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A Survey of Mercury Concentrations in the Cypress Creek and Upper Sabine River Basins of Northeast Texas

Field Operations Division

TEXAS NATURAL RESOURCE CONSERVATION COMMISSION

A Survey of Mercury Concentrations in the Cypress Creek and Upper Sabine River Basins of Northeast Texas

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ABSTRACT

The main objective of this study was to assess the geographical occurrence of mercury in northeast Texas. Sediment, soil, water, and fish samples were collected during 1993-95 from two river basins in northeast Texas that had factors favorable for mercury uptake (e.g. low pH, low alkalinity, low calcium, high total organic carbon, and seasonally flooded wetlands).

Mercury concentrations in water were highly variable (0.05-3.04 µg/L). Sediment concentrations were equally variable (<0.0l-1.57 mg/kg). Upland soils throughout the region had levels ranging from 0.01-o. 14 mg/kg. Fish samples from a large geographical area contained detectable concentrations of mercury. There was a significant difference ($P<0.05$) between mercury concentrations in largemouth bass muscle tissue from fish of different size groups from Caddo Lake. Mercury concentrations in largemouth bass from Caddo Lake were positively correlated to size ($r^2=0.71$). Bass from two natural lakes (Caddo and Pruitt lakes) had higher mercury concentrations than any other location, although it was unclear whether the differences were statistically significant.

The regulated communities' self-reporting data was reviewed for current point-source discharges of mercury to water. Mercury was not reported in any of the data.

Historical data relative to potential atmospheric loading are presented using tree cores from several counties in northeast Texas. No conclusions are attempted based on the limited data.

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INTRODUCTION

MERCURY IN THE ENVIRONMENT

Mercury is a naturally occuring element in the earth's surface and is present in low concentrations in all organisms. Mercury can enter the aquatic environment from areas of naturally occurring cinnabar (mercuric sulfide) formations, point-source discharges, and atmospheric deposition.

Geological

The average mercury concentration in minerals is 0.08 mg/kg; but mercuric sulfide, also known as cinnabar, can have levels up to 10,000 mg/kg. Cinnabar formations in Texas are found in Lower Cretaceous limestones like those in the Presidio County/Big Bend area. There was an active mercury mine there from 1900 through the 1970s. Cinnabar is present to a lesser extent in the Eagle Ford clays of the Upper Cretaceous (Sellers *et al*., 1978). The Eagle Ford clays roughly correspond to the Blackland Prairie region from central Texas to the Red River area.

Anthropogenic Sources

Major industrial processes that have introduced mercury into the environment include the production of chlorine, caustic soda, and slimicides. There is concern that sources of mercury loading are shifting from industrialized areas of the Northern Hemisphere to second and third world countries in the Southern Hemisphere. In addition to mining, current high-temperature industrial practices that release significant amounts of mercury into the atmosphere include emissions from coal-fired power plants; copper, lead, and iron smelters; cement manufacturing; and medical and municipal incinerators (Pacyna, 1987; Paasivirta, 1991; Keating, 1994). In spite of electrostatic scrubbers, 90-95% of the mercury in coal-fired generating plants is lost to the atmosphere (Hutchinson, 1987). Regulatory activity is being considered to set emissions standards for some of these major sources (Keating, 1994).

Atmospheric deposition is influenced by a complex combination of local, regional, and global emissions and transport/transformation processes. The relative contribution of local, regional, and global sources is site-specific and cannot be extrapolated to other sites (EPRI, 1994). Recent articles on global mercury cycling suggest that anthropogenic contribution to the total atmospheric mercury budget exceeds natural inputs (Linqvist and Rodle, 1985; Nriagu, 1989; Fitzgerald and Clarkson, 1991; Paasivirta, 1991; EPRI, 1994). Some models suggest about half of anthropogenically related emissons to the atmosphere are produced and deposited on a local or regional scale, while the other half contribute to the global mercury cycle (Fitzgerald, 1995).

Regardless of the source, mercury is naturally volatile. It is present in the atmosphere in a vapor form and is taken up on air currents and transported worldwide. The ultimate sink for atmospheric mercury is land (Fitzgerald, 1995). Effects from anthropogenic mercury loading to the aquatic environment via runoff from soils high in mercury concentrations have the potential to last for long periods of time after mercury emissions have been reduced.

Sediment

Mercury content in recently deposited sediments varies considerably, for example: Lake Ontario 0.31-1.0 mg/kg; lakes in Switzerland 0.01-2.23 mg/kg; and the Wisconsin River 0.4-2.7 mg/kg (Paasivirta, 1991). Background data for sediments collected along the East Coast and Gulf Coast of the United States show mercury values of 0.11 mg/kg at 1% aluminum (Al) concentration, and 0.21 mg/kg at 10% Al (Hansen *et al.*, 1993). The 85th percentile for mercury in sediment collected by the Texas Natural Resource Conservation Commission (TNRCC) from reservoirs statewide is 0.16 mg/kg, and 0.12 mg/kg from freshwater streams.

Since there are no numeric criteria for sediment metals, mercury concentrations must be evaluated using the literature. Long and Morgan (1990) published numeric levels for estuarine sediments that represented the 10th percentile of data that showed toxicity and sublethal effects. This value represents the level above which effects are first seen in sensitive species or life stages (Effects range low=ERL). They also published numeric levels for the 50th percentile, which represents the level above which effects were frequently, or always observed among most species (Effects range median=ERM). The ER-L and ER-M for mercury are 0.15 and 1.3 mg/kg, respectively (Long and Morgan, 1990).

Factors Affecting Biological Uptake

Mercury appears in aquatic organisms primarily as methylmercury. It is in this form that most mercury is transfered through the food web. Mercury is taken up differentially in the aquatic environment depending on a number of factors. The most conducive environmental conditions for the methylation and uptake of mercury into the aquatic food web include: low pH (<7), low alkalinity mg/L), low calcium (<15 mg/L), high total organic carbon (TOC), low chlorophyll-a (<10 µg/L), and significant seasonal fluctuations in water level (Rada *et al.*, 1989; Cope *et al.*, 1990; Wiener *et al.*, 1990; Lange *et al.*, 1993; Wiener, 1995). Methylmercury is produced initially by microbial activity. Although nearly insoluble in water, methylmercury forms colloids with humus. Humic material transfers mercury from soils to water, then to the food web via the microbial process. Wetlands are a ready source of dissolved organic carbon which complexes and transports mercury. Many east Texas lakes and sluggish stream systems exhibit characteristics that are favorable for mercury uptake into the aquatic environment. Newly impounded lakes, as well as lakes with high organic input, tend to have elevated levels of mercury in the aquatic fauna (Paasivirta, 1991; Wiener, 1995). Miller and Akagi (1979) suggest that as pH decreases, partitioning of mercury is shifted from sediment to water. A major pathway for mercury removal from solution is chemical binding with reduced sulphur, which has a high affinity for mercury. Lindberg *et al.* (1987) suggest that increased concentrations of some metals such as iron and manganese affect the non-biological methylation of mercury. Iron and manganese sulfides, known as acid volatile sulfides (AVS), are a reactive pool of solid phase sulfides that are available to bind with metals, such as mercury (Di Toro *et al.*, 1990). When bound by AVS, mercury is less available for uptake by aquatic organisms.

Fish Tissue

The National Study of Chemical Residues in Fish detected mercury at 92.2% of the 374 sites surveyed. Maximum, mean, and median concentrations in fish tissue were 1.80,0.26, and 0.17 mg/kg (U.S. EPA, 1992). Concentrations of mercury in fish from diverse locations nationwide did not change appreciably between 1976 and 1984 (Schmitt and Brumbaugh, 1990). Elevated mercury concentrations in fish have occurred in remote locations in Florida, Wisconsin, Ontario, and Scandinavia, where no known point sources or cinnabar formations exist (Evans, 1986; Johnson, 1987; Wiener *et al.*, 1990; Paasivirta, 1991; Lange *et al.*, 1993). Mercury concentrations in fish were positively correlated with sediment loading in the Ontario study (Johnson, 1987). In each study, the authors listed atmospheric deposition and increased mercury concentration in surface sediments as important processes in the uptake of mercury into the aquatic food web. Without historical data, it is impossible to know whether elevated mercury levels in fish from remote regions represent a recent phenomenon, or if this is an entirely natural process (Lindberg *et al.*, 1987).

Biomagnification of Mercury in the Environment

Mercury biomagnifies as it moves up trophic levels in the food web. Biomagnification by plankton from water has been measured as high as 100,000 times. Food web enrichment is apparent in plankton and aquatic insects, insect-eating fish, piscivorous fish, and fish-eating birds (Paasivirta, 1991).

Plants

Mercury uptake by plants apparently is low (Jenkins, 1981; Paasivirta, 1991), but the uptake is sufficient for plants to be included in an overall monitoring program (Jenkins, 1981). Mercury contamination downwind of an acetaldehyde plant in Japan has been documented from tree rings (Suzuki, 1994). Trees uptake mercury through the xylem, either from soil via the root system, or from direct deposition via foliage and bark. Some tree species are better than others for use in dendrochemical studies. Ideal species include those that are long-lived, grow over a wide range of sites, and have a large geographical distribution, a distinct heartwood with a low moisture content, a low number of rings in the sapwood, and low radial permeability (Cutter and Guyette, 1993). Species from northeast Texas that have these qualities include white oak (*Quercus alba*), post oak (*Q. stellata*), and eastern red cedar (*Juniperus virginiana*).

MERCURY (Hg) CONCENTRATIONS IN NORTHEAST TEXAS

Sediment

Several sediment surveys have been conducted in northeast Texas during the 1990s. TNRCC's Surface Water Quality Monitoring (SWQM) sediment data from 1990-95 include ranges and means&standard deviations ($\bar{x}\pm sd$) for:

- Mid-Caddo Lake (0401.0100)
  Hg <0.0l-1.57 mg/kg;
  0.37±0.55 mg/kg (n=7);
  total organic carbon (TOC) 1.4-4.5% (n=4).

- Big Cypress Creek/Lake 0' the Pines (0404.0020)
  Hg <0.01-0.45 mgkg;
  0.30±0.28 mg/kg (n=8);
  TOC 1.8-3.3% (n=2).

U.S. Army Corps of Engineers (COE) surveyed sediment during 1991 for the proposed Red River waterway project. COE data include:

- Lake O' the Pines $\bar{x}$ 0.176 mg/kg (n=6); maximum 0.405 mg/kg;
- Big Cypress Bayou $\bar{x}$ and maximum were both <0.1 mg/kg;
- Caddo Lake $\bar{x}$ 0.132 mg/kg (n=7), maximum 0.262 mg/kg (COE, 1994).

U.S. Fish and Wildlife Service (USFWS) data from a survey of the Sulphur River basin include:

- Days Creek/Sulphur River <0.0l-0.09 mg/kg (n=15) (Inmon ***et al.*** 1993).

Water

Water quality criteria in the Texas Surface Water Quality Standards for protection of aquatic life are 1.3 µg/L total mercury for freshwater chronic; 2.4 µg/L for freshwater acute (TAC, 1995).

TNRCC (1996) data include:

- mid Caddo Lake (0401.0100), 1990-93,
  dissolved mercury <0.2-0.74 µg/L (n=3);
- mid Caddo Lake (0401.0100), 1994-95,
  total mercury <0.01-0.02 µg/L (n=4);
- Big Cypress Creek/Lake O'the Pines (0404.0020). 1990-93,
  dissolved mercury <0.02-0.51 µg/L (n=5).
- Big Cypress Creek/Lake O'the Pines (0404.0020), 1994-95,
  total mercury 0.01-0.78 µg/L (n=3).

Mean concentrations of total mercury in water from the COE's 1991 study were:

- Lake O' the Pines 0.3 µg/L (n=6);
- Big Cypress Bayou <0.2 µg/L (n=8);
- Caddo Lake 0.2 µg/L (n=7) (COE, 1994).

Fish Tissue

TNRCC samples from a fixed-station at mid Caddo Lake (0401.0100) from 1990-94 include individual and composite samples of whole fish with mercury concentrations from:

- spotted gar, *Lepisosteus oculatus* 0.32 mg/kg (n=l);
- largemouth bass, *Micropterus salmoides* 0.33-0.45 mg/kg (n=9);
- chain pickerel, *Esox niger* 0.55 mg/kg (n=l) (TNRCC, 1996).

TNRCC samples from a fixed-station site at Big Cypress Creek/Lake O'the Pines (0404.0020) from 1990-95 include individual and composite samples of whole fish with mercury concentrations from:

- mixed sunfish, ***Lepomis*** spp. 0.13 mg/kg (n=7);
- channel catfish, ***Ictalurus** punctatus* 0.06-0.10 mg/kg (n=5);

- white bass, *Morone chrysops* 0.14 mg/kg (n=1);
- largemouth bass 0.15-0.22 mg/kg (n=3);
- carp, *Cyprinus carpio* 0.19 mg/kg (n=1);
- spotted gar 0.44 mg/kg (n=1) (TNRCC, 1996).

Texas Department of Health (TDH) sampled several areas in east Texas during 1994-95. Seven water bodies were sampled, and a variety of species were collected at each site. Selected results include individual fish with muscle tissue concentrations from:

Caddo Lake

- channel catfish 0.14-0.36 mg/kg (n=5), x 0.21 mg/kg;
- white bass 0.15-0.78 mk/kg (n=7), $\bar{x}$ 0.44 mg/kg;
- largemouth bass 0.21-1.63 mg/kg (n=23), $\bar{x}$ 0.83 mg/kg;
- freshwater drum, *Aplodinotus grunniens* 0.92-1.53 mg/kg (n=8), R 1.27 mg/kg.

Lake O' the Pines

- channel catfish 0.03-0.135 (n=7), $\bar{x}$ 0.06 mg/kg;
- largemouth bass 0.10-0.66 mg/kg (n=23), $\bar{x}$ 0.25 mg/kg.

Big Cypress Creek

- white bass 0.11-0.25 mg/kg (n=3), $\bar{x}$ 0.18 mg/kg;
- largemouth bass 0.12-0.94 mg/kg (n=5), $\bar{x}$ 0.44 mg/kg;
- freshwater drum 0.23-1.29 (n=3), $\bar{x}$ 0.60 mg/kg;
- bowfin, *Amia calva* 0.94-1.55 (n=2), $\bar{x}$ 1.20 mg/kg.

Toledo Bend Reservoir

- freshwater drum 0.17-0.60 (n=7), $\bar{x}$ 0.29 mg/kg;
- white bass 0.14-0.93 (n=6), $\bar{x}$ 0.39 mg/kg;
- largemouth bass 0.10-1.65 (n=48), $\bar{x}$ 0.90 mg/kg (TDH, 1996).

USFWS surveyed Days Creek and the Sulphur River near Texarkana and found composite, whole-fish concentrations ranging from:

- mixed sunfish 0.090 mg/kg (n=8);
- green sunfish, *Lepomis cyanellus* 0.317 mg/kg (n=2);
- yellow bullhead, *Ameiurus natalis* 0.078-0.200mg/kg (n=29);
- channel catfish 0.180 mg/kg (n=6);
- bowfin 0.200 mg/kg (n=1);
- largemouth bass 0.370 mg/kg (n=4);
- spotted gar 0.407-0.510 mg/kg (n=5) (Inmon ***et al.,*** 1993).

TDH conducted a similar survey in Days Creek and the Sulphur River with concentrations in individual and composite muscle tissue ranging from:

- striped bass, *Morone saxatilis* 0.31 mg/kg (n=1);
- freshwater drum 0.32 mg/kg (n=1);
- yellow bullhead 0.41 mg/kg (composite);

- largemouth bass 0.46 mg/kg (n=l);
- white bass 0.49 mg/kg (n=l) (TDH, 1996).

The Academy of Natural Sciences of Philadelphia (ANSP) conducted a survey of the Sabine River near Longview during October 1995. Individual muscle tissue concentrations ranged from:

- blue catfish, *Ictalurus furcatus* 0.13-0.43 mg/kg (n=8);
- channel catfish 0.14-0.22 mg/kg (n=4);
- longnose gar, *Lepisosteus osseus* 0.34-0.58 mg/kg (n=8) (ANSP, 1996).

HEALTH EFFECTS

Humans

Mercury in fish tissue is a human health concern over a large part of North America, Europe, and parts of the Southern Hemisphere Lindberg ***et al.,*** 1987). There are health advisories either banning or limiting the consumption of fish in 34 states (U.S. EPA, 1994). Mercury is a neurotoxicant. Early symptoms of long-term consumption of fish containing elevated mercury concentrations include tingling of the extremities; loss of coordination, and tunnel vision. Residence time for mercury in humans is relatively short (70-76 days), but is much longer in fish (400-1,000 days) (Paasivirta, 1991). There is added concern since fetuses and pregnant women are at increased risk of adverse neurological effects from exposure to methylmercury (World Health Organization (WHO), 1990).

Aquatic Organisms

Among aquatic species, mercury has caused behavioral modifications growth inhibition, reproductive impairment, decreased embryo-larvae survival, and a variety of neurological and enzymatic dysfunctions (Zillioux et ***al.,*** 1993).

Birds and Mammals

Fish-eating birds accumulate mercury at predictable rates, making them reliable indicators of ecological damage (Zillioux et ***al.,*** 1993). Bald eagles with elevated blood levels of mercury have been shown to have reduced reproductive success (Wiemeyer et ***al.,*** 1984). Mammals that consume mercury-laden fish such as the Florida panther, have been documented to have impaired reproductive success (Roelke et ***al.,*** 1991). Mink populations in South Carolina, North Carolina, and Georgia are declining and individual animals have been documented with elevated levels of mercury in the blood (Southern States Mercury Task Force (SSMTF), 1994).

HEALTH ADVISORIES

Screening Levels

Several screening levels are used to determine whether mercury concentrations in fish are safe for human consumption. The U.S. Food and Drug Administration (FDA) sets action levels for fish shipped in interstate commerce. FDA's action level is based on national consumption patterns which are protective of the general population, because on average the general population does not consume large amounts of fish, and because marketplace fish come from many sources which on average are low in mercury. FDA's action level underestimates exposure to certain subgroups of the population. The current FDA action level for mercury in fish in interstate commerce is 1.0 mg/kg. The U.S. Environmental Protection Agency EPA) uses risk-based formulas to establish screening levels. Due to uncertainty over possible increased risk to certain subpopulations, the EPA has recommended that the screening level for mercury in fish be lowered to 0.6 mg/kg while the reference dose is being reevaluated (U.S. EPA, 1993). Individual states can set their own screening values and issue their own advisories.

Texas

The TDH does not have a set mercury concentration in fish tissue at which it issues fish consumption advisories. Instead, TDH uses an aggregrate risk-based assessment that is protective of the most sensitive subgroups of the population, such as women of child-bearing age that are exposed to mercury in fish. Toxicologists use a reasonable maximum exposure level that considers the average mercury concentration by species, fish size, and geographical area.

TDH issued a fish consumption advisory for Caddo Lake during January 1995. The advisory recommends that people not consume largemouth bass greater than 18 inches in length, or freshwater drum of any size from Caddo Lake due to elevated mercury concentrations. In August 1995, TDH expanded the advisory to include consumption limits on chain pickerel from Caddo Lake; largemouth bass and hybrid striped/white bass from Toledo Bend Reservoir; largemouth bass >18 inches and hybrid striped/white bass from Sam Rayburn Reservoir, largemouth bass, white bass and freshwater drum from Steinhagen Reservoir; and largemouth bass >18 inches, flathead catfish ***Pylodictus olivaris,*** and bowfin from Big Cypress Creek (TDH, 1995).

Arkansas

Twelve counties in Arkansas have fish consumption advisories in effect for mercury. The Ouachita Mountains are volcanic in origin and have elevated mercury concentrations in soil and sediment compared to other parts of the state. In addition, approximately 50 mercury mines were in operation at one time in the three county area around Camden, Arkansas (SSMTP, 1994).

Louisiana

Louisiana has fish consumption advisories on the Ouachita River and 10 reservoirs in the northeastern part of the state. During the fall of 1993, largemouth bass from 12 northern Louisiana lakes were analyzed for mercury Two of 15 individual fish from Caddo Lake contained mercury concentrations in excess of Louisiana's screening level of 0.5 mg/kg. Prom Cross Lake, which is also in the Cypress River basin, 12 of 15 fish had mercury concentrations at or near 0.5 mg/kg. In the Sabine River basin, the upper part of Toledo Bend Reservoir was sampled and seven of 15 individuals contained >0.5 mg/kg mercury (SSMTP, 1994).

Other Southern States

In Oklahoma, McKee Creek Reservoir has a fish consumption ban in effect for mercury. Other southern states that have fish consumption advisories for mercury include Florida, Alabama, Georgia, Tennessee, Missouri, South Carolina, and North Carolina.

STUDY OBJECTIVES

The main objective of the present study was to assess the geographical occurrence of mercury in northeast Texas. Sediment, soil, water, and fish samples were collected from two river basins that had characteristics favorable for mercury uptake. Another objective included identifying possible point sources of mercury to water through a review of the regulated communities' self-reporting data. Possible changes in atmospheric mercury loading over the past 100 years also were of interest. An attempt was made to obtain historical data using tree cores from several counties in northeast Texas and sediment cores from a sphagnum bog marsh. Guidance for future studies was also formulated.

STUDYAREA

CYPRESS CREEK BASIN

Background Infomation

The Cypress Creek basin has the smallest drainage area of any river basin in Texas. The economy in the basin is dominated by agriculture and forest-related industries. Many streams in the basin have sluggish flow characteristics, seasonally low dissolved oxygen, and naturally high organic loading (TNRCC, 1994a).

TNRCC's Surface Water Quality Monitoring (SWQM) data for selected parameters from the Cypress basin from 1990 through 1995 include means (±sd) for:

- total alkalinity 29±29 mg/L (n=154);
- pH 6.7fo.7 S.U. (n=648);
- dissolved calcium 7±4 mg/L (n=96);
- chlorophyll-a 7±ll µg/L (n=l62);
- and TOC 8±2 mg/L (n=190) (TNRCC, 1996).

Mean (±sd) surface water quality data for selected parameters from Caddo Lake based on the SWQM database from 1990-95 include:

- total alkalinity 23±19 mg/L (n=12);
- pH 6.9±0.8 S.U. (n=48);
- dissolved calcium 5±l mg/L (n=5);
- chlorophyll-a 15±13 µg/L (n=l0);
- and TOC 8±2 mg/L (n=13) (TNRCC, 1996).

Sample Sites

Sample sites were chosen based on geographic location within the basin (Figure 1) as well as proximity to possible point sources. Lake Bob Sandlin (~6,000 ha) was filled in 1980 and is located on Big Cypress Creek in the upper portion of the basin, adjacent to a large lignite-fired power plant operated by Texas Utilities. Lake O' the Pines (~7,600 ha) was filled in 1959 and is located on Big Cypress Creek in the middle part of the basin. Lone Star Steel operates a secondary steel mill near the upper reaches of Lake O' the Pines. Pruitt Lake (<40 ha) is a natural lake located in the middle basin on Black Cypress Creek. Adjacent bottomland is typically flooded during winter and spring. There are no known point source discharges near Pruitt Lake, which was chosen as a control site. Benten Lake (<40 ha) is a natural lake adjacent to Big Cypress Creek, upstream of Caddo Lake. Benten Lake has only a minor amount of seasonally flooded wetlands.

Caddo Lake is located in the lower portion of the basin below Big Cypress Creek and Little Cypress Bayou. Caddo Lake originally was a natural lake formed in the early 1800s from a distributary channel of the Red River (COE, 1993). The lake was stabilized with a dam in 1914, and enlarged in 1971 to its current size. Big Cypress Creek and the upper end of Caddo Lake are swampy in nature, and the adjacent shorelines are typically flooded during winter and spring. One sample site was located in the Devil's Elbow/Carter Lake area of upper Caddo Lake. Another sample location was at mid-lake near an abandoned pipeline crossing, close to the Texas-Louisiana state line, where a variety of sediment and water samples have been collected for over 20 years by the TNRCC.

Regulated Dischargers

TNRCC-regulated industrial wastewater dischargers in the Cypress basin include the Longhorn Army Ammunition Plant (LAAP), which is located on the southeast shore of Caddo Lake. LAAP does not use mercury in any current processes and laboratory analyses of the permitted outfall show no mercury concentrations above the detection limit (DOA, 1995). Other industrial dischargers in the basin include Texas Utilities' lignite-fired electric power plant on Lake Monticello, Southwestern Power Company's (SWEPCG) coal-fired power plant on Welsh Reservoir, Pilgrim's Pride chicken processing plant located in Segment 0404 of Big Cypress Creek, and the Lone Star Steel company near the upper end of Lake O'the Pines. The following is a list of the larger municipal wastewater treatment plants (WWTPs) in the area, with permitted discharge volumes in million gallons per day (MGD):

- Mount Pleasant (2.53 MGD);
- Pittsburg (0.97 MGD & 0.20 MGD);
- Atlanta (1.1 MGD);
- Daingerfield (0.70 MGD);
- Jefferson (0.55 MGD);
- Hughes Springs (0.49 MGD);
- Naples (0.25 MGD).

Municipalities with discharges >1.0 MGD are required to test for metals during the TNRCC permit renewal process. Mount Pleasant's WWTP renewal application stated that mercury was below the detection limit in samples collected during July 1994 and June 1989. Data self-reported to the TNRCC since 1992 by Lone Star Steel showed mercury concentrations to be less than the detection limit in their discharge. Samples collected during 1995 at Jefferson and Hughes Springs WWTPs also showed mercury levels to be below the detection limit

UPPER SABINE RIVER BASIN

Background Information

The Sabine River has a much larger drainage basin than Cypress Creek, and in some years has the largest flow of any Texas river. The diverse economy in the upper part of the basin is centered around lignite mining, agriculture, manufacturing, and tourism. Sluggish flow characteristics and seasonally low dissolved oxygen are not a concern, at least in the upper portion of the basin. Brandy Branch and Martin Lake reservoirs have fish consumption advisories due to elevated levels of selenium associated with coal-fired power plants.

TNRCC's SWQM database from 1990-95 for the upper Sabine River basin (Segments 0505-0506) include means (± sd) for:

- total alkalinity 50±40 mg/L (n=95);
- pH 7.0±0.5 S.U. (n=164);
- dissolved calcium 20±12 mg/L (n=73);
- chlorophyll-***a*** 11±18 µg/L (n=88);
- TOC 11±3 mg/L (n=54) (TNRCC, 1996).

Sample Sites

Samples sites were chosen based on geographic location within the basin (Figure 2) and proximity to potential point sources. The Sabine River at SH 14 was chosen as an upstream control site. The Sabine River at SH 155 is close to the Chevron pipeline fuel terminal. The Sabine River at IH 20 is located immediately downstream of the City of Longview's WWTP discharge, and near Marathon-LaTourneau, a manufacturer of large earth-moving equipment. The Sabine River at SH 149 is located upstream of the Texas Eastman chemical plant. The Sabine River at the Atchinson Topeka & Santa Fe railroad crossing is located approximately 2 km downstream of the Texas Eastman plant. SWEPCG operates a lignite-fired electric generating plant on Brandy Branch Reservoir.

Regulated Dischargers

TNRCC-regulated industrial dischargers in the upper Sabine basin include Texas Eastman and SWEPCO. Texas Eastman does not use mercury in any process. Larger domestic WWTPs in the upper Sabine River basin and their permitted discharge volumes include:

- Longview (13.9 MGD);
- Kilgore (3.0 MGD);
- Gladewater (1.4 MGD);
- White Oak (1.0 MGD);
- Mineola (0.8 MGD);
- Hawkins (0.25 MGD).

Recent renewal applications for Texas Eastman, Longview, and Kilgore all show discharge concentrations of mercury to be below detection limits.

METHODS

Water

TNRCC routine sampling procedure was changed during 1993 to evaluate total rather than dissolved mercury Since mercury binds with colloidal organic particles, total mercury concentration in water better reflects the amount that is bioavailable. Samples from the Sabine basin were collected before total mercury was routinely analyzed. All samples were collected immediately beneath the surface in polyethylene jars, preserved with metals-free nitric acid, placed on ice, and sent to the TNRCC lab for analysis by cold vapor atomic absorption spectroscopy (CVAA) as detailed in EPA SWA-846 Method 7470.

Sediment

Surface sediment samples were collected with an Ekman dredge. Penetration of the dredge ranged from 50-200 mm The most recently deposited sediment (<50 mm) was collected with a teflon scoop. Multiple grabs (≥3) were taken and cornposited. Samples were placed in nitric-acid-rinsed glass containers that had Teflon-lids, placed on ice, and sent to the TNRCC lab for analysis by CVAA Spectroscopy as detailed in EPA SWA-846 Method 7471.

Soil

Surface soil samples were collected from 12 sites in 10 counties throughout the region. Natural areas such as state parks, national forests, and similar undeveloped areas were chosen. Samples of lignite and coal were collected from surface stock piles at the four fossil-fuel power plants within the region. These samples were placed in nitric-acid-rinsed jars placed on ice, and sent to the TNRCC lab where they were analyzed by CVAA as detailed in EPA SWA-846 Method 7471.

Fish

Fish were collected either by gill netting or boat-mounted electroshocking. Largemouth bass was the target species for the lakes within the Cypress River basin portion of the study area. Since bass are difficult to collect in rivers, channel catfish was the target species at the Sabine River sites. Lack of target species necessitated some substitution in both areas. TNRCC's water quality procedures manual recommends whole-body composites for surveys of ecosystem health, and individual muscle-tissue samples for health risk surveys (TNRCC, 1994). Since this was primarily a survey of ecosystem health, composite samples of three to five similar-sized individuals were utilized when possible. The EPA guidance manual for assessing chemical contamination in fish recommends using composite samples in screening studies and replicate composite samples from three size classes in follow-up surveys where concerns are located (U.S. EPA, 1993). Individual muscle tissue samples of largemouth bass also were collected over a wide size range on Caddo Lake in order to check the variability of mercury concentrations between and within certain size classes. Samples were wrapped in aluminum foil, placed on ice, and sent to the TNRCC lab. In the lab, fish were physically homgenized by two different techniques dependant upon whether muscle tissue (edible portion) or whole fish (the entire body, head, fins, and scales) analysis was requested. In the case of whole fish, specimens were chopped into 5-6 cm sections using a stainless-steel cleaver. These sections were then processed though a standard sausage grinder in three passes until a homogeneous fish paste with discrete particle sizes of 0.5 mm or less was produced. For muscle tissue, a large representative portion of fillet was cutfrom the whole fish. This was blended to a paste with discrete particle sizes of 0.5 mm or less. In each case, fish pastes were analyzed by CVAA according to EPA/6000/4-91/010 Series Method 245.6.

Benthic Macroinvertebrates

Benthic macroinvertebrates were collected with a D-frame net from submerged portions of cypress trees in the upper ends of Lake O'the Pines and Caddo Lake. Specimens were put in clean plastic sandwich bags, placed on ice, and sent to the TNRCC lab. In the lab, whole benthic macroinvertebrates were placed on a polypropylene cutting board and chopped to fine bits, 1 mm wide or finer, using a staiuless-steel cleaver. Bits were then homogenized until a paste with discrete particle sizes of 0.5 mm or less was obtained. The material was then analyzed by CVAA according to EPA/6000/4-91/010 Series Method 245.6.

Tree Cores

Cypress tree cores were collected at Pruitt Lake and upper Caddo Lake. A 10-mm diameter stainless-steel tree auger was placed perpendicular to the tree and manually turned until heartwood was felt. The core was aged with a field magnifying glass. Rings representing 30-year periods were broken off. The rings were then placed in clean glass screwcap vials, packed in ice, and sent to the TNRCC lab. A similar procedure was used to collect cores from cedar trees in Smith and Harrison counties. The largest individual trees in the area (minimum diameter of 0.5 m) were chosen. At the TNRCC lab, each section was halved along it's longitudinal axis to provide replicate samples of each core. The whole length of a longitudiual cut was required to be digested for each discrete aliquot to yield a representative sample. Core section masses were greater than typically analyzed. Consequently a five-fold scale up of reagent quantities from EPA/6000/4-9 l/010 Series Method 245.6 was required for preparation.

Sphagnum Bog Cores

Core samples were collected at Chester's bog on the Engling Wildlife Management Area in Anderson County. There is a small stream discharging from the downgradient end of the bog. Therefore, this bog is considered non-umbatropbic. A thick-walled PVC pipe was hammered perpendicular into the bog until resistance was felt a depth. The pipe was then twisted out and the core remained in the PVC pipe for shipment to the lab where the peat core section was halved along its longitudinal axis to provide replicate samples. Core section masses were of greater quantity than typically analyzed, and a similar five-fold scale up of reagent quantities was utilized in preparing bog core samples for CVAA analysis.

Data Analysis

Data anaylsis of muscle tissue from fish was complicated by the fact that some sites had composite samples within a size group and other sites had individual fish within those groups. The lack of similar data sets between sites did not allow for statistical comparisons. The only statistical analysis conducted was of largemouth bass size versus mercury concentration in muscle tissue from Lake. Small sample sizes did not allow for statistical analysis of sediment, water, or tree core data.

RESULTS AND DISCUSSION

Total Mercury in Water

Dissolved and total mercury in water concentrations were highly variable (Table 1). Values were less than Texas surface water quality criteria for protection of aquatic life, except for two from Pruitt Lake and upper Caddo Lake during June 1994. Both of these values exceeded the freshwater acute criterion (2.4 µ/L); and both were greater than the maximum concentrations observed by the Corps of Engineers during the 1991 survey on Caddo Lake. Limited number of samples made interpretation difficult. Elevated values may have reflected seasonality in mercury partitioning between sediment and water.

Mercury in Sediment

Sediment mercury values were variable (Table 2). Concentrations were:

- Caddo Lake <0.01-1.57 mg/kg;
- Lake O'the Pines 0.42-0.88 mg/kg.

Maximum concentrations were greater than those observed by the Corps of Engineers during the 1991 survey of Caddo Lake and Lake O'the Pines.

Upland Soil

Mercury concentrations in upland soil from 12 locations throughout east Texas included:

- Blanton Creek road, Red River County, 0.01 mg/kg;
- Davy Crockett national forest, Houston County, 0.02 mg/kg;
- Lake Fork reservoir, Wood County, 0.03 mg/kg;
- Atlanta state park, Cass County, 0.03 mg/kg;
- Black Bayou at CR4659, Cass County, 0.03 mg/kg;
- Tridens Prairie, Lamar County, 0.03 mg/kg;
- Tyler state park, Smith County, 0.04 mg/kg;
- Daingerfield state park, Morris County, 0.04 mg/kg;
- Engling wildlife management area, Anderson County, 0.05 mg/kg;
- East Fork Angelina River, Rusk County, 0.05 mg/kg;
- Pruitt Lake 0.09-0.11 mg/kg;
- Caddo Lake state park, Harrison County, 0.14 mg/kg.

Mercury in Coal

Mercury concentrations in fossil-fuel stockpiles near cooling reservoirs located at the region's four power plants (and the type of fuel) are:

- Lake Welsh (bituminous coal from Wyoming's Powder River basin) 0.03 mg/kg;
- Brandy Branch Reservoir (lignite) 0.18 mg/kg;
- Martin Lake (lignite) 0.30 mg/kg;
- Lake Monticello (lignite) 0.59 mg/kg.

Benthic Macroinvertebrates

The benthic macroinvertebrate community was dominated by crayfish, dragonflies (Macromiidae), and damselflies (Lestidae). Mercury concentrations were:

- Pruitt Lake 0.18 mg/kg;
- Lake O'the Pines <0.06 mg/kg;
- upper Caddo Lake 0.16 mg/kg.

Fish

Mercury concentrations in whole-body, composite bluegill ***(Lepomis mucrochirus)*** samples were:

- Lake Bob Sandlin 0.08 mg/kg (n=5);
- mid and upper Caddo Lake 0.29 mg/kg (n=5).

The target species for the Cypress basin sites was largemouth bass. Mercury in muscle tissue ranged from 0.03-1.01 mg/kg. In general, larger bass had higher mercury concentrations. Mercury concentrations in fish generally were lower from Lake Bob Sandlin and Lake O'the Pines, than from Caddo Lake (Table 3), although whether the difference was statistically significant was unclear.

Largemouth bass were difficult to collect from Pruitt Lake, so spotted bass ***(Micropterus punctulatus)*** were substituted. Spotted bass had mercury concentrations in muscle tissue of:

- 0.03 mg/kg (313 mm);
- 0.69 mg/kg (289 mm);
- 1.07 mg/kg (295 mm).

Based on age determinations from otoliths, spotted bass from Pruitt Lake and individual largemouth bass (350-450 mm) from Caddo Lake were of similar age (>=4 years old). It is possible that mercury may correlate more closely with age than with size of predatory fish. In general, it was observed that in several area lakes larger bass had higher concentrations of mercury in their muscle tissue than did smaller individuals from the same lakes (Table 3).

For largemouth bass from Caddo Lake, there was a highly significant difference in mercury concentrations between size groupings ($P<0.01$) (Table 4). Older and larger fish have higher mercury concentrations. Variability of mercury concentrations, as measured by the coefficent of variation (CV), was greatest in the <350 mm size class (CV 86%); followed by the 350-450 mm size class (CV 16%); then by the 450 mm class (CV 7%). As graphed, size of fish versus mercury concentration showed a strong positive relationship (r^2=0.71) (Figure 3).

Mercury concentrations in different tissues were compared from three individual largemouth bass from Caddo Lake. Highest concentrations were in muscle, followed by liver, then heart, in each individual.

The target species for the Sabine River sites (channel catfish) could not be collected from each site. Whole fish samples from various species from six sites have mercury concentrations from 0.12-0.23 mg/kg (Table 5). Concentrations were similar to those collected by ANSP in 1995.

Tree Ring Cores

Recent versus old annular rings from six cypress trees cores in Cass County (Pruitt Lake) and Harrison County Caddo Lake) did not show observable differences in mercury concentrations from approximately 0-30 years before present (1995-1965), to >60 years before present (4935) (Table 6). Annular rings from four cedar trees in Harrison and Smith counties were analyzed for mercury corresponding to 30-year intervals (Table 6). Mercury concentrations were approximately 10 times higher in bald cypress than in eastern red cedar. Concentrations appeared to be higher for the years 1935 - 1965 than for other time periods in cedar tree samples from Harrison County.

Sphagnum Bog Cores

Replicate samples of mercury concentrations from bog cores from the Engling Wildlife Management Area in Anderson County did not meet TNRCC'S laboratory quality control requirements. Therefore, the data are not presented in this report.

Spanish Moss

Spanish moss collected from upper Caddo Lake contained mercury at <0.20 mg/kg.

CONCLUSIONS

Based on cumulative results, it is concluded that mercury is present in the aquatic food web throughout the Cypress and upper Sabine basins, and throughout northeast Texas. At every sample location, a majority of the fish collected had detectable mercury concentrations in muscle tissue. Six of 22 individual largemouth bass muscle samples from the Cypress Basin exceeded the U.S. EPA screening level of 0.6 mg/kg; and two of 22 individual samples were at the FDA action level of 1.0 mg/kg. One of the control sites, Pruitt Lake, had some of the highest mercury concentrations in fish tissue.

Mercury concentrations vary in different species. The likelihood of finding higher mercury concentrations increases with increased position in the food web. Top predators typically contain the highest mercury concentrations, while older and larger individuals of a particular species generally have higher mercury concentrations than younger, smaller individuals.

No point source discharges of mercury to surface waters were identified from regulatory information reviewed, or from data collected. No naturally occurring geologic formations in the area were identified that contain elevated mercury concentrations. Therefore, a nonpoint source of mercury input to the environment is suspected.

None of the data collected suggest any local atmospheric source of mercury. Therefore, it is theorized that mercury is being deposited from global or regional atmospheric sources evenly over the entire area, and is being incorporated into the food web to a greater degree in certain areas where natural conditions are conducive to mercury methylation.

It is difficult to say if atmospheric mercury levels have changed based on limited tree core data.

RECOMMENDATIONS

Additional sampling should be performed in geographical areas of the state that support conditions favorable for the biological uptake of mercury.

It is difficult to collect enough individuals of the same species in a survey that covers large geographical areas and different environments, such as lakes and rivers. However, a single target species should be selected and every effort made to utilize it exclusively. Largemouth bass seem to concentrate mercury in muscle tissue to a greater extent than other sport fishes, and most top predators. Largemouth bass are present in most water bodies, although they are sometimes uncommon in rivers. Nightime electroshocking is recommended to increase the probability of collecting enough individuals to allow statistical comparisons between sites. Largemouth bass size affects mercury concentration in muscle tissue. Future studies should attempt to collect enough individuals over a variety of size ranges to allow statistical comparisons. Size ranges that correspond to common regulatory limits derived by the Texas Parks and Wildlife Department are suggested.

Further seasonal sampling of sediment mercury and total mercury in water is also recommended. AVS and simultaneously-extracted metals should be included in all sediment analysis. Seasonally flooded wetlands may have a role in mercury biomagnification in the food web, and future studies should target this habitat type. There are several oxbow lakes in East Texas that would make good sample locations.

Air-monitoring stations should be established in several regions across the state. Data is needed to determine if global or regional atmospheric mercury is a source of loading to the aquatic environment.

Additional tree core samples are needed to suppliment the current data set. Cores from trees near and remote from potential atmospheric point sources, and near air-monitoring sites, should be collected. Ten-year tree ring increments are suggested for analysis.

Mercury concentrations in fish-eating birds, reptiles, and mammals also should be investigated.

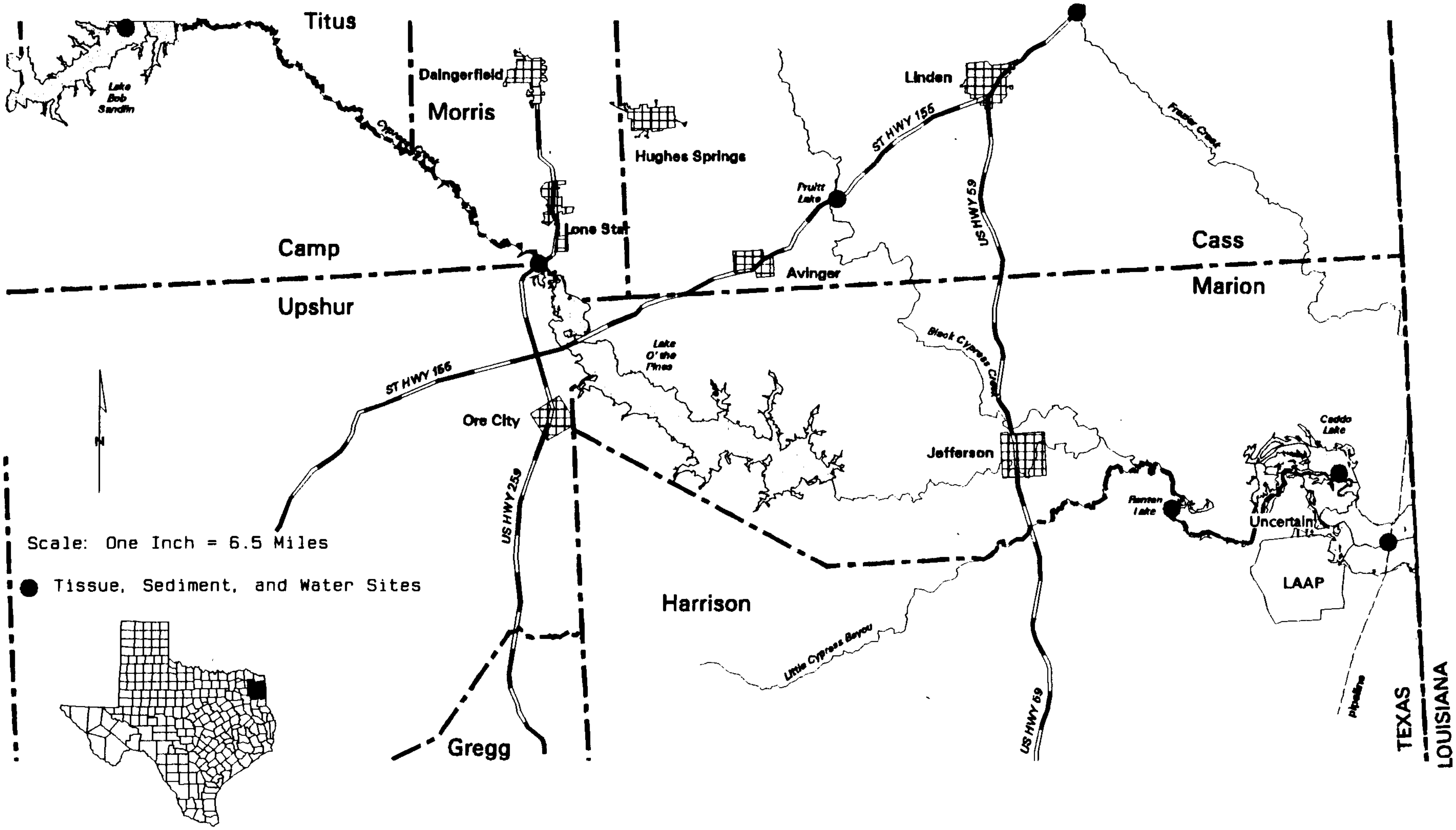

Figure 1. Sample Sites, Cypress River Basin, 1994-95.

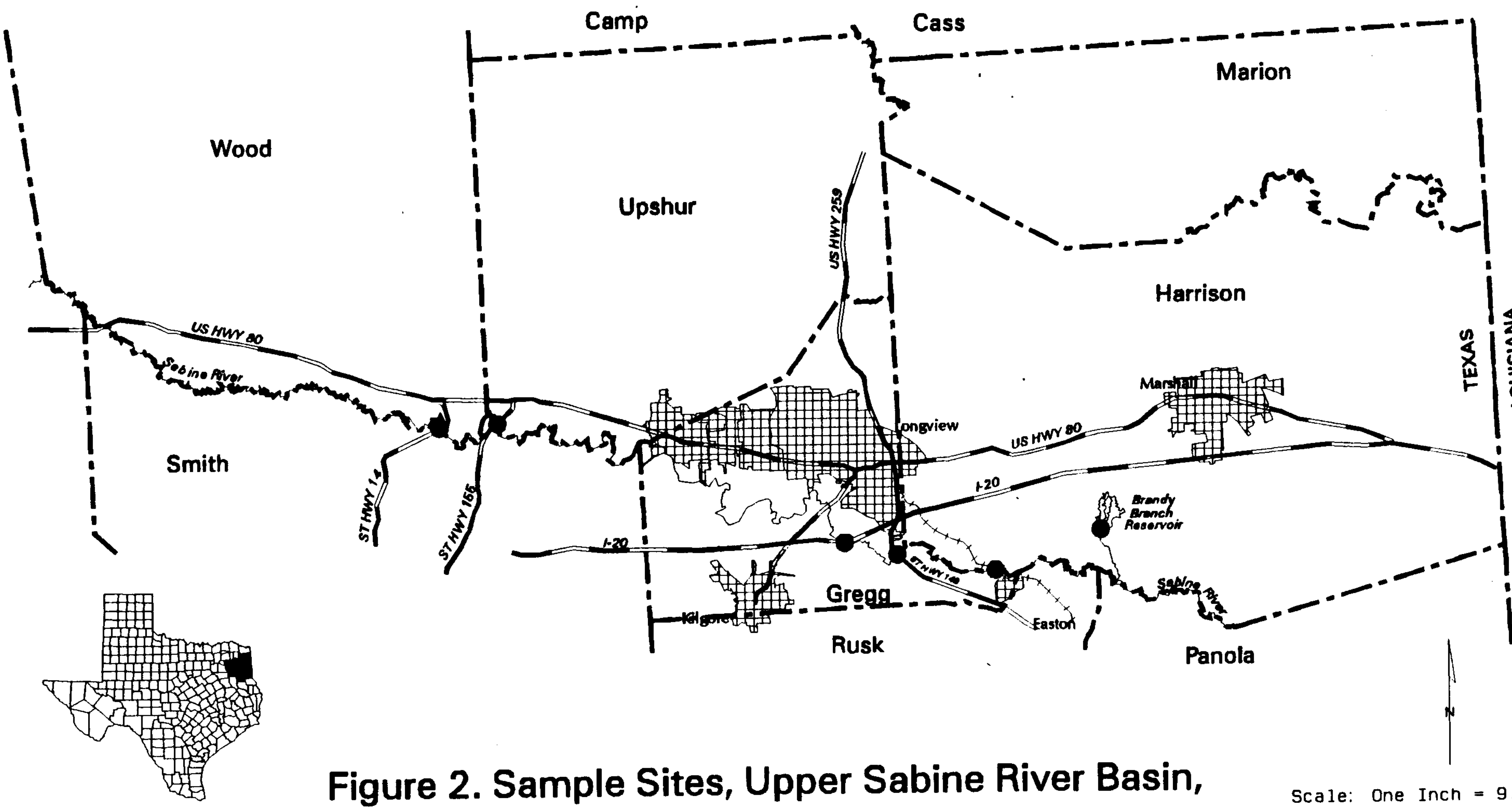

Figure 2. Sample Sites, Upper Sabine River Basin, August and September, 1993.

Scale: One Inch = 9.4 Miles

● Tissue, Sediment, and Water Site

Figure 3. Mercury (mg/kg) vs size (mm)

r-square = 0.71

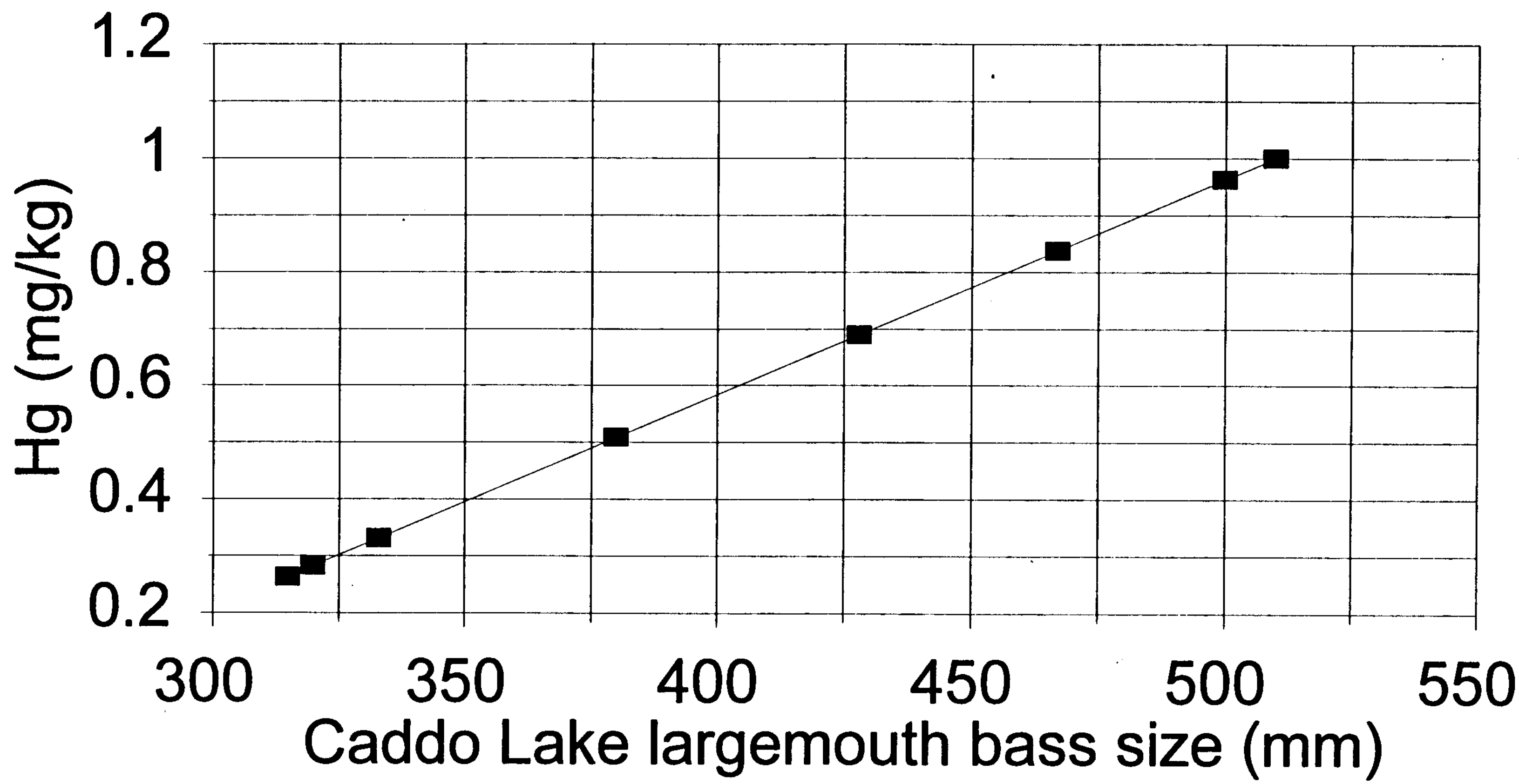

Table 1. Mercury (Hg) concentration in water, Cypress River basin (May-October 1994) and Sabine River basin (September 1993).

Location	Dissolved Hg (μg/L)	Total Hg (μg/L)
Upper Lake O'the Pines		.05-0.78
Mid Lake O'the Pines		0.05
Lower Lake O'the Pines		0.28
Pruitt Lake at SH 155		3.04
Upper Caddo Lake		<0.12-3.04
Mid Caddo Lake		<0.12
Sabine River at SH 14	0.83	
Sabine River at SH 155	0.74	
Sabine River at IH 20	0.83	
Sabine River at SH 149	<0.20	
Sabine River at ATSF R/R	<0.56	

Table 2. Mercury (Hg) concentrations in sediment, Cypress River basin (May-October 1994) and Sabine River basin (September 1993).

Location	Sediment Hg (mg/kg dry weight)
Lake Bob Sandlin at Monticello dam	<0.06
Upper Lake O'the Pines	0.42-0.88
Mid Lake O'the Pines	0.45
Pruitt Lake at SH 155	0.25
Upper Caddo Lake	<0.01
Mid Caddo Lake	<0.01-1.57
Sabine River at SH 155	0.15
Sabine River at SH 149	0.08

Table 3. Largemouth bass muscle mercury (Hg) concentrations, Cypress River basin, 1994-95.

Location	Size (mm)		Hg (mg/kg)
Lake Bob Sandlin	**<350**	314-340(n=5)	0.22
Lake O'the Pines		277	<DL
		320	<DL
Pruitt Lake		285	0.52
Benten Lake		307	0.03
		308	0.04
		343	0.07
upper Caddo Lake		315	0.23
		320	0.12
		333	0.68
Lake Bob Sandlin	**350-450**	372-417(n=5)	0.30
Lake O' the Pines		435	0.04
		377	0.39
upper Caddo Lake		380	0.46
		380	0.41
		428	0.56
Lake Bob Sandlin	**>450**	480-510(n=5)	0.40
Lake O' the Pines		460	0.47
upper Caddo Lake		467	0.99
		500	0.93
		510	1.01
		560	0.86

Table 4. Analysis of variance for difference in mercury concentrations between largemouth bass from three size ranges (<350; 350-450; >450 mm) from upper Caddo Lake during 1994.

	Degrees of Freedom (df)	Sum Squares	Mean Square	F
Treatment	2	0.72	0.36	12.0**
Error	7	0.20	0.03	
Total	9	0.92		

Table 5. Mercury (Hg) concentrations in whole fish, upper Sabine River basin, 1993.

Location	Species	Size (mm)	Hg (mg/kg)
Sabine River at SH 14	channel catfish	380-460	0.20
Sabine River at SH 155	blue catfish	405	0.21
Sabine River at IH 20	smallmouth buffalo	380	0.23
Sabine River at SH 149	channel catfish	unknown	0.13
Sabine River at ATSF R/R	channel catfish	unknown	0.12
	blue catfish	410	0.13
Brandy Branch Reservoir	largemouth bass	278-380	0.12

Table 6. Mercury (Hg) concentrations in annular rings from trees in northeast Texas.

Location Tree	Hg (mg/kg) Present-1965	Hg (mg/kg) 1965-1935	Hg (mg/kg) <1935
Cass County bald cypress	0.13 0.15 0.11	No Data (ND) ND ND	0.14 0.14 0.11
Harrison County bald cypress	0.10 0.05 0.11	ND ND ND	0.06 0.04 0.18
Harrison County red cedar	0.02 0.02	0.07 0.27	ND 0.01
Smith County red cedar	0.01 0.01	0.01 0.01	0.01 0.01

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