Mercury in North Mississippi Lakes: A Growing Concern?

D.B. Huggett[1,2], J.A. Steevens[1,2], J.C. Allgood[1], C.B. Lutken[3], C.A. Grace[3], and W.H. Benson[1,2]
[1]Department of Pharmacology,
[2]Environmental and Community Health Research,
[3]Mississippi Mineral Resources Institute
The University of Mississippi, University, Mississippi

INTRODUCTION

In 1997, 2,299 fish consumption advisories were issued in the United States (USEPA 1997a). A majority of these advisories resulted from tissue concentrations of mercury in excess of the Food and Drug Administration's 1.0 mg Hg/kg standard. Mercury contamination may occur as a result of natural geochemical and/or anthropogenic sources. Mercury accumulates in the tissues of seafood, via these sources, primarily in the form of methyl mercury. Once ingested, 95% of methyl mercury is absorbed from the gastrointestinal tract into the blood. Kidney damage, cardiovascular collapse and death may occur if 10 to 60 mg Hg/kg is consumed in a short period of time, while neurotoxicity results when smaller concentrations of methyl mercury are absorbed over a long period of time (USEPA 1997b).

Contaminated food is the major route of exposure for humans to methyl mercury (USEPA 1997b). Currently in Mississippi, there are fish consumption advisories for mercury (mercury $\geq$ 1.0 mg Hg/kg tissue) in seven water systems, including Enid Lake and the Yocona River below Enid Lake. In 1996, the Mississippi Department of Environmental Quality (MSDEQ) reported mean mercury concentrations in largemouth bass from Enid Lake of 1.07 mg Hg/kg tissue. MSDEQ data from Grenada and Sardis Lakes, which do not have fish consumption advisories, indicate that mean mercury concentrations in largemouth bass were on average 1.07 and 0.85 mg Hg/kg tissue, respectively. Not only do these data indicate that there may be a potential risk for people who consume these fish, but it is unknown if mercury contamination in these lakes results in adverse effects on survival, growth and reproduction of aquatic organisms. In addition, it is unclear as to why a health advisory has been issued for Enid Lake, while no health advisory has been issued for Sardis or Grenada Lakes.

Based on available fish tissue data, the environmental fate and effects of mercury in Enid, Sardis, and Grenada Lakes merits attention. The objective of this study was to evaluate the parameters associated with mercury in fish tissue and compare three regional lakes in Mississippi. To address the main objective and issues described above, the specific aims of the study were to: 1) determine concentrations of mercury in sediment collected from Enid, Sardis, and Grenada Lakes; 2) determine concentrations of mercury in fish collected from Enid Lake; and 3) evaluate potential human hazard from fish consumption in area lakes.

MATERIALS AND METHODS

Site Description

Sardis, Enid, and Grenada Lakes are located within three different watersheds in Central and North Mississippi. Sardis Lake (Figure 1) is located in North Mississippi, approximately 50 miles south of Memphis, Tennessee. There are 98,357 acres of land and water associated with this reservoir. Enid Lake (Figure 2), located 70 miles south of Memphis, Tennessee, is also located in North Mississippi. The smallest of the lakes, Enid Lake only has 44,036 acres of land and water. Located in Central Mississippi, Grenada Lake (Figure 3) is 99 miles south of Memphis, Tennessee. Grenada Lake has 90,379 acres of land and water in its boundaries.

Sampling Regime

Surficial sediments from Enid Lake were collected in the Fall of 1997 with a gravity corer. A ponar dredge was used to collect sediments from Enid, Sardis, and Grenada Lakes in the summer of 1998. Samples were placed in ziplock plastic bags and stored at 4° C until analysis. Fish were collected from several locations on Enid Lake and pooled as one sample, in the Fall of 1998, with an Electro-Shocker Model C-phase electronic shocking device. Fish species of similar size were filleted, placed in aluminum foil and stored at 4° C until analysis. Fish analyzed included: largemouth bass (*Micropterus salmoides*), hardhead catfish (*Arius felius*), carp (*Cyprinidae* spp.), gar (*Lepisosteus* spp.) and black crappie (*Pomoxis nigromaculatus*).

Analytical Methodology

Sediment and fish were analyzed for total mercury utilizing a Varian SpectraAA 20 flame atomic absorption spectrophotometer with a Varian VGA-76 vapor generation accessory. EPA methods 245.5, and 245.6 were utilized for all mercury analyses. Briefly, two to five grams of sediment or fish were weighed into a BOD bottle. Concentrated nitric acid (3 ml) and hydrochloric acid (2 ml) were added to each sample and allowed to digest for at least 6 hours. Potassium permanganate (15 ml of 5% *w/v* solution) and potassium persulfate (10 ml of 5% *w/v* solution) were then added to each sample and allowed to digest for 15 minutes. Distilled, deionized water (65 ml), 5 ml of a 12% *w/v* sodium chloride and 12% *w/v* hydroxyamine hydrochloride solution, and 50ul of antifoam "B" silicone emulsion were added to each sample so that the total sample volume was 100 ml.

Method detection limits for fish tissue and sediment were 100 ng Hg/kg. Prior to analyses of sediment and tissue samples, a three point calibration curve, which also included a laboratory reference blank, was established. In addition, matrix spikes (85 - 115% recovery), initial and continuing calibration verification samples (ICV and CCV), and certifiable reference standards were utilized for quality assurance.

Exposure Assessment and Consumption Limits

Human exposure to mercury through ingestion of contaminated fish was determined according to methods outlined by USEPA (1989). The following equation was utilized to calculate human exposure:

$$\text{Intake (mg/kg/d)} = \frac{CF \times IR \times EF \times ED}{BW \times AT},$$

where CF = mercury concentration in fish (mg/kg), IR = ingestion rate (kg/meal), EF = exposure frequency (meals/yr), ED = exposure duration (yrs), BW = body weight (kg), and AT = averaging time (ED x 365 d/yr). Ingestion rate utilized in this study was 0.227 kg/meal, which is equivalent to an 8 oz. fish meal. Average exposure frequency for the United States is 48 d/yr, while a 30 year exposure duration is typical for assessing noncarcinogenic effects. The average body weight for an adult is 70 kg, while 14.5 kg is average for children (USEPA 1989). In this analysis, it was assumed that 100% of mercury ingested is absorbed into the blood stream.

A hazard index (HI) for each fish species was calculated by dividing the intake (as calculated above) by the reference dose (RfD) for methyl mercury. An RfD is a value which estimates an exposure in which no toxic effects are to occur over a lifetime. The RfD for methyl mercury is 1.0×10^{-4} mg/kg/d. An HI less than one indicates that toxic effects are not predicted to occur to those who ingest the fish; however, if the HI is greater than one, effects are predicted to occur (USEPA 1989).

Monthly consumption limits for Enid Lake fish were determined using methods from USEPA (1997b). Limits were calculated utilizing the equation:

$$CR_{mm} = \frac{RfD \times BW}{C_m} \times \frac{30.44\ \text{d/mo}}{IR},$$

where RfD = reference dose (1×10^{-4} mg/kg/d), BW = body weight (70 or 14.5 kg), Cm = concentration in fish (mg/kg), IR = ingestion rate (0.227 kg/meal).

RESULTS AND DISCUSSION

Mercury in Sediment

Mean sediment concentration in Enid Lake in 1998 (0.088 ug Hg/g) was not significantly different than that measured in 1997 (0.154 ug Hg/g). Concentrations in Sardis (0.027 – 0.113 ug Hg/g) and Grenada Lakes (0.076 – 0.168 ug Hg/kg) were not significantly different than those observed in Enid Lake (0.041 – 0.116 ug Hg/kg). Sediment mercury concentrations in all three lakes investigated were similar, indicating that the amount or mass of mercury that Enid Lake is receiving is similar to the amount that Sardis and Grenada Lakes are receiving. These three lakes are in different watersheds, indicating that a point source for mercury contamination is not probable.

Concentrations in these sediments are much less than those reported by Bloom et al. (1999) at a chlor-alkali plant in Lavaca Bay, Texas. Sediment concentrations ranged from 5 to 790 ng Hg/g, with 1 to 2% of this being methyl mercury. However, concentrations in the porewater of these sediments were up to 80% methyl mercury. It was suggested that methyl mercury was bound to metal oxides, while inorganic mercury was bound to organic carbon and sulfides. Rood et al. (1995) reported concentrations of mercury in the Florida Everglades similar to data in the present study, with an average concentration of 120 ng Hg/g. The Florida Everglades also have a fish consumption advisory due to fish tissue exceeding 1.5 mg Hg/kg tissue. Atmospheric deposition, both global and regional, has been implicated as the source which has enriched Everglades sediments and caused consumption advisories. Mercury in Florida bays and estuaries ranged from 1 to 219 ng Hg/g with an average of 0.77% being methyl mercury (Kannan et al. 1998). Organic carbon was related to the concentration of total mercury, but not methyl mercury. There also was a positive correlation between total sediment mercury concentrations and those observed in fish.

Porewater concentrations are important to consider because porewater is thought to be the major route of exposure for benthic organisms (Muir et al. 1985). With up to 80% of mercury in the methylated state (Bloom et al. 1999), benthic organisms may readily accumulate mercury. When *Hexagenia rigida* was exposed to a bulk sediment concentration of 0.5 mg Hg/kg, it was noted that accumulation was 60 times higher when exposed to methyl mercury as opposed to inorganic mercury (Saouter et al. 1993). In addition, no changes in growth or survival were noted after a nine day exposure period. In Clear Lake, California, invertebrate diversity decreased with increasing sediment mercury levels (0.27-183 ug Hg/g) (Suchanek et al. 1995).

Mercury in Fish

Mean mercury concentrations in fish from Enid Lake ranged from 0.634 mg Hg/kg in carp to 1.89 mg Hg/kg in gar. These concentrations indicate that mercury accumulation is dependent on the trophic status of the fish in Enid Lake. Gar, black crappie and largemouth bass, which are predators, contained the highest concentrations while scavengers, such as catfish and carp, contained lower concentrations. It should be noted that one catfish and one carp contained concentrations greater than 1 mg Hg/kg tissue. The catfish was almost twice the size of the other catfish, indicating the accumulation also may be dependent on age and size. It has been shown that mercury accumulation is very size dependent (Jackson 1990); however, Henry et al. (1998) indicated that here was no relationship between mercury tissue concentrations and smallmouth bass size. Concentrations of mercury present in these fish differ greatly than those presented in other studies. Henry et al. (1998) reported total mercury concentrations in smallmouth bass (*Micropeterus dolomieui)* collected from Fumee Lake in the Upper Peninsula of Michigan that were less than the FDA action level (average was 0.22 ug/g). Water quality in Fumee Lake though (pH=8.41, alkalinity and hardness = 128 and 152 mg $CaCO_3$) indicates that methyl mercury formation and thus bioaccumulation is not likely. Fish collected from Florida bays and estuaries contained on average 1.41 ug/g total mercury, with up to 100% of the total mercury being methyl mercury. Fish collected near an inactive mercury mine in Clear Lake, CA, contained average mercury concentrations less than 0.85 ug Hg/g (Harnly et al.1997). The National Bioaccumulation Study (NBS) indicated that background concentrations in fish around the country was 0.16 ug/g, while fish in agricultural areas averaged 0.17 ug/g (Bahnick and Sauer 1994). Located in a predominately agricultural area, Enid Lake fish contained considerably higher mercury concentrations than those observed in the NBS (Bahnick and Sauer 1994). Specifically, average concentrations in carp and largemouth bass (0.11 and 0.46 ug/g, respectively) collected in the NBS were several orders of magnitude less than those observed in Enid Lake.

Niimi and Kissoon (1994) reported that liver, kidney and spleen accumulated more mercury than brain and muscle tissue in *Oncorhynchus mykiss*; however, only a small difference existed between muscle concentrations and total body burden concentrations. Total body burden concentrations between 10 to 20 mg Hg/kg may cause lethality (Niimi and Kissoon 1994).In addition, a maximum allowable toxicant concentration (MATC) for reproductive impairment of fathead minnow after a 41 week exposure to inorganic mercury was 1.4 mg Hg/kg tissue (Snarski and Olson 1982). For chronic effects, 1 to 5 mg Hg/kg has been proposed as a threshold concentration above which adverse effects may occur (Niimi and Kissoon 1994). Fish in this study (at least one from each species tested) contained muscle concentrations greater than 1 mg Hg/kg in the muscle, indicating that chronic mercury toxicity may be occurring in Enid Lake fish.

Exposure Assessment

Mean total mercury concentrations in largemouth bass, gar and black crappie exceeded the FDA action level of 1.0 mg Hg/kg; however, only one carp and one catfish analyzed had mercury concentrations in the edible fillet above the 1.0 mg Hg/kg FDA action level. Utilizing EPA guidelines, human adults are being exposed to as much as 8.0×10^{-4} mg Hg/kg/d, while childern may be exposed to 3.8×10^{-3} mg Hg/kg/d. Hazard indexes for both adults and children consuming any of these fish species are above 1, indicating that it is hazardous for both groups to consume Enid Lake fish. It is at least an order of magnitude more hazardous for children to consume these fish than adults. Consumption limits calculated for adults indicate that six, 0.227 kg meals of bass, crappie and gar may be eaten a year, while twelve catfish or eighteen carp meals may be eaten a year. Children should limit consumption to one meal of crappie, bass or gar a year, but may eat three meals of carp or catfish per year.

Fleming et al. (1995) investigated human consumption in the Florida Everglades and observed that average mercury hair concentrations for fishermen were 3.62 ug/g, which is considerably lower than the benchmark dose for effects of 11 ug Hg/g hair. Those identified to be at greatest risk were African Americans and those in lower socio-economic levels which were less likely to know about the mercury advisory in the Florida

Everglades.This may be equally true at Enid Lake where the consumption advisory is not adequately posted and subsistence fishing is considered high. However, Flemming et al. (1995) also noted that those who did know about the advisory (71%), did not change their consumption habits due to the advisory. Native Americans living near an inactive mercury mine site contained an average hair and blood concentration of 0.64 ug/g and 15.6 ug/L, respectively (Harnly et al. 1997). Blood concentrations less than 44 ug/L are considered safe (USEPA 1997b).

CONCLUSIONS

The mechanism by which mercury is entering and enriching North Mississippi Lakes is unknown. Sediment data from this study suggest that a point source discharge is not likely since all three lakes have similar sediment concentrations and are in different watersheds. Natural inputs, such as would be expected in select areas in Arkansas, do not seem likely based on the topography of the area. Atmospheric deposition has been implicated as the primary mechanism by which the Florida Everglades and many other areas have been contaminated with mercury. Swain et al. (1992) illustrated that in seven undisturbed lakes in Minnesota and Wisconsin, the average deposition to these lakes was 12.5 ug/yr/m which was well above the preindustrial value of 3.7 ug/yr/m.These areas, much like Sardis, Grenada and Enid Lakes, are remote with little or no industry in the immediate area. Rolfhus and Fitzgerald (1995) concluded that in coastal areas, only 5.4% of the total mercury deposited atmospherically is needed to maintain mercury concentrations in marine fish at 0.20 mg Hg/kg tissue. With atmospheric concentrations of mercury increasing 1.46 $\pm$ 0.17% a year and with up to 25% of mercury deposition to a terrestrial catchment entering a lake, one may hypothesize that atmospheric deposition may be responsible for at least part of the mercury enrichment in Northern Mississippi Lakes.

Sediment concentrations in Enid, Sardis and Grenada Lakes are similar to those in the Florida Everglades, where fish consumption advisories have been posted since 1989. While it is apparent that fish from Enid Lake are accumulating levels of mercury which may be hazardous to those consuming these fish, it is unknown whether fish from Sardis and Grenada Lakes are hazardous to human health. In addition, available data indicate that chronic toxicity may be occurring in Enid Lake fish, while toxicity to invertebrates is unknown. Further studies are needed to address the fate and effects of mercury in North Mississippi Lakes.

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Table 1.Concentrations of Mercury in Sediment (mg/kg) Collected from North Mississippi Lakes.

Lake	Year	Mean (SD)	Range
Enid	1997	0.154 (0.061)	0.067 – 0.476
Enid	1998	0.088 (0.028)	0.041 - 0.116
Sardis	1998	0.112 (0.170)	0.027 - 0.113
Grenada	1998	0.133 (0.028)	0.076 - 0.168

Table 2.Concentrations of Mercury (mg/kg) in Fish Collected from Enid Lake.

Fish Type	Mean (SD)	Range
Carp	0.634 (0.453)	0.352 - 1.218
Largemouth Bass	1.400 (0.300)	1.122 - 1.868
Gar	1.890 (0.307)	1.584 - 2.198
Black Crappie	1.690 (0.100)	1.590 - 1.790
Catfish	0.820 (0.567)	0.425 - 1.660

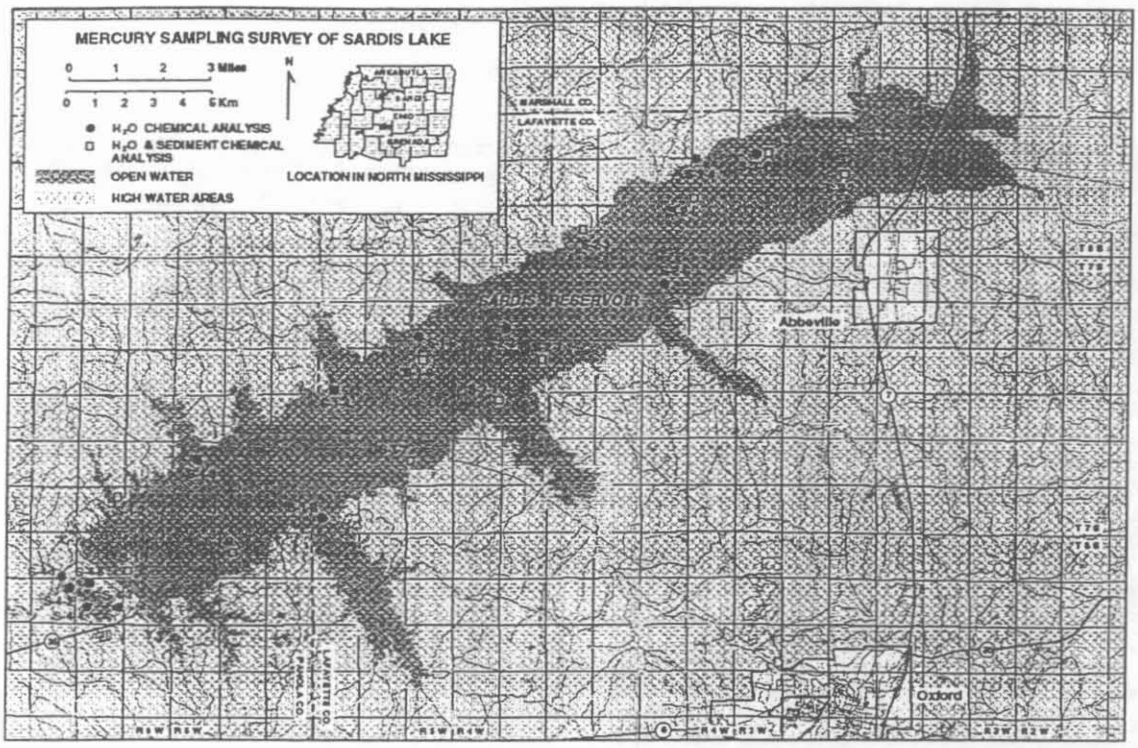

Figure 1.

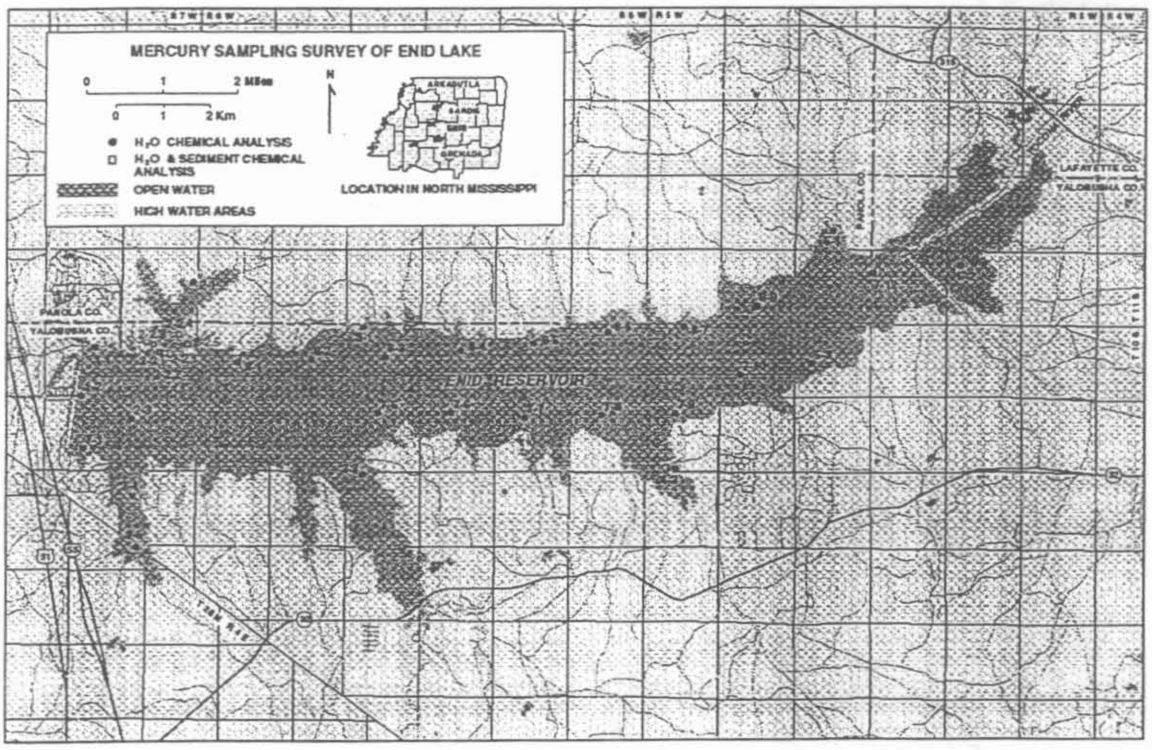

Figure 2.

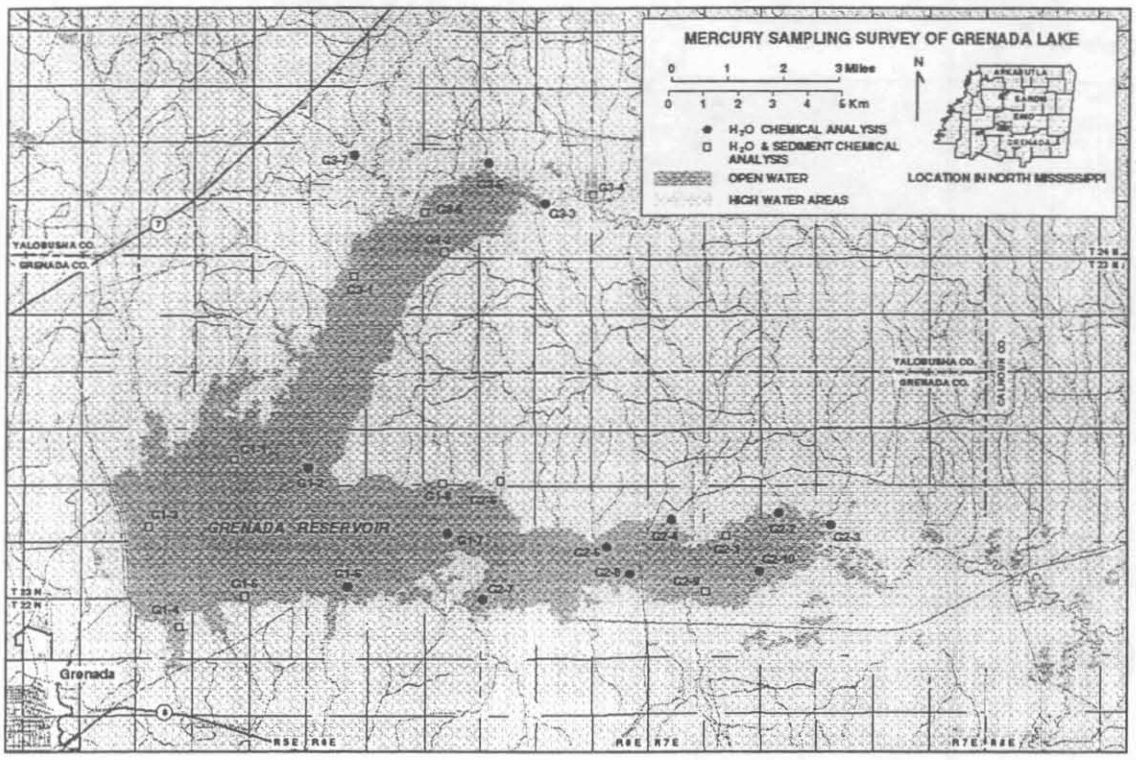
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Figure 3.

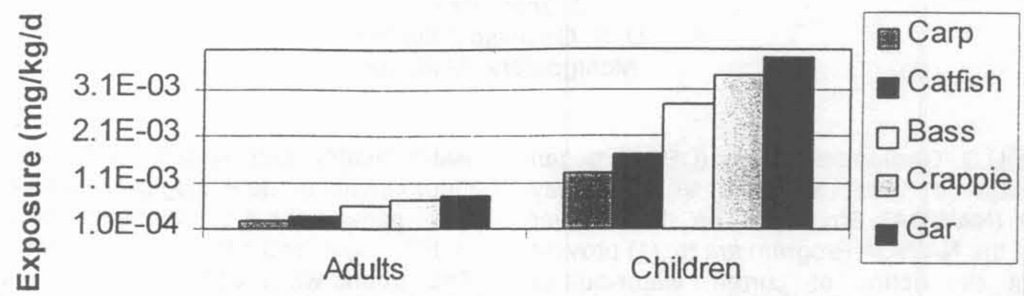

Fig. 4. Adult and Child Exposure to Mercury Through Consumption of Enid Lake Fish

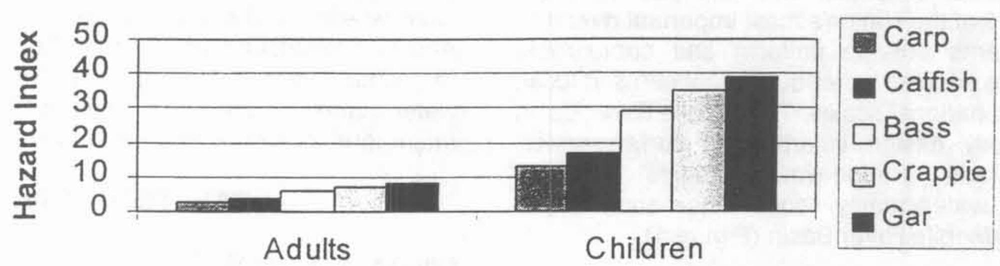

Fig. 5. Hazard Index for Adults and Childern Consuming Enid Lake Fish

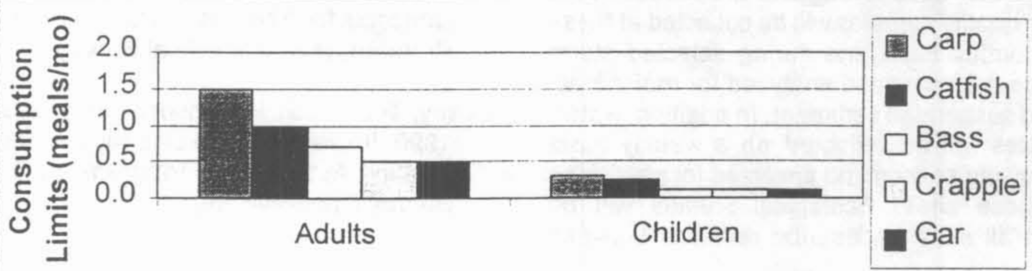

Fig. 6. Consumption Limits for Adults and Childern Consuming Enid Lake Fish