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MERCURY CONCENTRATIONS IN STREAMS OF EAST TEXAS

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ABSTRACT: Recent studies on potential mercury (Hg) contamination of fish from East Texas lakes and waterways have caused concern about mercury levels in East Texas waters. Historical records of Hg concentrations in 33 East Texas streams showed that median concentrations for each stream segment were no different than other U.S. streams. All the means and medians for stream segments having at least 20 recorded measurements were less than Texas (2.4 µg/L) water quality standards. Water samples collected in December 1995 and March 1996 from 6 different stream sites in Nacogdoches County had concentrations similar to historical records. Due to biological magnification, fish Hg levels can be 20,000 times greater than water Hg levels and levels are greater in large fish than in small fish. Although a recent study on sediment cores in 13 East Texas reservoirs and lakes suggested possible increases in mercury concentrations across the region, all Hg concentrations in water and sediment were far below Texas acute and chronic quality standards. No significant correlations were found between fish mercury concentrations and mercury concentrations in water or sediment. Potential agricultural inputs of Hg in East Texas are very low; the most likely source of Hg is atmospheric deposition from fossil fuel combustion and other industrial practices. The following may be considered to minimize potential health risks: 1) consume smaller fish from a variety of waterbodies, 2) increase consumption interval, 3) avoid eating skin and fatty tissues, and 4) limit consumption to quantities recommended by the Texas Health Department.
KEY TERMS*:* mercury; streams; reservoirs; fish; East Texas.

INTRODUCTION

Potential mercury contamination of fish in East Texas lakes and waterways has been an issue of great concern in recent years (Turner, 1995; Crowe, 1996; Twidwell, 2000). First, the Texas Department of Health (1995a) warned that consumers should limit consumption of largemouth bass and freshwater drum caught in Caddo Lake, Big Cypress Creek, Toledo Bend Reservoir, Sam Rayburn Reservoir and Steinhagen Lake to two 8-ounce servings a month for adults and two 4-ounce servings a month for children. For white bass or white/striped hybrid bass from Steinhagen Reservoir, the consumption should be limited to one 8-ounce serving for adults and one 4-ounce serving per month for children. In the November 6, 1995 issue, Texas Park & Wildlife's "News" also reported the concern and preventive measure of fish consumption for the cited water bodies in East Texas. However, the report indicated that there is no risk of mercury contamination from fishing, skiing, boating or other water-contact recreational activities. Then, in 1999, Texas Department of Health broadened its warning regarding possible mercury contamination of fish consumption to include Lake Pruitt in Case County and Lake Kimball in Hardin and Tyler Counties. The recommended maximum consumption levels are the same as the other East Texas streams.

More recently, 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 physiochemical variables in water and sediment (Twidwell, 2000). The results showed that total mercury concentrations in edible largemouth bass muscle tissue ranged from 0.04 to 2.1 mg/kg and nearly all largemouth bass from Pruitt Lake and Kimball Reservoir had mercury concentrations exceeding the 0.7 mg/kg screening level.

A number of questions about mercury levels in East Texas waters have been raised because of these reports. Is the mercury concentration in East Texas waters too high? Is the present mercury level higher than the past? If it is too high, what are the possible causes? This report examines the historical records of mercury concentrations and presents results of analyses of current mercury concentrations in a few streams in East Texas.

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MECURY AS A POLLUTANT

Mercury (Hg) is a naturally occurring element that can be emitted from coal burning, municipal incinerators, mining, chlorine-caustic soda plants, old oil wells, and runoff from farms using pesticides containing mercury. It is widely used in the processing of steel, electrical apparatus, industrial instruments, thermometers, batteries, photographic material, fungicides, papermills, algicides, ship's paints, etc. (Nriagu, 1979). Mercury has not been used in agricultural pesticides for food use since 1969 and registrations on all U.S. pesticides containing mercury were canceled in 1995.

Mercury concentrations are 0.1 µg/L or less in U.S streams (Hodges, 1977) and 0.02-0.2 mg/g in soils (Bohn et al. 1979). Fitzgerald (1979) summarized results of other studies to indicate 0.05 µg/L (Table 1) as an upper limit for natural fresh waters with no significant effects of pollution or mineralized Hg strata. However, many laboratories and surveys cannot detect a concentration at this scale. The detection limit for Hg in water by the U. S. Geological Survey (USGS) is 0.1 µg/L.

Table 1. Summary of background mercury concentrations in natural waters (Fitzgerald, 1979).

Natural Waters	Mercury Concentration, µg/L
Open ocean seawater	≤0.01
Coastal seawater	≤0.02
Estuarine seawater	≤0.05
Rivers and lakes	0.01-0.05
Rainwater	
Open ocean	0.001
Coastal ocean	0.01
Continents	≥0.05
Sediment interstitial water	0.1
Glacial water	0.01
Ground water	0.05

The U.S. Environmental Protection Agency (USEPA) sets the drinking water quality standard for mercury to be 2 µg/L. In Texas, the mercury concentration standards are 2.4 µg/L for acute aquatic life protection in fresh water and 0.0122 µg/L in water and fish for human health protection (Texas Natural Resource Conservation Commission, 1995). Accumulation of mercury in the body may affect the respiratory, central nervous, and cardiovascular systems. Symptoms may include tingling of the skin, incoordination, visual and hearing impairments, brain and tissue damages, and temporary insanity. Also, a developing human fetus is very sensitive to the neurotoxic effects of methylmercury. USEPA (1997) stated that about 1-3% of women of childbearing age consume sufficient quantities of methylmercury to place their offspring at risk.

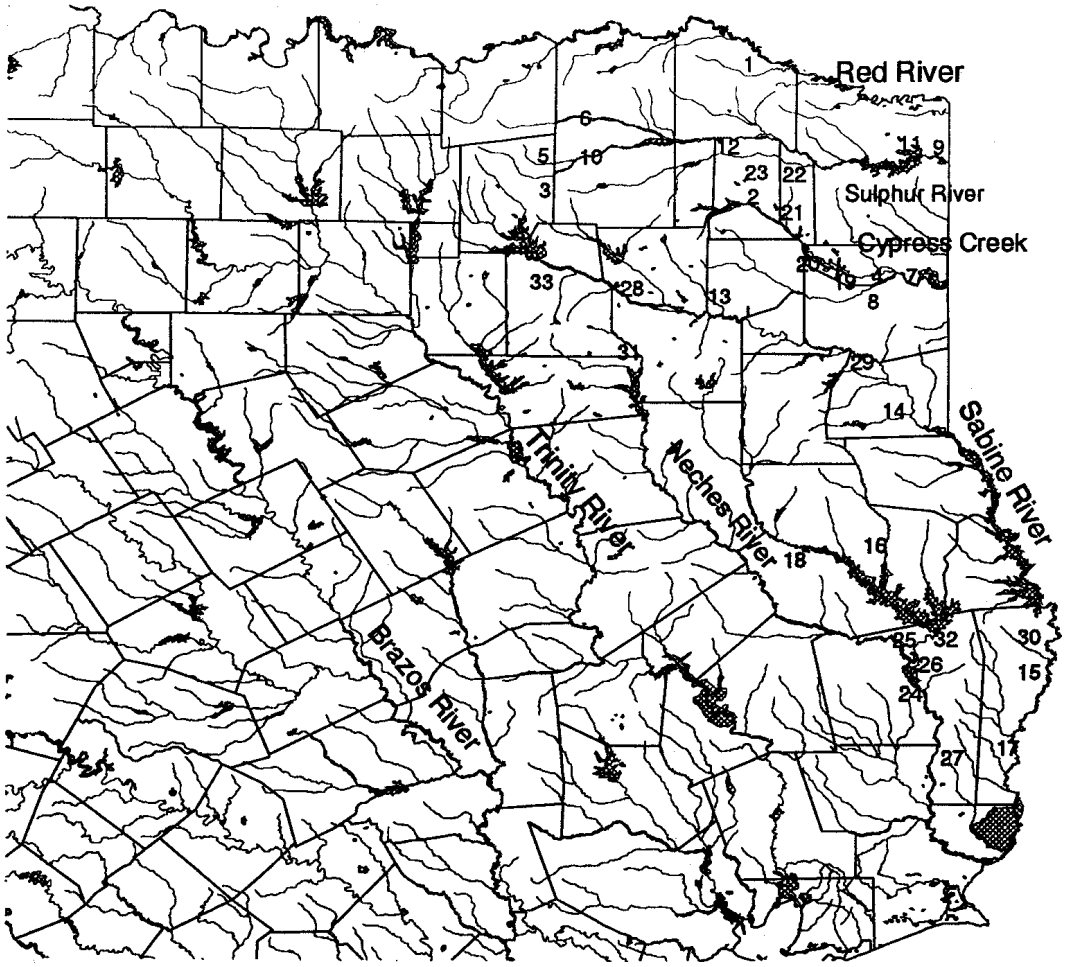
Chemically, elemental mercury is a heavy, liquid metal at common temperatures. Both organic and inorganic forms of mercury can be found in the environment. When inorganic mercury (such as $HgCl_2$, HgO, HgS, $HgSO_4$) is released into the water, it binds with sediment under anaerobic conditions. Inorganic mercury can be converted into organic methylmercury under aerobic conditions by microorganisms (Anderson, 1979; Pfeiffer et al. 1993). Methylmercury is soluble in fats but not in water and is very stable and very toxic to living organisms. It has a biological half-life in humans of about 72 days, compared to 5 days for inorganic mercury. Organic mercury is available in the food chain; and biological magnification may increase mercury concentrations in fish and shellfish to a level 100,000 times the mercury concentration found in water (Texas Department of Health, 1995a). Mercury is a significant hazard to human health only if contaminated fish is consumed in quantities exceeding the reference dose of 1µg/kg bw/day (USEPA, 1997)

HISTORICAL RECORDS

USGS is responsible for monitoring water quality of surface streams and ground waters in the United States. However, mercury monitoring is not a routine activity in their water quality program. Data are only available for a few streams and for a very limited number of measurements throughout the year. Examining Texas Water Resources Data published by USGS showed only 33 stream segments in East Texas having records on mercury concentration dating back to 1974 (Figure 1). There are no USGS records on mercury concentrations in East Texas streams before 1974.

The maximum mercury concentration of these records was 14 µg/L observed on May 3, 1988 in the Middle Sulphur River at Commerce, followed by 3.3 µg/L on June 18, 1986 in Big Sandy Creek near Big Sandy and 3.1 µg/L on December 15, 1992 in the N. Sulphur River near Cooper. However, as many as 25 out of the 33 (or 76%) stream segments have a maximum mercury concentration equal to or less than 1.0 µg/L.

The concentration of 14 µg/L in the Middle Sulphur River was greater than the second maximum value in the East Texas streams by 11.7 µg/L. Of the 18 measurements at the sampling site on the Middle Sulphur River, the second highest value was 0.2 µg/L and all the others were less than 0.1 µg/L. The extreme outlier of 14 µg/L does not seem to be representative of normal conditions. The flow on May 3, 1988 in the river was very low (0.05 cms), conductivity was high (492 µS/cm), and pH (8.1) was very high (US Geological Survey, 1988). These conditions may trigger complicated changes of inorganic mercury into high solubility of mercury (Morel & Herring, 1993).



1. Red River, DeKalb
2. Big Cypress Creek, Pittsburg
3. S. Sulphur River, Commerce
4. Big Cypress Creek, Jefferson
5. Mid Sulphur River, Commerce
6. N. Sulphur River, Cooper
7. Big Cypress Creek, Karnack
8. Little Cypress Creek, Jefferson
9. Sulphur River, Texarkana
10. S. Sulphur River, Cooper
11. Patman Lake, Texarkana
12. Sulphur River, Talco
13. Big Sandy Creek, Big Sandy
14. Sabine River, Beckville
15. Sabine River, Burkeville
16. Attoyac River, Chireno
17. Sabine River, Ruliff
18. Angelina River, Lufkin
19. Lake O'Pines, Jefferson
20. Lake O'Pines, above Dam, Jefferson
21. Big Cypress Creek, Lone Star
22. White Oak Creek, Omaha
23. White Oak Creek, Talco
24. Neches River, Town Bluff
25. Neches River, Rockland
26. Angelina River, HWY 63, Horger
27. Neches River, Evadale
28. Sabine River, Mineola
29. Sabine River, Tatum
30. Mill Creek, Burkville
31. Neches River, Chandler
32. Angelina River, Jasper
33. Sabine River, Wills Point

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Figure 1. Sites of the 33 stream segments with USGS historical records on mercury concentrations in East Texas

Median Hg concentration for each of the 33 stream segments was no different than other U.S. streams (Hodges, 1977). Simple statistics of mercury concentrations for stream segments having 20 measurements or more in the record are given in Table 2. All the means and medians were less than the EPA drinking water quality standard (2 µg/L) and the Texas fresh water quality standard for acute aquatic life protection (2.4 µg/L). Of the five different stream segments having the maximum mercury concentration higher than 2 µg/L in their entire records, all of the second maximum values were much less than 2 µg/L. No seasonal patterns were noticeable in the mercury concentrations in East Texas.

Table 2. Descriptive statistics of mercury concentrations (µg/L) for selected stream segments in East Texas.

Stream Segments	Mean	Median	Std. Dev.	Max	Min	No Obs	Period
Red River at DeKalb	0.13	0.05	0.20	0.9	0.00	48	1974-94
Little Cypress Creek, Jefferson	0.07	0.05	0.06	0.3	0.00	36	1982-94
S. Sulphur River, Cooper	0.09	0.05	0.09	.04	<0.1	38	1982-94
Sulphur River, Talco	0.05	0.05	0.05	0.2	0.0	41	1980-94
Sabine River, Beckville	0.07	0.05	0.06	0.2	0.00	29	1980-94
Sabine River, Burkeville	0.17	0.05	0.50	2.5	0.00	24	1974-85
Sabine River, Ruliff	0.09	0.05	0.10	0.5	0.0	46	1974-91
Neches River, Town Bluff	0.15	0.05	0.36	1.7	<0.1	21	1981-91
Neches River, Rockland	0.13	0.05	0.21	1.0	0.00	46	1974-94
Neches River, Evadale	0.10	0.05	0.22	1.5	0.00	47	1976-91

Note: All concentrations of <0.1 µg/L entered as 0.05 µg/L (or 50% of the detection limits) in the calculation (Newman et al. 1989

STREAM SAMPLES

Mercury concentrations in East Texas streams were also examined in two sampling schemes. One was conducted on December 12, 1995 in which six stream samples, six sediment samples, and one rainwater sample were collected from Nacogdoches County, Texas and were analyzed by Edna Wood Laboratories, Inc. Houston, Texas. The other was on March 11, 1996 in which water samples from the same six sampling sites in Nacogdoches County were collected for analyses in MBA Labs, Houston, Texas. Each water 1 L water sample was collected from the bank, kept in a polyethylene container, preserved by treating with HNO_3 (1 mL/L) to reduce pH below 2, and stored at 4°C before analysis. Sediment samples were collected for the surface 6 inches of the stream bottom using a soil core sampler. Both labs used the EPA approved Cold-Vapor Atomic Absorption Spectrometric Method. However, the mercury detection level is 1.0 µg/L in Edna Wood Labs and 0.2 µg/L in MBA Labs, both higher than the USGS detection limit (0.1 µg/L).

Results of the laboratory analyses are given in Table 3. All mercury concentrations at the six sampling sites were much less than the EPA and Texas water quality standards described previously. However, these were random samples for a few sites at two occasions. Their scientific implications in East Texas streams are very limited. A more systematic sampling scheme covering more stream segments and seasonal variations is necessary for a more comprehensive analysis of mercury concentrations in East Texas. Sediment samples augered from various depth levels in reservoirs may further provide insights on the temporal variation of mercury concentrations within the watershed.

Table 3. Mercury concentrations (µg/L) for samples collected in Nacogdoches County, Texas.

	December 12, 1995			March 11, 1996
Location	Water	Sediment	Rainwater	Water
Attoyac River at Hwy 7	<1.0	<0.02		0.2
Attoyac River at Hwy 103	<1.0	0.029		0.2
Waffelo Creek, Upstream	<1.0	<0.01		0.2
Waffelo Creek, Downstream	<1.0	<0.02		0.2
Terrapin Creek	<1.0	0.23		0.2
Angelina R. at Co. Rd 529	<1.0	<0.02		0.2
SFA Weather Station			<1.0	

In a more recent study, Twidwell (2000) examined mercury concentration-depth profiles in sediment cores collected from 11 reservoirs and two natural lakes in East Texas. In this study, mercury concentrations in surficial sections were higher than those in deeper strata in nearly every case. This implies that recent mercury inputs may be continuing to increase across the East Texas region. However, the same study also showed that there were no significant correlations between fish mercury concentrations and mercury concentrations in water or sediment. These relationships suggest that reservoir and lake processes are more important in controlling the production of methylmercury and its subsequent bioaccumulation in largemouth bass than mercury concentrations in water and sediment in East Texas.

IMPLICATIONS

Historical records of mercury concentrations in East Texas streams showed most readings below EPA and Texas water quality standards. Although there were a few samples had concentrations above these levels, no geographical patterns were noticeable. However, mercury concentration is not a routine measurement in the USGS surface water quality program. These sporadic measurements were usually separated by a few months or even longer than a year. It is difficult to track the causes of a single high mercury level while all the others are extremely low. Water samples collected in December 1995 and March 1996 from six different stream sites in Nacogdoches County had Hg concentrations similar to USGS historical records for East Texas streams. The historical records mentioned along with the current measurements seem to suggest no noticeable increases in Hg concentration in the last 20 years. In addition, mercury concentrations in marine fish tissues have remained relatively constant over the last 20 years (USEPA, 1997). However, these Hg concentrations in East Texas streams seem to be higher than those waters without pollution as reported by Fitsgerald (1979). But, because of the detection limits of the USGS and the present analysis, it is also difficult to determine if there is any Hg pollution involved in East Texas streams.

Many studies have shown that new reservoirs and flooding of adjacent land commonly causes an appreciable increase in the Hg content of fish (Bodaly et al. 1984; Kimmel and Groeger, 1986). This is due to either the release of Hg into aquatic ecosystem from flooded land or the enhancement of Hg mobility and bioavailability through microbial transformation of inorganic Hg into organic Hg, or both (Jackson, 1988). Although the recent study on sediment cores in 13 East Texas reservoirs and lakes suggested a continuing increase in mercury concentrations in recent years, all Hg concentrations in water and sediment were far less than the Texas acute and chronic quality criterion (Twidwell, 2000). Agricultural uses of mercury in East Texas have been relatively low. Furthermore, mercury-containing pesticides were deregistered in 1995, suggesting that agricultural activities are not likely to be responsible for current mercury levels. Atmospheric deposition from fossil fuel combustion associated with electrical power generation and petrochemical processing along the Gulf coast could be likely sources for atmospheric deposition, though a more comprehensive study involving precipitation and water sampling using very low detection limits would be required to determine the possible sources of mercury in East Texas waters.

No significant correlations were found between fish mercury concentrations and mercury concentrations in water or sediment. Fish mercury levels in six East Texas reservoirs range from 0.033 to 1.65 ppm (Texas Dept. of Health, 1995b). The average Hg concentrations among different species in these reservoirs are given in Table 4. The current U.S. Food and Drug Administration action level for mercury is 1 ppm based on considerations of health impacts (USEPA, 1997). This level is as much as 20,000 times greater than mercury levels in water. Generally, for a given a species, the concentration of mercury is greater in large fish than in small fish due to biological magnification. Consequently, the mercury concentration in stream sediments may be more important than the concentration in water. To reduce possible risks in health, one should consider the following: 1) consume smaller fish from a variety of waterbodies, 2) increase consumption interval, 3) avoid eating skin and fatty tissues, and 4) limit consumption of listed fish species to quantities recommended by the Texas Department of Health.

Table 4. Average mercury concentrations (ppm, wet weight) among different species in East Texas, 1994-95 (Texas Department of Health, 1995b).

Species	Caddo Lake	Big Cypress Creek	Lake O' Pines	Toledo Bend Reservoir	Sam Rayburn Reservoir	Steinhagen Reservoir
Largemouth Bass						
(All)	0.83	0.44	0.25	0.9	0.73	0.88
(14-18")		0.32	0.16	0.71	0.59	0.81
(>18")	1.10	0.94	0.55	1.06	0.82	1.19
White Bass	0.44	0.18		0.39		1.19
Crappie	0.47	0.06		0.402	0.3	0.55
Freshwater Drum	1.27	0.60		0.29	0.56	0.94
Blue Catfish				0.098	0.5	0.32
Channel Catfish	0.21	0.26	0.06	0.28	0.136	0.26
Flathead Catfish		0.95		0.28	0.5	
Common Carp		0.18	0.055			
Pickerel	0.96					
Sunfish	0.39					

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