MASSACHUSETTS FISH TISSUE MERCURY STUDIES:

LONG-TERM MONITORING RESULTS, 1999 - 2004

by

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PREFACE

In 1994, the first comprehensive Massachusetts statewide examination of mercury in freshwater fish was conducted (MassDEP 1997). This study was followed in 1999 by an investigation of fish mercury concentrations in a region of the state predicted to have regionally high atmospheric deposition of mercury (MassDEP 2003b). That study was complemented by a study of historical mercury deposition into one lake in this region through the analysis of a sediment core using radioisotope dating techniques (Wallace et al. 2004). Additional work addressing mercury emissions and deposition is ongoing.

A number of additional studies have been conducted as part of the Department's continuing efforts to better elucidate the status of the Commonwealth's freshwater fish populations and environments with respect to mercury contamination.

A long-term monitoring network of lakes was established in 2001 to provide temporal tracking of changes in the mercury contamination status of fish in the Commonwealth, particularly as comprehensive mercury use and emissions reductions efforts have been implemented in Massachusetts and regionally. Results from these lakes will also provide a perspective on the scale of natural variability in tissue mercury concentrations for comparison with other sources of variation. The results from the first 5 years of this effort are contained in this report and in particular highlight the changes in fish tissue mercury concentrations which have taken place in the high mercury deposition area during a period when emissions from major point sources of mercury to the atmosphere have declined substantially in Massachusetts and across the region.

Other studies completed as part of our overall effort include one of seasonal variation in fish tissue mercury concentrations, which was conducted to provide perspective on the magnitude of this source of variance in fish tissue mercury concentration measurements (MassDEP 2005). This information is intended to help more efficiently design monitoring studies. Another study was performed to help elucidate the ecological basis for varying fish mercury patterns seen in different lakes. This comparative food web mercury study was conducted in two similar lakes in close proximity, which have different levels of mercury in top predator fish (MassDEP 2003a).

Wildlife are integral parts of pond ecosystems. Piscivorous birds in particular are at risk from mercury exposure via the food chain. Loons have been a focus of attention in New England for aesthetic and ecological reasons. A first step in the process for 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 information on the state of knowledge on mercury in wildlife in the Commonwealth was performed as part of our overall program (Pokras and Tseng 2001).

The data generated from these studies on mercury concentrations in edible tissues of popular freshwater fish also permit more widespread screening of the Commonwealth's lakes for potential human health risks posed by eating mercury-contaminated fish from

these lakes. These health hazards are addressed through the issuance of fish consumption advisories by the Massachusetts Department of Public Health.

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ABSTRACT

This report describes the establishment of a network of lakes in Massachusetts for the long-term monitoring of temporal changes in mercury concentrations in edible tissues of two species of freshwater fish. This network provides environmental indicator data to help evaluate the effectiveness of state and regional mercury reduction programs overall and in a modeled mercury deposition "hotspot".

Fourteen lakes were identified as the core lakes for this program. They are located across the Commonwealth in order to give a breadth of geographic coverage. The monitoring plan calls for seven of these lakes to be sampled each year on a rotating basis so that all are sampled every two years. Additional lakes in an area of specific interest, northeast (NE) MA[1], were added in some years to give a total of 17 lakes addressed in this report. Largemouth bass (LMB; *Micropterus salmoides*) and yellow perch (YP; *Perca flavescens*) are sampled because they are known to accumulate mercury and they are caught and consumed by recreational and subsistence anglers. On each spring sampling visit to a lake, approximately 30 YP and 12 LMB are caught and analyzed using a statistically-based sampling design. This report covers data returns from 17 lakes for the years 1999 through 2004.

Over this period consistent and substantial statistically significant decreases in YP and LMB fish tissue mercury concentrations occurred in most lakes sampled. Of seventeen lakes with at least two years of YP data, mean mercury concentrations in YP decreased significantly in 13 of the waterbodies between the earliest and latest dates sampled. Nine of the lakes were located in northeastern Massachusetts (NE MA). In 8 of the 9 waterbodies in this area significant decreases in YP mercury were observed, ranging from -26.0 to -61.9%. The mean change for all 9 lakes was -32.4%. Five of the remaining eight lakes around the rest of the state also had statistically significant, but not as large, decreases in YP mercury, ranging from 20.1 to 28.0%, with an overall mean change for all 8 lakes of -15.4 %.

LMB mercury concentrations followed a similar pattern with 11 of 17 lakes throughout the state decreasing in tissue mercury concentrations. Eleven of the lakes sampled were in NE MA and mercury levels in LMB from 7 of those decreased significantly, ranging from -16.0 to -55.2%. Mercury levels in 3 of the 4 other lakes also decreased, but the changes were not statistically significant. The mean change in LMB mercury among all 11 of these lakes was -24.8 %. Four of the remaining six lakes located around the rest of the state also had statistically significant but smaller decreases in LMB tissue mercury concentrations. The range of these changes was 15.9 - 36.4%, with an overall mean for all six lakes of -19.0%.

[1] *This area was a former mercury deposition "hotspot" in the 1990s that has experienced a very substantial decline in mercury emissions from local point sources since 1999. Recent preliminary deposition modeling and monitoring indicate that these emission reductions have resulted in similarly large decrements in mercury deposition in the area as well.*

Although reduced, it is important to note that the overall mean mercury concentrations in YP and LMB in many of the sampled lakes, in particular those in the northeast part of Massachusetts, still exceed the level deemed as safe for consumption by pregnant women, nursing mothers and children. On an individual lake basis, even after the noted reductions, many continued to have fish containing unsafe levels of mercury.

The temporal pattern of fish tissue mercury concentration decreases was consistent. No significant decreases were seen over a period of one year. Decreases were observed in some waterbodies over a period of 3 years and were consistently observed at 4 years. The first year of monitoring occurred prior to substantial reductions in mercury emissions from Massachusetts, regional and local mercury point sources that occurred through the Massachusetts Zero Mercury Strategy (MAZMS) and the New England Governors and Eastern Canadian Premiers (NEG-ECP) Mercury Action Plan. In particular, mercury emissions in the NE MA deposition hotspot area are estimated to have decreased by about 87% between the late 1990's and 2004 due to new pollution controls on municipal solid waste combustors (MSWC) and the closure of medical waste incinerators (MWIs) and a MSWC in the area.

In conclusion, the study results are notable for the significant decreases in edible fish tissue mercury concentrations, in particular from waterbodies located in a mercury deposition hotspot area, that occurred within 36-48 months of the adoption and implementation of comprehensive state and regional plans that effectively reduced emissions of mercury. These reductions were achieved primarily through the imposition of stringent mercury emissions controls on MSWCs and MWIs, as well as reductions from other regional sources. These results suggest that mercury levels in fish from temperate water bodies can be significantly reduced over a relatively short timeframe if emission sources are effectively controlled. However, although reduced, overall average mercury concentrations in fish from many of the waterbodies sampled still exceed the recommended safe consumption level. As discussed in the Massachusetts TMDL Alternative Proposal[2] submitted to the USEPA in 2004, significant reductions from out-of-state mercury sources will likely be needed to achieve water quality and public health objectives in Massachusetts.

MassDEP will continue to monitor mercury concentrations from these waterbodies to assess the environmental results of mercury reduction efforts targeting coal-fired utilities as well as mercury pollution from consumer and industrial products and from dental offices, that are currently underway in Massachusetts and the northeastern U.S. and Canada. MassDEP is also working to address the contribution of upwind, out-of-state sources to mercury deposition in Massachusetts through additional monitoring and modeling.

[2] *A TMDL Alternative Regulatory Pathway Proposal for the Management of Selected Mercury-Impaired Waters: A Supplementary Document to the Massachusetts Year 2004 Integrated List of Waters*
http://www.mass.gov/dep/water/resources/mercalt7.doc

1.0 INTRODUCTION

The Commonwealth of Massachusetts has monitored fish contaminants, including mercury, since 1984. The primary goal of much of the early work was to identify fish populations that might pose unacceptable health risks to those consuming the fish. Sampling sites were not often revisited in subsequent years, methods and procedures had not been fully standardized until more recent years, and the level of sampling intensity was not sufficient to optimally support statistically-based comparisons between samples.

Starting in the autumn of 1994, a more rigorous and comprehensive approach to the study of fish tissue mercury concentration processes in Massachusetts was implemented in response to increased concern about mercury inputs to the environment and possible adverse human health effects as a result of consuming mercury contaminated fish. A statewide study was first performed to determine the distribution of mercury in the edible tissues of several freshwater fish species and the relationships of those concentrations to environmental characteristics (Rose et al. 1999). Based upon limited data collected for fish consumption advisory purposes showing signs of high mercury levels in fish and recognition of the presence of several likely sources of high atmospheric emissions of mercury in the northeast region of the state, a targeted, intensive study of the degree of mercury contamination of two species of freshwater fish was performed in the spring of 1999 (MassDEP 2003b). These two studies also provided the first good assessment of the degree of variability in mean fish tissue mercury concentration estimates which served as a basis for statistically-based designs of subsequent fish mercury studies. This information highlighted the importance of a number of factors contributing to variability in fish mercury concentrations estimates. If uncontrolled or unaccounted for, these sources of variability can mask the variation of interest (e.g., change due to controlling the source of the mercury). In order to better understand the magnitude of the contributions of these factors to variance in the data, several follow-on studies were designed and executed. One examined whether tissue moisture content was a significant source of variance in the data (MassDEP 2005); a second examined seasonal and fish reproductive state-related variance (MassDEP 2005) and a third, reported in this document, sought to document the scale of interannual variation, both in relation to natural factors, and also to changes in mercury inputs to the environment.

This report describes the establishment of a long-term monitoring network of lakes for fish tissue mercury monitoring in Massachusetts. The data from this effort are intended to provide several pieces of valuable information to help understand temporal edible fish tissue mercury concentration trends. This information will firstly 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 as a result of regional and national mercury emissions control efforts. The information will also address random year-to-year variation in fish mercury concentrations. In cases where data collected in different years are compared to evaluate the influence of some other variable (e.g., comparisons between urban and rural lakes), knowledge of the magnitude of random interannual variation would assist with the determination of the significance of differences attributed initially to other factors. The species monitored in

our program are largemouth bass (LMB; *Micropterus salmoides*) and yellow perch (YP; *Perca flavescens*).

Interannual variation of mercury in fish can reflect changes in mercury inputs to lake ecosystems, variation in internal processes such as mercury methylation rates, and biological and statistical variation. Interannual variation has been documented in fish tissue mercury concentrations in largemouth bass (LMB) between some, but not all years in some published studies. Lange et al. (1994) observed ~34% differences in yearly means in LMB in Florida. Jeremiason (2000) documented approximately 40% decreases in northern pike (*Esox lucius*) lake mean mercury concentrations over a >5 year period in Minnesota. Another study in remote Canadian Shield lakes did not detect interannual variation in LMB, northern pike, walleye, and cisco tissue mercury concentrations over a 3-year study period (Bodaly et al. 1993). The presence of this type of variation in a multi-year study would seem to be a function of the locale of the study, so that one generalization cannot apply to all situations. The limited information summarized above shows differences from 0-40% between years at the same location, likely reflective of year-to-year variability in environmental parameters and changes in mercury inputs. In the Everglades, multiyear monitoring data document a trend of decreasing mercury in biota. Overall, LMB and bird (great egret nestlings) monitoring data from the Florida Everglades document a significant decrease in mercury concentrations from 1990-2000 (Atkeson et al. 2003). These interannual reductions correlate with reductions in local mercury emission rates in South Florida of more than 90% since peaks in the late 1980s and early 1990s. However, the generalizability of these observations to temperate waterbodies is unclear.

The objectives of this work have been to establish a long-term monitoring network that will allow interannual variation and trends in mercury levels in fish to be assessed in temperate waterbodies and potential associations with changes in mercury emissions and deposition to be explored. In particular we have focused on a subset of lakes located in proximity to a number of historically large point sources of mercury emissions and in a predicted high mercury deposition area. This report evaluates early data returns on mercury concentrations in the two target species, spanning the 1999-2004 timeframe. Samples from a total of 17 lakes are assessed, including a subset from the predicted high mercury deposition area.

2.0 MATERIALS AND METHODS

2.1 PROGRAM DESIGN

The program objective is to document the magnitude and direction of year-to-year and long-term changes in edible muscle total mercury concentrations in LMB and YP in the designated monitoring lakes. Approximately half the lakes are to be sampled on a rotating annual cycle. Dependent upon the degree of interannual variation observed between years in the initial stages of the program and available financial resources, the duration between repeat samplings may be changed from two years in subsequent years

of the program. To date, in some years, additional numbers of lakes were sampled in regions of the state of particular interest, specifically the predicted high deposition area encompassing the northeast part of MA, in order to give more temporal and spatial resolution.

Fourteen lakes throughout the state (Figure 1) have been designated long-term fish tissue mercury monitoring sites. Figure 1 also shows additional lakes that have been sampled to augment the sampling effort at the 14 program lakes as well as any lakes initially started and then dropped for various practical reasons. Lakes were chosen using several criteria:

- lakes previously sampled;
- locations in representative ecoregions of the state;
- lakes in the predicted high mercury deposition area in northeast MA;
- lakes spanning the West–to-East distance across the state to reflect possible out-of-state long-range transported atmospheric inputs with prevailing winds;
- lakes positioned in urban and rural areas of the state;
- lakes recommended by Massachusetts Basin Team leaders;
- lakes having protected watersheds;
- heavily fished lakes;
- lakes providing habitat for species higher on the food chain.

2.2 FIELD SAMPLING

The protocols for collecting fish and water samples in the field and subsequent processing in the laboratory are shown in Figure 2 and Figure 3. Fish were collected in the spring of each year to control for the variability which can be introduced by seasonal changes in fish tissue mercury concentrations (MassDEP 2005).

Fish were collected with box nets, gill nets, trot lines, electroshocking and rod and reel. They were removed from the water, rinsed with ambient water, wrapped individually in aluminum foil, placed in polyethylene Ziploc© bags and placed on ice for delivery to the laboratory within 24 hours of collection.

In order to provide robust size/age ranges of LMB, a size spectrum of fish was collected. We sought to obtain YP greater than 20-25 cm total length to represent those consumed by anglers.

Required numbers of replicate fish were determined using sample size calculation algorithms in Statistica©. Estimates of variance in the data from our previous studies were used along with a desired confidence level of 0.10 and power of 80% to calculate required sample sizes. Our calculations and consideration of practical issues including analytical costs and concerns over potential overharvesting of resident fish populations, led us to seek 30 replicate YP per lake per sampling event and 12-15 LMB. Based on the variance values from our previous studies, these sample sizes were estimated to have an ability to identify differences in means of approximately 40-50% in LMB and 15-20% in

YP. In practice, there were occasions when it was not possible to obtain the desired numbers of fish.

Basic water quality measurements were obtained at one station at the deepest part of each lake at 1 m depth intervals with multiprobe field instruments. Temperature, pH, dissolved oxygen concentration and conductivity were measured. Dependent upon whether or not the water column was stratified at the time of sampling, either mid-epilimnion and hypolimnion water samples were taken or a single mid-depth sample was taken for analysis of major cations and anions (Na, K, Ca, Mg, Fe, Mn, SO_4, Cl), dissolved organic carbon content (DOC), total organic carbon content (TOC), nitrate+nitrite nitrogen, total phosphorus, and ammonia. The analytical techniques used for each and associated detection limits are provided in Table 1 and MassDEP (2005).

2.3 LABORATORY PROCEDURES

Fish were processed for analysis of mercury in lateral muscle in accordance with US EPA procedures (U.S. Environmental Protection Agency 1993). Total fish lengths and wet weights were recorded. The sex and reproductive condition of each fish was assessed by visual examination of gonads and classification as: Immature; Developing; Ripe; and Spent. Gonad wet weights were determined. Scales were removed from the fish for age analysis. Tissue moisture contents were determined on the 2001 fish for calculation of the dry weight basis of the mercury content of the tissues. These data are presented and analyzed in MassDEP (2005). Other details of handling and sample preparation are identical to those described in Rose et al. (1999). Mercury in tissues was analyzed using US EPA Method 245.6. A Perkin Elmer Flow Injection Mercury System was used for total mercury analysis. The method detection limit was 0.01 mg/kg and the reporting limit was 0.03 mg/kg. Recovery for mercury- spiked fish samples and precision of the analyses were 96.0 ± 11.1% and 5.5 ± 5.5 % (means ± 1 std. dev.). The reference standard for mercury in fish tissue was freeze-dried tuna tissue (BCR ref. std #463). The accuracy of analyses of that standard was 102.1 ± 12.7%. Mercury in all laboratory reagent blanks was less than the method detection limit.

The data presented for the spring of 2001 were a subset of a larger data set of results from 7 of the long-term monitoring lakes sampled in the spring, summer, and fall of 2001 and winter and spring of 2002. The larger seasonal dataset is presented and analyzed for seasonal variability in MassDEP (2005).

2.4 DATA ANALYSIS

Bivariate plots of individual fish mercury concentrations versus total fish length for each species for each lake in each year were examined to determine if there were any outliers. Outliers were either corrected if representing a data entry error or excluded if outlying the sphere of the remainder of the data. The criterion for exclusion was a subjective determination that a data point(s) fell well outside the range of others in the data set

and/or represented a mercury size relationship at odds with all the other data. Only one record was struck from the original data set. The anomalous mercury concentration value was set due to a lab fish processing error. Some fish were noted as "outliers" during data analysis and have been identified as such in the text. An examination of the bivariate plots revealed that in almost all cases, there was a positive linear correlation of fish length with tissue mercury concentrations. In order to adjust for the effects of this covariate prior to testing for mercury concentration differences between years, either an ANCOVA was performed for data sets consisting of greater than two years, or individual fish mercury concentrations were adjusted to the concentration of a standard-sized fish of that species and a t-test was performed for situations where two years were being compared. The first phase of an ANCOVA involved testing for individual lakes regression line slopes of mercury concentrations versus lengths for parallelism between different years (Sokal and Rohlf 1995). If there was no interaction between the covariate and the independent variable (length) and the classification variable (year), the second part of an ANCOVA analysis (an ANOVA) was performed, testing for between- year differences in mercury concentrations with an adjustment for length. In cases where there were significant differences between years, a Duncan's post hoc multiple range test was performed to identify which means differed from each other.

In those cases where there was a differential relationship between years of the mercury concentrations-length regression slope, the size effect was controlled for by deriving a predicted mercury concentration for a "standard-sized fish", defined as the arithmetic mean fish length over all fish sampled (339 mm for LMB; 243 mm for YP) in our study of mercury concentrations in fish from northeastern Massachusetts (MassDEP 2003a). In subsequent analyses for comparing data between lakes, the predicted mercury concentration of a standard-sized fish for a lake was used as a basis for comparison. This value was determined by regressing individual fish mercury concentrations on total body lengths for the fish species from a lake in a year, and then solving the regression equation for the predicted tissue mercury associated with the length of the standard-sized fish. Prior to running the regression analysis, plots of these two variables were examined for linearity: most of the mercury – length relationships approximated linearity. In order to retain individually-based fish data in analyses, thereby getting maximal statistical benefit out of the sample size "n" for the lake, individual fish mercury concentrations were also size-adjusted to the mercury concentration of a standard-sized fish. The rationale behind this adjustment is that the mercury-size relationship for each individual fish in the lake would follow the same relationship (slope of regression line) as that determined for all fish (least squares regression line). Lines having the same slope as the overall regression positioned to cross through each data point will have different intersection points with a vertical line at the standard-sized fish length (representing tissue mercury concentrations). This set of new size-adjusted data points for each fish for each lake was then available for use in subsequent analyses testing for interannual differences using either a one-way ANOVA or a t-test.

The fish tissue mercury concentration data or size-standardized mercury concentrations for ANCOVAs and ANOVAs for each species for each year for a lake were examined for the following characteristics to determine if they met the assumptions implicit in using parametric statistics for analysis of the data: normal distribution of the data; homogeneity

of error variances; independence of the means and variance. Normality was assessed through: generation of frequency histograms of individual fish tissue mercury concentrations and application of the Kolmogorov-Smirnov test for goodness of fit to normal distribution at $\alpha = 0.05$ (Sokal and Rohlf 1995); and generation of normal probability plots of these mercury concentrations for each lake. Homogeneity of error variances between lake tissue mercury concentrations was assessed with Levene's test (Sokal and Rohlf 1995). Plots of lake mean tissue mercury concentrations or size-standardized mercury concentrations versus associated standard deviations were examined to determine if means were correlated with errors. Violations of these assumptions of normality and errors for any species were addressed by applying $\log_{10}$ transformations to the individual fish tissue mercury concentration data prior to additional testing. For t-tests, the test was run on both the size-standardized value and a log-transformed value as a time-saving measure, rather than making all the determinations above. The same test outcome occurred in all cases except one.

All statistical evaluations in this study were performed with the Statistica/W©, Version 5.0 software package (StatSoft, Tulsa, OK, USA).

3.0 RESULTS

Lake physical and chemical variable data is presented in Appendix Table A-3, but is not analyzed in this report. Summarized lake means and standard deviations for fish mercury concentrations for each year of sampling are provided in Table 2 and plotted in Figure 4 for YP and Figure 5 for LMB. Slopes of the linear regressions of mercury concentration versus total lengths are also presented. In order to guide the reader in the types of tests performed to test for significant differences between annual means for a species at each monitoring lake, a checklist of tests performed is presented in Appendix Table A-2.

Annual means throughout the state ranged from 0.061 – 0.966 mg/kg (size-standardized values were 0.169-0.847 mg/kg) for YP and 0.070 – 1.633 mg/kg (size standardized values were 0.198 – 1.678 mg/kg) for LMB.

Of the 17 lakes having at least 2 sampling times with sufficient numbers of YP for comparison throughout the state, mercury concentrations in 13 of them decreased significantly between the first value in either 1999 or 2001 and the most recent values in 2004 (Table 3). Nine of the lakes were located in NE MA and 8 of those had significant decreases ranging from 26.0 – 61.9% with a mean change over all the lakes of –32.4%. Five of the remaining 8 lakes around the remainder of the state had mercury concentration decreases between the earliest and latest measurements (range: 20.1 – 28.0 %). The overall mean change across all of the eight lakes was –15.4%. Although a smaller percentage decrease, this mean was not significantly different ($\alpha = 0.05$) from that for the NE lakes.

The data trends for LMB were similar. Of the 17 lakes having at least 2 sampling times for comparison throughout the state, mercury concentrations in 11 of them decreased

significantly between the first value in either 1999 or 2001 and the most recent values in 2004. Eleven of the lakes were located in NE MA and 7 of those had significant decreases ranging from 16.0 to 55.2 %. The mean of the changes over all these lakes was –24.8%. Four of the remaining 6 lakes around the remainder of the state had mercury concentration decreases between the earliest and latest measurements (range: 15.9 – 36.4%). The mean change over all those lakes was –19.0%; not statistically significantly different from the NE MA mean ($\alpha > 0.05$).

The data sets for several lakes received closer, post analysis examinations as a result of their anomalous mercury concentrations in relation to predominant trends observed in all other lakes.

A fish kill of unknown origin in Pomps Pond in the spring of 2001 substantially reduced the fish populations in that lake. The few fish caught that year represented small young-of-the-year or year class 1+ fish. The YP from that lake 3 years later in 2004 were of intermediate size, representing primarily year class 3+ fish, while LMB were year class 2+ or 3+. Because of the relatively uniform sizes of these post fish kill fish, their mercury concentrations shown in Table 2 were not size standardized, nor were the interannual changes analyzed further because of the unique event which took place in this pond in the middle of our monitoring program.

YP from Johnsons Pond in NE MA initially had an apparent large temporal increase in tissue mercury concentrations between 1999 and 2004. On closer examination of the size and mercury distributions of the fish in the 1999 group and the 2004 group from this lake, it was apparent that size-standardization to a 243 mm fish wasn't appropriate, since all the fish in both years were smaller than the 243 mm size standard. There was therefore no practical basis for extrapolating to a larger fish beyond the range of measured lengths. In this case, for comparing the temporal differences between 1999 and 2004, a different approach was taken than for the rest of the data because all but one fish represented a narrow size range (210-234 mm total length) with mercury concentrations showing no relationship with size over this narrow interval. Size standardization of the 2004 group was therefore performed to the mean length of the 1999 group (221 mm). The 1999 unadjusted mercury concentration mean and the size-adjusted 2004 group mean were then compared with a t-test and were not significantly different (t-test, $p>0.05$), even though they differed by 8.3%. These data are presented in Table 2, Table 3 and Figure 4.

The LMB from Haggetts Pond exhibited a statistically significant decrease in size-adjusted tissue mercury concentration from 1999 to 2003. However, the 2004 sample represented a slight increase over the 1999 value (Figure 5). This anomalous pattern is not readily explainable. The 2005 sample, which has yet to be analyzed, will hopefully shed light on the mercury dynamics in this lake. The percentage changes in mercury concentrations presented in Table 3 for Haggetts Pond LMB are based on the 1999 – 2004 comparison, yet should be viewed cautiously. The values are reflected in the summary change value shown in Table 3.

The mean mercury concentrations for YP for Buckley Dunton Lake (2003 and 2004) in Table 2 on first examination were anomalous. Upon closer examination, the mercury concentrations of 5 of the 30 fish in 2003 fell well outside of the range of values for the other 25 fish (3 fold lower than expected concentrations based upon the mercury – size relationship exhibited by the other fish). These 5 fish were treated as outliers, removed from the regression of mercury concentration versus fish length for the 2003 fish and new size adjusted values generated based upon the new regression, the 2003 and 2004 size-adjusted data sets can be compared without the influence of these outliers. This slope based on censored data is the one shown in Table 2. The means (values shown in Table 2 were significantly different (t-test, $\alpha = 0.01$) and the direction of the change was an increase of 20.7% between 2003 and 2004 (Table 3).

4.0 DISCUSSION

Once released into the environment mercury persists and does not break down into harmless components like many other pollutants. It also bioaccumulates, or concentrates, into fish to levels up to a million times higher than in water. Although mercury is a natural element, the amount of this toxin circulating in portions of the biosphere, which interact with man today, is much higher than it was 100 years ago.

Mercury's potentially harmful health effects to humans received widespread international attention in the mid to late 1950s as a result of mass human poisonings from ingestion of contaminated fish and shellfish from Minamata Bay, Japan. Other subsequent poisoning episodes came from maternal consumption of breads made from methylmercury contaminated grains made into flour in Iraq in the early 1970s (Bakir et al. 1973) and from consumption of pork fed methylmercury-treated grains by a family in New Mexico in 1969 (Davis et al. 1994). These tragic events were the catalyst for efforts to address mercury as a global pollutant.

Mercury is a potent toxin that adversely affects people and wildlife. It can adversely affect the neurological system, kidneys, immune system and cardiovascular system. The brain and developing neurological system of the fetus and children are particularly sensitive to mercury and can be damaged by fairly low levels of exposure. Based on recent data from the United States (US) Centers for Disease Control, which measured mercury levels in the blood of women across the country, several hundred thousand newborns each year are at risk of mercury toxicity in the US because of their mother's exposure to mercury. This equates to over 10,000 newborns at risk each year in the Commonwealth. Human exposures to mercury are largely attributable to the consumption of contaminated fish, in which mercury has bioaccumulated.[3]

[3] *Wildlife can also be adversely affected by mercury, including loons, otters and fish eating mammals and even some songbirds. Data indicates that mercury exposures to loons may be high enough in the northeast to reduce their ability to reproduce and can even reach lethal levels in mink.*

Based on data from MassDEP's fish monitoring program, fish consumption advisories have been issued for over 100 specific waterbodies in MA. Overall, about 60% of all tested waterbodies have one or more species of fish with mercury concentrations that necessitate fish consumption advisories for sensitive subgroups including women of childbearing age, pregnant women, nursing mothers and children. More than 40% of the tested waterbodies require fish consumption advisories for the general public.

Mercury's serious environmental impacts, documented in the 1996 Mercury in Massachusetts Report (MassDEP 1996) and in the 1998 Northeast Regional Mercury Study[4], led MA, the other New England States and the Eastern Canadian Provinces to develop a regional strategy, the New England Governors and Eastern Canadian Premiers (NEG-ECP) Mercury Action Plan (MAP) targeting mercury pollution. The goals of the NEG-ECP MAP are to reduce New England and Eastern Canadian mercury emissions by 50% as of 2003 and by 75% as of 2010, with a long-term goal of virtual elimination. To further the goals of the regional MAP, Massachusetts adopted its own multi-agency Zero Mercury Strategy in 2000.

Under the umbrella of these initiatives mercury pollution in Massachusetts and the region has been dramatically reduced and a number of monitoring and research projects designed to evaluate progress and manage state priorities were implemented. These include collaborative efforts to establish mercury emission source inventories; measure emission reductions; monitor mercury deposition rates; model mercury deposition from local and distant sources; and track mercury levels in fish. Some of these activities have been completed, others are underway[5]. Monitoring these environmental indicators allows MassDEP to evaluate the effectiveness of policies and regulations to eliminate mercury pollution.

In order to help assess the effectiveness of mercury pollution reduction programs and to better document conditions in a region predicted to have had experienced elevated mercury deposition MassDEP focused considerable effort on the northeastern region of the state because of the following findings:

1. **Modeled atmospheric deposition rates of mercury for that region were the highest of those predicted for the northeastern U.S.**, in part attributable to a number of point sources of mercury emissions the area. As part of the regional effort in the mid to late 1990s to better understand mercury cycling in the region, the northeast states and eastern Canadian provinces conducted a computer modeling analysis of mercury deposition in the region (Northeast States/Eastern

[4] *This study provided important evidence of the need for concerted and coordinated actions to address mercury and was a key factor behind the decision to pursue a regional mercury action plan.*

[5] *MassDEP is collaborating with the; Northeast States for Coordinated Air Use Management (NESCAUM); US Environmental Protection Agency New England (EPA-NE); Environment Canada; University of Massachusetts (UMASS); University of Michigan; Northeast Waste Management Officials' Association (NEWMOA); New England Interstate Water Pollution Control Commission (NEIWPCC); New England Governor's Conference (NEGC); and Secretariate of the Eastern Canadian Premiers on a number of projects.*

Canadian Provinces 1998). Total predicted annual wet and dry deposition rates for the region of northeast Massachusetts and southern New Hampshire bounded by a 40 x 40 km grid cell from the model were greater than 100 ug Hg/m^2 (Figure 6). This rate was the highest modeled in the region and was greater than measured rates (21 – 83 ug/m^2/yr) across a variety of lakes in Vermont and New Hampshire (Kamman and Engstrom 2002).

2. **A geochronological history of mercury deposition to lake sediments in the area revealed augmented twentieth century deposition relative to other regional locations and especially increased deposition in the last two decades of that century.** Data from two lake bottom sediment cores provided an initial comparative picture of historical mercury deposition in the area (Figure 7) (Wallace et al. 2004). Lake Cochichewick located approximately 5-6 km to the east or southeast of the cluster of incinerators, which were operating in northeastern Massachusetts in the late 1990s, provided the picture for the predicted high deposition area. Echo Lake data came from unpublished work by Luce and Wallace and provide a comparative picture of historic mercury deposition in a more pristine, rurally-located lake approximately 45 km west southwest of downtown Boston. There are no local point sources of mercury emissions to the atmosphere in the region so that the mercury in the lake should reflect generalized atmospheric deposition of mercury to the lake and its watershed. Mercury concentrations in the sediments of the two lakes were similar from about 1850 through 1910 (with the exception of an unexplained spike in mercury content of the Echo Lake sediments in the early 1900s). Thereafter, mercury concentrations in Lake Cochichewick sediments increased about six times faster than those of Echo Lake. As of about 2001, the surface sediment concentrations in Lake Cochichewick were about 2.6 times those in Echo Lake (Wallace et al. 2004). Northeastern Massachusetts has an important history of industrialization dating back into the nineteenth century with the extensive burgeoning of textile mills and associated cities along major rivers such as the Merrimack River and subsequent urbanization through the twentieth century. Associated with urbanization have been activities associated with mercury emissions such as manufacturing, generation of domestic and industrial wastes, generation of combustion products to the atmosphere from widespread burning of coal for domestic heat, for coal gas production, for firing industrial boilers in the late nineteenth and first half of the twentieth centuries, and municipal-level solid waste combustion. The monotonic increase in the flux of mercury to the sediments of Lake Cochichewick throughout the twentieth century is shown in Figure 8 and approached an annual deposition rate of 90 ug Hg/m^2/yr in recent years, in concordance with the model-predicted value noted above.

3. **Early (1980s) limited sampling of various freshwater fish species from some lakes in the area suggested a fairly consistent picture of elevated mercury concentrations, sufficient to be of public health concern.**

4. **Lastly, the early data suggesting that the fish in this region had higher concentrations of mercury than those from other regions of the state, which led MassDEP to focus further sampling in this region, was confirmed by a statistically-based intensive sampling program of YP and LMB from 21 lakes in NE MA (MassDEP 2003b).**

The long-term monitoring effort, which started in 1999, focused on a number of waterbodies in this hotspot deposition area to investigate the magnitude and time course of possible reductions in response to the emission reductions set to occur in MA, New England and in the local area as a result of upcoming regulatory activities under the NEG-ECP MAP and MAZMS. By the early 2000s after the imposition of tighter incinerator mercury emissions limits, only two of three MSWCs were still in operation and no MWIs continued to operate in that part of MA. Concomitantly, better emissions controls were installed on remaining facilities in the area and across Massachusetts and New England. Overall, mercury emissions in New England and the Eastern Canadian Provinces decreased by about 54% between 1998 and 2003. During this period emissions in Massachusetts decreased by about 70% and those in the study area by about 87%. Although the exact timing of the pollution reductions over this period cannot be precisely evaluated due the large number of differing sources and regulatory requirements in play, the largest fraction occurred after 2000 when new regulations came into effect limiting emissions from MSWCs and MWIs in the New England states and MA.

The results for fish tissue mercury concentration changes over this timeframe were notable. Over the period 1999 through 2004, mean edible tissue mercury concentrations in YP and LMB exhibited fairly consistent decreases with 13 of 17 lakes across the state showing statistically significant decreases (Figure 4, Figure 5 and Table 3). The presentation of the lakes data has been segregated into two categories for interpretative purposes: (1) those lakes in northeastern Massachusetts subject to local atmospheric inputs of mercury from several large point sources of mercury; (2) those lakes throughout the rest of the state subject to more diffuse sources of mercury emissions to the atmosphere.

Within NE MA, YP had the largest overall decreases in mercury. The mean decrease over all 9 lakes sampled in that region was 32.4% and 8 of those had statistically significant decreases.

The picture for LMB was similar but not as complete because of smaller sample sizes due to difficulties capturing sufficient numbers of fish (or any fish in some cases). The variation was also greater between lakes for LMB than YP (Table 3). Four out of eleven lakes had no statistically significant changes (Figure 9). The statistically significant decreases ranged from 16 to 55.2% of the initial value. The average change was –24.8%.

For lakes around the remainder of the state, a fairly consistent picture of decreases in tissue mercury concentrations in both species was apparent (Figure 4 and Figure 5). Five out of eight had statistically significant decreases in YP tissue mercury concentrations

(Figure 9). The average change in tissue mercury concentrations for all lakes in the remainder of the state was -15.4%.

Tissue mercury concentrations in LMB in lakes around the rest of the state decreased by an average of 19.0% and statistically significant decreases occurred in four out of the six lakes sampled (Table 2, Table 3, Figure 9).

Comparison of the temporal changes in fish tissue mercury concentrations between those lakes from NE MA and those from the rest of the state should be performed cautiously because the starting points for both groups are somewhat different. Because of logistical and budget constraints, 1999 baseline testing was focused on the northeast deposition hotspot area. Thus, pre-incineration emissions reductions values (1999) were available for many of the NE MA lakes but not for lakes around the rest of the state. The first data point for these other statewide lakes was usually 2001, so that those lakes had a different baseline value for calculation of amount of change than the NE MA lakes. However, for those few lakes where a baseline mean was available in 1999, the 2001 value was not statistically different from the 1999 value (YP: Lake Cochichewick, Kenoza Lake; LMB: Lake Cochichewick, Stevens Pond) (Figure 5 and Table 2). Therefore, the 2001 values may not be bad representations of conditions in the lakes in 1999.

The timing of fish tissue mercury decreases is similar to the timing of mercury emissions reductions. The YP tissue mercury yearly means for Lake Cochichewick in NE MA plotted in Figure 10 were representative of the trends seen from most lakes (Figure 4 and Figure 5). Total statewide MSWC and MWI mercury emissions are plotted on the same time scale. The baseline level of emissions in the late 1990s prior to imposition of emissions controls is represented by the data through 2000 (approximately 3000 kg mercury per year). By 2003 after the emissions controls had been operational for 3 years, total emissions levels dropped by about an order of magnitude to approximately 300 kg per year. Fish tissue mercury concentrations showed a similar pattern. Means in 1999 and 2001 were statistically the same. In 2002 they decreased significantly ($\alpha = 0.01$) and they decreased even further in 2004. Overall, the concentrations decreased 47.4% between 1999 and 2004. The magnitude of reductions in mercury emissions is more significant in NE MA than in the remainder of the state because of the concentration of local major point sources in NE MA before 2000 and subsequent reductions in numbers of operating facilities and substantial reductions in the mercury emissions of those remaining.

The time scale over which fish tissue mercury concentration changes have been observed in relation to changes in anthropogenic mercury inputs is notable. While the closest samples that we have for comparison with post-2000 samples were from 1999, 12 months before the imposition of the emissions controls in 2000, we think that the 1999 samples were likely representative of fish tissue mercury concentrations at the time of imposition of the controls. We draw this conclusion for the same reason discussed in earlier paragraphs that we concluded that the 2001 samples were likely reflective of 1999 conditions in those lakes where we did not have 1999 samples: we cited data from 4 lakes with 1999 and 2001 data showing no differences between 2001 and 1999. Therefore we

reference our conclusions about the timing of mercury decreases to the 2000 date. The majority of the decreases occurred 36-48 months after the implementation of new emissions controls and accompanying decreases in mercury emissions. Until recently, the prevailing view has been that it would be many years (tens of years) before fish tissue mercury concentrations reflected any decreases in inputs of mercury into the environment through emissions and use reductions efforts. This perspective probably has come from the fact that mercury cycling in the environment is a complex process and substantial historical stores of mercury exist in the sedimentary environment. These stores would presumably serve as a reservoir for mercury to be reintroduced back into the aquatic ecosystem even after present day inputs are reduced.

Recent data from the Florida Everglades have recast the thinking on this issue. Fish (LMB) and birds (great egret nestlings) have more rapidly reflected decreases in local atmospheric mercury inputs than was previously anticipated (Atkeson et al. 2003). Greater than 80% decreases in the mercury content of these two species were documented over the decade from 1990-2000. Local mercury emission rates in South Florida decreased by more than 90% since peaks in the late 1980s and early 1990s. More stringent emissions control regulations for incinerators came into effect in mid 1992, which led to decreases in mercury emissions rates and closures of some incineration facilities in Florida. From the time emissions started to decrease, it took from 6-36 months before decreases in LMB tissue mercury concentrations were detected. Modeling indicated that changes in atmospheric deposition, inferred from sediment core data, may account for the recent changes in LMB mercury concentrations. The generalizability of these observations to temperate waterbodies is, however, unclear due to the rather unique attributes of the Everglades ecosystem.

The relative importance of old reservoir sources of mercury such as aquatic sediments versus newer mercury inputs has been elucidated by the results of experiments from isotopic mercury tracer field studies. They have indicated that newly deposited atmospheric mercury is more reactive (and bioavailable) than old mercury in lake systems (Hintelmann et al., 2002; Babiarz et al., 2003). These results indicated that mercury cycling in aquatic systems responds rapidly to changes in recent depositional inputs of mercury. This empirical field data and data from Florida lend support for the preliminary conclusion that decreases in fish tissue mercury concentrations seen in northeastern Massachusetts may be reflecting recent large decreases in the levels of mercury emissions to the atmosphere and subsequent deposition to aquatic ecosystems.

Our results for YP versus LMB in NE MA are concordant with the theory of preferential accumulation of recent mercury. YP in NE MA exhibited greater tissue mercury concentration decreases than did LMB (– 32.4 % versus – 24.8 % (Table 3). YP feed lower on the food web and would be more directly and quickly the recipients of smaller prey lower on the food chain, closer to benthic habitats where methylation of mercury probably takes place. LMB feed on small fish and therefore are at least one step further removed from the trophic levels where changes in mercury inputs would be see most quickly. Given also their longevity, perhaps their tissue stores of mercury are less labile than those of YP and would more strongly reflect the longer-term accumulation dynamics

of mercury than would the YP. This comparison does not hold for these two species in lakes from the remainder of the state, where the changes in recent mercury inputs were not as dramatic as those in NE MA.

While it is encouraging to see distinct, measurable environmental benefits associated with reduced mercury pollution, it should be noted that a potential human health hazard from ingestion of mercury-containing fish from many of the lakes sampled still exists because tissue concentrations have universally not decreased below the concentration limit of 0.5 mg Hg/kg (see Figure 4 and Figure 5) used by the state's Department of Public Health for issuing fish mercury consumption advisories. This situation is particularly the case with LMB, which tend to have higher tissue mercury concentrations than YP because of their higher trophic position.

In conclusion, the study results are notable for the significant decreases in edible fish tissue mercury concentrations, in particular from waterbodies located in a mercury deposition hotspot area, that occurred within 36-48 months of the adoption and implementation of comprehensive state and regional plans that effectively reduced emissions of mercury. These reductions were achieved primarily through the imposition of stringent mercury emissions controls on MSWC and MWI, as well as reductions from other regional sources. These results suggest that mercury levels in fish from temperate water bodies can be significantly reduced over a relatively short timeframe if emission sources are effectively controlled. However, although reduced, overall average mercury concentrations in fish from many of the waterbodies sampled still exceed the recommended safe consumption level. As discussed in the Massachusetts TMDL Alternative Proposal submitted to the USEPA in 2004, significant reductions from out-of-state mercury sources will likely be needed to achieve water quality and public health objectives in MA.

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TABLES

Table 1. Analytical Methods for Water Quality

Analyte	Method Reporting Limit, mg/L	Method
Na	0.02	EPA 200.7
K	0.07	EPA 200.7
Ca	0.01	EPA 200.7
Mg	0.005	EPA 200.7
SO_4	0.06	EPA 300
Cl	0.07	EPA 300
Fe	0.01	EPA 200.7
Mn	0.005	EPA 200.7
TOC	0.2	EPA 415.1
DOC	0.2	EPA 415.1
Alkalinity	0.25	EPA 310.1
NO_2	0.003	EPA 300.0
NO_3	0.002	EPA 300.0
NH_3	0.001	Standard Methods. 4500-NH_3 F
Tot. P	0.001	Standard Methods. 4500-P E

Table 2. Annual Lake Mean Fish Tissue Mercury Concentrations and Size-Adjusted Means.

AREA	SPECIES	LOCATION	YEAR	Mercury Concentration (m/kg)			slope*	Size Adjusted Hg Mean	
				Mean	n	Std.Dev.		Mean	Std.Dev.
NE MA	LMB	Baldpate Pond	1999	1.333	9	0.158	0.004	1.401	0.112
	LMB	Baldpate Pond	2004	0.421	15	0.173	0.003	0.696	0.126
	LMB	Chadwicks Pond	1999	1.170	12	0.288	0.000	1.172	0.288
	LMB	Chadwicks Pond	2004	0.986	15	0.442	0.005	1.054	0.277
	LMB	Lake Cochichewick	1999	0.579	9	0.186	0.004	0.554	0.163
	LMB	Lake Cochichewick	2001	0.699	12	0.458	0.006	0.626	0.191
	LMB	Lake Cochichewick	2004	0.363	14	0.219	0.002	0.416	0.095
	LMB	Haggetts Pond	1999	0.894	8	0.539	0.007	0.664	0.263
	LMB	Haggetts Pond	2003	0.180	12	0.127	0.001	0.260	0.091
	LMB	Haggetts Pond	2004	0.578	15	0.612	0.005	0.764	0.193
	LMB	Johnsons Pond	1999	0.607	9	0.149	0.004	0.563	0.069
	LMB	Johnsons Pond	2004	0.316	15	0.116	0.001	0.473	0.037
	LMB	Kenoza Lake	2001	1.104	5	0.254	0.003	0.948	0.207
	LMB	Kenoza Lake	2004	0.719	13	0.436	0.004	0.814	0.155
	LMB	Lake Attitash	1999	1.011	9	0.252	0.004	0.575	0.152
	LMB	Lake Attitash	2004	0.353	12	0.208	0.003	0.428	0.102
	LMB	Lake Saltonstall	1999	0.514	9	0.187	0.004	0.655	0.057
	LMB	Lake Saltonstall	2003	0.341	12	0.255	0.003	0.427	0.094
	LMB	Lowe Pond	1999	1.112	9	0.284	0.002	1.078	0.229
	LMB	Lowe Pond	2004	0.833	3	0.051	0.001	0.775	0.044
	LMB	Pomps Pond	1999	1.321	9	0.498	0.005	1.200	0.283
	LMB	Pomps Pond	2001	0.070	9	0.020	-	-	-
	LMB	Pomps Pond	2004	0.232	6	0.056	-	-	-
	LMB	Rock Pond	1999	1.633	9	0.212	0.007	1.678	0.170
	LMB	Rock Pond	2004	0.834	14	0.538	-0.001	0.752	0.530
	LMB	Stevens Pond	1999	0.612	9	0.165	0.002	0.571	0.125
	LMB	Stevens Pond	2001	0.427	11	0.355	0.003	0.561	0.208
	LMB	Stevens Pond	2004	0.318	9	0.141	0.002	0.404	0.056
	YP	Baldpate Pond	1999	0.606	9	0.228	-0.002	0.645	0.219
	YP	Baldpate Pond	2004	0.198	4	0.067	0.002	0.246	0.010
	YP	Chadwicks Pond	1999	0.664	9	0.208	0.001	0.674	0.208
	YP	Chadwicks Pond	2004	0.379	30	0.164	0.004	0.492	0.115
	YP	Lake Cochichewick	1999	0.321	9	0.093	0.000	0.321	0.093
	YP	Lake Cochichewick	2001	0.333	30	0.128	0.002	0.349	0.103
	YP	Lake Cochichewick	2002	0.235	26	0.137	0.002	0.226	0.074
	YP	Lake Cochichewick	2004	0.145	30	0.107	0.002	0.169	0.059
	YP	Haggetts Pond	1999	0.381	9	0.143	0.003	0.498	0.140
	YP	Haggetts Pond	2003	0.264	30	0.060	-0.001	0.238	0.058
	YP	Haggetts Pond	2004	0.233	30	0.063	0.001	0.310	0.057
	YP	Johnsons Pond	1999	0.301	9	0.059	-	-	-
	YP	Johnsons Pond	2004	0.177	34	0.126	0.004	0.326	0.085
	YP	Kenoza Lake	2001	0.790	29	0.370	0.009	0.535	0.164
	YP	Kenoza Lake	2002	0.966	27	0.279	0.009	0.497	0.149
	YP	Kenoza Lake	2004	0.411	30	0.221	0.003	0.396	0.141
	YP	Lake Attitash	1999	0.289	9	0.092	0.001	0.316	0.087
	YP	Lake Attitash	2004	0.150	30	0.076	0.001	0.208	0.065
	YP	Lowe Pond	1999	0.432	9	0.147	0.004	0.374	0.138
	YP	Lowe Pond	2004	0.286	30	0.132	0.002	0.268	0.096

Table 2 cont. Annual Lake Mean Fish Tissue Mercury Concentrations and Size-Adjusted Means.

AREA	SPECIES	LOCATION	YEAR	Mercury Concentration (m/kg)			slope*	Size-Adjusted Hg Mean	
				Mean	n	Std.Dev.		Mean	Std.Dev.
NE MA	YP	Pomps Pond	1999	0.536	7	0.180	0.003	0.474	0.175
	YP	Pomps Pond	2001	0.106	9	0.021	-	-	-
	YP	Pomps Pond	2004	0.121	9	0.046	-	-	-
	YP	Rock Pond	1999	0.859	9	0.180	-0.001	0.847	0.180
	YP	Rock Pond	2004	0.383	30	0.239	0.003	0.529	0.210
	YP	Stevens Pond	1999	0.457	9	0.085	-0.001	0.473	0.082
	YP	Stevens Pond	2001	0.061	1	0.000	-	-	-
	YP	Stevens Pond	2004	0.135	2	0.007	-	-	-
Rest of	LMB	Bare Hill Pond	1999	0.549	9	0.129	0.003	0.550	0.101
State	LMB	Bare Hill Pond	2004	0.533	12	0.423	0.004	0.536	0.244
	LMB	Echo Lake	2004	0.478	13	0.127	0.002	0.553	0.057
	LMB	Lake Lashaway	2003	0.522	12	0.385	0.004	0.594	0.126
	LMB	Massapoag Dunstable	1999	0.784	9	0.077	-0.001	0.742	0.057
	LMB	Massapoag Dunstable	2004	0.578	12	0.157	0.002	0.624	0.083
	LMB	Massapoag Sharon	2003	0.438	12	0.333	0.004	0.471	0.132
	LMB	Lake Nippenicket	2003	0.645	12	0.296	0.004	0.764	0.156
	LMB	North Watuppa Pond	2001	0.772	9	0.461	0.009	0.529	0.124
	LMB	North Watuppa Pond	2004	0.928	12	0.272	0.006	0.539	0.152
	LMB	Onota Lake	2001	0.241	21	0.106	0.001	0.300	0.063
	LMB	Onota Lake	2004	0.143	6	0.053	0.001	0.198	0.048
	LMB	Upper Reservoir	2001	0.716	5	0.111	0.000	0.727	0.111
	LMB	Upper Reservoir	2004	0.815	2	0.474			
	LMB	Lake Wampanoag	2001	0.856	14	0.395	0.004	0.805	0.201
	LMB	Lake Wampanoag	2004	0.511	14	0.264	0.003	0.587	0.114
	LMB	Wequaquet Lake	2001	0.554	30	0.297	0.003	0.612	0.129
	LMB	Wequaquet Lake	2004	0.842	12	0.351	0.005	0.389	0.156
	LMB	Wickaboag Pond	2003	0.291	12	0.336	0.003	0.423	0.202
	YP	Bare Hill Pond	1999	0.342	9	0.111	0.001	0.329	0.106
	YP	Bare Hill Pond	2004	0.190	30	0.057	0.002	0.263	0.041
	YP	Buckley Dunton Lake	2003	0.236	25	0.105	0.003	0.448	0.050
	YP	Buckley Dunton Lake	2004	0.212	29	0.083	0.004	0.541	0.055
	YP	Echo Lake	2004	0.253	18	0.135	0.002	0.376	0.043
	YP	Lake Lashaway	2003	0.227	15	0.110	0.001	0.299	0.093
	YP	Massapoag Dunstable	1999	0.428	9	0.157	0.004	0.418	0.109
	YP	Massapoag Dunstable	2004	0.253	30	0.119	0.002	0.327	0.084
	YP	Massapoag Sharon	2003	0.154	30	0.060	0.001	0.212	0.047
	YP	Lake Nippenicket	2003	0.344	30	0.079	0.002	0.416	0.064
	YP	North Watuppa Pond	2001	0.646	30	0.157	0.003	0.533	0.131
	YP	North Watuppa Pond	2002	0.388	30	0.089	0.001	0.375	0.074
	YP	North Watuppa Pond	2004	0.415	30	0.146	0.003	0.391	0.086
	YP	Onota Lake	2001	0.229	30	0.082	0.002	0.270	0.077
	YP	Onota Lake	2002	0.208	24	0.092	0.001	0.226	0.089
	YP	Onota Lake	2004	0.131	30	0.059	0.001	0.212	0.047
	YP	Upper Reservoir	2001	0.702	30	0.210	0.006	0.779	0.189
	YP	Upper Reservoir	2002	0.642	20	0.218	0.006	0.738	0.160
	YP	Upper Reservoir	2004	0.585	4	0.294	0.011	0.703	0.109

Table 2 cont. Annual Lake Mean Fish Tissue Mercury Concentrations and Size-Adjusted Means.

AREA	SPECIES	LOCATION	YEAR	Mercury Concentration (m/kg)			slope*	Size-Adjusted Hg Mean	
				Mean	n	Std.Dev.		Mean	Std.Dev.
Rest of	YP	Lake Wampanoag	2001	0.720	30	0.236	0.006	0.797	0.174
State	YP	Lake Wampanoag	2004	0.440	30	0.136	0.003	0.574	0.079
	YP	Wequaquet Lake	2001	0.489	30	0.129	0.004	0.413	0.094
	YP	Wequaquet Lake	2002	0.380	30	0.129	0.003	0.331	0.084
	YP	Wequaquet Lake	2004	0.296	30	0.091	0.002	0.330	0.048

* slope of regression line of mercury concentration versus length

Table 3. Mean Percent Changes in Mercury Concentrations in Species and Lake-Specific Tissue Mercury Concentrations Between First Monitoring Date and Latest Date

Area	Lake	Yellow Perch % Change[a]	Largemouth Bass % Change[a]
Northeast MA	Lake Attitash	**-34.3**	**-25.4***
	Baldpate Pond	**-61.9**	**-50.4**
	Chadwicks Pond	**-26.9**	-10.0
	Lake Cochichewick	**-47.4**	**-24.9**
	Haggetts Pond	**-37.9**	*15.1*
	Johnsons Pond	8.3	**-16.0**
	Kenoza Lake	**-26.0**	-14.2
	Lowe Pond	**-28.4***	**-28.1***
	Pomps Pond	-	-
	Rock Pond	**-37.5**	**-55.2**
	Lake Saltonstall	-	**-34.8**
	Stevens Pond	-	-29.3
	Group Mean:	**-32.4**	**-24.8**
Rest of State	Bare Hill Pond	**-20.1**	-2.5
	Buckley Dunton Lake	***20.7***	-
	Lake Massapoag- Dunstable	**-21.9***	**-15.9**
	North Watuppa Pond	**-26.6**	1.9
	Onota Lake	**-21.5***	**-34.1**
	Upper Reservoir	-5.3	
	Lake Wampanoag	**-28. 0**	**-27.1**
	Wequaquet Lake	-20.1	**-36.4**
	Group Mean:	**–15.4**	**-19.0**

[a] Bolded values represent statistically significant changes at $\alpha = 0.01$, unless noted with an '*' representing significance at $\alpha = 0.05$. Anomalous values noted in *italics*.

FIGURES

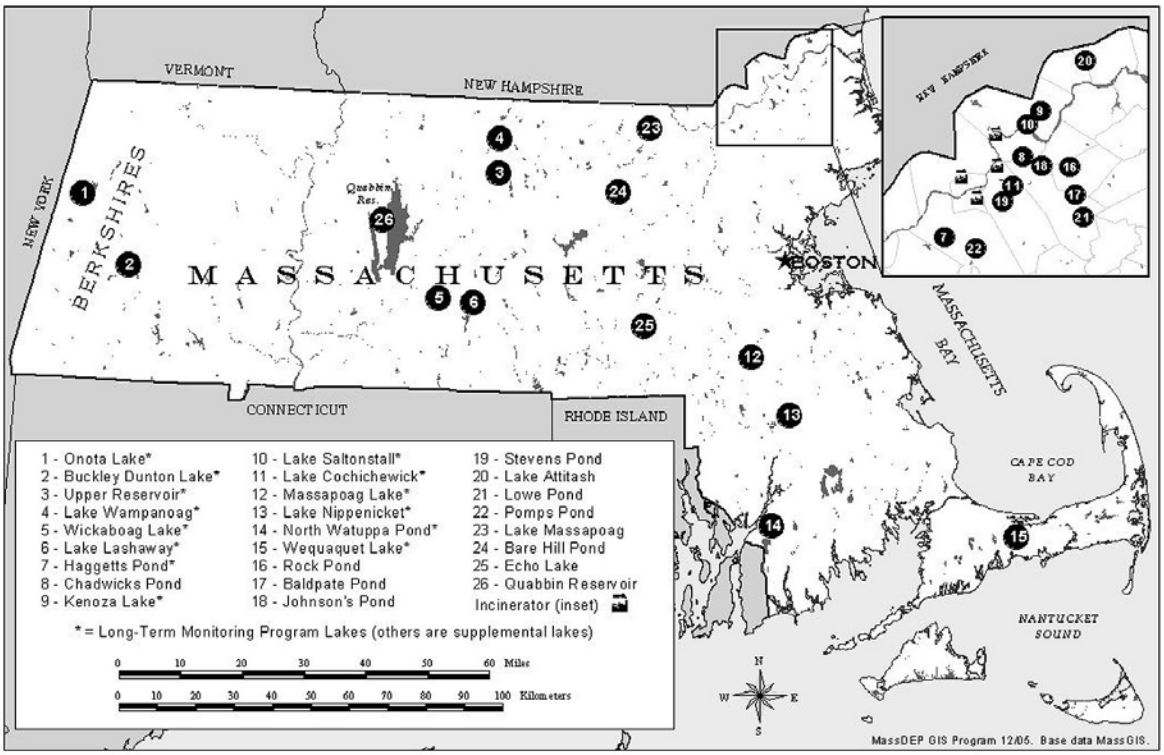

Figure 1. Locations of Long-Term Monitoring Lakes (Lake Saltonstall (#10) dropped in 2004 because of absence of YP and high LMB fishing pressure. Rock Pond (#16) substituted for it in 2005).

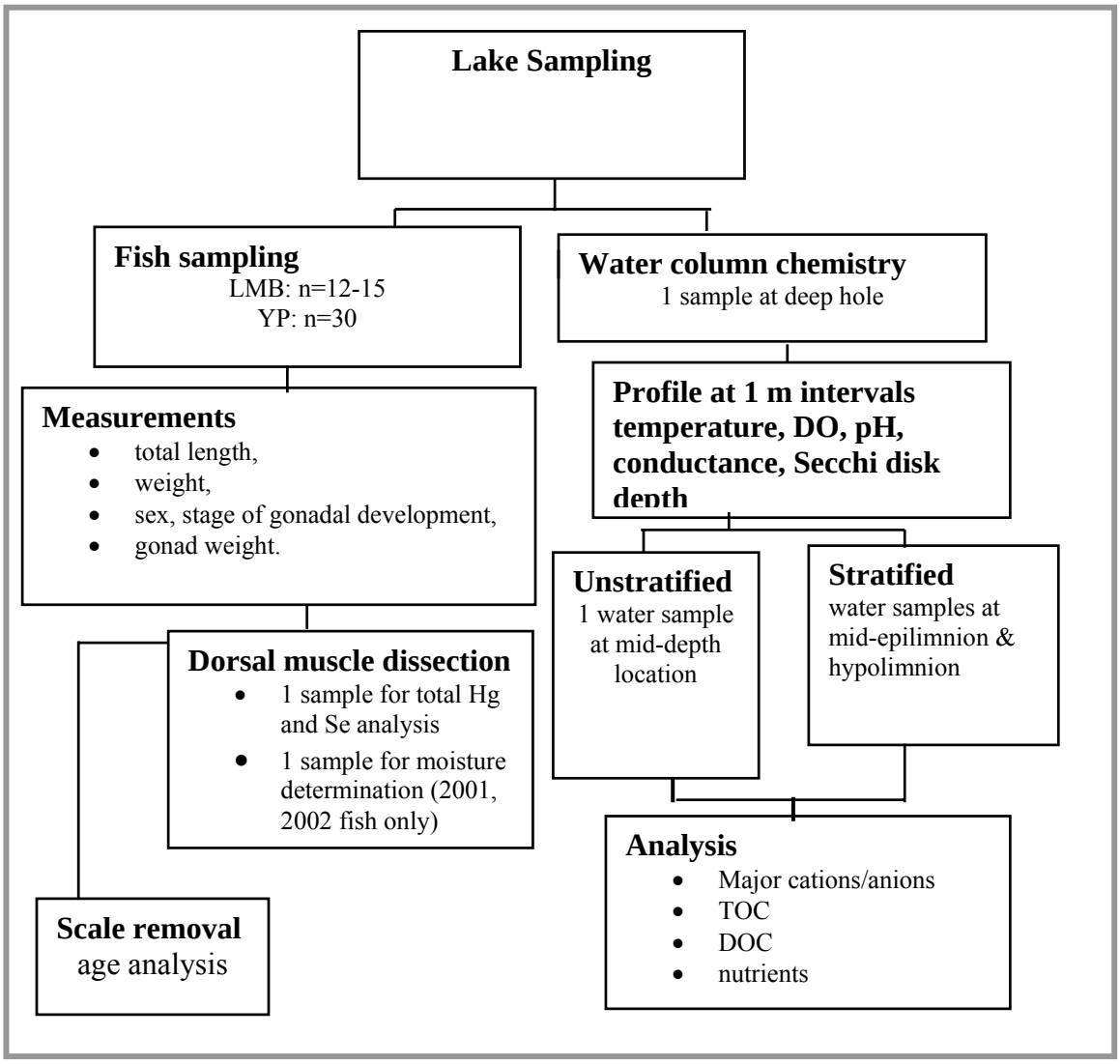

Figure 2. Field and Lab Handling Protocol.

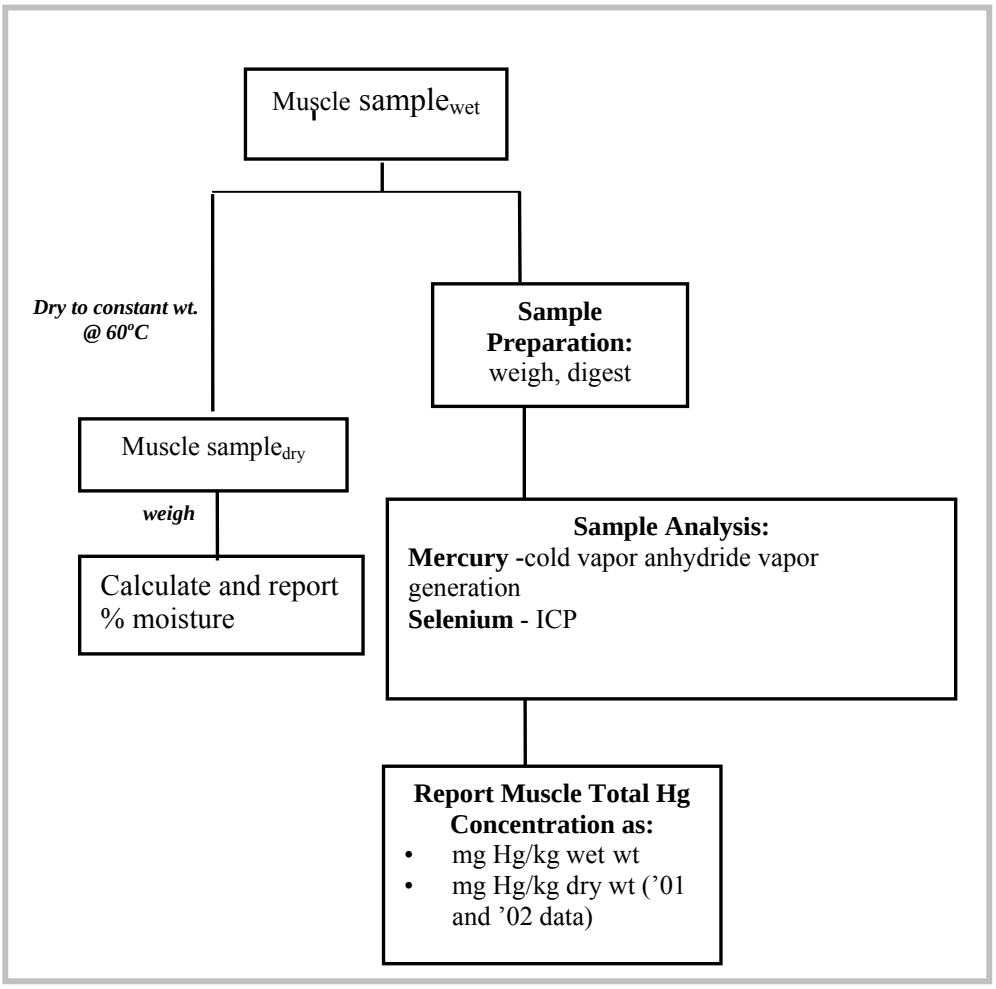

Figure 3. Fish Laboratory Processing Protocol

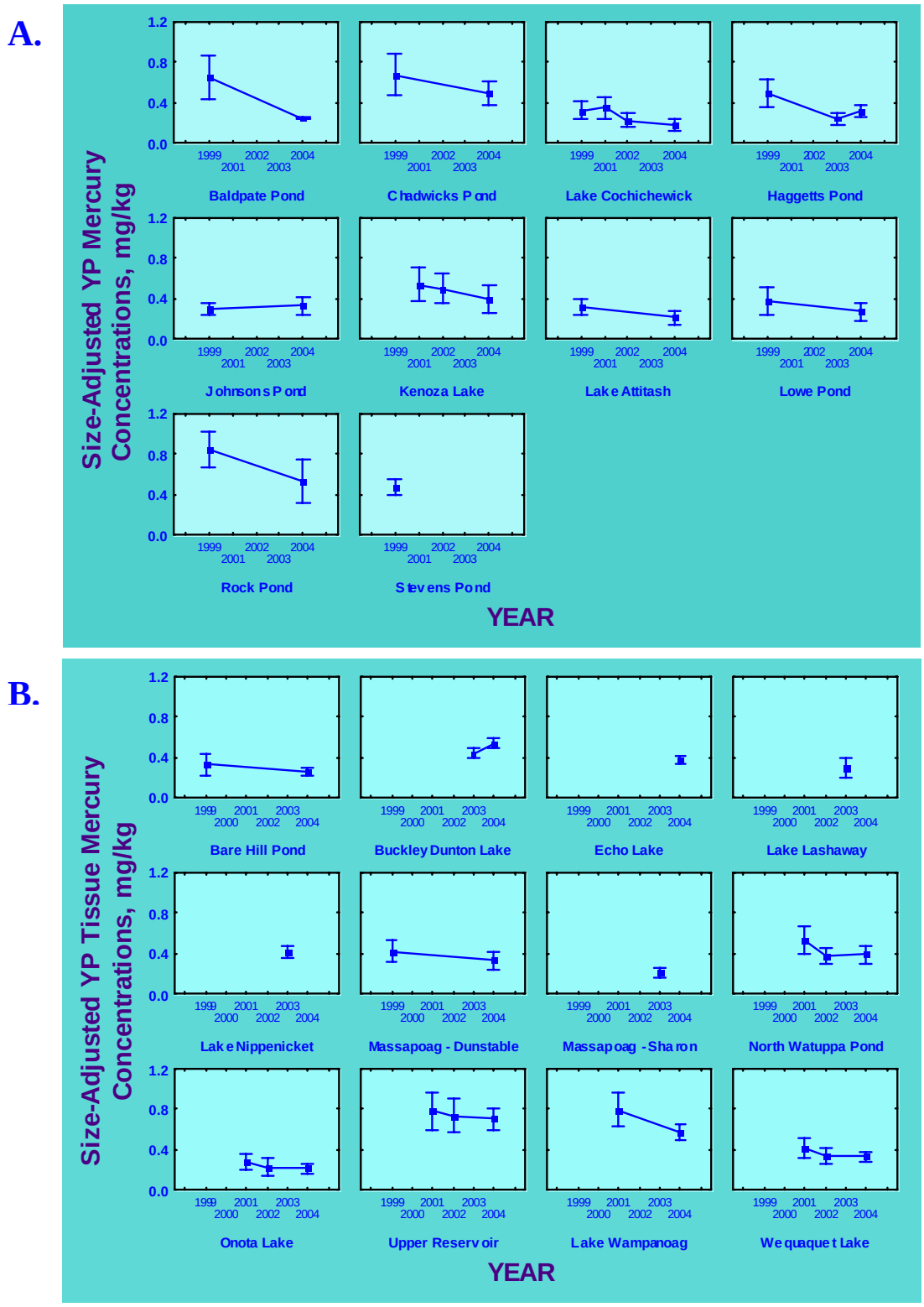

Figure 4. Size-Adjusted Annual Lake Mercury Concentration Means (± 1 std. dev.) for YP. A. Northeast Massachusetts Lakes; B. Statewide Lakes.

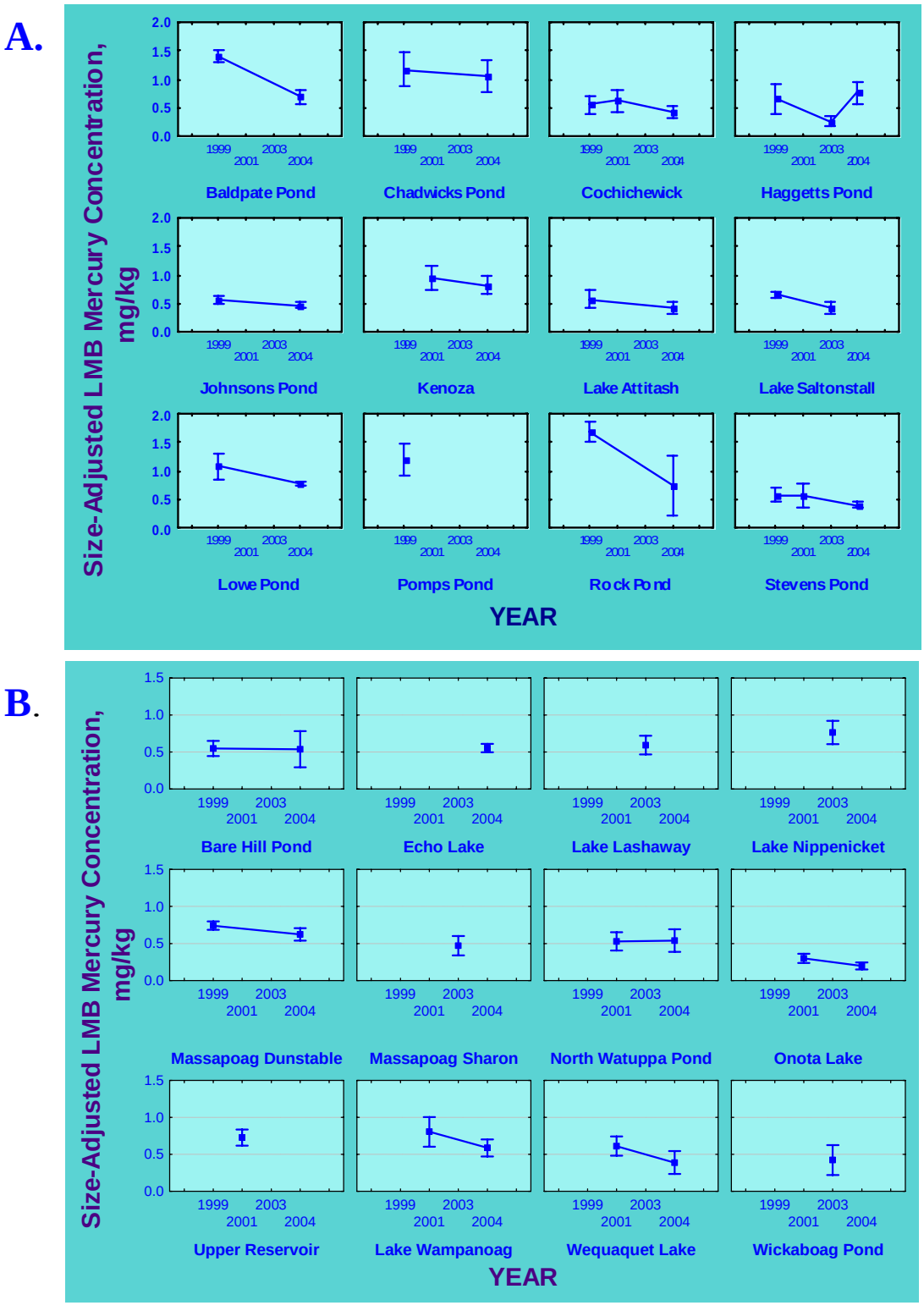

Figure 5. Size-Adjusted Annual Lake Mercury Concentration Means (±1 std. dev.) for LMB. A. Northeast Massachusetts Lakes; B. Statewide Lakes

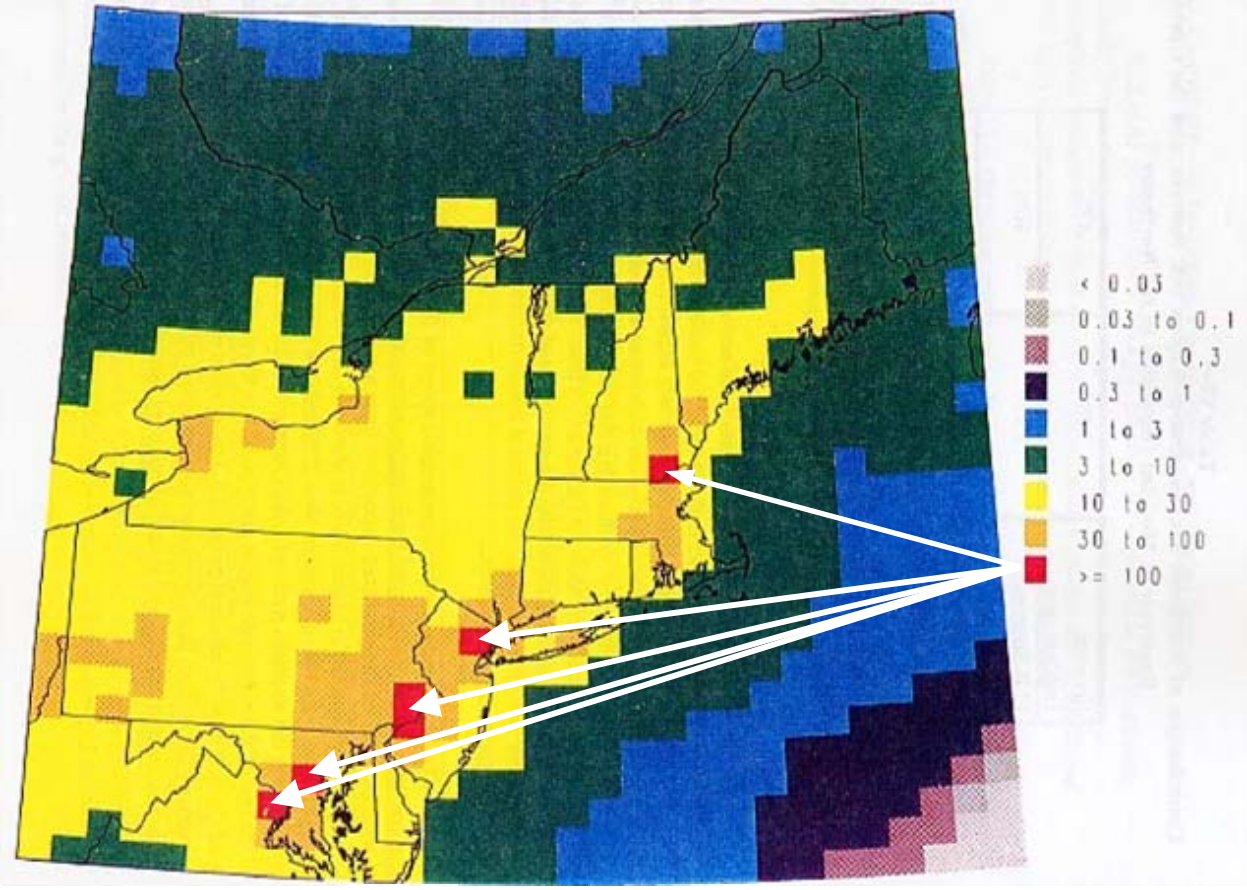

Figure 6. Predicted Annual Wet and Dry Mercury Deposition (ug/m2) from All U.S. Sources (Source: Northeast States/Eastern Canadian Provinces 1998)

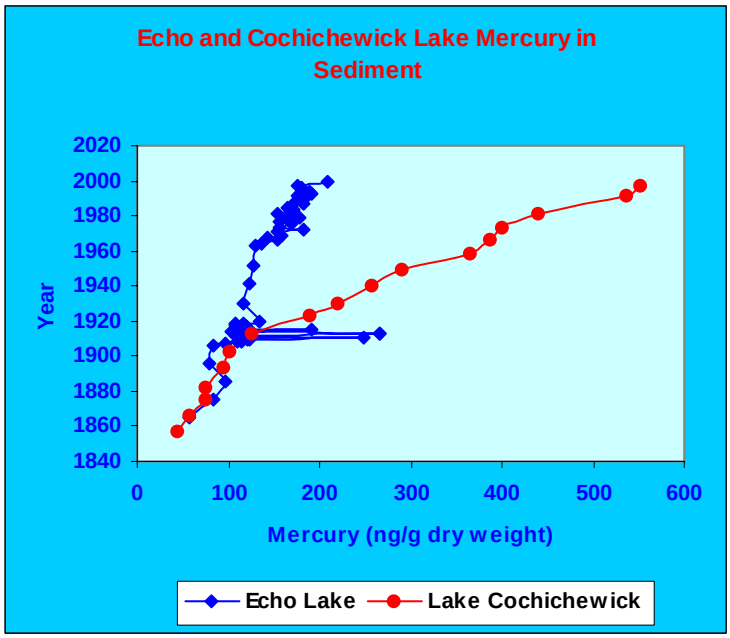

Figure 7. Sediment Core Mercury Concentration Versus Year for Echo Lake and Lake Cochichewick, MA. (Source: Wallace et al. 2004. Echo Lake Data from Wallace and Luce, unpublished data.).

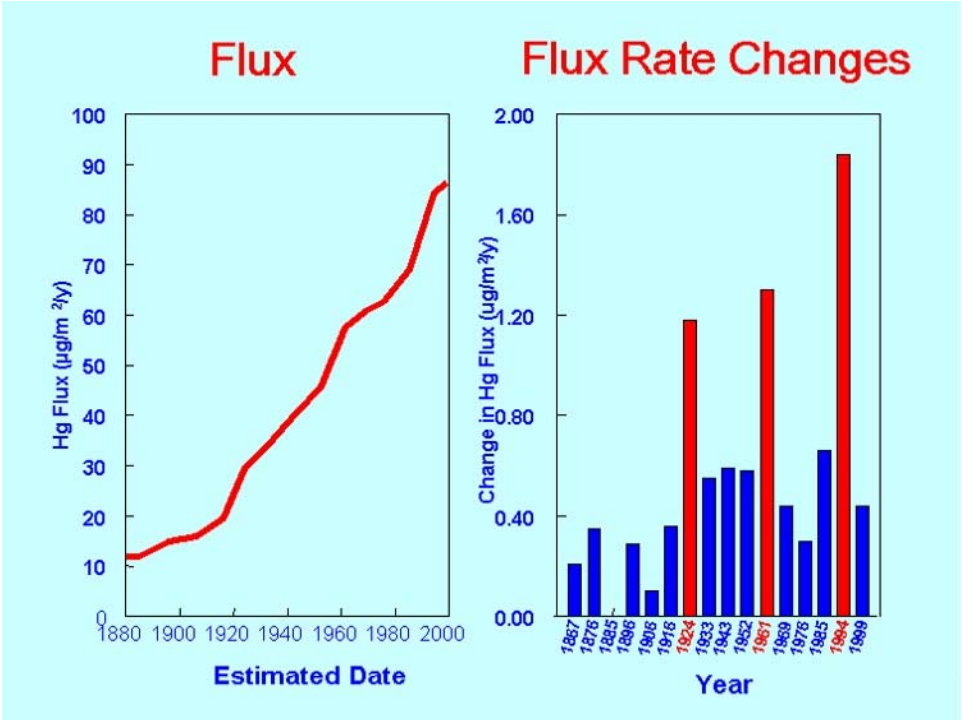

Figure 8. Mercury Fluxes Into Sediments of Lake Cochichewick Over The Last 120 Years (Source: Wallace et al. 2004)

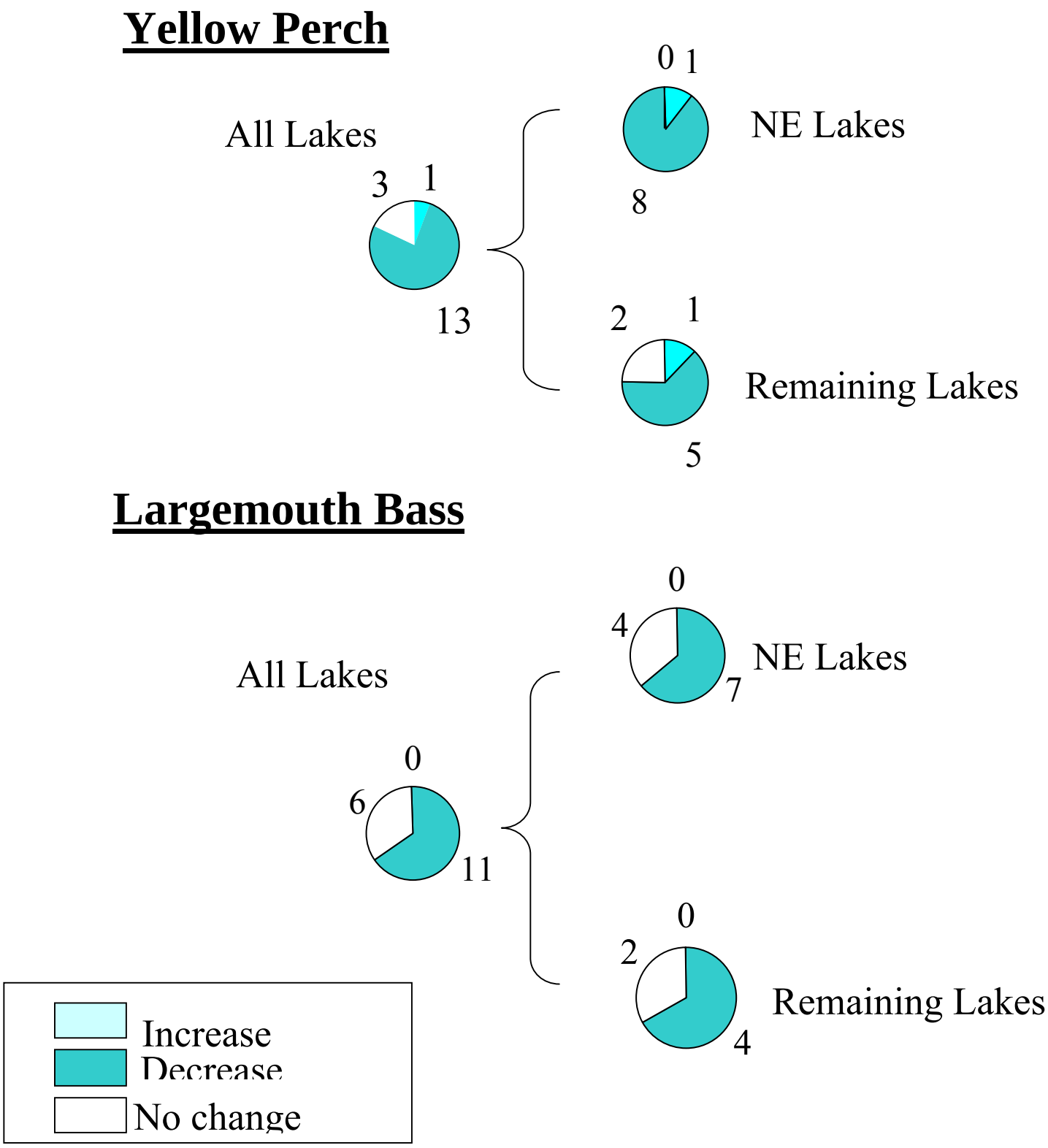

Figure 9. Numbers of Lakes Having Statistically Significant Increases, Decreases and No Significant Change in Fish Tissue Mercury Concentrations, 1999-2004. A. YP; B, LMB.

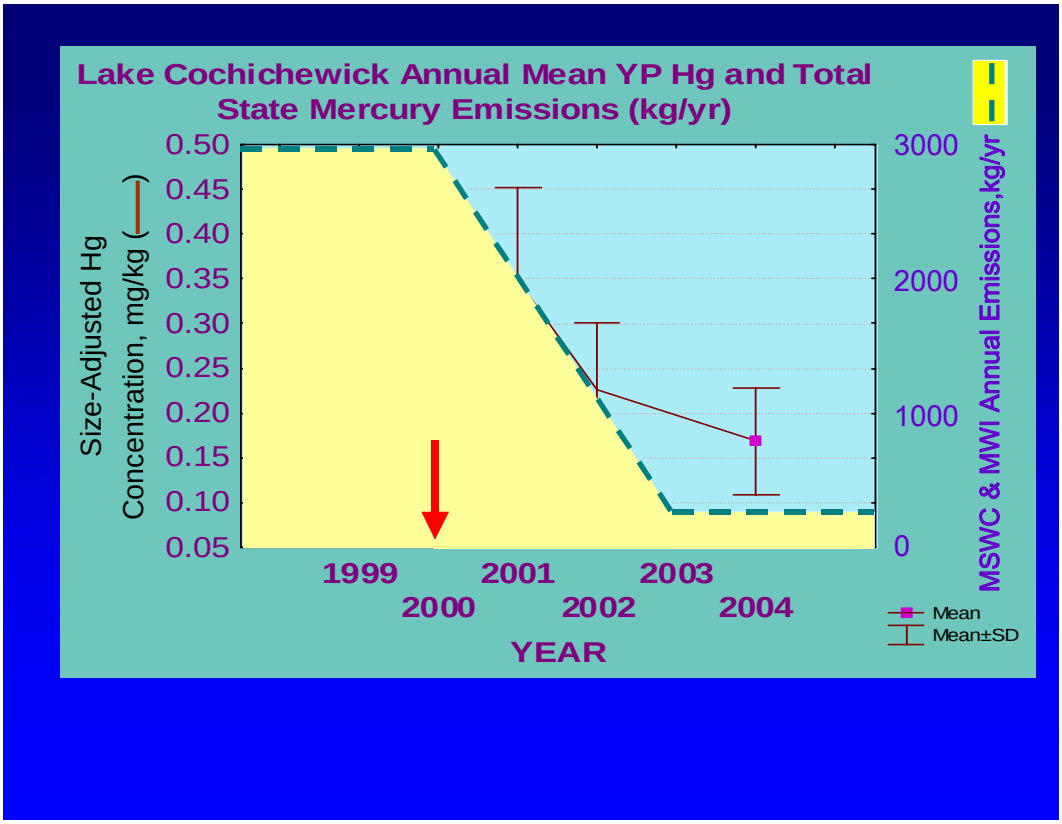

Figure 10. Representative Fish Tissue Mercury and Incinerator Emissions Changes Versus Time in NE MA.

APPENDIX

Table A-1. Long-Term Monitoring Lakes and Seasonal Variability Lakes (shown in bold)

Water Body	Acres	Town	Watershed	PALIS #	Year Sampled									
					1976	1995	1998	1999	2001	2002	2003	2004	2005	2006
Lake Wampanoag	218	Ashburnham Gardner	Nashua	81151		X			S,S,F	W		X		
Upper Reservoir	57	Westminster	Millers	35091		X				S		X		
North Watuppa Pond	1700	Fall River	Mount Hope Bay	61004		X		S,S,W	S,S	W,S		X	X	
Lake Cochichewick	555	North Andover	Merrimack	84008				X	S,S	W,S				
Kenoza Lake	287	Haverhill	Merrimack	84028					S,S,F	W,S		X		
Onota Lake	617	Pittsfield	Housatonic	21078	X				S,S	W,S	X	X		
Wequaquet Lake	654	Barnstable	Cape Cod	96333		X			S,S,F	W,S		X		
Lake Saltonstall	45	Haverhill	Merrimack	84059				X			X	▲See note		
Rock Pond¥2		Georgetown	Parker	91012				X				X	X	
Massapoag Lake	353	Sharon	Neponset	73030									X	
Buckley-Dunton Lake*	195	Becket	Westfield	32013		X						X		X
Haggetts Pond	214	Andover	Merrimack	84022				X		X	X	X	X	
Lake Nippenicket	354	Bridgewater	Taunton	62131						X			X	
Wickaboag Lake	320	West Brookfield	Chicopee	36166									X	
Lake Lashaway	270	North & East Brookfield	Chicopee	36079						X			X	
Baldpate Pond¥	55	Boxford	Merrimack	91001				X		X X		X		

Table A-1 cont. continued. Long-Term Monitoring Lakes and Seasonal Variability Lakes (shown in bold)

Water Body	Acres	Town	Watershed	PALIS #	1976	1995	1998	1999	2001	2002	2003	2004	2005	2006
Chadwicks Pond¥	161	Haverhill, Boxford	Merrimack	84006				X				X		
Echo Lake¥	123	Milford, Hopkinton	Charles	72035								X		
Quabbin Reservoir¥	25,000	Multiple towns	Chicopee	36129	1989								X	
Massapoag		Dunstable						X				X		
Johnson's Pond¥		Groveland Boxford						X				X		
Stevens Pond¥		North Andover						X				X		
Bare Hill Pond¥		Harvard						X				X		
Lake Attatash¥		Amesbury						X				X		
Lowe Pond¥		Boxford						X				X		
Pomps Pond¥		Andover						X	X[1]			X		

S,S,F,W designation in some cells for Year Sampled indicated that fish were sampled in Spring, Summer, Fall or Winter.

♠ Dropped in 2004 because no YP previously caught and small LMB population with heavy fishing pressure.

[1] Part of Food web study

[2] Added to Long-Term Monitoring List in 2005 as substitute for L. Saltonstall

* Substituted Buckley Dunton Lake for Yokum Pond because no fish caught in Yokum Pond which was originally chosen.

¥ Special sampling conducted in 2004. Not part of long-term monitoring group of 12 lakes.

Table A-2. Summary of Sampling Dates and Types of Interannual Means Difference Tests Performed

Water Body	1995	1998	1999	2001	2002	2003	2004	t-test YP	t-test LMB	ANOVA/ANCOVA YP	ANOVA/ANCOVA LMB
Lake Wampanoag	X			S,S,F	W		X	√	√	-*	-
Upper Reservoir	X			S,S,W	S		X	√ 01 v 02	-	-	-
North Watuppa Pond	X			S,S	W,S		X	√		√	
Lake Cochichewick			X	S,S	W,S		X			√	√
Kenoza Lake				S,S,F	W,S		X		√	√	
Onota Lake				S,S	W,S		X		√	√	
Wequaquet Lake	X			S,S,F	W,S		X		√	√	
Lake Saltonstall			X			X	♠See note		√		
Massapoag Lake -Sharon						X					
Buckley-Dunton Lake	X					X	X	√	-		-
Haggetts Pond			X			X	X			√	√
Lake Nippenicket						X		-	-	-	-
Wickaboag Lake						X		-	-	-	-
Lake Lashaway						X		-	-	-	-
Baldpate Pond			X				X	√	√		
Chadwicks Pond			X				X	√	√		
Echo Lake							X	-	-	-	-
Quabbin Reservoir							X(05)	-	-	-	-
Massapoag - Dunstable			X				X	√	√		

* not done because insufficient numbers of fish or samples.

♠ dropped in 2004 because no YP previously caught and small LMB population with heavy fishing pressure.

Table A-3. Physical/Chemical Parameters. All Units mg/L Unless Noted Otherwise

List of Chemical Symbols	
Alk	Alkalinity, as $CaCO_3$
Ca	calcium
Cl	chloride
DOC	dissolved organic carbon
Fe	iron
K	potassium
Mg	magnesium
Mn	manganese
Na	sodium
NH_3	ammonia nitrogen
NO_2	nitrite nitrogen
NO_3	nitrate nitrogen
SC	specific conductivity
SO_4	sulfate
TOC	total organic carbon
TP	total phosphorus

Location	Date	Season	T	DO	pH	SC,	Alk	Ca	Na	K	Mg	Mn	Fe
			°C			mS							
Kenoza Lake	2001	spring - prespawn	7.3	11.5		213.5							
Kenoza Lake	2001	spring	12.4	8.3	6.9	234.1	19.5	11.82	24.59	1.62	2.38	0.02	0.08
Kenoza Lake	2002	spring - postspawn	9.4	10.5	7.4	210.1							
Lake Cochichewick	2001	spring - prespawn	12.1	12.4		162.0							
Lake Cochichewick	2001	spring	17.7	121.6	7.5	106.4	14.4	7.61	17.04	1.68	2.04	0.13	0.12
Lake Cochichewick	2002	spring - postspawn	9.9	11.3	7.5	148.8							
Lake Wampanoag	2001	spring	16.2	8.1		106.2	0.2	2.16	14.57	0.44	0.48	0.13	0.26
North Watuppa Pond	2001	spring	16.6	8.8		75.9	1.6	2.58	8.49	0.39	0.81	0.03	0.07
North Watuppa Pond	2002	spring - postspawn	14.0	11.8	6.8	73.2							
Onota Lake Bottom	2001	spring	6.0	3.4	6.9	220.8	77.8	21.61	5.10	0.40	7.76	0.20	0.05
Onota Lake Top	2001	spring	7.5	9.5	7.7	189.8	65.1	18.80	4.45	0.33	6.60	0.01	0.03
Onota Lake	2002	spring - postspawn	9.5	5.3	8.0	208.0							
Upper Reservoir	2001	spring	16.6	4.3	4.6	64.0	-0.4	1.71	6.16	0.45	0.43	0.03	0.32
Upper Reservoir	2002	spring - postspawn	16.5	9.4	4.9	61.0							
Wequaquet Lake	2001	spring	17.6	5.1	6.6	109.1	3.3	1.22	12.37	0.96	2.00	0.01	0.08
Wequaquet Lake	2002	spring -postspawn	14.7	10.9	7.1	67.1							
Haggetts Pond	2003	spring	17.8	6.8	8.5	423.4	15.8	12.06	64.54	3.06	2.95	0.04	0.06
Lake Saltonstall	2003	spring	16.9	6.7	8.3	283.1	21.6	9.56	46.82	1.69	2.02	0.02	0.06
Lake Attitash	2004	spring	20.1	6.7	7.3	164.9	17.1	10.51	15.37	2.53	2.63	0.06	0.09
Baldpate Pond	2004	spring	12.7	7.2	8.2	194.9	21.4	10.89	29.63	1.87	3.03	0.07	0.20
Bare Hill Pond	2004	spring	19.2	6.7	7.1	193.3	8.9	6.97	21.18	1.29	1.70	0.11	0.21
Buckley Dunton Lake	2004	spring	20.8	8.7	6.8	26.3	2.1	3.23	2.15	0.48	0.56	0.03	0.25
Chadwicks Pond	2004	spring	15.8	10.9	7.8	154.3	24.3	11.10	16.02	1.69	2.78	0.17	0.08
Cochichewick	2004	spring	15.4	8.9	7.4	200.2	13.3	8.83	22.12	1.99	2.35	0.03	0.10
Johnsons Pond	2004	spring	15.2	5.2	6.7	134.7	26.6	11.39	13.15	1.97	2.66	0.19	0.12
Kenoza Lake	2004	spring	13.2	10.5	6.4	292.7	18.9	13.81	32.41	2.00	2.85	0.02	0.09
Massapoag Dunstable	2004	spring	13.8	6.5	6.6	103.3	20.9	14.39	19.01	2.21	2.05	0.16	0.17
Onota Lake epilimnion	2004	spring	20.0	8.6	8.7	156.8	69.6	22.16	5.18	0.44	7.10	0.01	0.04
Onota Lake hypolimnion	2004	spring	7.8	4.5	7.8	121.5	74.4	22.94	5.38	0.55	7.37	0.15	0.06
Rock Pond	2004	spring	17.6	5.0	6.5	223.8	19.1	11.09	23.17	1.56	2.67	0.12	0.64
Upper Reservoir	2004	spring	18.0	7.1	5.2	63.0	-0.6	1.92	7.37	0.56	0.46	0.02	0.43
Wampanoag	2004	spring	17.1	7.7	7.0	115.0	0.1	2.48	18.93	0.73	0.54	0.09	0.59
North Watuppa Pond	2004	spring	18.6	8.3	7.0	88.8	2.3	2.94	10.32	0.59	0.87	0.01	0.04
Wequaquet Lake	2004	spring	17.4	8.8	6.9	98.0	4.1	1.46	11.27	1.11	1.85	0.01	0.08

Location	Date	Season	Cl^-	$SO_4^=$	$NO3^-$	$NO_2^=$	NH_3	TP	DOC	TOC	Methods
Kenoza Lake	2001	spring - prespawn									* BDLs were entered as 1/2 DL.
Kenoza Lake	2001	spring	48.40	9.47	<.01		0.088	0.011	5.7	4.8	
Kenoza Lake	2002	spring - postspawn									* Anions- EPA method 300.0. DL in parentheses.
Lake Cochichewick	2001	spring - prespawn									(chloride=.07, sulfate=.06)
Lake Cochichewick	2001	spring	31.88	8.00		0.01	0.040	0.030	5.3	5.1	
Lake Cochichewick	2002	spring - postspawn									* Method 353.1 (.02). In 2003 they
Lake Wampanoag	2001	spring	25.78	4.40		0.005	0.040	0.010	4.8	3.1	were analyzed separately using
North Watuppa Pond	2001	spring	13.90	7.14		0.005	0.050	0.040	5.0	3.8	nitrite-N EPA Method 300.0
North Watuppa Pond	2002	spring - postspawn									MDL=2 ug/L.
Onota Lake Bottom	2001	spring	8.08	5.33		0.081	0.210	0.051	2.3	1.6	* Ammonia-N-Standard Methods 4500-NH3
Onota Lake Top	2001	spring	7.05	6.08		0.005	0.032	0.031	3.0	2.8	MDL=1 ug/L.
Onota Lake	2002	spring - postspawn									* TOC/DOC-EPA Method 415.1
Upper Reservoir	2001	spring	10.22	4.09		0.005	0.070	0.020	8.9	8.3	MDL=2 ug/L. Method for analysis for TOC/DOC.
Upper Reservoir	2002	spring - postspawn									is the same. The only difference is the method
Wequaquet Lake	2001	spring	20.00	7.16		0.005	0.010	0.030	3.9	2.4	of sampling. DOC is filtered, DOC is not.
Wequaquet Lake	2002	spring -postspawn									
Haggetts Pond	2003	spring	110.22	10.56	0.036	0.01	0.050	0.015	5.3	5.2	
Lake Saltonstall	2003	spring	77.12	5.44	0.034	0.012	0.022	0.014	4.2	4.2	
Lake Attitash	2004	spring	30.40	10.42	0.751	0.006	0.178	0.077	8.7	9.7	
Baldpate Pond	2004	spring	54.21	7.82	0.087	0.001	0.043	0.008	7.1	7.2	
Bare Hill Pond	2004	spring	41.31	5.60	0.029	0.001	0.429	0.010	5.8	5.8	
Buckley Dunton Lake	2004	spring	3.88	3.57	0.001	0.001	0.687	0.011	6.4	6.4	
Chadwicks Pond	2004	spring	30.77	6.24	0.001	0.001	0.014	0.010	8.2	8.2	
Cochichewick	2004	spring	42.37	9.43	0.001	0.001	0.021	0.011	6.4	6.8	
Johnsons Pond	2004	spring	25.80	6.65	0.002	0.001	0.076	0.018	9.2	9.2	
Kenoza Lake	2004	spring	64.66	10.99	0.002	0.001	0.011	0.008	8.3	8.1	
Massapoag Dunstable	2004	spring	44.05	5.98	0.071	0.001	0.291	0.007	4.8	4.4	
Onota Lake epilimnion	2004	spring	8.70	5.46	0.004	0.001	0.026	0.005	2.8	2.2	
Onota Lake hypolimnion	2004	spring	9.25	6.01	0.050	0.001	0.114	0.007	2.3	2.1	
Rock Pond	2004	spring	45.41	9.39	0.200	0.001	0.251	0.014	8.0	8.1	
Upper Reservoir	2004	spring	12.55	3.59	0.022	0.001	0.022	0.009	11.7	11.9	
Wampanoag	2004	spring	33.64	4.27	0.022	0.001	0.082	0.005	5.3	6.8	
North Watuppa Pond	2004	spring	17.95	6.84	0.002	0.001	0.309	0.008	4.0	3.8	
Wequaquet Lake	2004	spring	19.50	6.15	0.018	0.001	0.030	0.020	3.1	3.1	