grand Lake area. The LEAD Agency was concerned about mercury exposure because many of the area's residents rely on locally caught fish for food and because of the close proximity of the lake to several power plants. Study team members were interested in exploring ways to quantify the relationship between fish consumption and mercury exposure, as measured by mercury levels in the hair of residents who eat local fish.

Together, this research study sought to answer these questions:

- What are the levels of mercury in commonly consumed fish in the Grand Lake watershed?
- How much and what types of local fish are eaten by residents of the watershed?
- Are people who eat fish from the watershed exposed to high levels of mercury in their diet?

MERCURY IN THE ENVIRONMENT

Mercury is an important environmental contaminant with demonstrated linkages between exposure and health outcomes, particularly diminished neurological development in children (Grandjean et al. 1999; Karagas et al. 2012). The primary route of mercury exposure for U.S. residents is consumption of fish (Mahaffey et al. 2004). The largest source of mercury in the environment is emissions from power plants (about 50%; US EPA 2014), although there are a number of other important sources such as volcanic eruptions, mining, various manufacturing operations, incineration of wastes, batteries, and fluorescent lamps. Coal naturally contains trace levels of mercury that are emitted into the atmosphere when coal is burned. Depending upon its chemical form and climatic conditions, emitted mercury can travel for tens of miles, hundreds of miles, or even globally; therefore, the mercury deposited at a given location can have many sources, although local or regional sources often contribute the most significant portions to local concentrations (Carpi 1997; US EPA 2011).

After mercury is deposited on land, some portion finds its way to waterways through runoff (Weiner et al. 2003). In Oklahoma, almost all of the rivers are impounded by dams to create reservoirs, and mercury that enters these reservoirs and lakes through runoff and direct deposition tends to accumulate in sediments. Depending upon lake conditions, some of the deposited mercury is transformed by bacteria into an organic form, methylmercury, which is mobile in the environment and enters the aquatic food chain. Mercury concentrations can increase significantly up the food chain (a process known as biomagnification) ultimately reaching the top predator, which in some cases may be people. Methylmercury concentrations of a few parts per trillion in water can result in fish concentrations of methylmercury in parts per million, a million-fold increase in concentration.

Low oxygen conditions in many lakes, which are present in sediments, wetlands, and bottom waters during stratification, promote the transformation of inorganic mercury to methylmercury. More than 50% of Oklahoma lakes are classified as eutrophic (OWRB 2012), and this condition can contribute to stratification and low dissolved oxygen in hypolimnetic water (near the bottom), which is one of the factors that facilitates conversion of inorganic mercury to methylmercury. Furthermore, maps of atmospheric mercury deposition based on field measurements have shown that many parts of Oklahoma, especially counties in the southeastern portion of the state, regularly receive elevated mercury deposition (NADP 2014). With six coal-fired electricity generating plants operating within 60 miles of Grand Lake, several others in Oklahoma, and many more large plants upwind in Texas, elevated mercury deposition is expected in northeastern Oklahoma. An additional consideration is that mercury can be brought into the state in rivers that drain other states and then flow into Oklahoma. Taken together, natural and human-related sources of mercury contribute to the widespread presence of mercury in fish in all lakes where surveys have been completed. For these many reasons, studying mercury exposure through the consumption of fish in northeastern Oklahoma is warranted.

MERCURY EXPOSURE GUIDELINES

Mercury guidelines are based mostly on epidemiological studies that have examined the relationship between mercury exposure and health effects in people, particularly in populations exposed to high levels of mercury. From these studies, the US EPA has established a guideline for the amount of mercury that people can consume over a lifetime without having negative health outcomes. This amount is called a reference dose (RfD) and is measured as micrograms of methylmercury per kilogram of body weight per day (μg/kg/day). The RfD for methylmercury is 0.1 μg/kg/day and is specifically designed to protect unborn and young children. If a person consumes mercury at the RfD rate, it is estimated that they would have a blood methylmercury concentration of 5.8 micrograms per liter (μg/L) and a hair methylmercury concentration of 1 part per million (ppm).

A good way to monitor mercury intake is to measure the amount of mercury in hair. Individuals with hair mercury levels greater than 1 ppm are likely to have been exposed to mercury at levels above the RfD. Typically hair grows at a rate of around 1 centimeter per month, so the level of mercury in each centimeter of hair is an integrated measure of mercury exposure over the course of a month. Using hair mercury testing in this project allowed us to determine mathematical relationships between the amount of fish consumption reported by participants over the past three months, the amount of mercury in their hair, and the mercury levels in the fish they consumed.

The relationship between mercury intake and hair mercury concentration can be extended to establish guidelines for the levels of mercury in fish that can be consumed without exceeding the RfD. The US EPA has set a guideline for methylmercury in freshwater fish of 300 parts per billion (ppb) (US EPA 2001), while the Oklahoma Department of Environmental Quality (ODEQ) has set a standard of 500 ppb. These guidelines are based on the assumption that people consume 2-3 fish meals per month (8 ounces of fish per meal) and are intended for women of childbearing age and children who are more susceptible to mercury exposure. The main reason for the difference in guideline values stems from assumptions about other mercury exposure sources. National wide studies have shown that people who eat fish from both freshwater (lakes and streams) and saltwater (ocean) sources get about 40% of their mercury from ocean species such as tuna, swordfish, and shrimp (Sunderland 2007). Given this information, US EPA considers that non-local fish mercury sources should also be included in determining guidelines for mercury levels in freshwater fish. ODEQ guidelines do not include methylmercury exposure from ocean fish, hence the establishment of higher values.

An important consideration in determining appropriate consumption guidelines is the difference in mercury concentrations among various types of fish. Consuming large quantities of fish with low mercury levels can provide similar mercury intake as consuming smaller quantities of fish with high mercury levels. People who eat fish more frequently than twice per month should eat fish that, on average, have lower mercury than the US EPA guideline of 300 ppb in order to stay under the reference dose. For those eating fish 2-3 times per week, the amount of mercury should be substantially less than the standard in order to stay under the recommended reference dose. Ultimately, what matters is the total amount of mercury consumed daily, so consumers need to consider both mercury concentrations in fish and fish consumption rates.

Furthermore, it should be stressed that fish is an important source of dietary nutrients. The U.S. Food and Drug Administration (US FDA) and the EPA recently released draft recommendations that pregnant women eat at least 8 ounces and up to 12 ounces (2-3 servings) per week of a variety of fish that are lower in mercury to support fetal growth and development (US FDA 2014).

2. Study Methods

Grand Lake and Lake Hudson

Grand Lake, located in northeastern Oklahoma, was impounded in 1940 and covers approximately 41,800 surface acres (Figure 1). The 3 major tributaries are the Neosho River (68% of in-flow), Spring River (16% of in-flow), and Elk River (17% of in-flow). Its watershed has an area of approximately 10,300 square miles that encompasses parts of southeastern Kansas, southwestern Missouri, northwestern Arkansas, and northeastern Oklahoma. It is classified as eutrophic based on phosphorus concentrations (range: 0.028–0.092 mg/L).

Lake Hudson is located downstream of Grand Lake and was impounded in 1964. It covers approximately 11,000 surface acres and is also classified as eutrophic based on phosphorus concentrations (range: 0.01–0.126 mg/L) (OWRB 2014).

Objectives and approach

The goals of the research were to measure mercury levels in commonly-consumed fish within the Grand Lake watershed and to evaluate fish consumption and mercury exposure among people who eat fish from the lake. To support these goals, we measured mercury concentrations in over 1500 fish from Grand Lake, Lake Hudson, Neosho River, Spring River, and other tributaries. We also recruited 152 residents, including fishermen and their family members, who regularly consumed fish from the Grand Lake watershed. To evaluate mercury exposure in project volunteers, we measured mercury levels in hair samples from each participant at the time of recruitment and four additional times over the following year (roughly quarterly) to evaluate how exposure changed over the course of a year. Each participant also completed quarterly food frequency questionnaires (FFQs) to determine the types and quantities of the fish eaten as well as cooking and preparation methods. The questionnaire collected information about how often people consumed local fish caught from the Grand Lake watershed, as well as non-local fish such as tuna, shrimp, fish sticks, and salmon. Based on these results, we determined individual and collective fish consumption rates and the relative amount of each species consumed. After integrating these data, we were able to determine the relative levels of mercury exposure from local and non-local fish species and the relationships between fish consumption and hair mercury levels in our study participants.

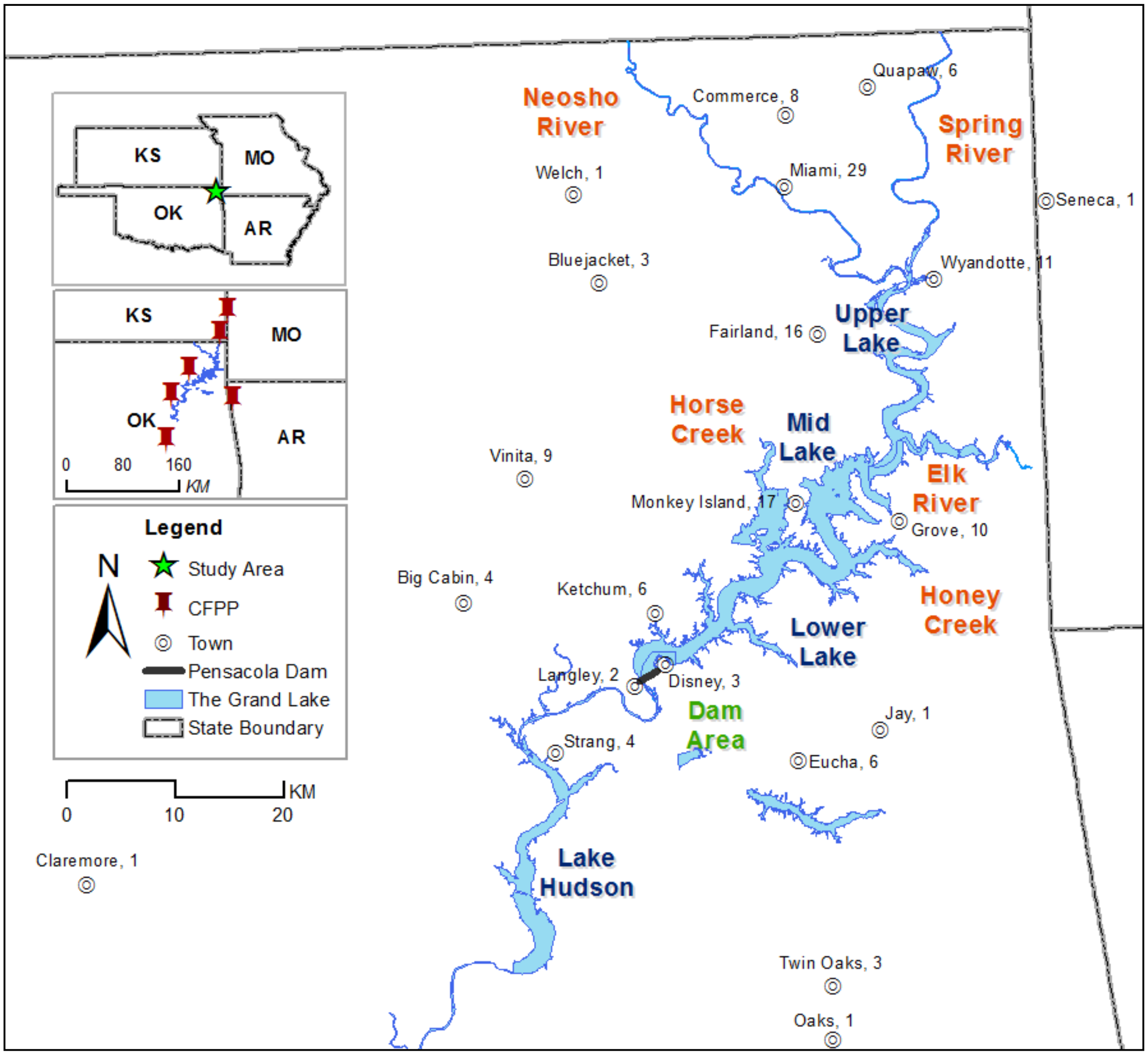


Figure 1. Map of Grand Lake, showing number of participants by town and proximity of coal-fired power plants (CFPP). Four towns located outside of the mapped area are not shown: Tulsa, 2; Chetopa (KS), 1; Joplin (MO), 2; Neosho (MO), 4.

METHODS

Fish sampling and testing

Between 2010 and 2013, over 1500 fish representing 30 species were collected from Grand Lake, Lake Hudson, Neosho River, Spring River, other tributaries, and local ponds. Filet samples from each of these fish were tested for mercury. Nearly half of these fish were collected by the Oklahoma Department of Wildlife Conservation as part of routine fish population surveys, primarily using gill nets. The rest of the samples were primarily provided by local fishermen and project volunteers who provided fish caught by hook and line and noodling. In addition, in 2012, study team members collected a series of small prey (forage) fish by seining to better understand the movement of mercury through the food chain and potential differences in mercury levels in fish throughout the watershed. Fish sampling and handling protocols were approved by the University of Oklahoma Health Sciences Center Institutional Animal Care and Use Committee.

Fish were weighed and measured and a small piece of tissue (about finger size) was harvested from each fish along the spine and just behind the head, and then shipped to the Trace Metals Laboratory at the Harvard School of Public Health for analysis using a DMA-80 Direct Mercury Analyzer (Milestone Inc., Shelton, CT). Eighty-four samples were further analyzed at the Trace Element Analysis Laboratory at Dartmouth College to determine the proportion of mercury present as methylmercury. Over 800 fish samples were analyzed at the Boston University Stable Isotope Laboratory to determine the relative amounts of carbon and nitrogen isotopes in these fish, as indicators of their position in the food chain and carbon sources in their diet.

Development of Food Frequency Questionnaire (FFQ)

Our FFQ was based on an FFQ used by Lincoln et al. (2011) and modified to reflect local fish species and to address the research questions for this project. Our FFQ asked about general and species-specific fish/shellfish consumption frequencies over the previous three months in 8 categories: never, once in last three months, once a month, two or three times a month, once a week, two or three times a week, four to six times a week, and once a day or more. We included fish species commonly caught from the Grand Lake watershed and commonly-consumed non-local, primarily saltwater, fish. In addition, we asked about both typical fish portion size and number of portions at a typical fish meal, after participants viewed a plaster model depicting filets of four different sizes (2, 4, 6, and 8 oz). The FFQ also included demographic questions and questions on fishing, sharing, and storage behaviors. The FFQ was reviewed and tested by a community advisory board and multiple focus groups that encompassed a range of ethnicities before administration to participants. A sample page from the questionnaire is provided in Figure 2, and the complete questionnaire can be viewed online at: http://www.grandlakemercurystudy.org/images/GLWMS_FFQ.pdf.

We enlisted the help of several groups in designing and field testing the questionnaire to ensure accuracy and relevance to the local community. For example, a list of fish species consumed locally was provided by the LEAD Agency staff and was modified and expanded after consultation with the Community Advisory Board, Council of Fishing Experts, and focus groups including American Indians, Hispanics, and Micronesian high school students, who also provided suggestions on appropriate means for recruiting participants and venues for sharing research findings.

	Largemouth bass	Smallmouth bass	White bass	Spotted bass	Striped bass	Crappie
(a). How often did you eat this type of fish over the past 3 months?						
Once a day or more						
4 to 6 times/week						
2 or 3 times/week						
Once a week						
2 or 3 times/month						
Once a month						
Once in last 3 mo.						
Never in last 3 mo.						
(b). Where did this fish come from? ***(check all that apply)***						
Grand Lake – Upper (UL)						
Grand Lake – Mid (ML)						
Grand Lake – Lower (LL)						
Dam Area (DA)						
Spring River (SR)						
Neosho River (NR)						
Elk River (ER)						
Honey Creek (HNC)						
Horse Creek (HSC)						
Other locations						
Restaurant, store or fish fry **(specify what percent came from a restaurant, store, fish fry or other event, if any)**	_____%	_____%	_____%	_____%	_____%	_____%

Figure 2. Sample questions from the food frequency questionnaire (FFQ).

Study participants

A concerted effort was made to reach the initial goal of 150 project volunteers (participants). The project focused on local residents who regularly consumed fish from the Grand Lake watershed with representation from different genders, ages, and ethnic groups. Participants were recruited through a variety of methods including personal contacts, fishing organizations, community fishing events, health fairs, pow wows, and other cultural events. In order to be eligible for the study, participants had to state that they ate fish from the watershed and that they were willing to take part in the study for one year. Some potential participants were turned away from the study since they did not eat any locally-caught fish. For example, we initially approached members of the local Micronesian community who frequently ate fish, but found that they only consumed non-local fish. Since selection of participants was not a random process, we are unable to conclude whether our participants accurately reflect the fish consumption patterns of other fish consumers in the area.

The hair of each participant was sampled at the time of recruitment and four additional times over the following year (roughly quarterly). A 2-cm length portion of the hair nearest the scalp was analyzed each time, which roughly corresponded to mercury exposure over the previous 1-3 months. At the same time, each participant completed an FFQ. In between visits, participants were asked to keep a diary of their fish consumption to aid in completion of subsequent FFQs. Hair samples were analyzed for mercury using the same instrument that was used to analyze the fish samples. Informed consent was obtained from every participant at the time of enrollment. All study materials and research protocols relating to human subjects were approved by the Office of Human Research Administration at Harvard School of Public Health and the Office of Human Research Participant Protection at the University of Oklahoma Health Sciences Center.

Data analysis

Based on these data, individual and collective fish consumption rates and the relative amount of each species consumed were determined. In addition, important information on the consumption rates of non-local fish such as tuna, shrimp, and fish sticks were collected. These data were combined with measured concentrations of mercury from locally-caught fish and values reported by US FDA (2014) to calculate mercury intake rates for project participants. Statistical models were developed to analyze the association between mercury in hair and fish consumption or estimated mercury intake, while accounting for other potential factors related to mercury exposure (i.e., gender, age, ethnicity, BMI, education, percent local fish consumed, season).

Report-back

Mercury results from each hair sample were reported back to participants within three months using a standardized reporting form that indicated the concentration of mercury in their hair (Figure 3). Each individual's results were placed into context compared to other benchmarks and guidelines, and participants also received general information about mercury and health. At the completion of the study, each participating household also received a fact sheet and a frequently-asked questions booklet (available on the project website) that summarized our key study findings, along with and a short film produced by Dartmouth College about mercury exposure and fish consumption. Similarly, when study participants or other community members donated fish samples, we reported back the mercury levels in the fish samples they donated, along with guideline values and information to interpret their results.

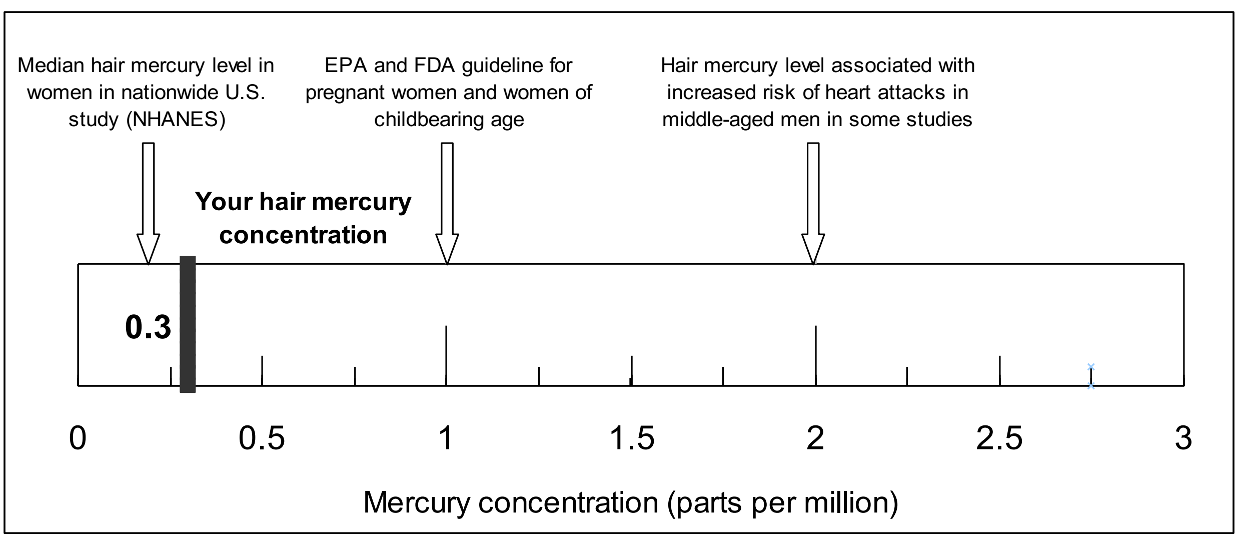


Figure 3. Graphic used in report-back letters to participants illustrating hair mercury test results

3. Grand Lake Watershed Fish Results

Mercury concentrations in commonly-consumed fish

Our results demonstrated that the levels of mercury in fish from Grand Lake and Lake Hudson were generally below health-based guideline values for moderate consumers of freshwater fish, and that there were substantial variations in mercury levels among the fish tested. These results help to identify fish most likely to contain elevated mercury levels as well as fish with low levels of mercury.

Mercury concentrations in fish varied considerably across species, with higher concentrations in fish at higher trophic levels (e.g., predator fish that eat other fish) (Table 1). On average, flathead catfish had the highest levels of mercury; however, these elevated levels were likely influenced by the relatively large size of the fish tested. Fish that typically eat plankton, algae, and small insects/crustaceans, such as spoonbill and shad, had much lower levels of mercury. Most mercury present in commonly-consumed fish was in the form of methylmercury, which represented above 90% for all species tested, with the exception of shad, which are not commonly consumed and reside lower on the food chain.

In general, mercury concentrations increased with fish length (Figure 4). This was an expected finding in keeping with known mechanisms of bioaccumulation of methylmercury in organisms. These results are very useful in setting advisories for the types and sizes of fish that consumers should consider in assessing dietary exposure to mercury. For example, flathead catfish greater than 40 inches in length are likely (greater than 50% chance) to have mercury levels above 300 ppb. Consumption of these larger, predator fish should be reduced for those who wish to lower their mercury intake.

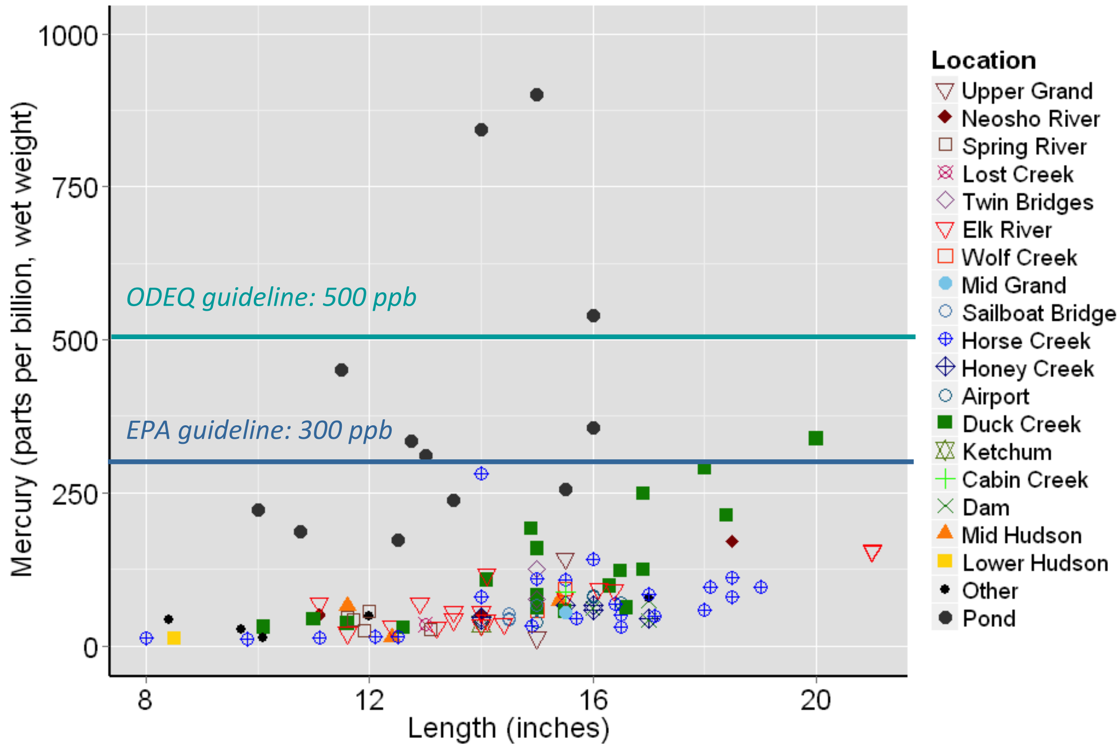
Fish collected from farm ponds tended to have higher levels of mercury than fish from the watershed. Preliminary testing of 27 fish (12 largemouth bass, 11 sunfish, 3 crappie, 1 channel catfish) collected from farm ponds in the vicinity of Grand Lake suggests that they tend to have higher mercury levels than fish from the watershed of comparable length (Figure 4). There are several possible explanations for this trend, including: accumulation of mercury over time in these ponds, which typically have no outlet; relatively high levels of organic material that promotes greater methylation of inorganic mercury; and greater interaction between the sediments and water column compared with the much larger lake and its tributaries.

Table 1. Mercury concentrations in Grand Lake watershed fish.
Mercury concentrations expressed in parts per billion (ppb) wet weight ± 1 standard deviation. Data not included for fish collected from ponds or from Lake Hudson. EPA guideline for moderate consumers of fish (2-3 times per month) is 300 ppb (US EPA 2001).

Species*	Number of samples	Average mercury concentration (ppb)	Range of mercury concentrations (ppb)	Percent of samples above 300 ppb	Average length (inches)
Top predator fish (consume fish)					
Flathead catfish	38	220 ± 150	46 – 700	24%	33
Largemouth bass	99	78 ± 60	11 – 340	1%	15
White bass	158	47 ± 34	6.5 – 190	0	12
Crappie	122	28 ± 20	10 – 140	0	11
Middle level fish (consume insects, small fish)					
Drum	29	130 ± 190	3.6 – 900	10%	13
Blue catfish	29	59 ± 56	15 – 400	2%	20
Channel catfish	112	49 ± 29	3 – 140	0	16
Sunfish	98	31 ± 27	8.0 – 190	0	6
Lower level fish (consume plankton, algae, insect larvae)					
Smallmouth buffalo	44	50 ± 39	8.9 – 170	0	17
Spoonbill	55	40 ± 24	2.7 – 110	0	40
Shad	39	15 ± 12	3.2 – 84	0	8

*includes species with at least 10 fish sampled

(a) largemouth bass



(b) flathead catfish

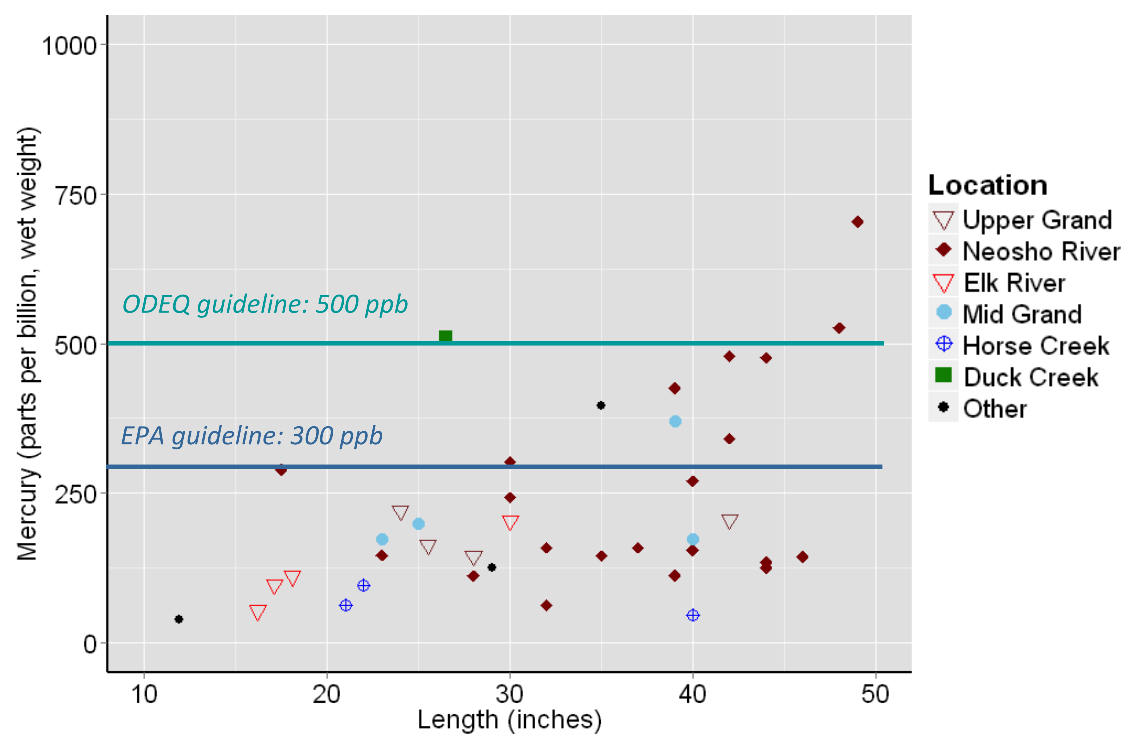


Figure 4. Mercury concentrations in (a) largemouth bass and (b) flathead catfish

Most fish had mercury levels below EPA's guideline for moderate consumers of freshwater fish. Only 3% of the fish we tested exceeded the US EPA guideline of 300 ppb, and the average concentrations for all fish species were below 300 ppb. Although average concentrations in all fish were below the US EPA guideline, 24% of flathead catfish, 1% of largemouth bass, 10% of drum, and 2% of blue catfish were above 300 ppb.

Mercury levels in fish from Grand Lake and Lake Hudson were relatively low compared to many other lakes in Oklahoma. Mercury levels in largemouth bass have been tested by Oklahoma Department of Environmental Quality in approximately 60 lakes since 2008. Grand Lake and Lake Hudson fish are at the low end of the data distribution among these lakes (Figure 5).

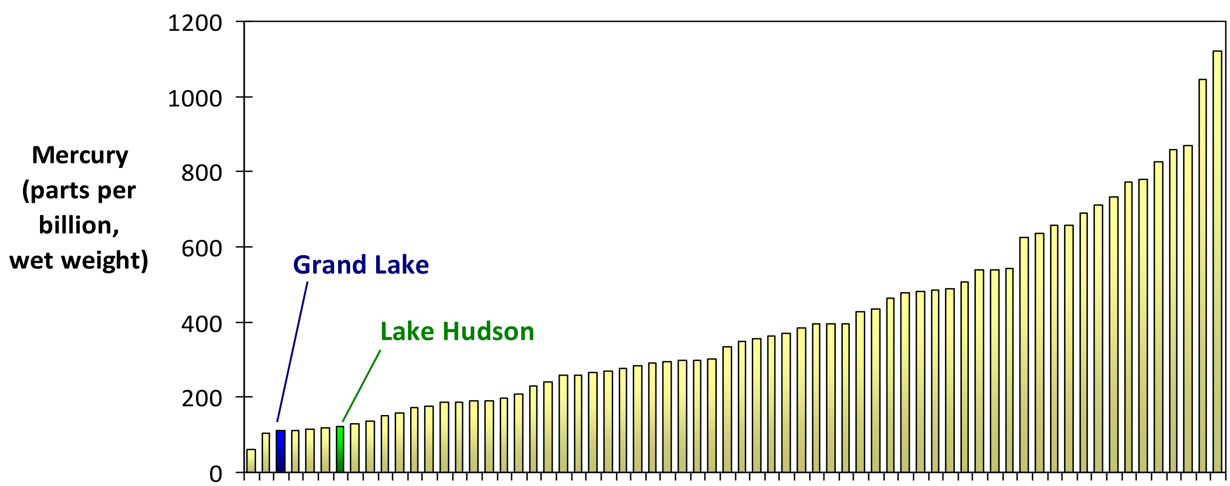


Figure 5. Length-normalized mercury concentrations in largemouth bass from 60 Oklahoma lakes. These concentrations represent modeled concentrations in a 14" largemouth bass, based on a methodology from the U.S. Geological Survey. Data courtesy of ODEQ.

Fish data were analyzed to determine whether mercury levels varied among the upper, middle, and lower regions of Grand Lake (Table 2). In general, it appears that mercury levels were higher in lower (downgradient) zones of the lake, although differences were not statistically significant. Since sample collection was incomplete for some species, it is not possible to make a firm determination of whether there are any spatial differences. More focused sampling efforts among major regions of the lake would allow for a more extensive analysis.

Table 2. Spatial distribution of mercury in fish from three areas of Grand Lake.
Mercury concentrations expressed as parts per billion (ppb) wet weight ± 1 stand deviation.

	Average mercury concentration* (ppb)		
Species	Upper	Middle	Lower
flathead catfish	227 ± 155	160 ± 110	--
largemouth bass	69 ± 42	64 ± 46	110 ± 84
blue catfish	60 ± 48	51 ± 77	67 ± 32
channel catfish	50 ± 27	42 ± 29	61 ± 30
white bass	48 ± 34	39 ± 18	55 ± 43
white crappie	26 ± 17	12 ± 2.8	--
bluegill	20 ± 4.6	21 ± 9.3	45 ± 54
shad	14 ± 6.1	20 ± 22	13 ± 5.0

*at least five fish tested

INTERPRETATION OF FISH MERCURY RESULTS

Another goal of the project was to develop lake-specific information about fish consumption for local community members. General guidelines do not adequately inform consumers about the amounts of different species of local fish that they might safely consume since mercury levels can vary widely among lakes. As demonstrated by the data in this report, the amount of mercury in fish varies by species and by fish size within species; therefore, guidelines that categorize all fish of all sizes as the same in terms of consumption guidelines do not accurately inform fish consumers about mercury exposure risks. For example, the data clearly show that consumers can eat a great deal more sunfish than large flathead catfish to stay within the reference dose of 0.1 micrograms of mercury per kilogram body weight per day. Although more accurate, species and size specific guidelines can be more confusing to the consumer. A convenient way to translate the information is to determine an individual consumer's own daily intake by considering their weight. The calculations conveniently determine that a person's weight in pounds (lb) is equivalent to the amount (in micrograms, µg) of mercury that they can consume per month from locally caught fish and stay below the reference dose. For instance, a 150 lb. person should consume no more than 150 µg of mercury per month from locally caught fish. Other site-specific guidelines based on data from this study can be seen in Table 3. Using average mercury concentrations analyzed in our fish samples (Table 3), a 150 lb. woman could monthly eat 25 servings of crappie (25 x 6 =150) or 4 servings of largemouth bass and 6 servings of blue catfish ((4 x 18 = 72) + (6 x 13 = 78) = 150)). A poster to communicate this information has been prepared and disseminated to areas of northeastern Oklahoma.

Table 3. Average mercury in 8-ounce servings of Grand Lake fish and interpretation for consumers of local fish.

Fish Type	Average amount of mercury in one 8-ounce serving (micrograms)	How often can women of childbearing age and children eat 8-ounce servings of the fish and stay below EPA's guideline?		
		50 pounds	150 pounds	200 pounds
Crappie	6	Twice a week	Once a day	Once a day
Sunfish	6	Twice a week	4 times a week	Once a day
Spoonbill	9	Once a week	4 times a week	4 times a week
White bass	10	Once a week	4 times a week	4 times a week
Channel catfish	10	Once a week	4 times a week	4 times a week
Smallmouth buffalo	11	Once a week	4 times a week	4 times a week
Blue catfish	13	Once a week	Twice a week	4 times a week
Largemouth bass	18	Twice a month	Twice a week	Twice a week
Drum	25	Twice a month	Once a week	Twice a week
Flathead catfish under 30"	40	Once a month	Once a week	Once a week
Flathead catfish 30" or longer	59	Once a month	Twice a month	Once a week

MERCURY LEVELS IN PREY (FORAGE) FISH

Since mercury bioaccumulation in fish depends on mercury levels in their prey, and since many fish commonly consumed by people are higher on the food web and eat small prey fish, mercury levels in prey fish were of particular interest. Furthermore, prey fish may provide a better indication of variations in mercury levels in food webs across locations since they have smaller ranges and are younger than larger predatory fish. We looked for variations in mercury levels in six species of prey fish (mostly 4-10 cm) over time and among different locations. Seining was conducted at three sites on five occasions between April and September in 2012 (upper Grand – near Sailboat Bridge; middle Grand – Horse Creek arm near Cleora; Lake Hudson – Snowdale Park).

In general, mercury concentrations were similar across locations, while species showed different trends across the sampling period. Shiners (predominantly *Notropis lutrensis*) and brook silversides (*Labidesthes sicculus*) were the most commonly collected species and provide a good illustration of ranges in trends observed (Figure 6). Mercury levels in shiners increased steadily at all sites over the course of the sampling season, indicating a slow trend of bioaccumulation from food sources. The mercury levels in brook silversides increased slightly at first then dropped by more than half, followed by a steady increase. The different trends among species may be related to a range of factors, such as: changes in diet that led to variations in mercury intake; changes in growth of individual fish that diluted or concentrated the mercury accumulated in their bodies; and the capture of younger fish in mid-summer collections. By contrast, mercury levels in small bluegill (*Lepomis macrochirus*) were relatively constant over time across all locations. These results suggest that, while there may be some differences in methylmercury in lake water and sediments among locations over time, mercury bioaccumulation in prey fish is species-specific and more dependent on physiological and ecological factors.

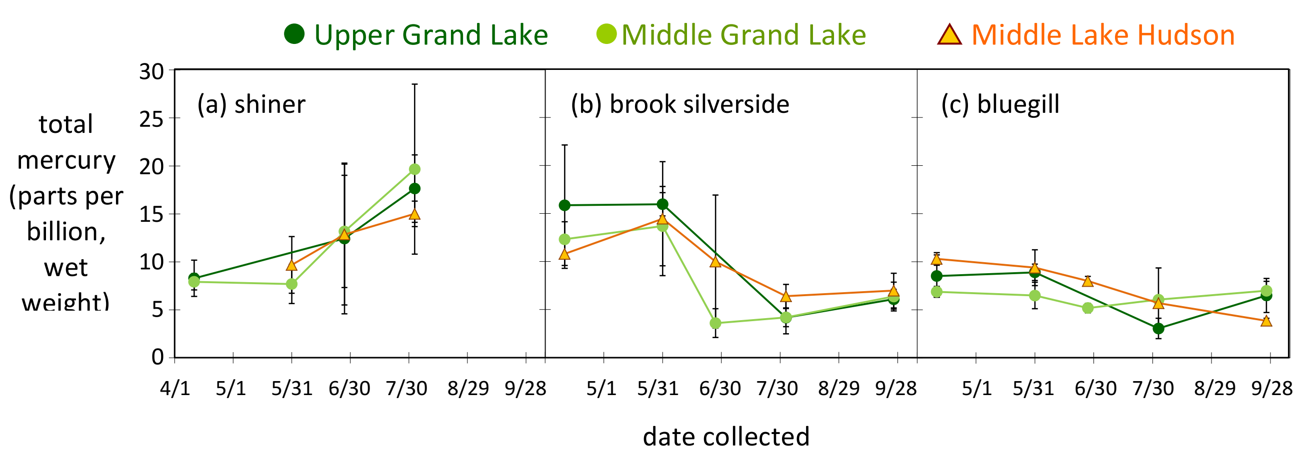


Figure 6. Variations in mercury levels in three species of prey fish.

4. Fish Consumption and Mercury Exposure Results

Demographics of study population

A total of 152 people volunteered to participate in the study, which met the goal of 150 participants. Participants were requested to fill out a food frequency questionnaire at the time of each hair sampling but not all participants did this on each of the 5 occasions. Overall, 611 FFQs (80% based on the initial sample size) and 599 hair samples were collected between July 2010 and March 2013. Participants were 55% men and 45% women with a median age of 55 and came from 20 towns, mostly in Ottawa, Delaware, Cherokee, and Mayes Counties. The largest racial/ethnic groups were white (63%) and American Indian (29%). Approximately 70% of participants were classified as overweight or obese, with a Body Mass Index (BMI) equal to or greater than 25.

Fishing behaviors

Of all participants, 79% reported fishing at least once during their five visits, and these anglers fished an average of 3.8 times per month, almost three times higher than the national average of 16 fishing trips per year (1.3 trips per month) among freshwater anglers (US FWS 2013). Another 14% of participants lived with a household member who had gone fishing. For each season, over two-thirds of participants reported fishing or living with someone who fished during the prior three months, ranging from 69% (fall) to 83% (spring). Most (77%) of anglers reported sharing their catch with others, including adults (62%) and children (38%) in their household and people outside their household (75%). A majority (72%) of participants froze some fish to eat later. Among 33 married couples, 74% of husbands went fishing, compared to only 34% of wives.

Fish consumption

To date, no published studies have quantified fish consumption levels in Oklahoma; therefore, an important component of this research was to determine fish consumption rates of local residents and their families. Without knowing actual rates of fish consumption, data collected from other areas, which may not accurately reflect local conditions, had to be used to develop guidelines. This is especially important when determining the relative contribution of local versus non-local sources of mercury in fish. Through the FFQs, the project gathered data on both the frequency at which individual fish species were eaten and the amounts of each type of fish that were consumed.

Fish consumption rates among participants were above those of the general U.S. population and similar to other studies of recreational anglers. Most participants ate fish 2-3 times a month (35%) or once a week (29%), and 20% ate fish at least 2-3 times a week (Figure 7). More than 30% of participants ate fish frequently (more than 2-3 times per week) while less than 30% ate fish once per month or less. Based on the FFQs, participants ate on average 58 grams (2.0 ounces) of fish per day which is less than half of the national average amount for fish consumers (129 grams or 4.5 ounces per day), although it should be noted that the national average includes people in coastal states who eat relatively large amounts of fish. Since fish consumption rates measured in this study were consistent with rates measured in other inland locations in the U.S., this indicates that our population was representative of other recreational anglers. The fact that most project participants do not eat fish at rates above the national average is an important factor to consider when interpreting the amount of mercury in hair and in extrapolating project findings to other areas.

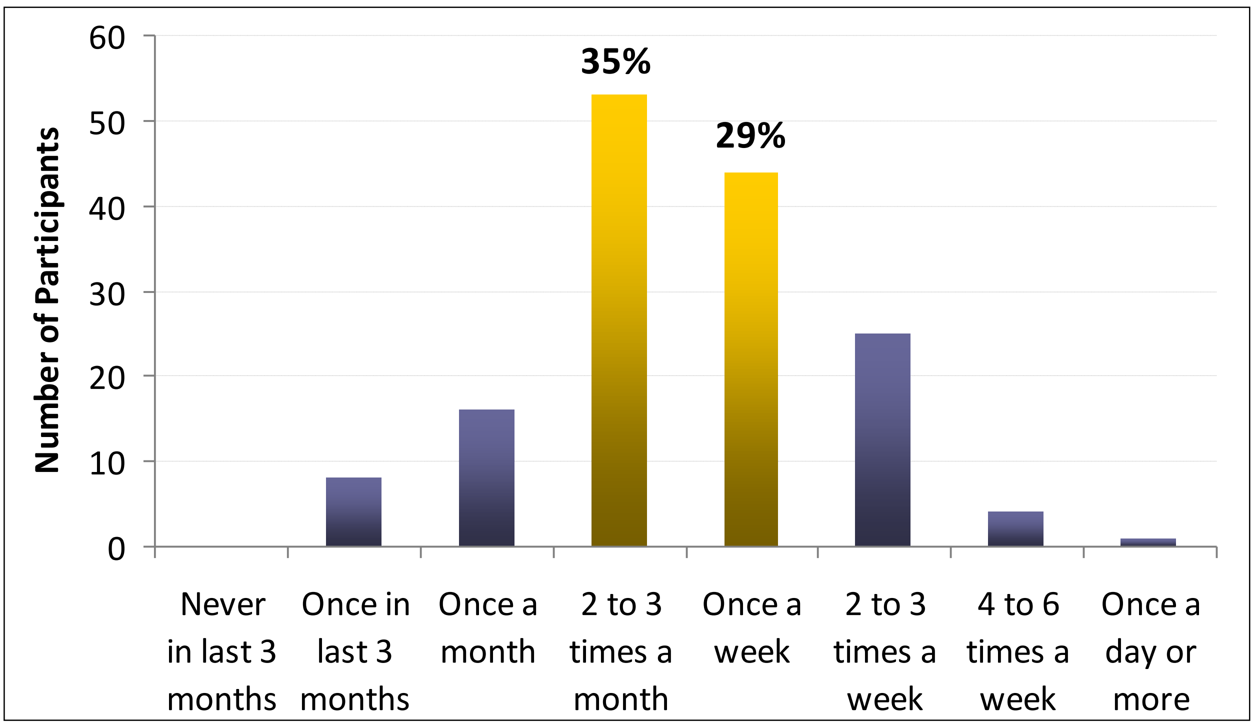


Figure 7. Fish consumption frequency reported by participants in their first food frequency questionnaire.

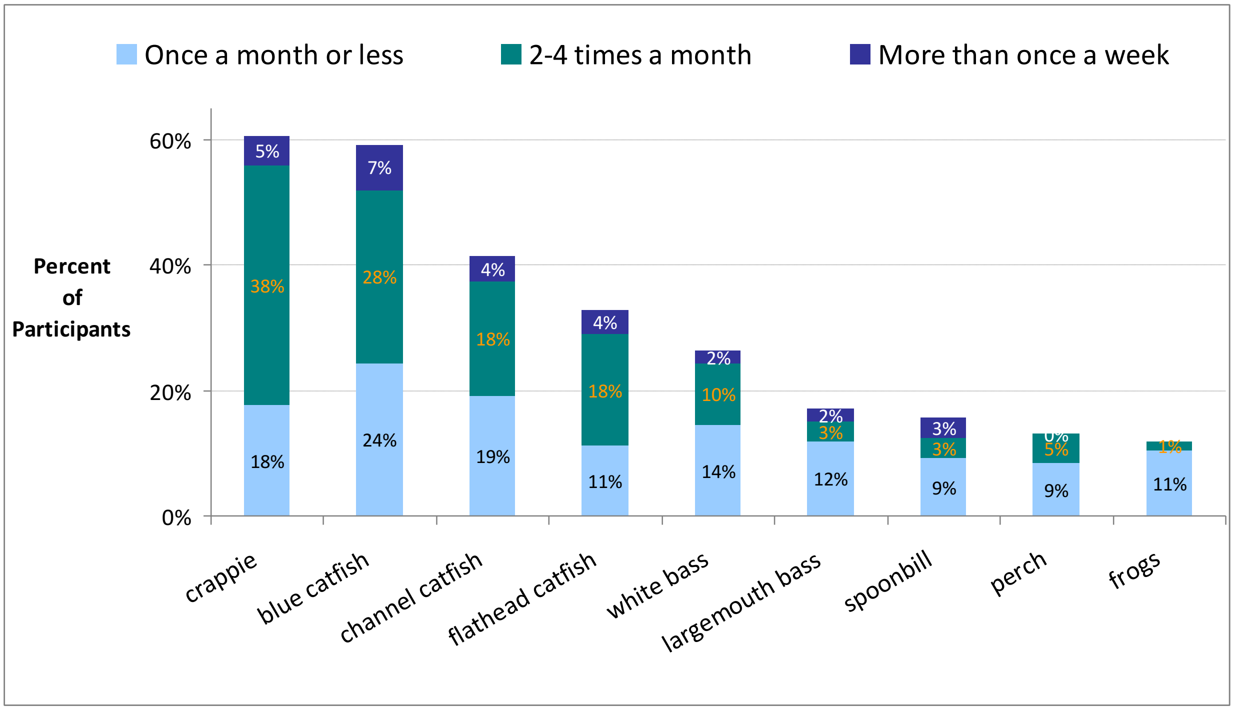
Among individual species, crappie was the most frequently consumed fish followed closely by blue catfish. Over 60% of participants reported eating crappie at least once per month and 59% reported eating blue catfish at least once per month; therefore, it is apparent that mercury levels in these two fish are an important part of the mercury exposure scenario. More than 20% of participants ate white bass, flathead, blue catfish, or crappie at least once per month (Figure 8a).

The most commonly eaten non-local fish were tuna, salmon, and tilapia, all of which were eaten on average at least once per month. Shrimp were eaten at least once per month by 54% of the population (Figure 8b) (in our analyses, shrimp were considered a non-local fish). Clearly, non-local fish represent a sizeable proportion of the total amount of fish consumed. Although mercury levels in non-local fish were not tested in this project, commercially available fish have been widely tested across the country and these data (US FDA 2014) were used to determine the relative contribution of mercury.

The consumption frequency of both local and non-local fish, and the amount of each fish consumed, were combined to develop an overall measure of fish consumption rates in our project participants. Consumption of local fish accounted for 69% of all fish eaten (Figure 9a). Among local fish, catfish were most heavily consumed (43% of total – blue catfish, 22%; channel catfish, 12%; flathead catfish, 9%), followed by crappie (11%). Consumption of non-local fish accounted for 31% of all fish eaten, with shrimp and tuna being eaten in approximately equivalent amounts. Some useful information can be gathered from these data. For instance, although catfish were less frequently eaten than crappie, they were eaten in larger amounts.

Finally, fish consumption rates were combined with known or measured mercury levels in fish to determine the relative contribution of mercury in the diet of project participants from all fish sources (Figure 9b). Locally-caught fish accounted for 60% of mercury intake, and non-local fish accounted for 40%. Among all fish, flathead catfish consumption accounted for 22% of total mercury exposure, while tuna consumption accounted for 31% of total mercury exposure. This disproportionate amount is due to the relatively high levels of mercury found in tuna. Taken together, catfish and tuna represent 73% of mercury exposure; therefore, it is apparent where reductions in overall mercury exposure can be obtained. Since larger flathead and blue catfish contained the most mercury, reducing the consumption of larger fish (especially flathead catfish over 40”) would have a significant effect on reducing mercury exposure.

(a) freshwater



(b) saltwater

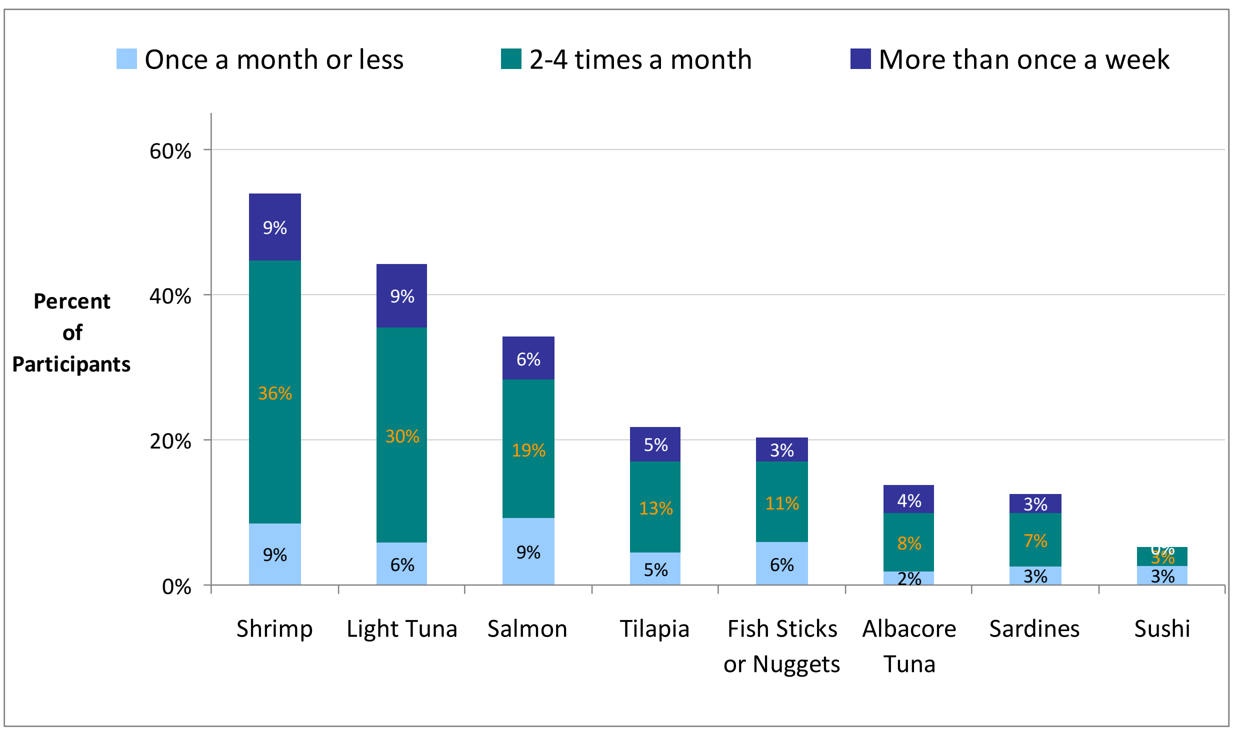


Figure 8. Types of (a) freshwater and (b) saltwater fish consumed by study participants.

Mercury levels in hair

Mercury values in the hair of participants were mostly below the EPA guideline of 1.0 ppm, and only 4.7% of measurements exceeded that value. This percentage is nearly identical to that made in a larger study of U.S. residents and slightly more than the 4.6% measured in the southern U.S. (including Oklahoma); however, this value also included coastal states (Mahaffey et al. 2004). Project participants had a higher rate than non-coastal residents in the U.S., of whom only 2.1% had hair mercury levels greater than 1.0 ppm. The median hair mercury level in participants was 0.15 ppm which is close to to the national average of 0.19 ppm (women 16-49 years).

The seasonal pattern of mercury concentration varied predictably from spring through winter (Figure 10). Average mercury values were relatively low in the spring, somewhat higher in summer, followed by a slow decline through fall and winter. It is expected that rates in spring would be low as they would reflect the antecedent colder weather and with some delay in increased fishing activity with warmer weather. Summer values should reflect greater levels of fishing in the warmer months and the data support this assumption. Fish consumption patterns are somewhat at odds with these expected findings. Consumption rates were highest in spring and summer; however, there was a significant drop during the fall. It is expected that fall fish consumption rates would be as high or higher with the onset of cooler weather, but this was not the case with study participants. Furthermore, it is expected that the seasonal pattern of mercury in hair would reflect the decrease in fish consumption. The reasons for this anomaly are unclear but may be due to differences in consumption patterns among individuals.

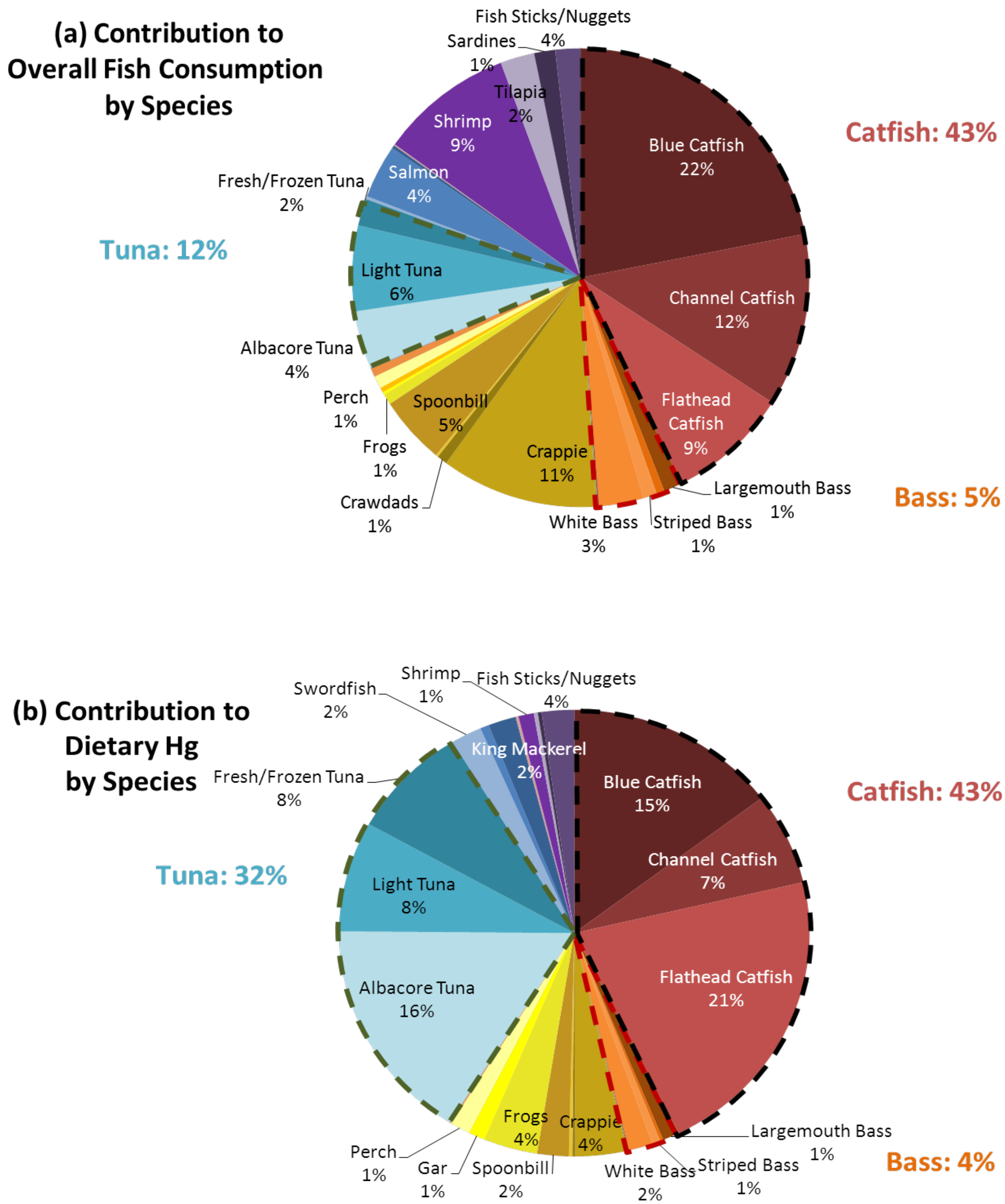


Figure 9. Species-specific contributions to (a) overall fish consumption and (b) mercury intake. Warm colors represent local freshwater species, and cool colors represent non-local saltwater species.

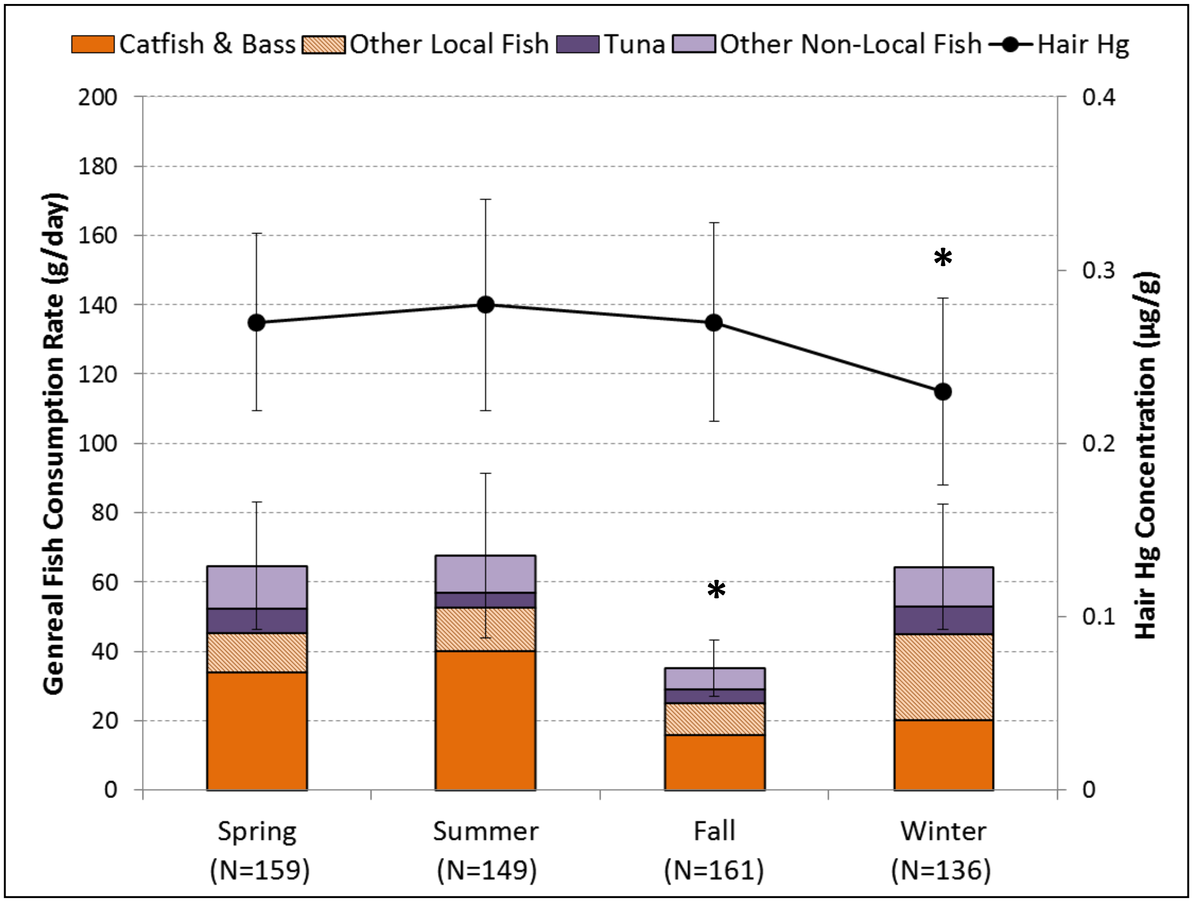


Figure 10. Fish consumption rates and hair mercury levels across all four seasons.

5. Discussion

As with any collaborative effort, the project sought to meet the goals of those with different interests tied to a common theme, in this case - the amount of mercury in fish and the level of health risk presented by consumption of those fish. In this regard, the project was successful in involving many different groups and meeting the goals of recruiting a variety of participants and developing an understanding of the relationship between their consumption of fish and resulting levels of mercury in their hair. Moreover, the project was able to identify future risks for mercury exposure in Grand Lake by assessing fish consumption levels for the most commonly eaten fish as well as developing quantitative measures of length versus mercury level for these fish. The latter was successfully used in developing site-specific and species-specific guidelines for fish consumption. Finally, the project was able to quantify consumption of non-local fish (e.g., seafood) among participants and these values could be helpful in determining appropriate fish consumption levels in relation to total daily mercury intake.

Unlike the general U.S. population, for whom a majority of mercury intake comes from seafood, our participants receive a majority of their dietary mercury from local fish, even though Grand Lake fish have relatively low levels of mercury. Based on our results, lake users can consume most fish at a rate of several times per month without exceeding mercury intake guidelines. It should be noted that this applies only to smaller fish and smaller species in general. While mercury levels in Grand Lake fish were generally below guidelines set by US EPA and ODEQ, larger fish and fish higher on the food web have elevated levels of mercury. Therefore, consumption of older, larger fish, especially flathead catfish, should be avoided for those who wish to limit their mercury exposure. Furthermore, about 5% of participants had elevated levels of mercury in their hair, so even in a lake with a relatively low level of mercury in fish, mercury exposure can be of concern if large quantities of high mercury fish are eaten. This raises concerns for the many lakes in Oklahoma that have higher levels of mercury. It should be noted that any reduction in mercury emission rates from local mercury sources in northeastern Oklahoma would likely reduce current mercury levels even further.

As seen in Figure 5, Grand Lake and Lake Hudson had some of the lowest mercury levels in largemouth bass that were measured in 60 lakes across Oklahoma between 2008 and 2012. At the beginning of the study, we anticipated that mercury levels in Grand Lake fish might be relatively high, given the number of local sources of mercury and maps of atmospheric deposition. Data collected from fish around the state have demonstrated that mercury levels in fish vary by location. Previous studies have shown that fish mercury levels can vary widely among lakes that receive similar inputs of mercury from the atmosphere depending on a combination of water quality, hydrological, and ecological conditions (Watras et al. 1995; Cabana et al. 1994). Bioaccumulation depends on water quality conditions, such as pH, dissolved oxygen, organic carbon, and sulfate that influence the conversion of inorganic mercury to methylmercury. Hydrological conditions that affect the extent and duration of stratification can affect mixing of methylmercury produced in the sediments with surface waters inhabited by fish. Ecological factors also affect mercury accumulation, such as food chain length, diet, and growth rates. The Grand Lake watershed has relatively little coverage by wetlands, where low oxygen conditions promote active mercury methylation, which may contribute to the relatively low levels of mercury in Grand Lake fish.

Despite the generally low levels of mercury for this species in Grand Lake, some individual fish had mercury levels greater than 300 ppb. A closer look at differences between Grand Lake and other state lakes (based on 2008-2012 surveys) reveals a similar pattern among species (Figure 11). Four lakes were chosen for this comparison based on geographic distribution across the state (Fort Supply Reservoir, Lake Eufaula, Lake Ellsworth, and Lake Hugo), and that fish sampling included similar fish to what were sampled in Grand Lake and Lake Hudson in this project. Average mercury values were much higher in all species for these four lakes. For example, average mercury levels in flathead catfish and largemouth bass for all four lakes exceeded 300 ppb. Given similar consumption patterns to what were measured during this project, it is anticipated that a substantial number of fishermen and their families would have mercury intake rates that exceed the health-based guidelines and far more than 5% would have hair mercury values exceeding 1.0 ppm. It should be noted that ODEQ has posted fish consumption advisories for a number of these lakes, which may influence the behavior of fishermen and fish consumers.

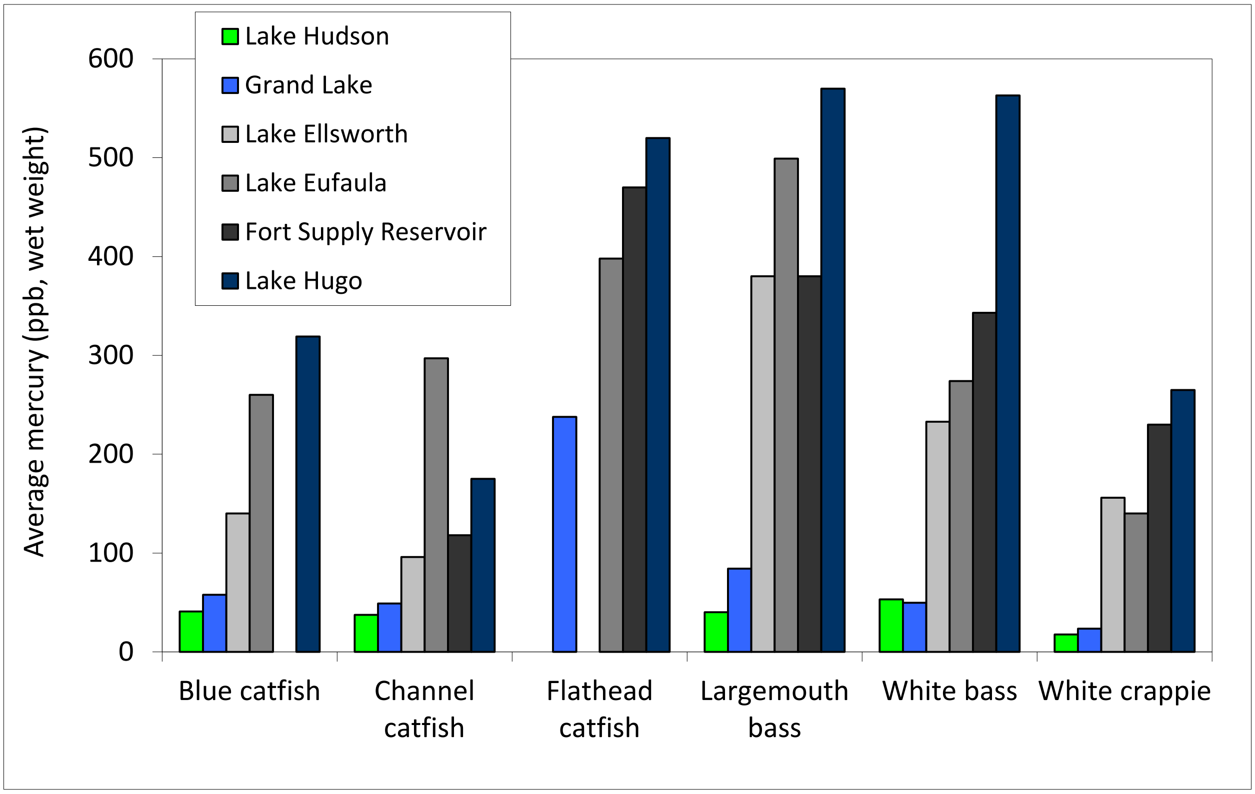


Figure 11. Mercury concentrations in 6 fish species from Grand Lake, Lake Hudson, and four other Oklahoma lakes.

Several recommendations can be made based on the findings of this research. First, mercury exposure from consumption of non-local, primarily saltwater, fish should be considered when setting fish consumption guidelines for local fish. Since reference dose values are based on total daily intake, neglecting a significant portion (>35% in this case) of daily intake may give a false impression that all mercury exposure comes from consumption of local fish. If consumers are to make the best choices to protect themselves, they should have complete information about…. Although data in this report were collected from one geographical area of Oklahoma and may not represent the entire state, consumption patterns measured were similar to national data, giving us added confidence that they are relevant for assessing statewide consumption patterns. A helpful approach for the future would be to gather more statewide data on fish consumption. This leads to the second recommendation, which is that additional hair samples should be taken in other areas, especially southeastern Oklahoma where fish mercury concentrations are much higher than in northeastern Oklahoma. A final recommendation is to identify and assess mercury exposure in local subpopulations with high rates of fish consumption. For instance, members of local Hmong populations also rely heavily on Grand Lake as a source of fish.

Beyond mercury, other contaminants in the Grand Lake watershed may pose human and ecological health concerns. Heavy metal contamination associated with legacy mining activities has led to elevated zinc, lead, and cadmium concentrations in parts of the Grand Lake watershed. Oklahoma DEQ has issued guidances for consumption of fish from Tar Creek and Grand Lake associated with lead bioaccumulation in some species of fish (ODEQ 2008). Another concern is the potential for blue green algae blooms, caused by highly elevated inputs of phosphorus. Certain types of blue green algae, such as the *Microcystis* bloom that impacted Grand Lake in 2011, can release harmful chemicals into the water and air and cause health effects in people and animals that come into contact with the water. While our testing of Grand Lake fish following the 2011 bloom did not show bioaccumulation of the microcystin toxin above the laboratory's detection limit (unpublished data), there is the potential for fish to bioaccumulate algal toxins (Poste et al. 2011). Our study provides an understanding of fish consumption patterns among people who rely on the waters of Grand Lake and can be used in evaluating risks associated with other contaminants in the lake. It also highlights the need to understand fish consumption patterns among anglers and their families in other Oklahoma lakes more severely impacted by mercury contamination.

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