

**ENVIRONMENTAL
MONITORING FOR MERCURY
IN MASSACHUSETTS**

**STUDIES STATUS REPORT
1994 - 2004**

November 2004

Office of Research and Standards
Massachusetts Department of Environmental Protection
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Boston MA 02108

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Executive Summary

The Commonwealth of Massachusetts has been conducting studies on understanding the dynamics and distribution of mercury in tissues of freshwater fish for two decades. The primary goal of the early work was to identify fish populations that posed unacceptable health risks to fish consumers. In 1994, the first comprehensive statewide examination of mercury in freshwater fish began. This study was followed in 1999 by a study of fish mercury concentrations in an area of the state thought to have regionally high atmospheric deposition of mercury. As part of continuing efforts to elucidate the status of the Commonwealth's freshwater fish populations with respect to mercury contamination, several additional studies have been carried out. These studies were designed to provide insight into long-term trends in freshwater fish mercury concentrations, to estimate the magnitude of seasonal variability in mercury measurements, to document the comparative differences in mercury partitioning in lake food webs, to summarize the state of knowledge of mercury in other wildlife, and to begin to discern the historical picture of mercury deposition to freshwater sediments. The data generated from the studies on mercury concentrations in edible tissue of popular freshwater fish permit more widespread screening of the Commonwealth's lakes for potential health threats posed by eating contaminated fish. Health threats are addressed through the issuance of fish consumption advisories by the Massachusetts Department of Public Health.

In 2001, a long-term monitoring protocol was developed for fish tissue mercury studies to enable charting of temporal changes in fish tissue mercury concentrations, and to determine whether recently imposed mercury emissions controls on municipal solid waste incinerators correlate with lower mercury in fish. A component of the study included intensified seasonal sampling in a subset of long-term monitoring lakes to document the magnitude of seasonal variation in fish tissue mercury concentrations. This information will be used to improve the designs of future fish sampling efforts and to provide a perspective on the scale of natural variability in tissue mercury concentrations for comparison with other sources of variation.

Previous work revealed that spatially proximate lakes similar in physical features could have fish with substantially differing amounts of mercury. In order to examine the underlying reasons for these differences, we selected two nearby similar ponds in northeastern MA for a comparative analysis of the food web concentrations of mercury and the environmental characteristics of each pond.

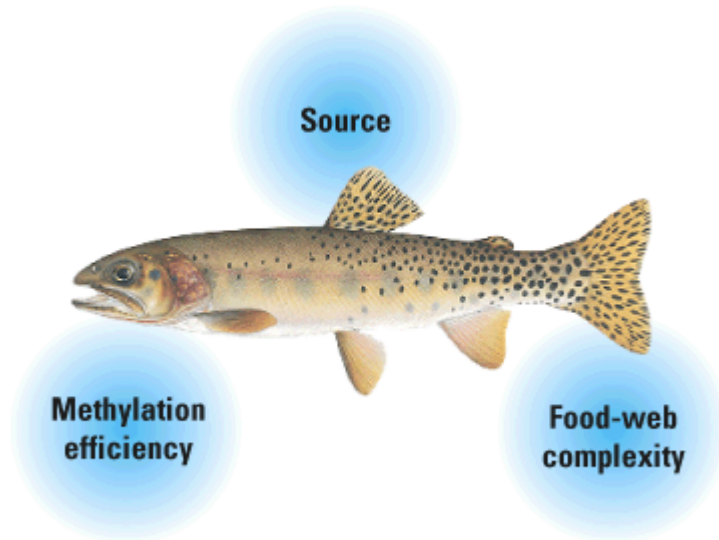
Piscivorous birds are at risk from mercury exposure via the food chain. Loons have been a focus of attention in New England for ecological and aesthetic reasons. A first step towards addressing threats of mercury to wildlife in Massachusetts is to have an understanding of the state of knowledge of mercury in indigenous non-fish vertebrates in the Commonwealth. A

compilation of this information for Massachusetts and recommendations for future monitoring has been assembled.

To provide a historical perspective to our work, a limited study of mercury deposition to the aquatic environment of northeastern MA was conducted.

Following a workshop held to consult with the research community on issues of sediment core analysis such as interpretation and costs, an analysis of the historical rate of mercury deposition to the bottom sediments of a freshwater lake in an area of the state having regionally high fish mercury concentrations was conducted, using isotopic dating of a sediment core.

Knowledge of the major sources of variance in fish mercury data can be used to improve the precision of fish tissue mercury studies. We conducted several studies of sources of variance in fish tissue mercury. Tissue moisture experiments were conducted to determine the variability due to sample preparation. A study of fish holding time was done to determine the stability of mercury in frozen fish samples over time. An investigation of statistical methods to compensate mathematically for the correlation of fish size with mercury, and the statistics to determine scientifically acceptable sample sizes were critical parts of the mercury studies.



Overview of Studies

The Division of Water Pollution Control at MA DEP began testing fish for contaminants in 1984. By then conventional water pollutants from point sources had been brought under control, and attention was turned to the evaluation of impacts from toxic substances. The initial fish testing programs developed into an Interagency working group that involved DEP's Office of Research and Standards, Wall Experiment Station, the Division of Fish and Wildlife, and the Department of Public Health. Members of the public were able to request fish testing at locations of concern through this program. If fish were found to contain contaminants, a fish advisory would be issued for the location by MA DPH. Test results showed a need for more comprehensive sampling to establish patterns and trends in fish contamination across the state.

Baseline Study of Fish Mercury

Fish reflect elevated mercury inputs to the environment. Approximately 40% of the rivers and lakes in Massachusetts sampled since 1983 are subject to fish consumption advisories as a result of mercury contamination of edible fish muscle.

When the extent of mercury contamination of Massachusetts' freshwater fishery resources became apparent in the early 1990s, MA DEP conducted a study to determine the distribution of mercury in freshwater fish tissue in non-source-impacted, largely rural Massachusetts lakes. That study sought to define a baseline for future studies and to identify possible environmental factors associated

with mercury in largemouth bass (*Micropterus salmoides*), yellow perch (*Perca flavescens*), and brown bullhead (*Ameiurus nebulosus*).

Fish Mercury Research in Northeastern Massachusetts.

In 1994, fish from a few of the lakes in the northeast part of the State were sampled as part of the State's interagency fish toxics surveillance program. The area had the State's highest concentration of point sources of atmospheric mercury emissions in the last two decades of the twentieth century: three municipal solid waste combustors and a medical waste incinerator. Some lakes were identified as having fish with tissue mercury concentrations greater than 0.50 mg/kg, the concentration above which the Massachusetts Department of Public Health issues fish consumption advisories. More restrictive advisories are issued for concentrations greater than 1 mg/kg.

In 1998 MA DEP initiated new municipal solid waste combustor rules that included stringent mercury emissions control regulations to lower mercury emissions up to 95%. A study of mercury in fish in lakes in and around the Merrimack Valley was initiated before the adoption of the new controls, so that the results would serve as an environmental baseline for comparison with fish tissue mercury monitoring results in the future, after the



emissions reductions. The study would also determine the need for additional consumption advisories, clarify possible spatial patterns in the occurrence of higher fish mercury concentrations, and allow comparison of fish contamination in a high mercury deposition area to more rural areas in Massachusetts and to regional New England data.

Mercury Initiative.

The New England Governors and Eastern Canadian Premiers adopted the Regional Mercury Action Plan in June 1998. The Plan takes an integrated, comprehensive approach that incorporates mercury pollution control and pollution prevention of air, water and land. The goals of the multi-agency Action Plan are to reduce New England mercury emissions by 50% as of 2003, by 75% as of 2010, and the virtual elimination of mercury emissions over the long term. A regional task force undertakes actions in emission reduction, source reduction, waste management, outreach and education, and monitoring and research.



The overall objective of the monitoring and research action category is to improve understanding of mercury sources and the

impact of mercury contamination on public health and ecosystem health. In addition, as the emissions control and other action categories commence, monitoring and research will track the response of the environment to the action initiatives. Key indicators for tracking the progress and success of the Regional Mercury Action Plan are studies of mercury in fish, wildlife, and lake sediments.

Long Term Monitoring Program.

Permanent locations for fish tissue mercury monitoring are required for documenting the manifestation of trends resulting from mercury reduction efforts. Established monitoring sites would provide several pieces of valuable information to help understand fish mercury trends. Resampling over time will provide a consistent, long-term record of mercury concentrations in fish across the state. The data will represent an indicator of the responses of the environment to changes in mercury inputs resulting from regional and national mercury emissions control efforts. The information will also characterize year-to-year and seasonal variation in fish mercury concentrations.

An accurate assessment of the causes of variation has been a major focus of the fish research program. Statistically robust sample sizes were developed to allow detection of geographic variation in a relatively small state. Sources contributing to the total variance were thoroughly investigated in the course of the research.

To document trends in time and space, analysis of sediment cores was initiated,

after consultation with the research community. Isotopic dating of a sediment core from a Merrimack Valley lake established a record of the history of mercury deposition, a history of increased deposition that was triggered by the

damming of the Merrimack River, and closely followed industrialization in Massachusetts.



Lowell, MA in the late 1800's.

Descriptions of Research Studies.

Title: Fish Monitoring Through Public Requests

Objectives: Massachusetts environmental agencies instituted a program by which the general public may request that fish from a lake or stream be sampled and analyzed for the presence of potentially toxic chemicals. The program screens fish from heavily fished locations for contaminants. If contaminants in the fish exceed health standards, a fish consumption advisory is posted at the waterbody. The program has been expanded to include waterbodies in watersheds as part of systematic watershed assessment.

Background: A growing awareness of the effects of environmental contamination of streams and lakes created an increased public demand for fish toxics monitoring data. Beginning in 1993, a formal protocol and request procedure was made available to the public by the following environmental agencies:

- Massachusetts Department of Environmental Protection

- Division of Watershed Management
- Division of Environmental Analysis / Wall Experiment Station
- Office of Research and Standards
- Massachusetts Department of Public Health, Bureau of Environmental Health Assessment
- Massachusetts Department of Fisheries, Wildlife and Environmental Law Enforcement

Program Description: An Interagency Committee was formed to conduct the program, and a Memorandum of Understanding was produced by the agencies involved to establish the structure of the Public Request Program. Applications for requesting fish testing at a waterbody were made available to the public, who then submit the request to DEP's Division of Watershed Management.



The Interagency Committee meets annually to prioritize the requests according to agreed upon criteria (fishing pressure and the potential for contamination). Three to five species of fish are collected, representative of the ecological niches in the waterbody, e.g., bottom feeders, water column feeders, and predatory fish. Three to five individuals of each species are composited for analysis. Fish samples are analyzed for arsenic, cadmium, lead, mercury, selenium, PCB arochlors and congeners, and organochlorine pesticides. Additional metals and organics may be analyzed for as necessary.

Summary of Results: Massachusetts' waterbodies are frequently found to be contaminated with mercury. Two hundred thirty six waterbodies have been assessed as part of this program. Ninety-eight of the waterbodies required a fish advisory for mercury contamination. Twenty five waterbodies required fish advisories for PCBs (13 advisories), DDT (6 advisories), chlordane (4 advisories), PAHs or dioxin (1 advisory each). One hundred thirteen locations did not require fish advisories. The Department of Public Health issued a statewide advisory regarding the dangers of consumption of freshwater fish, especially for pregnant women and young children, due to mercury contamination, as a result of the fish testing program.

By screening lakes for mercury and other contaminants in fish, it was found that fish contamination often occurred in lakes with no obvious source of pollution. The fish screening program demonstrated the need for a more directed study of lakes in different ecoregions across the state, using larger sample sizes of individual fish, rather than composites, to improve the

ability to detect geographic or environmental differences in the distribution of mercury in fish.

Program Status: The program continues to assess the water resources and suitability of fish for consumption in the Commonwealth.

Cost: Participants in the program are personnel employed at state agencies, and the program is conducted as part of their service. No additional costs or budget are associated with the program.

Work Products:

Reports: A yearly report describing the results of fish analysis is compiled by Robert Maietta, Division of Watershed Management, MA DEP.

Publications: Isaac, R.A., R.J. Maietta, A.S. Johnson, 1992. The role of fish tissue monitoring in evaluating and managing toxic substances: A summary of Massachusetts' Program. Ma Dept. of Environmental Protection, Bureau of Resource Protection, Division of Water Pollution Control.

Massachusetts Department of Public Health, Bureau of Environmental Health Assessment, 1994. Fact sheet on mercury in freshwater fish.

Massachusetts Department of Public Health, Bureau of Environmental Health Assessment, 2001. MDPH Issues New Consumer Advisories On Fish Consumption and Mercury Contamination. Press release.

Massachusetts Department of Public Health, Bureau of Environmental Health Assessment, 2004. Freshwater Fish Consumption Advisory List - April 2004. <http://mass.gov/dph/beha/fishlist.htm>

Public Presentations: Progress in monitoring Massachusetts Lakes is regularly presented at the New England Association of Environmental Biologists (NEAEB) annual meeting.

Massachusetts Department of Public Health, Bureau of Environmental Health Assessment also issues lake or river specific fish advisories when fish from a waterbody is determined to contain contaminants. The advisories are issued in five languages and are sent to the Boards of Health in the towns where the fish have been collected. Boards of Health are asked to post the fish advisories at the specific waterbody.



Title: Statewide Study of Lakes in Diverse Sub-ecosystems

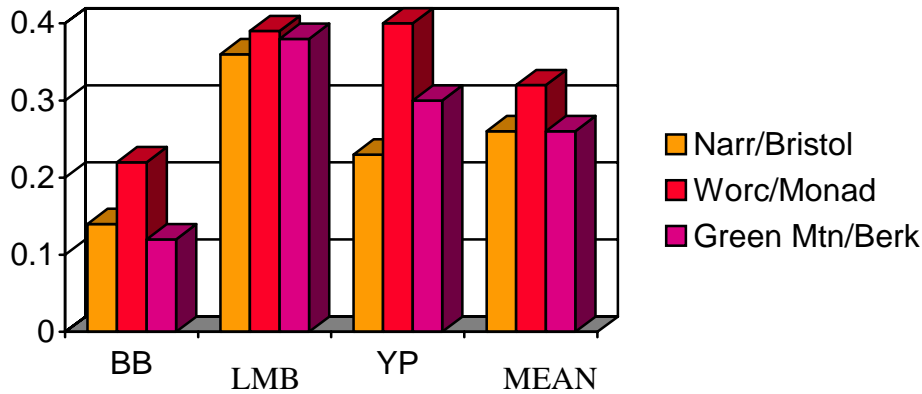
Objectives: The widespread mercury contamination in freshwater fish revealed in the screening program suggested a more comprehensive study was needed to understand the geographic distribution of mercury in fish and to determine what physical or hydrologic features may influence variation in mercury levels.

Background: The fish screening program conducted by three state environmental agencies showed that high levels of fish mercury existed in fish from lakes with no known sources of contamination. Ecologically-based geographic subdivisions had been investigated in some studies as being associated with differences in the bioaccumulation of metals by fish. Trophic status of lakes was also thought to influence mercury bioaccumulation in fish.

Study Description: The sediment, water and 3 species of fish from 24 of Massachusetts' relatively least-impacted waterbodies were sampled and analyzed to determine the patterns of variation in edible tissue mercury concentrations and the relationships of these patterns to characteristics of the sediment, water and water bodies (lake, wetland and watershed areas). Sampling was apportioned among three different ecological subregions and among lakes of differing trophic states, ranging from oligotrophic to eutrophic. We sought to partition the variance to discover if these broadly defined concepts are suitable predictors of mercury levels in fish.

Summary of Results: Average muscle mercury concentrations were 0.14 mg/kg wet weight in the bottom feeding brown bullheads (*Ameiurus nebulosus*) (range=0.01-0.79 mg/kg); 0.31 mg/kg in the omnivorous yellow perch (*Perca flavescens*) (range=0.01-0.75 mg/kg); and 0.40 mg/kg in the predaceous largemouth bass (*Micropterus salmoides*) (range=0.05-1.1 mg/kg). Statistically significant differences in fish mercury concentrations between ecological subregions in Massachusetts existed only in yellow perch, although there was a suggestion of such a relationship in brown bullhead. The productivity level of the lakes (as deduced from Carlson's Trophic Status Index) was not a strong predictor of tissue mercury concentrations in any species. A highly (inversely) correlated environmental variable was pH with yellow perch and brown bullhead tissue mercury. Largemouth bass tissue mercury concentrations were most highly correlated with the weight of the fish (+), the weight (+) and mercury concentrations (-) of yellow perch in the same lake and the magnitude of surface areas, watershed and wetland areas associated with lake (+). These results are generally consistent with existing knowledge of freshwater fish tissue mercury dynamics and are notable for demonstrating spatially correlated differences in tissue mercury concentrations across ecological subregions on a scale less than about 150 miles.

Mercury (ppm) in Fish from Three Ecological Subregions



Project Status: This research project is complete. The results have been used in subsequent studies as a representative baseline of fish mercury in the state.

Cost: \$65,000.

Work Products:

Reports: Fish Mercury Distribution in Massachusetts Lakes. Final Report. MA DEP, Office of Research and Standards, May 1997.

Publications: Rose, J., M.S. Hutcheson, C.R. West, O. Pancorbo, K. Hulme, A. Cooperman, G. DeCesare, R. Isaacs, and A. Screpetis, Fish Mercury Distribution in

Massachusetts, USA Lakes. *Environmental Toxicology and Chemistry*, Vol. 18, No. 7, pp. 1370-1379, 1999, SETAC.

Public Presentations: Fish Mercury Distribution in Massachusetts Lakes. Presented at the Society of Environmental Toxicology and Chemistry World Congress, Vancouver, British Columbia, Canada, 1995.

Fish Mercury Distribution in Massachusetts Lakes. Presented at the Boston Risk Assessment Group, Cambridge, MA, March 1997.

Title: Merrimack River Valley Fish Mercury Study

Objectives: Fish from 26 lakes in northeast Massachusetts were sampled in order to:

- Determine if human health fish consumption advisories for mercury were necessary;
- Examine the relationships between levels of fish tissue total mercury concentrations in the study area and other regions of the state and country; and
- Examine the possible contribution of local sources of atmospheric mercury to the local fish mercury concentrations.

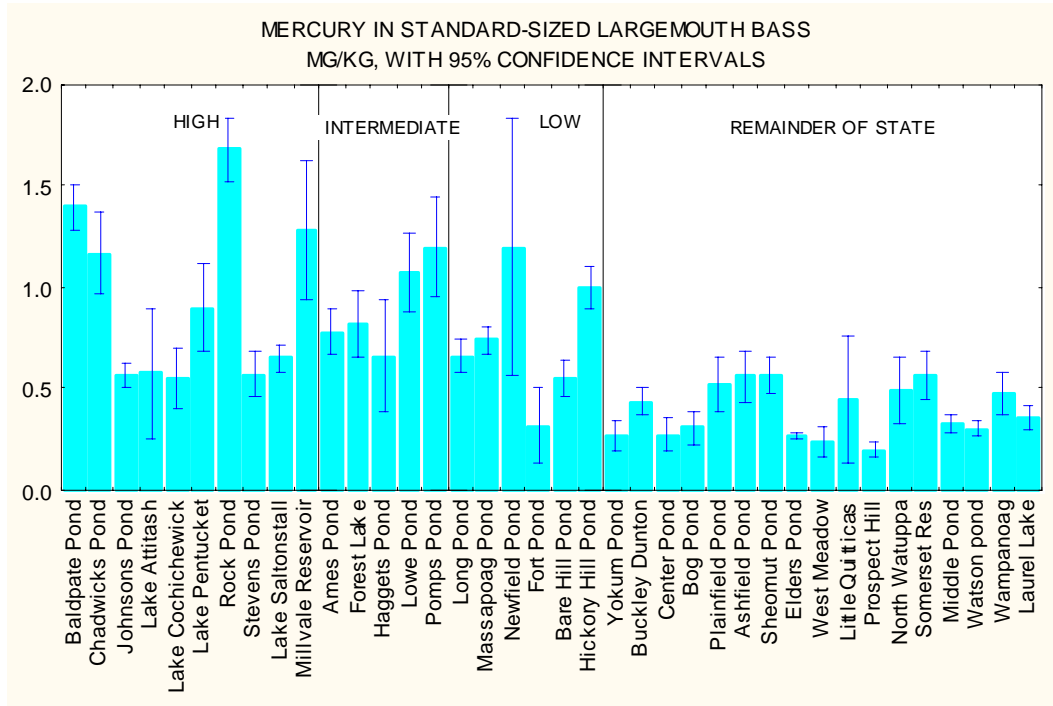
Background: In a regional report on mercury in the northeast states, this area of Massachusetts was predicted to have the highest level of atmospheric mercury deposition in the northeast U.S., on the basis of atmospheric dispersion modeling of sources of mercury emissions in the U.S. Until recently three municipal waste combustors and one medical waste incinerator were located in this area. The area has a long history of industrialization, with mercury releases occurring as early as the nineteenth century.

Study Description: The study area was delineated into downwind, near-field upwind and far upwind areas based upon prevailing wind patterns vis-à-vis the four incinerators. Largemouth bass (LMB) (*Micropterus salmoides*) and

yellow perch (YP) (*Perca flavescens*) were the primary species sampled. Concentrations of mercury in fish tissue were compared with data from elsewhere in the State and between these sub-areas to determine whether any differences could be potentially attributed to the incinerators.

Summary of Results: Mercury concentrations in LMB (mean 0.89 ± 0.43 mg/kg [n=192]) in the study area were in the top fourth of LMB mercury values derived for more rural, non-local-source-impacted Massachusetts lakes in the west, central and southeastern parts of the state. Because of the elevated mercury concentrations, all but one of the lakes in the study design in which LMB were caught warranted fish consumption advisories for LMB (concentrations >0.5 mg/kg). This particular lake was located farthest upwind of the incinerators. In other parts of the state, fewer than 50% of the waterbodies tested in a previous study required fish consumption advisories due to mercury. YP mercury concentrations (mean 0.44 ± 0.21 mg/kg, n=152) were similar to, or slightly greater than those from more rural regions of the state. YP mercury concentrations from 65% of the lakes were below the threshold for issuing a fish consumption advisory.

Fish Mercury Levels in High, Medium and Low Deposition Areas Compared to the Remainder of the State



There was no obvious relationship between LMB or YP fish tissue mercury concentrations and their locations relative to prevailing wind patterns and the incinerators (see above graph of mercury in fish populations in lakes located in projected high, medium and low deposition areas in relationship to incinerators). LMB tissue concentrations correlated with the mercury content of their prey, YP, and water temperature. Tissue concentrations did not correlate with lake water pH, conductivity or dissolved oxygen concentration. The study results therefore suggest that the tissue concentrations of mercury in LMB in the study area reflect the predicted higher atmospheric mercury deposition rate for this region, which has urban and rural areas, and that these concentrations are

greater than those for more rural areas of the state having lower predicted atmospheric deposition rates of mercury. Although no relationship could be discerned between the major point sources in the area and fish mercury concentrations, the resolution of the approach used (prevailing wind analysis) may have limited power to detect such effects.

Project Status: The directed study of the northeast has been completed. Several of the lakes have since been included in the long-term monitoring initiative and the seasonal variability study.

Cost: \$36,000

Work Products:

Reports: Fish Mercury Levels In Northeastern Massachusetts Lakes (<http://www.mass.gov/dep/bwp/hgrees.htm>).

Publications: Field Concentrations of Fish Muscle Mercury in a Regionally High Mercury Deposition Area. Draft manuscript to be submitted to *Science of the Total Environment*.

Public Presentations:

- U.S. Environmental Protection Agency, Atlantic Ecology Division, Narragansett, RI. 4/01. Invited Seminar. Mercury in Freshwater Fish in Massachusetts: Past and Future Studies.
- University of Connecticut, School of Pharmacy, Toxicology Program. Storrs, CT., 4/02. Invited Seminar. Sources of Mercury in Freshwater Fish.
- University of Massachusetts Boston, Environmental, Coastal and Ocean Studies Program. Invited Seminar. 2/03. Ecological and Land Use Perspectives in the

Distribution of Mercury in Freshwater Fish in Massachusetts.

- North Atlantic Chapter of Society of Environmental Toxicology and Chemistry, Annual Meeting. 4/03. Mystic, CT. Historic Mercury Inputs and Modern Spatial Patterns in Fish Tissue Mercury Concentrations in Massachusetts.
- Northeast States for Coordinated Air Use Management, Northeast Regional Science Policy Workshop. 5/04. Kennebunkport, ME. Massachusetts Mercury Monitoring Study: Results from a Regionally High Deposition Area.
- Boston Risk Assessment Group, Northeast Chapter of Society of Risk Analysis. Boston. 7/04. Fish Tissue Mercury Concentration Trends in Northeastern Massachusetts.
- U.S. Environmental Protection Agency, Annual Regional Risk Assessors Meeting. 6/04, Boston, MA. Tissue Mercury Concentration Trends in Northeastern Massachusetts.



Title: Mercury Bioaccumulation in the Food Webs of Two Northeastern Massachusetts Freshwater Ponds.

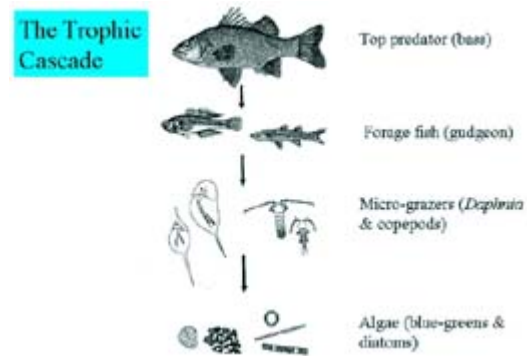
Objectives: A food chain study was conducted of the ecosystems in two small, similar lakes located within a few miles of each other, to gain a greater understanding of the process of mercury bioaccumulation in fish, and to help determine the pathways it takes in our freshwater lakes.

Background: The recent documentation of high levels of mercury in fish from northeastern Massachusetts freshwater lakes and ponds called attention to the need for additional study of mercury bioaccumulation in these environments. Mercury is known to bioaccumulate in lake ecosystems.

Fish studies in Massachusetts have focused on large numbers of lakes across wide geographic areas. The present study was designed to examine pond characteristics at a smaller scale, in hopes of observing relationships not perceptible in studies that encompass the variation associated with lakes in different geographic and climatic regions.

The ponds, Pumps Pond in Andover and Stevens Pond in North Andover, lie in the airshed of an urban area that conducts centralized incineration of municipal wastes. Thus, atmospheric deposition of mercury should be roughly equal into the ponds.

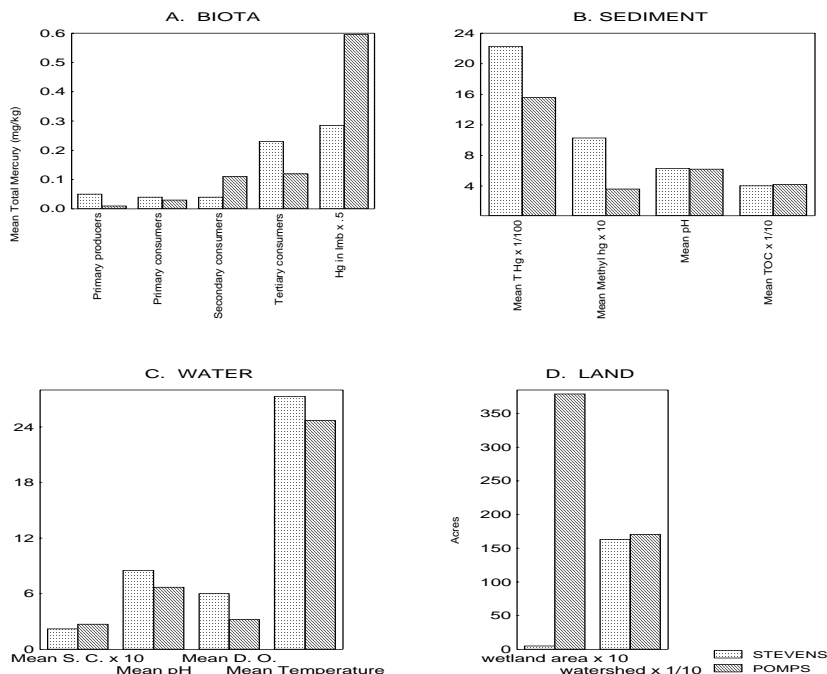
In a sampling event in 1999, largemouth bass from Pumps Pond contained higher amounts of tissue mercury than those from Stevens Pond. We assume that factors that influence mercury uptake in



the ponds would be measurably different, since mercury concentrations in largemouth bass were different. Likewise, factors that are the same in both ponds are assumed not to have a significant influence on mercury uptake. By examining mercury content at each trophic level, we anticipated finding differences or similarities between ponds that correlated with tissue mercury concentrations in largemouth bass or other trophic groups. Ponds that bore many physical, biological and geographic similarities should increase the probability that variables favoring mercury bioaccumulation would be observable.

Study Description: Organisms representative of four trophic levels in the ponds were collected and analyzed for mercury to determine the comparability of the ponds and the mercury levels present in the ponds' broadly defined trophic niches, and sediments. Physical characteristics of the ponds including the area of wetlands in the ponds' watersheds were determined.

Partitioning of Mercury in Two Ponds



Summary of Results: The predominant measured physical difference between Stevens Pond and Poms Pond is the area of wetlands adjacent to the ponds. The large extent of the adjacent wetlands at Poms Pond and the near absence of wetlands at Stevens Ponds suggest that processes taking place in the wetlands may modify the partitioning of mercury in the ponds in such a way as to make it more bioavailable. Higher bioaccumulation rates in Poms Pond may be reflective of increased mercury bioavailability due to mercury methylation within wetlands surrounding the ponds.

Project Status: Project complete. Resampling of higher trophic levels is anticipated as part of Long Term Monitoring Study.

Cost: \$10,000

Work Products:

Reports: Mercury Bioaccumulation in the Food Webs of Two Northeastern Massachusetts Freshwater Ponds. MA DEP, Office of Research and Standards, 2002.

Title: Assessment of Mercury in Massachusetts Wildlife.

Objectives: A major focus of this study was to develop a comprehensive plan for further studies on mercury in Massachusetts lakes. To facilitate that goal, this study set out to collect information on mercury levels in non-fish vertebrate species of Massachusetts, and to recommend further studies that could be considered to document mercury accumulation. The study undertook a detailed literature search and compilation of information concerning mercury in non-fish vertebrates; and conducted a field survey of lakes that have habitat, food supply and other niche requirements for common loons.

Background: Studies conducted thus far show that mercury bioaccumulates in fish. Knowledge of the impact to fish-eating wildlife in Massachusetts was inadequate.

Study Description: The study compiles mercury bioaccumulation data for bald eagles, common loons, river otters, mink, voles, mice, snapping turtles and other reptiles, with bibliographies for each group. Loon surveys were conducted on several lakes, and lakes with breeding loon pairs were mapped. Additional lakes were designated likely to support loons or potentially able to support loons. Next steps for further research and biomonitoring are included.

Summary of Results: The lake survey identified many lakes capable of supporting loon populations. Detailed recommendations as to how

loon populations could be established on suitable lakes by the Division of Fish and Wildlife are given in depth. Future research should:

- Continue to sample bald eagle chicks annually at about 10 weeks of age;
- Continue to sample adult and chick common loons;
- Begin studies of snapping turtles.

Project Status: Project complete. The suggested next steps are under consideration for continuation. Coordination with wildlife agencies is in progress.

Cost: \$14,180

Work Products:

Reports: Mercury in Non-fish Vertebrates in Massachusetts: Compilation of Existing Resources and Recommendations for the Future. Tufts School of Veterinary Medicine. Available from MA DEP ORS.



Title: Lake Sediment Mercury Deposition Study

Objectives: To determine to what extent differences in mercury fluxes to an ecosystem (in this case surface waterbodies in Massachusetts) can help explain differences in mercury concentrations in the fish residing in these systems. The specific goal of this study is to determine the feasibility of using isotope geochronological techniques to establish the recent (100 year) history of anthropogenic mercury additions to fresh water lakes in the Commonwealth.

Background: Emission and deposition rates of mercury can be quite variable, with depositional rates of mercury in a local region dependent on both distant and local emission sources, and regional and local atmospheric transport. Most of the supply of mercury to freshwater systems is thought to be derived from atmospheric input, because the retention of mercury in most watersheds surrounding such systems is extremely efficient (>90% retention).

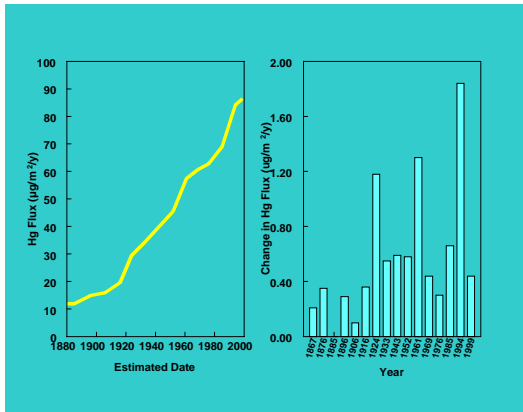
By comparing mass flux rates of mercury to different lakes and ponds it may be possible to better understand both local and regional heterogeneity of primarily atmospheric mercury fluxes to such systems and determine the linkage, if any, to local and regional heterogeneity in fish body burdens of mercury. The information provided by this approach can be used to both validate atmospheric models predicting mercury transport and deposition as well as contribute to the primary goal of understanding the environmental variables influencing mercury concentrations in tissues of fish

and other organisms using these ecosystems.

Study Description: Isotope (^{210}Pb and ^{137}Cs) geochronology was used to date a sediment core taken from Lake Cochichewick in North Andover, Massachusetts, known to have relatively high mercury concentration in fish resident in the lake. Historical changes in mercury contamination of the lake were determined using the mass accumulation rates determined by isotope geochronology and measurement of mercury concentrations downcore.

Summary of Results: The data clearly show a low and slowly increasing concentration of mercury before the 1900s and then a rapid increase in concentration beginning in the late 1800s and early 1900s. Concentrations are highest at the top of the core and are over an order of magnitude higher than those observed in the deeper part of the core that are more characteristic of relatively pristine areas (~20 - 30 ng/g dry weight). However, the uppermost section of the core analyzed in this work represents a time period of about 4 years or the period from 1997 to the date of collection in May 2001. Lack of temporal resolution at the surface of the core may mask any decrease in concentration occurring over the last few years.

The highest single increase was observed for the core section representing the period between 1990 and 1996, shortly after the construction of the incinerators in the 1980s near Lake Cochichewick. Whether



Mercury fluxes into sediments of Lake Cochichewick over the last 120 years. Panel on right indicates estimated change in sediment flux for each dated core section. Note the absence of any decrease in mercury concentration over the last decade.

this jump in Hg flux was wholly in response to the emissions from these incinerators cannot be conclusively determined by the limited data from a single sediment core, but does argue for closer scrutiny of the importance of these and possibly other local and regional sources.

Project Status: Project complete. Additional lake sediment cores are planned at strategically located lakes, and fish sampling and analysis has been conducted in lakes where sediment cores have been obtained.

Cost: \$18,000

Work Products:

Reports: Determination of Recent Inputs of Mercury to Lakes/Ponds in the Merrimack Valley Using Sediment Cores – A Feasibility Study, by G. Wallace, S. Oktay, F. Pala, M. Ferraro, M. Gnatek, & D. Luce. Dept. of Environmental, Coastal and Ocean Sciences, U Mass Boston, Boston, MA.

Publications: Manuscript in preparation.

Public Presentations:

- University of Massachusetts Boston, Environmental, Coastal and Ocean Studies Program. Invited Seminar. 2/03. Ecological and Land Use Perspectives in the Distribution of Mercury in Freshwater Fish in Massachusetts.
- North Atlantic Chapter of Society of Environmental Toxicology and Chemistry, Annual Meeting. 4/03. Mystic, CT. Historic Mercury Inputs and Modern Spatial Patterns in Fish Tissue Mercury Concentrations in Massachusetts.
- Northeast States for Coordinated Air Use Management, Northeast Regional Science Policy Workshop. 5/04. Kennebunkport, ME. Massachusetts Mercury Monitoring Study: Results from a Regionally High Deposition Area.
- Boston Risk Assessment Group/Northeast Chapter of Society of Risk Analysis. Boston. 7/04. Fish Tissue Mercury Concentration Trends in Northeastern Massachusetts.

Title: Sediment Coring Workshop

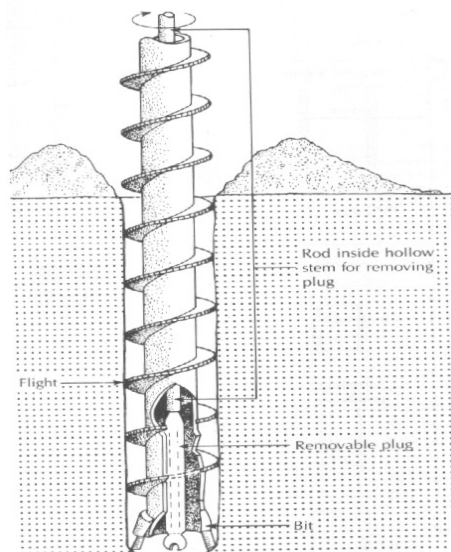
Objectives: A one-day workshop was convened, attended by regional scholars in sediment geochemistry and the ORS staff, to:

- Solicit input from the research community on the geochemical factors that may influence the flux of mercury into ponds within the Commonwealth of Massachusetts;
- Investigate a range of dating techniques used in coring;
- Become familiar with problems encountered in sediment core analysis and interpretation;
- Receive expert advice on the value and effectiveness of a sediment coring program for evaluating mercury deposition in Massachusetts lakes;
- Compare costs of sediment coring programs.

Background: The MADEP completed two major investigations of mercury in freshwater fish in ponds in the Commonwealth. Based on the results of the investigations on mercury in freshwaters performed to date, the MADEP proposed that a sediment coring program be developed and implemented in order to:

- 1) Further develop a historical perspective on mercury inputs in freshwater environments,
- 2) Differentiate between local and regional sources of mercury,

- 3) Evaluate the variation in mercury concentrations between different waterbodies and aquatic organisms, and
- 4) Establish a long term monitoring program.



Study Description: The workshop consisted of summary presentations of the mercury investigations that have been sponsored by MA DEP, presentations of the invitees, and findings of their research. Directed discussions for the remainder of the day included experiences with sediment coring, the laboratory analysis of sediment samples and data analysis and interpretation. The purpose of the workshop discussion session was to discuss any practical concerns for the

planning and implementation of future sediment coring programs. Presenters from the research community were:

Dr. Gordon Wallace – University of Massachusetts at Boston;

Dr. Tim Parshall – Harvard Forest, Harvard University;

Dr. Gabe Benoit – Yale School of Forestry and Environmental Studies;

Dr. John Colman – United States Geological Survey.

Summary of Results: Following the review of the recommendations made during the workshop a Work Plan for the Sediment Coring Program was to be prepared. A solicitation would then be issued for sampling and analytical services to support the Sediment Coring Program. The timing of the issuance of any Request

for Proposals (RFPs) and the implementation of the proposed Sediment Coring Program would be dependent upon the availability of funding for the study by the Commonwealth with the potential for joint funding with other institutions and agencies.

Project Status: Workshop completed. Additional lakes are being investigated as potential candidate lakes for coring.

Cost: \$3,500

Work Products:

- Improved knowledge of the use of sediment coring for analysis of mercury deposition.
- Improved contacts with regional experts in the field.

Reports:

Sediment Coring Workshop, June 26, 2001, Tower Hill Botanical Garden, Boylston, Massachusetts, Letter Report to MA DEP, Office of Research and Standards. Normandeau Associates.

Title: Fish Mercury Seasonal Variability Study

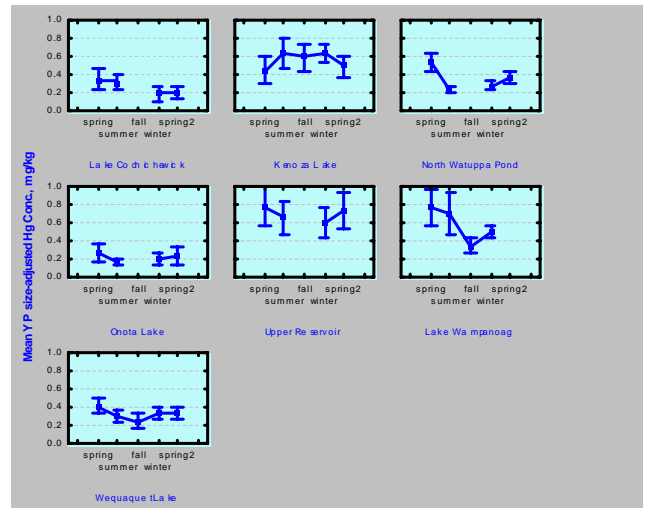
Objectives: The objectives of this study were to document the concentrations of fish tissue mercury in each of the major seasons and determine if and how they vary from season to season. The knowledge gained from this project is intended to tell us whether time of year is a critical variable to be factored into planning future fish sampling efforts.

Background: Massachusetts has been monitoring the concentrations of mercury in fish tissue throughout the state for approximately 20 years. The primary goal of much of this work has been to identify fish populations that might pose unacceptable health risks to those consuming the fish. Sampling sites have not often been revisited in subsequent years and until the late 1980's, methods and procedures were not fully standardized, and sampling intensity was not designed to permit rigorous comparisons of between-year trends. A long term monitoring plan was devised to address this problem. During the initial sampling years of the long term monitoring study described later, a study of seasonal variation in fish tissue mercury was conducted.

Study Description: LMB and YP were sampled and analyzed for mercury at 7 of the long-term study lakes. Sampling times were spring (pre- and post-spawn), summer, autumn, winter and spring again (pre-and post-spawn).

Summary of Results: Maximum seasonal differences between YP edible muscle tissue mercury concentration means for a lake ranged from 20 to 112% and LMB mean differences ranged from

26 to 107%. Values were generally highest in the spring. The substantial variation in the raw data means suggests that seasonal variation may be an important source of variation in fish tissue mercury data sets that should be considered in the design or interpretation of these types of data sets.



Seasonal Mean Size-Adjusted Tissue Mercury Concentrations (mg/kg) in Yellow Perch

Project Status: Field and laboratory work complete. Draft report complete.

Cost: \$73,000

Work Products:

Reports: Preliminary Study Summary - Long-term Monitoring Program of Fish Mercury in Massachusetts Lakes and Study of Seasonal Variation in Fish Mercury, Section A-“Fish Mercury

Studies: Long-term Monitoring Program and Directed Studies 2001-2003. MA DEP, Office of Research and Standards, 2004.

Suggestions For Study Design And Data Interpretation Improvement North Atlantic SETAC Meeting Poster, 2004.

Publications: Manuscript in preparation.

University of Massachusetts Boston, Environmental, Coastal and Ocean Studies Program. Invited Seminar. 2/03. Ecological and Land Use Perspectives in the Distribution of Mercury in Freshwater Fish in Massachusetts.

Public Presentations: Sources Of Variation In Fish Tissue Mercury Concentration Estimates And



With regard to mercury analysis variation associated with different holding times, insignificant variation in mercury levels occurred after two months. After one year, the mercury content of approximately 50% of the samples decreased significantly.



Project Status: The tissue moisture content study is complete. Wet weight measurements are now standard operating procedure. The holding time study will be repeated using larger sample sizes.

Cost: This work was conducted by personnel at Wall Experiment Station, and was supported with funds for the seasonal variability study.

Work Products: The tissue moisture study results have been incorporated into laboratory standard operating procedure. Modification of the laboratory standard operating procedure for mercury analyses to recognize holding times for up to 60 days for research projects is provisionally incorporated into laboratory standard operating

procedure for research projects. The standard US EPA 28 day holding time will continue to be observed for enforcement sensitive or public health advisory related work.

Public Presentations:

Sources Of Variation In Fish Tissue Mercury Concentration - Estimates And Suggestions For Study Design And Data Interpretation Improvement. North Atlantic SETAC Meeting Poster, 2004.

Boston Risk Assessment Group, Northeast Chapter of Society of Risk Analysis. Boston. 7/04. Fish Tissue Mercury Concentration Trends in Northeastern Massachusetts.

Title: Long Term Monitoring Program for Fish Tissue Mercury Trends

Objectives: A program to monitor fish mercury in selected lakes was initiated to determine whether the levels of mercury in fish are decreasing over time as a result of increased controls on mercury emissions sources; and, to sample waterbodies on a cyclic basis to identify other trends in fish mercury bioaccumulation.

Background: Early studies of fish tissue mercury in Massachusetts were conducted to provide broad or local spatial assessment of the need for fish consumption advisories. Locations were seldom revisited. As both the fish mercury work intensified and the state pressed for tighter controls on mercury emissions and sought to reduce mercury use in the Commonwealth, the need for a program to document long-term trends in fish tissue mercury concentrations became apparent.

Study Description: Beginning in 2001, the Office of Research and Standards selected 18 permanent locations for fish tissue mercury monitoring in Massachusetts to provide vital information to help understand fish mercury dynamics (Appendix 1). This information would provide a consistent, long-term record of mercury concentrations in fish across the state. The data would represent an indicator of the responses of the environment to changes in mercury inputs as a result of mercury emissions control efforts. The information on year-to-year variation in fish mercury concentrations could be determined using long term data. In cases where data collected in different years are compared to evaluate the influence of some other variable (e.g., urban versus rural comparisons),

knowledge of the magnitude of inter-annual variation would assist with the determination of the significance of differences attributed initially to other factors. Half of the lakes are sampled every year, so that data will be available for any particular lake every other year.

Summary of Results: Preliminary analysis of year-to-year temporal trends in fish mercury concentrations for lakes in northeastern MA has indicated that there have been statistically significant decreases in tissue mercury concentrations in both LMB and YP from 1999 through 2004.

Project Status: Data collection and analysis is ongoing. Two additional lakes in the northeast have been added to the original list of lakes to provide a more detailed picture of longer-term temporal trends in the Merrimack River Valley, where substantial reductions in atmospheric emissions of mercury have come to pass over the last few years. Other lakes were added from two areas of particular interest: Echo Lake in Hopkinton, where a dated record of mercury from sediment coring of this rural lake exists; and the Quabbin Reservoir, where atmospheric mercury deposition measurements have been made.

Cost: \$68,500 for 2004.

Work Products:

Reports:

- Fish Mercury Studies Status Report to March 31, 2004.
- Draft Final Report. Massachusetts Fish Tissue Mercury Studies: Long-Term Monitoring Results 1999 – 2004. MA DEP, Office of Research and Standards, (report in preparation).

Publications: Manuscript in preparation.

Public Presentations:

- Northeast States for Coordinated Air Use Management, Northeast

- Regional Science Policy Workshop. 5/04. Kennebunkport, ME. Massachusetts Mercury Monitoring Study: Results from a Regionally High Deposition Area.
- Boston Risk Assessment Group/Northeast Chapter of Society of Risk Analysis. Boston. 7/04. Fish Tissue Mercury Concentration Trends in Northeastern Massachusetts.
- Annual Meeting European SETAC. 5/05. Lille, France. Temporal Responses of Fish Tissue Mercury Concentration Responses to Local Atmospheric Mercury Emissions Reductions from Incinerators.

Lake Cochichewick, North Andover, with Municipal Waste Combustor in the Background



Title: Sources Of Variation In Fish Tissue Mercury Concentration - Suggestions For Study Design And Data Interpretation Improvement

Objectives: Assemble the information we have learned concerning sources of variation in fish studies in one place, to show how to improve the efficiencies of fish tissue mercury sampling programs. Knowledge of the major sources of variance in the data can be helpful and used to improve the precision of fish tissue mercury predictions in waterbodies by lowering the variance where possible.

Background: The objective of many studies of edible fish tissue mercury concentrations is to provide estimates of lake population mean mercury concentrations for comparison with human health or ecologically-based tissue concentration exposure limits or to compare between different sample groups. Insufficient attention may be given to the inherent variability in fish tissue mercury concentrations in the design of studies and during the interpretative phase of studies. The implications of conclusions reached or actions taken based upon false positives or negatives from such studies may have substantial public health and ecological consequences (e.g., unnecessary closure of a fishery or failure to protect public health with a warranted fish consumption advisory or to detect an ecological threat).

$$\sigma_x^2 = \frac{1}{N-1} \sum_i (x_i - \bar{x})^2$$

Study Description: This work component takes information gathered from the various Mercury Initiative sponsored projects and summarizes it in a practical format which can be used by others in the future to improve study designs and the quality of data interpretation. Sources of variation in fish tissue mercury concentrations which are considered include the correlation of mercury with fish size, the season in which the fish were collected, the degree of tissue hydration, the reproductive status of fish, and calculation of sample sizes.

σ	Power
50	.52
75	.26
100	.17
125	.12
150	.10

Summary of Results: The following recommendations have come out of our studies and experiences:

- perform analyses on individual fish using statistically based numbers of fish in order to preserve variance information and provide sufficient confidence and power in the study design;
- look for mercury concentration – size relationships and adjust for this covariate if necessary, using ANCOVA or size-standardization;
- try and obtain comparison samples (temporal or spatial) at the same time of year. Aggregated tissue mean mercury concentrations

calculated over seasons will have higher variance than same season means, and

- depending on the intended use of the data, recognize that spring samples of yellow perch and largemouth bass will give the highest estimate of population mercury concentrations.

Projected Status: Work in progress.

Cost: NA

Work Products:

Publications: Manuscript for peer review publication planned.

Public Presentations:

“Sources of Variation In Fish Tissue Mercury Concentration Estimates And Suggestions For Study Design And Data Interpretation Improvement.” – poster presentations at:

- North Atlantic Chapter of the Society of Environmental Toxicology and Chemistry, Annual Meeting. 4/03. Mystic, CT.
- Boston Risk Assessment Group/Northeast Chapter of Society of Risk Analysis. Boston. 7/04.

Appendix 1. Waterbodies for Long-term Monitoring

Waterbody	Years Sampled	Acres	Town	PALIS #
Onota Lake	1976 2001-2002 2004	617	Pittsfield	21078
Lake Wampanoag	1995 2001-2002 2004	218	Ashburnham Gardner	81151
Upper Reservoir	1995 2001-2002 2004	57	Westminster	35091
North Watuppa Pond	1995 2001-2002 2004	1700	Fall River	61004
Wequaquet	2001-2002 2004	654	Barnstable	96333
Lake Cochichewick	1999 2001-2002 2004	555	North Andover	84008
Kenoza Lake	1998 1999 2001-2002 2004	287	Haverhill	84028
Lake Lashaway	1994 2003	270	North & East Brookfield	36079
Wickaboag Pond	1995 2003	320	West Brookfield	36166
Lake Nippenicket	1978 2003	354	Bridgewater	62131
Massapoag Lake	1979 2003	353	Sharon	73030
Haggetts Pond	1999 2003	214	Andover	84022
Buckley-Dunton Reservoir	1995 2003	195	Becket	32013
Lake Saltonstall	1999 2003	45	Haverill	84059
Baldpate Pond	1999 2004	55	Boxford	91001
Chadwicks Pond	1999 2004	161	Haverhill/Box ford	84006
Echo Lake	2004	123	Milford/ Hopkinton	72035
Quabbin Reservoir	1989 2004	25,000	Petersham, New Salem, Ware, Belchertown, Pelham, Hardwick, Shutesbury	36129

