# Fish Contaminant Assessment of White, Muskegon, and Pentwater Lakes

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#### **Executive Summary**

The restoration targets for the Restrictions on Fish and Wildlife Consumption BUI in Muskegon Lake and White Lake Areas of Concern (AOCs) required a survey of mercury and PCB levels in the lakes and a comparison of contaminant concentrations to Pentwater Lake (a reference site). For the removal of the BUI, mercury and PCB concentrations in largemouth bass and carp should not show a statistically significant difference between the reference site and the AOC. The results of the 2011 sampling of largemouth bass and carp for Muskegon Lake, White Lake, and Pentwater Lake and comparisons with historical data found the following relationship between contaminant levels in the target fish species between the AOC and the reference system:

#### White Lake

- No statistically significant difference in mercury concentrations in largemouth bass and carp between the AOC and the Pentwater Lake reference site.
- No statistically significant difference in PCB concentrations in largemouth bass and carp between the AOC and the Pentwater Lake reference site.
- No statistically significant difference in the length and weight of largemouth bass and carp between the AOC and the Pentwater Lake reference site.
- An increasing trend in mercury concentrations in largemouth bass from 2006 2011 was found in the AOC and the Pentwater Lake reference site. This appears to be a regional phenomenon since the trend is present in the AOC and the reference site. A decreasing trend in PCB concentrations in largemouth bass from 2006 2011 was found in both the AOC and the Pentwater Lake reference site.
- A decreasing trend in mercury (1984 2011) and PCB concentrations (1980-2011) in carp was found in the AOC.

#### **Muskegon Lake**

- No statistically significant difference in mercury and PCB concentrations in largemouth bass between the AOC and the Pentwater Lake reference site.
- No statistically significant difference in mercury and PCB concentrations in carp between the AOC and the Pentwater Lake reference site.
- No statistically significant difference in the length and weight of largemouth bass and carp between the AOC and the Pentwater Lake reference site.
- Mercury concentrations in largemouth bass from 1986 20011 showed little change. Because mercury concentrations in largemouth bass from the Pentwater Lake reference site showed and increasing trend from 2006-2011, regional factors such as atmospheric deposition may be influencing the metal's distribution. A decreasing trend in PCB concentrations in largemouth bass from 2002 2011 was found in the AOC.
- A decreasing trend in mercury (2002 2011) and PCB concentrations (2002 2011) in carp from was found in the AOC.

These data demonstrate that restoration targets have been met in both AOCs and that the Restrictions on Fish and Wildlife Consumption BUI can be removed from the Muskegon Lake and White Lake AOCs.

#### Fish Contaminant Assessment of White, Muskegon, and Pentwater Lakes

#### Introduction

White Lake and Muskegon Lake are Areas of Concern (AOCs) that have the Restrictions on Fish and Wildlife Consumption Beneficial Use Impairment (BUI) in place due to historic discharges of anthropogenic chemicals and their accumulation in fish tissue. Both AOCs have developed numerical targets for delisting this BUI and need to assess the progress of restoration activities. The targets require fish contaminant monitoring data to support the removal of the BUI. Fish were collected and analyzed in 2006 from both AOCs and a reference system, Pentwater Lake (Rediske et al., 2009). The data from a second sample set is required to remove the BUI. The Annis Water Resources Institute (AWRI) collected common carp and largemouth bass in this project and analyze fish fillets for PCB congeners and mercury in a manner consistent with the MDEQ Fish Contaminant Monitoring Program. The delisting targets established by the Public Advisory Councils for the two AOCs utilize Pentwater Lake as a reference system and required that contaminant concentrations should not show a statistically significant difference between the reference site and the AOC. AWRI conducted statistical analyses on the 2006 and 2011 data to determine if the numerical targets were met for delisting the BUI.

#### **Methods**

#### Fish Collection

AWRI collected 20 largemouth bass (30-40 cm) and 20 common carp (40-55 cm) from White Lake, Muskegon Lake, and Pentwater Lake from the locations shown in Figures 1-3. Littoral habitats were sampled with boat electrofishing (Smith-Root 5.0 GPP control box). The same locations sampled in 2006 were evaluated in this investigation. Once identified and measured (length), fish were euthanized with a lethal dose (250 mg/L) of a standard fish anesthetic (tricaine methanesulfonate) kept on ice in the field and returned to the laboratory where they were wrapped in foil and frozen until analyzed.

#### Fish Tissue Preparation.

Fish tissue samples were prepared according to methods described by Jude et al., 2010. Fish tissue was prepared by homogenization in a stainless steel blender or Hobart 4822 grinder. Fish were filleted before homogenization and prepared as individual samples. Carp were analyzed as a skin off fillet and largemouth bass as a skin on fillet. Homogenized fish tissue (20 g) was mixed with 40 g. of sodium sulfate, spiked with the surrogate standards (CB-65 and CB-166; Cambridge Isotope Laboratories, Cambridge, Massachusetts, USA) and extracted in a soxhlet for 8 hours with 50:50 dichloromethane:hexane (Honeywell-Burdick & Jackson). Extracts were reduced to less than 2 mL using a Labconco Rapidvap and brought up to a 5 mL final volume with hexane. A 1-mL aliquot of the extract was used for gravimetric determination of lipids. A second 1-mL aliquot was passed through a column containing 10 g of 45% acidic silica gel

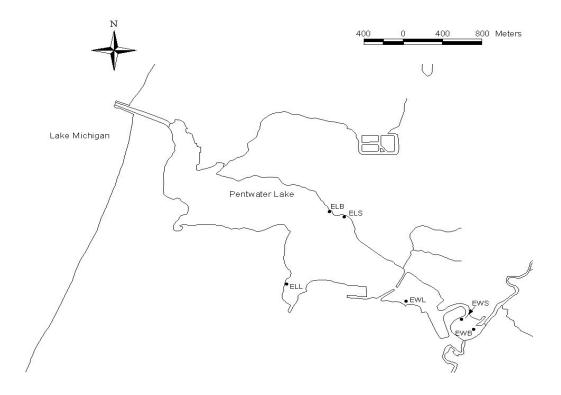


Figure 1. Pentwater Lake fish sampling locations (2006 and 2011).

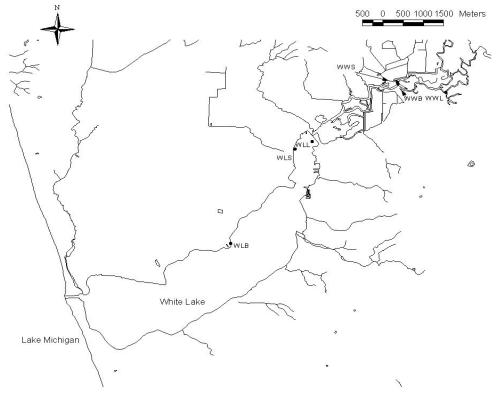


Figure 2. White Lake fish sampling locations (2006 and 2011).

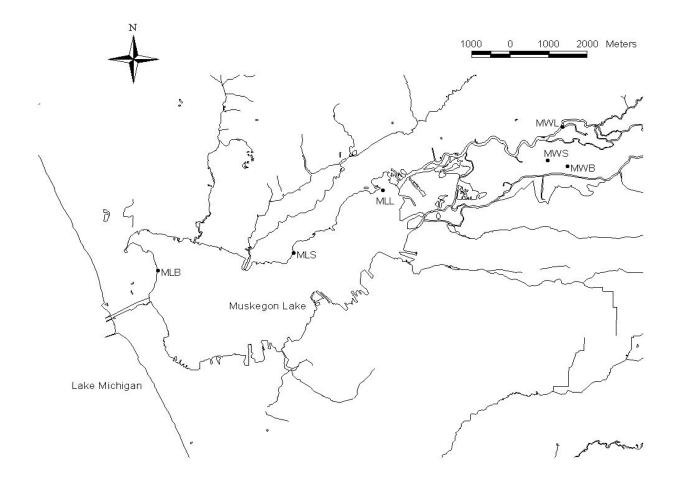


Figure 3. Muskegon Lake fish sampling locations (2006 and 2011).

(Kiesel gel, mesh size 230-400, Merck, Darmstadt, Germany) and a thin layer of sodium sulfate at the top. The column was cleaned with 150 mL of hexane prior to the transfer of sample extracts. Samples were then eluted with 200 mL of hexane and concentrated as above.

#### PCB Congener Analysis

Fish tissue samples were analyzed according to methods described by Jude et al., 2010. Identification and quantification of individual PCB congeners was accomplished with an Agilent 6890 series, high-resolution, gas chromatograph coupled to a 5973N quadrapole mass spectrometer. Separation was achieved by a fused-silica, capillary column coated with DB-XLB (60-m × 0.25-mm i.d.) at 0.25-µm film thickness (J&W Scientific, Folsom, CA). Column oven temperature was programmed from 80 to 160 °C at a rate of 40 °C/min and then to 170 °C at 10 °C/min, to 250 °C at 4 °C/min, and then to 296 °C at 8 °C/min with a final hold time of 10 min. Injector and transfer line temperatures were held at 260 and 250 °C, respectively. Hydrogen was

used as the carrier gas and <sup>13</sup>C-decachlorobiphenyl was used as an internal standard. The mass spectrometer was operated in negative chemical ionization mode using methane as a reagent gas. PCB congeners were determined by selected ion monitoring (SIM) at the two most intensive ions of the molecular ion cluster. Reported concentrations were not corrected from the recoveries of surrogate standards. Procedural blanks were passed through the whole analytical procedure to check for interferences and laboratory contamination. The instrument was calibrated by use of individual congener standards at five concentration levels from AccuStandard (New Haven, CT, USA). Concentrations for each of the 83 PCB congeners were determined. The following congeners were analyzed in the fish samples:

• PCB Congeners 6, 8, 9, 15, 16, 17, 18, 22, 25, 26, 28, 31, 32, 33, 37, 40, 41, 42, 44, 45, 46, 47, 48, 49, 52, 56, 60, 63, 64, 66, 70, 71, 74, 77, 82, 84, 87, 91, 92, 95, 97, 99, 101, 105, 110, 118, 123, 128, 132, 135, 136, 137, 138, 141, 144, 146, 149, 151, 153, 156, 158, 163, 170, 171, 172, 174, 176, 177, 178, 179, 180, 183, 185, 187, 190, 193, 194, 195, 196, 199, 202, 203, 206, 209.

Some congeners were reported as a pair if a co-elution existed. When this condition occurred, the response factor of the more dominate aroclor congener was used. Calibration accuracy was based on the ability to analyze Aroclor standards and obtain predicted amounts and ratios obtained by Frame et al. (4). Additional calibration verification was done using the West Coast Fish Studies standard supplied by AccuStandard. Appropriate quality control samples (blanks, matrix spikes, and duplicates) were analyzed to ensure precision and accuracy. Method blanks were run at a frequency of 1 per 20 samples and the mean  $\pm$  SE was 0.39  $\pm$  .05  $\mu$ g/kg. Matrix spikes were analyzed at a 5% frequency and recoveries ranged from 93 to 101%. Total PCBs was reported as the sum of the individual congeners. Lipid normalized PCBs was reported by dividing by the % lipid concentrations.

Mercury in fish tissue was analyzed by cold vapor atomic absorption according to EPA (1999) methods (3050/7140). Initial calibration was accomplished by analyzing 5 standards and a blank. Appropriate quality control samples (blanks, matrix spikes, and duplicates) and continuing calibration checks were analyzed to ensure precision and accuracy. Trace Analytical (Muskegon, MI) performed the mercury analyses.

Statistical analyses were performed using SPSS and SYSTAT.

#### **Results and Discussion**

#### White Lake

Ten largemouth bass samples were analyzesd from White Lake and Pentwater Lake for mercury and PCBs. For carp, ten samples were analyzed for mercury and twenty were analyzed for PCBs. The additional set of ten carp was analyzed to improve the statistical confidence of the data. The results of the PCB and mercury analyses in largemouth bass are shown in Table 1. Mean fish lengths for Pentwater and White Lake were 38.8 cm and 39.1 cm, respectively. Mean

Table 1. The results of PCB and Mercury Analyses Conducted on Largemouth Bass from White Lake and Pentwater Lake, 2011.

Species	Site	Length (cm)	Weight (g)	% Lipid	Total PCBs(ug/kg)	Lipid Normalized PCBs (ug/kg)	Mercury(mg/kg)
Largemouth Bass	Pentwater Lake	41.5	1050	0.28	12.8	4561	0.30
Largemouth Bass	Pentwater Lake	40.0	1082	0.73	31.2	4275	0.32
Largemouth Bass	Pentwater Lake	37.1	858	0.65	25.3	3897	0.25
Largemouth Bass	Pentwater Lake	41.8	982	0.41	19.5	4754	0.58
Largemouth Bass	Pentwater Lake	33.0	558	0.75	18.7	2487	0.33
Largemouth Bass	Pentwater Lake	40.1	974	0.66	24.9	3776	0.46
Largemouth Bass	Pentwater Lake	39.8	1003	0.50	12.9	2576	0.44
Largemouth Bass	Pentwater Lake	37.0	810	0.56	14.04	2507	0.34
Largemouth Bass	Pentwater Lake	37.3	844	0.61	28.08	4603	0.42
Largemouth Bass	Pentwater Lake	40.2	1133	0.66	23.5	3562	0.51
Pentwater Lake	Mean	38.8	929	0.58	21.1	3700	0.40
	Std. Error	0.9	56	0.05	2.2	299	0.03
Largemouth Bass	White Lake	41.5	870	0.46	13.1	2848	0.48
Largemouth Bass	White Lake	40.0	892	0.80	27.9	3484	0.32
Largemouth Bass	White Lake	37.1	1324	0.61	21.2	3471	0.49
Largemouth Bass	White Lake	41.8	906	0.47	17.1	3628	0.47
Largemouth Bass	White Lake	36.0	1030	0.44	20.2	4586	0.26
Largemouth Bass	White Lake	40.1	1009	0.56	25.8	4609	0.52
Largemouth Bass	White Lake	39.8	865	0.61	28.0	4584	0.42
Largemouth Bass	White Lake	37.0	964	0.50	20.4	4080	0.32
Largemouth Bass	White Lake	37.3	1036	0.51	16.4	3212	0.24
Largemouth Bass	White Lake	40.2	1030	0.83	34.6	4166	0.53
White Lake	Mean	39.1	993	0.58	22.5	3867	0.41
	Std. Error	0.7	45.1	0.05	2.2	209	0.04

fish weights for Pentwater and White Lake were 929 g and 993 g, respectively. There was no statistical difference between the fish lengths (t test; p>0.05) and weight (t test; p>0.05). Mean mercury concentrations for Pentwater and White Lake were 0.40 mg/kg and 0.41 mg/kg, respectively. There was no statistical difference between mercury concentrations in largemouth bass (t test; p>0.05). An Analysis of Covariance (ANCOVA) was performed on the mercury data with size as the covariant. There was no statistical difference in mercury concentrations with respect to site (p>0.05) however the concentrations did vary with size (p=0.017). The statistical power of the ANCOVA was 0.05. Boxplots of the mercury data are shown in Figure 4. Based on these results, there is no statistically significant difference between mercury concentrations in largemouth bass between Pentwater Lake and White Lake.

Mean PCB concentrations in largemouth bass for Pentwater Lake and White Lake were  $21.1\mu g/kg$  and  $22.5~\mu g/kg$ , respectively. There was no statistical difference between  $\log_{10}$  transformed PCB concentrations (t test; p>0.05) and lipid normalized PCB concentrations (t test; p>0.05) between the lakes. An ANCOVA was performed on the PCB data with size as the covariant. There was no statistical difference in PCB concentrations with respect to site (p>0.05). The concentrations did not vary with size (p>0.05). The statistical power of the ANCOVA was 0.071. An ANCOVA also was performed on the lipid normalized PCB data with size as the covariant. There was no statistical difference in PCB concentrations with respect to site (p>0.05). The concentrations did not vary with size (p>0.05). The statistical power of the ANCOVA was 0.067. Boxplots of the PCB and lipid normalized PCB data are shown in data Figures 5 and 6, respectively. The statistical power of the ANCOVA was 0.29. Based on these results, there is no statistically significant difference between PCB concentrations in largemouth bass between Pentwater Lake and White Lake.

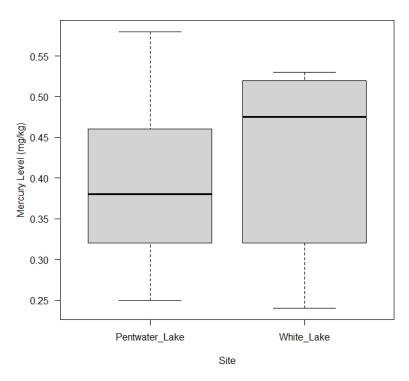


Figure 4. Box Plots of Mercury Concentrations in Largemouth Bass from Pentwater Lake and White Lake, 2011.

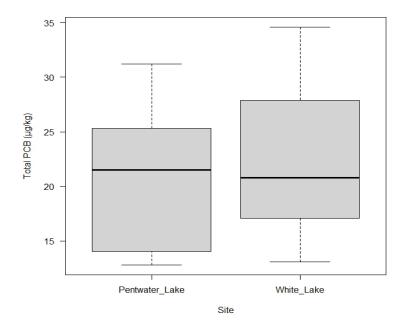


Figure 5. Box Plots of PCB Concentrations in Largemouth Bass from Pentwater Lake and White Lake, 2011.

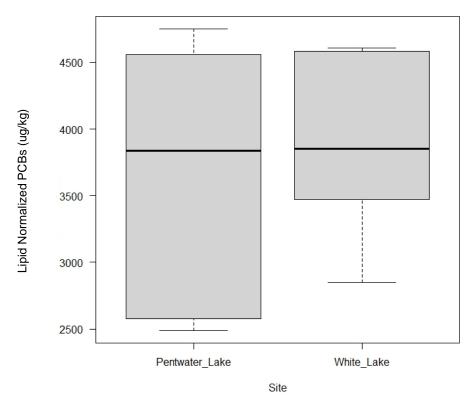


Figure 6. Box Plots of Lipid Normalized PCB Concentrations in Largemouth Bass from Pentwater Lake and White Lake, 2011.

The results of the PCB and mercury analyses in carp are shown in Table 2. Mean fish lengths for Pentwater Lake and White Lake were 64.2 cm and 65.6 cm, respectively. Mean fish weights for Pentwater Lake and White Lake were 3930 g and 4228 g, respectively. There was no statistical difference between the fish lengths (t test; p>0.05) and weight (t test; p>0.05). Mean mercury concentrations for Pentwater and White Lake were 0.13 mg/kg, respectively. There was no statistical difference between mercury concentrations in carp (t test; p>0.05). An ANCOVA was performed on the mercury data with size as the covariant. There was no statistical difference in mercury concentrations with respect to site (p>0.05) or size (p>0.05). The statistical power of the ANCOVA was 0.17. Boxplots of the mercury data are shown in Figure 7. Based on these results, there is no statistically significant difference between mercury concentrations in carp between Pentwater and White Lake.

Mean PCB concentrations in carp for Pentwater Lake and White Lake were  $107~\mu g/kg$  and  $109~\mu g/kg$ , respectively. There was no statistical difference between  $log_{10}$  transformed PCB concentrations (t test; p>0.05) and lipid normalized PCB concentrations (t test; p>0.05) between the lakes. An ANCOVA was performed on the  $log_{10}$  transformed PCB data with size as the covariant. There was no statistical difference in PCB concentrations with respect to site (p>0.05) however the concentrations did vary with size (p=0.001). The statistical power of the ANCOVA was 0.058. An ANCOVA also was performed on the  $log_{10}$  transformed lipid normalized PCB

Table 2. The results of PCB and Mercury Analyses Conducted on Carp from White Lake and Pentwater Lake, 2011.

Species	Site	Length (cm)	Weight (g)	% Lipid	Total PCBs(ug/kg)	Lipid Normalized PCBs(ug/kg)	Mercury(mg/kg
Carp	Pentwater Lake	63	3800	8.0	94	1169	0.21
Carp	Pentwater Lake	59.3	2380	6.0	60	998	0.14
Carp	Pentwater Lake	55.5	1530	7.81	69	878	0.06
Carp	Pentwater Lake	65.5	4880	11.27	66	588	0.16
Carp	Pentwater Lake	64.5	3970	6.25	44	701	0.16
Carp	Pentwater Lake	65.5	3860	7.8	89	1141	0.09
Carp	Pentwater Lake	68.0	4650	9.1	322	3542	0.17
Carp	Pentwater Lake	68.5	4990	8.0	126	1577	0.20
Carp	Pentwater Lake	62.4	3740	5.6	44	794	0.09
Carp	Pentwater Lake	57.3	2950	6.35	41	639	0.05
Carp	Pentwater Lake	65.0	3860	5.53	307	5552	NA
Carp	Pentwater Lake	65.2	4080	6.49	261	4024	NA
Carp	Pentwater Lake	67.1	4650	6.65	188	2827	NA
Carp	Pentwater Lake	66.2	4590	7.65	96	1261	NA
Carp	Pentwater Lake	71.4	4990	3.98	86	2168	NA
Carp	Pentwater Lake	68.3	5400	9.44	85	900	NA
Carp	Pentwater Lake	62.8	4540	3.02	72	2388	NA
Carp	Pentwater Lake	63.0	3520	4.69	37	790	NA
Carp	Pentwater Lake	62.6	3200	4.31	26	605	NA
Carp	Pentwater Lake	63.0	3010	2.51	25	1000	NA
	Mean	64.2	3930	6.5	107	1677	0.13
	Std. Error	0.9	223	0.5	20.8	311	0.02
Carp	White Lake	66.8	4360	9.78	180	1840	0.12
Carp	White Lake	63.5	3460	6.53	60	921	0.08
Carp	White Lake	64.5	3910	8.61	141	1633	0.09
Carp	White Lake	68.4	4710	5.00	53	1055	0.11
Carp	White Lake	62.2	3400	5.67	53	932	0.07
Carp	White Lake	65.1	4030	6.17	44	708	0.08
Carp	White Lake	68.2	4760	5.96	94	1577	0.15
Carp	White Lake	69.8	4990	8.05	233	2890	0.18
Carp	White Lake	69.7	4590	5.23	56	1061	0.20
Carp	White Lake	65.6	4990	6.5	69	1056	0.18
Carp	White Lake	75.6	5990	10.7	437	4084	NA
Carp	White Lake	70.3	6240	4.58	102	2233	NA
Carp	White Lake	70.5	5820	10.2	217	2127	NA
Carp	White Lake	63.3	4200	2.88	93	3212	NA
Carp	White Lake	62.5	3460	6.01	86	1431	NA
Carp	White Lake	73.2	5610	6.25	60	954	NA
Carp	White Lake	58.1	3270	3.74	50	1332	NA
Carp	White Lake	58.8	2040	2.60	60	2308	NA
Carp	White Lake	48.9	1810	4.04	32	790	NA
Carp	White Lake	67.0	2910	3.43	54	1574	NA
	Mean	65.6	4228	6.1	109	1686	0.13
	Std. Error	1.4	282	0.5	22.1	206	0.02

#### NA=not analyzed

data with size as the covariant. There was no statistical difference in PCB concentrations with respect to site (p>0.05) however the concentrations did vary with size (p=0.017). The statistical power of the ANCOVA was 0.15. Boxplots of the PCB and lipid normalized PCB data are shown in data Figures 8 and 9, respectively. Based on these results, there is no statistically significant difference between PCB concentrations in carp between Pentwater Lake and White Lake.

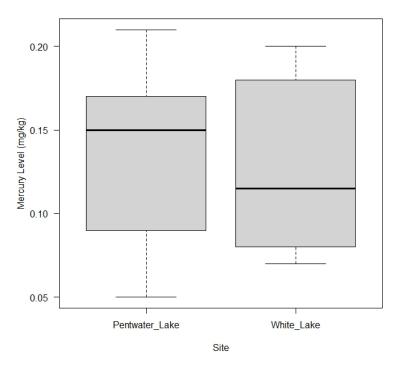


Figure 7. Box Plots of Mercury Concentrations in Carp from Pentwater Lake and White Lake, 2011.

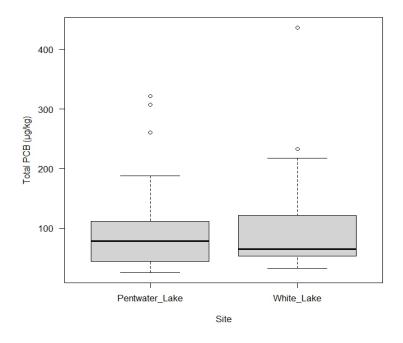


Figure 8. Box Plots of PCB Concentrations in Carp from Pentwater Lake and White Lake, 2011.

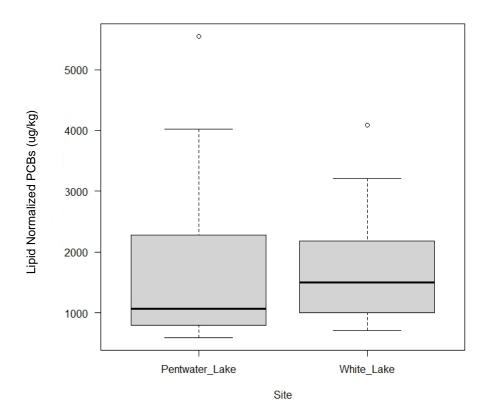


Figure 9. Box Plots of Lipid Normalized PCB Concentrations in Carp from Pentwater Lake and White Lake, 2011.

A comparison of 2006 and 2011 mercury and PCB data for largemouth bass and carp are shown in Figures 10 and 11, respectively. Mercury levels in largemouth bass increased in both White Lake and Pentwater Lake from 2006 (Rediske, 2009) to 2011 (Figure 10). The fact that an increase was observed in both lakes indicates that this was a regional issue and not related contamination in the White Lake AOC. PCB concentrations in largemouth bass decreased in both lakes over the same time period (Figure 11).

Historical mercury and PCB data are available from the MDEQ Fish Contaminant Data base (http://www.deq.state.mi.us/fcmp/) in addition to the 2006 information (Rediske 2009). Mercury and PCB data for White Lake carp are displayed in Figures 12 and 13, respectively. Mercury concentrations show a slight decreasing trend with a change occurring from 2004-2006 (Figure 12). In 2002, 85,000 cubic yards of sediment contaminated with mercury and other metals was removed from the Tannery Bay area of White Lake. PCB concentrations in carp show a greater degree of decline from 1980 – 2011 (Figure 13). In 2003, 12,000 cubic yards of PCB contaminated sediment was removed from the former Hooker Chemical Outfall in White Lake.

The White Lake Public Advisory Council (WLPAC) developed targets for delisting the Restrictions on Fish and Wildlife Consumption BUI (WLPC 2008). The targets require two

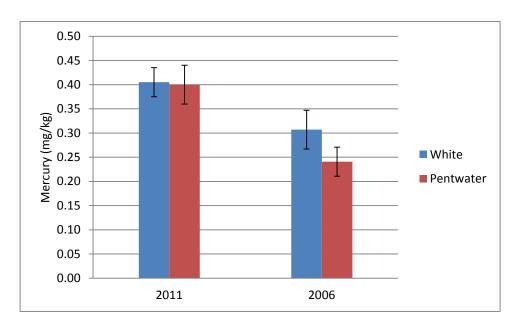


Figure 10. Comparison of Mercury Concentrations in Largemouth Bass from Pentwater and White Lake 2006 and 2011.

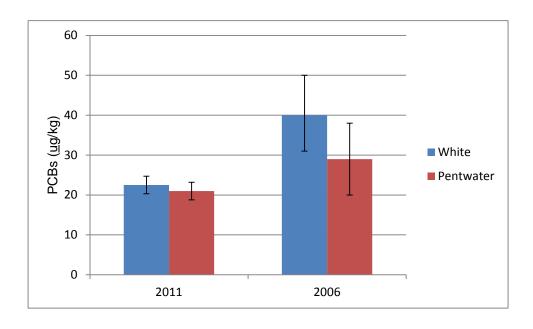


Figure 11. Comparison of PCB Concentrations in Largemouth Bass from Pentwater and White Lake 2006 and 2011.

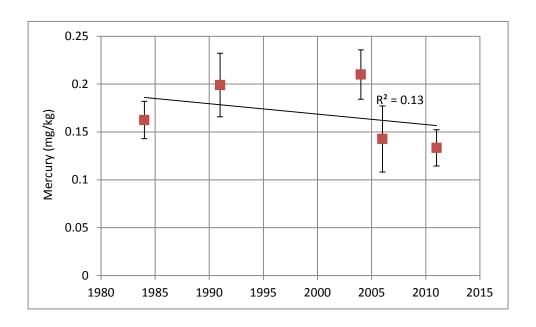


Figure 12. Comparison of Historical Mercury Concentrations in Carp from White Lake 1980 - 2011.

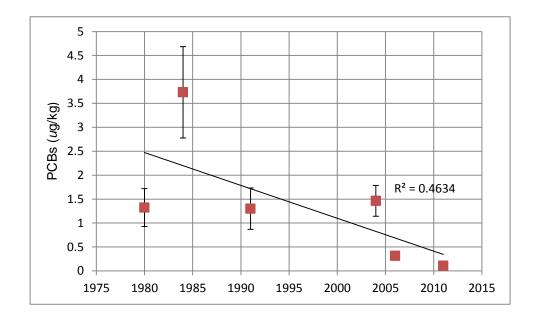


Figure 13. Comparison of Historical PCB Concentrations in Carp from White Lake 1980 - 2011.

rounds of fish sampling 5 years apart that show there was no statistical difference in mercury and PCB concentrations in carp and largemouth bass from White Lake and Pentwater Lake, non-AOC drowned river mouth lake. The results from 2006 found no statistically different results between PCB and mercury concentrations in the target fish species from Pentwater and White Lakes. Since the 2011 sampling also found no statistically different results between PCB and mercury concentrations in carp and largemouth bass from Pentwater and White Lake, the restoration targets for BUI removal have been achieved. In addition, historical data show a decreasing trend in PCBs in largemouth bass and carp and mercury in carp. Mercury largemouth bass appears to be increasing from 2006-20011 in White Lake; however the same pattern was present in the reference lake, suggesting a regional issue.

#### Muskegon Lake

Ten largemouth bass samples were analyzes analyzed from Muskegon Lake and Pentwater Lake were analyzed for mercury and PCBs. For carp, ten samples were analyzed for mercury and twenty were analyzed for PCBs. The additional set of ten carp was analyzed to improve the statistical confidence of the data. The results of the PCB and mercury analyses in largemouth bass are shown in Table 3. Mean fish lengths for Pentwater and Muskegon Lake were 38.8 cm and 38.1 cm, respectively. Mean fish weights for Pentwater and Muskegon Lake were 929 g and 839 g, respectively. There was no statistical difference between the fish lengths (t test; p>0.05) and weight (t test; p>0.05). Mean mercury concentrations for Pentwater Lake and Muskegon Lake were 0.40 mg/kg and 0.30 mg/kg, respectively. There was no statistical difference between mercury concentrations in largemouth bass (t test; p>0.05). An Analysis of Covariance

Table 3. The results of PCB and Mercury Analyses Conducted on Largemouth Bass from Muskegon Lake and Pentwater Lake 2011.

Species	Site	Length (cm)	Weight (g)	% Lipid	Total PCBs(ug/kg)	Lipid Normalized PCBs (ug/kg)	Mercury(mg/kg)
Largemouth Bass	Pentwater Lake	41.5	1050	0.28	12.8	4561	0.30
Largemouth Bass	Pentwater Lake	40.0	1082	0.73	31.2	4275	0.32
Largemouth Bass	Pentwater Lake	37.1	858	0.65	25.3	3897	0.25
Largemouth Bass	Pentwater Lake	41.8	982	0.41	19.5	4754	0.58
Largemouth Bass	Pentwater Lake	33.0	558	0.75	18.7	2487	0.33
Largemouth Bass	Pentwater Lake	40.1	974	0.66	24.9	3776	0.46
Largemouth Bass	Pentwater Lake	39.8	1003	0.50	12.9	2576	0.44
Largemouth Bass	Pentwater Lake	37.0	810	0.56	14.04	2507	0.34
Largemouth Bass	Pentwater Lake	37.3	844	0.61	28.08	4603	0.42
Largemouth Bass	Pentwater Lake	40.2	1133	0.66	23.5	3562	0.51
Pentwater Lake	Mean	38.8	929	0.58	21.1	3700	0.40
	Std. Error	0.9	56	0.05	2.2	299	0.03
Largemouth Bass	Muskegon Lake	34.3	632	0.52	24.7	4746	0.15
Largemouth Bass	Muskegon Lake	41.0	1088	0.30	10.6	3530	0.61
Largemouth Bass	Muskegon Lake	38.2	785	0.44	13.3	3030	0.29
Largemouth Bass	Muskegon Lake	38.5	846	0.54	11.7	2172	0.30
Largemouth Bass	Muskegon Lake	36.6	724	0.42	14.7	3490	0.14
Largemouth Bass	Muskegon Lake	37.8	834	0.41	13.1	3198	0.25
Largemouth Bass	Muskegon Lake	44.2	1318	0.38	12.0	3163	0.57
Largemouth Bass	Muskegon Lake	35.4	569	0.37	8.43	2278	0.27
Largemouth Bass	Muskegon Lake	37.7	829	0.57	20.5	3600	0.13
Largemouth Bass	Muskegon Lake	37.1	768	0.66	17.2	2612	0.26
Muskegon Lake	Mean	38.1	839	0.46	14.6	3182	0.30
	Std. Error	0.9	73	0.04	1.6	249	0.06

(ANCOVA) was performed on the mercury data with size as the covariant. There was no statistical difference in mercury concentrations with respect to site (p>0.05) however the concentrations did vary with size (p=0.001). The statistical power of the ANCOVA was 0.32. Boxplots of the mercury data are shown in Figure 14. Based on these results, there is no statistically significant difference between mercury concentrations in largemouth bass between Pentwater and Muskegon Lake.

Mean PCB concentrations in largemouth bass for Pentwater Lake and Muskegon Lake were 21.1 μg/kg and 14.6 μg/kg, respectively. PCB concentrations were significantly lower in Muskegon Lake (t test; p=0.02) and there was no significant difference in lipid normalized PCB concentrations (t test; p>0.05) between the lakes. An ANCOVA was performed on the PCB data with size as the covariant. PCB concentrations were significantly different with respect to site (p=0.019). The concentrations did not vary with size (p>0.05). The statistical power of the ANCOVA was 0.68. An ANCOVA also was performed on the lipid normalized PCB data with size as the covariant. There was no statistical difference in PCB concentrations with respect to site (p>0.05). The concentrations did not vary with size (p>0.05). The statistical power of the ANCOVA was 0.21. Boxplots of the PCB and lipid normalized PCB data are shown in data Figures 15 and 16, respectively. Based on these results, there is no statistically significant difference between PCB concentrations in largemouth bass between Pentwater and Muskegon Lake.

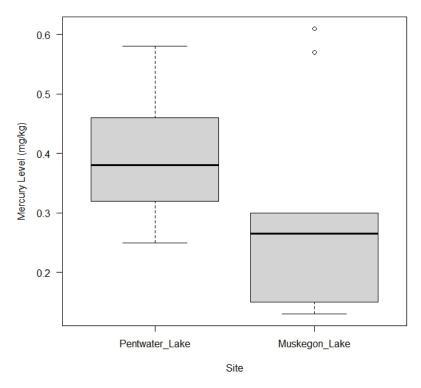


Figure 14. Box Plots of Mercury Concentrations in Largemouth Bass from Pentwater Lake and Muskegon Lake, 2011.

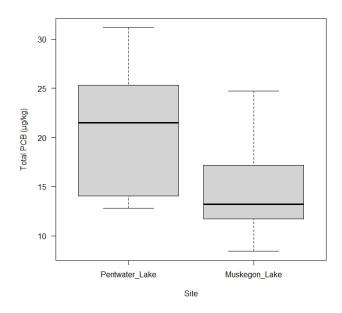


Figure 15. Box Plots of PCB Concentrations in Largemouth Bass from Pentwater Lake and Muskegon Lake, 2011.

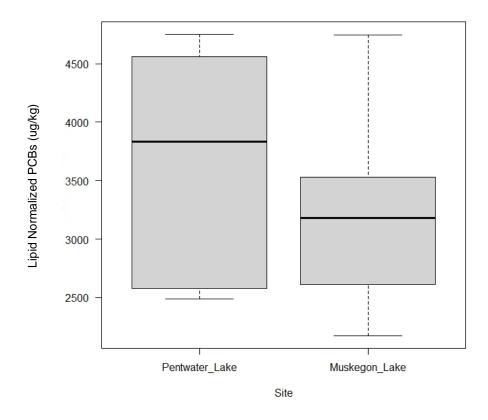


Figure 16. Box Plots of Lipid Normalized PCB Concentrations in Largemouth Bass from Pentwater Lake and Muskegon Lake, 2011.

The results of the PCB and mercury analyses in carp are shown in Table 4. Mean fish lengths for Pentwater Lake and Muskegon Lake were 64.2 cm and 65.4 cm, respectively. Mean fish weights for Pentwater Lake and Muskegon Lake were 3930 g and 4107 g, respectively. There was no statistical difference between the fish lengths (t test; p>0.05) and weight (t test; p>0.05). Mean mercury concentrations for Pentwater Lake and Muskegon Lake were 0.13 mg/kg, respectively. There was no statistical difference between mercury concentrations in carp (t test; p>0.05). An ANCOVA was performed on the mercury data with size as the covariant. There was no statistical difference in mercury concentrations with respect to site (p>0.05), however size was significant (p=0.013). The statistical power of the ANCOVA was 0.64. Boxplots of the mercury data are shown in Figure 17. Based on these results, there is no statistically significant difference between mercury concentrations in carp between Pentwater Lake and Muskegon Lake.

Mean PCB concentrations in carp for Pentwater Lake and Muskegon Lake were  $107 \,\mu g/kg$  each. There was no statistical difference between sin transformed PCB concentrations (t test; p>0.05) and  $\log_{10}$  transformed lipid normalized PCB concentrations (t test; p>0.05) between the lakes. An ANCOVA was performed on the sin transformed PCB data with size as the covariant. There was no statistical difference in PCB concentrations with respect to site and size (p>0.05). The statistical power of the ANCOVA was 0.087. An ANCOVA also was performed on the  $\log_{10}$  transformed lipid normalized PCB data with size as the covariant. There was no statistical difference in PCB concentrations with respect to site (p>0.05) however the concentrations did vary with size (p=0.007). The statistical power of the ANCOVA was 0.05. Boxplots of the PCB and lipid normalized PCB data are shown in data Figures 18 and 19, respectively. Based on these results, there is no statistically significant difference between PCB concentrations in carp between Pentwater Lake and Muskegon Lake.

Historical mercury and PCB data are available from the MDEQ Fish Contaminant Data base (http://www.deq.state.mi.us/fcmp/) in addition to the 2006 information (Rediske 2009). Mercury data for largemouth bass and carp are shown in Figures 20 and 21, respectively. Mercury levels in largemouth bass showed minimal change from 1986 to 2011 (Figure 20). The group of fish analyzed in 2006 was smaller than rest of the populations displayed in Figure 10, and consequently, the drop in concentration probably was influenced by size differences. Mercury concentrations in carp decreased in both lakes over the same time period (Figure 21). PCB data for Muskegon Lake largemouth bass and carp are displayed in Figures 22 and 23, respectively. PCB concentrations in largemouth bass show a strong decreasing trend from 1986-2006 (Figure 22). While size differences again may have influenced the 2006 data, a strong decreasing trend still was present (r<sup>2</sup>=0.87). PCB concentrations in carp show a similar degree of decline from 2002-2011 (Figure 23).

The Muskegon Lake Watershed Partnership (MLWPC) developed targets for delisting the Restrictions on Fish and Wildlife Consumption BUI (MLWPC 2007). The targets require two rounds of fish sampling 5 years apart that show there was no statistical difference in mercury and PCB concentrations in carp and largemouth bass from Muskegon Lake and Pentwater Lake, non-AOC drowned river mouth lake. The results from 2006 found no statistically different results

Table 4. The results of PCB and Mercury Analyses Conducted on Carp from Muskegon Lake and Pentwater Lake 2011.

Species	Site	Length (cm)	Weight (g)	% Lipid	Total PCBs(ug/kg)	Lipid Normalized PCBs(ug/kg)	Mercury(mg/kg
Carp	Pentwater Lake	63	3800	8.0	94	1169	0.21
Carp	Pentwater Lake	59.3	2380	6.0	60	998	0.14
Carp	Pentwater Lake	55.5	1530	7.81	69	878	0.06
Carp	Pentwater Lake	65.5	4880	11.27	66	588	0.16
Carp	Pentwater Lake	64.5	3970	6.25	44	701	0.16
Carp	Pentwater Lake	65.5	3860	7.8	89	1141	0.09
Carp	Pentwater Lake	68.0	4650	9.1	322	3542	0.17
Carp	Pentwater Lake	68.5	4990	8.0	126	1577	0.20
Carp	Pentwater Lake	62.4	3740	5.6	44	794	0.09
Carp	Pentwater Lake	57.3	2950	6.35	41	639	0.05
Carp	Pentwater Lake	65.0	3860	5.53	307	5552	NA
Carp	Pentwater Lake	65.2	4080	6.49	261	4024	NA
Carp	Pentwater Lake	67.1	4650	6.65	188	2827	NA
Carp	Pentwater Lake	66.2	4590	7.65	96	1261	NA
Carp	Pentwater Lake	71.4	4990	3.98	86	2168	NA
Carp	Pentwater Lake	68.3	5400	9.44	85	900	NA
Carp	Pentwater Lake	62.8	4540	3.02	72	2388	NA
Carp	Pentwater Lake	63.0	3520	4.69	37	790	NA
Carp	Pentwater Lake	62.6	3200	4.31	26	605	NA
Carp	Pentwater Lake	63.0	3010	2.51	25	1000	NA
	Mean	64.2	3930	6.5	107	1677	0.13
	Std. Error	0.9	223	0.5	20.8	311	0.02
Carp	Muskegon Lake	60.9	3120	4.87	23	473	0.15
Carp	Muskegon Lake	62.6	3460	3.29	15	457	0.08
Carp	Muskegon Lake	65.8	3400	3.65	30	816	0.10
Carp	Muskegon Lake	65.8	4030	7.08	112	1578	0.19
Carp	Muskegon Lake	59.8	2930	4.39	19	426	0.09
Carp	Muskegon Lake	63.9	3670	6.83	188	2747	0.18
Carp	Muskegon Lake	66.7	3740	6.91	75	1081	0.25
Carp	Muskegon Lake	64.4	3740	7.21	99	1379	0.10
Carp	Muskegon Lake	70.5	5670	8.65	162	1876	0.11
Carp	Muskegon Lake	58.0	3175	5.53	114	2055	0.07
Carp	Muskegon Lake	61.2	3630	5.52	91	1649	NA
Carp	Muskegon Lake	70.0	4510	9.07	210	2315	NA
Carp	Muskegon Lake	70.4	5930	9.19	150	1632	NA
Carp	Muskegon Lake	66.9	4990	6.97	235	3372	NA
Carp	Muskegon Lake	69.4	5670	7.57	125	1651	NA
Carp	Muskegon Lake	69.0	5200	6.62	121	1828	NA
Carp	Muskegon Lake	67.5	4200	5.12	131	2559	NA
Carp	Muskegon Lake	61.2	3570	4.77	109	2289	NA
Carp	Muskegon Lake	64.5	3350	4.53	34	760	NA
Carp	Muskegon Lake	68.5	4160	5.59	104	1860	NA
Mean	Mean	65.4	4107	6.2	107	1640	0.13
Std. Error	Std. Error	0.9	211	0.4	14	185	0.02

NA=not analyzed

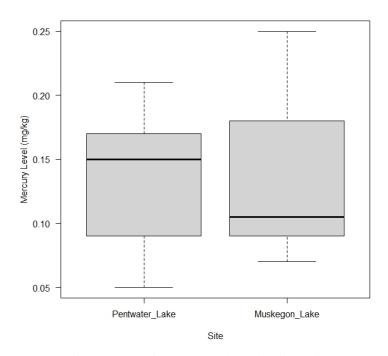


Figure 17. Box Plots of Mercury Concentrations in Carp from Pentwater Lake and Muskegon Lake, 2011.

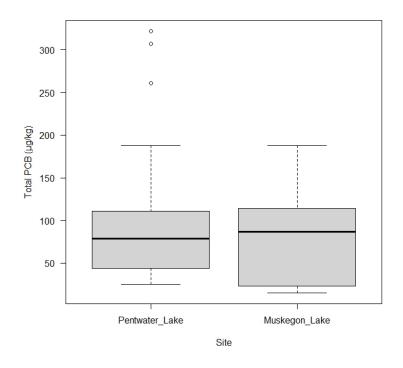


Figure 18. Box Plots of PCB Concentrations in Carp from Pentwater Lake and Muskegon Lake, 2011.

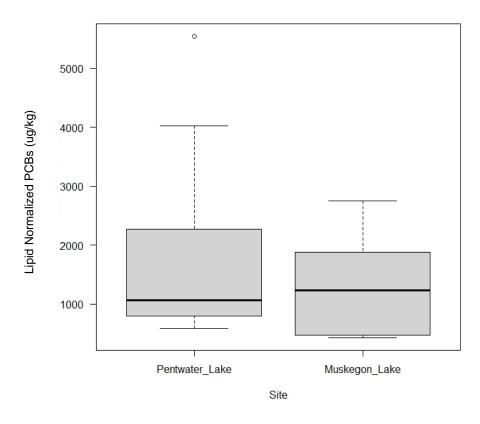


Figure 19. Box Plots of Lipid Normalized PCB Concentrations in Carp from Pentwater Lake and Muskegon Lake, 2011.

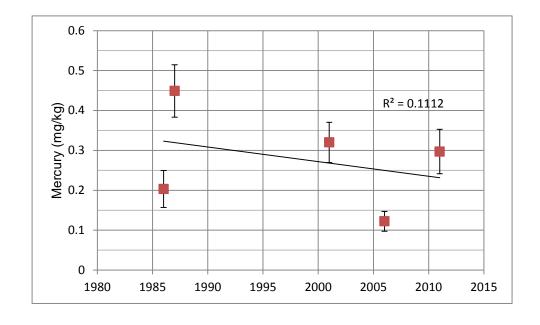


Figure 20. Comparison of Historical Mercury Concentrations in Largemouth Bass from Muskegon Lake 1986 - 2011.

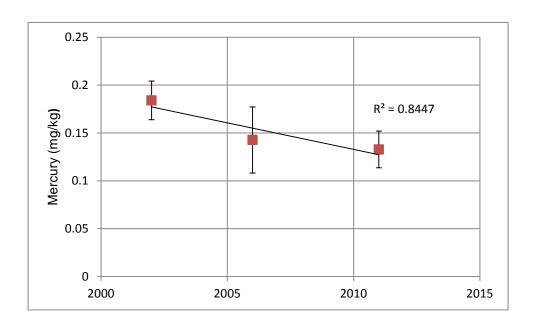


Figure 21. Comparison of Mercury Concentrations in Carp from Muskegon Lake 1986 - 2011.

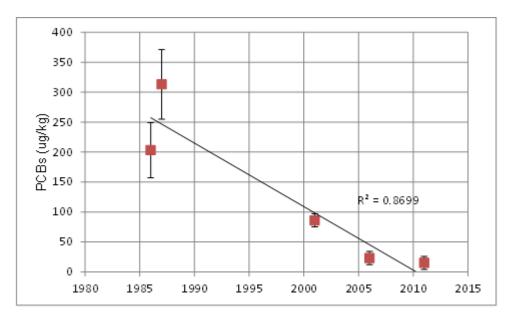


Figure 22. Comparison of Historical PCB Concentrations in Largemouth Bass from Muskegon Lake 2002 - 2011.

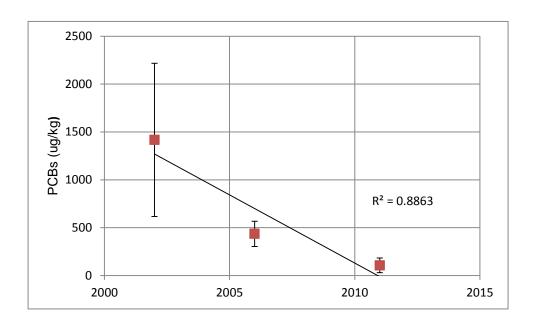


Figure 23. Comparison of Historical PCB Concentrations in Carp from Muskegon Lake 2002 - 2011.

between PCB concentrations in the target fish species from Pentwater Lake and Muskegon Lakes, however the fish from Pentwater Lake were significantly larger and contained more lipids. Mercury was significantly lower in largemouth bass from Muskegon Lake ( $\rho$ =0.01) than Pentwater Lake and there was no statistical difference between levels in carp. Since the 2011 sampling also found significantly lower PCB concentrations and similar mercury levels in carp and largemouth bass from Muskegon Lake and Pentwater Lake with fish of comparable size distributions, the restoration targets for BUI removal have been achieved. In addition, historical data show a decreasing trend in PCBs in largemouth bass and PCBs and mercury carp. While a decreasing trend in mercury concentrations of largemouth bass could not be demonstrated in the historical data set due to smaller fish sizes in 2006, the Pentwater Lake reference site showed an increasing trend over the same time period. This suggests that regional factors such as atmospheric deposition are resulting in increasing or steady state mercury levels in largemouth bass.

#### **Conclusions**

The restoration targets for the Restrictions on Fish and Wildlife Consumption BUI in Muskegon Lake and White Lake Areas of Concern required a survey of mercury and PCB levels in the lakes and a comparison of concentrations to Pentwater Lake (a reference site). For the removal of the BUI, mercury and PCB concentrations in largemouth bass and carp should not show a statistically significant difference between the reference site and the AOC. The results of the 2011 sampling of largemouth bass and carp for Muskegon Lake, White Lake, and Pentwater Lake and comparisons with historical data found the following relationship between contaminant levels in the target fish species between the AOC and the reference system:

#### White Lake

- No statistically significant difference in mercury concentrations in largemouth bass and carp between the AOC and the Pentwater Lake reference site.
- No statistically significant difference in PCB concentrations in largemouth bass and carp between the AOC and the Pentwater Lake reference site.
- No statistically significant difference in the length and weight of largemouth bass and carp between the AOC and the Pentwater Lake reference site.
- An increasing trend in mercury concentrations in largemouth bass from 2006 2011 was found in the AOC and the Pentwater Lake reference site. This appears to be a regional phenomenon since the trend is present in the AOC and the reference site. A decreasing trend in PCB concentrations in largemouth bass from 2006 2011 was found in both the AOC and the Pentwater Lake reference site.
- A decreasing trend in mercury (1984 2011) and PCB concentrations (1980-2011) in carp was found in the AOC.

#### Muskegon Lake

- No statistically significant difference in mercury and PCB concentrations in largemouth bass between the AOC and the Pentwater Lake reference site.
- No statistically significant difference in mercury and PCB concentrations in carp between the AOC and the Pentwater Lake reference site.
- No statistically significant difference in the length and weight of largemouth bass and carp between the AOC and the Pentwater Lake reference site.
- Mercury concentrations in largemouth bass from 1986 20011 showed little change. Because mercury concentrations in largemouth bass from the Pentwater Lake reference site showed and increasing trend from 2006-2011, regional factors such as atmospheric deposition may be influencing the metal's distribution. A decreasing trend in PCB concentrations in largemouth bass from 2002 2011 was found in the AOC.
- A decreasing trend in mercury (2002 2011) and PCB concentrations (2002 2011) in carp from was found in the AOC.

These data demonstrate that restoration targets have been met in both AOCs and that the Restrictions on Fish and Wildlife Consumption BUI can be removed from the Muskegon Lake and White Lake AOCs.

#### References

- EPA 1999. Test Methods for Evaluating Solid Waste Physical/Chemical Methods. U.S. Environmental Protection Agency. SW-846, 3rd Edition.
- Jude. D.J. R. Rediske, J. O'Keefe, S. Hensler, J. Giesy. 2010. PCB concentrations in walleyes and their prey from the Saginaw River, Lake Huron: A comparison between 1990 and 2007. Journal of Great Lakes Research. 36(2): 267-276.

- MLWP 2007. Final Delisting Targets for the Muskegon Lake Area of Concern. Muskegon Lake Watershed Partnership Muskegon, MI.
- Rediske, R.R. 2009. Assessment of Persistent Bioaccumulative Toxic Chemicals in Michigan Fish from Several Trophic Levels.. Report to Michigan Department of Environmental Quality. Lansing, MI.
- WLPAC 2008. Final Delisting Targets for the White Lake Area of Concern. White Lake Public Advisory Council. Whitehall, MI.