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May 2000
AS-180

Bioaccumulation of Mercury in the Selected East Texas Water Bodies

Monitoring Operations Division

TEXAS NATURAL RESOURCE CONSERVATION COMMISSION

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Bioaccumulation of Mercury in Selected East Texas Water Bodies

Prepared by

Stephen Twidwell

Surface Water Quality Monitoring Team
Laboratory and Mobile Monitoring Section
Monitoring Operations Division

AS-180
May 2000

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ABSTRACT

Largemouth bass *Micropterus salmoides* were collected from 11 reservoirs and two natural lakes in East Texas to determine relationships between mercury concentration in fish and physicochemical variables in water and sediment. Diverse water bodies, widely distributed spatially over the East Texas region, with a broad range in size (156-13,660 hectares), pH (4.7-8.7 std. units), and alkalinity (< 5-78 mg/L) were sampled. Total mercury concentrations in edible largemouth bass muscle tissue ranged from 0.04 to 2.1 mg/kg and were positively correlated with fish size. Nearly all largemouth bass from Pruitt Lake and Kimball Reservoir had mercury concentrations in tissue that exceeded the 0.7 mg/kg screening level. Mean and median mercury concentrations in largemouth bass tissue were significantly different among the study water bodies. Mean mercury tissue concentrations from Kimball Reservoir and Pruitt Lake exceeded the 0.7 mg/kg screening level and were significantly different from one another and the remaining water bodies. The Texas Department of Health (TDH) conducted follow-up sampling of the two water bodies and issued consumption advisories in April 1999 due to consistently elevated mercury concentrations in fish tissue. Chemical characteristics of the water bodies strongly influenced the bioaccumulation of mercury in largemouth bass. Although sampling of other species was limited, spotted bass *Micropterus punctulatus* and freshwater drum *Aplodinotus grunniens* also bioaccumulated mercury to elevated concentrations in edible muscle tissue, while channel catfish *Ictalurus punctatus*, white crappie *Pomoxis annularis*, black crappie *Pomoxis nigromaculatus*, and bowfin *Amia calva* had lower mercury concentrations. Mercury concentrations in largemouth bass, standardized to length (406.4 mm; 16 in) for comparisons ranged from 0.09 to 1.22 mg/kg and were positively correlated with total organic carbon in water and negatively correlated with pH, hardness, calcium, total dissolved solids, sulfate, magnesium in water, and manganese in sediment. No significant correlations were observed between standardized mercury concentrations in fish and mercury concentrations in water or sediment. These relationships suggest that mercury concentrations in water and sediment are not nearly as important as are in-reservoir and lake processes which control the production of methylmercury and its subsequent bioconcentration and bioaccumulation in largemouth bass in East Texas reservoirs and lakes. Linear regression revealed that pH accounted for 51 percent of the variation in mercury concentrations among standardized fish. Multiple regression revealed that pH and total organic carbon in water accounted for 61 percent of the variation in standardized fish mercury concentrations. Mercury concentrations in fish were significantly higher in reservoirs and lakes with calcium less than 10 mg/L, hardness less than 40 mg/L, or sulfate less than 10 mg/L. Mercury concentration-depth profiles in sediment cores revealed that mercury concentrations in surficial sections were elevated above those in deeper strata in nearly every case. Statistically significant trends for decreasing mercury concentrations with increasing depth were found for six reservoirs and one lake. These trends suggest that recent mercury inputs may be continuing to increase across the East Texas region. Mercury concentration-age profiles in tree cores from ten sites near the study reservoirs and lakes were generally inconclusive and failed to corroborate trends observed in sediment cores.

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REVIEW OF PREVIOUS MERCURY STUDIES

Mercury (Hg) has long been recognized as one of the more toxic metals, but only since about 1950 has it been identified as a serious pollutant in freshwater and marine aquatic environments. Mercury can exist in a number of different forms in the environment: Hg(0) (elemental, metallic, or gaseous), Hg_2(I)(mercurous), and Hg(II) (mercuric). Hg(0) is the only metal that is a shiny, silver-white, odorless liquid at room temperature. Some evaporation of metallic mercury occurs at room temperature and above to form mercury vapor, a colorless, odorless gas that is nearly insoluble (60 μg/L) in water. Mercury can combine with other elements such as chlorine, oxygen, or sulfur to form numerous inorganic and organic compounds (USEPA, 1980). Mercurous mercury is rarely stable under normal environmental conditions. Hg(0) can hydrolyze to form mercuric dihydroxide [$Hg(OH)_2$] . Hg(II) can hydrolyze to form the species mercuric hydroxide ion [$Hg(OH)^+$], $Hg(OH)_2$, and mercuric trihydroxide ion [$Hg(OH)_3^-$] (Rubin, 1976). The relative amounts of each species are dependent on pH. Except in the atmosphere, most of the mercury encountered in water, soils, and sediment is in the form of inorganic mercuric salts and organomercurics. Examples of Hg complexes include $Hg(OH)_2$ under alkaline conditions and $HgCl_2$ under acidic conditions. In alkaline sediments under moderately reducing conditions, dimethyl mercury (CH_3HgCH_3), which is insoluble and highly volatile, can form. At neutral and lower pH values, monomethyl mercury (CH_3Hg) can form. Under strongly reducing conditions (e.g., in the presence of hydrogen sulfide), mercury can precipitate as mercuric sulfide (HgS), which is insoluble (Nriagu, 1979).

Most commonly encountered mercury compounds in the environment include mercuric chloride ($HgCl_2$), $Hg(OH)_2$, HgS, mercuric hydroxide chloride (HgOHCl), methylmercury chloride (CH_3HgCl), and methylmercury hydroxide (CH_3HgOH). Other organic mercurics, such as CH_3HgCH_3 and phenylmercury (C_6H_5Hg), are generally encountered in smaller fractions (USEPA, 1997). Most inorganic mercury compounds are white powders or crystals, except for HgS (known as cinnabar), which is naturally red and turns black after exposure to light.

Liquid [Hg(0)] is heavier than water, and at one time it was thought that the metal would quickly precipitate to the bottom sediment and remain there in an innocuous state. However, based on stability constants and the solubility of liquid mercury in freshwater, liquid mercury would dissolve and form $Hg(OH)_2$ and HgOHCl without precipitating to the bottom sediment. In water, mercury has a strong affinity to particulates which tend to remove it from the water column to the sediment. Hg(0) can be oxidized in sediment to inorganic, divalent mercury [Hg(II)](Wood, 1974). Furthermore, bacteria have been found capable of transforming (methylating) Hg(II) to an organic form (methylmercury; CH_3Hg; MeHg) in freshwater sediments (Gilmour et al., 1992) and marine sediments (Compeau and Bartha, 1985). Compared to Hg(0) or Hg(II), MeHg is more soluble in water, more biologically reactive, and more toxic to aquatic life and wildlife. Largely due to methylation, mercury is considered a much more serious threat to the aquatic environment than was originally thought.

During the past decade, new trends about mercury contamination have emerged. Investigations during the late 1980s in the northern states and Canada found that fish, often in remote areas, commonly have elevated levels of mercury (Wiener, 1987). More recent studies in the southern region of the U.S. have shown widespread mercury contamination of fish from streams, lakes,

reservoirs, wetlands, estuaries, and the Gulf of Mexico. Human exposure to MeHg comes almost exclusively from consumption of fish and fish products (Fitzgerald and Clarkson, 1991). The increased trend in contamination is evident in the number of mercury consumption advisories that have been issued by 40 states, including Texas (USEPA, 1997).

The Global Mercury Cycle

Movement of mercury into the atmosphere and its subsequent deposition in the global biogeochemical cycle may be described in a variety of mass balance formulations (Figure 1)(Mason et al., 1994). Present estimates indicate the global atmosphere contains approximately 5 x 10^6 kg of mercury, with 98 percent existing in the gaseous elemental state [Hg(0)] and about two percent in the particulate inorganic divalent state [Hg(II)] (Mason, et.al., 1994). Human-related (anthropogenic) emissions (4 x 10^6 kg/y) in point sources alone rival or exceed natural inputs (3 x 10^6 kg/y). The total influence of human activities extends beyond direct mercury emissions, since portions of terrestrial and aquatic emissions include a "recycled" anthropogenic component.

Anthropogenic sources account for about 4,000 x 10^3 kg/y of estimated annual global point source emissions (Figure 1). About one-half of anthropogenic-related mercury emissions is produced and deposited on a local/regional scale, while the other one-half contributes to the global cycle. The particle-bound Hg(II) is deposited locally and may establish regional concentration gradients in nearby soil (Nater and Grigal, 1992). In the atmosphere, mercury exists almost entirely in the relatively insoluble gaseous Hg(0) state, which can be transported for long distances from the source and remain for a year or more (Fitzgerald and Mason, 1997; Hall, 1995). This is sufficient time for atmospheric mercury to be distributed globally before returning to land, reservoirs, lakes, oceans, and ice. As a result, while the principal emissions of mercury are from point sources concentrated in industrialized areas near population centers, mercury pollution is truly global. Historical records from dated lake sediment cores provide compelling evidence that remote areas receive significant inputs of anthropogenic mercury by long-range atmospheric transport (Mason et al., 1994). Atmospheric transport and deposition has also been shown to be the main source of mercury in fishes from remote, pristine lakes with no known direct local source of mercury (Hakanson et al., 1988; Wiener et al., 1990; Glass et al., 1991; Linberg et al., 1991).

Mercury was used extensively in industrial applications from 1900 to 1970 in processes for printing, pharmaceutical preparations, paint and varnish additives (antifouling and mildew proofing), explosives, electric light bulbs, batteries, dental supplies, pesticides, and speciality cleaning products (USEPA, 1997). All uses of mercury as a bactericide or fungicide were banned after August 1978, except for treatment of textiles for outdoor use, control of brown mold on new lumber, and as an in-can preservative in water-based paints. Although substantial reductions in industrial uses have occurred in recent years, mercury is still utilized in the production of chlorine, caustic soda, paints, batteries, thermometers/barometers/ manometers, and electric equipment (switches, lamps, arc-rectifiers). Mercury is a by-product of recycling paper, batteries, paints, and rubber and is a component in household wastes (Clement and Kagel, 1990).

Anthropogenic sources of mercury emissions come primarily from industrial fuel combustion, waste incineration, industrial processes (chlor-alkali and pulp industries), metal production, and refining

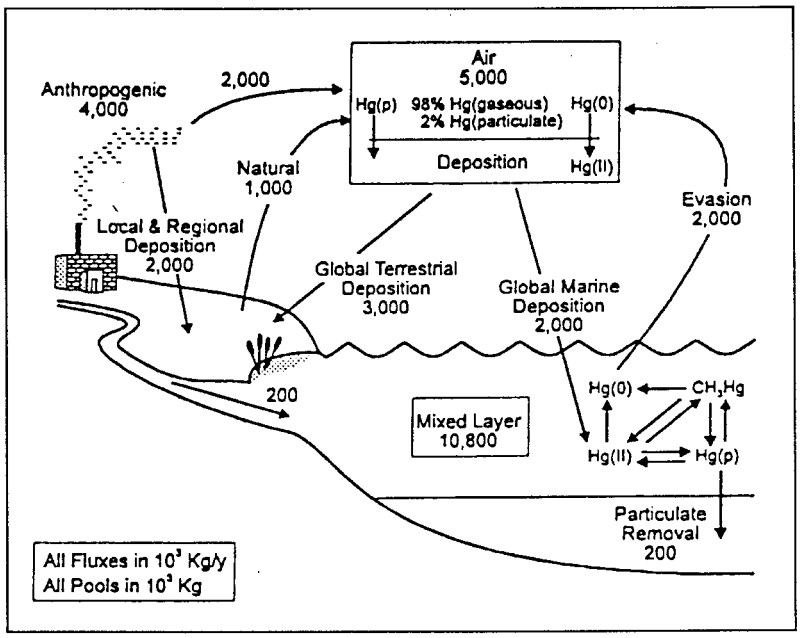


Figure 1. Global Mercury Cycle with Estimated Budgets and Fluxes (Source: Mason, et.al., 1994)

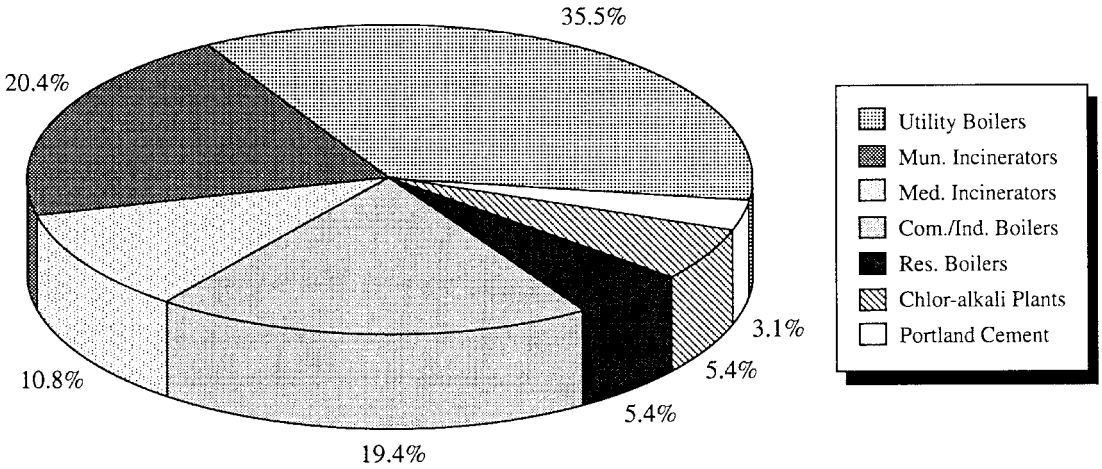


Figure 2. Percent Contributions of Major Mercury Anthropogenic Emissions Sources (Source: USEPA, 1997)

Of the estimated 144 Megagrams (Mg) (158 tons) of mercury emitted annually into the atmosphere by anthropogenic sources in the United States, approximately 87 percent is from combustion point sources, 10 percent is from manufacturing points sources, 2 percent is from area sources, and 1 percent is from miscellaneous sources (Table 1). Seven specific source categories account for nearly all of the total anthropogenic emissions (USEPA, 1997)(Figure 2).

About one-third of current annual global mercury emissions are thought to cycle from the oceans to the atmosphere and back again to the oceans (Figure 1). Emissions of mercury from terrestrial sources, which include volatilization from vegetation, degassing from geologic materials, particulate and vapor emissions from volcanic and geothermal activity, wind-blown particulate matter, forest fires, and agricultural burning account for about 10 percent of current annual emissions (Figure 1). A large proportion of the oceanic and terrestrial natural emission estimates consist of recycled mercury from previously deposited anthropogenic and, to a lesser extent, natural emissions (Fitzgerald, 1995).

In the atmosphere, approximately 98 percent of total mercury is in the elemental gaseous state [Hg(0)], where it is slowly oxidized to the divalent mercuric state [Hg(II)]. Most of this oxidation occurs in fog and cloud droplets, with ozone thought to be the main oxidant in the process (Munthe, 1992). The return of mercury from the atmosphere to the earth's surface occurs chiefly through wet precipitation of dissolved Hg(II) (Watras et al., 1995). Adsorption of mercury on aerosols such as soot, which are subsequently dry-deposited, is another depositing mechanism, especially over land (Mason et al., 1994).

Once oxidized, atmospheric mercury is deposited to land ($3{,}000 \times 10^3$ kg/y) and water ($2{,}000 \times 10^3$ kg/y). The greater proportion of mercury deposited to land probably reflects the proximity of sources of mercury to land, since water precipitation is three times less on land than on the oceans (Morel et al., 1998)(Figure 1). In oceanic waters, most of the Hg(II) is reduced to Hg(0) and returned to the atmosphere, with only a small fraction permanently exported to the sediment (Mason et al., 1994). In reservoirs and lakes, mercury is transferred from the water column by sedimentation and volatilization. Similar processes occur on land, resulting apparently in a smaller return of reduced mercury ($1{,}000 \times 10^3$ kg/y) to the atmosphere, since the terrestrial environment becomes a principal sink. As much as 80 percent of mercury deposited on land is retained in watersheds where it is mobilized slowly to enter fresh and coastal waters (Lindqvist and Rodhe, 1991). Recent estimates indicate that of the approximately 181,436 metric tons of mercury emitted to the atmosphere since 1890, about 95 percent resides in terrestrial soils, about three percent in surface waters, and two percent in the atmosphere (EPRI, 1994). Thus, the effects of anthropogenic mercury loadings may persist and affect aquatic biota for long periods after reduction or cessation of mercury emissions.

The Role of Acid Rain

Increased levels of mercury in fish from acidified lakes, but not in other lakes from the same region that receive of similar mercury loads from atmospheric deposition indicates that increased mercury deposition cannot be the entire explanation for elevated levels in fish (Gilmour and Henry, 1991). Inorganic mercury entering water bodies must be converted to MeHg prior to accumulation. For this reason, elevated MeHg levels in fish from acid-impacted lakes suggest a link between the process of MeHg production or bioaccumulation and acid deposition.

Table 1

Point Source Estimates of National Mercury Emission Rates by Category
1994-1995

Sources of Mercury	1994-1995 Mg/yr[a]	1994-1995 tons/yr[a]	% of Total Inventory[a]
Area Sources	**3.1**	**3.4**	**2.2**
Lamp breakage	1.4	1.5	1.0
General laboratory use	1.0	1.1	0.7
Dental preparations	0.6	0.7	0.4
Landfills	0.07	0.08	0.1
Mobile sources	b	b	b
Paint sources	b	b	b
Agricultural burning	b	b	b
Point Source	**141.0**	**154/7**	**98.7**
Combustion Sources	**125.3**	**137.7**	**86.9**
Utility boilers	47.2	51.8	32.8
Coal	(47)[c]	(51.6)	(32.6)
Oil	(0.02)	(0.2)	(0.1)
Natural gas	(<0.1)	(<0.1)	(0.0)
Municipal waste combustors[g]	26.9	29.6	18.7
Commercial /industrial boilers	25.8	28.4	17.9
Coal	(18.8)	(20.7)	(13.1)
Oil	(7.0)	(7.7)	(4.9)
Municipal waste incinerators	14.6	16.0	10.1
Hazardous waste combustors[d]	6.4	7.1	4.4
Residential boilers	3.3	3.6	2.3
Oil	(2.9)	(3.2)	(2.0)
Coal	(0.04)	(0.5)	(0.3)
Sewage sludge incinerators	0.9	1.0	0.6
Wood fired boilers[e]	0.2	0.2	0.1
Crematories	<0.1	<0.1	0.0
Manufacturing Sources	**14.4**	**15.6**	**10.0**
Chlor-alkali	6.5	7.1	4.5
Portland cement	4.4	4.8	3.1
Pulp and paper manufacturing	1.7	1.9	1.2
Instruments manufacturing	0.5	0.5	0.3
Secondary Hg production	0.4	0.4	0.3
Electrical apparatus	0.3	0.3	0.2
Carbon black	0.3	0.3	0.2
Lime manufacturing	0.1	0.1	0.1
Primary lead production	0.1	0.1	0.1
Primary copper production	<0.1	<0.1	0.0
Fluorrescent lamp recycling	<0.1	<0.1	0.0
Batteries	<0.1	<0.1	0.0
Primary mercury production	b	b	b
Mercury compounds	b	b	b
Byproduct coke	b	b	b
Refineries	b	b	b
Miscellaneous Sources	**1.3**	**1.4**	**0.9**
Geothermal power	1.3	1.4	0.9
Turf products	f	f	f
Pigments, oils, etc.	f	f	f
Total	**144**	**158**	**100**

[a]Numbers do not add exactly due to rounding; [b]Insufficient information to estimate 1994-95 emissions; [c]Parentheses denote subtotal within larger point source category; [d]For the purpose of this inventory, cement kilns, that burn hazardous waste for fuel are counted as hazardous waste combustors; [e]Includes boilers only; does not include residential wood combustion(wood stoves); [f]Mercury has been phased out of use; [g]EPA has finalized emissions guidelines for these source categories which will reduce mercury emissions by at least 90 percent over 1995 levels.

Buffering capacity and acid rain (acid deposition) are two factors which largely control vulnerability of water bodies for mercury bioaccumulation. The carbonate system, which is composed of carbon dioxide , carbonic acid, and carbonate ions, largely controls buffering in natural waters. This system neutralizes acids and bases so as to reduce the fluctuations in pH. In the southern U.S. and east Texas, many water bodies typically have low alkalinity (< 50 mg/L as $CaCO_3$) making them susceptible to low pH. Acidic precipitation resulting from industrial pollution can reduce the pH of poorly buffered waters significantly (Likens, et al., 1972). The solubility and toxicity of most metals increase as pH is lowered.

When rain reacts with sulfur and nitrogen oxides emitted into the atmosphere from pollution sources acidic rain is created. The entire eastern portion of the U.S. has been shown to have acid rain (NADP, 1998). A pH of 5.6 is considered normal for rainwater. Within Texas, weekly rainfall data from 11 sites across the state indicates acidity increases from west to east, with the lowest average acidity (pH = 5.98) at a site in Big Bend and the highest average acidity at Longview (pH = 4.81) (Gise, et al., 1993). This trend reflects the decreasing prevalence of acid-neutralizing materials, principally $CaCO_3$ in Texas soils from west to east across the state. Also important is the increasing amount and density of emission (nitrogen and sulfur oxides) sources in the eastern half of the state. The combination of long-term acid deposition and low water body buffering capacity (resulting from low carbonate levels) results in an acidic condition, making some water bodies in east Texas more vulnerable to mercury bioaccumulation.

Methylation of Mercury

Regardless of the source, once in water mercury has a strong affinity for particulates and organic matter and is mostly found in reservoir and lake bottom sediment (Sorensen et al., 1990; Gilmour et al., 1992; and Zillioux et al., 1993). Sorption of mercury onto particles is an important mechanism for removal of mercury from the water column (Bodek et al., 1988). Particles that act as sorbents for mercury ions include clay mineral, clay-sized grains, hydrous iron and manganese oxide grain coatings, and organic detritus (Perwak et al., 1980).

Methylation of mercury involves the transfer of a methyl group from an organic compound to the metal ion and is catalyzed by microorganisms (Morel et al., 1998). Sulfate-reducing bacteria have been shown to be important mediators of mercury methylation in anoxic marine and freshwater sediments and water (Compeau and Bartha, 1985; Gilmour et al., 1992). Methylation of mercury is enzymatically mediated by sulfur-reducing bacteria in the presence of methylcobalamin (vitamin B12)(Choi et al., 1994). Fermentation during anaerobic conditions produces the cobalamin necessary for methylation (Zillioux et al., 1993). While most MeHg is produced biologically by bacteria, abiotic processes (e.g., humic and fulvic acids in solution) appear to methylate the mercuric ion; however, contributions are generally considered minor (Nagase et al., 1982).

Methylation has been detected in most compartments of the aquatic freshwater environment including aerobic and anaerobic sediments, water, and epifauna (Gilmour and Henry, 1991). Methylation occurs near the sediment-water interface and in shallow sediments with rates in sediment much greater than in the water column (Gilmour et al., 1992). Aerobic conditions favor sediment uptake of both Hg(II) and MeHg, whereas anoxic conditions favor their release (Regnell, 1994; Regnell et al., 1996). Methylation in sediments is important because it often results in

remobilization of precipitated or sorbed mercury, and it creates a more toxic and easily bioaccumulated form. MeHg is 100 times more toxic than inorganic mercury (Spry and Wiener, 1991). Seasonally, methylation in surface sediments tends to increase in spring to late summer months when bottom waters become anoxic (Jackson et al., 1980; Korthals and Winfrey, 1987).

The MeHg available for bioaccumulation in a reservoir or lake results from a balance between production by methylation and losses due to conversion to dimethylmercury and demethylation. MeHg is lost by conversion to dimethylmercury, which readily volatilizes to the atmosphere (Craig and Moreton, 1984). Formation of MeHg is favored in slightly acidic water in which anaerobic decay and formation of methane is occurring (Balshaw-Biddle and Earley, 1994). Slightly alkaline to higher pH waters favor production of dimethylmercury (Fagerstrom and Jernelov, 1972). Demethylation of MeHg is an inducible, enzymatic microbial process (Summers, 1986). The same types of bacteria, sulfate reducers and methanogens, appear involved in demethylation of freshwater sediments (Oremland et al., 1991). Demethylation inhibits the accumulation of MeHg under aerobic conditions and favors a build-up under anaerobic conditions (Merian, 1991).

Factors That Affect Methylation

With the exception of extreme southern Florida, mercury concentrations in precipitation and wet deposition rates across southern states show little spatial variation (NADP, 1998). Local biogeochemical differences in water bodies apparently cause the variation in observed mercury concentrations in fish. These differences help explain why fish from two reservoirs within the same basin less than fifty miles apart have substantially different mercury tissue concentrations (e.g., Caddo Lake and Lake O' Pines; Table 9). The total mercury concentration is not as important as the mercury available for methylation. Clearly, along with microbial activity, mercury solubility and speciation in sediment pore waters probably determines methylation rates in sediments (Gilmour, 1995).

Factors that influence methylation and ultimately mercury in fish include low pH, low alkalinity, low calcium, high color and dissolved organic carbon (DOC), reservoir formation (age), stratification (low dissolved oxygen), and fluctuating water levels. The presence of an anoxic hypolimnion creates conditions favorable for activity by sulfate-reducing bacteria and allows flux of inorganic mercury from sediments (Korthals and Winfrey, 1987). There is little information on how DOC may affect methylation rates, but its concentration influences dissolved mercury speciation, and increased DOC levels in the water column result in lower mercury methylation rates (Gilmour, 1995). Presumably, ligand formation between dissolved mercury and DOC in the water column makes mercury unavailable for methylation by bacteria (Gilmour and Henry, 1991). Mercury is removed from the water column and deposited to sediments along with the particulate organic carbon, a process enhanced by lower pH (Jackson et al., 1980; Schindler et al., 1980). Organic carbon generally has the opposite effect in sediments, enhancing both the methylation rate and bacterial activity (Callister and Winfrey, 1986).

Color and alkalinity are unlikely to directly influence methylation, but are highly correlated with water quality characteristics that do affect mercury cycling and bioaccumulation in reservoirs and lakes (Swain and Helwig, 1989). Color is a surrogate measure of DOC, including humic and fulvic acids, which chelate metals and transport them from wetlands (Gorham et al., 1984), catalyze

methylation (Kemp et al., 1978), directly methylate mercury (Nagase et al., 1982), and may act as an energy source for methylating bacteria (Verta, 1984).

Low pH of water causes increased solubility of dissolved metals and results in a greater proportion of total aqueous metal as the free metal ion (Spry and Wiener, 1991). Lower pH is also generally associated with greater mobility of metals out of sediment (Schindler et al., 1980). Data from reservoir and lake surveys indicate that mercury concentration in fish is often inversely related with pH at the sediment-water interface and in the water column (Winfrey and Rudd, 1990). In Lake Ontario, the rate of methylation at low pH ($\leq$ 4.5) was found to be about seven-times that occurring at high pH ($\geq$ 8.5)(Xun et al., 1987).

The calcium ion apparently competes with other metal cations for binding sites on gill surfaces, decreasing the direct uptake of cationic metals by fish (Wicklund and Runn, 1988; McDonald et al., 1989). Low calcium in water probably enhances the uptake of MeHg, since the permeability of fish gill membranes is inversely related to aqueous calcium concentration (Rodgers and Beamish, 1983). Alkalinity is generally regarded as a surrogate for calcium and pH, with which alkalinity is highly correlated (Swain and Helwig, 1989).

Methylation of mercury has been shown to be accelerated in newly formed reservoirs due to sudden inundation of organic matter and exposure of soils containing mercury (Abernathy and Cumbie, 1977; Cox et al., 1979). Water level fluctuations may enhance the redistribution of mercury to reservoirs and lakes from their surrounding watersheds, making more mercury available for methylation. Periodic inundation of wetlands and marshy headwater areas by fluctuating water levels may allow mobilization of mercury with dissolved and particulate organic carbon (Lee and Hultburg, 1990; Mierle, 1990). This process is enhanced in low-pH reservoirs and lakes, since mercury binds with organic material more strongly at low pH (Winfrey and Rudd, 1990).

The availability of mercury in sediment for methylation is also affected by sulfur, which forms highly insoluble sulfides with many metals, including mercury, under anaerobic conditions. The toxicity of mercury, when at high concentrations in sediment, is controlled by binding to acid-volatile sulfide (AVS). In cases where the molar concentration of AVS exceeds that of the metals, including mercury, metals are bound through the formation of insoluble metal-sulfide complexes, making them less biologically available (Ankley, 1996). Selenium has also been shown to bind with mercury in sediment and interferes with mercury methylation (Jin et al., 1997) and bioaccumulation (Turner and Swick, 1983).

Bioaccumulation and Biomagnification of Mercury

In order to enter the aquatic food chain, beginning with bacteria, mercury must be transported across the lipid membrane that surrounds unicellular organisms. The microbial uptake of mercury is a key step in both methylation and bioaccumulation. To yield high concentrations in fish, MeHg is efficiently taken up by bacteria at the bottom of the food chain where it is retained and passed on to their predators. Many trace metals are efficiently accumulated by bacteria and phytoplankton, but they are not retained. These metals diffuse out of as readily as they diffuse into bacterial cells, or they become biologically associated with particulate cellular material which is excreted by higher organisms (Morel et al., 1998). Inorganic mercury is absorbed by fish less efficiently than MeHg

from both food and water, and if absorbed, is eliminated more rapidly than MeHg (Boudou, and Ribeyre, 1985). As a result, the average proportion of MeHg over inorganic mercury increases from about 5 percent in the water column to 15 percent in phytoplankton, 30 percent in zooplankton, and 95 percent in fish (Watras and Bloom, 1992).

Since MeHg is taken up more rapidly than it can be eliminated, bioaccumulation occurs (McKim et al., 1976). Consequently, mercury tends to accumulate in fish over time and larger-sized fish tend to have higher tissue concentrations (Huckabee et al.,1979). For example, Crowe (1996) demonstrated that mercury in largemouth bass muscle tissue from Caddo Lake was highly correlated with the size of fish (Figure 3). MeHg concentrates in the skeletal muscle of fish due to mercury binding to sulphdryl groups in protein (Boudon and Ribeyre, 1985). Once in the muscle, the MeHg can not be cut out or removed by cooking (Armstrong et al., 1995).

The accumulation of MeHg in higher organisms results mainly from the ingestion of food ($\geq$ 90 %) rather than direct uptake from water ($\leq$10 %)(Spry and Wiener, 1991). Total mercury concentrations in fish are often correlated with concentrations in organisms of lower trophic levels (Suns et al., 1987; Allared and Stokes, 1989). MeHg biomagnifies in aquatic food chains, since an incremental increase in concentration occurs at each successive level, and the food sources for organisms higher in the food chain are progressively more concentrated with mercury. Consequently, piscivorous fish species generally contain the highest mercury concentrations in aquatic systems (MacCrimmon et al., 1993; Wren et al., 1983). In the southern states, mercury concentrations in alligator *Alligator mississipiensis* are often higher than in piscivorous fish (Jaqoe et al., 1998).

The nature of biomagnification is clearly shown in a Finnish lake (Figure 4). The mercury content in plankton feeders (vendace) increases through the small predators (perch), the zoobenthos predator (burbot), and in the large predator (pike). Fish-eating birds (merganser and black-throated diver) contain more mercury than the herring gull which also consumes terrestrial species and garbage (Sarkka et al., 1978). Biomagnification can then account for the observation that fish can contain more than 1 mg/kg (one part per million) mercury in their tissue while residing in water with less than 0.001 μg/L (one part per trillion) mercury (Zillioux et al., 1993).

The structure of the foodweb often determines the transfer efficiency of MeHg from bacteria to top predators. The number of trophic levels between predators and prey is critical for biomagnification. In North American lakes, the presence of planktivores such as lake herring, shad, rainbow smelt, and mysids, which increases the number of trophic levels, leads to higher mercury concentrations in top predators (Akeilaszek and Haines, 1981; Cabana and Rasmussen, 1994).

Mercury Toxicity and Health Effects

Mercury is toxic to a wide variety of aquatic organisms depending to a large degree upon the type of mercury species tested (USEPA, 1980). Reported 4-day LC_{50}s for inorganic mercury range from 33 μg/L in 2-month old rainbow trout *Salmo gairdneri* at hardness ranging from 82-132 mg/L (Hale, 1977) to 687 μg/L in adult white suckers *Catostomus commersoni* at a hardness and alkalinity of 18 and 63 mg/L, respectively (Duncan and Klaverkamp, 1983). This range includes toxicity data from

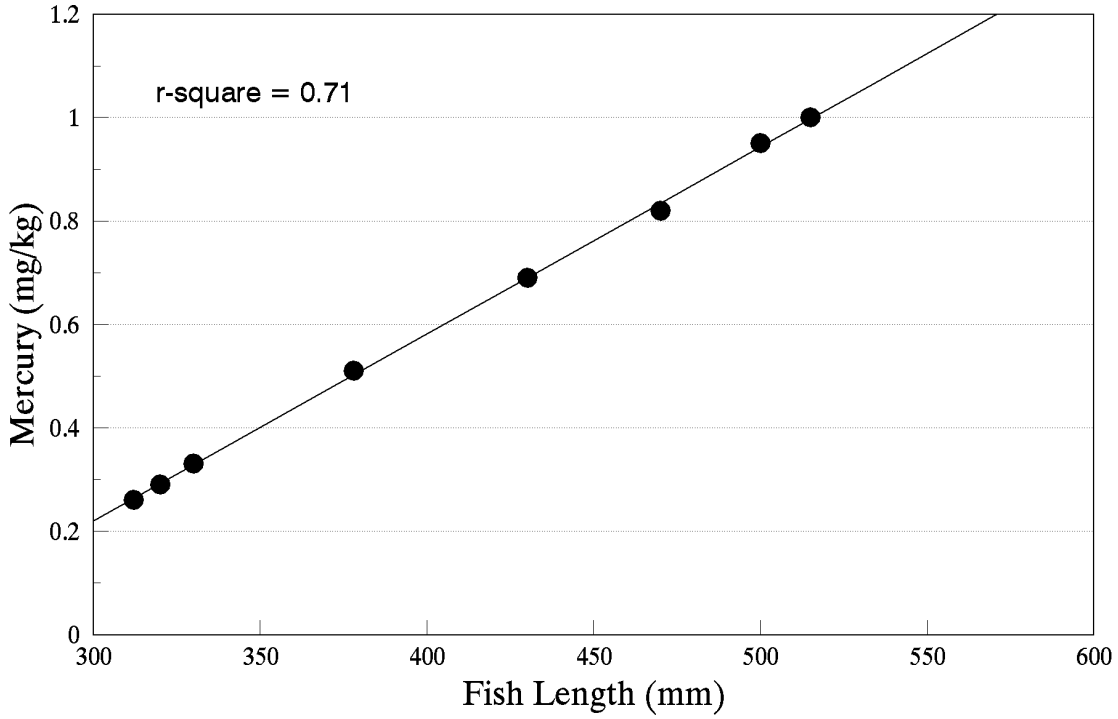


Figure 3. Relationship Between Largemouth Bass Length and Mercury Tissue Concentrations Showing Significant Bioaccumulation in Caddo Lake (Source: Crowe, 1996)

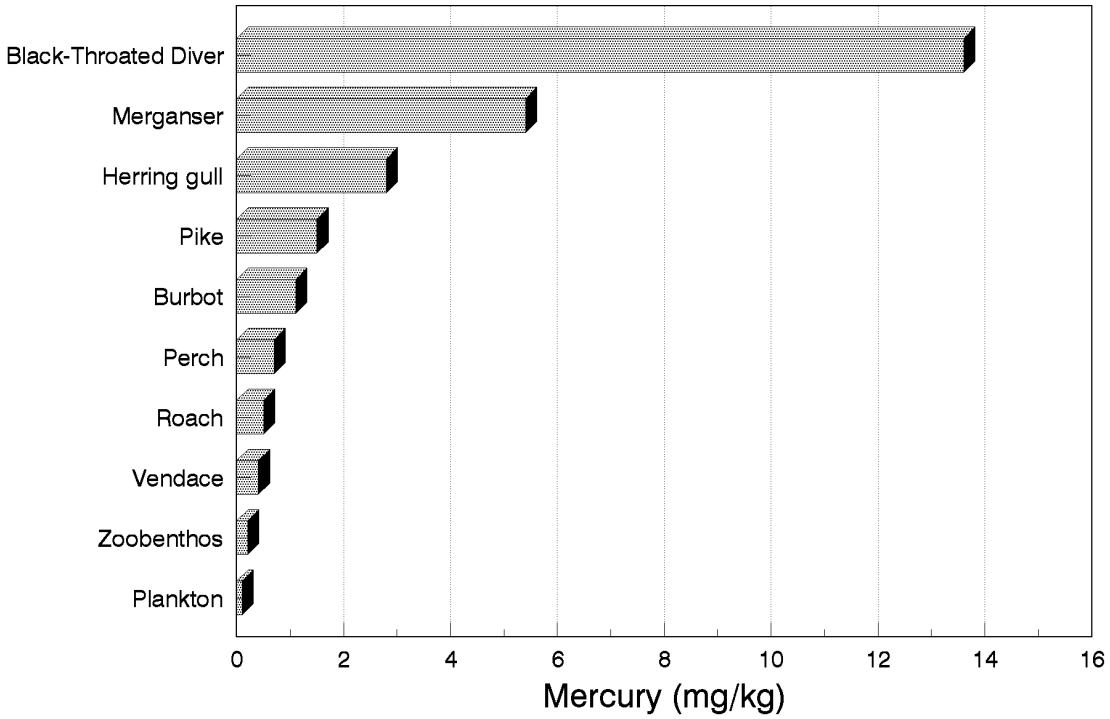


Figure 4. Biomagnification of Mercury Across Several Trophic Levels in a Finnish Lake (Source: Sarkka et al., 1978)

different species , water hardness, and temperatures. Embryo-larval stages are much more sensitive than older fish to inorganic mercury (Birge, et al., 1979, Weis and Weis, 1987). Organic forms of mercury are much more toxic than inorganic forms, and toxicity of mercury is slightly greater in freshwater than saltwater. Mercuric chloride, for example, is 7-fold more toxic than mecuric chloride to rainbow trout (Matida, et al., 1971). Phenylmercuric chloride is 30-fold more toxic than inorganic mercury in 24-hour exposures (MacLeod and Pessah, 1973). In contrast to short-term exposures, chronic exposures suggest similar effect levels for inorganic (Snarski and Olson, 1982) and MeHg (McKim, et al., 1976).

The Texas Natural Resource Conservation Commission (TNRCC) has developed total recoverable acute (2.4 μg/L; 24-hour average) and chronic (1.3 μg/L; seven-day average) criteria in the Texas Surface Water Quality Standards (TSWQS) to protect freshwater aquatic life from the toxic effects of mercury. For total recoverable mercury to protect saltwater aquatic life, the TNRCC has derived slightly lower acute (2.1 μg/L) and chronic (1.1 μg/L) criteria (TNRCC, 1997). Mercury criteria developed by the TNRCC to protect human health from consumption of water and fish are generally two full orders of magnitude lower. For the protection of human health from the toxic properties of mercury ingested through drinking water and freshwater fish, or from freshwater fish alone, the TNRCC has established a criterion of 0.0122 μg/L. The human health criterion (0.025 μg/L) for protection against consumption of saltwater fish is roughly double the freshwater criterion.

Mercury is a pollutant to which aquatic life, wildlife, and humans can be exposed through the environment. The most likely exposure route for humans is through consumption of fish that are contaminated with MeHg. The effects of exposure of humans to mercury have been well documented through accidental environmental contamination events in Sweden (1952), Japan (1955), and Iraq (1956, 1960, and 1970-71) and through studies of populations in New Zealand, Faroe Islands, and Seychelle Islands that rely on a diet of fish (containing mercury) as the major source of protein in their diet (ATSDR, 1999).

MeHg in the human diet is almost completely absorbed into the bloodstream. Gastrointestinal absorption of MeHg is usually more than 90 percent of oral intake regardless of age, including the neonatal stage. Once absorbed, MeHg is widely distributed to all organs and tissues, and can easily cross naturally occurring protective barriers such as the placental barrier and the blood-brain barrier (ATSDR, 1999).

MeHg is a neurotoxin that attacks the central nervous system, potentially impacting the sensory, visual, and auditory functions and those areas concerned with coordination, especially the cerebellum. Elemental mercury vapor produces an acute pneumonitis. Early signs of MeHg poisoning have been shown to appear when concentrations approach 50 mg/kg in hair or 200 μg/kg in whole blood. Inorganic mercury salts can cause acute gastrointestinal distress and renal failure. Eye and skin irritation can also occur. Early symptoms of mercury poisoning include malaise, blurred vision, tunnel vision, deafness, loss of coordination, impaired taste and smell, tremor, and tingling of the extremities. Humans are particularly sensitive to the adverse effects of MeHg during prenatal life and infancy due to sensitivity of the developing nervous system. Mercury has not been shown to cause cancer in humans (ATSDR, 1999).

THE MERCURY PROBLEM IN TEXAS

Initial Concerns About Mercury in Texas Waters

The TNRCC first became concerned about possible mercury contamination of Texas fishes in the summer of 1992. Routine fish tissue monitoring in neighboring states documented mercury contamination along the Ouachita River in southeastern Arkansas and northeastern Louisiana (Armstrong et al, 1995; LDEQ, 1995). Additional fish tissue monitoring revealed that mercury contamination was more widespread and not restricted to the Ouachita River Basin. By the summer of 1993, fish consumption advisories were in place for several southern Arkansas rivers and lakes due to mercury contamination. In the fall of 1993, the Louisiana Department of Environmental Quality (LDEQ) found elevated mercury levels (> 0.5 mg/kg) in largemouth bass from Caddo Lake (Segment 0401) and Toledo Bend Reservoir (Segment 0504), two water bodies that form portions of the Texas-Louisiana state boundary.

Only two water bodies in Texas (Lavaca and Cox bays) were known to be contaminated in 1992 when concern first arose over possible mercury contamination of Texas fishes. Mercury was released into Lavaca Bay (Segment 2453) from 1966 to 1970 by a local manufacturing plant (TDH, 1996). This discharge of mercury also impacted nearby Cox Bay. The mercury came from a chlor-alkali unit at the plant. From 1970 to 1979, wastewater containing mercury was released to lagoons adjacent to the plant. The chlor-alkali unit ceased operation in 1979. Sampling of Lavaca Bay by the Texas Department of Health (TDH) for more than 20 years has shown that fish and crabs contain elevated mercury levels, primarily in waters near the manufacturing plant. The TDH issued an aquatic life closure in April 1988 that covers an area around the plant in Lavaca Bay and adjacent Cox Bay. The aquatic life closure prohibits the taking of any fish or crabs from the affected area. The aquatic life closure for Cox Bay was rescinded by the TDH in January 2000 after analysis of fish and shellfish indicated that mercury had decreased to acceptable concentrations (TDH, 2000).

Historical Fish Tissue Studies in Texas

Several fish tissue studies in Texas, which included assessment of mercury, preceded the present study. From late 1984 to early 1985, the U.S. Fish and Wildlife Service (FWS) collected a total of 315 composite samples of whole fish from 109 sites nationwide, which were analyzed for metals (Schmitt and Brumbaugh, 1990). Seven of the sites sampled were located in Texas. Metal concentrations in fish tissue in 1984-85 were compared to earlier two-year periods (1976-77, 1978-79, 1980-81) to evaluate temporal and geographic trends. The national mercury geometric mean ($\leq$ 0.12 mg/kg) and 85th percentile ($\leq$ 0.19 mg/kg) did not change during 1976-84. The mercury geometric means from each of the seven Texas sites also did not change.

From 1986-89, the EPA conducted the "National Study of Chemical Residues in Fish", a one-time screening investigation to determine the prevalence and sources of selected bioaccumulative pollutants in fish (USEPA, 1992). A total of 388 sites was sampled nationwide; 314 were "targeted" sites thought to be influenced by various point and nonpoint pollutant sources. The remaining 74 sites were located away from pollution sources and selected to provide an indication of background concentrations. Of ten sites located in Texas, nine were considered targeted to point and nonpoint

pollutant sources. In general, bottom feeders were analyzed as whole-body composite samples to determine the occurrence of study chemicals. Game fish were analyzed as fillet composite samples to indicate potential risks to human health from fish consumption. Although 119 species were collected, most belonged to 14 different species. Carp *Cyprinus carpio* was the most frequently collected bottom feeder and largemouth bass was the most frequently collected game fish. Tissue samples were primarily analyzed for organic pollutants, as mercury was the only metal included. Mercury was detected in at least one sample from 92 percent of the sites, making it one of the most frequently detected pollutants. The national maximum, mean, and median mercury tissue concentrations reported from the study were 1.8, 0.26, and 0.17 mg/kg, respectively. The nationwide median for background sites only was lower (0.09 mg/kg). Ten of the sites with the highest 10th percentile mercury concentrations were located near paper mills, four near Superfund sites, and most of the remaining sites were from industrial areas. Mercury was one of the pollutants that was not detected in high enough concentrations to pose a human health concern for the average fish-eating human population.

From 1993 through 1994, the FWS conducted a toxic substances study of three reservoirs (Caddo Lake, Lake O' Pines, and Cypress Springs) located in northeast Texas (Giggleman, et al., 1998). Mercury was included in a suite of parameters evaluated in sediment, fish, macroinvertebrates, and birds. Fish samples consisted of whole body composite and individual fillet samples. Whole body grass shrimp *Palaemonetes sp.* comprised the macrobenthos samples. Feather, kidney, and liver tissues from nestling great blue herons *Ardea herodias* were analyzed. Sediments from Caddo Lake and Lake O' Pines exceeded the TNRCC statewide 85th percentile (0.19 mg/kg) for reservoirs. Mercury in whole body composite samples from all three reservoirs exceed the FWS predator protection level (0.1 mg/kg); however, fillet samples contained concentrations below the FDA action level (1.0 mg/kg). Largemouth bass, followed by spotted gar *Lepisosteus oculatus*, contained the highest mercury concentrations of any fish tested. Grass shrimp samples from two of nine sites on Caddo Lake also exceeded the predator protection limit. Elevated mercury concentrations were also found in nestling great blue herons from Caddo Lake.

From 1993 through 1994, the TNRCC conducted a study of mercury in Cypress Creek and upper Sabine River basins in northeast Texas (Crowe, 1996). Water, sediment, soil, coal, fish tissue, benthic macroinvertebrate tissue, sphagnum bog cores, Spanish moss, and tree cores were sampled for mercury. Sampling was conducted at Lake Bob Sandlin, Lake O' Pines, Pruitt Lake, Benten Lake, and Caddo Lake in the Cypress Creek basin and at two sites on the Sabine River and Brandy Branch Reservoir in the Sabine River basin. Mercury concentrations in sediment were highly variable among the sampling sites (range <0.01-1.57 mg/kg). Upland soils contained less variable mercury concentrations (range 0.01-0.14 mg/kg). Mercury concentrations in tree cores did not show observable differences from >60 years before present. Composite macrobenthos samples, which were comprised mainly of crayfish, dragonflies and damselflies, contained generally low mercury concentrations (range <0.06-0.18 mg/kg). Mercury concentrations in largemouth bass for the Cypress basin sites ranged from 0.03 to 1.01 mg/kg and generally increased with size of fish. A strong statistical relationship was demonstrated between mercury concentrations and size in largemouth bass from Caddo Lake. Spotted bass in Pruitt Lake also showed a similar relationship of increasing mercury concentrations with size of fish. Significant correlations were also found between age of fish (based on otoliths) and tissue mercury concentrations for spotted bass from Pruitt

Lake. Whole body channel catfish samples from the Sabine River basin contained generally low mercury concentrations (range 0.12-0.23 mg/kg).

TNRCC Historical Mercury Review

The TNRCC initially decided to conduct a review of historical mercury in fish tissue data to determine if Texas fishes were contaminated. The TNRCC's Surface Water Quality Monitoring Program (SWQMP) provides an integrated statewide evaluation of physical, chemical, and biological systems in relation to human health concerns, ecological condition, and designated uses assigned to water bodies in the TSWQS. A variety of water quality data are routinely collected at approximately 600 sites statewide by staff of 14 of the16 TNRCC regional offices and by Texas Clean Rivers Program participants.

Toxic chemical contaminants are sometimes assimilated through aquatic food chains and subsequently bioaccumulated in fish tissues. The SWQMP uses fish tissue monitoring to identify areas where water quality or sediment may be contaminated, and to detect and evaluate toxic chemical levels in fish that may be harmful to humans. Fish tissues are routinely checked for the presence of 40 pesticides, 31 volatile organic substances, 63 semivolatile organic substances, and seven metals, including mercury. Due to high costs for laboratory analysis toxic substances, analysis of metals in tissues is included at only about 20-50 sites each year by the TNRCC. The TNRCC focuses its fish tissue monitoring in areas likely to be contaminated that may include: sites near wastewater discharges with documented receiving water or effluent toxicity, sites that have shown recurrent ambient water and/or sediment toxicity, sites near large industrial or domestic outfalls, sites that receive high nonpoint source loads, sites with exceptional recreation uses, sites adjacent to hazardous waste or superfund facilities, sites downstream of major metropolitan areas, and sites that exhibit biological impairment.

The objective of most of the TNRCC's routine monitoring of fish tissue at fixed stations is to evaluate ecosystem health. This type of monitoring is generally conducted at sites impacted by point and/or nonpoint sources in order to document contaminant accumulations in fish. The sampling framework the TNRCC utilizes to evaluate ecosystem health is shown in Table 2.

In addition to fish tissue monitoring routinely conducted at fixed sites, the TNRCC collects fish for tissue analysis during special studies targeted to identification of toxic substances. These studies are conducted primarily in areas where data collected during routine fixed station monitoring indicate potential problems with accumulations of toxic chemicals in water, sediment, or fish tissue. Special studies may be used by the TNRCC to evaluate sources, fate, and distribution of toxic chemical contaminants in selected water bodies. Sampling strategies for special studies often involve collection of data to evaluate ecosystem health and human health risk.

Fish tissue data collected during routine fixed station and special study monitoring events are stored in an Ingres database in the SWQM module of the TNRCC integrated database system (TRACS). The SWQMP database contains data dating back to 1968 which have been collected by the TNRCC, as well as other agencies.

Table 2

TNRCC Fish Tissue Sampling Framework for Evaluation of Ecosystem Health

Sampling Information	Sampling Description
Where to Sample	Areas impacted by point or nonpoint sources
Type of Sample	Whole body composites
Number of Species	2-3 for each sampling event
Number of Individuals	3-5 of same species
Size of Individuals	Various size groups with individuals varying in total length by no more than 10 percent within a sample
Sampling Frequency	Once per year for two years
Example Species	**Freshwater:** bass, sunfish, carp, catfish, forage fish **Saltwater:** hardhead catfish, forage fish

Table 3

Number of Sites Sampled, Number of Mercury in Tissue Analyses,
and Type of Fish Tissue Sampled by the TNRCC, 1984-1993

Year	Number of Sampling Sites	Number of Hg Samples	Tissue Type Sampled	
			Fillet	Whole Body
1984	22	25	0	25
1985	26	28	0	28
1986	26	27	0	27
1987	26	30	0	30
1988	30	41	0	41
1989	53	69	6	63
1990	49	128	57	71
1991	29	38	2	36
1992	24	36	5	31
1993	52	94	22	72
Totals		**516**	**92 (18 %)**	**424 (82 %)**

In the ten-year period from 1984-1993 prior to the start of this study, the TNRCC collected a total of 516 fish tissue samples, which included analysis of mercury, at fixed stations and during special studies (Table 3). The smallest number of annual mercury in tissue samples (25) was collected in 1984 and the most (128) in 1990. Analysis of mercury in tissue was more commonly conducted on whole body fish samples (424 samples; 82 % of total) than on edible muscle portions (92 samples; 18 % of total). Channel catfish, carp, and largemouth bass were the species most frequently submitted to the laboratory for analysis of mercury in tissue during the ten-year period (Table 4). Collectively, these three fish species accounted for 41 percent (211 samples) of all samples submitted to the laboratory for analysis of metals in tissue.

Of these three species, only one channel catfish whole body sample (West Fork Trinity River; 0.83 mg/kg), and one largemouth bass whole body sample (White River Lake; 0.65 mg/kg) had mercury concentrations greater than 0.5 mg/kg (Table 5). The highest mercury concentration (0.34 mg/kg) in carp occurred in a fillet sample collected from Town Lake (Segment 1429). Mean and median mercury concentrations in whole body and fillet samples for each of the three species indicate that central tendency values have been much lower than the few elevated maxima. Mean and median mercury concentrations in tissue are highest for largemouth bass whole body and fillet samples.

A plot of maximum mercury in tissue concentrations in whole body and fillet samples from all species collected at 93 sites from 1984-1993 indicates no distinct statewide geographical trend (Figure 5). The highest mercury in tissue levels are scattered throughout the state rather than clustered within one region. The highest mercury in tissue concentrations reported were in a red drum *Sciaenops ocellatus* whole body sample (5.12 mg/kg) from Lavaca Bay, three red drum and one black drum *Pogonias cromis* whole body samples from Lavaca Bay (> 1.5 mg/kg), a spotted gar whole body sample from Caddo Lake (1.04 mg/kg), a walleye *Stizostedion vitreum* whole body (0.88 mg/kg) sample from Lake Mackenzie, a channel catfish whole body sample (0.83 mg/kg) from the West Fork Trinity River, a longnose gar *Lepisosteus osseus* whole body sample (0.73 mg/kg) from the Rio Grande near Presidio, a walleye fillet sample (0.67 mg/kg) from Lake Meredith, a largemouth bass whole body sample (0.64 mg/kg) from White River Lake, a longnose gar whole body sample (0.56 mg/kg) from the Sabine River near Longview, and a shortnose gar *Lepisosteus platostomus* whole body sample (0.55 mg/kg) from the Trinity River near Dallas.

There was little indication from historical TNRCC fish tissue data to suggest that many fish or water bodies were contaminated by mercury. A plot of the median mercury in tissue concentrations for all fish species collected at the aforementioned 93 sites indicates only those from Lavaca Bay were uniformly high, exceeding 0.5 mg/kg. (Figure 6). Further, for most of the water bodies (74 of 93; 80%) sampled by the TNRCC during the ten-year period, median mercury in tissue concentrations for all fish species were less than 0.2 mg/kg (Figure 7). Finally, about 75 percent of all fish collected during the ten-year period from the 93 water bodies had mercury concentrations ≤ 0.2 mg/kg, and about 95 percent had concentrations ≤ 0.5 mg/kg (Figure 8).

Table 4

Fish Species Most Frequently Analyzed for Mercury in Tissues
by the TNRCC, 1984-1993

Scientific Name	Common Name	Number of Samples	Percent of Total
Ictalurus punctatus	Channel catfish	87	17
Cyprinis carpio	Carp	65	13
Micropterus salmoides	Largemouth bass	59	11
Arius felis	Hardhead catfish	28	5
Dorosoma cepedianum	Gizzard shad	19	4
Micropogon undulatus	Atlantic croaker	18	4
Stizostedion vitreum	Walleye	16	3
Lepisosteus osseus	Longnose gar	14	3
Pylodictis olivaris	Flathead catfish	12	2
Ictalurus furcatus	Blue catfish	11	2

Table 5

Summary of TNRCC Fish Tissue Sampling Results, 1984-1993

Common Name	Tissue Type	Number of Samples	Number of Fish	Mercury in Tissue (mg/kg)			
				Minimum	Maximum	Mean	Median
Channel catfish	Fillet	18	45	0.06	0.28	0.13	0.15
	Whole	69	140	0.01	0.83	0.12	0.12
Carp	Fillet	30	81	0.09	0.34	0.18	0.18
	Whole	35	103	0.02	0.29	0.13	0.15
Largemouth bass	Fillet	12	26	0.06	0.48	0.24	0.22
	Whole	47	114	0.02	0.64	0.22	0.24

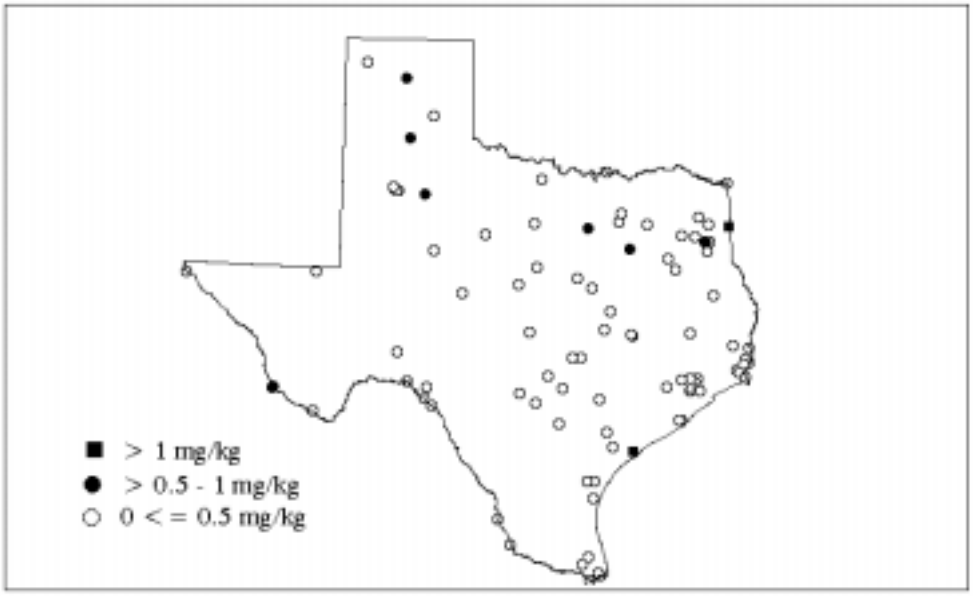


Figure 5
Distribution of Maximum Mercury in Tissue Concentrations
for All Species at 93 TNRCC Sites, 1984-1993

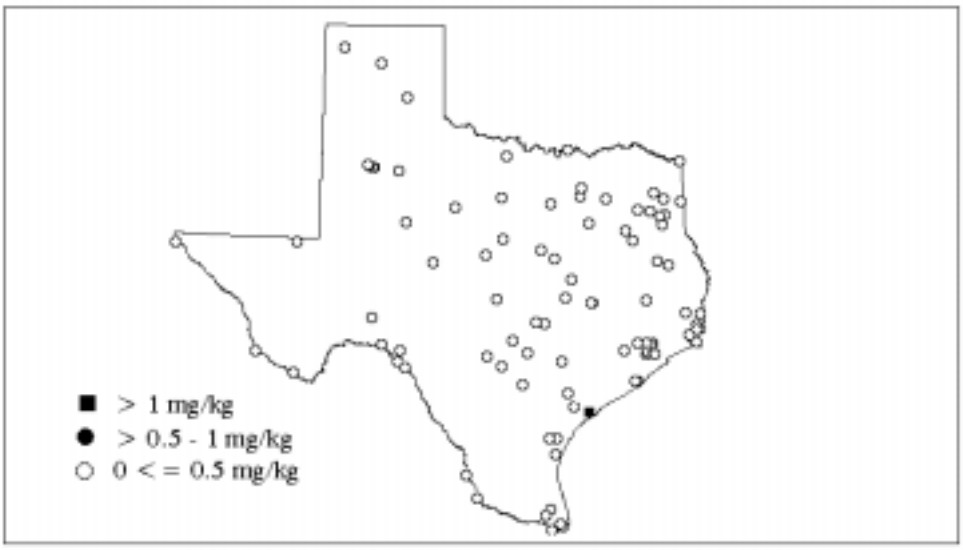


Figure 6
Distribution of Median Mercury Tissue Concentrations
for All Species at 93 TNRCC Sites, 1984-1993

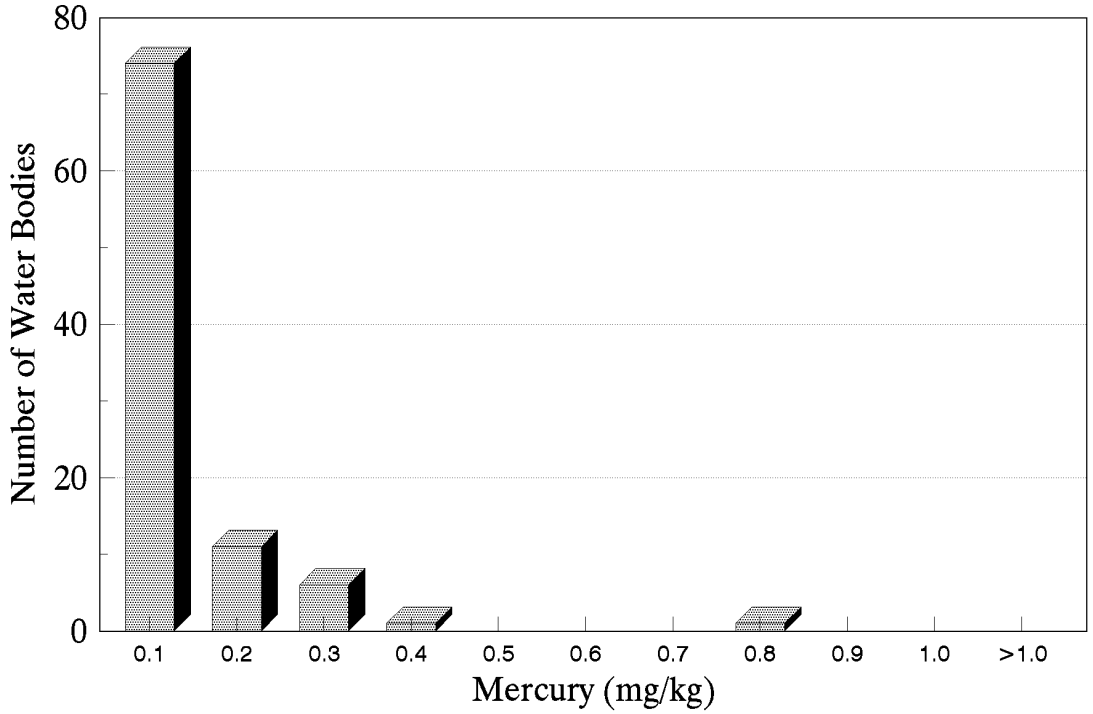


Figure 7. Median Mercury Tissue Concentrations for All Fish Species at 93 Sites Monitored by the TNRCC, 1984-1993

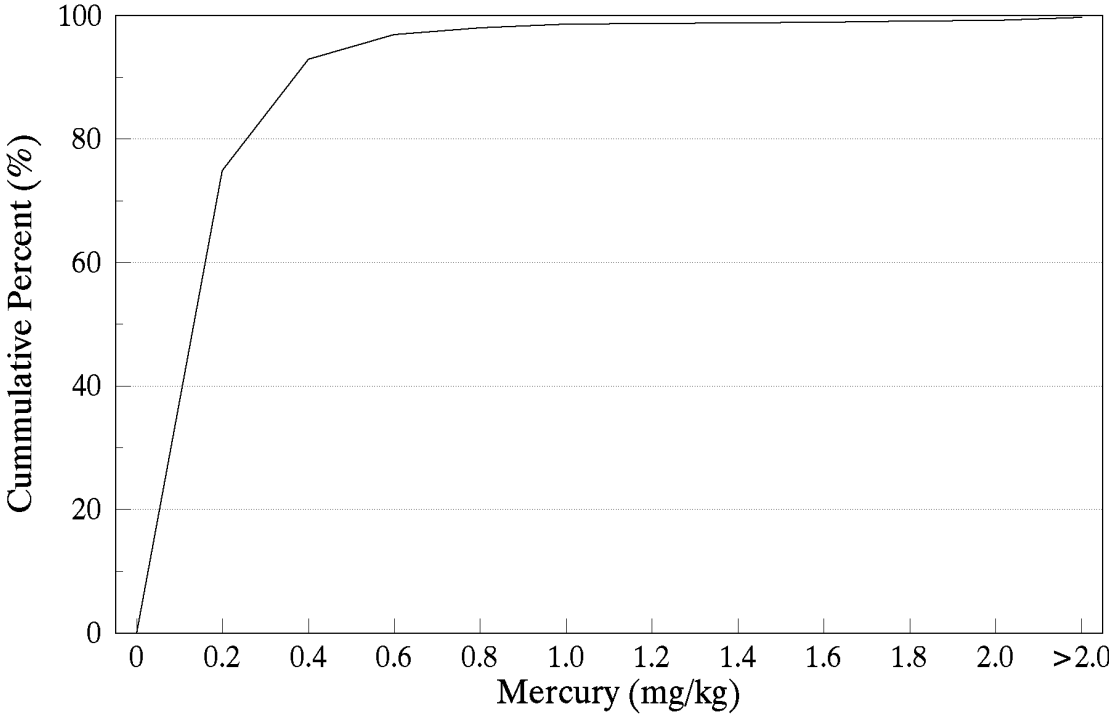


Figure 8. Cummulative Frequency of Occurrence for Maximum Mercury Tissue Concentrations for all Fish Species from 93 Sites Monitored by the TNRCC, 1984-1993

TDH Historical Mercury Review

The TDH Seafood Safety Division, which also has a fish and shellfish sampling program, conducted a review of their historical fish and shellfish tissue data in the fall of 1992 to determine if mercury contamination was indicated. The primary goal of the TDH program is to provide information about contaminants in fish and shellfish which may be harmful to human health. The TDH uses surveys by biologists in fresh and coastal waters, laboratory analysis of samples, and risk assessments of data to determine if consumption advisories or aquatic life closures are warranted at specific sites. The TDH sampling program does not involve routine monitoring at fixed stations, but rather responds to site-specific contaminant concerns. The TDH usually analyzes for a large number of chemical contaminants (organic substances and metals) in fish and shellfish tissue samples, since samples cannot be determined to be safe for human consumption if only selected compounds are evaluated.

The TDH monitoring is generally conducted at sites impacted by point and/or nonpoint sources that are commonly used for recreational fishing. The sampling framework the TDH utilizes to evaluate human health risk (Table 6) is very different from the one utilized by the TNRCC to evaluate ecosystem health (Table 2). The major difference is that the TDH analyzes individual fillets of edible muscle tissue, while the TNRCC has generally analyzed whole body composite samples. The TDH periodically publishes their historical fish and shellfish tissue data (TDH, 1998a).

Nearly all of the fish and shellfish tissue samples analyzed for mercury prior to 1994 by the TDH were from bay systems along the Texas Gulf Coast (Table 7). The TDH collection efforts in Texas bays were made primarily to document background mercury in tissue concentrations for comparison to Lavaca Bay near the time that the TDH issued an aquatic life closure in 1988 (Wiles, 1999). Fishes from Lake Meredith (Canadian River) and Red Bluff Reservoir (Pecos River) were targeted for mercury analysis following issuance of consumption advisories for mercury in fish in the upper portions of the two watersheds in New Mexico in 1991 (NMHED, 1991). The TDH conducted studies of metals in fish tissue from Braunig and Calaveras lakes in 1985 and Pine Island and Cow bayous in 1987. For the other water bodies sampled by the TDH, some metals in tissue samples were collected during studies primarily targeted for evaluation of toxic organic compounds.

In addition to Lavaca Bay, 16 of the other 37 water bodies had fish and/or shellfish with mercury concentrations in edible tissues for some samples that exceeded 0.5 mg/kg (Table 7). The highest mercury concentrations were found in Lavaca Bay oysters *Crassostrea virginica* (8 mg/kg), black drum and red drum (> 5 mg/kg), and gafftopsail catfish *Bagre marinus* (> 3 mg/kg). Fillets from large black drum from Matagorda Bay (5.9 mg/kg) and Espiritu Santo Bay (1.5 mg/kg), and a spotted seatrout *Cynoscion nebulosus* from the Laguna Madre (1.7 mg/kg) were also notably elevated. Evaluation of the mercury in tissue data by the TDH indicated much lower central tendencies for all fish and shellfish species than maximum concentrations in each water body. The mercury in tissue concentrations were low enough that the TDH concluded the risk of consuming fish and shellfish by humans from the 37 water bodies was not sufficiently high to trigger the issuance of consumption advisories or aquatic life closures (Table 7).

Table 6

TDH Fish and Shellfish Sampling Framework for Evaluation of Human Health Risk

Sampling Information	Sampling Description
Where to Sample	Areas impacted by point and nonpoint sources that are commonly used for recreational or commercial fishing
Type of Sample	Individual fillets of muscle tissue
Number of Species	2-4
Number of Individuals	10-30 per species (analyzed separately)
Size of Individuals	Greater than legal limit if possible
Sampling Frequency	Once for each event, except where a consumption advisory or aquatic life closure is issued when sampling may be continued for an indefinite period
Example Species	**Freshwater:** bass, crappie, catfish, suckers **Saltwater:** oysters, shrimp, crabs, Atlantic croaker, sand trout, speckled trout, red drum, black drum, flounder, catfish

Table 7

Summary of TDH Fish Sampling, Consumption Advisories and Aquatic Life Closures, 1970-1993

Water Body	TNRCC Segment	Date(s) of Collection	Number of Samples	Number of Species	Minimum Hg Level (mg/kg)	Maximum Hg Level (mg/kg)	Mercury Advisory/ Closure Status
Lake Meredith	0102	4/91	21	4	0.04	0.53	None Issued
Sulphur River	0301	6/93	12	9	0.19	0.76	None Issued
Cow Bayou	0511	7/87	5	4	0.29	0.47	None Issued
Brakes Bayou	0601	11/93	6	5	0.03	0.17	None Issued
Neches River Tidal	0601	11/93	4	3	0.028	0.14	None Issued
Pine Island Bayou	0607	8/87	4	4	0.36	0.69	None Issued
Trinity River	0804/05/19 /24/41	8/87-4/88	36	14	0.04	0.53	None Issued
San Jacinto River Tidal	1001	5/90	3	1	<0.04	0.13	None Issued
Clear Creek	1102	10/93	14	8	0.03	0.32	None Issued
Houston Ship Channel	1005/06/07	4/72-5/90	9	2	0.06	0.42	None Issued
Town Lake	1429	2/85	13	4	0.02	0.14	None Issued
Braunig Lake	1911	8/85	20	6	<0.02	0.058	None Issued
Calaveras Lake	1911	8/85	13	6	<0.02	0.073	None Issued
Donna Reservoir	2202	5/93	6	4	<0.02	0.34	None Issued
Red Bluff Reservoir	2312	4/91	10	3	<0.04	0.34	None Issued
Sabine Lake	2412	7/70-2/82	38	9	<0.02	0.24	None Issued

Table 7 TDH Fish and Shellfish Mercury in Tissue Data Collected 1970-1993 (Continued)

Water Body	TNRCC Segment	Date(s) of Collection	Number of Samples	Number of Species	Minimum Hg Level (mg/kg)	Maximum Hg Level (mg/kg)	Mercury Advisory/ Closure Status
Tabbs Bay	2426	6/83-5/90	6	3	<0.05	0.08	None Issued
Galveston Bay	2421/39	5/78-4/92	288	12	<0.01	0.53	None Issued
Trinity Bay	2422	8/74-5/90	38	9	<0.01	0.27	None Issued
East Bay	2423	5/70-10/84	18	6	0.01	0.25	None Issued
West Bay	2424	11/69-8/90	65	9	<0.01	0.21	None Issued
Clear Lake	2425	6/72	8	4	0.05	0.27	None Issued
Bastrop Bay	2433	4/83	5	2	0.07	0.17	None Issued
East Matagorda Bay	2441	7/70-6/83	33	9	<0.01	0.85	None Issued
Matagorda Bay	2451	7/70-3/83	36	9	0.01	5.9	None Issued
Powderhorn Lake	2451	2/81-7/84	9	5	0.03	0.27	None Issued
Tres Palacios Bay	2452	8/70-8/83	38	9	<0.01	0.57	None Issued
Lavaca Bay	2453	7/70-10/91	1387	16	<0.01	8.0	Aquatic Life Closure Issued 4/20/88
Espiritu Santo Bay	2461	7/70-7/84	40	9	<0.01	1.5	None Issued
San Antonio Bay	2462	7/70-3/85	43	10	<0.01	0.93	None Issued
Mesquite Bay	2463	7/70-6/83	27	7	<0.02	0.2	None Issued
Aransas Bay	2471	7/70-7/84	42	8	0.01	0.55	None Issued
Copano Bay	2472	7/70-6/84	34	9	<0.02	0.7	None Issued
Corpus Christi Bay	2481	2/71-7/84	17	9	0.02	0.53	None Issued

Table 7 TDH Fish and Shellfish Mercury in Tissue Data Collected 1970-1993 (Continued)

Water Body	TNRCC Segment	Date(s) of Collection	Number of Samples	Number of Species	Minimum Hg Level (mg/kg)	Maximum Hg Level (mg/kg)	Mercury Advisory/ Closure Status
Nueces Bay	2482	2/71-7/84	33	8	<0.02	0.43	None Issued
Laguna Madre	2491	5/77-7/87	55	11	0.01	1.7	None Issued
Baffin Bay	2492	5/77-7/87	20	6	0.05	0.74	None Issued
South Bay	2493	5/79-8/84	28	5	<0.02	0.72	None Issued

TPWD Historical Mercury Review

Prior to 1994, the Texas Parks and Wildlife Department (TPWD) did not have a routine fish tissue monitoring program in place. However prior to 1994, TPWD conducted two major special studies that involved analysis of trace metals, including mercury, in fish tissues. The TPWD conducted a study (1985-1989) to evaluate contaminants in fish tissues collected from reservoirs adjacent to coal-fired steam electric generating plants (Cantu and Moss, 1998). Arsenic, chromium, mercury, selenium, and zinc were included in the parametric coverage due to their association with coal and potential toxicity to aquatic organisms. The majority of samples collected during the project were individual liver and muscle tissues from largemouth bass and bluegill *Lepomis macrochirus*. Threadfin shad *Dorosoma cepedianum* whole body samples were also evaluated if they were included in the catches. Reservoirs sampled in the study included: Bob Sandlin, Brandy Branch, Braunig, Calaveras, Cypress Springs, Fairfield, Fayette County, Gibbons Creek, Limestone, Martin Lake, Monticello, Twin Oaks, and Welsh. Mean mercury concentrations in largemouth bass and bluegill fillets and threadfin shad whole body samples for all the reservoirs were quite low (<0.2 mg/kg). The study documented that mercury concentrations in tissues were not strongly influenced by sex of the fish. Correlations between mercury accumulation and size of fish were generally positive, with higher tissue residues found in larger fish. The TPWD study did not reveal problems with elevated mercury concentrations in fish tissues from the 13 reservoirs. However, the study provided the basis for issuance by the TDH of consumption advisories for selenium in fish for Brandy Branch, Martin Lake, and Welsh Reservoirs (TDH, 1998b).

The TPWD also conducted a contaminant study of Christmas Bay, Espiritu Santo Bay, and South Bay (1991-1993) to document background conditions in minimally impacted bays. Individual liver, muscle, and whole body samples from hardhead catfish *Arius felis*, red drum, southern flounder *Paralichthys lethostigma*, spotted seatrout, and Atlantic croaker *Micropogon undulatus* were sampled during the study (Sager, 1995). Oysters and blue crabs *Callinectes sapidus* were also sampled from the three bays. Mercury concentrations in finfish, crab, and oyster tissues from the three bays were often less than the reporting limit (<0.1 mg/kg) and mean concentrations for most species were less than 0.2 mg/kg (Table 8). Mercury concentration (0.61 mg/kg) in a spotted seatrout fillet from Espiritu Santo Bay was the only sample in the study that exceeded 0.5 mg/kg. Results of the TPWD study reflected generally low mercury concentrations in edible tissues for all species sampled from the three minimally impacted bays.

Conclusions From TNRCC, TDH, and TPWD Historical Mercury Data Reviews and Early Mercury Studies

By the spring of 1993, based on reviews of historical mercury concentrations in fish and shellfish tissues, the TNRCC, TDH, and TPWD concluded there was no compelling evidence to suggest widespread, or even regional, contamination. There were few streams and rivers, reservoirs, or bays that contained fish and shellfish with elevated mercury concentrations. Those with elevated mercury were erratically distributed spatially. With the exception of Lavaca Bay, none of the water bodies had consistently elevated mercury concentrations in fish and shellfish tissue. However, several events and studies would soon begin to change the perception that fish from the eastern portion of the state were not contaminated.

Table 8

TPWD Mercury Concentrations in Finfish, Crab, and Oyster Tissue
from Christmas, Espiritu Santo, and South Bays, 1991-1993

Water Body	Common Name	Tissue Type	Number of Samples	Mercury in Tissue (mg/kg)		
				Minimum	Maximum	Mean
Christmas Bay	Spotted seatrout	Fillet	15	<0.1	<0.1	0.05
	Red drum	Fillet	23	<0.1	0.1	0.1
	Southern flounder	Fillet	14	<0.1	0.1	0.06
	Hardhead catfish	Fillet	7	<0.1	0.1	0.08
	Atlantic croaker	Fillet	5	<0.1	0.1	0.06
	Blue Crab	Muscle	60	<0.1	<0.2	0.07
	Oyster *	Whole Body	10	<0.1	<0.1	0.05
Espiritu Santo Bay	Spotted seatrout	Fillet	28	<0.13	0.61	0.19
	Red drum	Fillet	24	<0.13	0.33	0.26
	Southern flounder	Fillet	11	<0.13	0.15	0.08
	Hardhead catfish	Fillet	17	<0.13	0.16	0.07
	Atlantic croaker	Fillet	5	<0.1	0.1	0.06
	Blue Crab	Muscle	61	<0.13	<0.13	0.065
	Oyster *	Whole Body	10	<0.13	<0.13	0.065

Table 8 TPWD Mercury in Tissue Data from Christmas, Espiritu Santo, and South Bays (Continued)

Water Body	Common Name	Tissue Type	Number of Samples	Mercury in Tissue (mg/kg)		
				Minimum	Maximum	Mean
South Bay	Spotted seatrout	Fillet	23	<0.2	0.2	0.2
	Red drum	Fillet	22	<0.1	<0.1	<0.1
	Southern flounder	Fillet	2	<0.1	<0.1	<0.1
	Hardhead catfish	Whole Body	18	<0.2	0.3	0.21
	Atlantic croaker	Fillet	2	<0.1	<0.1	<0.1
	Blue Crab	Muscle	18	<0.1	<0.1	<0.1
	Oyster *	Whole Body	3	<0.1	<0.1	<0.1

* All oyster samples were composited

Representatives of southern states from Texas on the west to North Carolina on the east met in Baton Rouge, Louisiana in March 1994, and created the Southern States Mercury Task Force (SSMTF). This group was formed to exchange information concerning mercury monitoring programs, analytical results, and risk communication/management of contamination in fish (SSMTF, 1994). Later in the year (September 1994), the EPA sponsored a "National Forum on Mercury in Fish" in New Orleans, Louisiana (USEPA, 1995). The primary purpose of this forum was to transfer "state-of-the-science" information about mercury to states and other parties involved monitoring, formulation of risk assessments, and issuance of fish consumption advisories.

Important information concerning mercury that was transferred through the forums includes: (1) mercury is concentrated primarily in the muscle tissue, (2) large, older, predatory fish species at the top of aquatic food chains are individuals most likely to be contaminated by mercury, and (3) environmental conditions that favor the formation of organic mercury and its uptake by aquatic organisms include acidic water, substantial organic matter in water and sediment, anoxic conditions near the water/sediment interface, and abundant sulfate in water.

The TNRCC, TDH, and TPWD concluded that environmental conditions in most East Texas streams and reservoirs were similar to those of Arkansas and Louisiana and would favor the formation and uptake of organic mercury by aquatic organisms (Sager and Nava, 1994). The TNRCC also realized that its ecosystem-health-based fish tissue monitoring program was not structured to efficiently detect mercury contamination. Large, individual, top predatory fish were not being targeted by the TNRCC for collection in the monitoring program (Table 2). Whole body analyses of composite samples, which tend to dilute individual tissue concentrations, were preferred for monitoring purposes by the TNRCC. Most other southern states were also using ecosystem-health-based monitoring programs similar to Texas' prior to 1994. The sustained use of this fish tissue monitoring strategy by southern states helps to explain why the mercury contamination problem was not discovered earlier. The TDH, which utilizes a human health risk type of monitoring strategy, failed to detect mercury contamination of fish in East Texas due to limited sampling in the area (Table 7).

The initial concern of the TNRCC, TDH, and TPWD, once information became available regarding the behavior of mercury in the aquatic environment, was that a large regional area of the state could possibly be affected. With limited funding available, the TNRCC regional office in Tyler began a study in September 1993 to evaluate mercury concentrations in water, sediment, and fish tissues of selected water bodies in the Cypress and Sabine River Basins in Northeast Texas (Crowe, 1996). This TNRCC special study used a human health risk approach where individual fillets and composites of fillets from predatory fish were analyzed for mercury. In September 1994, the TDH focused a human health risk investigation of fish collected from reservoirs, streams, and bayous located near the Texas-Louisiana border.

The early findings of the TNRCC study, which targeted collection of predatory fish, and the TDH study, which targeted collection of fish with variable feeding habits, were very similar. They both indicated that mercury is usually found in muscle tissues of some predatory fish, and that concentrations increase with increasing size of fish in some water bodies. Results of TDH sampling revealed consistently elevated mercury concentrations in fish from five of 14 water bodies (Table 9). Based on these results a collective consumption advisory was issued by the TDH in November

1995. The advisory recommends limiting consumption of affected fish from Caddo Lake, Big Cypress Creek upstream of Caddo Lake, and Toledo Bend, Sam Rayburn, and B.A. Steinhagen reservoirs (TDH, 1998b).

Table 9

Summary of TDH Fish Sampling, Mercury Consumption Advisories, and Aquatic Life Closures, 1994-1997

Water Body	TNRCC Segment	Date(s) of Collection	Number of Samples	Number of Species	Minimum Hg Level (mg/kg)	Maximum Hg Level (mg/kg)	Mercury Advisory/ Closure Status
Lake Wright Patman *	0302	7/96	22	5	<0.08	0.84	None Issued
Caddo Lake *	0401	8/94-4/95	54	8	0.09	1.6	Consumption Advisory Issued 11/02/1995
Big Cypress Creek *	0402	4/95	19	9	<0.03	1.5	Consumption Advisory Issued 11/02/1995
Lake O' Pines *	0403	8/94-4/95	33	4	<0.01	0.46	None Issued
Toledo Bend Reservoir *	0504	7/94-5/95	84	8	0.09	1.1	Consumption Advisory Issued 11/02/1995
Martin Creek Lake *	0505	11/96-3/97	34	6	<0.04	0.25	None Issued
Brandy Branch Reservoir *	0505	2/97	17	3	0.10	0.38	None Issued
Lake Tawakoni *	0507	5/96-3/97	31	7	0.04	0.48	None Issued
Lake Fork Reservoir *	0512	3/96	32	6	0.04	0.98	None Issued
Neches River Tidal	0601	1/94	10	5	0.09	0.29	None Issued
B.A. Steinhagen Reservoir *	0603	7/94-5/95	61	8	0.13	1.5	Consumption Advisory Issued 11/02/1995
Lake Palestine *	0605	11/96	26	7	<0.05	0.57	None Issued

Table 9 TDH Fish and Shellfish Mercury in Tissue Data Collected 1994-1997 (Continued)

Water Body	TNRCC Segment	Date(s) of Collection	Number of Samples	Number of Species	Minimum Hg Level (mg/kg)	Maximum Hg Level (mg/kg)	Mercury Advisory/ Closure Status
Sam Rayburn Reservoir *	0610	7/94-6/95	94	8	0.08	1.6	Consumption Advisory Issued 11/02/1995
Taylor Bayou *	0702	8/94	20	8	0.22	0.85	None Issued
Trinity River	0805	2/94	21	7	<0.02	0.35	None Issued
Fosdic Lake	0806	8/94	7	3	<0.01	0.46	None Issued
Echo Lake	0806	8/94	8	5	<0.03	0.14	None Issued
French Lake	0806	8/94	4	2	0.23	0.74	None Issued
Cement Creek	0806	8/94	5	2	<0.03	0.51	None Issued
Lake Como	0829	8/94	15	4	0.11	0.26	None Issued
Lake Conroe *	1012	6/96	21	5	<0.09	0.46	None Issued
Clear Creek	1102	11/93	14	8	<0.02	0.32	None Issued
Brazos River Tidal	1201	1/96-5/96	40	9	0.05	0.68	None Issued
Colorado River	1429	3/94	13	1	<1.0	<1.0	None Issued
Twin Lakes	2103	2/97	8	3	<0.06	0.34	None Issued
Rio Grande	2302	3/94	13	6	<0.02	0.15	None Issued
Sabine Lake	2412	1/94	10	4	<0.07	0.244	None Issued
Galveston Bay	2421/39	6/94	10	5	<0.059	0.394	None Issued

Table 9 TDH Fish and Shellfish Mercury in Tissue Data Collected 1994-1997 (Continued)

Water Body	TNRCC Segment	Date(s) of Collection	Number of Samples	Number of Species	Minimum Hg Level (mg/kg)	Maximum Hg Level (mg/kg)	Mercury Advisory/ Closure Status
Lavaca Bay	2453	2/93-5/96	149	8	<0.039	4.42	Aquatic Life Closure Issued 04/29/1988 and Modified 01/13/2000
Brownsville Ship Channel	2494	4/97	24	6	0.05	0.31	None Issued
Gulf of Mexico	2501	7/96-8/97	148	1	0.22	1.6	Consumption Advisory Issued 06/10/1997

* Water bodies in East Texas originally targeted by the TDH for analysis of mercury in fish tissue.

THE TNRCC'S SPECIAL STUDY OF MERCURY BIOACCUMULATION IN SELECTED EAST TEXAS WATER BODIES

Directives of the Study

This special water quality study was accomplished in accordance with the Texas Water Code, Section 26.127 and Section 104(b) of the Federal Clean Water Act. The report is an integral part of the State of Texas Water Quality Strategy published in 40 Code of Federal Regulations (CFR) 35.1551-2 pursuant to Section 303(e) of the Federal Clean Water Act and is utilized for the purposes listed below.

Purposes of the Study

The purposes of this special water quality study were to provide the TNRCC with a valid information source to:

1. improve geographical definition of the area of East Texas affected by mercury contamination in fish;

2. determine cause and effect relationships between field measurements, water chemistry, and sediment chemistry and accumulations of mercury in fish tissue;

3. clarify conditions that are conducive for biological transformation (methylation) of elemental mercury to organic forms; and

4. evaluate retrospective accumulations of mercury in sediment cores and long-lived trees.

Funding for the Study

EPA specifically allocated Regional Discretionary Funds for investigating mercury in east Texas (based on recent evidence of bioaccumulation problems in neighboring Arkansas and Louisiana), and notified the TNRCC that it was eligible for funding under Section 104(b) of the Clean Water Act to carry out this project. The amount of the award was $50,000 (with 5 percent match by the State), for a total project funding amount of $52,632. A quality assurance project plan (QAPP) was completed by the TNRCC and approved by the EPA prior to initiation of sampling (TNRCC, 1996).

METHODS USED IN EVALUATION

Field and laboratory procedures used during the present study are described in detail in the SWQM Program Water Quality Monitoring Procedures Manual (TNRCC, 1999a). Laboratory analyses of water, sediment, fish tissue, sediment cores, and tree cores were conducted using approved EPA methods by the TNRCC Laboratory in Houston, Texas. Parametric coverages, sampling frequencies, and spatial relationships of sampling locations were consistent with the objectives of the study and the QAPP (TNRCC, 1996).

Field Measurements

A Hydrolab Scout™ multiprobe instrument was utilized for monitoring water temperature, pH, dissolved oxygen, and specific conductance. The instrument was calibrated prior to field use and post-calibrated following monitoring activities each day. At each sampling site, the multiprobe sonde was placed directly in the water and allowed to reach equilibrium. Measurements were made in profile at depths of 0.3 m (1 ft) below the surface and at each 1.5 m (5 ft) interval below the surface. Secchi disk transparency was measured directly in-reservoir. The disk was lowered on the shaded side of the boat until it disappeared from view. The disk was then slowly raised until it was visible. The mathematical average of the two measurements was computed and recorded.

Water Chemistry

Samples of near surface water at each sampling site were collected by directly immersing three new clean one liter (quart) cubitainers approximately 0.3 m (1 ft) beneath the water surface. One of the samples was analyzed for chorophyll *a* content. Analysis of suspended solids, nutrients, alkalinity, and salts was conducted on a second sample. Analysis of total organic carbon, ammonia nitrogen, total Kjeldahl nitrogen, and total phosphorus was conducted on the third sample, which was preserved in the field with concentrated sulfuric acid to a pH < 2 standard units. All water samples were immediately placed on ice in a darkened chest where a temperature of < 4 ^{o}C was maintained during shipment to the TNRCC Laboratory.

Metals in Water

Dissolved and total metals samples were collected using clean techniques involving a pump and in-line filter apparatus. All materials (tubing, filter, containers, gloves) used in the procedure were handled with latex-gloved hands and transported to sampling sites in plastic ziplock bags. Once on-site, two personnel using “clean hands, dirty hands” techniques loaded the materials onto a pump-holding apparatus. One end of the tubing was attached to a cartridge filter. A middle portion of the tubing was threaded around the pump head, and the terminal end of the tubing was inserted into a holder on an extension arm of the apparatus to allow sampling beneath the water surface. The peristaltic pump was turned manually and the tubing and filter were flushed with ambient water. The filter was then placed over the mouth of an opened, one liter capacity, high density plastic container and filled. The containers were pre-acidified with 5 mL of metals-grade HNO_3/H_2O preservative in a 1:1 solution. Following collection of the dissolved metals sample, the in-line filter was removed and a total metals-in-water sample was collected in using the same apparatus and tubing. The

containers were returned to plastic ziplock bags and placed on ice for shipment to the TNRCC Laboratory. Aluminum, cadmium, chromium, copper, nickel, and zinc concentrations in water were analyzed using inductively coupled plasma atomic emission spectroscopy (ICP-AES) methods. Arsenic, lead, selenium, and silver concentrations were determined by graphite furnace atomic absorption (GFAA). Mercury concentrations were analyzed using cold vapor atomic absorption (CVAA).

Sediment Samples

Sediment samples were collected using an Ekman dredge. Multiple grabs (3-5) were made at each site. Water overlying the sediment was decanted carefully by tilting the dredge to one side. Each grab sample was deposited into a clean shallow plastic pan by simultaneously opening the jaws of the dredge. The most recently deposited upper portions ($\leq$5 cm; 3 inches) of the grab samples were transferred to the jars with a clean plastic scoop. A composite was produced by transferring equal portions of successive grabs to two detergent-rinsed pint glass jars until they were filled. The jars were sealed with teflon-lined lids, leaving no head space between the sediment and the lids. The samples were placed on ice for shipment to the TNRCC Laboratory. Analysis of metals in sediment was conducted on one jar, while grain size, total organic carbon, and acid volatile sulfide were analyzed from the other. Aluminum, cadmium, chromium, copper, manganese, nickel, silver, and zinc concentrations in sediment were determined by ICP-AES. Arsenic, lead, and selenium concentrations were determined by GFAA, while CVAA was used for mercury analyses.

Fish Tissue Samples

Collection of fish for metals in tissue analysis followed guidance provided by EPA (USEPA, 1993). Largemouth bass were targeted for collection due to its popularity as a sportfish, trophic position as a piscivorous predator, and demonstrated tendency to bioaccumulate mercury. At each reservoir site, the goal of the collection effort was to continue until ten largemouth bass within a size range of 305 to 457 mm (12 to 18 inches) were obtained. This size range encompasses the most common sizes of largemouth bass caught by anglers. Other large predatory fishes (crappie, channel and flathead catfish, freshwater drum, gar, and bowfin) were also collected and considered for analysis, particularly in reservoirs where the target species was difficult to obtain.

Fish were collected by using boat-mounted electrofishing equipment. Netted fish were retained in a live well until sampling was completed. Only live or freshly dead fish (red gills) within the prescribed size range were selected from the catch for submittal to the laboratory. The remaining fish were returned to the water body. The total length of each fish was measured in the field. Each fish was then individually wrapped in aluminum foil (dull side to the fish). All individually foil-wrapped fish within each species were placed in plastic bags, which were taped and labeled before placement on ice for shipment to the TNRCC Laboratory. In the laboratory, a fillet was removed from each individual fish. The muscle tissue was blended to a thickened homogenate. Chromium, copper, lead, and selenium concentrations in tissue were determined by ICP, and CVAA was utilized for mercury analysis. Total mercury was analyzed since more than 95 percent of the total mercury in the muscle tissue of freshwater fish is MeHg (Bloom, 1992).

Sediment Cores

One sediment core was collected at or near the area of maximum depth in each reservoir with a Phleger gravity corer. The corer is essentially a large, hydrodynamically-shaped lead weight (28 kg; 65 lbs). A stabilizing fin assembly attached to the upper portion of the weight minimizes turbulence, vibration, and planing as the corer falls through the water. An internal check valve near the top of the corer opens by flow of water through the corer during descent and closes to prevent washout of the sediment as the corer is raised. At each site, a core catcher was inserted into the bottom of a clean, clear plastic liner 98 cm (38 in) in length. The plastic liner with the attached core catcher was inserted into a galvanized steel coring tube 100 cm (39 in) in length and 37 mm (1.4 in) in internal diameter. A core cutter was then attached to the bottom of the coring tube to hold the plastic liner in place and aid in penetrating the sediment. The coring tube was threaded onto the bottom of the lead weight. The corer was raised and lowered with a manually operated winch. The corer was lowered to six meters (20 ft) above the bottom of each reservoir site. The brake on the winch was released, allowing the corer to free-fall, forcing penetration of the coring tube into the sediment. The leaves of the core catcher opened at the bottom of the plastic liner as the coring tube penetrated the bottom, allowing sediment to enter the plastic tube liner. The leaves were pressed closed by the weight of the trapped sediment as the corer was retrieved to the surface.

At the surface, the coring tube was removed from the lead weight while held in vertical position. The core cutter was removed from the bottom of the coring tube and the plastic liner was removed. Plastic caps were placed over the top and bottom ends of the plastic liner tube. The plastic tubes were held in vertical position and the overlying water was removed with tubing attached to a peristaltic pump. The core catcher at the bottom of the plastic liner was removed and the liner was placed on top of a rubber plunger mounted on a 105 cm (41 in) steel rod. The plastic liner was pulled downward on the plunger to force the sediment core upward in the tube. The sediment core was slowly extruded out the top of the liner into pre-cut 5 cm (1.95 in) sections of liner tube placed on top. A stainless steel spatula was used to cut the sediment core between pieces of the plastic tubing. The cut piece of sediment was then extruded from the 5 cm piece of tubing into a plastic ziplock bag. This process was repeated until all of the sediment core was removed from the liner. The pieces of sediment core were placed on ice for shipment to the TNRCC Laboratory where mercury concentrations were determined by CVAA.

Tree Cores

Bole wood samples were collected from the largest standing eastern red cedar *Juniperus virginiana* trees located in cemeteries throughout East Texas. A 12 mm (0.47 in) diameter, 30 cm (11.7 in) long, three-threaded incremental steel borer was used to extract core samples. The threads and outside of the coring bit were first lubricated with beeswax to facilitate penetration into the tree. The bit was attached to the handle and placed perpendicular to the tree and manually turned clockwise until the desired depth was achieved. A concaved-shaped, steel core extractor sleeve was inserted onto the top of the core between the core and boring bit tube. The borer was then turned one half revolution in a counter clockwise direction to break the core from the tree and position the extractor sleeve under the core. The extractor sleeve was then pulled from the borer with the detached core resting on top. The borer bit was promptly removed from the tree. The cores were placed in plastic

ziplock bags and stored on ice until returned to the laboratory. The cores were frozen until aging and sectioning was done.

Each core was sanded with aluminum oxide paper to highlight growth rings. Cores were aged with the aid of a dissecting microscope. Ten-year sections were cut manually with a steel coping saw. The sections were placed in glass vials and shipped to the TNRCC Laboratory for analysis of total mercury by CVAA.

Statistical Analyses

All statistical analyses were performed using Minitab™ (Minitab, 1999). Regressions of mercury concentrations in tissue and total length of fish were determined for populations of largemouth bass in each of the study reservoirs and lakes. Distributions of all variables used in statistical analyses were tested for normality by inspection of normal probability plots. Plots were performed for all variables with untransformed values and $\log_{10}$ transformations. Variables were transformed when the linearity of the probability plot was improved after transformation. Regressions were used to predict the expected concentrations of mercury (*E*Hg) at a standardized length of 406 mm (16 inches). This length was chosen because it is near the grand mean of total length (396 mm or 15.8 inches), was within the lengths of largemouth bass sampled in most populations, and is a size that most anglers would utilize for consumption. For water bodies in which no significant relationship between total length and mercury in tissue was found, mean mercury concentrations were used (Bodaly et al., 1993).

The effects of field measurements, water chemistry, and sediment chemistry variables on *E*Hg were evaluated through calculation of a correlation matrix of all variables and linear and multiple regression. The best-fit model was selected based on the multiple coefficient of determination (R^2). Independent variables were included only if regression coefficients were significant ($P \leq 0.05$).

STUDY AREA

Water bodies for the present study were selected in coordination with representatives of the TDH, TPWD, and TNRCC regional offices in Tyler and Beaumont to reduce duplication of monitoring effort among agencies and to maximize the number of sites monitored. All of the study sites were located in East Texas, which is the area considered most conducive for formation and uptake of organic mercury by fish (Figure 9). Most of the water bodies are located in the south central plains ecoregion (Pruitt, Ellison Creek, Monticello, Murvaul, Cherokee, Nacogdoches, and Pinkston), two are located in the Texas blackland prairies ecoregion (Pat Mayse and Cooper), two are located in east central Texas plains ecoregion (Houston County and Cedar Creek), and two are located in the south central plains ecoregion (Kimball and Shallow Prong) (Omernik and Gallant, 1987).

The budget allocated to the project allowed sampling of only 13 water bodies. The 13 selected water bodies are hydrologically diverse, representing 11 constructed reservoirs and two natural lakes (Shallow Prong and Pruitt)(Table 10). All of the reservoirs and lakes are used for recreation. Municipal water supply is the primary use for eight of the reservoirs and industrial supply is the primary use for two (Ellison Creek and Monticello)(Table 11). Ellison Creek was the oldest (1943) constructed reservoir sampled, while Lake Cooper (1991) was the youngest. At least one reservoir or lake was sampled in seven of the major river basins that drain East Texas. Sampling was conducted in a series of seven, two or three day efforts between April and November of 1997.

Water bodies for the study were selected primarily to expand the geographical area to the north, west, and south of locations where mercury consumption advisories had been issued by the TDH (Figure 9). Lake Monticello was included due to its location adjacent to a coal-fired electric generating plant. Lake Cherokee was sampled due to nearby coal-fired electric generating plants on Martin Lake (to the south) and Brandy Branch Reservoir (to the northwest) and presence of a gas-fired electric generating plant adjacent to the lake. Ellison Creek Reservoir was included due to the proximity of a secondary steel manufacturing plant. Lake Cooper was selected due to its young age. Several water bodies were selected due to their small sizes and unusual watershed characteristics. For instance, Pruitt and Shallow Prong are natural lakes with very swamp-like headwater regions. Lake Kimball is a very small reservoir which was constructed to encourage adjacent residential development.

Morphometric Features of Study Reservoirs and Lakes

Collectively, the study reservoirs and lakes are morphologically diverse (Table 11). Cedar Creek Reservoir is the largest water body in nearly every aspect with the largest capacity, surface area, length, shoreline length, maximum depth, and drainage area. Pruitt Lake is the smallest in surface area, length, and shoreline length, while Shallow Prong Lake has the smallest drainage area, capacity, and maximum depth. Morphometric features of the other reservoirs and lakes ranged between the sets of extremes characterized by Cedar Creek Reservoir and the two natural lakes (Pruitt and Shallow Prong).

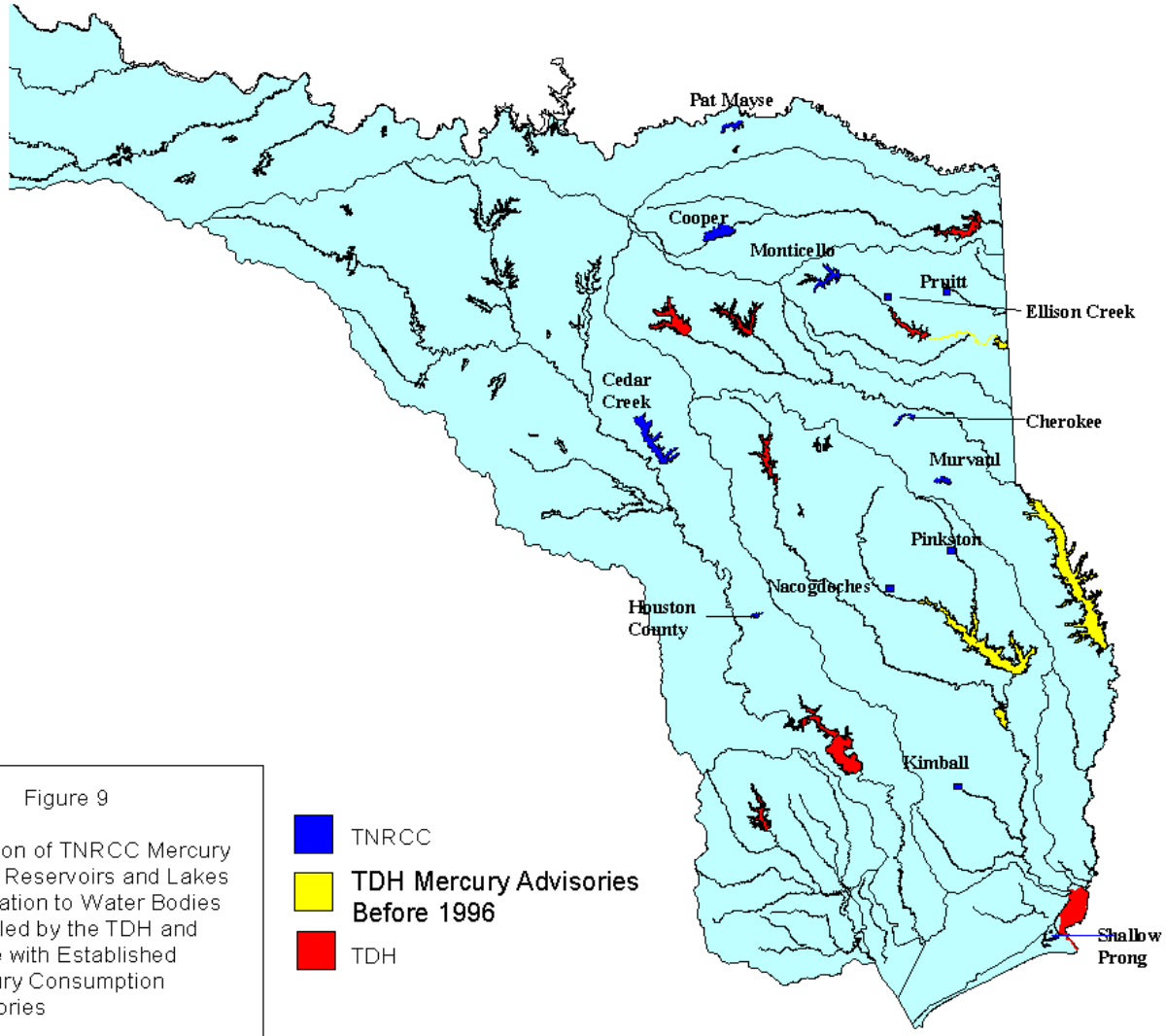


Figure 9

Location of TNRCC Mercury Study Reservoirs and Lakes in Relation to Water Bodies Sampled by the TDH and Those with Established Mercury Consumption Advisories

Table 10

Locations and Sampling Dates of Reservoirs and Lakes

Reservoir of Lake	River Basin	Segment	County	Date Sampled
Pat Mayse	Red	0209	Lamar	10/15/97
Cooper	Sulphur	0307	Delta	10/14/97
Pruitt	Cypress	0402	Cass	08/25/97
Ellison Creek	Cypress	0404	Morris	08/26/97
Monticello	Cypress	0408	Titus	07/29/97
Murvaul	Sabine	0509	Panola	08/27/97
Cherokee	Sabine	0510	Gregg	07/30/97
Kimball	Neches	0608	Hardin	06/11/97
Nacogdoches	Neches	0611	Nacogdoches	11/04/97
Pinkston	Neches	0612	Shelby	11/05/97
Shallow Prong	Neches–Trinity	0701	Jefferson	06/10/97
Houston County	Trinity	0813	Houston	04/30/97
Cedar Creek	Trinity	0818	Henderson	04/29/97

Table 11

Morphometric Features of Mercury Study Reservoirs and Lakes

Reservoir or Lake	Began Operation	Primary Use	Drainage Area (km^2)	Capacity (m^3)	Surface Area (hectares)	Reservoir Length (km)	Shoreline Length (km)	Maximum Depth (m)
Pat Mayse	1967	Municipal	5,993	153,569,505	2,427	14.8	39.9	13.7
Cooper	1991	Municipal	19,280	382,381,900	7,819	51.5	18.7	13.7
Pruitt		Recreation	1,516	2,486,476	156	2.6	5.8	3.7
Ellison Creek	1943	Industrial	2,000	30,467,203	614	6.4	18.7	10.7
Monticello	1973	Industrial	3,720	49,462,949	810	7.7	12.9	12.8
Murvaul	1956	Municipal	3,720	56,512,344	1,547	10.0	27.4	7.6
Cherokee	1948	Municipal	877	57,603,983	1,614	14.2	29.6	9.1
Kimball	1964	Recreation	128	13,287,110	492	3.2	7.1	5.5
Nacogdoches	1977	Municipal	228	50,745,778	895	12.9	23.8	12.2
Pinkston	1977	Municipal	50	9,103,156	212	3.2	10.3	10.1
Shallow Prong		Recreation	25	2,254,584	251	4.5	7.7	1.8
Houston County	1966	Municipal	1,282	24,053,055	519	6.4	19.3	6.7
Cedar Creek	1966	Municipal	25,850	837,786,408	13,669	31.5	130.0	13.7

RESULTS AND DISCUSSION

Field Measurements

Field measurements were made at each site, since they affect many chemical and biological processes. Single measurements made each site represent a very limited snapshot of water quality conditions within the water bodies. These single measurements they cannot be used to evaluate compliance with the TSWQS or determine support of designated uses. The types and quantity of data required to determine standards compliance and use support are described in detail elsewhere (TNRCC, 1995, 1997, and 1999b).

Dissolved oxygen is one of the most important indicators of water quality for aquatic life. Most aquatic organisms depend upon dissolved oxygen to support metabolism. When dissolved oxygen in water falls below 3-5 mg/L, some sensitive species may not survive and/or reproduce. Dissolved oxygen is a particularly sensitive water quality constituent because chemicals present in the water, physical and biological processes, and temperature exert major influences on its availability.

Water temperature is also an important indicator of general water quality, since it directly affects the rates of most chemical and biological processes. Temperature affects the dissolved oxygen content of water (oxygen levels decrease as temperature increases) and influences the rate of photosynthesis by aquatic plants, metabolic rates of aquatic organisms, and sensitivity of aquatic organisms to toxic substances, parasites, and many diseases.

The acidity of water is measured by pH on a scale of 0.0 to 14.0 standard units. A pH measurement of 7.0 indicates neutral conditions; > 7.0, alkaline conditions; and < 7.0, acidic conditions. Most aquatic organisms flourish in water with a pH range of 6.0-9.0. Aqueous pH affects many chemical and biological processes in water and strongly influences toxicity and bioavailability of metals. At low pH, metals become more mobile and available for uptake by aquatic life, and increased metals availability at low pH can become toxic to sensitive aquatic species. Photosynthesis by aquatic plants removes carbon dioxide from water, which often substantially increases pH during daylight hours. Waters enriched with active planktonic algae often exhibit pH values > 8.5.

Conductivity is a measure of the ability of water to pass an electrical current. Conductivity in water is influenced by the presence of inorganic dissolved ions such as chloride and sulfate ions, which carry a negative charge or calcium and magnesium ions, which carry a positive charge. Conductivity is affected by temperature: the warmer the water, the higher the conductivity. For this reason, specific conductance is reported as conductivity at 25 ^{o}C. Specific conductance in lakes and reservoirs is primarily determined by the geology of watersheds that contribute inflow.

Turbidity is a measure of water clarity, or the degree to which suspended in water decreases the passage of light. The Secchi disk provides a convenient method for measuring light penetration, and thus transparency. Turbidity increases water temperature because suspended particles absorb heat. Increased temperatures lower dissolved oxygen concentrations, because oxygen is less soluble in warm water. Turbidity may also result in a more direct decrease in dissolved oxygen, due to the fact that light penetration is required for photosynthesis by aquatic plants, which produce oxygen.

Six of the reservoirs (Pat Mayse, Cooper, Murvaul, Cherokee, Houston County, and Cedar Creek) have been established by the TNRCC as classified segments. These water bodies have been designated for a high aquatic life use which is independently protected by a dissolved oxygen criterion of 5 mg/L (24-hour mean). Due to the absence of available data, the remaining five reservoirs and Shallow Prong Lake are presumed to support a high aquatic life use. For general screening purposes, the 24-hour 5 mg/L dissolved oxygen criterion was established as an instantaneous minimum screening level and is used as a yardstick to gauge general water quality of the study reservoirs and lakes. Instantaneous dissolved oxygen measurements from the mixed surface layer (MSL) at each site were averaged and compared to the 5 mg/L screening level (Table 12).

Dissolved oxygen concentrations in the msls from all 13 study reservoirs and lakes exceeded 5 mg/L (Table 12). At the times of sampling, dissolved oxygen in most of the water bodies was generally mixed, with surface to bottom measurements varying < 3 mg/L. Monticello, Cherokee, Murvaul, and Pat Mayse Reservoirs and Pruitt Lake were strongly stratified with dissolved oxygen concentrations < 1 mg/L in the bottom water. Pinkston Reservoir was weakly stratified with a dissolved oxygen top-to-bottom range near 5 mg/L and bottom concentrations < 2.5 mg/L.

Water quality criteria for water temperature (maximum level) and pH (minimum-maximum range) are established in the TSWQS to safeguard general water quality against the influences of point and nonpoint sources of pollution rather than for protection of a specific use. A surface temperature maximum criterion of 33.8 ^{o}C (93 ^{o}F) is assigned to several of the classified reservoirs in the TSWQS and was used to gauge general thermal conditions in the study reservoirs and lakes. Surface water temperatures from the two lakes and most of the reservoirs were less than the criterion (Table 12). Surface water temperatures were highest for reservoirs with associated industrial uses (Monticello, Ellison Creek, and Cherokee). Surface water temperature was exceptionally high, 40 ^{o}C (104 ^{o}F), in Lake Monticello on the day of sampling. The elevated temperature contributed to observed stressed and dead gizzard shad *Dorosoma cepedianium* in many coves of the reservoir. An unusual period of several cloudy, very calm days preceded the sampling effort and contributed to a build-up of heat in the reservoir. Some of the heat would normally have been dissipated due to surface cooling by prevailing winds.

While most of the reservoirs may become thermally stratified at some point in the summer, at the time of sampling, only Monticello, Cooper, Murvaul, Pat Mayse, and Pinkston exhibited well-developed thermoclines, where water temperature rapidly declined with depth. Vertical temperature profiles from the other reservoirs and two lakes declined gradually over the entire water column, indicating partially mixed thermal conditions.

A pH range of 6.0-9.0 standard units encompasses the normal seasonal variability in most East Texas reservoirs and lakes and was used to gauge general acidity of the water bodies sampled. With the exception of Lake Kimball and Ellison Creek Reservoir, pH measurements within the mixed surface layers of the study reservoirs and lakes indicated slightly alkaline conditions (pH >7.0) (Table 12). The minimum and maximum ranges of pH values measured in vertical profiles were fairly narrow (<1 unit) for most of the reservoirs and lakes. Only Monticello and Murvaul Reservoirs had surface

Table 12

Field Measurement Data from Study Reservoirs and Lakes

Parameter		Reservoir/Lake												
		Cedar Creek	**Houston County**	**Shallow Prong**	**Kimball**	**Monticello**	**Cherokee**	**Pruitt**	**Murvaul**	**Ellison Creek**	**Cooper**	**Pat Mayse**	**Nacogdoches**	**Pinkston**
Date		4/29/97	4/30/97	6/10/97	6/11/97	7/29/97	7/30/97	8/25/97	8/27/97	8/26/97	10/14/97	10/15/97	11/04/97	11/05/97
Time		1300	1250	1500	1255	1445	1415	1815	0900	1425	1505	0955	1415	1545
Total Depth (m)		11.2	5.5	0.8	4.0	11.6	8.5	3.1	6.7	10.1	13.7	13.7	13.1	9.0
Temperature (°C)	Mean	17.2	18.7	27.5	23.8	34.7	26.4	26.6	27.8	29.3	22.6	21.5	18.6	16.6
	Range	16.8-17.5	17.0-19.3	27.2-27.8	22.2-28.4	25.7-40.0	17.8-33.5	25.4-28.2	25.1-29.6	29.0-31.1	22.2-23.1	18.5-22.2	18.4-18.8	13.3-17.7
Dissolved Oxygen (mg/L)	Mean MSL	7.7	7.2	5.1	5.3	7.8	6.9	5.2	10.1	6.6	7.2	5.5	7.9	6.9
	WC Range	6.9-7.8	5.6-7.3	4.3-5.8	4.1-5.3	0.1-8.4	0.2-7.3	0.9-5.2	0.4-10.1	4.4-6.9	6.8-7.6	0.2-5.7	7.6-8.4	2.5-7.4
pH (Std. Units)	Range	7.5-7.8	7.0-7.4	7.3-7.5	4.7-5.3	7.3-8.5	7.0-7.8	6.8-7.4	7.3-8.7	6.5-6.8	7.7-7.8	7.0-7.5	7.8-8.2	7.3-7.9
Specific Conductance (μmhos/cm)	Mean	169	87	319	38	231	105	80	150	136	219	172	93	103
	Range	164-170	87-88	319-319	37-40	222-260	94-138	78-85	136-188	135-137	218-220	160-220	91-94	71-212
Secchi Disk (m)		0.46	1.16	0.43	0.46	0.37	0.85	0.79	0.82	1.22	0.61	0.85	1.80	1.22

MSL = mixed surface layer; WC = water column; shaded cell indicates exceedance of screening level

pH values greater than 8.5 and greater than a one unit change from surface to bottom. On the basis of overall pH values, only Lake Kimball and Ellison Creek Reservoir are characterized as acidic (pH <7.0).

Specific conductance in East Texas reservoirs and lakes is generally lower than other areas of the state due primarily to lower dissolved solids concentrations. With the exception of Shallow Prong Lake, located on the coastal plain, all of the study water bodies had mean water column specific conductance measurements <250 μmhos/cm (Table 12). The two lakes (Kimball and Pruitt) and Houston County and Nacogdoches Reservoirs had mean specific conductance measurements <100 μmhos/cm. The specific conductance measurements in vertical profile indicate density stratification in Monticello, Cherokee, Murvaul, Pat Mayse, and Pinkston reservoirs. For the other reservoirs and two lakes, specific conductance varied little from top to bottom, indicating well-mixed conditions.

Due to turbidity and color normally found in East Texas water, Secchi disk transparency was low (< 0.9 m; <3.0 ft.) in most of the study reservoirs and lakes (Table 12). Transparency was $\geq$1.2 m ($\geq$ 4.0 ft) in Ellison Creek, Pinkston, and Nacogdoches reservoirs.

Water Chemistry

Numeric water quality criteria for nutrients and chlorophyll *a* in water have not been developed by the TNRCC, but their involvement in aquatic plant growth and proliferation warrants their consideration in assessment of water quality. In addition, elevated ammonia nitrogen concentrations are toxic to aquatic life, deplete dissolved oxygen resources through bacterial nitrification, and are frequently indicators of recent sewage pollution. Chlorophyll *a* is the primary photosynthetic pigment and is present in all algae and is often used as an estimate of algal biomass.

The TNRCC and the Clean Rivers Program (CRP) have established screening levels for nutrients and chlorophyll *a* for different water body types based on statewide 85th percentiles from the TNRCC SWQM database (TNRCC, 1999b). Nutrient and chlorophyll *a* screening levels for reservoirs and lakes are shown in Table 13. Although the TNRCC and the CRP have not established screening levels for total (TSS) and volatile suspended solids (VSS), total organic carbon (TOC), alkalinity, hardness, calcium, or magnesium, their 85th percentile values are provided in Table 13 and were used to screen general water quality of the study reservoirs and lakes.

Nutrient concentrations in the study reservoirs and lakes were generally low, with only four isolated exceedances of screening levels (Table 13). In response to the low nutrient concentrations, chlorophyll *a* concentrations were all substantially less than the screening level (22.7 μg/L). Chlorophyll *a* concentrations in six of the 13 water bodies were less than the analytical reporting limit (1 μg/L), and the highest concentration found was in Kimball Reservoir (8 μg/L). Due in part to the low density of phytoplankton, TSS and VSS, and TOC concentrations were also generally low. Only two exceedances of TOC screening level were found (18 mg/L for Shallow Prong Lake and Kimball Reservoir). All study reservoirs and lakes were considered to have low alkalinity since values were substantially less than the 85th percentile (215 mg/L), and all but Cooper Reservoir had less than 50 mg/L. Kimball Reservoir had exceptionally low alkalinity (< 5 mg/L). All of the

Table 13

Water Chemistry Data from Study Reservoirs and Lakes

Parameter (mg/L)	85th Statewide Percentile Screening Level	Reservoir/Lake												
		Cedar Creek	Houston County	Shallow Prong	Kimball	Monticello	Cherokee	Pruitt	Murvaul	Ellison Creek	Cooper	Pat Mayse	Nacogdoches	Pinkston
Ammonia-N	0.11	0.06	0.03	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	0.05	0.14	0.22	0.17
Nitrite + Nitrate-N	0.43	0.299	0.245	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.086	0.09	0.01	<0.1	<0.1
Kjel-N	1.08	0.83	0.69	1.71	0.78	0.69	0.36	0.54	0.87	0.42	0.56	0.60	0.41	0.54
O-Phosphorus	0.09	<0.06	<0.06	<0.06	<0.06	<0.06	<0.06	0.05	<0.06	<0.06	0.03	0.01	<0.06	<0.06
T-Phosphorus	0.18	0.11	0.03	0.09	0.03	0.06	0.05	0.14	0.05	0.02	0.05	0.04	0.01	0.02
Chlorophyll *a*	22.7	<1.0	<1.0	4.89	8.03	5.87	<1.0	7.71	19.7	3.47	<1.0	5.43	<1.0	<1.0
TSS	75	10	4	4	7	2	2	7	7	4	10	3	3	4
VSS	22	2	<1	2	3	2	2	4	5	1	3	2	2	2
Chloride	SS	11.3	11	67	5	19	11	6	12	9	6	4	7.26	5.73
Sulfate	SS	17.4	10	6	4	43	10	11	12	35	11	13	11	5.24
TDS	SS	131	111	194	55	142	73	91	87	108	134	104	82	101
TOC	12.83	7	7	18	18	6	12	9	10	<1	6	6	6	6
Alkalinity	215	42	8	40	<5	32	20	12	30	12	78	46	16	18
Hardness	174	51.2	17.3	43.2	6.9	60.8	28.1	21.4	35.8	45.2	81.9	56.7	28.9	21.5
Calcium	73	15.5	3.2	8.2	1.7	11	6.8	5.2	7.1	11.9	27.9	19.4	5.2	4.1
Magnesium	25	3.1	2.2	5.5	0.6	8.1	2.7	2	4.4	3.8	2.9	2	3.9	2.7

SS = Segment specific criterion; all values expressed as mg/L excepting chlorophyll *a* (μg/L); shaded cell indicates exceedance of screening level

reservoir and lake waters were considered soft, with total hardness concentrations < 100 mg/L, substantially less than the statewide 85th percentile (174 mg/L) for statewide reservoirs. Hardness concentrations were low primarily due to low concentrations of calcium, magnesium, chloride, and sulfate.

Total dissolved solids (TDS) is a measure of inorganic materials dissolved in water. TDS is highly correlated with chloride and sulfate concentrations and specific conductance measurements. Chloride and sulfate occur naturally in water, as the result of leaching from common minerals in the watershed. Chloride, sulfate, and TDS criteria in the TSWQS represent annual averages of all values reported for the entire segment. Instantaneous grab sample data collected during the present study can not be used for determination of standards compliance. However, the criteria were used as yardsticks to gauge the general salt content of the study reservoirs and lakes. Specific chloride, sulfate, and TDS criteria have been established for seven of the study water bodies. For the others, the criteria of the nearest downstream water body were used for comparison purposes.

Chloride, sulfate, and TDS concentrations from the 13 study reservoirs and lakes were less than established criteria and reflect generally low salt content. The highest chloride and TDS concentrations (67 mg/L and 194 mg/L, respectively) were found in Shallow Prong Lake, and the highest sulfate concentration (43 mg/L) occurred in Ellison Creek Reservoir. Kimball Reservoir had the lowest chloride (5 mg/L), sulfate (4 mg/L), and TDS (55 mg/L) concentrations.

Metals in Water

Aquatic organisms require minute quantities of most metals for their metabolism. However, exposure to high concentrations may induce adverse effects. The TSWQS establish acute and chronic criteria for most metals to protect aquatic life from exposure to concentrations that may induce lethal or sublethal effects. Acute and chronic metals criteria were developed by the TNRCC to protect assigned aquatic life uses in the TSWQS. Metals that accumulate in aquatic organisms may potentially cause risks to human consumers. The TSWQS also establish human health criteria (water concentrations) to prevent contamination of drinking water, fish, and other aquatic life and ensure they are safe for human consumption. More than one instantaneous grab water sample is required to determine compliance with acute and chronic aquatic life criteria and the human health criteria (TNRCC, 1999b). Only one grab sample was collected during this study at each site, so comparison to the TSWQS are not warranted. However, the criteria were used as yardsticks to gauge metals concentrations in the study reservoirs and lakes.

Criteria for mercury and selenium are based on total recoverable concentrations due to their ability to bioaccumulate (Table 14). Dissolved fractions for the other metals are used as criteria, since they are generally the more toxic forms. For six of the metals (cadmium, chromium, copper, lead, nickel, and zinc), where the relationship to toxicity is defined as a function of hardness, acute and chronic criteria were computed from long-term, segment-specific, 15th percentile hardness data (TNRCC, 1995).

Most metals concentrations in the study reservoirs and lakes were low, with 117 of 143 (82%) reported as less than minimum analytical limits (MALs) (Table 14). Total selenium and dissolved,

Table 14

Metals in Water Data from Study Reservoirs and Lakes

Metal (μg/L)	Criterion			Reservoir/Lake												
	Acute Aq Life	Chronic Aq Life	Human Health	Cedar Creek	Houston County	Shallow Prong	Kimball	Monticello	Cherokee	Pruitt	Murvaul	Ellison Creek	Cooper	Pat Mayse	Nacogdoches	Pinkston
Aluminum	991	----	----	240	413	219	893	<41	<41	<41	48	<41	<40	<40	<41	<41
Arsenic	360	190	50	<2	<2	<2	<2	2.59	<2	<2	<2	<2	2.38	2.72	<6	<2
Cadmium	*	*	5	<5	<5	<5	<5	<5	<5	<5	5	<5	<5	<5	<5	<5
Chromium	*	*	100	<3	<3	<3	<3	<3	4	<3	<3	<3	<3	<3	<3	<3
Copper	*	*	----	<3	<3	<5	<5	<3	<3	<3	4	<3	<3	<3	<3	<3
Lead	*	*	5	<1	<1	<1	3.75	<1	<1	<1	<1	<1	<1	<1	<1	<1
T Mercury	2.4	1.3	0.0122	0.011	<0.010	0.019	0.013	0.021	0.012	0.013	0.018	<0.010	<0.010	<0.010	<0.010	0.018
Nickel	*	*	----	<11	<11	<11	<11	<11	<11	<11	<11	13	<11	<11	<11	<11
T Selenium	*	*	50	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2	<2
Silver	0.92	----	----	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<2.5	<2.5	<0.5	<0.5
Zinc	*	*	----	<4	<4	6	10	<4	<4	4	<4	<4	<4	5	26	<4

* Criterion a function of hardness (see Texas Surface Water Quality Standards); metals reported as the dissolved fraction except for mercury and selenium; shaded cell indicates exceedance of acute, chronic, or human health criterion.

free-ionic silver were not detected in any of study reservoirs and lakes, and only one detection was found for dissolved cadmium, chromium, copper, lead, and nickel. Total mercury was the most commonly detected metal (8 of 13 water bodies). The levels of total mercury detected were not considered even remotely toxic to aquatic life, since they were substantially less than the acute and chronic criteria (Table 14)(Figure 10). However, total mercury concentrations in six of the study reservoirs and lakes (Shallow Prong and Pruitt Lakes and Kimball, Monticello, Murvaul, and Pinkston reservoirs) exceeded the human health criterion (0.012 μg/L) (Table 14) (Figure 11). The human health criterion is established in the TSWQS as a long-term average concentration, so additional samples would be required to determine if the criterion is violated. Dissolved aluminum and zinc concentrations exceeded MALs in five of 13 water bodies. Samples from Murvaul and Kimball Reservoirs contained the largest number of detected metals (4 of 11 metals). Dissolved lead from Kimball Reservoir (3.75 μg/L) was considered potentially toxic, since the concentration exceeded the aquatic life chronic criterion (0.21μg/L).

Sediment Chemistry

Bottom sediments consist of mineral particles, organic material, and water. Sediment deposits form primarily from the settling of material from the overlying water. Mineral particles include rock fragments and mineral grains that result from natural erosion of terrestrial materials. Mineral components in reservoir sediments are composed primarily of clay, silt, sand, and gravel that enter from tributary streams. Organic matter from decaying or dead aquatic plants and animals usually comprises a small volume of sediment, but the organic fraction is an important component because it regulates the sorption and bioavailability of many contaminants. The spaces between sediment particles are occupied by interstitial water.

Movement of materials into and out of sediments is controlled by physical, chemical, and biological processes. The porosity (volume of spaces between particles) and permeability (ability of water to move between, into, and out of spaces) of sediment are physical factors that largely control movement of materials. Gravels and sands are the most permeable; clays are the least permeable. The coarse fractions ($\geq$ sand) are generally noncohesive and not associated with chemical contamination. The fine fractions (silts and clays) are composed of particles with a relatively large surface-to-volume ratio and surface electric charges that cause these particles to be more chemically and biologically reactive than coarser materials. These physical properties increase the likelihood of sorption and desorption of contaminants. Chemical accumulations are most often associated with fine sediment. In general, sediment-sorbed contaminants are more persistent, less mobile, and occur at higher concentrations than those in the overlying water.

Criteria have not been adopted for the wide array of contaminants in sediment. The EPA has developed procedures to generate criteria for selected toxicants in sediment; however, they have targeted only a few parameters, and criteria have not been adopted. Sediment criteria have not been developed by the TNRCC in the TSWQS. The TNRCC and the CRP have developed sediment screening levels to identify water bodies with elevated sediment metals concentrations (TNRCC, 1999a). Sediment screening levels were derived from long-term monitoring data and represent statewide 85th percentiles for each metal (Table 15). The percentiles indicate concentrations below which 85 percent of concentrations have occurred. For example, if the 85th percentile is 0.87 mg/kg, 85 percent of statewide concentrations were equal to or less than 0.87 mg/kg. Separate screening

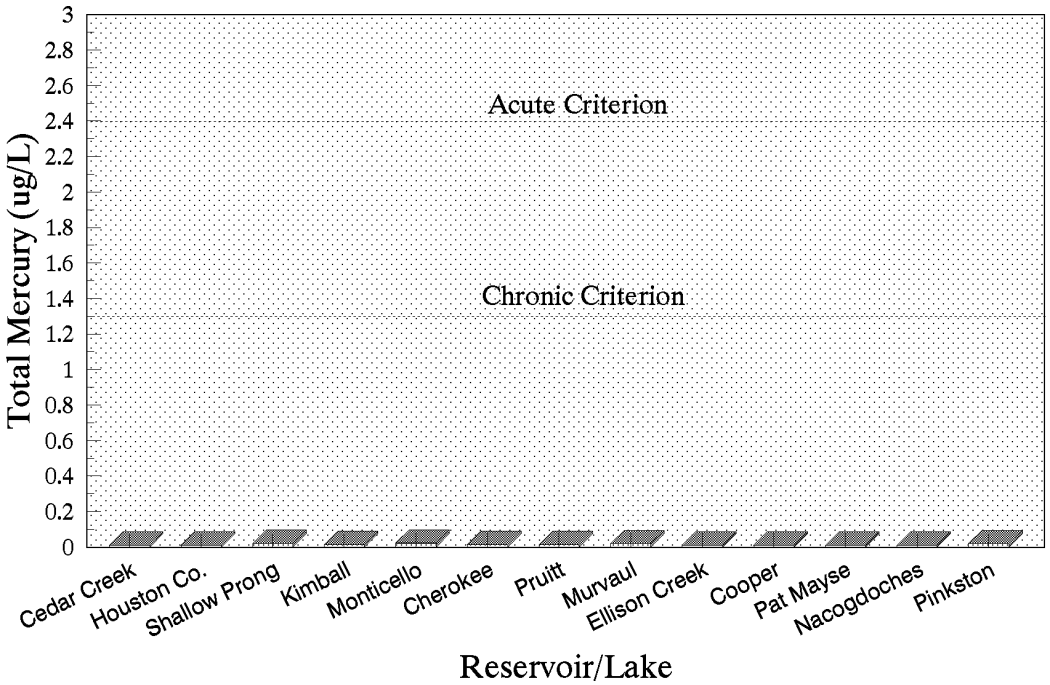
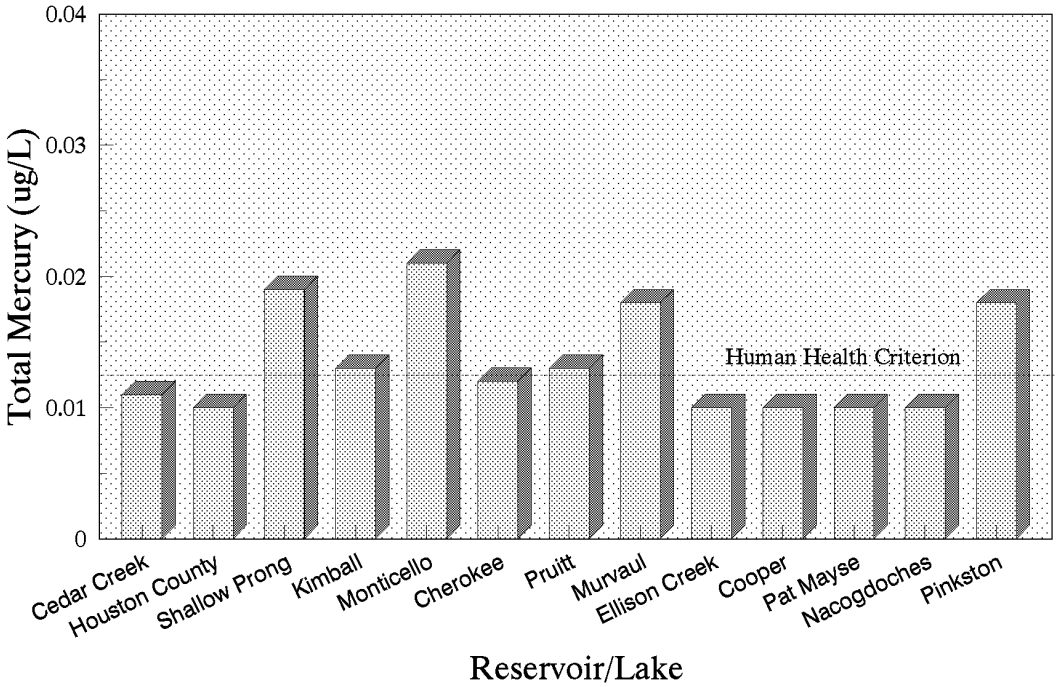


Figure 10. Comparison of Total Mercury in Water Concentrations From 13 East Texas Reservoirs and Lakes Against Acute and Chronic Aquatic Life Criteria



soFigure 11. Comparison of Total Mercury in Water Concentrations From 13 East Texas Reservoirs and Lakes Against the Human Health Criterion for Freshwater

Table 15

Metals in Sediment Data from Study Reservoirs and Lakes

Sediment Metal (mg/kg)	Screening Level TEL/85th/PEL/ERM (mg/kg)	Reservoir/Lake												
		Cedar Creek	Houston County	Shallow Prong	Kimball	Monticello	Cherokee	Pruitt	Murvaul	Ellison Creek	Cooper	Pat Mayse	Nacogdoches	Pinkston
Aluminum	----/28,800/----/----	25,900	4,930	11,200	11,200	28,700	25,300	26,500	26,300	25,400	28,600	19,000	20,900	20,100
Arsenic	5.9/35.4/17.0/70.0	6.86	5.76	4.66	0.79	2.7	39.9	5.73	8.01	32.8	13.5	15.0	90.7	28.0
Barium	----/293.0/----/----	199	62.7	170	79.8	258	345	365	361	262	214	276	241	328
Cadmium	0.59/2.0/3.53/9.6	0.08	0.12	<0.05	0.02	<0.06	<0.06	0.4	0.26	9.9	<0.03	0.14	0.18	0.13
Chromium	37.3/38.0/90.0/370.0	23.3	5.99	18.6	9.85	22	30.6	20.6	23.2	85.3	25.3	22.1	39.5	24.4
Copper	35.7/25.0/197.0/270.0	12.4	2.27	13.2	2.86	24.8	140	10.8	14.6	146	14.4	17.7	10.1	18.6
Lead	35.0/62.2/91.3/218.0	25.4	25.4	35.9	16.6	20	30.7	28.3	30	383	<10.9	<13.4	34.8	26.3
Manganese	----/1,380/----/----	1,070	243	585	119	2,590	1,300	1,060	3,310	2,960	992	1,660	662	1,070
Mercury	0.174/0.194/0.486/0.71	0.072	0.034	0.105	0.074	0.116	0.126	0.164	0.103	0.364	0.033	0.047	0.090	0.124
Nickel	18.0/27.7/35.9/51.6	18	6.38	7.73	3.72	26.6	22.9	19.1	29	82.2	15	18.5	35	26.1
Selenium	----/2.0/----/----	0.66	<0.27	1.78	1.69	<1.22	<6.28	<1.64	<2.59	4.76	3.44	3.77	<2.05	<3.83
Silver	----/1.6/----/3.7	<0.81	<0.36	<1.89	<1.68	<1.75	<0.88	<1.0	<0.91	<3.0	<0.94	<1.15	<1.05	<1.69
Zinc	123.1/144.0/315.0/410.0	66.7	22.4	61.4	24	96.7	167	94.2	86	1,480	65.7	52.8	114	68.7

Shaded cell indicates exceedance of screening level; ----/ indicates absence of screening level

levels are established for freshwater streams, tidal streams, estuaries, and reservoirs. Statewide 85th percentiles do not indicate biological effects (lethal or sublethal), but rather indicate areas where metals concentrations are elevated relative to others in the state for a given water body type.

In addition to statewide percentiles, threshold effects levels (TELs), probable effects levels (PELs) and the effects range-median (ERM) category developed by NOAA to assess potential effects of metals in sediment were also used to screen sediment quality (Long and Morgan, 1991; Long et al., 1995; NOAA, 1999; Kohl, 1999). The freshwater TEL is based on benthic community metrics and toxicity test results. The TEL is calculated as the geometric mean of the 15th percentile concentration of the toxic effects data set and the median of the no-effects data set. The TEL, therefore, represents the concentration below which adverse biological effects are rarely expected to occur. PELs are used to identify compounds which are likely elevated to toxic concentrations. Freshwater PELs are based on benthic macroinvertebrate community metrics and toxicity test results. The PEL, as the geometric mean of the 50th percentile of impacted, toxic samples and the 85th percentile of non-impacted samples, is the level above which adverse biological effects are frequently expected. The ERM category is based on the 50th percentile for toxicity and sublethal effects to aquatic organisms of each metal. Metals concentrations in sediment equivalent to and above the ERM value represent a probable-effects range within which adverse biological effects are expected to occur frequently (Figure 12). Although ERM values were derived from marine sediments, they are often used to evaluate the quality of freshwater sediment. Generally, TELs represent the lowest metals concentrations used for sediment screening purposes, followed in ascending order by the 85th percentiles, PELs, and ERM values (Table 15).

The 13 study reservoirs/lakes contained sediments composed mainly of fine particles, with silts and clays comprising >50 percent of their mass (Table 16). The sediment sample from Houston County Reservoir was the lone exception where sand particles dominated composition. The sediments were hydrous, since water comprised >50 percent of their content. Most of the sediments were highly organic, with TOC concentrations exceeding the statewide 85th percentile in 4 of 13 water bodies. TOC concentrations in sediments from Cedar Creek, Houston County, Monticello and Kimball reservoirs were generally about half the others. The sediments from these four reservoirs were composed of coarser materials (higher sand content).

Since metals tend to accumulate in sediment, they were almost always detected (143 of 169 separate measurements; 85%) in samples from the 13 study reservoirs and lakes (Table 15). Silver was the only metal that was not detected in any of the sediment samples. Aluminum was present in sediments from all the reservoirs and lakes, but the concentrations were less than the only screening level (85th percentile). Metals concentrations in sediment from Houston County, and Kimball reservoirs were less than screening levels for all metals. Only one metal exceeded screening levels in Cedar Creek (arsenic), and Shallow Prong Lake (lead) sediments. The occurrence of elevated metals concentrations in sediment was in general erratic among the study water bodies. Arsenic and nickel exceeded screening levels in eight of the study water bodies, while barium and manganese exceeded screening levels in four. With respect to individual water bodies, sediment from Ellison Creek Reservoir had generally the highest concentrations for most metals, as concentrations for ten metals exceeded screening levels. Cadmium (9.9 mg/kg), lead (383 mg/kg), nickel (82.2 mg/kg), and zinc (1,480 mg/kg) concentrations in Ellison Creek Reservoir sediment exceeded all four

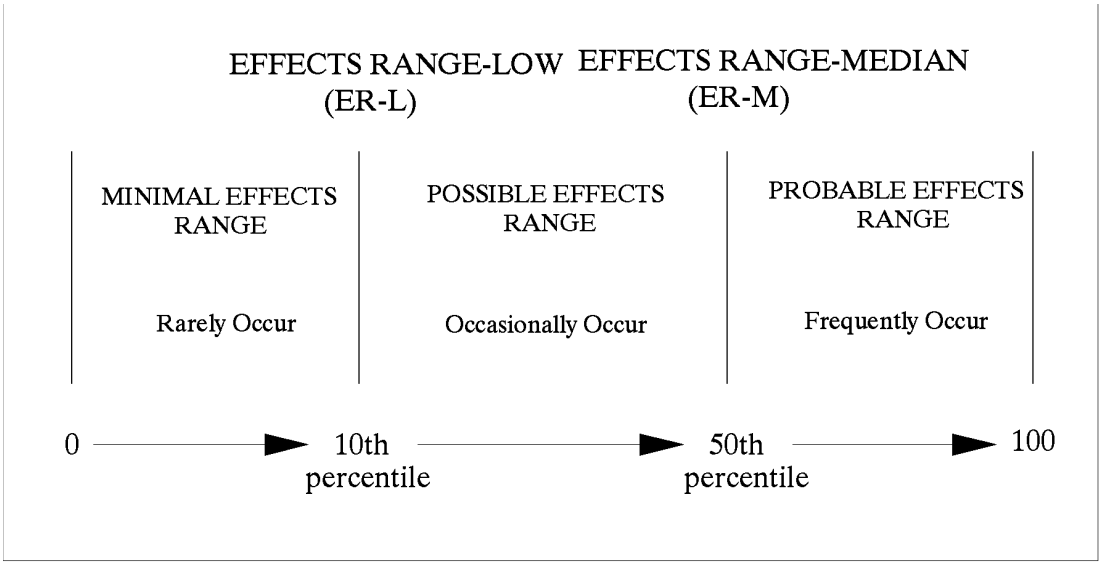


Figure 12. Biological Effects Range of Sediment-Sorbed Contaminants. Compiled by Kohl (1999), after Long and Morgan (1991) and Long et al. (1995)

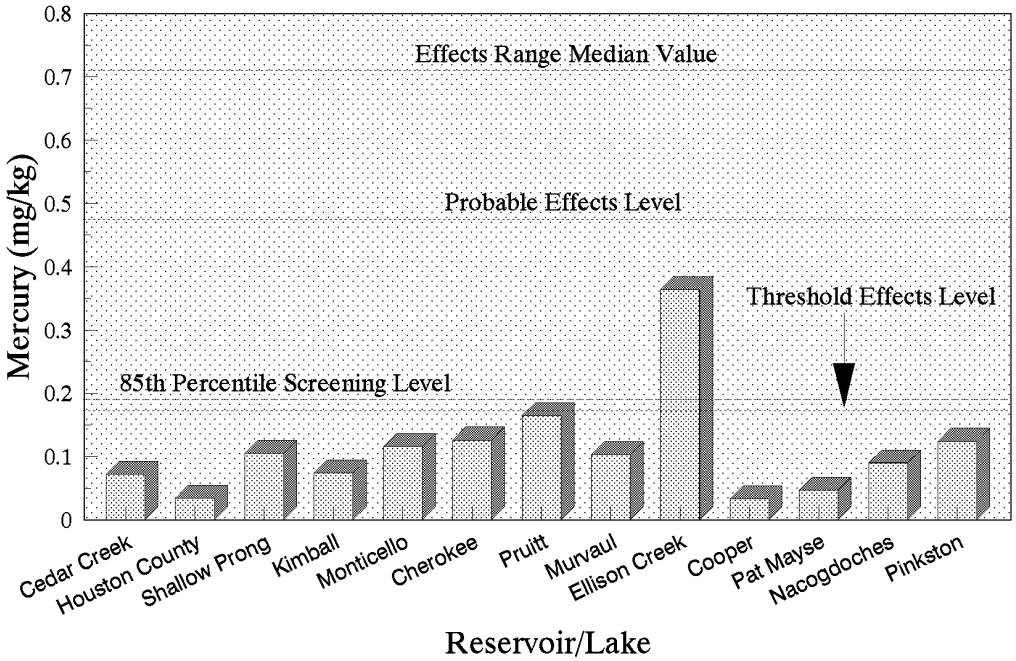


Figure 13. Comparison of Mercury in Sediment Concentrations From 13 East Texas Reservoirs and Lakes Against TEL, 85th Percentile, PEL and ERM Screening Levels

Table 16

Sediment Conventional Parameter Data from Study Reservoirs and Lakes

Parameter	Screening Level	Reservoir												
		Cedar Creek	**Houston County**	**Shallow Prong**	**Kimball**	**Monticello**	**Cherokee**	**Pruitt**	**Murvaul**	**Ellison Creek**	**Cooper**	**Pat Mayse**	**Nacogdoches**	**Pinkston**
% Clay	None	43.1	15.1	47.4	33.0	45.3	23.7	65.0	41.7	47.8	59.1	78.1	65.2	----
% Silt	None	23.6	3.6	47.4	28.3	36.4	53.8	19.2	52.2	38.3	38.5	21.9	34.8	----
% Sand	None	33.3	81.3	5.2	38.7	18.3	22.5	15.8	6.1	13.9	2.4	0	0	----
% Gravel	None	0	0	0	0	0	0	0	0	0	0	0	0	----
SEM (mmol/kg)	None	0.0963	0.331	0.78	0.263	1.13	3.09	0.896	1.27	28.8	0.806	0.873	1.63	1.36
AVS(mmol/kg)	None	3.22	0.93	1.97	0.59	39.2	25.1	11.9	24.5	73.1	15.0	37.1	20.8	392
SEM/AVS	None	0.30	0.36	0.40	0.46	0.03	0.12	0.08	0.05	0.39	0.05	0.02	0.08	0.004
TOC (mg/kg)	55,800	23,200	25,100	125,000	23,500	31,000	45,900	52,800	80,000	73,900	35,400	47,000	46,800	88,300
% Solids	51.5	34.9	46.4	13.8	35.5	18.9	16.8	25.9	18.4	12.7	22.8	14.0	16.8	11.6

SEM = Simultaneously Extracted Metals; AVS = Acid Volatile Sulfide; TOC = Total Organic Carbon; shaded cell indicates exceedance of screening level

screening levels. Despite the elevated sediment metals concentrations, historical water and sediment toxicity testing from Ellison Creek Reservoir and Big Cypress Creek downstream of the reservoir has revealed only rare instances of toxicity (TNRCC, 1999c). Arsenic in sediment (90.7 mg/kg) from Nacogdoches Reservoir was the only other metal that exceeded all four screening levels. Ellison Creek Reservoir was the only water body where mercury in sediment (0.364 mg/kg) exceeded screening levels (Figure 13).

Arsenic in sediment from four reservoirs (Cedar Creek, Murvaul, Cooper, and Pat Mayse), lead in sediment from Shallow Prong Lake, and nickel in sediment from four reservoirs (Monticello, Cherokee, Pat Mayse, and Pinkston) and Pruitt Lake exceeded only the TELs, the lowest of the screening levels (Table 15). The TELs and TNRCC and CRP statewide 85th percentile concentrations were in general agreement in identifying sediments with elevated metals concentrations and/or predicted potential effects. For those metals where both sets of screening levels exist and only the TELs or 85th percentiles were exceeded, elevated concentrations were identified by both sets in 10 of 20 samples. Exceedance of PELs and ERM values, which suggest probable adverse biological effects, was restricted to Cherokee Reservoir (arsenic), Nacogdoches Reservoir (arsenic), Pinkston Reservoir (arsenic), and Ellison Creek Reservoir (arsenic, cadmium, lead, nickel, and zinc).

Analysis of simultaneously extracted metals (SEM) and acid-volatile sulfides (AVS) has been shown to account for pore water concentrations and relative bioavailability of metals in sediments (DiToro et al, 1990). Divalent metals (copper, lead, cadmium, zinc, nickel, mercury, and silver) in sediment would not be predicted to cause toxicity across a wide range of sediments when the concentrations of SEM are lower than the concentration of AVS. In cases where the ratio SEM:AVS is <1, divalent metals in sediment are bound through the formation of insoluble metal-sulfide complexes making them biologically unavailable. All SEM concentrations were lower than AVS concentrations in sediments from the 13 study reservoirs and lakes, suggesting that divalent metals were not bioavailable at the time of sampling (Table 16).

Metals in Fish Tissue

Screening levels for evaluation of metals concentrations in fish tissue were developed by the TNRCC and the CRP from human health criteria (in water) in the TSWQS (TNRCC, 1999b). Screening levels for arsenic, cadmium, chromium, copper, and selenium were developed by the TDH and are slightly lower than concentrations that trigger issuance of consumption advisories. Although the Food and Drug Administration (FDA) action level for mercury in edible fish tissue is 1.0 mg/kg, a more conservative screening criterion (0.7 mg/kg) was used to evaluate concentrations in fish from the study water bodies. The 0.7 mg/kg criterion was derived with the current reference dose (0.1 μg/kg/day) in EPA's Integrated Risk Information System (IRIS) and the fish ingestion rate for freshwaters (10 g/day) specified in the TSWQS.

Concentrations of total metals in individual axial muscle fillets were determined for 137 largemouth bass, 17 channel catfish, 10 white crappie, six freshwater drum, three black crappie, three bowfin, and two spotted bass (Tables 17-29). At least 10 individuals of the target species, largemouth bass, were collected from study water bodies, excepting Cherokee Reservoir and Pruitt Lake, where six were collected. Mean total length of all largemouth bass was 396 mm (15.8 in) over a range in size

Table 17

Metals in Fish Tissue Data from Cedar Creek Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	368	<1.01	<0.101	0.087	0.231	<0.708	0.121	<1.42
	381	<0.99	<0.099	0.11	0.118	<0.693	0.107	<1.39
	394	<1.02	<0.102	0.124	0.149	<0.713	0.111	<1.43
	432	<0.988	<0.099	0.178	0.189	<0.692	0.113	<1.38
	432	<1.01	<0.101	0.061	0.101	<0.706	0.274	<1.41
	445	<1.02	<0.102	<0.061	0.187	<0.714	0.133	<1.43
	451	<1.0	<0.10	0.173	0.177	<0.701	0.151	<1.40
	451	<0.006	<0.10	0.097	0.227	<0.697	0.191	<1.39
	451	1.59	<0.101	0.139	0.185	<0.707	0.213	<1.41
	533	<1.02	<0.102	0.126	0.325	<0.712	0.248	<1.42
Ictalurus punctatus (channel catfish)	356	1.09	<0.102	0.161	0.194	<0.696	0.196	<1.39
	419	1.13	<0.102	0.15	0.332	<0.713	0.048	<1.43
Pomoxis annularis (white crappie)	292	<0.996	<0.10	0.157	0.222	<0.697	0.048	<1.39
	318	1.6	<0.102	0.157	0.208	<0.713	0.074	<1.43
	356	<1.01	<0.101	0.178	0.179	<0.710	0.137	<1.42

Table 18

Metals in Fish Tissue Data from Houston County Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	368	1.11	<0.102	0.187	0.248	<0.668	0.149	<1.34
	381	1.21	<0.102	0.17	0.294	<0.713	0.249	<1.43
	394	1.73	<0.098	0.166	0.264	<0.685	0.468	<1.37
	406	1.53	<0.098	0.18	0.549	<0.688	0.307	<1.38
	406	1.26	<0.106	0.158	0.353	<0.739	0.409	<1.48
	432	<1.03	<0.103	0.167	0.286	<0.719	0.320	<1.44
	451	<1.03	<0.103	0.133	0.355	<0.722	0.505	<1.44
	451	<0.998	<0.10	0.193	0.213	<0.699	0.411	<1.40
	457	1.55	<0.101	0.153	0.347	<0.706	0.582	<1.41
	470	1.62	<0.102	0.195	0.161	<0.677	0.335	<1.35
	483	1.32	<0.10	0.141	0.464	<0.699	0.332	<1.40
	495	<1.01	<0.101	0.141	0.548	<0.710	0.634	<1.42

Table 19

Metals in Fish Tissue Data from Shallow Prong Lake

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	312	1.57	<0.103	0.132	0.349	<0.722	0.196	<1.44
	318	1.07	0.099	0.13	0.259	0.696	0.221	<1.39
	375	<1.04	0.128	0.065	0.211	<0.728	0.375	<1.46
	381	<0.998	0.102	0.087	0.183	<0.699	0.264	<1.40
	381	<0.971	0.157	0.111	0.234	<0.680	0.416	<1.36
	381	<1.10	0.148	0.105	0.265	<0.768	0.268	<1.54
	419	1.48	<0.098	0.083	0.142	<0.688	0.576	<1.38
	432	<1.01	<0.101	0.085	0.195	<0.707	0.362	<1.41
	457	<1.01	<0.101	0.127	0.185	<0.706	0.347	<0.141
	460	1.14	<0.098	0.151	0.514	<0.689	0.429	<1.38
Ictalurs punctatus (channel catfish)	349	<0.996	0.108	<0.060	0.173	<0.697	0.126	<1.39
	419	1.06	<0.103	0.095	0.228	<0.719	0.067	<1.44
	445	<0.994	0.114	0.147	0.221	<0.696	0.107	<1.39
Pomoxis annularis (white crappie)	241	1.52	<0.10	0.12	0.164	<0.70	0.264	<1.40
	292	<0.990	<0.099	0.13	0.22	<0.693	0.466	<1.39
	279	<1.01	0.102	0.105	0.127	<0.710	0.394	<1.42

Table 19 (Continued)

Metals in Fish Tissue Data from Shallow Prong Lake

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Pomoxis annularis (white crappie)	279	<1.04	0.116	0.096	0.21	<0.726	0.267	<1.45
Aplodinotus grunniens (freshwater drum)	325	1.09	<0.102	0.123	0.246	<0.714	0.110	<1.43
	330	1.34	<0.101	0.199	0.245	<0.710	0.125	<1.42
	371	1.32	0.188	0.14	0.187	<0.714	0.100	<1.43

Table 20

Metals in Fish Tissue Data from Kimball Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	330	<0.981	<0.098	0.126	0.275	<0.686	1.02	<1.37
	337	<0.996	0.109	0.111	0.25	0.793	0.811	<1.39
	337	2.06	<0.104	0.122	0.221	<0.726	1.11	<1.45
	349	1.45	0.11	0.1	0.262	<0.678	1.10	<1.36
	356	1.42	<0.102	0.083	0.158	<0.716	1.03	<1.43
	356	<0.996	0.164	0.147	0.366	<0.697	0.917	<1.39
	368	<1.01	<0.101	0.088	0.225	<0.708	1.10	<1.42
	375	<1.02	<0.102	0.153	0.167	<0.714	0.931	<1.43
	375	1.46	<0.099	0.133	0.192	<0.696	1.30	<1.39
	419	1.97	<0.101	0.099	0.293	<0.707	1.05	<1.41
	635	<0.095	<0.098	0.148	0.305	<0.689	2.10	<1.38

Table 21

Metals in Fish Tissue Data from Monticello Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	324	<0.992	<0.099	0.075	0.305	<0.694	0.099	1.69
	324	<0.990	<0.099	<0.059	0.496	<0.693	0.104	1.51
	343	<1.05	<0.105	0.139	0.268	<0.735	0.126	1.97
	349	<1.05	<0.101	0.096	0.32	<0.707	0.121	1.7
	394	1.14	<0.101	0.071	0.371	<0.704	0.154	1.41
	413	<1.07	0.107	0.106	0.197	<0.748	0.107	1.69
	417	<0.979	<0.098	0.088	0.198	<0.685	0.124	1.64
	424	1.37	<0.10	0.118	0.273	<0.70	0.121	1.63
	451	<0.988	<0.099	0.061	0.191	<0.692	0.329	1.84
	488	<1.04	<0.104	0.094	0.395	<0.731	0.215	1.72
	514	<1.05	<0.105	0.089	0.212	<0.732	0.166	1.71
	521	<1.02	<0.102	0.166	0.304	<0.711	0.358	2.01
Ictalurus punctatus (channel catfish)	381	1.15	<0.106	0.1	0.335	<0.742	0.037	<1.48
	457	<1.0	<0.10	0.081	0.336	<0.703	0.046	<1.41
	464	1.06	<0.10	0.326	0.184	<0.697	0.034	<1.39
	483	<0.983	<0.098	0.074	0.32	<0.688	0.038	<1.38
	521	<1.03	<0.103	0.098	0.245	<0.722	0.036	<1.44

Table 22

Metals in Fish Tissue Data from Cherokee Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	305	1.0	<0.10	<0.06	0.282	0.758	0.098	<1.40
	330	<1.0	<0.10	<0.06	0.156	<0.7	0.213	1.64
	376	<0.980	<0.098	<0.059	<0.059	<0.686	0.183	<1.37
	391	<1.02	<0.106	<0.061	0.129	<0.714	0.249	<1.43
	391	<1.0	<0.10	<0.06	0.214	<0.7	0.165	1.46
	472	<0.995	<0.133	0.063	0.106	1.13	0.308	1.74

Table 23

Metals in Fish Tissue Data from Pruitt Lake

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	248	<1.02	< 0.102	<0.061	0.193	<0.711	0.945	<1.42
	299	<1.01	<0.101	0.071	0.663	<0.710	0.582	<1.42
	299	<1.02	<0.102	0.104	0.303	<0.717	0.694	<1.43
	311	<0.973	<0.097	0.107	0.318	<0.681	0.766	<1.36
	318	<0.998	<0.10	0.115	0.348	<0.699	0.874	<1.40
	330	<0.979	<0.098	0.108	0.361	<0.685	0.758	<1.37
Micropterus punctulatus (spotted bass)	292	1.41	<0.10	0.127	0.232	<0.700	0.817	<1.40
	324	<0.994	<0.099	<0.060	0.273	<0.696	0.826	<1.39
Ictalurus punctatus (channel catfish)	394	<0.992	<0.120	0.067	0.136	<0.694	0.202	<1.39
Aplodinotus grunniens (freshwater drum)	533	1.12	<0.099	0.136	0.801	<0.690	0.794	<1.38
	546	<0.967	< 0.097	<0.058	0.327	<0.677	1.53	<1.35
	749	<1.02	<0.102	<0.061	0.584	<0.717	1.34	<1.43
Amia calva (bowfin)	426	<1.01	<0.101	0.103	0.224	<0.708	0.383	<1.42
	445	1.02	<0.101	0.097	0.276	<0.704	0.412	<1.41
	540	<0.998	<0.10	0.096	0.253	<0.699	0.405	<1.40

Table 24

Metals in Fish Tissue Data from Murvaul Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	292	<1.04	<0.104	0.096	0.25	<0.725	0.106	<1.45
	305	<1.01	<0.101	0.074	0.106	<0.706	0.099	<1.41
	311	<1.02	<0.102	0.061	0.182	<0.711	0.149	<1.42
	324	1.27	<0.10	0.124	0.274	<0.699	0.125	<1.40
	356	<1.01	<0.101	0.068	0.074	<0.704	0.160	<1.41
	413	<0.981	<0.098	0.064	0.374	<0.686	0.146	<1.37
	419	<1.02	<0.102	0.072	0.281	<0.716	0.321	<1.43
	438	1.8	<0.102	<0.061	0.237	<0.714	0.224	<1.43
	438	<0.954	<0.095	<0.057	0.262	<0.668	0.215	<1.34
	443	<0.971	<0.097	<0.058	0.133	<0.680	0.314	<1.36
	469	<1.02	<0.102	0.071	0.257	<0.711	0.240	<1.42
	508	1.23	<0.103	0.096	0.218	<0.722	0.229	<1.44
	527	1.89	<0.099	0.177	0.262	<0.694	0.330	<1.39
Ictalurus punctatus (channel catfish)	362	<1.01	<0.101	<0.060	0.170	<0.706	0.071	<1.41
	413	<1.03	<0.12	0.089	0.228	<0.722	0.045	<1.44
Pomoxis nigromaculatus (black crappie)	267	<0.979	<0.098	<0.058	0.206	<0.685	0.096	<1.37
	269	< 0.981	< 0.098	< 0.059	0.124	<0.686	0.046	< 1.37
	279	1.15	<0.13	<0.061	0.117	<0.708	0.059	<1.42

Table 25

Metals in Fish Tissue Data from Ellison Creek Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	318	<0.992	<0.099	<0.060	<0.262	<0.694	0.065	<1.39
	324	<1.02	<0.102	0.098	0.184	<0.717	0.071	<1.43
	343	1.19	<0.098	<0.059	0.187	<0.689	0.150	<1.38
	343	<1.03	<0.103	0.160	0.359	<0.722	0.072	<1.44
	381	1.43	<0.101	0.137	0.234	<0.707	0.073	<1.41
	406	1.76	<0.098	0.108	0.238	<0.688	0.095	<1.38
	426	<0.986	<0.099	0.142	0.603	<0.800	0.102	<1.38
	432	1.11	<0.10	<0.060	0.179	<0.699	0.096	<1.40
	445	1.22	<0.098	0.108	0.289	<0.685	0.120	<1.37
	489	<0.996	<0.100	<0.060	0.178	<0.697	0.043	<1.39
	495	<0.979	<0.098	0.066	0.134	<0.685	0.114	<1.37
	498	<1.02	<0.102	<0.061	0.183	<0.717	0.054	<1.43
Ictalurus punctatus (channel catfish)	508	<0.994	<0.099	<0.060	0.157	<0.696	0.023	<1.39
	533	<1.01	<0.101	<0.061	0.149	<0.707	0.040	<1.41
	546	<1.03	<0.103	<0.062	0.120	<0.723	0.051	<1.45

Table 26

Metals in Fish Tissue Data from Cooper Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	305	<1.06	<0.106	0.125	0.315	<0.739	0.073	<1.48
	311	1.09	<0.100	0.062	0.352	<0.701	0.074	<1.40
	318	<1.02	<0.102	<0.061	0.27	<0.717	0.081	<1.43
	324	<0.994	<1.39	0.085	0.253	<0.696	0.127	<1.39
	330	1.13	<0.097	0.082	0.263	<0.676	0.099	<1.35
	356	<0.983	<0.098	0.088	0.337	<0.688	0.102	<1.38
	362	<1.04	<0.104	0.093	0.393	<0.728	0.102	<1.46
	368	1.11	<0.105	0.100	0.195	<0.737	0.158	<1.47
	419	<1.02	<0.102	<0.061	0.495	<0.717	0.174	<1.43
	470	1.69	<0.097	0.059	0.221	<0.676	0.333	<1.35
Ictalurus punctatus (channel catfish)	394	<1.04	<0.104	0.065	0.265	<0.725	0.073	<1.45
	457	<1.01	<0.101	0.103	0.316	<0.710	0.071	<1.42
	483	<1.02	<0.102	0.065	0.344	<0.717	0.144	<1.43
	508	<0.969	<0.097	0.082	0.271	<0.678	0.229	<1.36

Table 27

Metals in Fish Tissue Data from Pat Mayse Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	305	<0.971	<0.097	0.101	0.504	<0.680	0.095	<1.36
	311	<0.981	<0.098	0.097	0.379	<0.686	0.083	<1.37
	324	<1.06	<0.106	<0.064	0.335	<0.742	0.132	<1.48
	343	<1.04	<0.104	0.093	0.229	<0.728	0.131	<1.46
	356	<1.05	<0.105	0.084	0.432	<0.732	0.144	<1.46
	356	<1.01	<0.101	0.077	0.357	<0.710	0.158	<1.42
	362	<1.04	<0.104	0.069	0.311	<0.728	0.137	<1.46
	381	<0.942	<0.094	0.076	0.339	<0.659	0.153	<1.32
	400	<1.02	<0.102	0.077	0.251	<0.714	0.127	<1.43
	419	<1.06	<0.106	0.105	0.307	<0.740	0.145	<1.48

Table 28

Metals in Fish Tissue Data from Nacogdoches Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides (largemouth bass)	368	1.08	<0.098	<0.059	0.502	<0.686	0.283	<1.37
	368	<1.04	<0.104	0.084	0.250	<0.729	0.309	<1.46
	381	1.76	<0.104	<0063	0.643	<0.731	0.348	<1.46
	381	1.14	<0.105	0.066	0.420	<0.753	0.251	<1.47
	406	1.12	<0.101	0.072	0.405	<0.708	0.303	<1.42
	406	1.09	<0.106	0.076	0.430	<0.739	0.328	<1.48
	413	1.15	<0.100	0.093	0.490	<0.703	0.413	<1.41
	419	<1.04	<0.104	0.084	0.526	<0.731	0.453	<1.46
	419	<0.988	<0.099	0.079	0.537	<0.692	0.322	<1.38
	445	<0.973	<0.097	0.074	0.498	<0.681	0.315	<1.36
	470	<0.953	<0.095	0.093	0.198	<0.667	0.586	<1.33
	483	<1.03	<0.103	0.083	0.503	<0.722	0.317	1.47

Table 29

Metals in Fish Tissue Data from Pinkston Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Micropterus salmoides	311	1.74	<0.097	0.082	0.31	<0.678	0.344	1.4
	318	1.62	<0.094	0.075	0.323	<0.658	0.347	<1.32
	356	<0.938	<0.094	0.059	0.398	<0.657	0.398	<1.31
	368	<1.05	<0.105	0.084	0.259	<0.732	0.382	<1.46
	394	1.33	<0.094	0.087	0.594	<0.658	0.497	<1.32
	419	0.967	<0.097	0.294	0.294	<0.677	0.598	<1.35
	413	<0.988	<0.099	0.062	0.276	<0.692	0.413	<1.38
	419	<0.988	<1.38	0.092	0.276	<0.692	0.476	<1.38
	419	<0.979	<0.098	<0.059	0.242	<0.685	0.377	<1.37
	432	<1.06	<0.106	0.085	0.255	<0.743	0.401	<1.49
	470	<1.02	<0.102	<0.061	0.228	<0.713	0.556	<1.43
	470	<1.04	<0.104	0.092	0.314	<0.725	0.471	<1.45
	527	<0.981	<0.098	0.062	0.251	<0.686	0.810	<1.37
Ictalurus punctatus (Channel catfish)	406	<0.979	<0.098	0.070	0.305	<0.685	0.064	<1.37
	470	0.998	<0.100	0.076	0.295	<0.697	0.057	<1.39
	546	<1.02	<0.102	0.086	0.284	<0.711	0.074	<1.42
Pylodictus olivaris (Flathead catfish)	381	<1.0	<0.100	<0.060	0.256	<0.701	0.223	<1.40
	413	<0.998	<0.100	<0.060	0.263	<0.699	0.179	<1.40

Table 29 (Continued)

Metals in Fish Tissue Data from Pinkston Reservoir

Fish Species	Size (mm)	Metal (mg/kg wet weight)						
		Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Selenium**
		3.0	**0.5**	**100**	**40**	**1.25**	**0.7**	**2.0**
Pylodictus olivaris (Flathead catfish)	470	<0.967	<0.097	<0.058	0.221	<0.677	0.193	<1.35
	591	<1.07	<0.107	<0.064	0.169	<0.746	0.160	<1.49

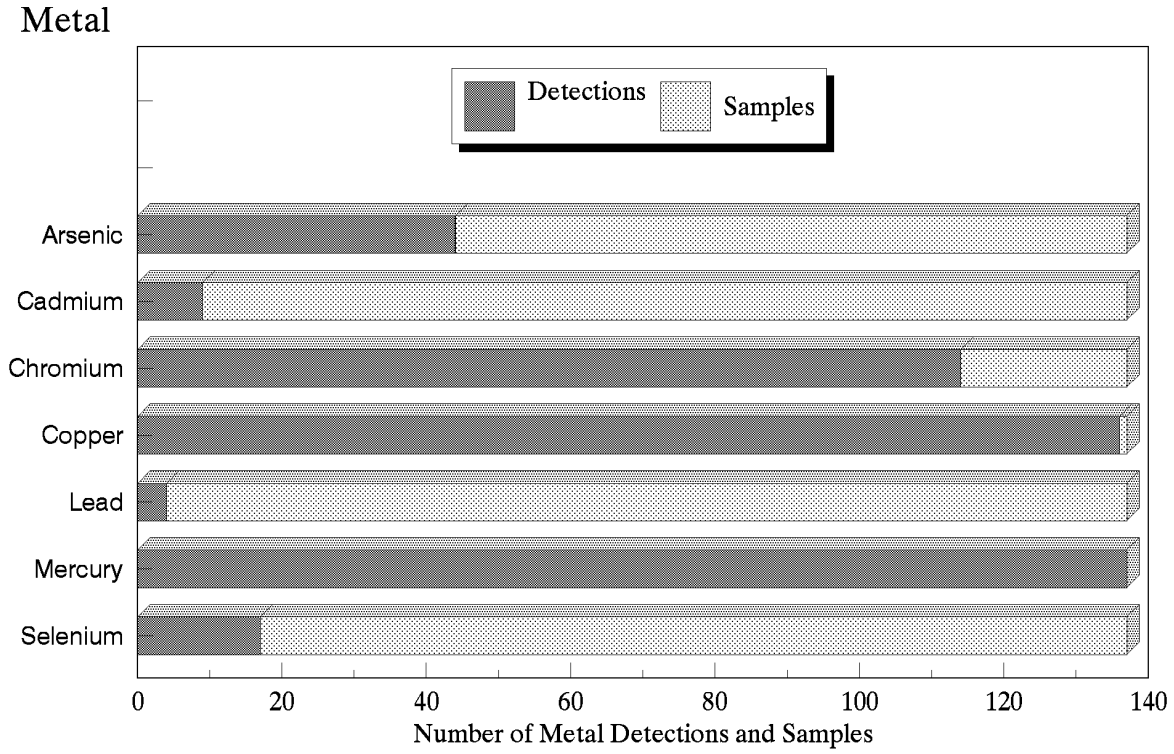


Figure 14. Numbers of Detections for Seven Metals in 137 Largemouth Bass Filet Samples From 13 East Texas Reservoirs and Lakes

of 248 to 635 mm (10 to 25 in). The range in largemouth bass total lengths was greatest (305 mm; 12.2 in) for Kimball Reservoir and lowest (82 mm; 3.3 in) for Pruitt Lake.

Of the seven metals analyzed in largemouth bass edible tissues, arsenic (44/137; 32%), cadmium (91/137; 7%), lead (4/137; 3%), and selenium (17/137; 12%) were not commonly detected (Figure 14)(Tables 17-29). Mercury was detected in all (137/137; 100%) largemouth bass tissue samples, and chromium (114/137; 83%) and copper (136/137; 99%) were also commonly detected. With the exception of selenium and mercury, metals concentrations in the 137 largemouth bass samples were less than screening levels. Occurrence of quantifiable selenium in largemouth bass tissue was common in Monticello and Cherokee reservoirs, two reservoirs associated with electric power generation (Tables 21 and 22). Only one largemouth bass sample, from Monticello Reservoir, had a selenium concentration (2.01 mg/kg) that exceeded the 2.0 mg/kg screening level.

Mercury was the only metal that exceeded the screening level (0.7 mg/kg) with some regularity (16/137; 12%) in all fish species (Tables 17-29; Figures 15-27). However, only largemouth bass from Kimball, and Pinkston reservoirs and Pruitt Lake exceeded the screening level (Figures 18, 21, and 27). All 11 largemouth bass collected from Kimball Reservoir and four of six collected from Pruitt Lake exceeded the screening level (Tables 20 and 23; Figures 18 and 21). In Pinkston reservoir, only the largest individual bass had an elevated mercury concentration (Table 29; Figure 27).

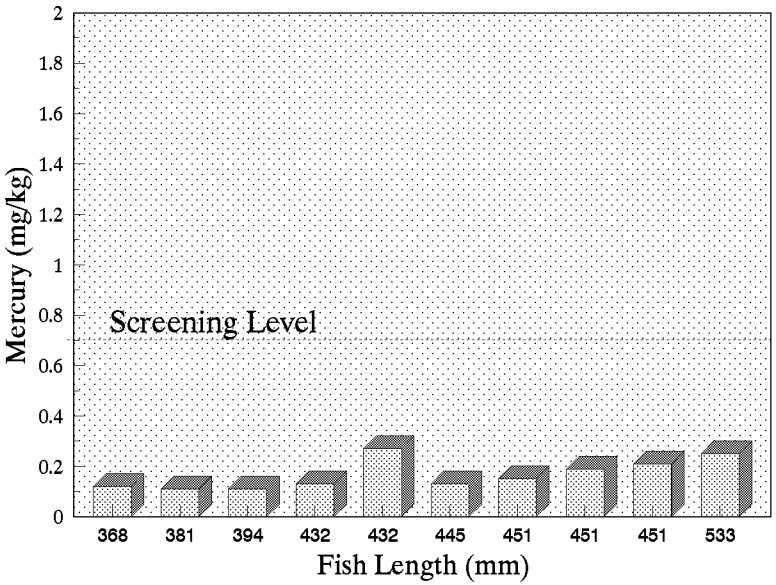


Figure 15. Mercury in Muscle Tissue of Largemouth Bass from Cedar Creek Reservoir

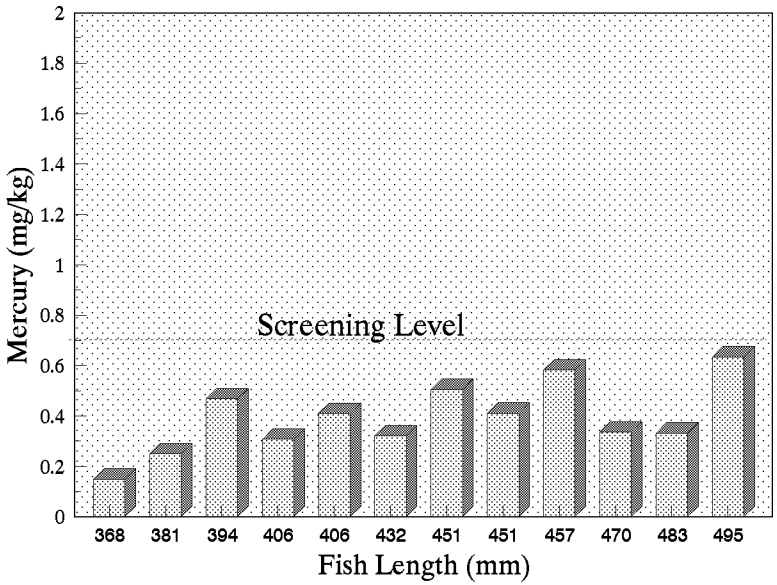


Figure 16. Mercury in Muscle Tissue of Largemouth Bass from Houston County Reservoir

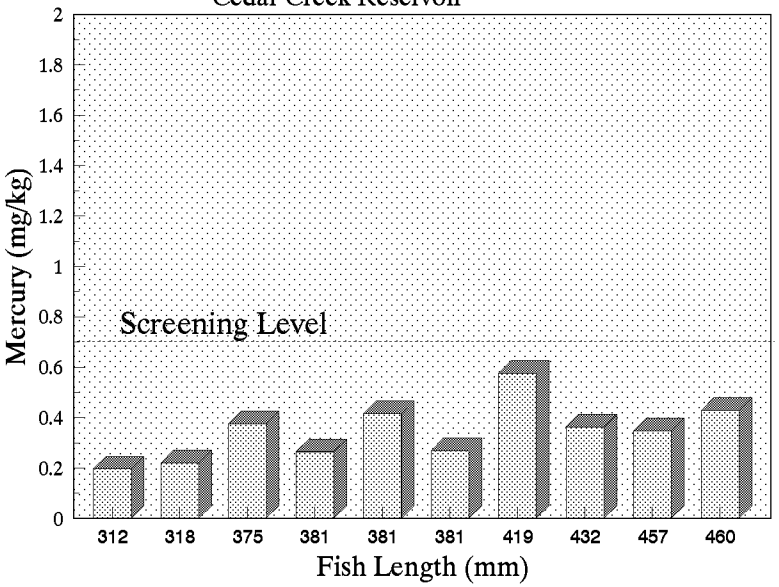


Figure 17. Mercury in Muscle Tissue of Largemouth Bass From Shallow Prong Lake

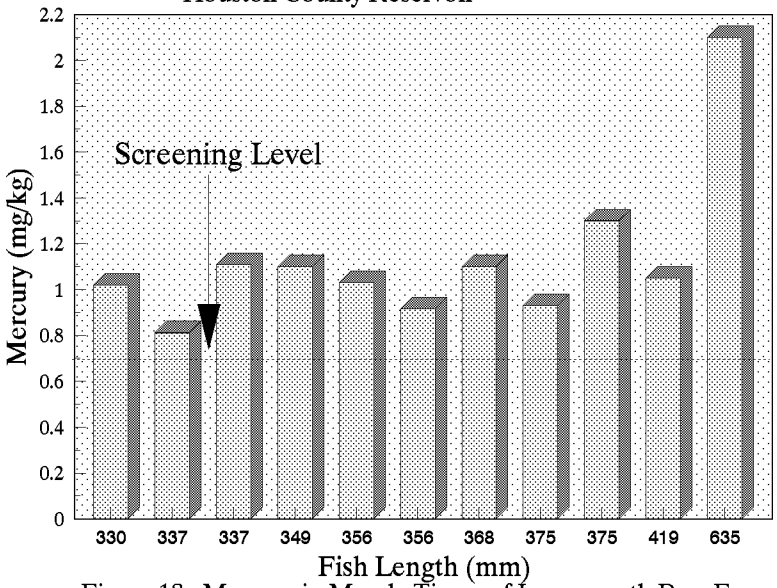


Figure 18. Mercury in Muscle Tissue of Largemouth Bass From Kimball Reservoir

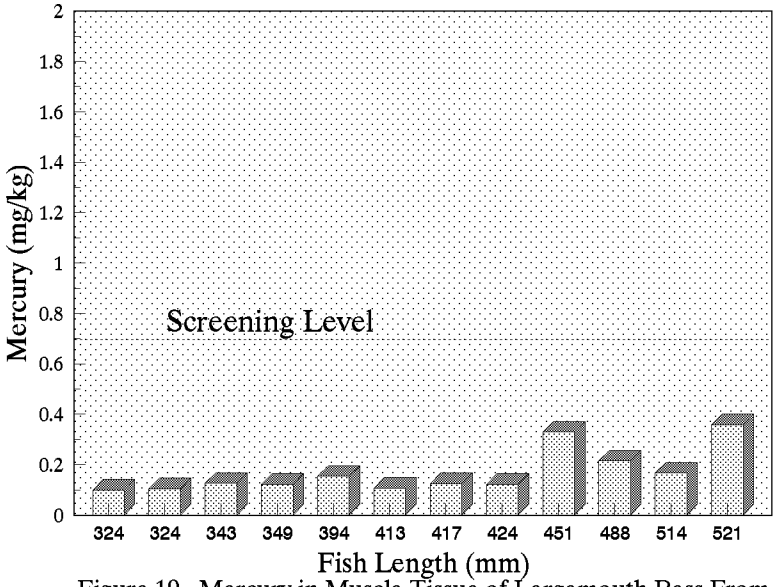


Figure 19. Mercury in Muscle Tissue of Largemouth Bass From Monticello Reservoir

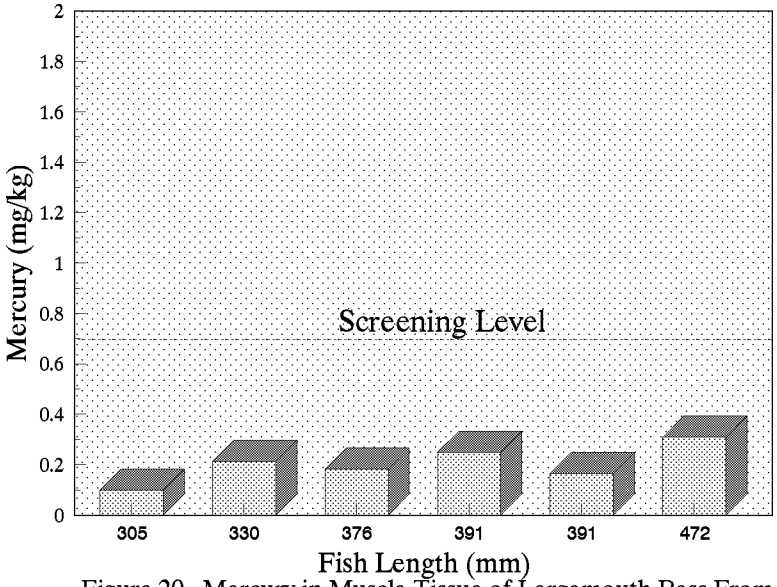


Figure 20. Mercury in Muscle Tissue of Largemouth Bass From Cherokee Reservoir

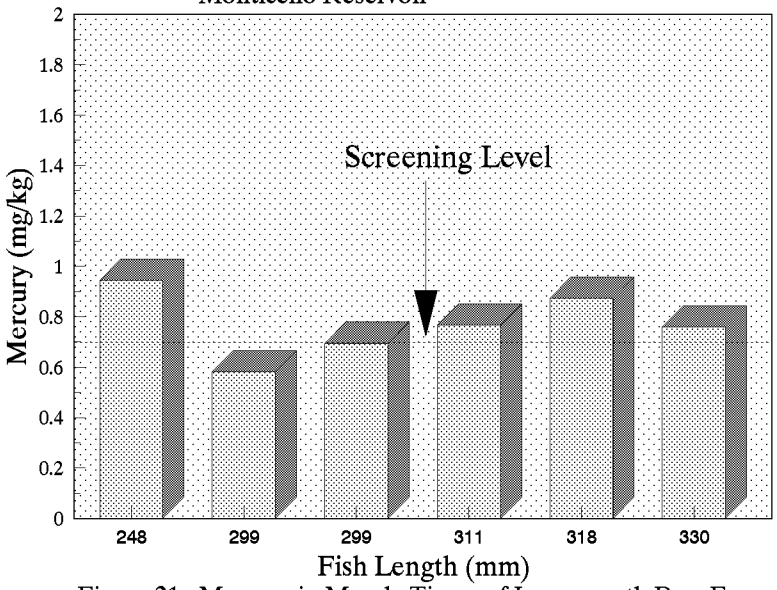


Figure 21. Mercury in Muscle Tissue of Largemouth Bass From Pruitt Lake (Black Cypress Bayou)

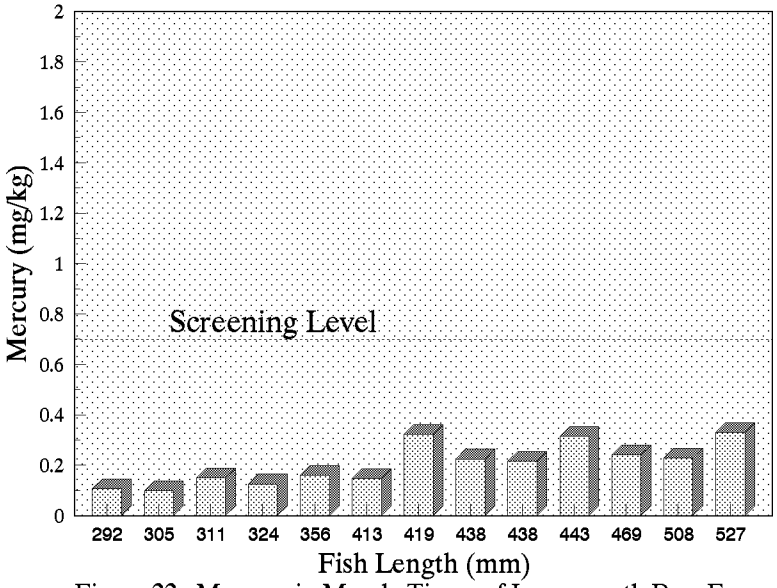


Figure 22. Mercury in Muscle Tissue of Largemouth Bass From Murvaul Reservoir

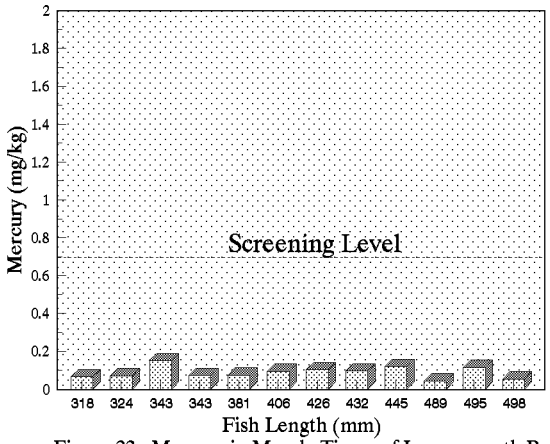


Figure 23. Mercury in Muscle Tissue of Laregemouth Bass From Ellison Creek Reservoir

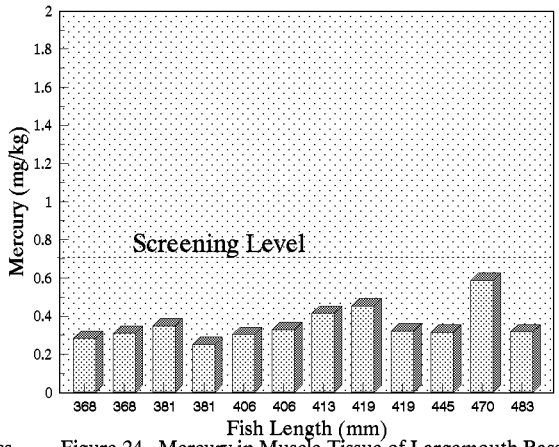


Figure 24. Mercury in Muscle Tissue of Largemouth Bass Nacogdoches Reservoir

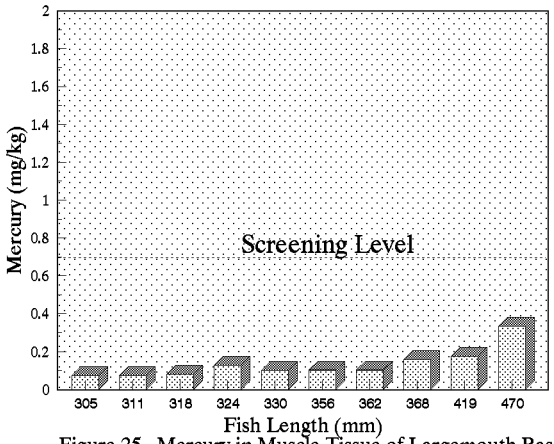


Figure 25. Mercury in Muscle Tissue of Largemouth Bass From Cooper Reservoir

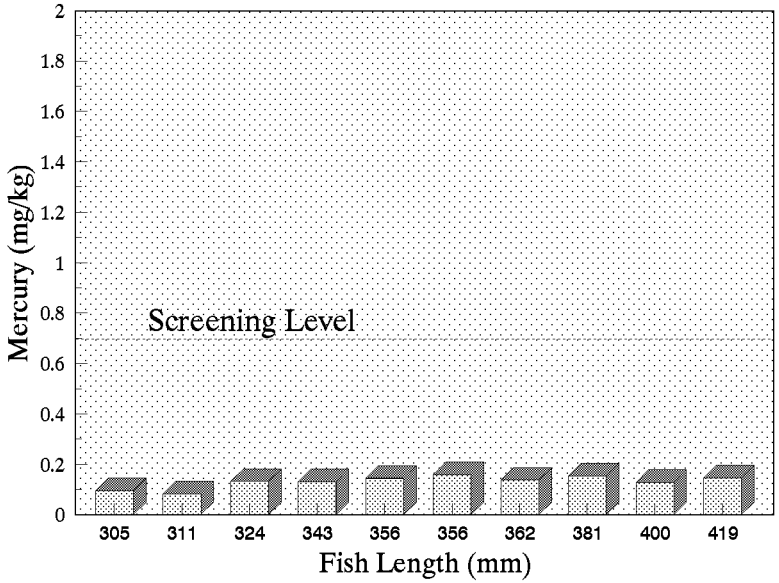


Figure 26. Mercury in Muscle Tissue of Largemouth Bass From Pat Mayse Reservoir

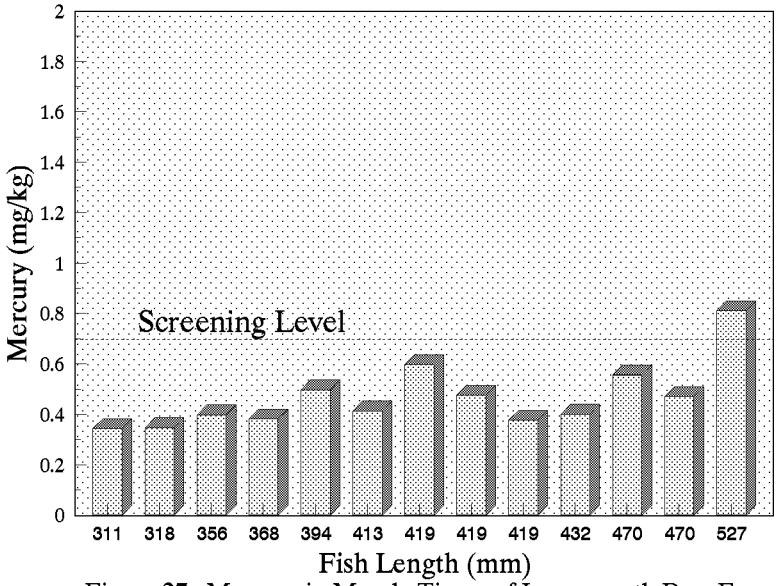


Figure 27. Mercury in Muscle Tissue of Largemouth Bass From Pinkston Reservoir

The mercury in tissue data from this study were provided to the TDH for evaluation in March 1998. The TDH conducted follow-up sampling of Kimball Reservoir and Pruitt Lake in September 1998 and also found consistently elevated mercury concentrations in fish tissue. The TDH subsequently performed a risk assessment, and issued consumption advisories in April 1999 covering all fish species for the two water bodies. The consumption advisories suggest that adults should consume no more than two eight-ounce meals per month, while children should consume no more than two four-ounce meals per month (TDH, 1999).

Boxplots of mercury in tissue data indicated skewness in about half the water bodies, so a nonparametric approach was used to test for differences (Figure 28). Median mercury in tissue concentrations from Kimball Reservoir and Pruitt Lake also exceeded the 0.7 mg/kg screening level (Figure 28). Differences in median mercury in tissue concentrations among the 13 reservoirs and lakes were determined to be significant (Kruskal-Wallis test, $P < 0.05$). Nonparametric multiple comparisons could not be performed due to unequal number of samples among the reservoir and lakes (Tables 17-29). Significant differences among mean mercury tissue concentrations were found using analysis of variance (ANOVA) ($P < 0.01$). Multiple comparisons (Tukey's Mulitple Comparison Test of unequal sample sizes; $P < 0.05$) indicated that mean mercury tissue concentrations in largemouth bass from Kimball Reservoir and Pruitt Lake were different from one another and the remaining water bodies (Table 30). Considerable overlap of the range of mercury tissue concentrations occurred among Pinkston, Houston County, Nacogdoches, and Cherokee Reservoirs and Shallow Prong Lake. Mean mercury tissue concentrations for the remaining water bodies were not significantly different from one another.

Mercury concentrations in axial muscle tissue of 137 individual largemouth bass ranged from 0.04 to 2.1 mg/kg (Tables 17-29). Mercury concentrations increased with total length of fish in all study water bodies excepting Ellison Creek Reservoir and Pruitt Lake (Table 31). No evidence of bioaccumulation was found in largemouth bass from Ellison Creek Reservoir. In Pruitt Lake, all the largemouth bass appeared stunted in size, possibly due to the small size of the lake. Total length was used to determine the expected mercury concentration (*E*Hg) for a harvestable-sized fish (16 inches). Regression coefficients (r) for regressions of mercury in tissue concentrations against total length within individual reservoirs and lakes ranged from -0.39 for Pruitt Lake to 0.92 for Kimball Reservoir. Examples of mercury concentrations in largemouth bass versus total length are plotted with regression lines in Figure 29 for two study reservoirs. *E*Hg concentrations in largemouth bass ranged from 0.09 mg/kg in Ellison Creek Reservoir to 1.2 mg/kg in Kimball Reservoir and exceeded the 0.7 mg/kg screening level in Kimball Reservoir and Pruitt Lake.

Chemical reservoir and lake water and sediment variables most highly correlated ($P<0.05$) with *E*Hg were pH, hardness, total organic carbon (TOC), calcium, TDS, sulfate, alkalinity, and magnesium in water and manganese in sediment (Table 32). Most conspicuous was the absence of significant correlations between *E*Hg and mercury in water or mercury in sediment.

The best one-variable linear regression model had pH as the independent variable ($r^2 = 0.51$), yielding the equation (Figure 30):

$$EHg = 2.39 - 0.291\ pH$$

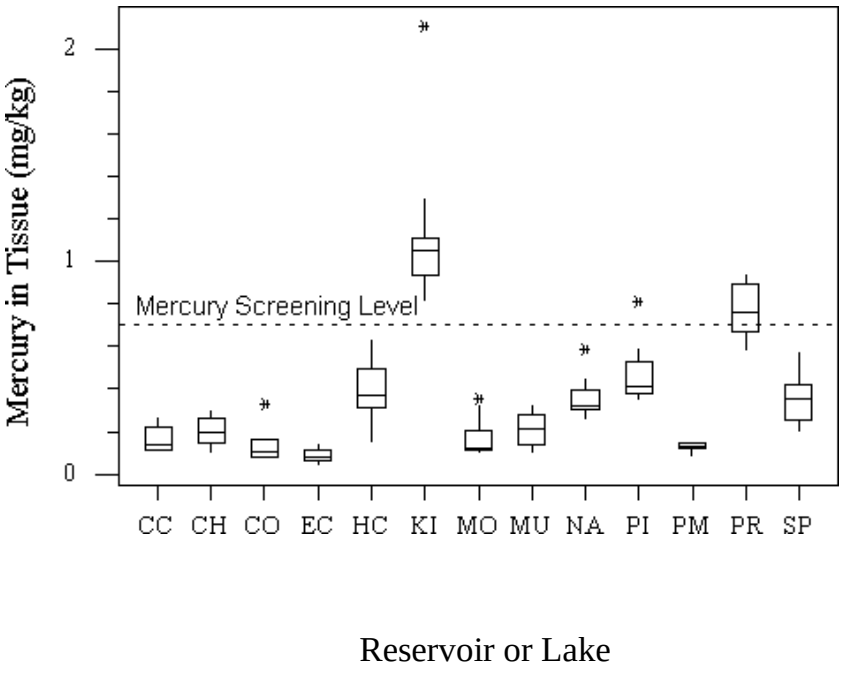


Figure 28. Comparison of Mercury Medians and Ranges in Largemouth Bass Tissue Among Study Reservoirs and Lakes. Asterisk indicates observations between 1.5 and 3 times the interquartile range. Names of Reservoirs and Lakes are Abbreviated

Table 30

Relationship of Largemouth Bass Mean Mercury Concentrations Among Study Reservoirs/Lakes. Names of Reservoirs/Lakes are Abbreviated. Mercury Means with Boxes Shaded Along the Same Row are not Significantly Different.

Reservoir/Lake												
KI	**PR**	**PI**	**HC**	**NA**	**SP**	**CH**	**MU**	**CC**	**PM**	**MO**	**CO**	**EC**

Table 31

Actual Mean and Expected Mercury (*E*Hg) Concentrations in Axial Muscle Tissue of Largemouth Bass and Correlation Coefficients for Regressions of Mercury in Tissue Against Total Fish Length from 13 East Texas Reservoirs/Lakes during 1997

Reservoir/Lake	Number of Fish	Actual Mercury Tissue Concentration (mg/kg)		*E*Hg (mg/kg)	Pearson's Correlation Coefficient (r)
		Mean	Range		
Cedar Creek	10	0.17	0.11-0.27	0.11	0.66
Houston County	12	0.39	0.14-0.63	0.22	0.62
Shallow Prong	10	0.35	0.20-0.58	0.40	0.65
Kimball	11	1.13	0.81-2.10	1.22	0.92
Cherokee	6	0.20	0.10-0.31	0.25	0.82
Pruitt	6	0.77	0.58-0.95	0.77[a]	-0.39
Ellison Creek	11	0.09	0.04-0.15	0.09[a]	0.07
Murvaul	12	0.19	0.09-0.32	0.21	0.76
Monticello	12	0.17	0.10-0.36	0.17	0.73
Cooper	10	0.13	0.07-0.33	0.21	0.92
Pat Mayse	10	0.13	0.08-0.16	0.18	0.65
Nacogdoches	12	0.35	0.25-0.59	0.32	0.51
Pinkston	13	0.47	0.34-0.81	0.46	0.79

[a] Actual mean mercury concentration used in place of *E*Hg where no significant relationship between mercury concentration in tissue and total length of fish was observed .

The regression slope was highly significant ($P<0.01$). Moreover, multiple regression revealed a significant ($P<0.05$) two-variable regression with pH and water column TOC as independent variables which explained 61 percent of the variation in *E*Hg (Figure 31). Addition of other independent variables did not significantly improve the multiple coefficient of determination (r^2) of the two-variable model. The equation was:

$$EHg = 1.59 - 0.211\ pH + 0.0282\ TOC$$

Although other variables did not improve the regression model, the *E*Hg in largemouth bass from reservoirs and lakes with calcium concentrations < 10 mg/L was significantly higher ($P< 0.05$) than in fish from reservoirs and lakes with calcium ≥10 mg/L (Table 33). The *E*Hg was significantly higher ($P<0.05$) in reservoirs and lakes with hardness values < 40 mg/L than in reservoirs and lakes

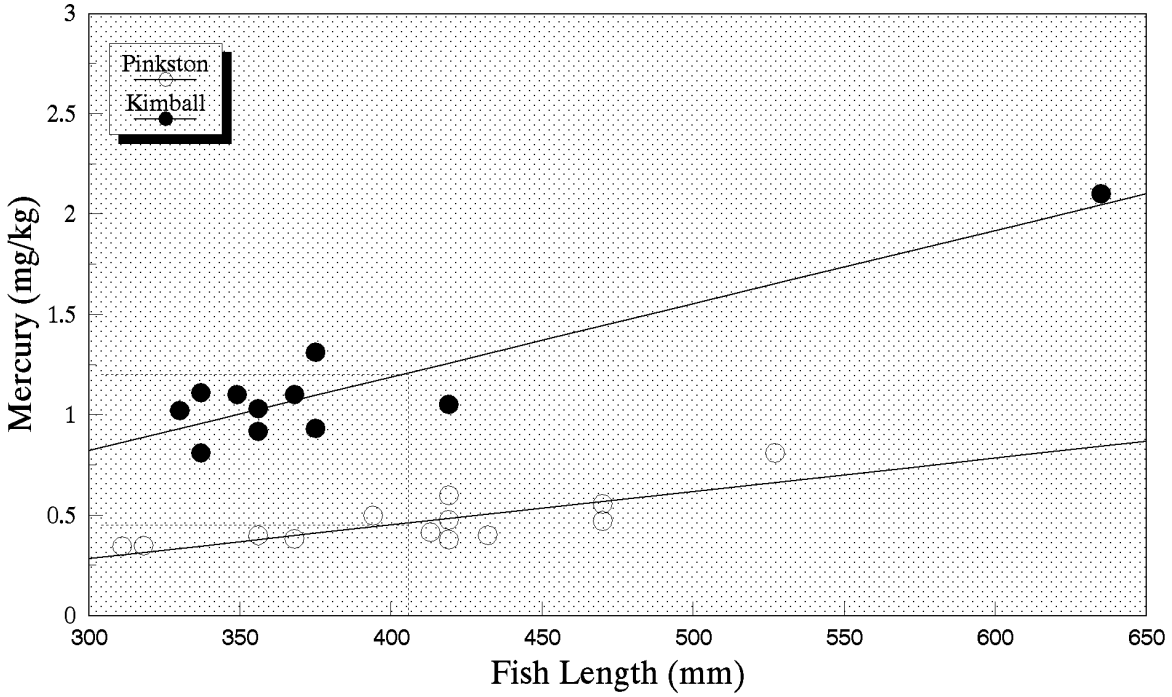


Figure 29. Examples of the Distribution of Mercury Concentrations in Largemouth Bass with Fish Length for Pinkston and Kimball Reservoirs. The Regression Equation for Pinkston Reservoir is Hg=9.2477 + 14.6919 (length); r2=0.62; and for Lake Kimball it is Hg=4.9272 +9.0465 (length); r2=0.84. Dashed lines show the expected mercury concentrations for Pinkston (0.46 mg/kg) and Kimball (1.22 mg/kg) at the standard length (406.4 mm).

with hardness concentrations ≥40 mg/L. Similarly, the *E*Hg was significantly higher (P< 0.05) in reservoirs and lakes with sulfate concentrations < 10 mg/L than in reservoirs and lakes with sulfate concentrations ≥40 mg/L.

Although numbers of samples were more limited, mercury concentrations in 23 channel catfish tissues from eight of the study water bodies were substantially less than the screening level (Figure 32). Likewise, no elevated concentrations were found in white crappie tissue from Cedar Creek Reservoir or Shallow Prong Lake, or in black crappie from Murvaul Reservoir (Figure 33). Three large bowfin from Pruitt Lake also had low mercury concentrations in tissue (Table 23). Freshwater drum from Shallow Prong Lake also had low mercury concentrations (<0.13 mg/kg), while three larger individuals from Pruitt Lake had concentrations that exceeded the screening level (Figure 34). The only two spotted bass collected during the study, from Pruitt Lake, had elevated mercury concentrations in tissue similar to levels for largemouth bass from the same water body (Table 23).

The inverse relationship between ambient water pH and mercury concentrations in largemouth bass muscle tissue in the present study is consistent with previous observations of elevated mercury concentrations in fish. Such a relationship has been documented for largemouth bass (Lange et al.,1993; Armstrong et al., 1995; Hanten et al., 1998), smallmouth bass *Micropterus dolomieui* (McMurtry et al., 1989), pumpkinseeds *Lepomis gibbosus* (Wren and MacCrimmon, 1983),

Table 32

Pearson Correlation (r) of Expected Largemouth Bass Mercury Concentration (*E*Hg), and Level of Significance (P) with Water and Sediment Variables From East Texas Study Reservoirs/Lakes

Variable	r	P
Field Measurements		
pH	-0.743	0.004
Water Chemistry		
Hardness	-0.690	0.009
Total Organic Carbon	0.674	0.012
Calcium	-0.620	0.024
Total Dissolved Solids	-0.609	0.027
Sulfate	-0.598	0.031
Magnesium	-0.599	0.031
Sediment Chemistry		
Manganese	-0.593	0.033

walleye (Scheider et al., 1979; Wiener et al., 1990), and yellow perch *Perca flavescens* (Cope et al., 1990).

The relationship between acidity of East Texas reservoirs and lakes and mercury concentration in largemouth bass tissue may have resulted partly from the effects of water chemistry on the microbial production of MeHg. There are several ways that pH of water may contribute to mercury accumulation in fish. Microbial production of MeHg near the sediment–water interface is inversely correlated with reservoir and lake pH (Xun et al., 1987). Experimental acidification studies of Wisconsin lakes resulted in higher mercury methylation rates in water and sediment than in control lakes (Winfrey and Rudd, 1990). Partitioning of MeHg between the water column and sediment has been shown to be affected by pH, with the release of MeHg greater at low pH than at high pH (Miller and Akagi, 1979). The uptake of mercury by largemouth bass also may have been enhanced by the greater permeability of biological membranes to MeHg, since permeability is inversely related to the waterborne calcium concentration, which is typically low in low-alkalinity, low pH reservoirs and lakes (Rodgers and Beamish, 1983). The efficiency of direct uptake of waterborne MeHg at all trophic levels and passage through the food chain would presumably be greater in low alkalinity reservoirs and lakes (Cope et al., 1990).

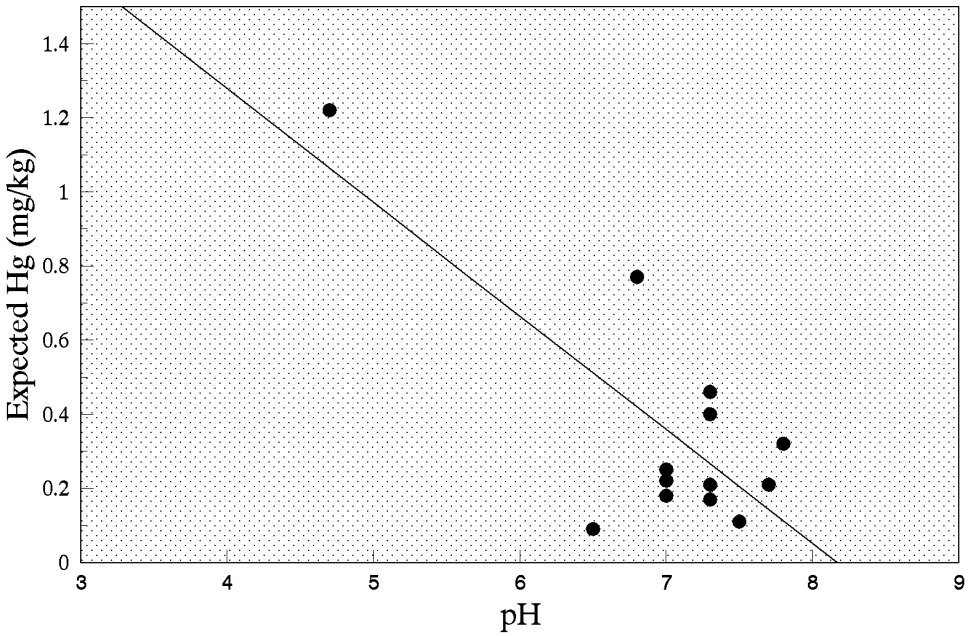


Figure 30. Relationship Between Expected Mercury Concentrations (EHg) and Ambient pH in 13 East Texas Reservoir and Lakes. The Regression Equation is 2.39 - 0.291(pH); r2 = 0.51.

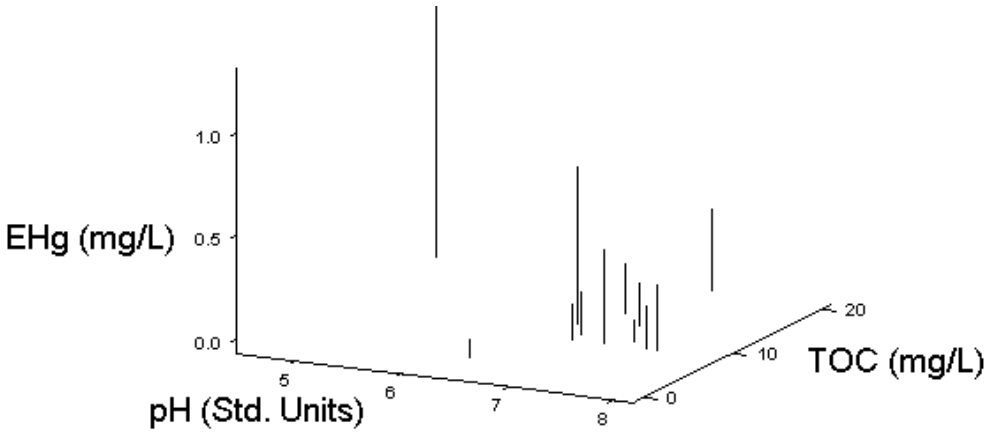


Figure 31. Expected Mercury Concentrations (EHg) in Largemouth Bass in Relation to pH and TOC in 13 East Texas Reservoir/Lakes.

Table 33

Median Expected Mercury Concentrations (*E*Hg) for Calcium, Hardness and Sulfate Categories from 13 East Texas Reservoirs and Lakes. Within each category, mercury concentrations with different letters are significantly different (P <0.05) using multiple Mann-Whitney tests.

Category	**Median *E*Hg (mg/kg)**	**Range, N**
Calcium (mg/L)		
< 10	0.36 z	(0.21-1.22, 8)
≥10	0.17 y	0.09-0.21, 5)
Hardness (mg/L)		
< 40	0.32 z	(0.22-1.22, 7)
≥ 40	0.18 y	(0.11-0.4, 6)
Sulfate (mg/L)		
< 10	0.40 z	(0.22-1.22, 5)
≥ 10	0.20 y	0.09-0.77, 8)

In the present study, largemouth bass with the highest mercury tissue concentrations were collected from Kimball Reservoir, which had the lowest pH (4.7 std. units), alkalinity (< 5 mg/L), hardness (6.9 mg/L), calcium (1.7 mg/L), and magnesium (0.6 mg/L) of any water body sampled (Tables 12 and 13). In contrast, reservoirs and lakes with higher pH, alkalinity, and calcium (Cooper, Pat Mayse, and Cedar Creek Reservoirs) had much lower mercury concentrations in fish tissue. High pH favors production of more volatile dimethyl mercury (Fagerstrom and Jernelov, 1972). In addition, elemental mercury volatilizes faster at high pH (Steffan et al., 1988).

Total organic carbon in water probably also contributed to mercury accumulations in largemouth bass. Water level fluctuations are common in East Texas reservoirs and lakes due typically to extended dry summer periods and high inflows during spring and fall months. These fluctuations play an important role in the cycling of nutrients, minerals, and organic substances in impoundments. Reservoirs and lakes that receive greater input of total organic carbon from their watersheds have been shown to receive increased mercury loadings (McMurtry et al., 1989). Periodic inundation and flushing of wetland areas may allow mobilization of mercury with total organic carbon (Mierle, 1990). This process is enhanced in low pH reservoirs and lakes, since mercury binds with organic matter more strongly at low pH (Jackson et al., 1980; Winfrey and Rudd, 1990). Mercury is bound to organic detritus and is often removed from the water column through deposition to sediments, where organic carbon enhances both methylation and bacterial activity (Callister and Winfrey, 1986).

Mercury in Sediment Cores

Sites sampled on the 13 reservoirs and lakes were generally assumed to be depositional over the life of the water bodies. Sediment cores were collected from the lower, deeper portions of each water

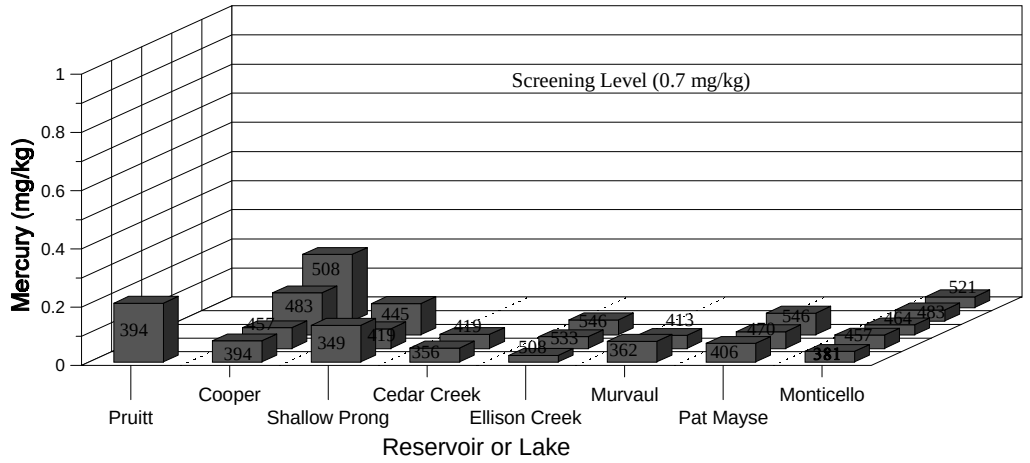


Figure 32. Mercury in Muscle Tissue of Individual Channel Catfish From Eight Reservoirs and Lakes Compared to the 0.7 mg/kg Screening Level. Fish Lengths (mm) Provided Boxes

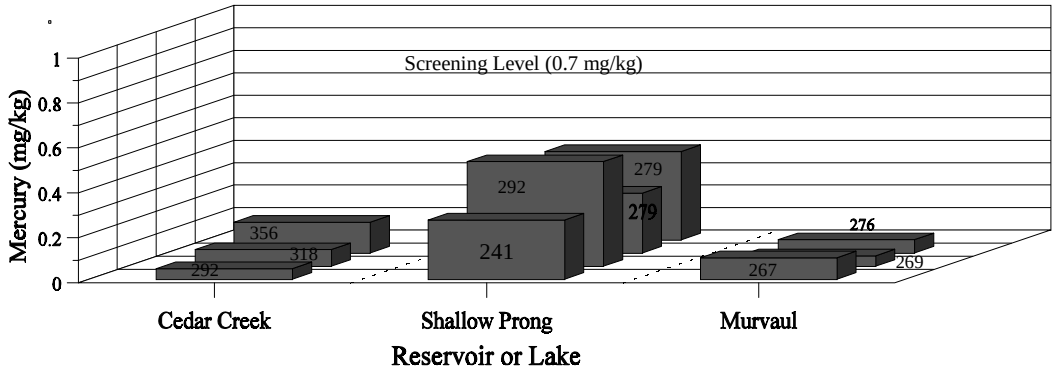


Figure 33. Mercury in Muscle Tissue of Individual Crappie From Cedar Creek and Murvaul Reservoirs and Shallow Prong Lake Compared to the 0.7 mg/kg Screening Level. Fish Lengths (mm) Provided in Boxes.

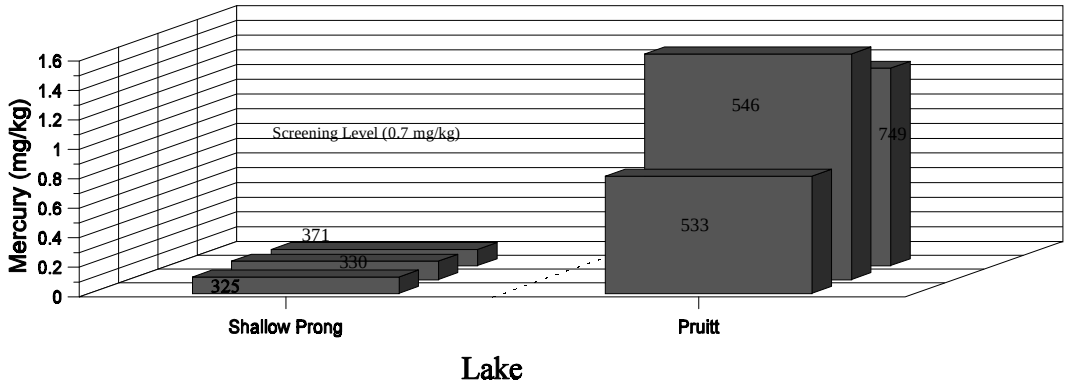


Figure 34. Mercury in Muscle Tissue of Individual Freshwater Drum from Pruitt and Shallow Prong Lakes Compared to the 0.7 mg/kg Screening Level . Fish Lengths (mm) Provided in Boxes.

body where influences of wind action and other surface activities would have minimal effects on sediment deposits. Kimball Reservoir and Pruitt Lake were considered exceptions, because during high flow their bed sediments could be subjected to disturbance by scouring. Sediment cores from those two water bodies were the shortest (0.15 m) of those collected (Table 34). The cores were collected according to standard protocol to allow general comparison of mercury concentrations over depth. No attempts were made to age the core sections or determine sedimentation rates.

Mercury concentration-depth profiles in sediment cores from all the study reservoirs and lakes exhibited a general decrease in concentration with depth into the sediment layer (Table 34). Plots of the mercury profiles in the 13 core samples show similar signatures with concentrations decreasing with increasing depth (Figures 35-47). Although all mercury profiles possessed a general similarity, there were distinct differences in surface maximum mercury concentrations. The highest mercury concentration (0.35 mg/kg) in the surface layers of the sediment cores was from Ellison Creek Reservoir, while the lowest surface concentration (0.015 mg/kg) was from Kimball Reservoir. With the exception of Ellison Creek Reservoir and Pruitt Lake, all mercury concentrations at the surface of the cores were less than 0.1 mg/kg.

As shown by detailed core stratigraphy (Figures 35-47), mercury concentrations in surficial core sections were elevated above those in deeper strata in nearly every case. Top to bottom differences in the cores were greatest for Ellison Creek Reservoir (0.32 mg/kg) and least for Kimball Reservoir (0.002 mg/kg). Houston County Reservoir was the only water body where the mercury concentration at the surface of the sediment core was less than the concentration at the bottom. Statistically significant decreasing trends of mercury concentrations with increasing depth were found in sediment cores from Cedar Creek ($r = -0.94$, $P < 0.005$) , Monticello ($r = -1.0$, $P < 0.001$), Cherokee ($r = -0.97$, $P < 0.001$), Ellison Creek ($r = -0.96$, $P < 0.001$), Pat Mayse ($r = -0.94$, $P < 0.005$), and Nacogdoches ($r = -0.91$, $P < 0.002$) reservoirs and Shallow Prong Lake ($r = -0.77$, $P < 0.001$).

Recent sediment core studies from the Rio Grande in Texas (USGS, 1997), the upper midwest portion of the U.S. (Rada et al., 1989; Swaim et al., 1992; Hurley et al., 1994), and Canada (Lockhart et al., 1993) have shown higher mercury concentrations in recently deposited reservoir and lake layers than in deeper, older layers. These findings suggest that mercury inputs may be continuing to increase. Mercury concentrations found in surficial sediments of East Texas reservoirs and lakes compare favorably with those observed during other studies in Texas, upper midwestern states, and Canada, which also indicate trends for higher mercury concentrations near the surface of sediment cores. The similarity of surficial mercury concentrations in sediments across East Texas suggests that local inputs generally are not particularly important. The exception is Ellison Creek Reservoir, where high mercury concentrations in sediment may originate from a secondary steel manufacturing facility located nearby.

Mercury in Tree Cores

Eastern red cedar *Juniperus virginiana* trees were sampled in areas as close as possible to the study reservoirs and lakes sampled in this study (Figure 48). A few were sampled in central Texas in order to evaluate possible regional differences. Eastern red cedar trees were customarily planted when

Table 34

Mercury Concentrations (mg/kg) in Sediment Cores from Study Reservoirs and Lakes

Sediment Depth	Reservoir/Lake												
	Cedar Creek	Houston County	Shallow Prong	Kimball	Monticello	Cherokee	Pruitt	Murvaul	Ellison Creek	Cooper	Pat Mayse	Nacogdoches	Pinkston
0 - 0.05 m	0.067	0.048	0.089	0.015	0.029	0.047	0.150	0.089	0.351	0.035	0.047	0.093	0.020
0.05 - 0.10 m	0.066	0.028	0.095	0.010	0.022	0.035	0.064	0.049	0.114	0.036	0.049	0.079	0.018
0.10 - 0.15 m	0.062	0.022	0.094	0.012	0.010	0.035	0.049	0.044	0.248	0.020	0.041	0.039	0.020
0.15 - 0.20 m	0.061	0.019	0.069		0.009	0.029		0.049	0.229	0.015	0.034	0.036	0.023
0.20 - 0.25 m	0.059	0.067	0.042		0.008	0.033		0.042	0.065	0.016	0.011	0.030	0.018
0.25 - 0.30 m	0.060		0.056			0.015		0.056	0.061	0.022	0.008	0.016	0.019
0.30 - 0.35 m			0.047			0.019		0.048	0.036			0.017	0.018
0.35 - 0.40 m			0.057			0.016		0.046	0.032			0.019	
0.40 - 0.45 m			0.048			0.011			0.031				
0.45 - 0.50 m			0.023			0.007			0.032				
0.50 - 0.55 m			0.039						0.027				
0.55 - 0.60 m			0.021						0.027				
0.60 - 0.65 m			0.027						0.031				
0.65 - 0.70 m			0.050										

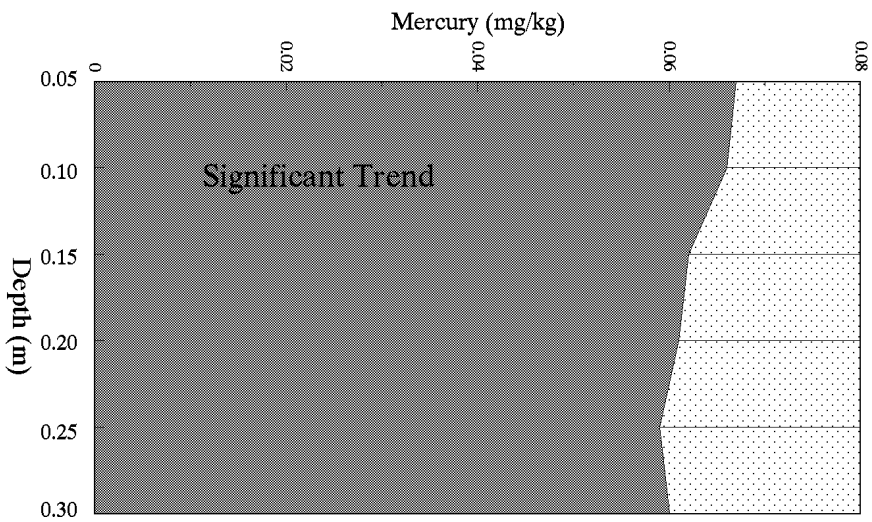


Figure 35. Depth Distribution of Mercury in Cedar Creek Reservoir Sediment

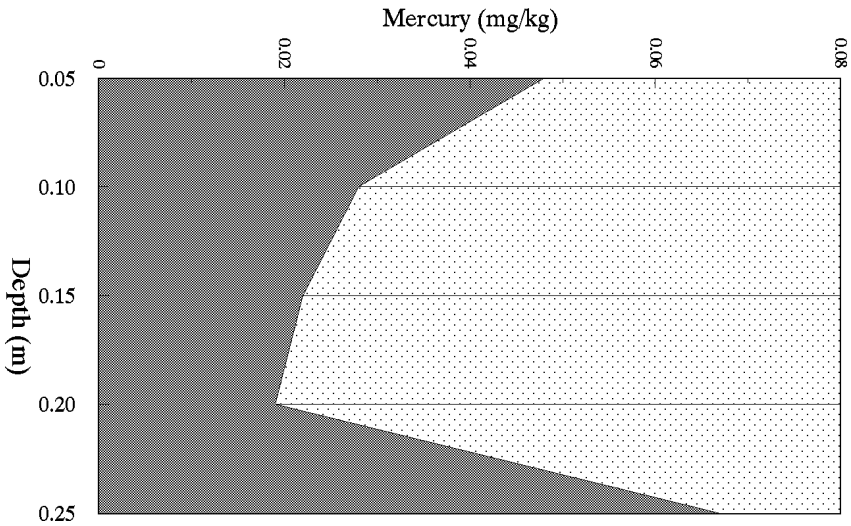


Figure 36. Depth Distribution of Mercury in Houston County Lake Sediment

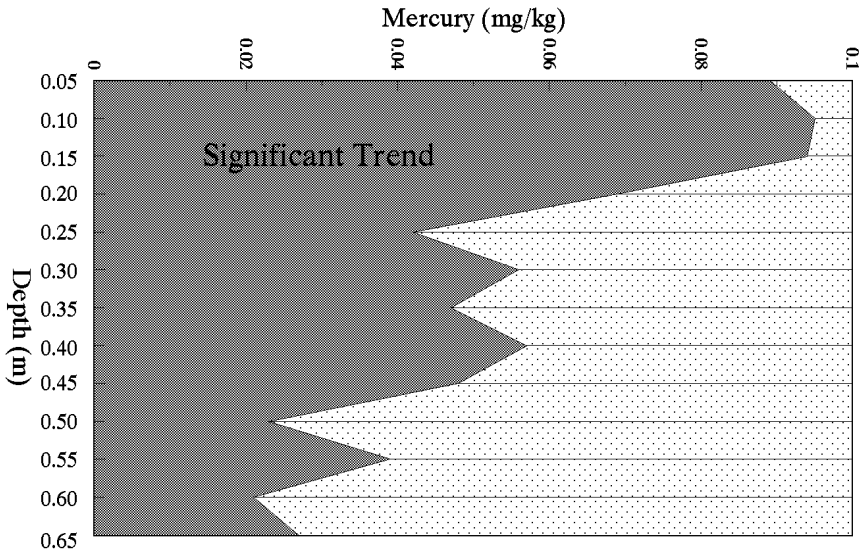


Figure 37. Depth Distribution of Mercury in Shallow Prong Lake Sediment

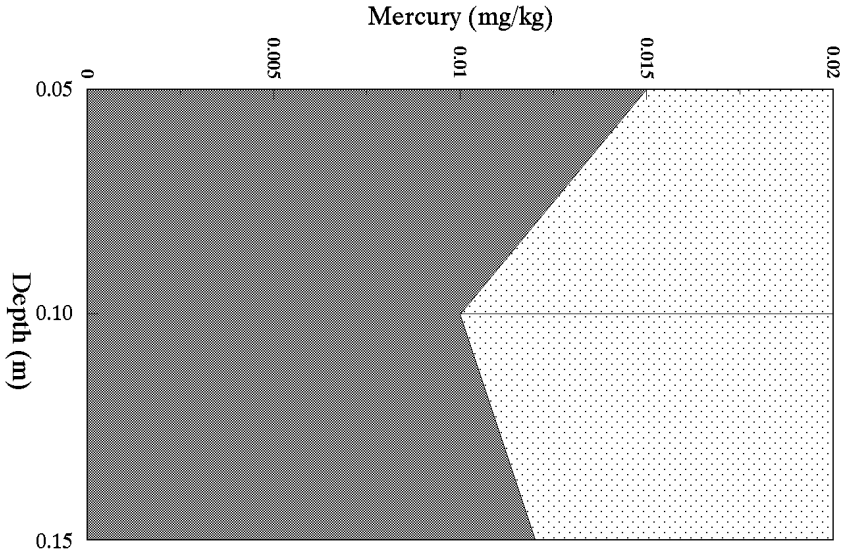


Figure 38. Depth Distribution of Mercury in Kimball Reservoir Sediment

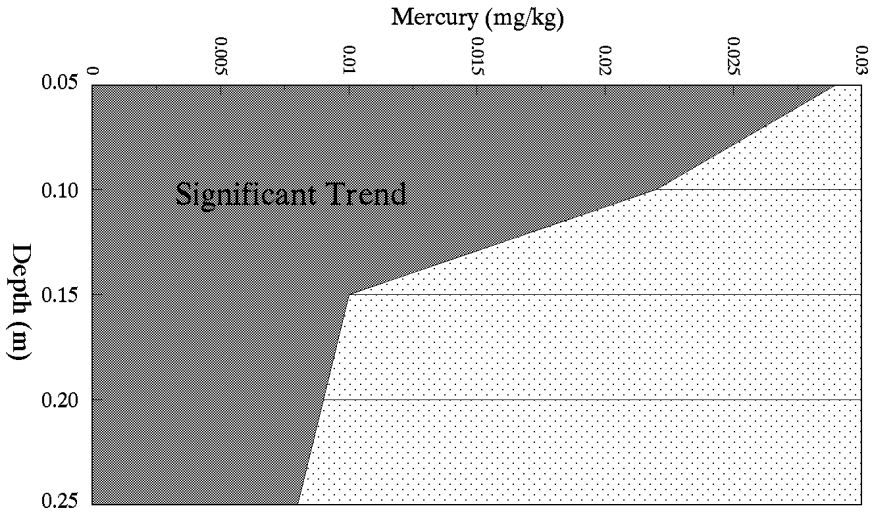


Figure 39. Depth Distribution of Mercury in Monticello Reservoir Sediment

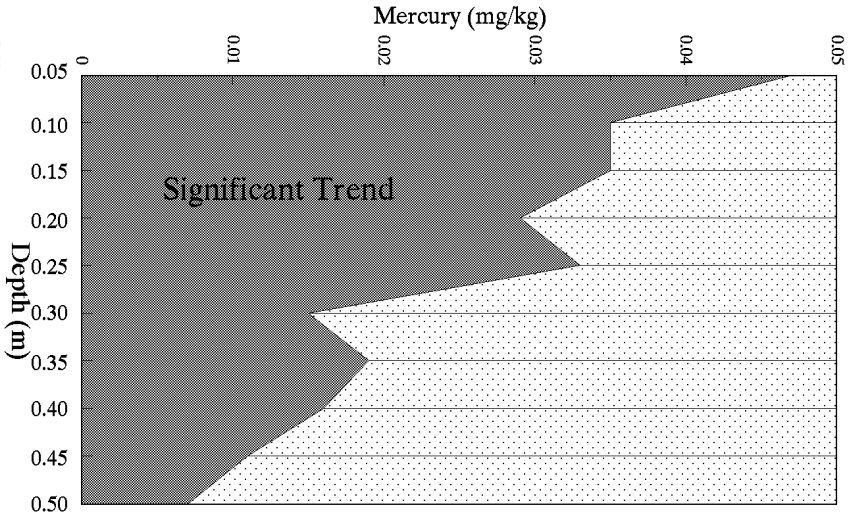


Figure 40. Depth Distribution of Mercury in Cherokee Reservoir Sediment

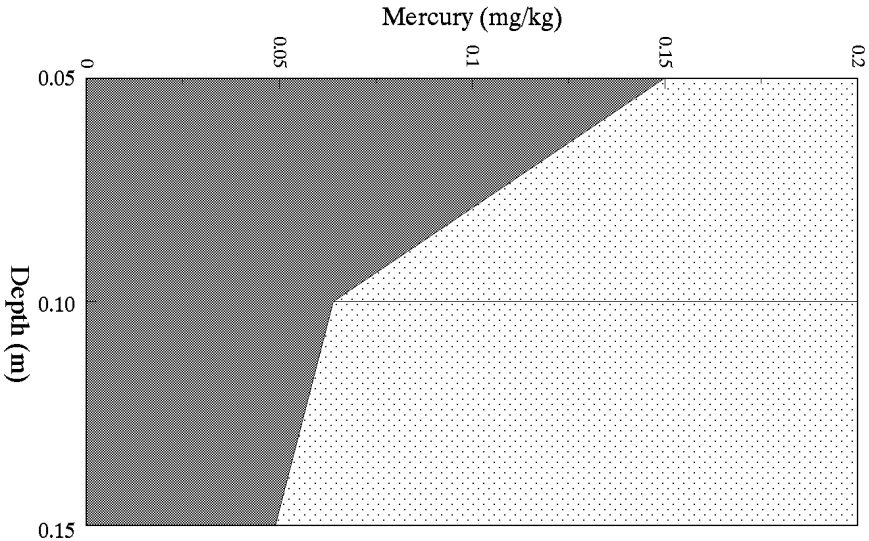


Figure 41. Depth Distribution of Mercury in Pruitt Reservoir Sediment

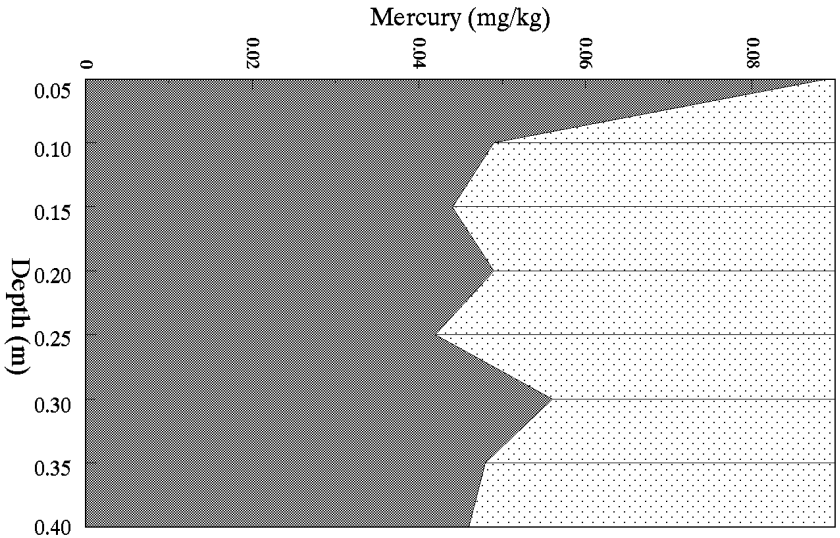


Figure 42. Depth Distribution of Mercury in Murvaul Reservoir Sediment

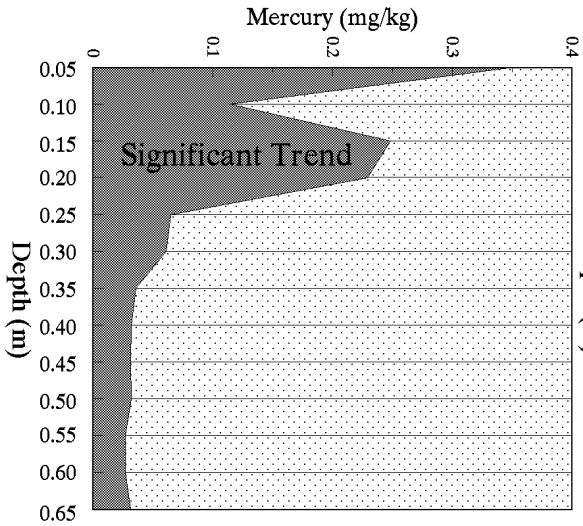


Figure 43. Depth Distribution of Mercury in Ellison Creek Reservoir Sediment

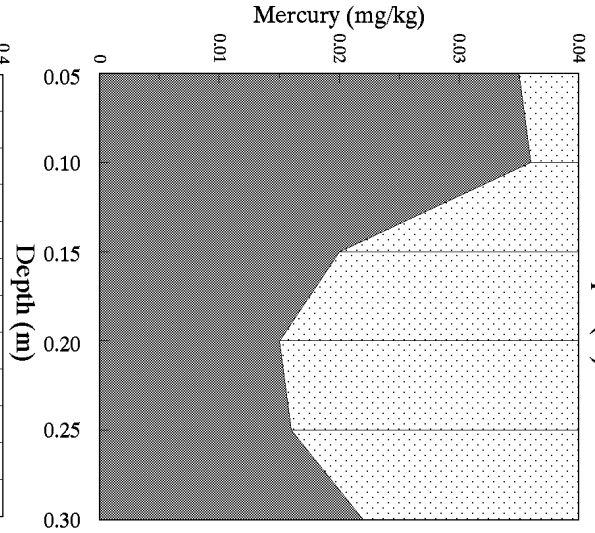


Figure 44. Depth Distribution of Mercury in Cooper Reservoir Sediment

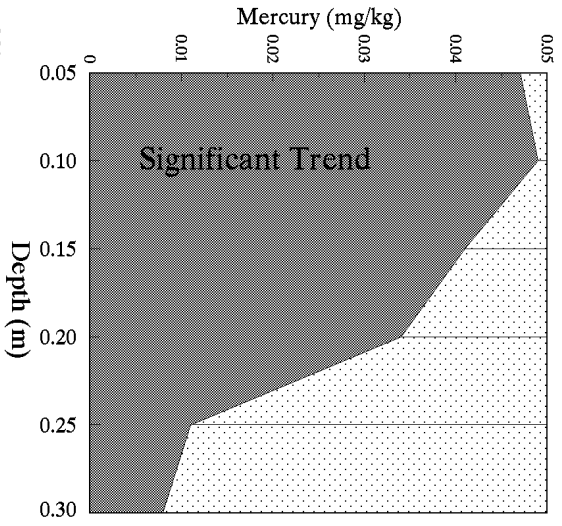


Figure 45. Depth Distribution of Mercury in Pat Mayse Reservoir Sediment

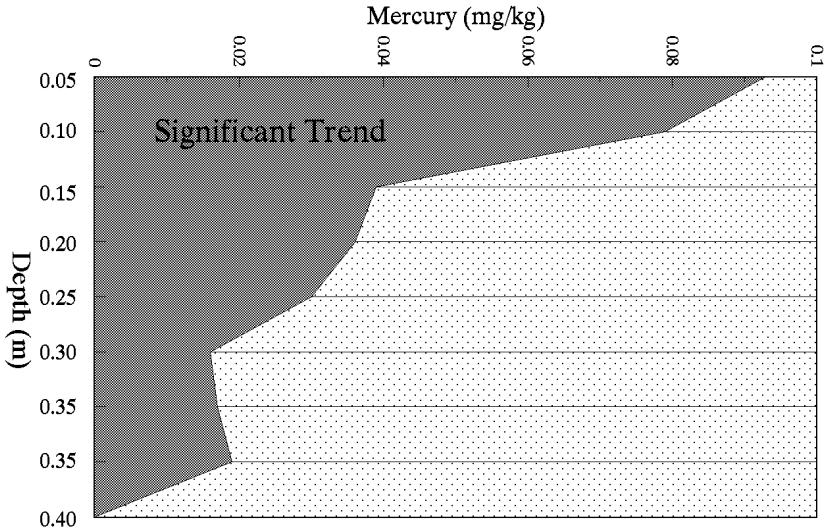


Figure 46. Depth Distribution of Mercury in Nacogdoches Reservoir Sediment

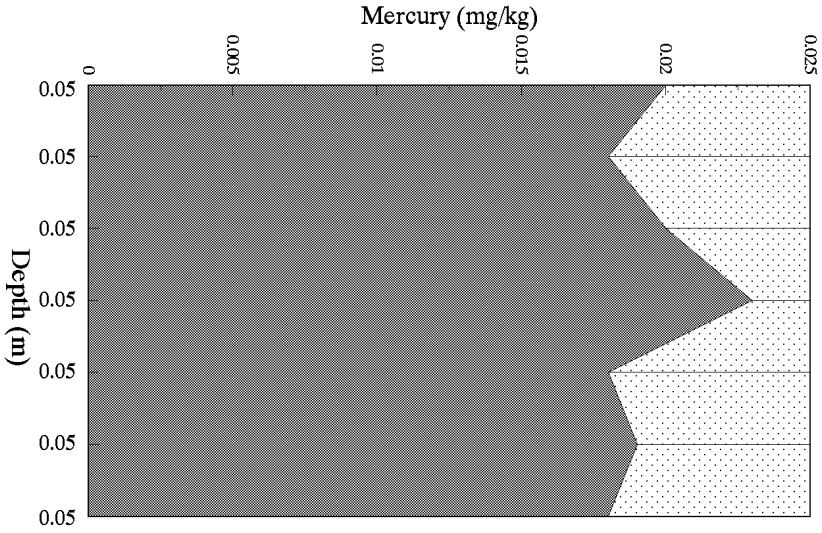


Figure 47. Depth Distribution of Mercury in Pinkston Reservoir Sediment

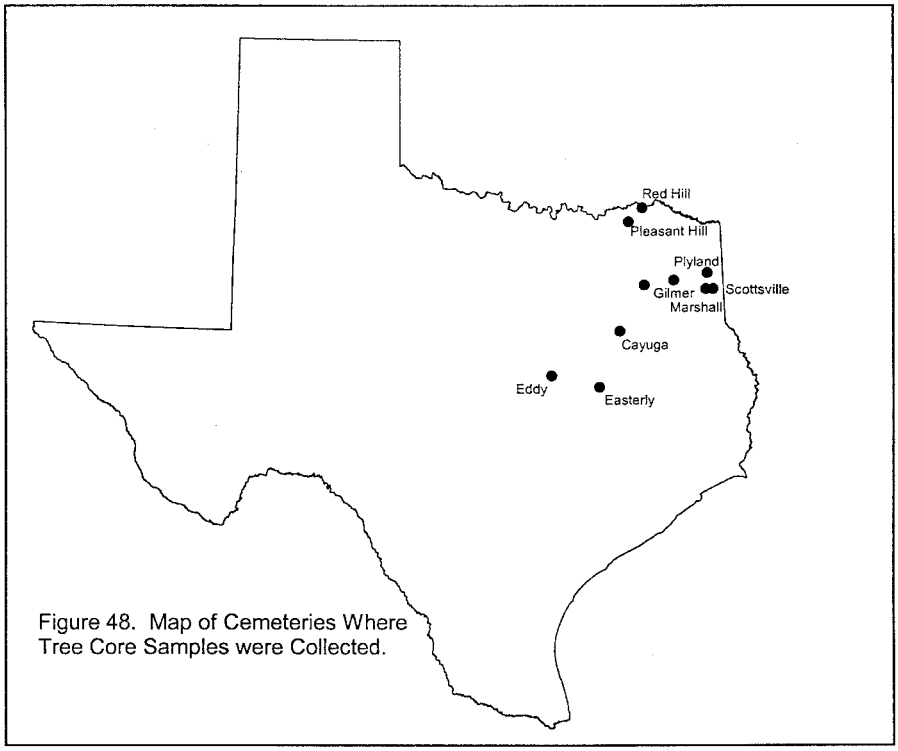


Figure 48. Map of Cemeteries Where Tree Core Samples were Collected.

cemeteries were first laid out across Texas. The trees readily highlight an area as a cemetery and provide shade and green foliage throughout the year.

Red cedar can be a very long-lived species with documented life spans in excess of 1,000 years (Guyette and McGinnes, 1987). The heartwood of eastern redcedar has a relatively low moisture content (McGinnes and Dingeldein, 1969), very low radial permeability (Stamm, 1970), and is considered very durable. Annual growth rings of softwood and hardwood trees have been employed with some success to reconstruct the historical deposition of metals and other contaminants (Baes and McLaughlin, 1984; Guyette et al., 1992). Recently, mercury concentrations in eastern redcedar have been used to evaluate temporal changes near contaminated industrial facilities in the U.S. (Turner and Bloom, 1997) and Japan (Kawabe et al., 1991).

In dendrochemical studies the chemical makeup of the annual woody increment is assumed to partially reflect the chemistry of the local environment (Amato, 1988). This assumption is complicated by the movement of elements within the woody stem of a tree. Absorbed elements enter the xylem through the roots from the soil solution, through the foliage following direct deposition, or through the stem following deposition on the bark (Cutter and Guyette, 1993). The occurrence of mercury in the atmosphere is largely in the gaseous elemental form and is not substantially taken

up through tree roots (Lindqvist, 1991). Uptake by foliar and bark may be far more important for mercury than for other metals (Turner and Bloom, 1997).

Mercury concentration-age profiles in tree cores from the ten tree sites exhibited few general trends (Table 35). For most of the eastern red cedar trees, recent mercury concentrations were very similar to concentrations from the oldest sections of the cores. The highest mercury concentration (0.072 mg/kg) occurred in a core section (1928-38) from the Easterly site, while the lowest concentration (0.003 mg/kg) occurred in a core section (1958-68) from the Mineola site. Tree cores from none of the cemetery sites had statistically significant decreasing trends of mercury concentrations with increasing age of incremental tree rings. In fact, the only statistically significant trends were for increasing mercury concentrations with increasing age of incremental tree rings at the Marshall ($r = 0.90$, $P < 0.001$) and Red Hill ($r = 1.00$, $P < 0.001$) sites.

The tree core data failed to corroborate trends noted for sediment cores, which suggested mercury inputs to the water bodies may be increasing. This suggests that analysis of red cedar tree rings may not be able to resolve temporal differences in ambient atmospheric mercury concentrations, except near strong industrial sources.

Table 35

Mercury Concentrations (mg/kg) in Tree Cores

Approximate Age	Cemetery									
	Easterly	Gilmer	Cayuga	Eddy	Marshall	Pleasant Hill	Plyland	Red Hill	Mineola	Scottsville
1988-1998	0.029	0.044	0.022	0.027	0.016	0.025	0.008	0.015	0.006	0.004
1978-1988	0.055	0.035	0.034	0.039	0.022	0.020	0.009	0.016	0.005	*
1968-1978	0.062	0.035	0.021	0.043	0.022	0.031	0.017	0.016	0.006	*
1958-1968	0.040	0.066	0.019	0.034	0.028	0.033	0.011	0.018	0.003	*
1948-1958	0.047	0.042	0.021	0.039	0.035	0.036	0.009	0.016	0.004	*
1938-1948	0.045	0.069	0.023	0.025	0.039	0.031	0.011	0.018	*	*
1928-1938	0.072		0.022		0.043	0.031	0.008	0.019	*	
1918-1928	0.020		0.023		0.046			0.018	*	
1908-1918			0.025		0.048			0.021		
1898-1908					0.041			0.018		

* No result due to severe matrix interference from cedar oils

SUMMARY AND CONCLUSIONS

Due to atmospheric deposition, mercury contamination of fish in East Texas water bodies has probably been occurring for many decades. Contamination problems were revealed in Texas only after mercury-contaminated fish were found in the neighboring states of Arkansas and Louisiana. The practice employed by the TNRCC SWQM Program of collecting a variety of fish at different sites from differing trophic levels and size groups and conducting toxic substance analyses on whole body portions contributed to the failure to identify the mercury problem. The ecosystem-health fish monitoring program used by the TNRCC is an efficient procedure for monitoring a broad spectrum of toxic substances in fish tissue. However, this approach failed to identify mercury contamination in Texas fish, since the metal concentrates in the muscle tissue of large-sized, piscivorous fish. Inadequate numbers of fish monitored annually for toxic substances by the TNRCC, TDH, and TPWD also caused the mercury problem to go undetected in Texas.

Texas would benefit from having a coordinated, statewide fish tissue monitoring program targeted primarily to evaluate human health risk. Unfortunately, none of the three major state resource protection agencies currently have funding to develop and administer such a program. A recent attempt by the TDH to secure additional funding for a statewide fish tissue monitoring program was rejected by the Texas Legislature. The TPWD is the only agency that is presently aggressively sampling fish in East Texas water bodies to determine if they are contaminated with mercury.

The 11 reservoirs and two natural lakes in East Texas sampled during the present study had waters of very good quality. Field measurements from most of the water bodies were within acceptable ranges and attained established criteria or screening levels. All had dissolved oxygen concentrations >5 mg/L in the mixed surface layer. Monticello Reservoir, which receives a heated water effluent from a power plant located nearby, had exceptionally high maximum water temperatures near the surface. The high water temperatures were due to a sustained weather pattern that allowed heat to accumulate in the reservoir coupled with the discharge of warm effluent from the power plant. Due to low buffering capacity, water in Kimball Reservoir was acidic (pH <5.5 std. units). Most of the reservoirs and lakes sampled were considered to have low alkalinity (<50 mg/L) and soft water (hardness <100 mg/L). Nutrient and chlorophyll *a* concentrations were generally low, with only few exceedances of screening levels.

Most of the 11 metals sampled in water from each of the reservoirs and lakes were low in concentration with 82 percent of the analyses reported as less than minimum analytical limits. Total mercury was the most commonly detected metal (8 of 13 water bodies). Mercury in water concentrations were substantially less than acute and chronic aquatic life criteria, indicating they were not even remotely toxic to aquatic life. However, mercury in water concentrations from six water bodies exceeded the human health criterion. These exceedances suggest fish from these water bodies may potentially be harmful if consumed by humans. Additional sampling would be required to determine if the criterion is actually violated, since it is established as a long-term average concentration in the TSWQS, and only instantaneous samples were collected during this study.

Sediments from the 13 reservoirs and lakes were composed primarily of fine particles . Most were highly organic. Metals in sediments were nearly always detected (>85 % for all analyses); however, exceedances of screening levels were erratic among water bodies. Sediment from Ellison Creek

Reservoir was most contaminated (high levels of barium, cadmium, copper, lead, manganese, mercury, nickel, and selenium) evidently in relation to the location of a steel manufacturing facility adjacent to the reservoir. Historical toxicity testing of water and sediment from the reservoir and Big Cypress Creek downstream has revealed only rare instances of toxicity. Sediments from all water bodies had SEM concentrations lower than AVS concentrations, indicating that divalent metals may be bound in insoluble sulfide complexes and would not be predicted to cause toxicity.

Analysis of largemouth bass is an appropriate approach for assessing mercury bioavailability in streams, reservoirs, lakes, and ponds throughout Texas (as well as other Southern States). The species is widespread and abundant, and samples can usually be collected with minimal effort. Of seven metals analyzed in 137 largemouth bass fillet samples, arsenic, cadmium, lead, and selenium were not commonly detected (<35 % of all analyses). Mercury, chromium, and copper were frequently detected (>80 % of all analyses). Mercury was the only metal detected in all largemouth bass, and concentrations exceeded the 0.7 mg/kg screening level in Houston County, Kimball, and Pinkston reservoirs and Pruitt Lake. In Houston County and Pinkston reservoirs only the largest individual bass had elevated mercury concentrations, while all of the largemouth bass from Kimball Reservoir and five of six from Pruitt Lake had elevated concentrations. Total mercury concentrations in largemouth bass muscle tissue ranged from 0.04 to 2.1 mg/kg and were positively correlated with fish size in all but two water bodies. No evidence of bioaccumulation was found in fish from Ellison Creek Reservoir. In Pruitt Lake, largemouth bass appeared stunted in size, possibly due to over-population and the relatively small size of the lake. Largemouth bass from Pruitt Lake, although smaller and more uniform in size than those from the other study water bodies, may actually have been older. This anomaly indicates a need for determining age as well as length of fish in bioaccumulation studies.

Reservoir and lake variables most strongly related to mercury concentrations in largemouth bass included low pH, low concentrations of hardness, calcium, magnesium, TDS, and sulfate in water, high concentrations of TOC in water, and low concentrations of manganese in sediment. The pH of water alone accounted for 51 percent of the variation in expected mercury concentration in largemouth bass. Reservoir and lake pH and TOC in water combined explained 61 percent of the expected mercury concentration. The highest expected mercury concentration (1.22 mg/kg) in largemouth bass came from Kimball Reservoir, which had the lowest pH, alkalinity, hardness, calcium, and magnesium in water, shared the highest TOC concentration in water, and had the lowest concentration of manganese in sediment. In stark contrast was the lack of correlation between expected mercury concentration in largemouth bass and mercury concentrations in water or sediment. Of the six water bodies that had mercury concentrations in water that exceeded the human health criterion, only Kimball Reservoir and Pruitt Lake had largemouth bass with consistently elevated mercury concentrations. This lack of correlation suggests that exceedance of the mercury-in-water human health criterion may not be a good predictor of mercury bioaccumulation in fish. However, additional sampling would be required to determine if the human health criterion is violated, since it is established a long-term average in the TSWQS. The observed relationships suggest that mercury concentrations in water and sediment are not nearly as important as are in-reservoir/lake processes which control the production of MeHg and its subsequent bioconcentration and bioaccumulation in largemouth bass in East Texas reservoirs and lakes.

The occurrence of elevated mercury concentrations in largemouth bass in East Texas does not appear as widespread as originally thought. In the present study, two of 13 water bodies had largemouth bass with consistently elevated mercury concentrations; the TDH found five of 14 water bodies with consistently elevated mercury concentrations in largemouth bass and other species. The water bodies in which mercury concentrations in largemouth bass were not consistently elevated were generally more alkaline. Waters with pH of 7.0 and higher favor the formation of dimethyl mercury which readily volatilizes to the atmosphere. Piscivorous fish from numerous low alkalinity, soft-water East Texas streams, reservoirs, lakes, and ponds, that have not been sampled, have the potential to accumulate mercury burdens exceeding levels considered safe for human consumption. The present study and earlier efforts indicate that mercury content in largemouth bass is fairly predictable, provided pH and TOC in water are known. Evaluation of ambient water pH would serve as a quick, inexpensive predictive method for selecting water bodies to sample to determine if fish are contaminated with mercury.

Research elsewhere indicates that atmospheric deposition accounts for high mercury concentrations in surface sediments and is likely to explain observations of elevated MeHg concentrations in freshwater fish from remote lakes. The general uniformity in wet deposition of mercury across the southern states, and uniformity in mercury content in surficial sediment core sections observed in the present study suggest that atmospheric deposition is the predominant source of mercury across the East Texas region. Local sources may override atmospheric deposition, as was suggested in Ellison Creek Reservoir, which was constructed next to a steel manufacturing plant. Mercury concentrations in sediment in that reservoir were approximately double the screening levels. One of the primary reasons for including Monticello Reservoir in the present study is the adjacent location of a coal-fired steam electric generating plant. Combustion of coal as a fuel has been shown to be one of the dominant point emission sources of mercury. However, mercury concentrations in sediment from Monticello Reservoir were less than screening criteria. It is unlikely that atmospheric inputs of mercury to the low-pH reservoirs and lakes were greater than those to the high-pH-water bodies. Although not age-dated, mercury concentration-depth profiles in sediment cores showed that mercury concentrations in surficial sections were elevated above those in deeper strata in nearly every case. Statistically significant decreasing trends in mercury concentrations with increasing depth in sediment cores from seven of the study water bodies suggest that mercury inputs may be increasing across the East Texas region.

Mercury contamination of fish across East Texas most likely will persist, since mercury is a naturally occurring element that does not decay, and in-reservoir and lake conditions of some water bodies provide favorable conditions for methylation of mercury. Even if all anthropogenic sources of mercury emitted to the atmosphere were eliminated, large reserves stored in terrestrial soils and emissions from natural sources would continue to provide mercury for cycling. Unfortunately, once mercury enters a stream or impoundment, it can remain for decades, cycling between the sediment and biota, provided conditions that favor methylation are present. The mercury contamination problem is manageable, however, through continued monitoring to identify problem water bodies and issuance of consumption advisories to advise the public of the potentially harmful effects of consuming mercury-contaminated fish.

Emissions of mercury to the atmosphere originate world-wide from anthropogenic and natural sources. Ultimately, anthropogenic emissions will have to be dramatically reduced to affect

reduction in bioaccumulation of mercury in fish. Data on industrial demand for mercury show a general decline in domestic mercury use since demand peaked in 1964. According the EPA's *Mercury Study Report to Congress* (USEPA, 1977) for industrial or manufacturing sources that use mercury in products or processes, the overall consumption of mercury is generally declining. Industrial consumption of mercury has declined by about 75 percent between 1988 [1,503 Megagrams (Mg)] and 1996 (Mg 372). Much of the decline can be attributed to the elimination of mercury as a paint additive and the reduction of mercury in batteries. Mercury from emission sources should show similar declines, now that EPA has finalized mercury emission limits for municipal waste combustors and medical waste incinerators. When fully implemented, these emission limits will reduce mercury emissions from these sources by an additional 90 percent over 1995 levels. Continued reductions in nitrogen and sulfur dioxides from point emission sources which burn fossil fuels are anticipated through more restrictive limits. These reductions will help to reduce acid rain which makes water bodies in east Texas more vulnerable to mercury bioaccumulation.

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