

# Assessment of Mercury Concentrations in Water and Fish Tissue Analysis in Kaw Lake, Oklahoma, 2022

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## Abstract

The Kaw Nation has been collecting water, sediment, and fish samples from Kaw Lake and upper Arkansas River from 2007 to present to examine the concentrations of Mercury and other heavy metals to protect the health of the tribal members. Kaw Lake is in the North-Central part of Oklahoma. Kaw Lake is a permanent water body constructed in 1976 by the Army of Corps of Engineers. The Lake is consistently fed by the Arkansas River and other tributaries as runoff coming all the way from Colorado through Kansas to Kaw Lake of Oklahoma. The Lake has a surface area of 26.64 square miles (69 km<sup>2</sup>) and shoreline of 168 miles (270 km) with a total drainage area of 56,345 square miles (145,393 km<sup>2</sup>) and an average water depth of 8 meters. The water and fish samples were collected from 7 sites of Kaw Lake, once in a month and the fish samples once in a year during summertime, early July to end of July. The fish samples focused on 5 sport, predator, and bottom dwelling species of large consumable size, greater than 200 mm length and 560 grams weight. The five fish sampled were Catfish, White bass, Largemouth and Smallmouth bass, Black and White crappie. The fish and water samples were sent to Accurate Environmental Labs for detailed analysis. Predator species were analyzed as fillet and the bottom dwellings as a whole fish using EPA Method 7471A-M. Mercury from Water and Fish Tissue Samples were analyzed by Atomic Fluorescence Spectrometry. The laboratory analysis indicated that all the Mercury concentration in the fish samples except in Blue Catfish and Spotted Bass fall below the Maximum Contaminant Level (MCL) of 0.5 mg/kg.

## Keywords

Mercury Contamination, Fish Tissue, Bioaccumulation

## 1. Introduction

Mercury is a naturally occurring metal found throughout the environment. Most of the mercury found in the environment is in the form of metallic and inorganic mercury compounds [1] [2]. Metallic and inorganic mercury enters the air from mining deposits of ores that contain mercury, from the emissions of coal-fired power plants, burning municipal and medical waste, production of cement, and from uncontrolled releases in factories that use mercury [2]. Metallic mercury is liquid at room temperature, but some of the metal will evaporate into the air and can be carried long distances. In air, the mercury vapor can be changed into other forms of mercury and can be further transported to water or soil in rain or snow [3]. Inorganic mercury may also enter water or soil from the weathering of rocks that contain mercury, from factories or water treatment facilities that release water contaminated with mercury, and from incineration of municipal garbage that contains mercury (for example, in thermometers, electrical switches, or batteries that have been thrown away). Inorganic or organic compounds of mercury may be released to the water or soil if mercury-containing fungicides are used [4] [5].

Microorganisms such as bacteria, phytoplankton, and fungi convert inorganic mercury to methylmercury. Methylmercury released from microorganisms can enter the water or soil and remain there for a long time, particularly if the methylmercury becomes attached to small particles in the soil or water. Mercury usually stays on the surface of sediments or soil and does not move through the soil to underground water. If mercury enters the water in any form, it is likely to settle to the bottom where it can remain for a long time [5] [6].

Fish have two routes of mercury uptake. Fish concentrate mercury from water and through their diet (bioaccumulation). Fish typically accumulate only small amounts of methyl mercury through gill tissue and directly from the water column [7]. Majority of the mercury accumulation occurs through the food web or food chain. Bioaccumulation of mercury in fish is of concern because of potential human health effects from fish consumption, as well as potential effects of on fish eating phytoplankton [8]. The form of mercury that bioaccumulates in the food chain is methylmercury. Inorganic mercury does not accumulate up the food chain to any extent. When small fish eat the methylmercury in food, it goes into their tissues. When larger fish eat smaller fish or other organisms that contain methylmercury, most of the methylmercury originally present in the small fish will then be stored in the bodies of the larger fish. As a result, the larger and older fish living in contaminated waters build up the highest amounts of methylmercury in their bodies [9]. Saltwater fish (especially sharks and swordfish) that live a long time and can grow to a very large size tend to have the highest levels of mercury in their bodies. Plants (such as corn, wheat, and peas) have very low levels of mercury, even if grown in soils containing mercury at significantly higher rates than background levels. Mushrooms, however, can accumulate high levels if grown in contaminated soils [10]. Mercury levels in fish tend to

increase with age and size because of slow elimination of methyl mercury and increased intake as fish grow to larger size. Therefore, older, larger fish typically have higher mercury concentration in their tissue than younger fish of the same species [2].

The objective of this study was to examine and determine the concentration, distribution, and bioaccumulation of mercury in water and fish tissue in Kaw Lake of Oklahoma (**Figure 1**).

## 2. Materials and Methods

### 2.1. Study Design

The Kaw Nation Environmental Department in close cooperation with the Oklahoma Department of Wildlife Conservation (ODWC) conducted water, sediment, and fish samples from 4 sites of Kaw Lake. The sampling was conducted in the early mornings of late summer where most of the sport fish are harvested, and the level of the lake was down. The sampling was done in the southern part of the Lake at Sandy, Sarge and Washunga (**Figure 1**). Kaw Lake is a permanent water body constructed in 1976 by the Army of Corps of Engineers. The Lake is consistently fed by the Arkansas River coming all the way from Colorado through Kansas to Kaw Lake of Oklahoma. Other tributary creeks bring flood from agricultural areas, mining and water and wastewater treatment areas into the Lake as well. The Lake has a surface area of 69 km<sup>2</sup> and shoreline of 168 miles (270 km) with a total drainage area of 56,345 square miles (145,393 km<sup>2</sup>), Army Corps of Engineers [11]. The Lake has an average depth of 8 meters. The fish sampling was carried on a ODWC's motorboat where an electric shocker is mounted. To meet the National Fish Tissue Study Objectives, a composite sample of predator and bottom dwelling fish were decided to be sampled. Five individuals per composite were collected, all of which were large enough to provide sufficient tissue for analysis of the target analytes (**Table 1**). Each composite samples of the predators were more than 560 grams of edible tissue and 560 grams of total body tissue of bottom dwellers (**Table 2**). Based on US EPA 1995 guidance each composite had the same fish species and reached legal requirement of harvestable size, or weight with the smallest individual is no less than 75% of the largest individual [12] [13].

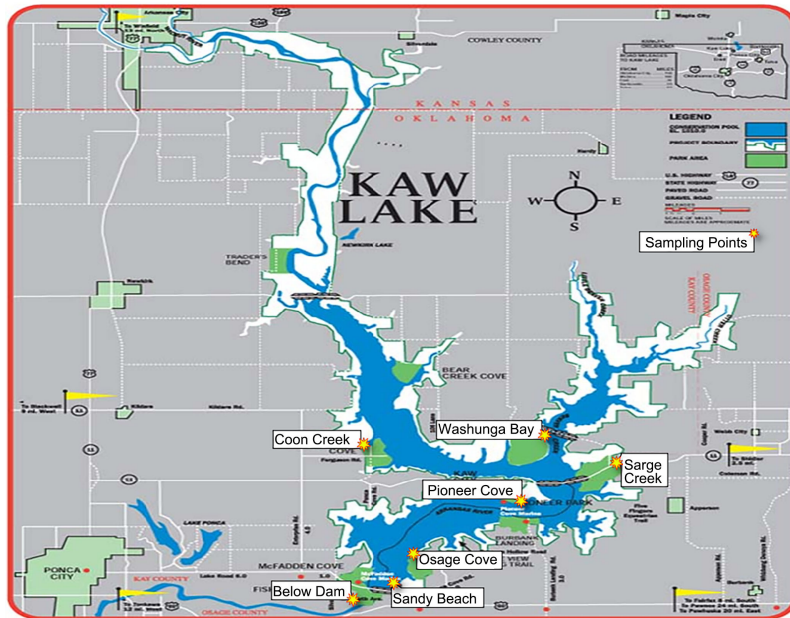
### 2.2. Field Sampling

The protocol for collecting fish and water samples in the field and subsequent processing in the laboratory are shown in **Figure 2**, **Table 1** and **Table 2**. The fish samples were collected with box nets, gill nets, trot lines, electroshocking and rod and reel. The samples were removed from the water, rinsed with ambient water, wrapped individually in aluminum foil, placed in polyethylene Ziploc<sup>®</sup> bags and placed on ice and delivered to Stillwater Accurate Laboratory within 24 hours of collection.

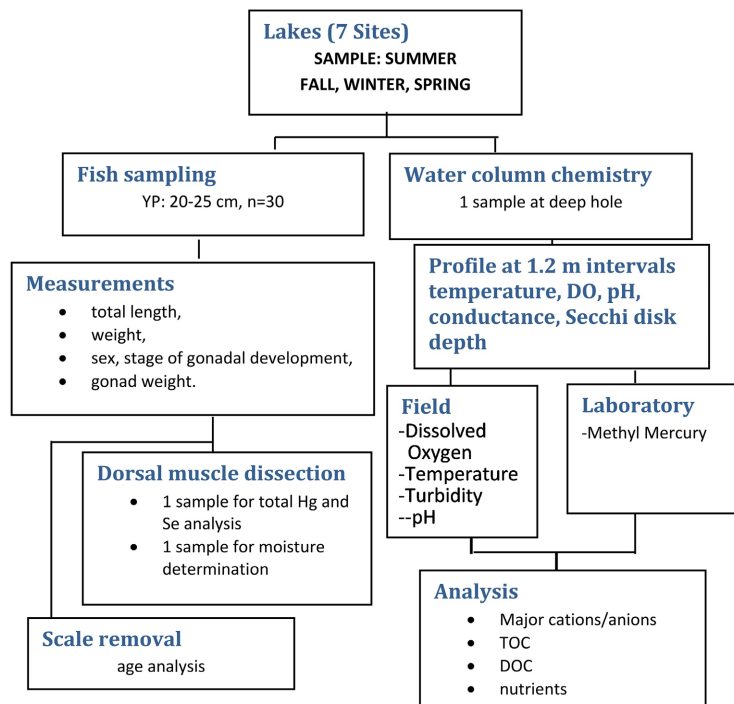
Justification for selecting target species were based on US EPA guidance for

assessing chemical contaminants data for fish advisories, Volume 1: Fish Sampling and Analysis, 3<sup>rd</sup> Edition, [14].

- 1) Abundance and preference of consumption in the study area.
- 2) May potentially accumulate high concentration of chemicals.
- 3) Species easy to identify.
- 4) Adult species are large enough to provide adequate tissue for analysis.



**Figure 1.** Locations of water, sediment and fish sampling sites of kaw lake (Map: ACOE).



**Figure 2.** Field and lab handling protocol.

**Table 1.** Target chemicals and analytic methods.

Analytic Method	Target Chemical
Total Mercury by oxidation, purge and trap, and cold vapor atomic spectrometry (Method 1631, Revision B with Appendix A—Digestion procedures for Total Mercury in tissue, Sludge, sediment, and soil.	<ul style="list-style-type: none"> <li>• Mercury</li> </ul>
Arsenic specialization Arsenic Generation Chromatographs and Atomic Adsorption Spectrometry (Method 1632, Revision A)	<ul style="list-style-type: none"> <li>• Arsenic (III)</li> <li>• Arsenic (IV)</li> <li>• Dimethylarsenic Acid (DMA)</li> <li>• Monomethylarsenic (MMA)</li> <li>• Total inorganic arsenic</li> </ul>
Organochlorine pesticides by Gas Chromatography/ Halide Specific Detector (GC/HSD) method 1656, Revision A).	<ul style="list-style-type: none"> <li>• 2,4' DDD Kepone</li> <li>• 2,4' DDE Methoxychlor</li> <li>• 4,4' DDT, Aldrin Dieldrin</li> <li>• Endosulfine</li> </ul>

**Table 2.** Target species.

	Family Name	Common Name	Scientific Name
Predator/ Game Fish Species	Centrarchidae	• Largemouth Bass	• <i>Micropterus salmoides</i>
		• Smallmouth Bass	• <i>Micropterus dolomieu</i>
		• Black Crappie	• <i>Promoxis nigromaculatus</i>
		• White Crappie	• <i>Promoxis annulants</i>
	Percichthyidae	White Bass	• <i>Morone Chrysops</i>
Bottom Dwelling Fish Species	Cyprinidae	Common Carp	• <i>Ictalurus carpio</i>
	Ictaluridae	Channel Catfish	• <i>Ictalurus punctatus</i>
		Blue Catfish	• <i>Ictalurus furcatus</i>

Basic water quality measurements (**Table 3**) were conducted at four stations of the lake at 1.2 m depth intervals with multiprobe field instruments. Temperature, pH, turbidity, dissolved oxygen, salinity, total dissolved solids, and conductivity were measured. Water samples were taken from 4 feet (1.2 m), 8 feet (2.4 m) and 12 feet (3.6 m) and composited to have adequate representation of the lake strata and the samples were sent for more analysis of major cations and anions (Na, K, Ca, Mg, Fe, Mn, SO<sub>4</sub>, Cl), dissolved organic carbon content (DOC), total organic carbon content (TOC), nitrate, nitrite nitrogen, total phosphorus, and ammonia. The analytical techniques used for each, and associated detection limits are provided in **Table 3** and **Table 4**.

**Table 3.** Analytical methods for water testing.

Analyte	Method Reporting Limit, mg/L	Method
Na	0.02	EPA 200.7
K	0.07	EPA 200.7
Ca	0.01	EPA 200.7
Mg	0.005	EPA 200.7
SO <sub>4</sub>	0.06	EPA 300
Cl	0.07	EPA 300
Fe	0.01	EPA 200.7
Mn	0.005	EPA 200.7
TOC	0.2	EPA 415.1
DOC	0.2	EPA 415.1
Alkalinity	0.25	EPA 310.1
NO <sub>2</sub>	0.003	EPA 300.0
NO <sub>3</sub>	0.002	EPA 300.0
NH <sub>3</sub>	0.001	APHA, 1998. Method 4500-NH <sub>3</sub> F
Tot. P	0.001	APHA, 1998. Method 4500-P E

### 2.3. Laboratory Procedure

According to US EPA guidelines [15] and [16], the lab analyzed different tissue fractions for predator composites (fillets) and bottom dweller composites (whole bodies) (Table 1 and Table 2) to obtain target chemicals. Analyzing fish fillets provides information for human health, while whole body analysis produces information for ecosystem health [16] [17].

Fish were processed for analysis of mercury in lateral muscle in accordance with US EPA procedures [13]. Total fish lengths and wet weights were recorded. The sex and reproductive condition of each fish was assessed by visual examination of gonads and classified as: immature; developing; ripe and spent. Gonad wet weights were recorded. Tissue moisture contents were determined for calculation of the dry weight basis of the mercury content of the tissues (Figure 2). The frozen fish tissue was thawed, chopped into manageable size (1 - 2 cubes) and added into the sample processor to completely homogenize the sample. Individual fish was composited and digested through Atomic Fluorescence Spectrometry. The whole fish was also eviscerated as described in US EPA Method 7471A [17] and (Figure 3).

Moisture content was determined on a duplicate tissue sample of the same size and from the same portion of the fillet as the sample for mercury analysis. Individual samples were gently blotted on laboratory tissue paper and their wet weights determined. They were then dried overnight at 104°C in an aluminum weighing dish and weighed again. The moisture content as a percent was calculated from the wet and dry weights of the tissues. Mercury concentrations of the samples were then calculated using the wet weight and the dry weight values.

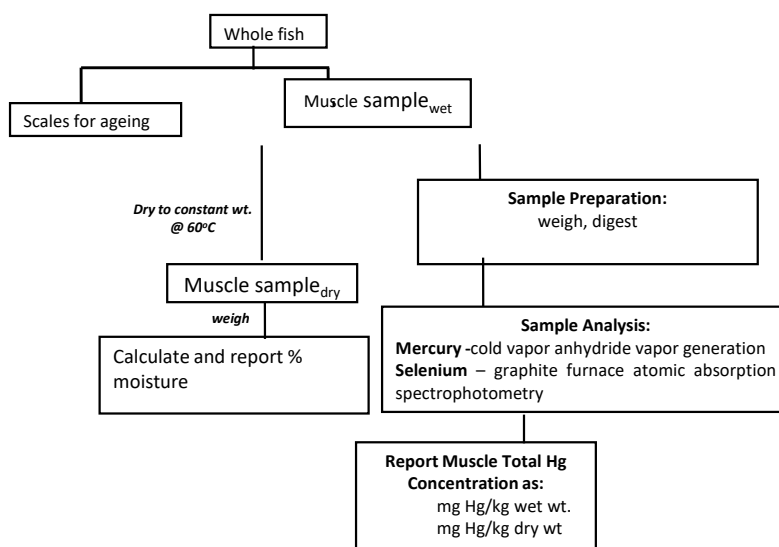


Figure 3. Laboratory processing protocol whole fish tissue analysis.

### 3. Results

#### 3.1. Water Quality Parameters or Measurements

To better understand patterns, dynamics, and changes in mercury accumulation in fish, Kaw Nation also collects water chemistry and in-situ measurements from each site. Kaw Nation measured the water chemistry parameters listed in Table 3 and Table 4 during the summer of 2008-2015.

Table 4. Water quality parameters or measurements.

Year	Temp (C degree)	Conductivity (uS/cm)	DO (mg/L)	pH	Turbidity (NTU)	Chlorophyll (ug/L)	Salinity (ppt)	TDS
2008	16.43	536	9.51	7.84	16.5	6.4	0.29	292
2009	24.33	807	7.40	7.98	9.2	4.7	0.41	547
2010	28.29	396	8.87	8.14	21.1	5.6	0.18	258
2011	26.18	326	10.46	8.51	13.8	7.5	0.64	842
2012	22.67	568	6.85	8.20	13.0	3.4	0.27	369
2013	26.07	763	6.05	8.10	12.8	3.7	0.37	496
2014	26.65	584	6.38	8.00	25.4	2.7	0.27	341
2015	26.71	158	7.60	8.51	7.5	4.4	0.29	394

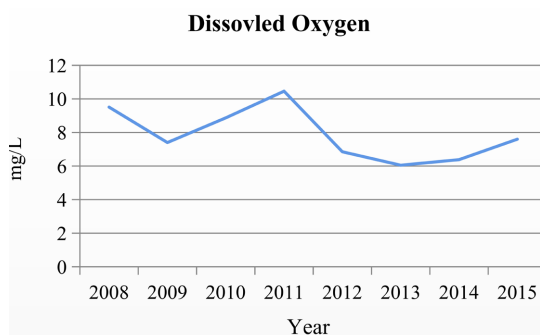
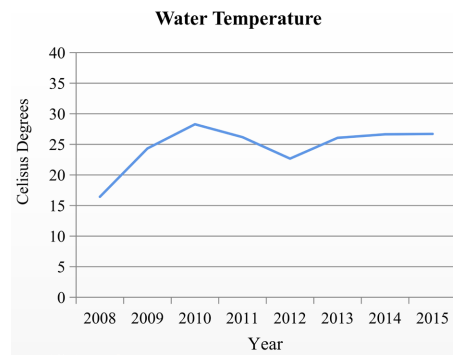
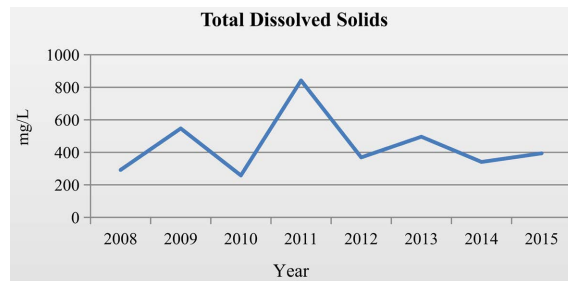


Figure 4. Dissolved oxygen of Kaw Lake.

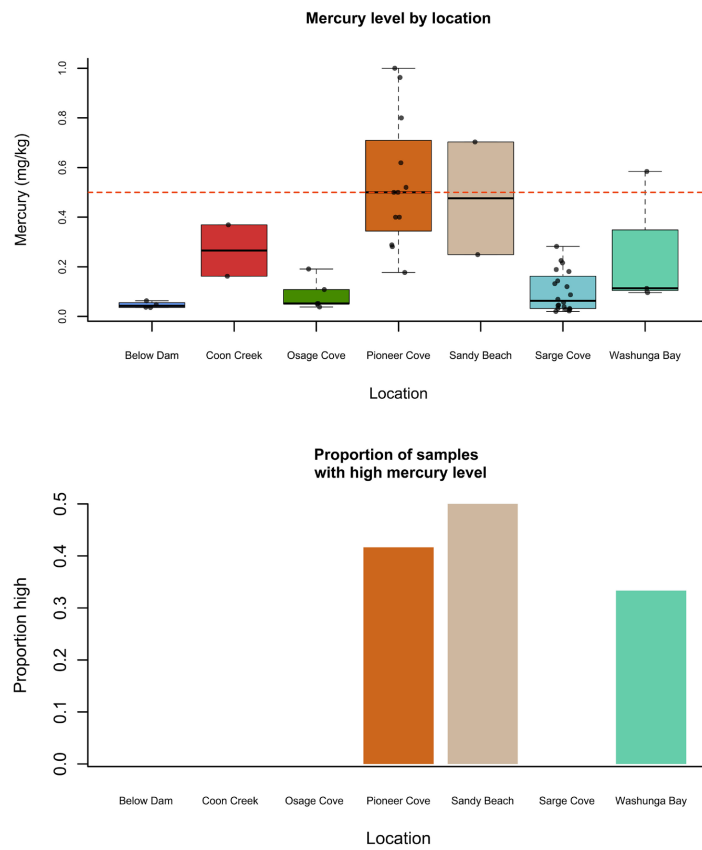


**Figure 5.** Water temperature of Kaw Lake.



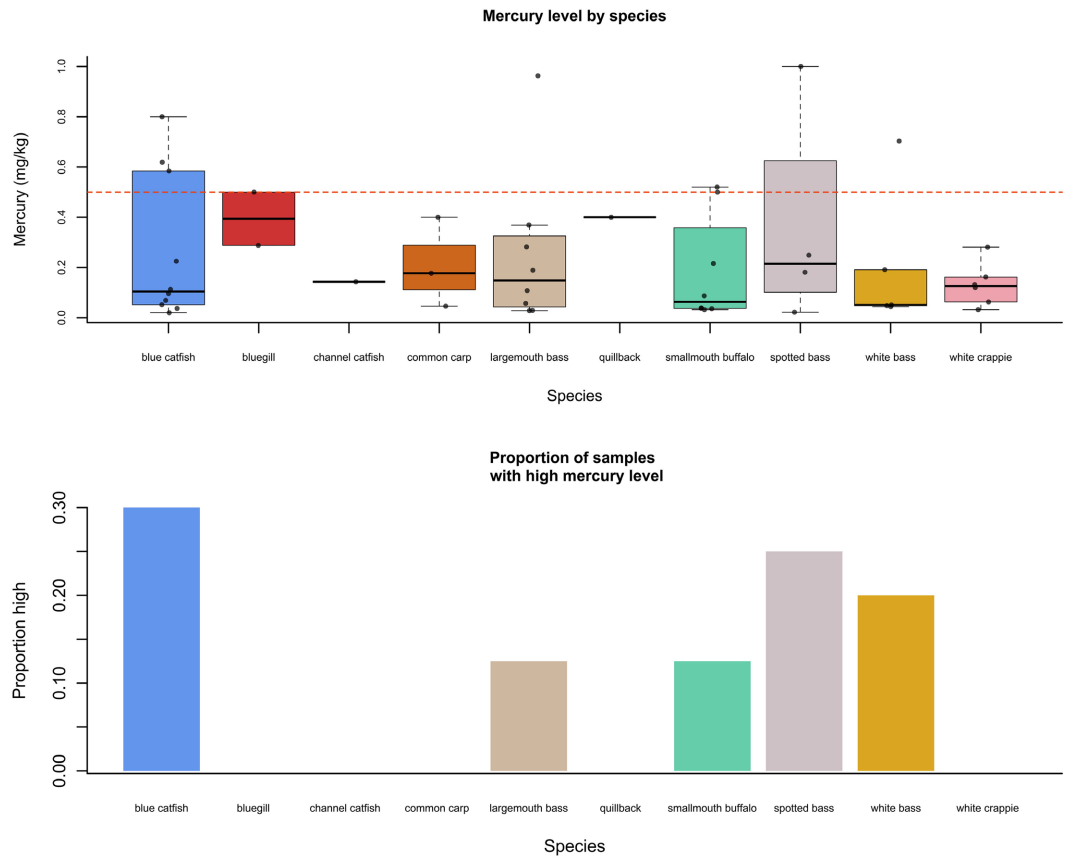
**Figure 6.** Total dissolved solids of Kaw lake.

### 3.2. Fish Tissue Analysis

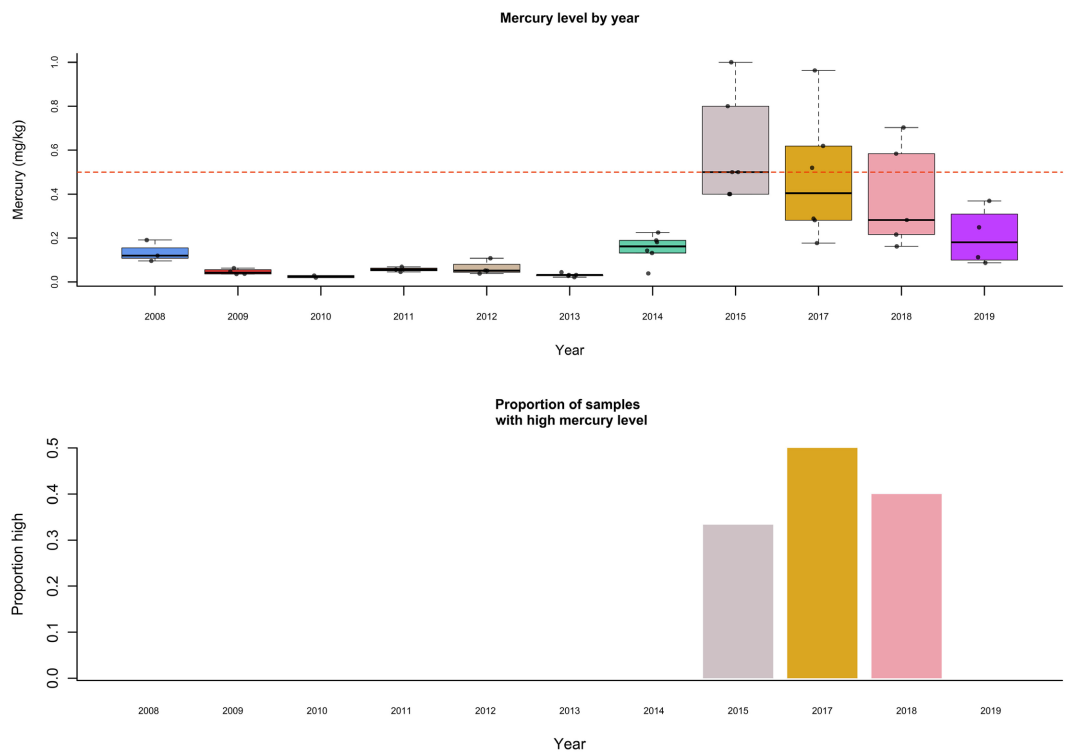


**Figure 7.** Mercury level by location. - - - - - = 0.5 mg/kg of Mercury, MCL.

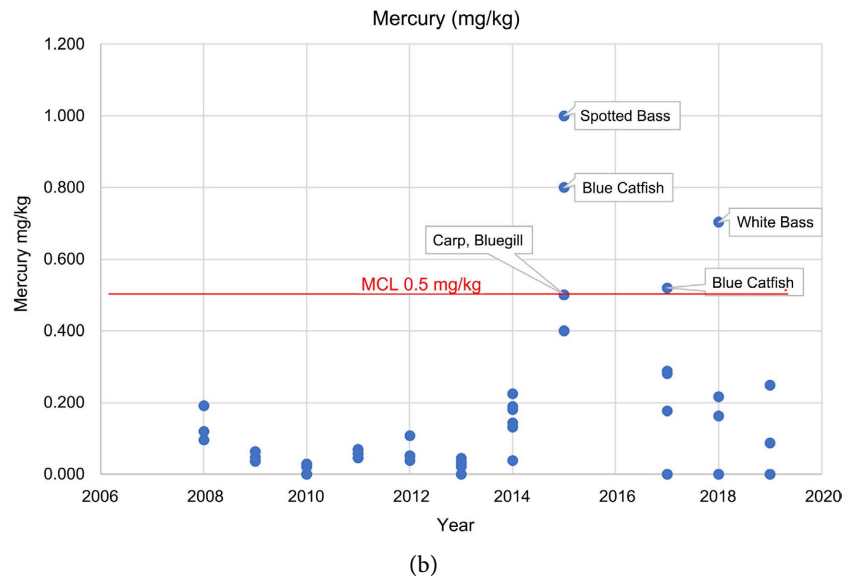




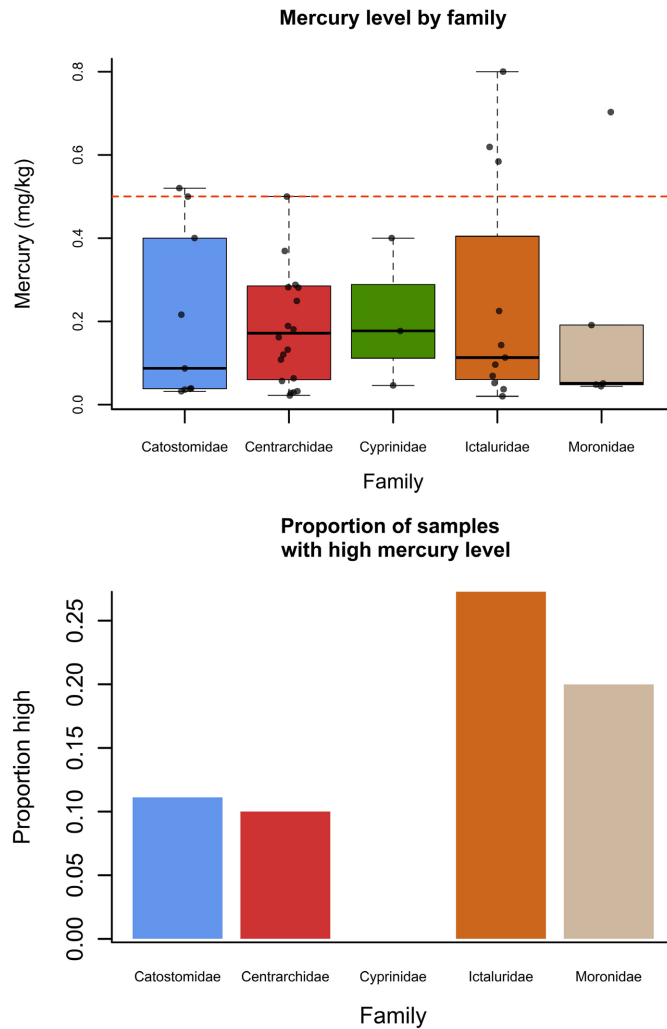
**Figure 8.** Mercury level by species. - - - - = 0.5 mg/kg of Mercury, MCL.



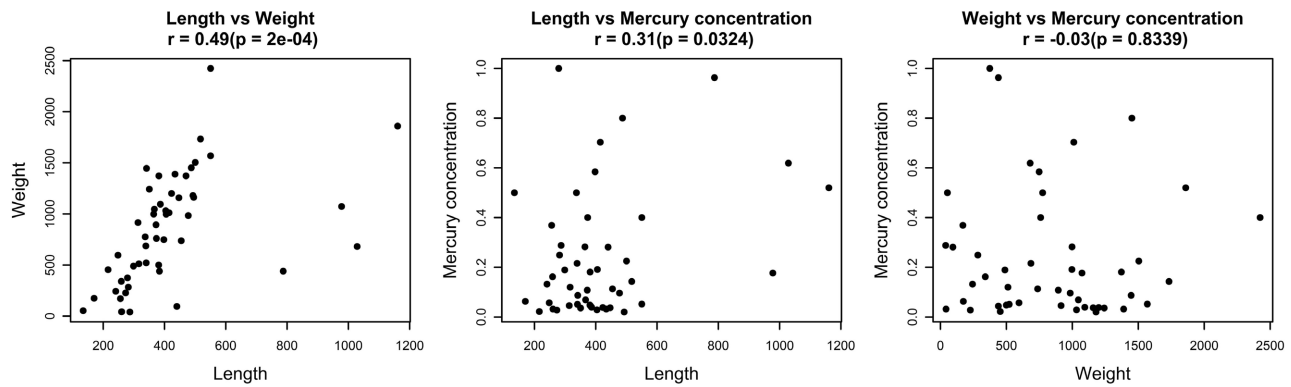
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**Figure 9.** Mercury level by year. ----- = 0.5 mg/kg of Mercury, MCL.



**Figure 10.** Mercury level by family.



**Figure 11.** Mercury concentrations by length and weight.

#### 4. Discussion

The presence and abundance of fish species can be related to water chemistry, physical habitat, and land-use activities to provide a more complete picture of water quality across the watershed. Although fish communities may have a high degree of natural variability, they can be useful indicators of ecosystem health [18]. Berkman and others [19] recommended fish be given consideration in biological water-quality monitoring of streams because they generally are perceived by the public to be ecologically relevant, and they are directly related to legislative mandates because of human health and endangered species concerns. Stream habitats vary from cool, clear, and forested headwater streams that have crystalline bedrock with high gradients and coarse substrates less vegetated, to that have low gradients and fine substrates of the Kaw Lake. Some of the major water quality parameters that can have influence on fish population are listed in **Table 4** and **Figures 4-6**.

Dissolved oxygen, temperature and total dissolved solids as indicated in **Figures 4-6**, are major indicators of water quality parameters and have significant impact on the survival and distribution of aquatic species including fish species. Water with high concentrations of dissolved materials above 800 mg/L have lower DO concentrations ( $<7$  mg/L) and could be a contributing factor for fish kills. Similarly, water with high temperature decrease oxygen solubility in water and creates stressful conditions for the survival of fish species. Direct discharge of pollutants including mercury from point source and non-point sources into river or lake would decrease water quality and at the same time, the fish species would be exposed to consume mercury through the food chain [20].

In this study we analyzed 5 species from top of the chain fish and generally these species have higher concentrations of mercury than those of lower trophic levels [21] [22]. Difference in mercury accumulation found in these fish species may be related to morphological difference, life cycle and food items of each species [23] [24]. Bioaccumulation of methyl mercury might be one factor for the increase in Hg concentrations in lake [25].

The feeding behavior of predatory species might contribute for an increased concentration of Hg. The other factor could be the nature of mobility or move-

ments such as migratory or sedentary [26]. Fish from lentic system have mercury contents higher than fish from the same trophic levels in lotic system, with the same concentration MeHg in water [27]. The other possible factor might be the land use or deforestation. Human factor may play an important role in the concentration of Hg. Impact of hydroelectric dam in increased MeHg and bioaccumulation in fish are not fully studied.

While fish consumption is the main source of exposure to methylmercury, the Oklahoma Department Environmental Quality (ODEQ) advises that the benefits and risks of eating fish should be balanced, since fish are an excellent source of high-quality protein and omega-3 fatty acids and are low in saturated fat. To protect Oklahomans from mercury poisoning, ODEQ issues fish consumption advisories which recommend limits for consumption of fish caught in Oklahoma waters. The Oklahoma Department of Environmental Quality have developed a downloadable fish advisory card. Although the ODEQ does not issue advisories on “Fish you buy” such as salmon, cod, pollock, sole, shrimp, mussels, and scallops, these species generally have lower mercury levels and are therefore considered safe for consumption [28].

#### **Bioaccumulation of Mercury**

Mercury in water is converted to methylmercury by bacteria and other processes. Fish absorb methylmercury from their food and from water as it passes over their gills. Mercury is tightly bound to proteins in all fish tissue.

According to the Vermont Department of Environmental Conservation [29], methyl mercury accumulates as you move up the food chain:

- 1) Methylmercury in the water and sediment is taken up by tiny animals and plants known as plankton.
- 2) Small fishes eat large quantities of plankton over time.
- 3) Large predatory fish consume many smaller fish, accumulating methylmercury in their tissues. The older and larger the fish, the greater the potential for high mercury levels in their bodies.
- 4) Fish are caught and eaten by humans and animals, causing methylmercury to accumulate in their tissues.

The Kaw Nation Environmental Department (KNED) has been monitoring the levels of mercury in fish tissue since 2001. Measurable concentrations of mercury have not been observed in all the samples collected from lakes and rivers within the watershed.

#### **Mercury Level by Location:**

Of all the fish sampling locations, **Figure 3**. Pioneer Cove and Sandy beach showed high levels of mercury concentrations above 0.5 mg/kg - 0.7 mg/kg followed by Washunga Bay with 0.35 mg/kg of mercury. Pioneer and Bay are the two busiest camping sites. Pioneer is a marine place where there are many boat activities that could discharge toxic chemicals to the air and water. The lowest site where there are low mercury concentrations is Below the Dam, Osage Cove and Sarge Creek or Cove (**Figure 7**).

#### Mercury Level by Species:

Fish species that fall below 0.5 mg/kg are the blue Catfish and Spotted bass, **Figure 2**. The proportion of fish sample species with a high mercury level are 30% Blue catfish followed by 25% of the spotted bass (**Figure 8**).

#### Mercury Level by Year:

Of all the fish species higher concentrations of mercury observed in the year 2015, 2017 and 2018 among spotted bass, white bass, and white crappie due to higher flooding. Total dissolved solids including mercury concentrations showed elevated levels (**Figure 9**).

#### Mercury Level by Fish Family:

Based on the fish family analysis **Figure 1**, all the fish species including Smallmouth buffalo (Catostomidae), Common carb (Cyprinidae), Blue catfish (Ictaluridae) and White bass (Moronidae) are all below 0.5 mg/kg of mercury concentrations. Out of all the fish family analyzed 25% of the sample proportion is blue catfish followed by 20% of the White bass. The other two-family species, the Smallmouth buffalo and White crappie (Centrarchidae) are in the proportion of 10% with high level of mercury (**Figure 10**).

#### Mercury Level by Length and Width:

The mercury concentrations were analyzed by correlating the weight and length of fish species as shown in **Figure 11**. The concentration of mercury in length and weight have shown good correlation as per US EPA guidelines of [13] that all edible size of fish greater than 560 grams of weight showed mercury concentrations.

## 5. Conclusions

Water samples and sport fish of edible size were collected for Hg analysis from 4 sites of Kaw Lake. Water sample was collected from each of the 4 sites. Five species of sport fish mainly, white bass, white and black crappie, Catfish, striped bass, large and small mouthed bass were analyzed for Hg concentration.

Mercury concentrations in the water sampled ranged below the quantification limit in all the 4 lake sites. All samples were less than US EPA aquatic life criteria (0.002 mg/L).

Concentration of Hg in fish tissue collected from 4 Kaw Lake sites was analyzed by family, species, and locations. Blue catfish and spotted bass were found higher in Methyl Mercury concentration 0.5 mg/kg as the Maximum Contaminants Level (MCL) set by US EPA. Pioneer and Washunga Bay sites where there are many marine activities have shown a higher concentration of methyl mercury above the US EPA MCL of 0.5 mg/kg.

Fish species heavier than 560 grams in weight and over 400 mm in length meaning at edible size have shown good correlation for mercury concentrations. Fish with over 400 mm of length have shown to have higher mercury concentrations indicating length to be one of the good indicators for mercury concentrations. Weight alone didn't show high mercury concentrations that could be because of limited fish sample size. According to Oklahoma Department of Envi-

ronmental Quality [28], fish tissue that had less than 0.5 mg/kg of mercury concentrations been edible at any time without any advisory note. According to Kaw Nation's of over 8 years laboratory fish tissue analysis, all small mouth buffalo, common carb, white bass are edible without any restriction. Fish tissue of blue catfish and spotted bass with mercury concentrations of 0.5 to 1.0 mg/kg are advised to be consume only two times per month.

Finally, this study agrees with the US Environmental Protection Agency Fish Advisories [30] that fish is an excellent, low-fat source of protein and other nutrients and an important part of a balanced diet. But some fish also contain unsafe levels of mercury. The amount of mercury in fish varies depending on the type of fish; their size, weight, and age; what they eat; and where they live. Smaller, non-predatory fish with shorter life spans tend to have lower levels of mercury. Larger, older fish that eat smaller fish tend to have the highest levels. Fish with an average level of less than 0.5 milligram of mercury per kilogram of body weight are considered safe for eating.

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### Author's Contributions

All authors equally contributed to this work.

### Ethics

This article is original and contains unpublished materials. The corresponding author confirms that all the other authors have read and approved the manuscript and no ethical issues involved.

### Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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