FINAL REPORT

FISH MERCURY LEVELS IN NORTHEASTERN MASSACHUSETTS LAKES

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ABSTRACT

Fish from 26 lakes in northeast Massachusetts were sampled in order to: 1) determine if human health fish consumption advisories for mercury were necessary; 2) examine the relationships between levels of fish tissue total mercury concentrations in the study area and other regions of the State and country; and 3) examine the possible contribution of local sources of atmospheric mercury to the local fish mercury concentrations. In a recent regional report on mercury in the northeast states, this area of Massachusetts was predicted, on the basis of atmospheric dispersion modeling of sources of mercury emissions in the U.S., to have the highest level of atmospheric mercury deposition in the northeast U.S. This area of Massachusetts has had until recently three municipal waste combustors and one medical waste incinerator and has a long history of industrialization, with mercury releases occurring as early as the nineteenth century. 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. 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.

Largemouth bass (LMB) (*Micropterus salmoides*) and yellow perch (YP) (*Perca flavescens*) were the primary species sampled. Brown bullhead (BB) (*Ameiurus nebulosus*), chain pickerel (CP) (*Esox niger*), and yellow bullhead (YB) (*Ictalurus natalis*) were also obtained for advisory screening. Mercury concentrations in LMB (mean 0.89 ± 0.43 mg/kg [n=192] for all individuals) in the study area were in the top quartile of LMB mercury values derived for more rural, non-local-source-impacted Massachusetts lakes (the Green Mountains/Berkshire Highlands, the Worcester/Monadnock Plateau, and the Narragansett/Bristol Lowlands subecoregions in the west, central and eastern parts of the state respectively). Because of these 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 far upwind of the incinerators. In those 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, if not

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.

There was no obvious relationship between LMB or YP fish tissue mercury concentrations and their locations relative to prevailing wind patterns and the 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 urbanized 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) is of limited power to detect any such effects. Long-term monitoring of fish tissue and sediment mercury concentrations which are underway will provide additional information on this issue.

EXECUTIVE SUMMARY

Massachusetts (MA) has sampled 189 fresh waterbodies since 1983 as part of a statewide program to identify freshwater fish populations with concentrations of various chemicals, including mercury, that could be harmful to humans. Fish taken from 85 of the lakes and rivers have had sufficiently high, elevated levels of edible tissue mercury to warrant posting of mercury fish consumption advisories for these water bodies.

The extent of mercury contamination of our freshwater fishery resources became apparent in the early 1990s, and state agencies then began to address the mercury problem in its entirety. The Massachusetts Department of Environmental Protection's Office of Research and Standards (ORS) initiated a study in 1995 (Rose et al., 1999) to determine the distribution of mercury in freshwater fish tissue in lakes that are not impacted by known sources of mercury. The 1995 study lakes were located in three rural ecoregions of the west, central and southeastern parts of the state (the Green Mountains/Berkshire Highlands, the Worcester/Monadnock Plateau, and the Narragansett/Bristol Lowlands). The study sought to characterize a baseline for future investigations and to identify possible environmental factors associated with mercury in fish.

One of the products of the regional focus on mercury issues was a 1998 examination of sources of mercury in the state and regional rates of atmospheric deposition of mercury. The Northeast States/Eastern Canadian Provinces Mercury Study (1998) used a computer model that combined emission source information with wind and weather data to predict mercury deposition rates across New England. Elevated mercury deposition was predicted in an area extending from the northeast region of Massachusetts, including the Merrimack River Valley, into southern New Hampshire and Maine. The model assessed mercury emissions from sources outside New England (e.g. coal-fired utilities in the Midwest) and within the region (e.g., municipal waste combustors, medical waste incinerators and other combustion facilities).

The predicted high mercury deposition rate in this urbanized area of the state, combined with public requests for additional fish testing in the same area, gave rise to the present study. This study addressed concerns about public health risks from human consumption of potentially mercury-contaminated fish from the northeast region of Massachusetts. It also compared fish tissue mercury concentrations in the study area to those of fish from other regions of the State, as well as the geographical distribution of fish mercury with respect to identified local point sources of atmospheric emissions of mercury. The study was designed to sample lakes located at increasing distances from four mercury emissions point sources operating over the last approximately 20 years (3 municipal waste combustors and 1 medical waste incinerator) in far upwind, upwind and downwind directions (based on prevailing wind direction). This was the first extensive fish testing effort around an urbanized area of Massachusetts and the first in New England targeting an area of predicted maximal mercury deposition. Largemouth bass (LMB) (*Micropterus salmoides*) and yellow perch (YP) (*Perca flavescens*) were the primary species sampled. Brown bullhead (BB) (*Ameiurus nebulosus*), chain pickerel (CP) (*Esox niger*) and yellow bullhead (YB) (*Ictalurus natalis*) were also obtained for advisory screening.

The study goals were to:

1. sample fish from lakes in northeast Massachusetts where the public has access to fishing to determine the need for fish consumption advisories;

2. determine whether the frequency of advisories is greater in this area than across the state as a whole;

3. establish a baseline for fish mercury concentrations in order to evaluate trends, thereby providing an environmental results-based indicator of the success of mercury source control efforts

4. compare mercury concentrations in fish from the region with those from other more rural parts of the State;

5. determine if there are geographic differences in fish mercury concentrations within the study area related to the locations of the major point sources of mercury emissions vis-à-vis prevailing wind direction;

6. determine whether predicted high atmospheric deposition rates of mercury for the area were mirrored by fish tissue mercury concentrations.

7. evaluate the accuracy of a model, developed by MA DEP, to predict mercury levels in fish based on measures of water quality.

Of the 26 lakes studied, LMB were caught successfully in 24. 23 of these warranted fish consumption advisories on the basis of mercury levels in LMB. The one lake not meriting an advisory was located far upwind of the area with the incinerators. Mercury levels in YP and BB were generally lower than those in LMB in about 65% of the lakes, which did not warrant advisories for those species. Two lakes lacked the target species for the study and thus were not issued a fish consumption advisory. Only one lake did not receive a fish consumption advisory, even though all target species were collected from the lake.

In other more rural parts of the state, fewer than 50% of the waterbodies tested in a previous study required fish consumption advisories due to mercury levels in LMB. In contrast to LMB, YP mercury concentrations from 65% of the present study lakes were below the threshold for issuing a fish consumption advisory.

In this study, the overall mean mercury concentration over all fish caught was 0.89 ± 0.43 (1 std. dev.) mg/kg for LMB, ranging from 0.18 - 2.5 mg/kg. In YP, mean mercury concentration was 0.44 ± 0.21 mg/kg, ranging from 0.12 - 1.1 mg/kg. In BB, mean mercury concentration was 0.28 mg/kg, ranging from 0.10 - 0.52 mg/kg.

The LMB mean mercury concentrations per lake in the study area were higher overall than those observed elsewhere in the State, with values falling in the top 25% of those observed for the three rural ecoregions of Massachusetts previously sampled. YP and BB mean mercury concentrations were also greater in these northeast lakes compared to the lakes in the earlier study. The values for these two species were lower than those for LMB, consistent with our experience that these species do not tend to bioaccumulate mercury to the same degree as LMB. These results are also consistent with those of other studies, which have found that LMB and other predatory, long-lived fish species at or near the top of the food web have higher mercury concentrations than fish from lower trophic levels.

Geographic differences in fish mercury levels were not related to the proximity or location of the lakes, vis-à-vis prevailing winds, to the mercury point sources located in the area. However, the only lake in this study area in which LMB were caught and which did not require a consumption advisory for LMB was located far upwind from the point sources considered. Seasonally and weather system-dependent shifts in wind directions associated with wet deposition events may have complicated the atmospheric mercury dispersion pattern.

A model developed by MA DEP in the course of earlier fish studies, relating mercury in fish to water quality parameters, did not accurately predict mercury levels in fish in the present study.

In conclusion, this study has documented that greater mercury concentrations exist in the muscle of mercury-accumulating fish in a region of the state which has a long history of industrialization, historic point sources of mercury emissions and a predicted high atmospheric deposition rate of mercury. The study did not discern a relationship among the locations of lakes, their observed fish mercury concentrations, and the locations of the point sources considered. However, these results should be interpreted cautiously, as distances from the point sources vis-à-vis prevailing wind patterns is a crude measure of the potential for atmospheric mercury deposition which is also dependent on wet deposition events.

Additionally, the relationship between fish tissue mercury concentrations and mercury inputs to the environment is complex and poorly understood, modulated by numerous biological, physical and chemical factors (e.g., lake mercury methylation rates, lake wetland and watershed areas, lake water chemistry, bedrock geology in the region, fish physiology, variations in wind patterns and precipitation events).

The elevated mercury levels recorded in LMB in the study area may result from one or a combination of the following factors: 1) higher rates of air deposition, attributable to multiple sources, including distant and local point sources (e.g., the incinerators located in the area), a possibility that is consistent with the air deposition modeling results discussed previously; 2) the greater degree of overall urbanization and industrialization of lands within the airshed in this study, compared to those evaluated previously (e.g., Green Mountains/Berkshire Highlands, Worcester/Monadnock Plateau, Narragansett/Bristol Lowlands); 3) unique physical, chemical or biological characteristics of the lakes in the area, which might increase the bioavailability of mercury. Further research, some of which is underway, is needed to differentiate among these possibilities. Of particular interest is the extent to which mercury deposition to these lakes, as well as New England as a whole, has responded to the substantial reductions in mercury emissions that have been achieved as a result of the New England Governors and Eastern Canadian Premiers Mercury Action Plan and the MA Zero Mercury Strategy. Under these plans mercury emissions have been reduced by >50% in New England and Eastern Canada and by close to 70% in MA. Unfortunately, commensurate reductions in mercury emissions from out-of-region sources have not occurred. Regional efforts are underway to assess the current contributions of out-of-region sources to mercury deposition in New England and to assess changes in mercury deposition and fish contamination. Because of the cycling of past emissions and ongoing inputs, particularly from out-of-region sources (e.g. coal-fired utilities), it may take many years to see reductions in fish tissue mercury concentrations.

INTRODUCTION

Northeast Massachusetts (MA) has an important history of industrialization dating back into the nineteenth century with the extensive burgeoning of mills along major rivers, including the Merrimack River in northeast Massachusetts and southern New Hampshire. Most of this industry is now gone and the infrastructure for the mills is slowly being converted to non-manufacturing uses. Many of the older, larger towns are still densely populated areas, yet surrounding lands are relatively undeveloped. Associated with urbanization have been manufacturing activities, generation of domestic and industrial wastes, and generation of combustion products to the atmosphere from heating, energy generation and waste destruction. This region was recently identified through the use of an air deposition model as having the highest predicted annual levels of recent wet and dry atmospheric deposition of mercury (>100 ug/m^2) in the northeast United States (Northeast States/Eastern Canadian Provinces, 1998). This mercury input has added to the historical input of mercury and other chemicals to the environment.

Mercury deposited from the atmosphere is thought to come from longer-range transport and near-field point sources (Mason et al., 1994). The atmospheric mercury can come from anthropogenic or natural sources, such as volcanoes and earth crustal off gassing. Long-range transport-derived deposition should be relatively uniform across the entire area of northeast Massachusetts. The area which was the focus of the present study 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 (MSWC) and a medical waste incinerator (MWI) (Figure 1). Prior to that period, most trash incineration was conducted on a more dispersed municipal level. Only two facilities (MSWC) are still operational. These types of facilities were subject to new, stricter limitations on their atmospheric emissions of mercury starting in 2000. Zones downwind from major point sources (e.g., smelters, tailings piles and power stations (Goodman and Roberts, 1971) may be subject to increased deposition of a variety of contaminants. Past 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 probably all contributed to a relatively high background mercury signature in the environment.

Fish reflect elevated mercury inputs to the environment. Approximately 44% 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, Rose et al. (1999) 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 (LMB) (*Micropterus salmoides*), yellow perch (YP) (*Perca flavescens*), and brown bullhead (BB) (*Ameiurus nebulosus*). The general order of mercury concentrations in the three species was LMB>YP>BB. The study lakes were apportioned among more rural areas of three subecoregions of the state: the western Green Mountains/Berkshire Highlands, the more centrally located Worcester/Monadnock Plateau, and the eastern Narragansett/Bristol Lowlands. Fish tissue mercury concentrations only varied significantly among areas in YP and were highest in the Worcester/Monadnock Plateau lakes. The level of primary production in each lake was not a strong predictor of fish tissue mercury concentrations. YP and BB tissue mercury concentrations correlated inversely with the pH of the lake waters. LMB tissue mercury concentrations correlated most highly with the weight of the fish, the size of the lakes, and the areas of surrounding wetlands and watersheds. Predictive numerical models for each species' tissue mercury concentrations were developed from the data set. The best predictors of tissue mercury concentrations differed between species: lake water calcium concentration and water temperature for YP; fish size and the mercury concentrations in YP for LMB; and dissolved organic carbon concentration and lake water pH for BB.

In 1994, fish from a few of the lakes in the northeast part of the State were sampled as part of the State's routine fish toxics surveillance program. 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 (MA DPH, 1995) issues fish consumption advisories. More restrictive advisories are issued for concentrations greater than 1 mg/kg.

In 1998 MA DEP initiated new MWC rules that included stringent mercury emissions control regulations to lower mercury emissions up to 95%. This study was initiated before the adoption of the new controls, so that the results will serve as an environmental baseline for comparison with fish tissue mercury monitoring results in the future after the emissions reductions. The other reasons for the study were to determine the need for additional consumption advisories, to examine possible spatial patterns in the occurrence of higher fish mercury concentrations, and to compare the fish contamination situation in this localized, geographical region to more rural areas and to regional New England data. The region predicted to have higher atmospheric deposition rates of mercury may be considered an "urban airshed", containing both urban and rural land use types. The lakes sampled within this airshed were in primarily rural settings. The objectives of the study were divided into two categories:

Category 1 – Public Health-Based Lake Survey for Mercury Advisory Consideration

1. Sample fish where fishing takes place from lakes in northeast Massachusetts to determine the need for fish consumption advisories; and

2. determine whether the frequency of necessary advisories is greater in this area than in other, more rural areas of the State.

Category 2 – Regional Geographic Comparisons

3. Establish a baseline for fish mercury concentrations in order to evaluate trends, thereby providing an environmental results-based indicator of the success of mercury source control efforts;
4. Determine if there are any spatial patterns in fish mercury concentrations within the study area related to the locations of the major historic point sources of mercury emissions;

5. determine how well measured mercury concentrations match those predicted by a fish tissue mercury prediction model developed by MA DEP; and

6. compare mercury concentrations in fish from the study area with those from other parts of Massachusetts.

MATERIALS AND METHODS

STUDY DESIGN

The two broad categories of study objectives dictated the details of the study design. One set of objectives consisted of a broader survey of additional species and some additional lakes to determine whether fish consumption advisories for mercury were needed to protect public health. The second set of objectives consisted of and was met by a more intensive sampling program on a subset of the lakes sampled in the first category. For the latter category, more LMB and YP fish were obtained and analyzed individually to support the statistically-based evaluation of the relation between atmospheric mercury inputs and fish tissue mercury concentrations.

The study area was subdivided on the basis of the potential for atmospheric deposition of mercury from potentially major, local emission sources (Figure 1). While the area delineated by the regional deposition modeling project as the high deposition zone covered a large part of the study area, it was defined, in part, by the minimum spatial resolution of the model used to predict deposition. Actual patterns of mercury deposition within this artificially designated zone may not be uniform because of local point source contributions and variations in local wind directions and associated precipitation events.

The subdivision of the study area was made to improve the efficiency of the study design for the second category of objectives. The (Northeast States/Eastern Canadian Provinces, 1998) air deposition model used smaller grid areas within its study area to allow for more refined projection of deposition specific to the conditions of each grid area. We stratified the high deposition area defined by the model's grid cell into three areas (Figure 1): an area in the predominant downwind direction (Gaylor and Swirsky Gold, 1998) from the major mercury point sources (an arc from about 0-90^{o} centered on the Lawrence MSWC; an area upwind of these sources (within approximately about 7.3 km and in direction from 90-360^{o} centered on the Lawrence MSWC); and farther upwind (> 15.5 km from the western-most located incinerators) of the modelling grid, influenced likely only by deposition of mercury transported longer distances. The locations of lakes upwind and downwind of the point sources might reasonably be expected to represent the potential for lesser and greater atmospheric deposition of mercury, respectively, from the point sources. Upwind/downwind evaluations were made from a wind rose compiled from meteorological data collected from Nov. 1989 – Nov. 1990 at Ward Hill, Haverhill, MA (DiNardi et al., 1991).

The fish species analyzed were LMB, YP, BB, yellow bullhead (YB) (*Ictalurus natalis*) and chain pickerel (CP) (*Esox niger*). The lakes sampled in this study (Table 1) were chosen on the basis of: size of lake (4 hectares minimum size), availability of fish species, fishing pressure, access, and proximity to other lakes.

Category 1 – Public Health-Based Lake Survey for Mercury Advisory Consideration

The objective of this part of the work was to provide fish tissue mercury concentration data for lakes in the study area to permit evaluation of the need for fish consumption advisories to protect public health. Twenty-six lakes were sampled (fourteen from the downwind area, six from the upwind and six from the far upwind areas).

Category 2 – Regional Geographic Comparisons

The objectives of this section of the study were to: (1) quantitatively compare individual fish edible muscle concentrations of mercury in LMB and YP from lakes downwind, upwind and far upwind from potential major point sources of mercury emissions; (2) to examine spatial patterns in species mercury concentrations; and (3) compare data obtained in this study to previous studies around the State and elsewhere. This work was limited to these 2 species because LMB are known to be among the highest bioaccumulators and they are representative of an upper level trophic group. YP are ubiquitous native omnivores. Our previous work in Massachusetts lakes has shown that other species such as BB are less likely to accumulate mercury (Rose et al., 1999).

Other possible determinants of the fish mercury concentrations beyond geographically-based differences being investigated could act as data confounders. These include season, fish size/age, reproductive condition, physiological condition, water temperature, pH and conductivity, amount of available organic matter in the water, trophic status of the lakes and extent of watershed and wetlands feeding into the lakes. Many of these variables are independent and their effects on fish mercury concentrations may be difficult to differentiate.

In the design phase, the anticipated relationship between fish size and tissue mercury concentrations was addressed by limiting field collections to those fish which might be kept and consumed by anglers (i.e., minimum sizes to keep and upper cutoffs to restrict size-related variability) (e.g., LMB between 30 and 36 cm in total length and YP between 20 and 25 cm in total length). For budgetary reasons, the study was performed in the spring, unintentionally coincident with spawning season. It was therefore not possible to design the study to control for seasonally varying variables such as fish reproductive condition, fish condition index, and water temperature in the experimental design. However, relationships among these variables and tissue mercury concentrations were evaluated during data interpretation.

One determinant of lake water pH is the nature of the surrounding soils. Since the study area lies within one subecoregion of the state (Griffith et al., 1993), we did not expect any large pH differences between lakes as a result of surrounding soil differences between lakes, although there may be other site-specific factors which could be responsible for differences in pH values between lakes. The potential relationships among lake water pH, conductivity and fish mercury concentrations are explored in the data interpretation phase.

SAMPLING PROTOCOL

Category 1 – Public Health-Based Lake Survey for Mercury Advisory Consideration

The Commonwealth's fish sampling protocol for its fish toxics testing program (Isaac et al., 1992), which calls for fish composite analysis, was used on five of the 26 lakes because the resultant data were to be used for advisory analysis only. Most of these fish were processed identically to those for Category 2 objectives except for compositing of fillets for 2-3 fish. One to six individuals each for the 5 species of fish were sought from each lake with a target of 3 fish per species.

Category 2 – Regional Geographic Comparisons

A subset of 21 of the 26 lakes sampled in this study (Table 1) was subjected to more intensive sampling for geographic difference evaluation than those lakes where fish were used for advisory screening. Nine fish of each species were sought from each lake. We have previously determined that this number would be adequate to address the sampling needs for these types of comparisons, given the variability in fish tissue mercury concentrations in the state (Rose et al., 1999). Ten, five and six lakes were sampled from each of the downwind, upwind and far upwind sampling regions, respectively (Table 1). A greater number of lakes were sampled from the downwind region in order to provide sufficient coverage to investigate spatial patterns in fish mercury within that region.

FIELD METHODS

Fish were sampled with box nets, gill nets, trot lines, electroshocking, and rod and reel. Electroshocking was the preferred method because of its efficiency and the lessened chance for injury or tissue deterioration associated with some of the other methods. Fish 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. During field collections at each lake, depth profiles of water temperature, dissolved oxygen concentration, pH, and conductivity were made at one meter depth intervals throughout the water column from one station in each lake located over the deepest portion of the lake. The list of water quality parameters was a subset of that used in our previous work (Rose et al., 1999), reflecting that study's identification of the important environmental determinants of tissue mercury concentrations. Field sampling took place between 14 April and 26 May 1999, except for Hovey's Pond (fished 17 June 1999).

LABORATORY PROCEDURES

Fish were processed for analysis of mercury in lateral muscle in accordance with U.S. EPA procedures (U.S. Environmental Protection Agency, 1993). Total fish lengths and wet weights were recorded. Scales were removed from LMB and YP for age analysis. Pectoral spines were removed from BB for age analysis. Other details of handling and sample preparation are described in Rose et al. (1999). A Perkin Elmer Flow Injection Mercury System was used for total mercury analysis. Recovery for spiked fish samples and precisions of the analyses were 103.3 ± 9.1% and 4.0 ± 3.8% (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 102.5 ± 4.7%. Mercury in all laboratory reagent blanks was less than the method detection limit (MDL) of 0.02 mg/kg.

DATA ANALYSIS METHODS

Category 1- Public Health-Based Lake Survey for Mercury Advisory Consideration

Mean species tissue mercury concentrations based either on individual fish analyses or composite sample analyses gathered in this study from each of the 26 lakes were used for evaluating each lake's fish consumption health risk status. The determination of the need for fish consumption advisories for various groups of consumers was performed by MA DPH according to methods described in (MA DPH, 1995), consisting essentially of using a concentration of 0.50 mg/kg as the point at and above which advisories are required.

The frequency of advisories for LMB and YP for the lakes in northeast Massachusetts (not including the three far upwind lakes in Harvard, Lancaster and Lunenburg) was compared to the frequencies of advisories required for the two species from lakes sampled in our previous Massachusetts study of more rural lakes (Rose et al., 1999). Fisher's Exact Test was used for comparison of frequencies (one-sided test, $p = 0.05$) (Sokal and Rohlf, 1995). The LMB tissue mercury concentrations of theoretical standard-sized fish (see section below on Preliminary Data Evaluation) were used to eliminate variation due to fish size in this analysis. Standardization for size was not done for the Kenoza Lake sample, as it was a composite. While this adjustment is justifiable for comparison of frequencies of advisories, it is not appropriate to adjust for size when evaluating the acceptability of fish for human consumption from a particular lake. Each lake's standard-sized LMB tissue mercury concentrations, or the lake mean of all individual YP mercury concentrations were compared to categories of health risk defined by the MA DPH (1995): 1) concentrations below the MA DPH defined advisory cutoff concentration of 0.50 mg/kg; 2) concentrations between 0.50 – 1.0 mg/kg; and 3) concentrations >1.0 mg/kg. MA DPH uses the higher value as another cutoff for more restrictive advisories.

<u>Category 2 - Regional Geographic Comparisons</u>

Preliminary Data Evaluation

Bivariate plots of individual fish mercury concentrations versus age, length or weight for all lakes for each species and then for each lake individually were examined for relationships between the variables. In order to determine if there was a differential effect of fish size (using length as an indicator) on mercury concentrations for each species, tests of parallelism of regression line slopes (Sokal and Rohlf, 1995) of $\log_{10}$-transformed tissue mercury concentration versus length were performed on the data for separate lakes.

Two procedures were applied to the LMB data to remove the confounding effect of size noted on preliminary evaluation from other between groups comparisons of mercury concentrations. One approach was to censor the data set. Although fish size objectives were predefined for field sampling in an effort to minimize the anticipated size effect, in practice, fish over a larger size range were obtained and included in the data set. Through visual examination of plots of mercury concentrations versus length for all fish, and the frequency distribution of these values, a target size range of 27-38 cm total length was identified. Use of fish within this range represents a compromise between trying to retain as many fish as possible in the analysis for statistical power reasons and narrowing of the size range to remove the effect of size. Only fish falling within this target size range were used in subsequent analyses as noted where there was a possibility of size being a confounder in the analysis.

The other approach used to adjust for the effect of LMB size was to derive a predicted mercury concentration for a "standard-sized LMB": defined as the arithmetic mean fish length over all fish sampled. 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. It was determined by regressing individual fish mercury concentrations on body lengths for fish from the lake, and 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. In those data sets which were nonlinear, both variables were $\log_{10}$-transformed prior to running the regression analysis.

Verification of Assumptions for Use of Parametric Statistics

The fish tissue mercury concentration data for each species were examined with the following techniques to determine if they were normally distributed: 1) generation of frequency histograms of individual fish tissue mercury concentrations and application of Kolmogorov-Smirnov test for goodness of fit to normal distribution at a = 0.05 (Sokal and Rohlf, 1995); 2) generation of normal probability plots of these mercury concentrations for each lake; 3) examination of plots of lake mean tissue mercury concentrations versus associated standard deviations to determine if means were correlated with errors; and 4) inspection of error variances between lake tissue mercury concentrations for homogeneity (Bartlett's test (Sokal and Rohlf, 1995)). 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. The justification for the use of this particular transformation was that frequency distributions of tissue mercury concentrations were skewed to the right and means were correlated with standard derivations. Transformations removed the dependence of the variance on the mean and made the distributions more symmetrical, i.e., normal.

Mercury Concentrations, Fish Condition, and Reproductive Condition

The condition of fish was assessed with a condition factor calculated for individual fish as its weight divided by its cubed length (W/L^3) (Ricker, 1975). Data for all fish caught were included in the analysis because there was no apparent relationship between fish condition and length or age. W/L^3 values were examined for normality prior to further statistical evaluation. Potential differences in each species condition factor between areas and between lakes within areas were examined with a nested analysis of variance (ANOVA) of this variable between upwind-downwind areas nested in ponds with replicated measurements ($p = 0.05$) (Sokal and Rohlf, 1995). The relationship between condition index and mercury level in all fish was visually examined in bivariate scatterplots of the data points for these two variables for each species and generation of Pearson's cross-correlation coefficients.

Possible relationships between the reproductive condition of fish and their respective tissue mercury concentrations were examined with bivariate plots of each lake's mean tissue mercury concentrations for target-sized LMB and all YP versus reproductive condition for each species, sex, and reproductive condition segregated by sex. Two-way ANOVAs were performed with log-transformed mercury values for these fish to test for differences in each species mercury concentrations between sex and reproductive condition.

Spatial Variation in Mercury Concentrations in Northeast Massachusetts

A nested ANOVA with unequal sample sizes (Sokal and Rohlf, 1995) was used to test for differences in each species' mercury concentrations in lakes among downwind, upwind and far upwind potential deposition areas. The first level of classification was deposition area. Lakes were the subordinate classification category, and $\log_{10}$-transformed mercury concentrations were the dependent variable. Individual fish within lakes represent replicated observations of mercury concentration in fish for that lake. The analyses were run for the target-sized LMB, because there were no replicate observations for the "standard sized" fish predicted mercury concentration, and for all YP. In cases where there were less than three fish per lake, the lake's data were omitted from the analyses because of either the low number of replicate fish per lake or the absence of enough fish (>1) to calculate within-lake variance estimates.

Mercury in Northeast Massachusetts Lake Fish Versus Other State Fish

The mercury concentrations for all YP and standard-sized LMB for lakes in northeast Massachusetts were compared against the data for these species from our study of 24 rural, non-source-impacted lakes throughout Massachusetts (Rose et al., 1999). Mean YP mercury concentrations for each lake were calculated. The LMB Mercury concentration data for each of the lakes in that study were standardized (as described above for northeastern Massachusetts lakes) to the concentrations associated with the standard-sized fish identified for this study. The 24 lake mercury concentrations for each species were then used to identify the 25th and 75th percentile concentrations from frequency distributions of these species fish tissue mercury concentrations. These cutoff points defined three ranges of mercury concentrations which we called "low" (<25th percentile), "medium" (25-75th percentile) and "high" (>75th percentile). For each species, the numbers of lakes from this study falling into each of the three ranges based on sampling from rural, non-source-impacted lakes were then tabulated using the mercury values for each lake in this study. For YP, lake mean tissue mercury concentrations for the rural study were compared against those of the northeast Massachusetts study area (using all lakes sampled) using a one-sided t-test (H_o: $\mu_{NE\ MA} \leq \mu_{rural}$ vs. H_a: $\mu_{NE\ MA} > \mu_{rural}$). For LMB, the same test was performed on log-transformed standard-sized fish mercury concentration values (using only data from lakes where data on individual fish were available).

Fish Mercury and Lake Water Quality

The relative importance of lake water quality parameters to the variability observed in tissue mercury concentrations between lakes was assessed with a factor analysis (Sokal and Rohlf, 1995) using tissue mercury concentrations, fish condition index, and the physical variables. This multivariate statistical procedure provides a means for identifying intervariable correlation structures among numerous variables. A large, multivariate data set is reduced through this procedure to one of fewer, new, abstract variables called factors. These factors are constructed to be independent of each other, and to represent those groups of original variables in the data set which are most highly intercorrelated in terms of their variance patterns. Similar variance patterns may be inferred to represent one basis for commonality in the processes linking those groups of variables. The upwind-downwind areas were numerically coded for this analysis with downwind having the lowest numerical value and far upwind having the highest value. Pearson's product moment correlation matrices were calculated for each species mercury concentration and environmental variables. A 'varimax' normalized rotation strategy was used to improve the separation of variables on factors. Initially, the factor analysis was computed for two factors. The number of factors was increased iteratively until mercury in the species being analyzed scored highly on only one factor. Appendix A contains a more detailed general description of the concepts behind factor analysis and the interpretation of the results of these types of analyses.

Predictive numerical models (multiple regression equations) of fish muscle mercury concentrations for each species based upon lake water characteristics such as pH, conductivity, etc., had been developed from the data obtained with sampling in three subecoregions in the state in and reported on in MA DEP (MA DEP, 1997) (Table 2). These equations were applied to the present study to predict tissue mercury concentrations in the northeast Massachusetts lakes. The predictive equation for YP employed lake calcium concentrations which were not measured in the present study. A surrogate variable measured in this study was used: pH. The relationship between pH and calcium in the subecoregion study was determined with a linear regression analysis ($[Ca^{++}] = -6.54+1.495*pH$, $r^2=0.49$, sig. at $p=0.05$). This relationship allowed lake pHs measured in the present study to be used to predict calcium concentrations which then were used in the equation for mercury. These predicted concentrations are compared to the measured concentrations for that species in the lakes of the present study. Data for Center Pond, Yokum Pond and Ashfield Pond were not included because of anomalously high calcium values greater than 10 mg/L. Those ponds also had high chloride and conductivity values. Prospect Hill Pond data were also omitted because of an anomalously high pH value of 10.5.

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

RESULTS

Twenty-six lakes were sampled in northeast MA. Collected fish included 203 LMB, 160 YP, 43 BB, 11 CP and 9 YB.

CATEGORY 1 - PUBLIC HEALTH-BASED LAKE SURVEY FOR MERCURY ADVISORY CONSIDERATION

Mean mercury concentrations based on all YP caught (Table 3 and Appendix B Table B1) from 65% (11/17) of the study area lakes were below the threshold for issuing a fish consumption advisory for mercury (Table 3, Figures 2 and 3A). The YP in the remaining six ponds had mercury concentrations from 0.5 ppm - 1 ppm. None of the ponds contained YP with mean mercury levels greater than 1 ppm. The frequency of advisories (35%) warranted for this species in northeast Massachusetts where mercury concentrations were >0.50 mg/kg was greater than those needed for fish from the three subecoregions sampled by Rose et al. (1999) ($p=0.05$, Figure 3B).

Five of the six ponds requiring mercury based fish consumption advisories are approximately 4 – 14 km northeast to east of the incinerators, and the sixth pond is about 9 km SSE of the incinerators (Figure 3A). Four of the five are not directly in the predominant downwind direction (Northeast States/Eastern Canadian Provinces, 1998) from the incinerators in this area.

Four of the ponds sampled are 5.6 - 14 km in the predominant direction downwind from the incinerators, yet did not require fish advisories based on mercury in YP. One pond approximately 6 km upwind of the incinerators required a fish advisory based on mercury in YP.

For LMB, fish advisories were warranted for all but one of the ponds that were tested in the upwind and downwind directions (Table 3, Figures 2 and 3A, Appendix B Table B1) where this target species was present. The frequency with which advisories for LMB in the study area ponds (Hg >0.50 mg/kg) were warranted is greater than that for the ponds in more rural subecoregions sampled in our earlier study (p=0.05, Figure 4). As with YP populations, the distribution of ponds and their associated mercury advisories for LMB bears no evident relationship to the location of the incinerators (Figure 3A).

CP from all 4 lakes where they were caught would require fish consumption advisories for mercury (3 downwind, 1 upwind). YB for Chadwicks Pond in the downwind area merited an advisory, whereas those individuals of this species from two upwind ponds did not require advisories. BB obtained in 15 lakes had not accumulated enough mercury to warrant fish consumption advisories except in 2 lakes (1 downwind and 1 far upwind) (Table 3).

CATEGORY 2 - REGIONAL GEOGRAPHIC COMPARISONS

Summarized results are contained in Appendix B Table B1.

Preliminary Data Evaluation

The slopes of individual lake regression lines of YP mercury concentrations versus length were unequal (p=0.05). Closer examination of the plots for individual lakes did not reveal any consistent pattern in mercury – length relationships, with most having no relationship. YP mercury concentrations over all lakes were not related to fish length (Figure 5). All YP data were therefore used in any subsequent between-lake comparison of tissue mercury concentrations because of the absence of a size confounding effect.

Mercury concentrations in LMB were positively correlated with fish length (Figure 5). Slopes of individual lake regression lines of mercury versus length were significantly different between lakes (p = 0.05). This situation indicated that results from any further tests with the raw data set would be confounded by the effect of the length of the fish in each sample. A target size range of 27-38 cm was then chosen to narrow the size range of fish included in subsequent analyses using individual fish data in order to minimize the confounding effect of size on the comparative analyses between lakes. This range represented 77% (n=148) of all the individual LMB analyzed.

The total length of the “standard-sized” LMB for this data set was 33.9 cm. Central tendency estimates of tissue mercury concentrations generated for all fish caught, for fish in the “target range” and for “standard-sized” fish were very similar for almost all ponds (Appendix B Table B1). Variation about the central tendency estimates was usually greatest for the whole data set. The mean percentage difference between estimated mean mercury concentrations using the second and third methods of controlling for the differential effects of size on LMB tissue mercury concentrations (limiting the size of fish included in the calculations and use of a calculated mercury concentration for a standard-sized fish) over all lakes was 9 ± 13 % (mean ± 1 std. dev.) (Appendix B Table B1), ranging between 0 and 48%. This comparison indicates that either method produces relatively similar estimates of mean LMB mercury concentrations in a lake independent of fish size. The “target range” is perhaps more subjective since it relies upon the analyst’s judgment to identify an appropriate size range of fish to include in the analysis. The “standard-sized” fish approach uses all of the data obtained and employs a more objective process for identifying a central estimate.

Verification Of Assumptions

The raw mercury concentration values for both species (and the target size range concentrations for LBM) did not meet the criteria outlined in the methods section for use in ANOVAs and other types of parametric statistical procedures. Values were, therefore, $\log_{10}$-transformed before use in subsequent procedures; the transformations rectifying the violations of assumptions that existed with the untransformed data.

Mercury Concentrations, Fish Condition, And Reproductive Condition

YP caught in this study were 17.3 - 33.8 cm long (mean = 24.2 cm) and weighed from 52.3 to 378.3 g (mean = 176.0). These fish ranged in age from year-class 2 to 11, with the majority of fish in the fifth to seventh year class. The condition indices of these fish did not vary significantly between areas, but did vary significantly between lakes within areas (nested ANOVA: $F_{Areas} = MS_{Area}/MS_{Pond} = 0.067$ with 2,14 df, NS; $F_{Ponds} = MS_{Ponds}/MS_{Error} = 12.78$ with 14, 135 df, p=0.05) (Figure 6B).

The mean (± 1 std. dev.) YP mercury concentration of all individual YP sampled in this study was 0.44 ± 0.21 mg/kg. Concentration values ranged from 0.12 - 1.1 mg/kg. Condition index was independent of the muscle mercury concentrations (r^2=0.001) (Figure 7). Sixty seven and eight tenths percent (103/152) of YP were female. However, 3% of the fish were not classified as to sex. Ninety-six percent of the fish had spent gonads. Neither sex nor reproductive stage had a significant relationship with YP tissue mercury concentrations nor was there a significant interaction between the two variables (2-way ANOVA: $F_{sex} = 2.71$ with 1,131 df; $F_{condition} = 0.13$ with 1,131 df respectively, $F_{sex\ x\ condition} = 3.35$ with 1,131 df; for each p>0.05) (Figure 8).

The LMB sampled were 24.2 - 53.2 cm long (mean = 33.9 cm) and weighed from 151.5 - 2392.3 g (mean = 584.6). The youngest fish caught was in the first year-class, the oldest in the 14th. Most fish were in the third through fifth year-classes. There were no significant differences in condition indices between the three areas, but differences between lakes within each area were significant (nested ANOVA: $F_{Areas} = MS_{Area}/MS_{Pond} = 2.299$ with 2,18 df, p > 0.05; $F_{Ponds} = MS_{Ponds}/MS_{Error} = 3.24$ with 18,171 df, p=0.05) (Figure 6A).

The mean (±1 std. dev.) mercury concentration for all LMB caught (n=192) was 0.89 ±0.43 mg/kg. Concentrations ranged from 0.18 – 2.5 mg/kg. The condition of these fish was independent of their muscle mercury concentrations (R^2 = 0.025) (Figure 7). Forty-seven and three tenths percent (70/148) of all target-sized LMB caught were identified as female. The majority of these LMB were in a prespawning condition (40.5% developing and 49.3% ripe). Only one fish of all fish sampled was in a spent condition. Mercury concentrations of fish did not differ significantly between the two sexes, nor between those fish in different reproductive states (conclusion limited to pre-spawning fish characterized as developing or ripe) (2-Way ANOVA: F_{sex} = 0.74 with 1,127 df and $F_{condition}$ = 0.31 with 1,127 df, p>0.05)(Figure 8). There were too few spent and immature fish to include in between-group testing.

Spatial Variation In Mercury Concentrations In Northeast Massachusetts

There was no obvious geographical pattern in tissue mercury concentrations between lakes for either species (Figure 1). YP showed modest variation in mercury concentration values and no spatial pattern. Differences between the mean YP mercury concentrations of lakes within each of the three designated study areas (Figure 2) were not significant (nested ANOVA: F = MS_{AREA}/MS_{LAKES} = 1.39 with 2,14 df, p>0.05). Differences between lakes were significant (F = 7.57 with 14,135 df, p>0.05). The single fish from Crystal Lake in Haverhill was not included in this analysis.

The highest concentrations of LMB tissue mercury in the data set were in lakes which were both upwind and to an easterly angle of 45^o of the prevailing SW- NE wind direction in the area (Figure 1). Differences between the target LMB mercury concentrations of lakes within each of the three study areas (Figure 2) were not significant (nested ANOVA: F = MS_{AREAS}/MS_{LAKES} = 1.49 with 2,17 df, p>0.05). Differences between lakes were significant (F = 29.87 with 17,120 df, p=0.05). Lake Attatash in Amesbury was omitted from the nested ANOVA because it had fewer than three fish within the target size range for interpretation.

Mercury Concentrations In Northeast Massachusetts Lake Fish Versus Statewide Mercury Concentrations

The 25th percentile and 75th percentile concentrations defining the distribution of mercury concentrations from more rural, non-source-impacted lakes were 0.20 and 0.38 mg/kg for YP and 0.27 and 0.49 mg/kg for standard-sized LMB.

The YP mercury concentrations from eight of the northeast Massachusetts ponds (Figure 3B) fell into the medium range of the rural lake values. The YP mercury concentrations of the remaining nine study area ponds were in the high range of the rural lake values. None of the northeastern Massachusetts lake values were in the low range of rural lake values. Northeast Massachusetts YP mean lake mercury concentrations as a group were significantly greater than those of more rural lakes ($t=3.22$, 40 df, $p = 0.05$) (Figure 9).

Seventy-five percent of the LMB populations sampled in the 1997 study (Rose et al., 1999) contained less than 0.49 mg/kg mercury. All of the standard-size LMB lake mean tissue mercury concentrations from the northeast study area ponds, except Fort Pond, far upwind, were greater than this value (the high range of the more rural pond values). The northeast study area lake fish tissue mercury concentrations for standard-sized LMB as a group were significantly greater than those of lakes from more rural areas ($t=4.97$, 32 df, $p<0.05$).

Although the data set for BB was not intended for detailed statistical analysis, there were sufficient numbers of samples to enable a comparison of lake mean concentrations between the northeast Massachusetts lakes and those of more rural areas of the rest of the state. Mean lake mercury concentrations (log-transformed) of fish from the northeast study area were significantly greater than those from more rural areas lakes (1 sided t-test: $t=2.89$, 35 df, $p=0.05$).

Fish Mercury And Lake Water Quality

The only strong relationships between fish tissue mercury concentrations and the environmental variables or the condition of the fish were for the LMB mercury concentrations, YP condition and water temperature (Table 4, Factor 1). YP condition and water temperature had similar variance patterns. The high factor loading scores of both YP and LMB tissue mercury concentrations on the same Factor (3) indicated that they had similar variance patterns and were positively correlated. The moderate, negative condition index factor score for LMB on this factor can be interpreted as indicating that the mercury concentrations in the two principal fish species studied were inversely related to the condition index of LMB. Fish mercury concentrations and the probable depositional areas of mercury from the incinerators related to up– and downwind directions are independent, as shown by their loadings on different independent factors. None of the lake environmental variables were significantly different when grouped and compared by upwind and downwind lakes, and by far upwind lakes to downwind lakes (t-test, $p>0.05$). Other relationships between variables which were revealed by the factor analysis are: similarities between water conductivity and the defined potential depositional areas for mercury (i.e., Factor 2. higher conductivities associated with lakes in more upwind directions) (Figures 3 and 6F); and between water pH and dissolved oxygen concentration (Factor 4). The apparent relationship between conductivities and assigned depositional areas is likely a data artifact because of the anomalously high conductivity value of 647 µS/cm for Ames Pond. When lake conductivities are grouped by depositional area, there are no significant differences between the groups, with or without the high value included (1-Way ANOVAs: $F_{2,18}=2.28$, $p>0.05$; $F_{2,17} = 2.38$, $p>0.05$ respectively).

Predicted mercury concentrations for YP and LMB tissues using the model from the rural lakes study and values of physical variables measured in this study were substantially greater than those actually measured (median of 5x for YP and median of 5.6x for LMB)(Results not presented).

DISCUSSION

The results from this study provide a valuable perspective on the ecological and human health ramifications of mercury inputs to urban areas. Regionally elevated tissue mercury concentrations in some fish species were consistent with projections from limited prior fish sampling and from modeling of atmospheric deposition inputs of mercury to the ecosystem, yet there were no obvious relationships apparent between the concentrations of mercury in the tissues of these species in lakes in northeast Massachusetts and the locations of those lakes with respect to recognized point atmospheric emission sources of mercury (MSWC and MWI) (Figure 1). LMB contained more mercury than YP from the same water bodies, consistent with the results of our earlier work in Massachusetts (Rose et al., 1999). The high LMB tissue mercury concentrations translate into a public health risk when these fish are consumed by humans. The concern is particularly focused upon pregnant women, infants and young children, who are all susceptible to the toxicological effects of mercury. Using criteria employed by the MA DPH, fish consumption advisories for mercury were warranted for LMB for all but one of the lakes in the study area in which this species was caught . This lake, Fort Pond in Lancaster, was in the far upwind area out of the region of high predicted atmospheric deposition of mercury. The frequency of advisories for northeastern lakes is greater than that seen in more rural parts of the state. A number of consumption advisories for mercury in YP were warranted. The frequency of advisories was barely significantly greater than that required in more rural areas.

Atmospheric inputs of mercury to terrestrial and aquatic ecosystems are recognized to come from long-range transport and local sources. The elemental (Hg^o) and ionized (Hg (II)) oxidation states of inorganic mercury are the primary forms of mercury in the atmosphere. The predominantly elemental, gaseous Hg^o is transported back to earth mainly through dry deposition, whereas the more reactive, water soluble Hg (II) is removed from the atmosphere much more quickly through wet and dry processes (Lin and Pehkonen, 1999). With current levels of control technology in the United States on point sources of mercury such as combustion facilities, most of the Hg(II) is believed to be captured, and the emitted mercury is in the Hg^o state, having a longer atmospheric residence time and therefore longer time and further distance transport before deposition (Lin and Pehkonen, 1999). Some unknown, but probably large, proportion of the mercury historically emitted by emission sources in northeast Massachusetts would therefore be expected to have been transported out of the immediate downwind areas. The local deposition component would reflect dry deposition and precipitation scavenging of particulate mercury.

A number of studies, but not all, have reported or suggested gradients of decreasing mercury in various media with distance from point atmospheric sources (Table 5). The absence of a clear relationship between fish mercury concentrations and locations of mercury emission point sources in northeast Massachusetts suggests that the fate of mercury in the ecosystem from introduction to sequestration in biological tissues is a complex one, modified by many intervening processes, such as biological methylation and water and sediment geochemical processes. The presence of multiple sources makes inferring relationships between inputs from any one source and concentrations in fish in any particular lake difficult. The association of precipitation events, which would wash mercury from the atmosphere, and wind directions at the times of those events would also tend to "smear out" the pattern of dispersion of mercury if precipitation was not primarily associated with predominant winds (SW → NE).

There were interspecific differences in the degree of fish tissue mercury contamination in the study area. LMB had tissue mercury concentrations on average 2.2x higher than values from more rural, non-local-source-impacted regions of the state, and had similarly elevated concentrations comparable to LMB in a number of other studies (Figures 2 and 9). The general level of mercury contamination of YP from northeast Massachusetts was similar to that of YP from more rural regions of the state and from other areas of the country (Figure 9).

The urban-rural differences in fish tissue mercury concentrations for LMB, while intuitively consistent with the expected situation from high predicted atmospheric mercury inputs from the urban region, are counter to the results of several studies elsewhere. In a pilot, unpublished, nationwide study of fish tissue mercury concentrations from 20 watershed basins throughout the United States, mercury concentrations in predominantly bass species ranked as follows when categorized by predominant land use of the lands surrounding lakes sampled: agriculture/forest >> mine-impacted > agriculture > urban (Brumbaugh et al., 2001). In the urban watersheds, lake sediment mercury concentrations (both total and methyl mercury) often ranked high, while fish tissue concentrations ranked low. Mercury loading to urban Minnesota lakes was 35% greater than in rural areas (Swain, 2000). The highest mercury concentrations in predacious fish occurred however in more rural parts of the State (Jeremiason, 2000). This situation was attributed to greater production rates of methyl mercury in the more rural lakes than in the urban lakes (Jeremiason, pers. comm.). LMB tissue mercury concentrations in fish from drainage lakes in highly urbanized areas of Connecticut were less than those in fish from rural areas (Hanten et al., 1998). Total mercury in atmospheric precipitation was higher in the Connecticut urban areas than in more rural areas of the State (Carley and Perkins, 2000). Regional differences in LMB tissue mercury concentrations were attributed to the influences of geology on lake water chemistry, rather than urbanization and atmospheric deposition (Hanten et al., 1998). The Connecticut urbanized lakes were in areas of either metamorphosed limestone forming a marble valley, or sedimentary rock held together by carbonate materials. The lakes in northeastern Massachusetts occur in glacial sediments over metamorphic and granitic bedrock and therefore have less buffering capacity and a tendency for lower pHs than the Connecticut urban lakes. Since fish tissue mercury concentrations have often been related to water pH in the literature (Grieb et al., 1990); (Driscoll et al., 1994); (Watras et al., 1998)), the differences between Connecticut urban and Massachusetts urban lakes and their respective rural counterparts may reflect the geologic influences from underlying bedrock. The two state comparative studies suggest that while inputs (atmospheric or otherwise) of mercury to waterbodies may play a role in the final mercury concentrations seen in fish, in-lake or watershed and wetland biogeochemical processes, including methylation, may play a more important role in the production and availability of organic forms of mercury for absorption by invertebrates and fish. Nationally, the ranking of fish tissue mercury concentrations by land use was paralleled by the methyl mercury water concentrations (Brumbaugh et al., 2001), supporting the idea of the importance of methylation. We have recently supported work which has documented that the sediments in one lake in the downwind zone of our study area have had a greater mercury deposition rate since shortly after 1910 ((Wallace et al., 2003)) than has occurred in a more rural lake elsewhere in the state (Luce and Wallace, unpublished data). This information supports the predictive modeling results for the region.

The results from other comparative studies on LMB (Figure 9) further support the conclusion that LMB tissue mercury concentrations in northeast Massachusetts are high. The general level of mercury in LMB tissues in Maryland lakes located as far as 15 km from a coal-fired power plant was less than that seen in northeast Massachusetts (0.43 ppm maximum in LMB from the

Maryland lakes vs. 2.5 ppm maximum in the northeastern Massachusetts lakes of this study) (Pinkney et al., 1997). The Oregon reservoir data (Park and Curtis 1997) represent a water body not influenced by point sources of mercury. However, fish in reservoir impoundments have high tissue mercury concentrations as a result of mobilization of mercury sequestered in flooded organic matter (Bodaly et al.,1984). This comparative value from a reservoir might therefore be biased high relative to values for seepage lakes. In a preliminary analysis of nationwide tissue mercury data for LMB (n=50), Brumbaugh et al. (2000) identified a mean tissue concentration of 0.51 mg/kg in age 3 fish. The corresponding mean value for this study's age 3 fish was 0.80 mg/kg. YP concentrations measured in this study were similar to those in other studies in Minnesota, Wisconsin and New York.

A number of factors could potentially influence the levels of mercury in fish tissue in this and other comparative studies beyond those discussed above (e.g., mercury inputs, geology, water pH, dissolved organic carbon content, alkalinity, hardness). Potentially important among these are fish size and age, food chain length, year-to-year variation, seasonal factors, fish condition index, reproductive state and the sex of the fish.

Older, larger, predacious fish (LMB, CP) tend to accumulate more mercury as they age (Rose et al., 1999). Data from the present study indicate that differences in mercury concentrations between smallest and largest fish may be about one order of magnitude. This degree of difference illustrates the importance of controlling for age or size as a confounder when interpreting tissue mercury concentration in some species of fish. Failure to account for this source of variability in the data can lead to incorrect conclusions derived from comparisons between fish tissue mercury concentrations from different samples. We corrected for the potential effect of fish size two different ways: by censoring data, and by standardizing for fish size. Use of censored data on individual fish results in fewer numbers of fish tissue mercury concentrations used in analysis. Use of a standard-size fish's mercury concentration uses the information on all fish gathered to generate a regression-derived single mercury concentration for a standard-sized fish.

Interannual variation can reflect changes in mercury inputs/losses to lake ecosystems, variation in internal processes such as mercury methylation rates, and biological and statistical variation. Little year-to-year variation was seen in LMB, northern pike, walleye and cisco tissue mercury concentrations over a three year study period in remote, northwestern Ontario lakes (Bodaly et al., 1993). Park and Curtis (1997) recorded substantial interannual variation, but comparisons were based on weak statistics. In Minnesota, where mercury loading to lakes has decreased by approximately 30% since 1970 (Swain, 2000), northern pike tissue mercury concentrations have decreased over a greater than 5 year period (Jeremiason, 2000). We are unable to quantify the likely contribution of interannual variation on the data presented in this report.

We have designed and initiated a project to chart the long-term trends in freshwater fish tissue mercury concentrations in a group of sentinel lakes in Massachusetts. This project will provide data on the magnitude of interannual variation in this parameter in LMB and YP in the state and will allow for tracking the directions of any changes in fish tissue mercury concentrations since the state's vigorous initiatives to reduce mercury use and emissions in the Commonwealth. A total of 14 lakes were identified for long-term monitoring, with half of them to be sampled every 2-3 years. The first 7 were sampled intensively each season from the spring of 2001 through the spring of 2002. Data obtained represent the first data point for the long-term picture and also are used as described in subsequent paragraphs to examine seasonal variation in the tissue mercury concentrations of these fish. The remaining 7 lakes were sampled in the summer of 2003. These data sets are presently being analyzed. The sampling cycle will be repeated in the spring of 2004 with 7 lakes.

Seasonal factors may result in apparent changes in tissue mercury concentrations. Temperature and photoperiod changes throughout the year drive the fish reproductive cycle. The time of year, seasonal temperature, reproductive state of the fish and their physiological condition are interrelated and their relative influences with respect to interpreting tissue mercury concentration data may be important. Associated changes in fish physiology and biochemistry take place as lipids are stored and mobilized, differential growth in tissues takes place and the overall condition of individuals changes as gametes are shed. The relationships between all of these factors and variation in tissue mercury concentration or body burden of mercury have not been well examined. We noted that water temperature, YP condition index and mercury concentrations in LMB all had similar variance patterns. Bodaly et al. (1993) observed a correlation of fish tissue mercury concentrations with water temperature. Nimi (1983) recorded lower mercury concentrations in fish before spawning. The substantial natural physiological changes associated with the annual reproductive cycle in fish may result in apparent changes in tissue mercury concentration differences. The physiological condition of a fish may reflect either a toxicological effect of mercury, or some lake-specific fish growth characteristic, independent of a mercury effect. Fish in poor or robust condition may have altered mercury uptake and elimination kinetics. The lessened or greater amount of soft tissue per unit of body length, or altered levels of tissue hydration may result in variable tissue mercury concentrations even where the total mercury body burden remains the same.

Seasonal and interannual variation enter into the interpretation of the present data set because this and the previous Massachusetts study (Rose et al., 1999) were conducted in the spring and autumn respectively of different years. We have no estimate of the influence of seasonal or year-to-year differences on our measurements of fish tissue mercury concentrations because the literature is conflicting on this point. Bidwell and Heath (1993) recorded no changes in fish tissue mercury concentrations over the seasons. Francesconi et al. (1997) saw summer to winter differences in mercury concentrations of 4 out of 8 marine species that they studied in Australia. Park and Curtis (1997) saw little evidence of a seasonal pattern in LMB tissue mercury concentrations in two Oregon reservoirs.

To address this uncertainty in the role of seasonal factors in influencing fish tissue mercury concentrations, we designed a study to provide data on the magnitude of seasonal variation in LMB and YP tissue concentrations. This data set (mentioned above as the first data set in the long-term monitoring program) included pre- and post-spawn spring, summer, fall, winter and pre- and post- spawn spring sampling from 2001 through 2002 in 7 lakes. This data set is presently under analysis.

In the present study, species condition indices varied among lakes, but not in any explainable pattern. Condition indices and tissue mercury concentrations in the same species were not correlated (Figure 7). However, LMB mercury concentrations had a similar variance pattern to YP condition. The condition factors of fish within defined populations are integrated measures of the health status of that population. This variable may reflect food availability, interspecific competitive interactions for food, resources and habitat, or the presence of disease or pollution. The variation observed may have been due to short-term seasonally driven factors, since sampling took place over about a six-week period during or after spawning season when water temperatures were rapidly changing. The relatively high factor scores of YP condition index and water temperatures on the same factor in the factor analysis (Table 4) suggest that these two variables are correlated, supporting a conclusion that even within the narrow time frame of this study, YP were becoming more robust (higher condition index) as water temperatures rose.

Mercury concentrations did not vary between sexes, or with the reproductive state of either LMB or YP (Figure 8). This conclusion for the reproduction state of YP should be tempered by the fact that the sample sizes for the ripe females and males were only 2 and 3 respectively. In the limited reporting in the literature of fish tissue mercury concentrations differentiated by sex, no differences were reported between sexes for northern pike in Lake Champlain (Friedmann et al., 1996). That study had small sample sizes, thereby limiting the power of the study to detect differences, and did not control for fish size effects on mercury concentrations, thereby confounding any sex-related differences with size-related differences. The conclusions of the present study concerning tissue mercury and reproductive state of fish are preliminary because of insufficient numbers of fish for either species both before and after spawning. The large majority of LMB were in the prespawning state, and YP were in the postspawning state.

The information collected in this study on the water chemistry of the lakes did not identify any relationships between fish tissue mercury concentrations and lake chemical characteristics. Fish tissue mercury concentrations in the study lakes were also poorly predicted by a numerical model developed with a statewide data set to predict fish tissue mercury based on lake water chemistry variables. Our previous work on least-impacted Massachusetts lakes revealed that properties of individual lakes, such as pH, watershed and wetland areas, were more important for determining fish tissue mercury concentrations than were small-scale ecoregional differences (Rose et al., 1999). Most of the lakes sampled in this study from downwind or upwind areas had relatively similar, neutral or slightly basic pH values, while the pH values of the far upwind reference ponds were generally more acidic (pH<7) (Figure 6E). On the basis of the influence of pH alone, one would therefore not expect significant differences in fish tissue mercury between lakes in the downwind and upwind areas, yet might expect higher concentrations from far upwind because of the lower pH waters. This was not the case. There were significant differences in fish tissue mercury concentrations between lakes within depositional areas, yet no significant differences between lakes grouped by depositional areas. The LMB mercury concentrations in several of the lower pH, far upwind ponds were lower than those of most all other ponds (Figure 6E). The apparent lack of relationships between observed high LMB mercury tissue concentrations in the study area and lake pH may simply reflect higher mercury loads in ponds in the study area, or the influence of uncharacterized processes favoring the methylation of mercury in the lakes, or other as yet unidentified processes. To begin to address some of these issues we have conducted a modest study of mercury concentrations in the food chain and sediments in two closely situated lakes in the study area where previous sampling had indicated marked differences in fish mercury concentrations between the ponds. We also characterized conditions which would be associated with or reflect the degree of methylation in the two lakes ((MA DEP, 2003)).

This study confirmed a number of previously recognized generalities about fish and mercury. Longer-lived, more predacious fish such as LMB and chain pickerel (Table 3) accumulate higher concentrations of mercury than fish such as YP, BB, and YB. LMB mercury concentrations are positively correlated with the size of the fish. This study contributed insights into some of the relationships that are not yet fully understood: 1) high regional fish tissue mercury concentrations exist in the same region where high atmospheric mercury deposition is predicted to take place; 2) neither fish sex differences nor condition of the fish as measured by a morphometric-based condition index are confounders of tissue mercury concentrations in LMB and YP. This study also provided information which does not always match conclusions reached elsewhere: an absence of an apparent spatial relationship between the mercury concentrations in fish in the study lakes relative to the locations of those lakes vis-à-vis large point sources of atmospheric mercury emissions in the area. Over the last 22 years, this traditionally urbanized region where this study was conducted has had a concentration of 3 municipal solid waste combustors and 1 medical waste incinerator. These types of facilities are recognized as major potential contributors to mercury emissions in the US (U.S. Environmental Protection Agency, 1997) and Massachusetts (Smith and Rowan-West, 1996). The study did not provide additional insight about the potential for relationships between fish mercury concentrations and: 1) interannual and seasonal variation; 2) fish spawning period; and 3) chemistry of lake water and sediment (to the extent measured in this study for all three items). However, studies launched in the last two years will provide this information.

The major findings of this study are that fish tissue mercury concentrations in our study area appear to reflect model-predicted inputs of mercury within the regional airshed from atmospheric deposition, but do not reflect more localized inputs from what were considered major mercury emissions sources within the area. Better understanding of mercury bioaccumulation in fish in Massachusetts could be facilitated in the future by the availability of modelled mercury deposition estimates on a more refined areal grid, actual measurements of atmospheric mercury deposition, chronological histories of mercury deposition rates in the sedimentary record of lake sediment cores, direct measures of mercury methylation rates in the study lakes, measurement of organic matter content and concentrations of organic and inorganic mercury in surface waters.

SUMMARY AND CONCLUSIONS

The study goals were to:

- sample fish from lakes in northeast Massachusetts where the public has access to fishing to determine the need for fish consumption advisories;
- determine whether the frequency of advisories is greater in this area than across the state as a whole;
- establish a baseline for fish mercury concentrations in order to evaluate trends, thereby providing an environmental results-based indicator of the success of mercury source control efforts;
- compare mercury concentrations in fish from the region with those from other more rural parts of the State;

- determine if there are geographic differences in fish mercury concentrations within the study area related to the locations of the major point sources of mercury emissions vis-à-vis prevailing wind direction;
- determine whether predicted high atmospheric deposition rates of mercury for the area were mirrored by fish tissue mercury concentrations.
- evaluate the accuracy of a model, developed by MA DEP, to predict mercury levels in fish based on measures of water quality.

LMB and YP were the primary target species and brown bullhead, yellow bullhead and chain pickerel were secondary species. The study area in northeast Massachusetts was delineated into three regions based upon their locations with respect to a cluster of four major mercury point sources in the region (3 municipal waste combustors and 1 medical waste incinerator): far upwind, upwind, and downwind as inferred from predominant wind directions.

Of the 26 lakes studied, 23 warranted fish consumption advisories on the basis of mercury levels in LMB. Mercury levels in YP and BB were generally lower than those in LMB in about 65% of the lakes, which did not warrant advisories for those species. Two lakes lacked the target species for the study and thus were not issued a fish consumption advisory. Only one lake did not receive a fish consumption advisory, even though all target species were collected from the lake. In addition, we found that:

- Size-standardized LMB mercury concentrations in all lakes sampled in northeast Massachusetts were in the top quartile of size-standardized LMB tissue mercury concentrations from more rural lakes around the state (>0.49 mg/kg). The mean (± 1 standard deviation) LMB mercury muscle concentration was 0.89 ± 0.43 mg/kg (n=192).
- The LMB mercury concentrations in all ponds in the study area within the zone of predicted high atmospheric deposition of mercury warranted fish consumption advisories for mercury, a frequency greater than has been required after monitoring in more rural parts of the state.

- YP mercury concentrations were in the middle to high ranges (25^{th} – 75^{th} percentile and > 75^{th} percentile) of values for YP from more rural lakes in the state. The mean (±1 std. dev.) YP muscle mercury concentration was 0.44 + 0.21 mg/kg (n=152).

- Mercury in YP from 35% of the predicted high atmospheric deposition area study area lakes was above the threshold (>0.5 mg/kg) for issuing an advisory. This frequency of required advisories was just barely greater than that required from monitoring of this species from rural parts of the state.
- There was no obvious geographical pattern in the lake fish tissue mercury concentrations with respect to the upwind-downwind study design, or to the locations of the major point sources in relation to the lakes.
- LMB tissue mercury concentrations were correlated with water temperature and the condition of YP. There was also a positive relationship between mercury in LMB, that in YP and an inverse relationship of these two variables with LMB condition index (weight/total $length^3$).
- This study was designed to assess whether fish mercury concentrations were related to local major atmospheric mercury emission sources. This study was not designed to control for potentially confounding variables such as: seasonal and interannual differences in fish tissue mercury concentrations; reproductive state of the fish; in-lake mercury methylation rates; and mercury inputs to specific lakes. The influences of these and other variables remain to be examined in future work.

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Table 1. Study Design Details

Lake	Area*	Species	Individuals	Composites	Advisory Analysis	Geographic Analysis
Baldpate Pond, Boxford	D	LMB	X		X	X
		YP	X		X	X
		BB	X	X	X	
Chadwicks Pond, Haverhill	D	LMB	X		X	X
		YP	X		X	X
		CP	X		X	
		YB	X		X	
Johnsons Pond , Groveland	D	LMB	X		X	X
		YP	X		X	X
Lake Attitash, Amesbury	D	LMB	X		X	X
		YP	X		X	X
		BB		X	X	
Lake Cochichewick, N.Andover	D	LMB	X		X	X
		YP	X		X	X
Lake Pentucket, Haverhill	D	LMB	X		X	X
		BB		X	X	
Lake Saltonstall Haverhill	D	LMB	X		X	X
Millvale Reservoir, Haverhill	D	LMB	X		X	X
		BB		X	X	
Rock Pond, Georgetown	D	LMB	X		X	X
		YP	X		X	X
		BB		X	X	
Stevens Pond, N.Andover	D	LMB	X		X	X
		YP	X		X	X
		BB		X	X	
Kenoza Lake, Haverhill	D	LMB	X	X	X	
		YP	X		X	
		CP	X		X	
Hoveys Pond, Boxford	D	LMB	X		X	
		YP	X		X	
		CP	X		X	
Stevens Pond, Lawrence	D	BB		X	X	
Towne Pond, Boxford/N Andover	D	BB		X	X	
Ames Pond, Tewksbury	U	LMB	X		X	X
		BB		X	X	
Forest Lake, Methuen	U	LMB	X		X	X
		YP	X		X	X
Haggets Pond, Andover	U	LMB	X		X	X
		YP	X		X	X
Lowe Pond, Boxford	U	LMB	X		X	X
		YP	X		X	X
		BB		X	X	
Pomps Pond, Andover	U	LMB	X		X	X
		YP	X		X	X
		BB		X	X	
Crystal Lake, Haverhill	U	LMB		X	X	
		YP		X	X	
		CP		X	X	
Long Pond, Dracut	FU	LMB	X		X	X

Lake	Area*	Species	Individuals	Composites	Advisory Analysis	Geographic Analysis
		YP	X		X	X
		BB		X	X	
Lake Massapoag, Dunstable	FU	LMB	X		X	X
		YP	X		X	X
		BB		X	X	
Newfield Pond, Chelmsford	FU	LMB	X		X	X
		YP	X		X	X
		BB		X	X	
Bare Hill Pond, Harvard	FU	LMB	X		X	X
		YP	X		X	X
		BB		X	X	
Fort Pond, Lancaster	FU	LMB	X		X	X
		YP	X		X	X
		YB		X	X	
Hickory Hills Pond, Lunenburg	FU	LMB	X		X	X
		YP	X		X	X
		YB		X	X	

* D – downwind, U – upwind, FU – far upwind

Table 2. Predictive Equations for Mercury in Massachusetts Fish

Species	Model
Yellow Perch	[HG] = 2.883 + 0.001*[CA^{++}] – 0.046* TEMP
Largemouth Bass	[HG] = -0.227 + 0.0005*LMB MEAN WT
	– 0.367*YP MEAN [HG] – 0.004*YP MEAN WT

Source: MA DEP , 1997

Table 3. Advisory Screening Tissue Mercury Concentrations in Northeast Massachusetts Lakes.

Species	Depositional Area	Lake	Mean Hg (mg/kg)	s	n
LMB	Downwind	Kenoza Lake	1.80	---	Composite of 3
	Downwind	Hoveys Pond	0.88	0.14	3
	Upwind	Crystal Lake	1.04	0.20	3
YP	Downwind	Kenoza Lake	1.01	0.13	6
	Downwind	Hoveys Pond	0.57	---	1
	Upwind	Crystal Lake	0.33	---	1
CP	Downwind	Kenoza Lake	1.4	0.23	6
	Downwind	Hoveys Pond	0.95	---	1
	Upwind	Crystal Lake	0.56	---	1
	Downwind	Chadwicks Pond	1.1	0.14	3
YB	Downwind	Chadwicks Pond	1.1	.24	3
	Far upwind	Hickory Hills	0.47	---	Composite of 3
	Far upwind	Fort Pond	0.14	---	Composite of 3
BB	Downwind	Lake Attitash	0.22	---	Composite of 3
	Far upwind	Lake Massapoag	0.24	0.05	2
	Far upwind	Long Pond	0.52	---	Composite of 3
	Downwind	Stevens Pond, N.A.	0.16	---	Composite of 3
	Downwind	Lake Pentucket	0.73	---	Composite of 3
	Upwind	Pomps Pond	0.1	---	Composite of 3
	Downwind	Baldpate Pond	0.37	---	Composite of 3
	Downwind	Rock Pond	0.39	---	Composite of 3
	Far upwind	Newfield Pond	0.09	---	Composite of 3
	Far upwind	Bare Hill Pond	0.15	---	Composite of 3
	Upwind	Ames Pond	0.14	---	Composite of 3
	Upwind	Lowe Pond	0.21	---	Composite of 3
	Downwind	Stevens Pond, L.	0.35	---	Composite of 3
	Downwind	Millvale Res.	0.26	---	Composite of 2
	Downwind	Towne Pond	0.12	---	Composite of 5

KEY: LMB=Largemouth bass, YP =Yellow perch, CP=Chain pickerel, BB=Brown bullhead, YB=Yellow bullhead, comp=composite sample, s=standard deviation, n=number of individuals in sample, N.A.=North Andover, L.=Lawrence.

Table 4. Factor Scores for Environmental and Fish Condition Variables.

Variable	Factor 1	Factor 2	Factor 3	Factor 4
depositional area	-0.02	**0.86**	-0.17	-0.22
water temperature	**0.83**	-0.00	0.17	-0.13
dissolved oxygen	-0.45	-0.20	0.24	**0.68**
pH	0.17	0.13	0.02	**0.90**
conductivity	-0.10	**0.83**	0.03	0.25
Hg conc. in LMB	**0.60**	0.07	**0.65**	0.21
Hg conc. in YP	0.01	-0.18	**0.90**	0.08
LMB W/L^3	0.17	-0.47	-0.52	0.45
YP W/L^3	**0.80**	-0.22	-0.14	0.13
% explained variation	21.7	19.9	18.3	18.5

Table 5. Results of Environmental Gradient Studies from Point Atmospheric Mercury Sources

Environmental Component	Gradient Observed	No Gradient Observed	Reference
Atmosphere:			
Hg^o concentration	?		(Iverfeldt 1991)
Hg deposition	?		(Nater and Grigal 1992)), (Dvonch; Vette; Keeler; Evans, and Stevens 1995), (Keeler and Hoyer 1997), (Mason; Lawson, and Sullivan 1997)
Soils:		?	(Meneses; Llobet; Granero; Schuhmacher, and Domingo 1999;Schuhmacher M.; Granero; Belles; Llobet, and Domingo 1996)
	?		(Johansson; Aastrup; Andersson; Bringmark, and Iverfeldt 1991), (Nater and Grigal 1992)
Terrestrial Vegetation:	?		(Steinnes and Andersson 1991)
lichens	?		(Kurttio; Pekkanen; Alfthan; Paunio; Jaakkola, and Heinonen 1998)
wild berries, mushrooms		?	(Schuhmacher M. and others 1996), (Kurttio and others 1998), (Meneses and others 1999)
Birds:			
Anhingas, white ibis eggs &		?	(Rumbold; Bruner; Mihalik, and Marti

nestlings			1997)
Aquatic Environments:			
sediments	?		(Engstrom and Swain 1997), (Swain 2000)
fish		?	Pinkney et al. (1997)

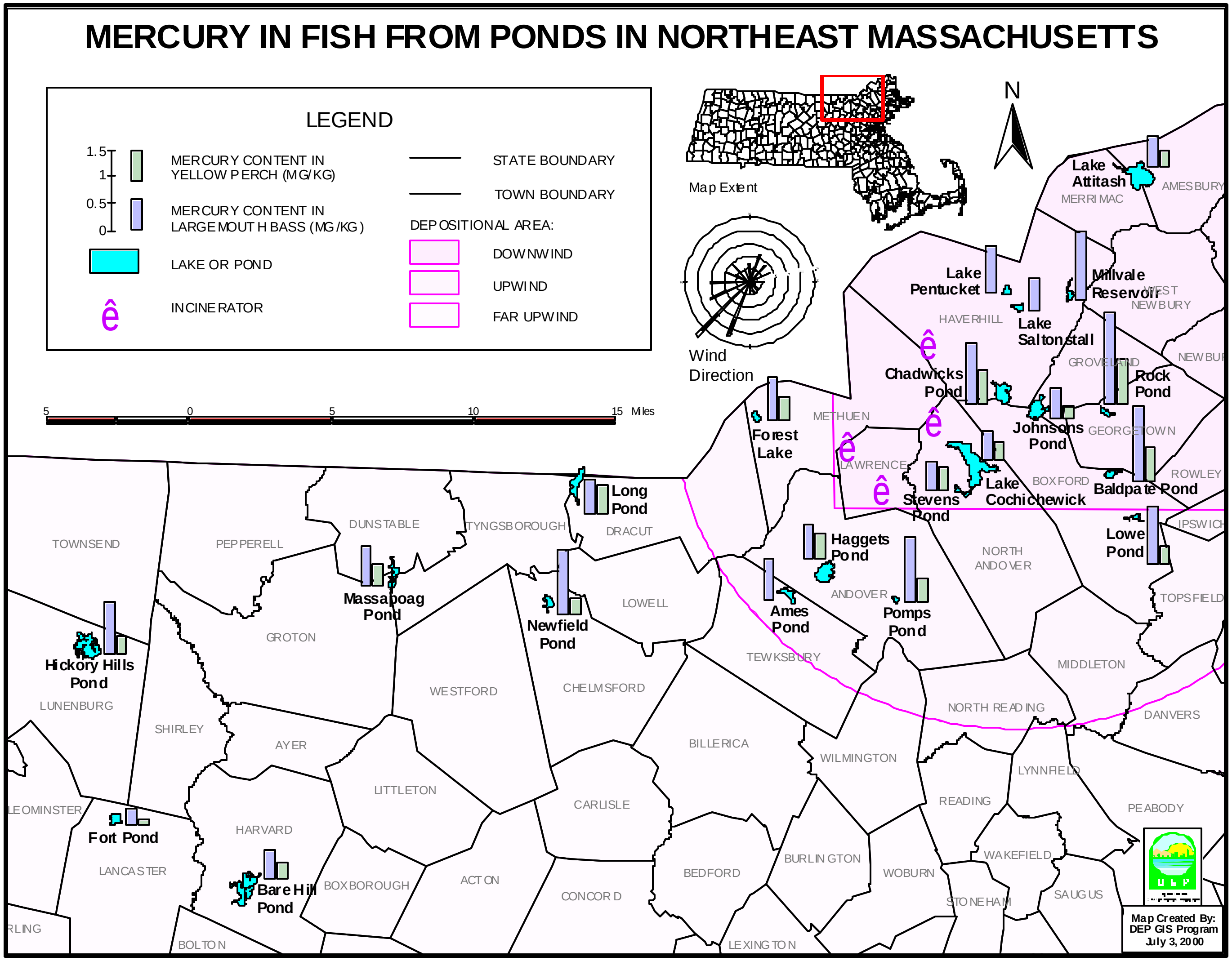


Figure 1. Mean Mercury Concentrations for Target-Sized LMB and All YP in Northeast Massachusetts Study Lakes. Potential Mercury Depositional Areas Noted in Relation to Point Source Locations

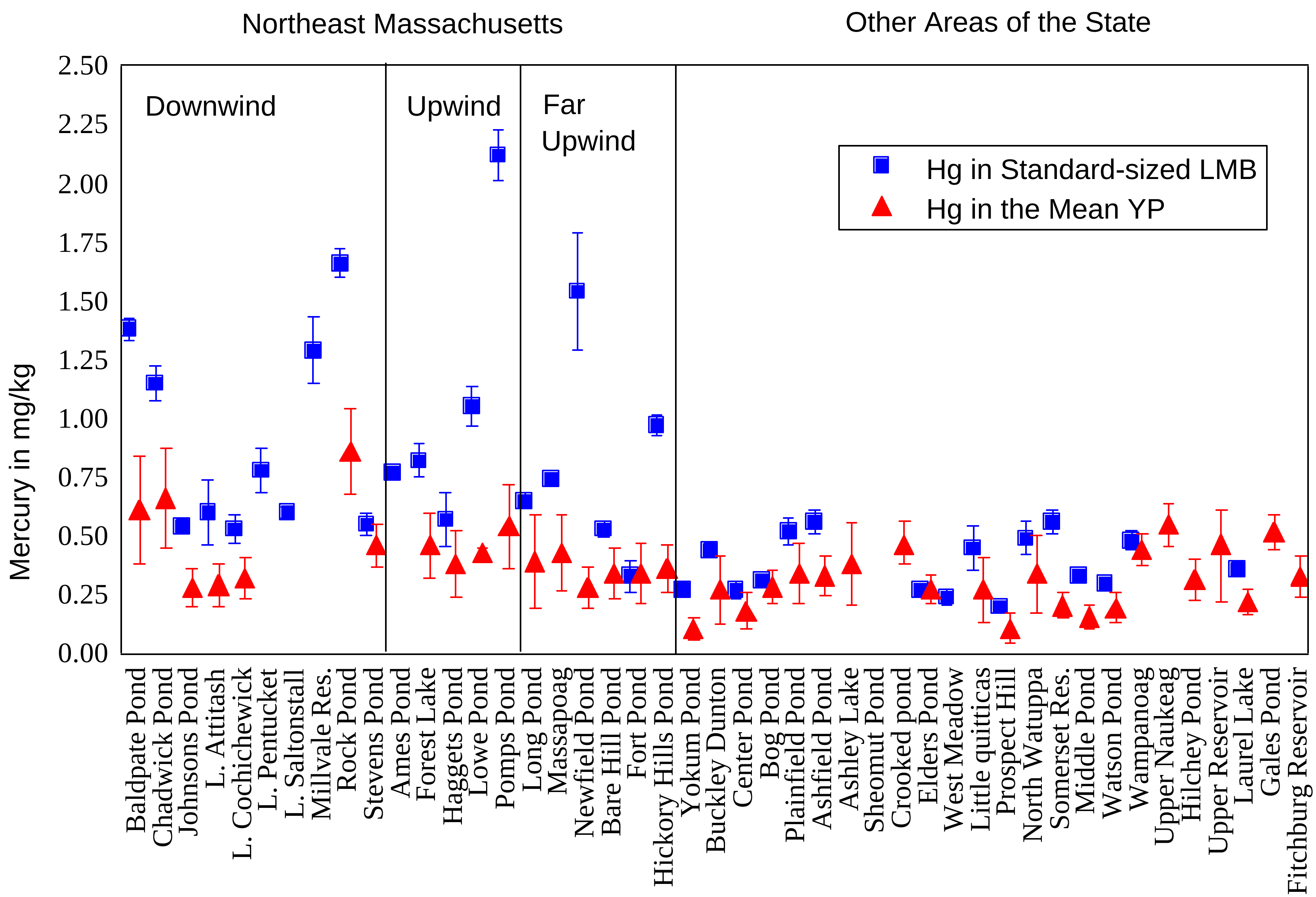
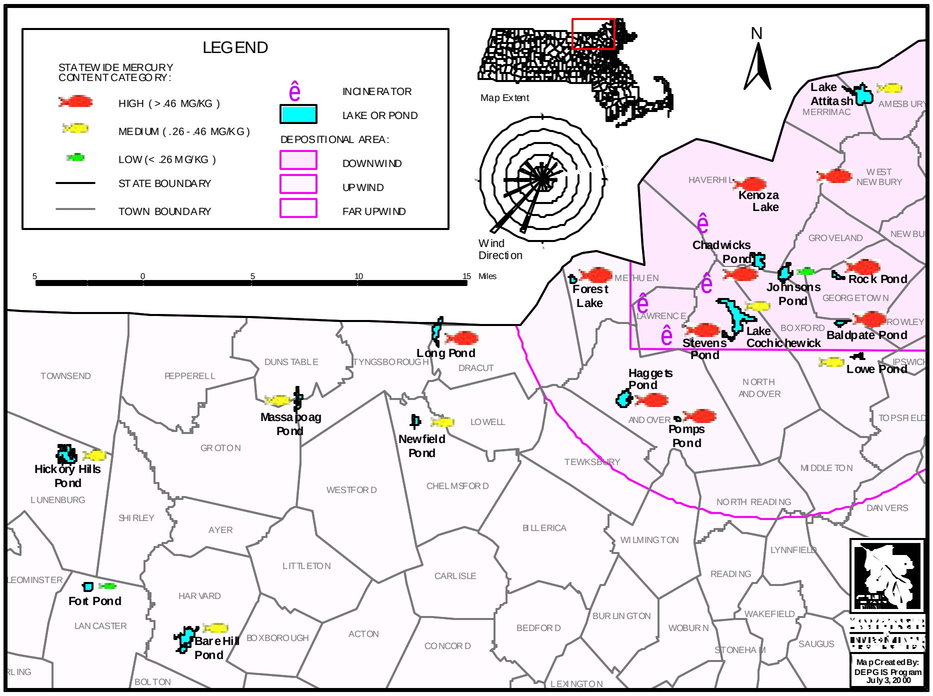


FIGURE 2. FISH SPECIES MERCURY CONCENTRATIONS (±1 STANDARD DEVIATION) BY LOCATION.

A.



B

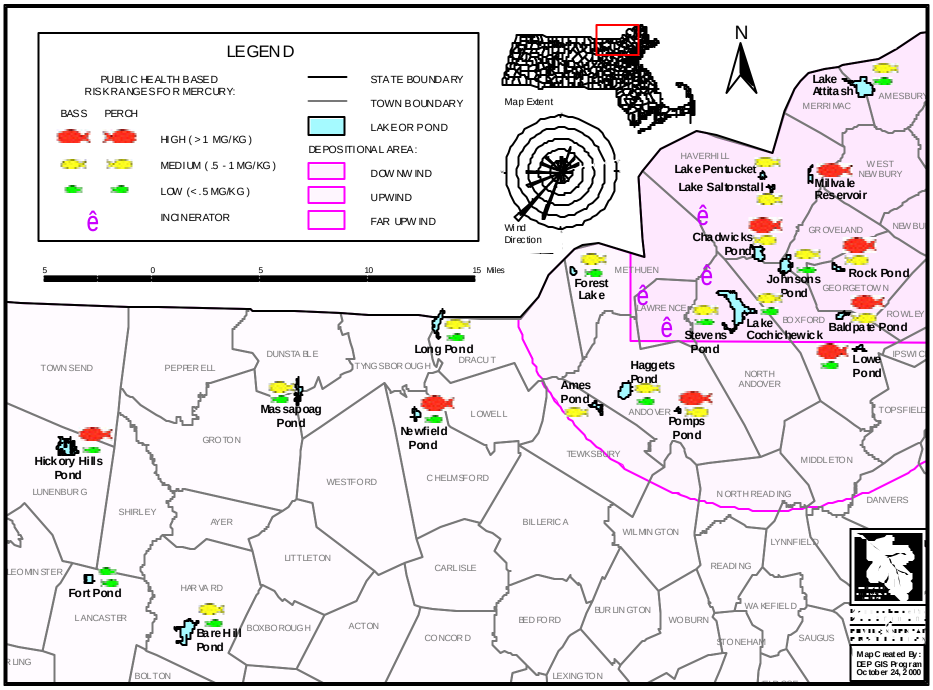


Figure 3. Fish Mercury Concentrations in Northeast Massachusetts. A) Based on Public Health Risk Criteria; B) YP Values Compared to 1997 Study Values (Rose et al., 1999).

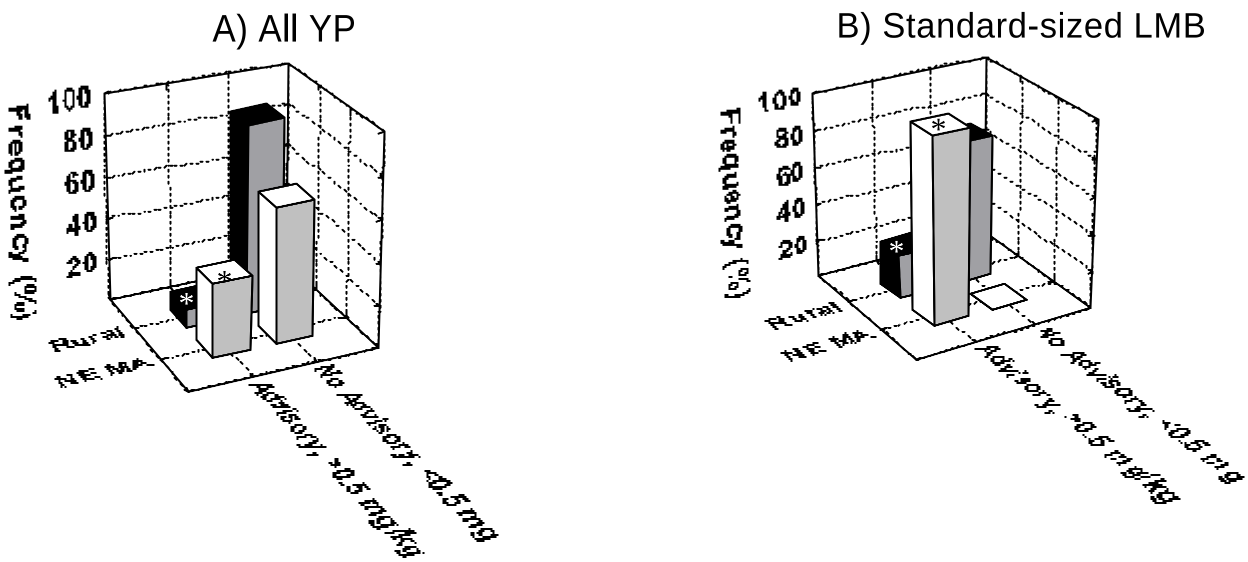


Figure 4. Frequencies of Fish Consumption Advisories Based on Fish Tissue Mercury Concentrations (>0.5 mg/kg). Northeast Massachusetts Versus Rural Areas. A) all YP; B) standard-sized LMB. * NE Massachusetts advisory frequency significantly greater than Rural Areas, p=0.05.

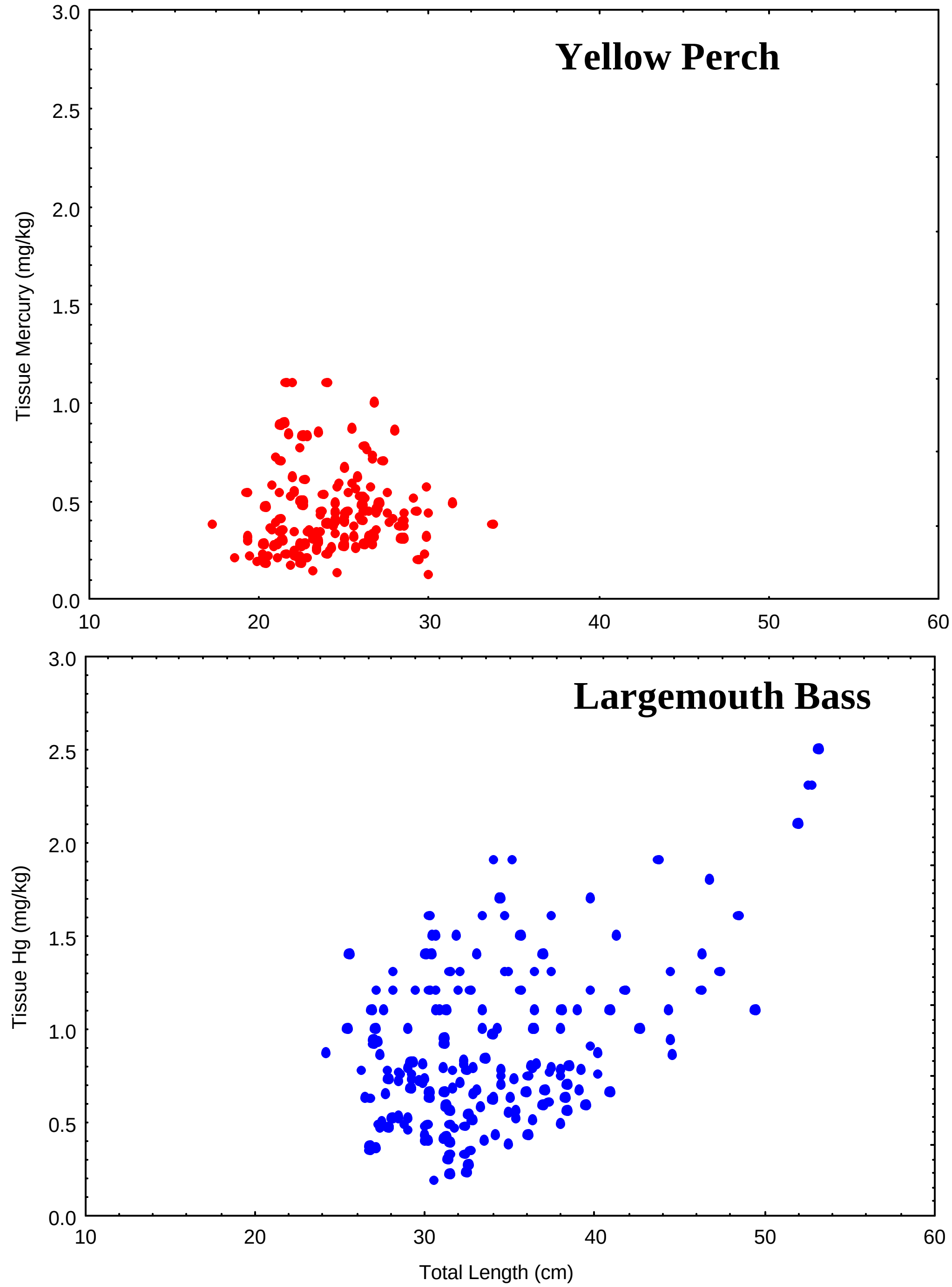


Figure 5. Total Fish Mercury Concentrations Versus Total Fish Length

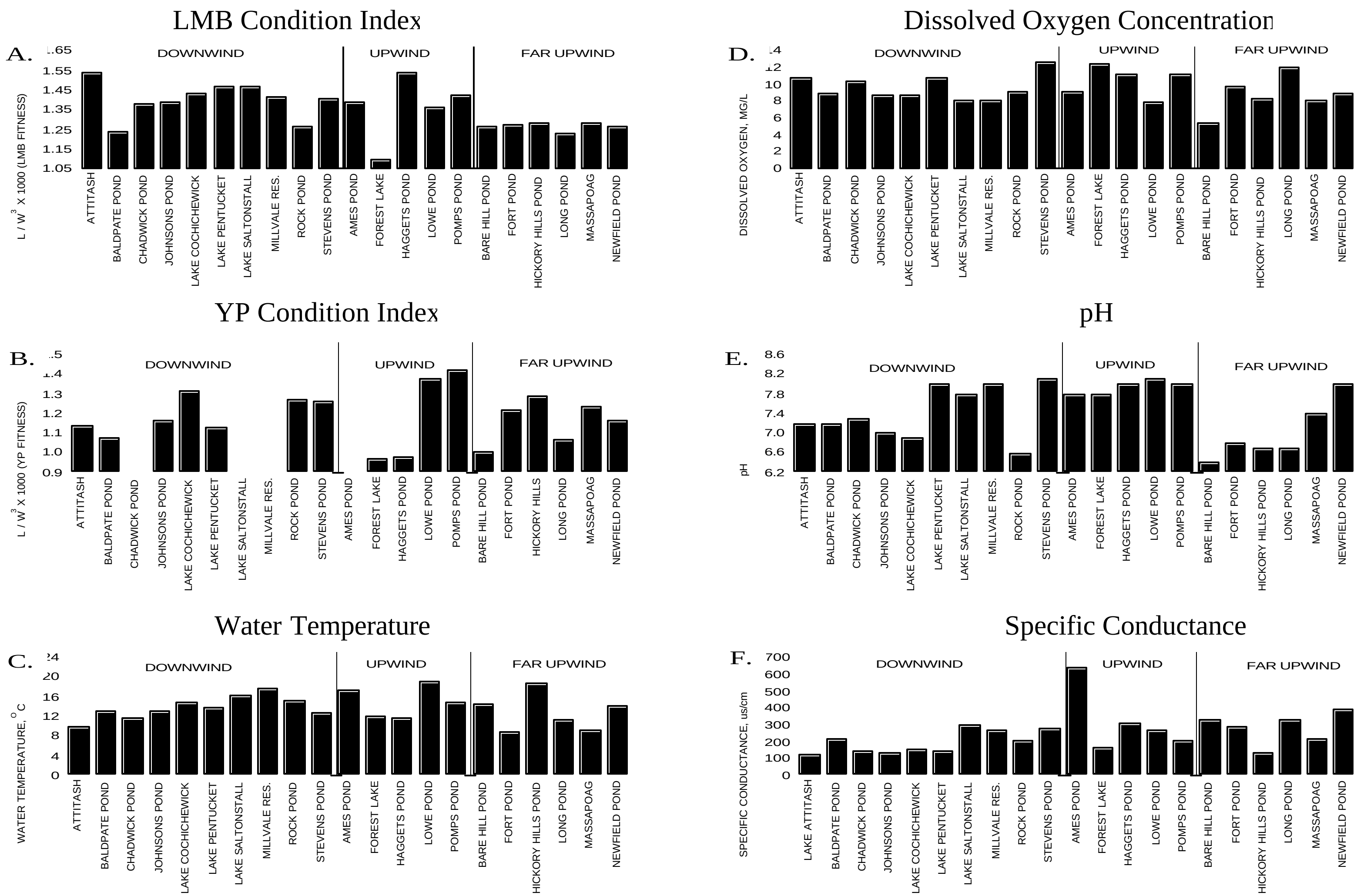


Figure 6. Study Variables Plotted by Study Area

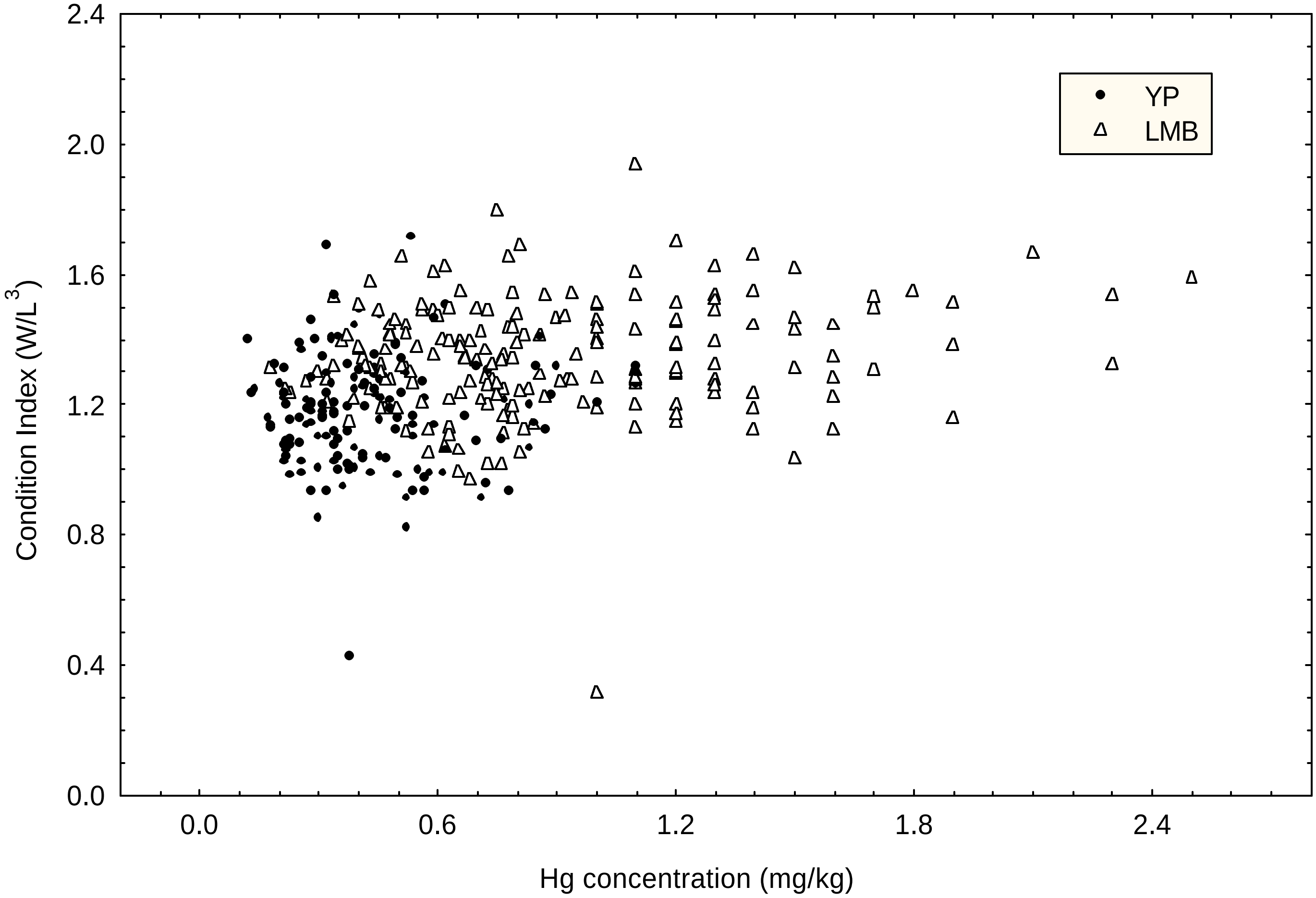


Figure 7. Fish Condition Index (weight/length^3) Versus Fish Muscle Mercury Concentration

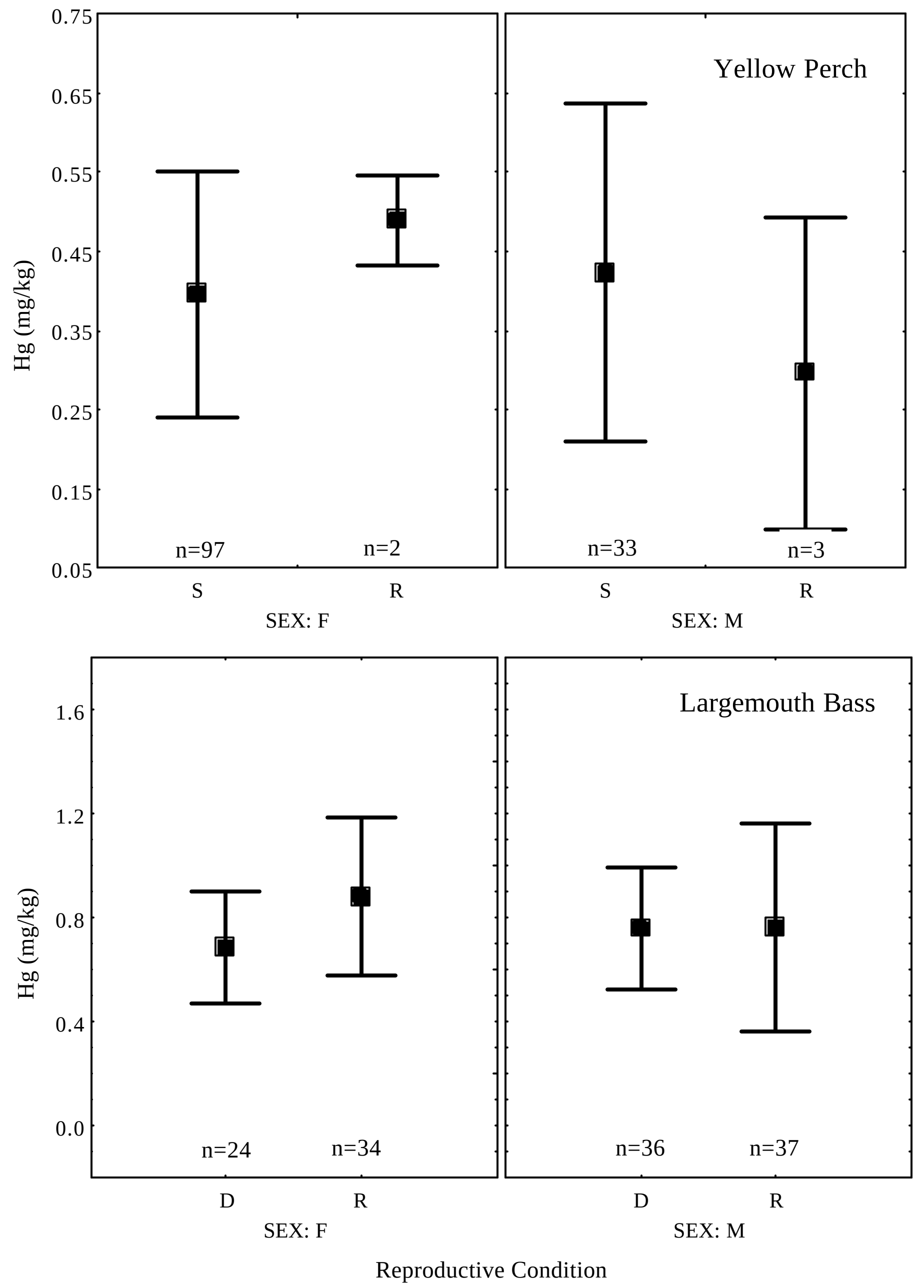


Figure 8. Mean Species Mercury Concentrations (means ± 1 std. dev.) Stratified by Sex and Reproductive Condition Based on All YP and 27-38 cm Target Range LMB. D-developing, R-ripe, S-spent gonads.

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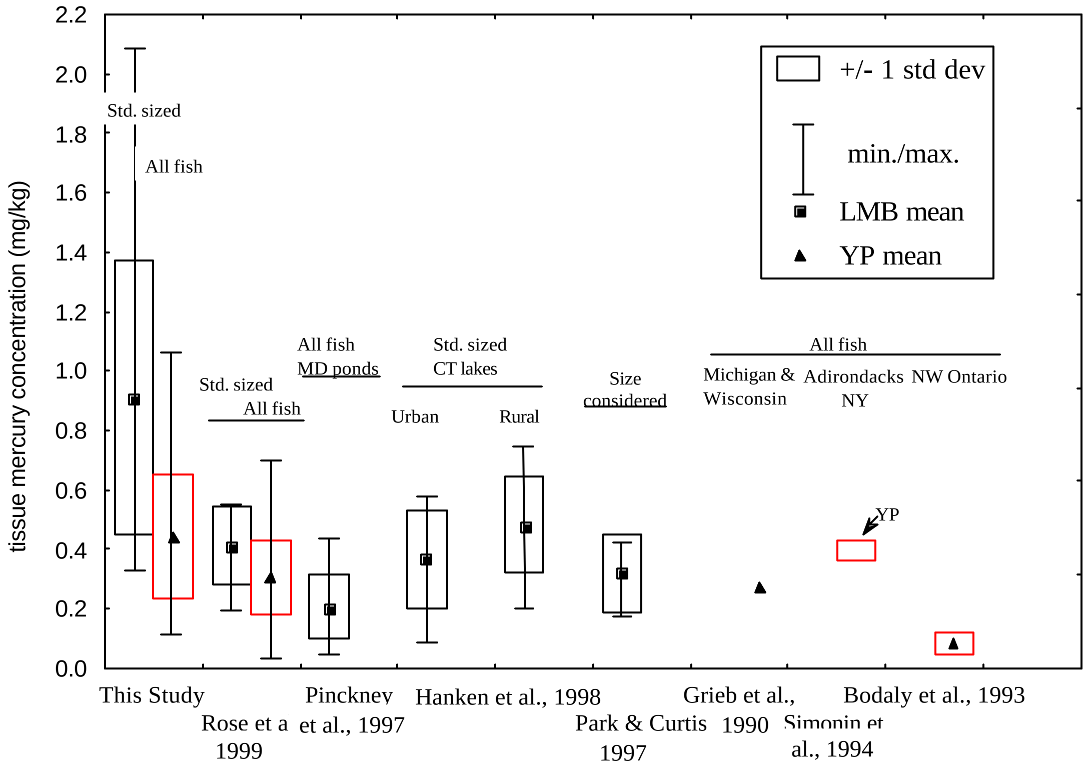

Figure 9. Comparative LMB and YP Muscle Mercury Concentrations. Means ±1 standard deviation, ranges.

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APPENDIX A

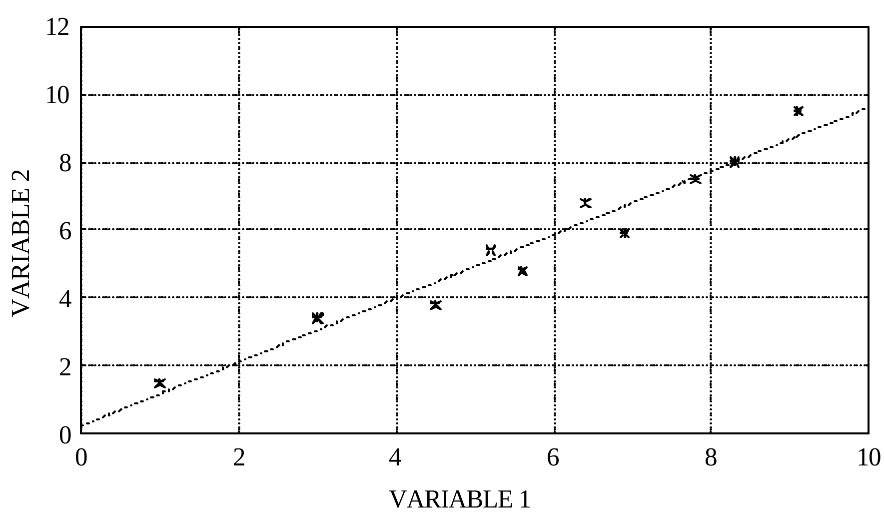
Factor Analysis Description

FACTOR ANALYSIS DESCRIPTION

The term “Factor analysis” (FA) actually represents a collection of mathematical techniques that can be used with sets of variables to detect underlying patterns of relationships among the variables or to reduce the size of the data set. Because FA is applied to sets of variables, it is referred to as a multivariate procedure. This brief discussion of Factor Analysis refers to classical Factor Analysis. The reader should consult more detailed statistics texts for discussion of other types of FA which are available.

In order to explain the basis for FA, it is useful to return to the simple correlation concepts used for individual pairs of variables. A regression line on an x-y plot between two variables represents the best summary of the linear relationship between the two variables (Figure A1).

Figure A1. BIVARIATE X-Y PLOT AND LINEAR REGRESSION



If a new variable could be defined that would approximate the regression line of the plot, then it would capture the essence of the correlation between the two variables. Two variables would be reduced to one factor. When interrelationships between more than two variables have to be discerned in data sets, new correlations or factors for each pair can be developed. This sequential, bivariate approach for looking at all possible pairs of variables quickly outstrips our ability to conceptually link all the interrelationships and discern any underlying patterns of variance relationships in the data. FA is a statistical technique which moves beyond the limitations of the bivariate approach, and which provides a means for identifying intervariable correlation structures among numerous variables by extending the basic idea of the derived factor for a two variable relationship to multiple variables.

FA reduces the size of a data set of variables to a new set of fewer variables called factors. The factors are constructed to be independent of each other in terms of correlations and to represent those original variables in the data set which are most highly correlated in their patterns of variance. For example, a complex data set of variables from an ecology study might includes variables such as a species density in a particular habitat (Clayton; Perritt; Pellizzari; Thomas; Whitmore; Wallace; Ozkaynak, and Spengler 1993), the density of its prey (Zhou and Weis 1998), the mean annual air temperature (ATEMP), the density of a particular plant species (PLANT), the median grain size of the soil (GRAIN), and the water content of the soil (SH2O). The researcher finds this number of variables too many to interpret when all possible intercorrelations are considered (Table A1) and wonders if any of these variables have similar patterns of variance which would indicate some commonality in the processes which link those groups of intercorrelated variables.

Table A 1. Sample Correlation Matrix for All Variables

	SPDEN	PREY	PLANT	ATEMP	GRAIN	SH2O
SPDEN	**1.00**	0.65	0.65	0.14	0.15	0.14
PREY	0.65	**1.00**	0.73	0.14	0.18	0.24
PLANT	0.65	0.73	**1.00**	0.16	0.24	0.25
ATEMP	0.14	0.14	0.16	**1.00**	0.66	0.59
GRAIN	0.15	0.18	0.24	0.66	**1.00**	0.73
SH2O	0.14	0.24	0.25	0.59	0.73	**1.00**

A FA on the data set eventually extracts or derives two factors (Table A2). Factor 1 is composed of the variance in both the species density and its prey density and density of vegetation . The analysis indicates that these three have similar variance patterns and even can convey whether they are positively or inversely related. In this example, they are all positively related as indicated by positive values in the table. The second factor identified could result from the similar variance patterns in mean annual temperature, soil grain size and water content. This relationship might make intuitive sense from our understanding of ecology, but in other types of data sets, the underlying relationships between variables may not be known and the objective of the analysis would be to identify these patterns and perhaps fortuitously reduce the complexity of the data set.

The sequence of steps in a FA, some of which were omitted for simplification in the description in the previous paragraph, is illustrated by the sample data set just discussed:

1) preparation of a matrix of correlation coefficients between all variables in the data set (e.g., Table A1);

Table A2. Factor Loadings on Rotated Axes

Variable	FACTOR 1	FACTOR 2
SPDEN	**0.862**	0.052
PREY	**0.890**	0.110
PLANT	**0.886**	0.153
ATEMP	0.062	**0.846**
GRAIN	0.107	**0.903**
SH2O	0.141	**0.870**
Variance Total	2.375	2.326
Proportion of Total	0.393	0.388

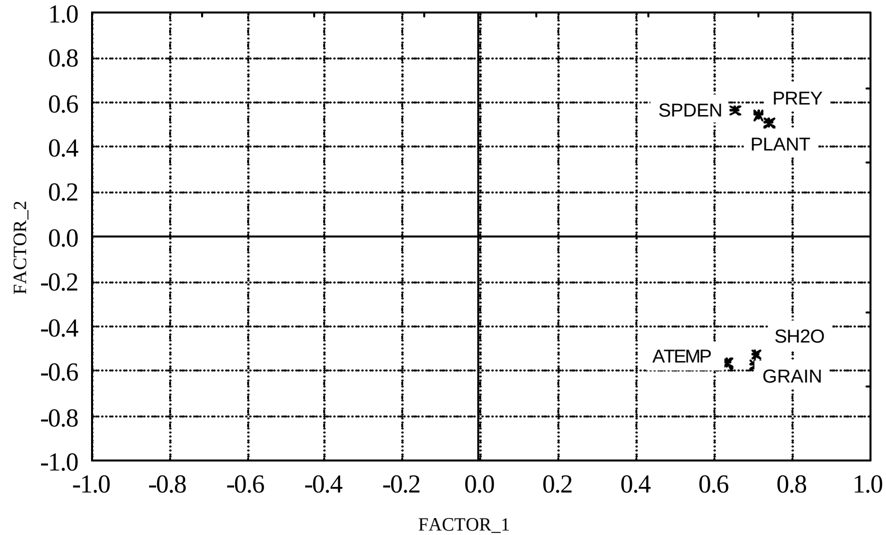
2) extraction of an initial set of factors on the basis of interrelationships exhibited in the data. Each variable will have a varying correlation with each factor referred to as its factor “score” or “loading” (Table A3).

Table A3. Factor Loadings on Unrotated Axes

Variable	Factor 1	Factor 2
SPDEN	0.654	0.564
PREY	0.715	0.541
PLANT	0.742	0.508
ATEMP	0.634	-0.563
GRAIN	0.706	-0.573
SH2O	0.708	-0.526
Variance Total	2.89	1.79
Proportion of Total	**0.48**	**0.299**

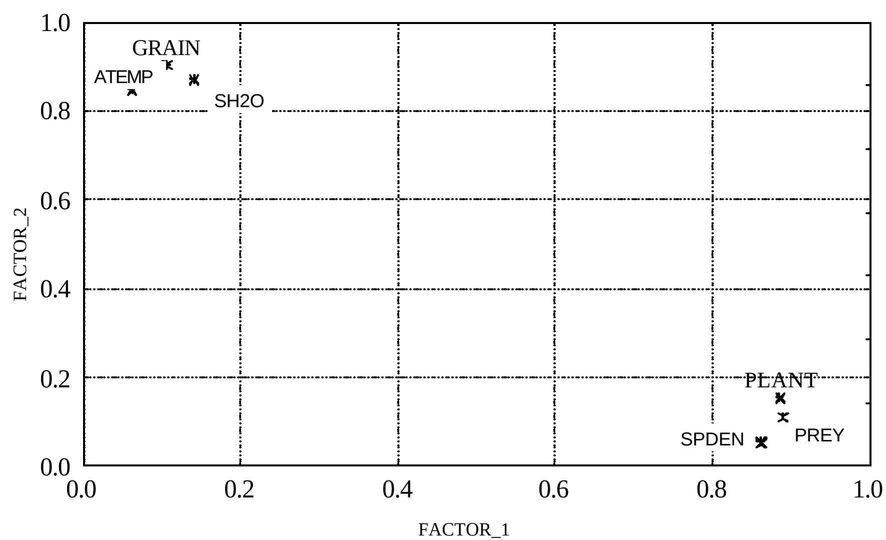
This extraction process is performed so that the factors are independent of (uncorrelated with or orthogonal to) each other. The first extracted factor accounts for the largest possible amount of variance in the data set. Each additional factor extracted accounts sequentially for the largest possible amount of remaining variation independent of the previously derived factors. Note that at this stage of the analysis, each variable may have a relatively high score on both factors. In addition, the proportion of total variance explained by each factor is given at the bottom of each Factor column. These relationships can be graphically represented by bivariate plots of the correlation scores of each original variable on each pair of factors (Figure A2 for Factor 1 versus Factor 2).

Figure A2. Factor 1 and 2 Loadings on Unrotated Axes



3) rotation in n-dimensional space of the axes for each pair of Factors about the points, while their relative positions are maintained, so as to achieve a simpler and more meaningful factor pattern. Such a pattern is one where the correlations for one set of intercorrelated original variables clearly have high correlations for one factor and low correlations on the other factors (Figure A3).

Figure A3. Rotated Factor Loadings on Factors 1 and 2.



The final product is a rotated factor matrix (Table A2) containing values for each variable which are both regression weights and correlation coefficients versus the inferred factor. These loadings represent the regression coefficients of factors to describe a given variable. For example, the equation to describe a specific variable in terms of the new factors could be:

SPDEN = 0.862 * Factor 1 + 0.052 * Factor 2

In common with regression analysis, the independent variables (i.e., the hypothetical factors) are said to control or account for a certain percentage of the variance in the dependent variables. The variance of SPDEN due to Factor 1 is the square of the factor score contained in the factor matrix. The total variance in a variable accounted for by all the factors is given by the sum of squares of the respective factor loadings.

It is also possible to determine the importance of a given factor in terms of the amount of total variance in the data set that it accounts for. This is accomplished by squaring each factor score, summing down in the table across variables, and dividing the total by the number of variables in the data set. For example, in the final solution, Factor 1 accounts for 39.3% of the total variance in the data set on the rotated axes (Table A2). Since the variables SPDEN, PREY, and PLANT have the highest factor scores, the are responsible for the majority of the variance in Factor 1 and have common patterns of variance themselves.

APPENDIX B

Table B1. Central Tendency Tissue Mercury Concentrations Estimates* (mg/kg) and Physical Data Summary

Sp.	Area	Lake	All Fish $\overline{x}_{Hg}$	All Fish s	All Fish n	Standard (A) Hg	Target –Size Fish (B) $\overline{x}_{Hg}$	Target –Size Fish (B) s	Target –Size Fish (B) n	%? A? B
LMB	Down-wind	Baldpate Pond, Boxford	1.33	0.16	9	1.38	1.33	0.16	9	3
		Chadwicks Pond, Haverhill	1.17	0.29	12	1.15	1.14	0.16	8	1
		Johnsons Pond, Groveland	0.61	0.15	9	0.54	0.57	0.15	7	6
		Lake Attitash, Amesbury	1.01	0.25	9	0.60	0.63	---	1	5
		Lake Cochichewick, N.Andover	0.58	0.19	9	0.53	0.53	0.11	8	0
		Lake Pentucket, Haverhill	1.30	0.76	10	0.78	0.81	0.38	6	4
		Lake Saltonstall Haverhill	0.51	0.19	9	0.60	0.47	0.04	7	22
		Millvale Reservoir, Haverhill	1.12	0.18	9	1.29	1.20	0.14	4	7
		Rock pond, Georgetown	1.63	0.21	9	1.66	1.63	0.21	9	2
		Stevens Pond, N.Andover	0.61	0.17	9	0.55	0.53	0.08	3	4
	Upwind	Ames Pond, Tewksbury	0.73	0.26	10	0.77	0.79	0.01	2	3
		Forest Lake, Methuen	0.71	0.07	9	0.82	0.73	0.07	7	2
		Haggets Pond, Andover	0.89	0.54	8	0.57	0.55	0.16	3	4
		Lowe Pond, Boxford	1.11	0.28	9	1.05	0.97	0.16	6	8
		Pomps Pond, Andover	1.32	0.50	9	1.14	1.10	0.31	6	48
	Far Upwind	Long Pond, Dracut	0.65	0.11	9	0.65	0.63	0.10	8	3
		Massapoag Pond, Dunstable	0.78	0.08	9	0.74	0.79	0.06	6	7
		Newfield Pond, Chelmsford	0.66	0.10	9	1.54	0.66	0.10	9	44
		Bare Hill Pond, Harvard	0.55	0.13	9	0.53	0.55	0.13	9	4
		Fort Pond, Lancaster	0.29	0.07	9	0.33	0.29	0.07	9	12
		Hickory Hills Pond, Lunenburg	0.95	0.19	9	0.97	0.95	0.04	7	1

Table B1 continued.

Sp.	Area	Lake	All Fish			Standard (A)	Hg Water Quality Parameters			
			$\bar{x}_{Hg}$	s	n		T	DO	pH	Cond.
YP	Down-wind	Baldpate Pond	0.61	0.23	9	0.64	13.1	8.9	7.2	220
		Chadwicks Pond	0.66	0.21	9	0.67	12	10.5	7.3	150
		Johnsons Pond	0.28	0.08	10	0.26	13.1	8.7	7.0	138
		Lake Attitash	0.29	0.09	9	0.32	10.2	10.8	7.2	128
		L. Cochichewick	0.32	0.09	9	0.32	15.0	8.8	6.9	156
	Down-wind	Lake Pentucket	---	---	0	---	14.1	10.9	8.0	152
		Lake Saltonstall	---	---	0	---	16.3	8.2	7.8	301
		Millvale Reservoir	---	---	0	---	17.8	8.1	8.0	274