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FISH MERCURY DISTRIBUTION IN MASSACHUSETTS, USA LAKES

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Abstract—The sediment, water, and three species of fish from 24 of Massachusetts' (relatively) least-impacted water bodies were sampled to determine the patterns of variation in edible tissue mercury concentrations and the relationships of these patterns to characteristics of the water, sediment, and water bodies (lake, wetland, and watershed areas). Sampling was apportioned among three different ecological subregions and among lakes of differing trophic status. We sought to partition the variance to discover if these broadly defined concepts are suitable predictors of mercury levels in fish. Average muscle mercury concentrations were 0.15 mg/kg wet weight in the bottom-feeding brown bullheads (*Ameiurus nebulosus*) (range = 0.01–0.79 mg/kg); 0.31 mg/kg in the omnivorous yellow perch (*Perca flavescens*) (range = 0.01–0.75 mg/kg); and 0.39 mg/kg in the predaceous largemouth bass (*Micropterus salmoides*) (range = 0.05–1.1 mg/kg). Statistically significant differences in fish mercury concentrations between ecological subregions in Massachusetts, USA, existed only in yellow perch. The productivity level of the lakes (as deduced from Carlson's Trophic Status Index) was not a strong predictor of tissue mercury concentrations in any species. pH was a highly (inversely) correlated environmental variable with yellow perch and brown bullhead tissue mercury. Largemouth bass tissue mercury concentrations were most highly correlated with the weight of the fish (+), lake size (+), and source area sizes (+). Properties of individual lakes appear more important for determining fish tissue mercury concentrations than do small-scale ecoregional differences. Species that show major mercury variation with size or trophic level may not be good choices for use in evaluating the importance of environmental variables.

Keywords—Mercury Fish Perch Bullhead Bass

INTRODUCTION

During the past 10 years, a growing awareness of the problem of high mercury concentrations in freshwater fish has generated a proliferation of studies at the international [1–3], national [4,5], and state [6,7] levels.

Massachusetts has surveyed contaminants in freshwater fish since 1983 [8], focusing primarily on areas of known or suspected contamination or on areas where biological effects were observed. These studies have shown that the variation in fish mercury contamination is relatively high in surface waters. Concentrations have been sufficiently high in some species to warrant the issuance of Fish Consumption Advisories for specific water bodies as well as a statewide health advisory cautioning pregnant women of the possible health risk associated with eating fish from Massachusetts freshwater bodies (excluding stocked and farm-raised fish).

Many factors contribute to the dynamics of contaminant accumulation in fish populations. An ecoregional approach partially explained geographic variation in fish mercury concentrations [9]. Lake productivity and lake trophic status affect the accumulation of persistent pollutants in fish [10]. The complexity of the definitions of ecoregion and lake trophic status makes these concepts potentially apt descriptors for ecosystems, which are inherently complex systems.

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Two ecoregions and 13 ecological subregions have been delineated in Massachusetts [11]. Shared components of ecoregions included soils, vegetation, climate, geology, and physiography. Patterns of animal migration and land use were also used to delineate ecoregions. Lakes in Massachusetts are either glacial (~10,000 years old) or they date back to the last mountain-building episode, roughly 200 million years ago. Most lakes were altered in colonial times to increase their utility to industrious New Englanders. The ecoregion concept may prove to be an effective tool for statistical analysis, research, and assessment of environmental resources, because it characterizes relatively homogeneous geographic regions, incorporating more information than do individual physical or chemical measurements.

In this study, yellow perch (*Perca flavescens*), largemouth bass (*Micropterus salmoides*), and brown bullheads (*Ameiurus nebulosus*) were sampled for muscle mercury concentration determinations in 24 lakes not likely to have been affected by nonpoint sources (e.g., landfills, industrial facilities, hazardous waste sites, wastewater treatment facilities). We also attempted to determine the relative degrees of influence on these concentrations of geographic location as well as lake biological, physical, and chemical characteristics.

MATERIALS AND METHODS

Lake selection

The lakes chosen for sampling were identified on the basis of the region of the state in which they were located and the

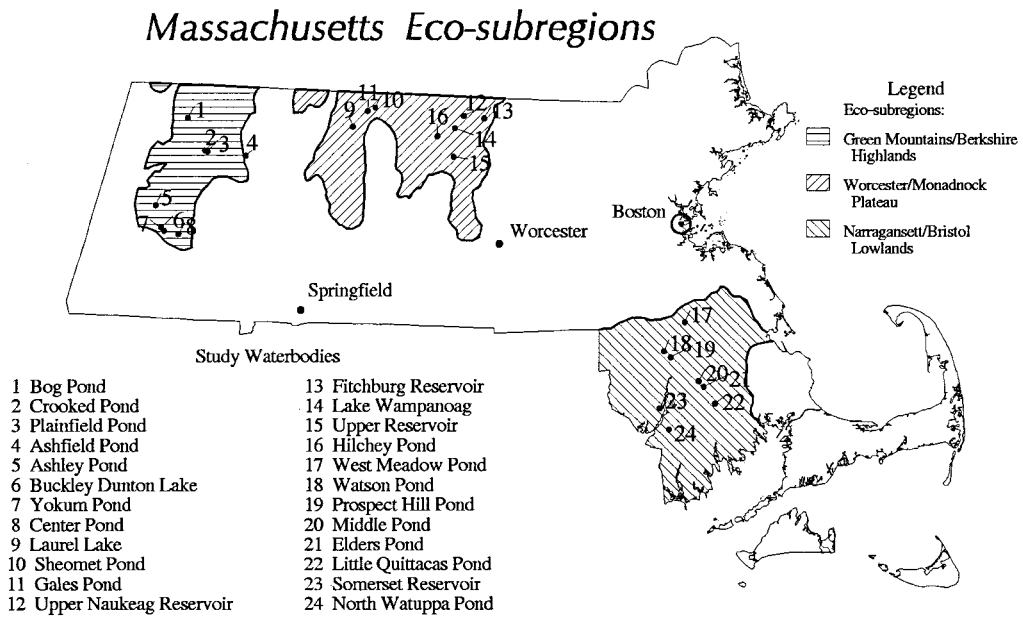

Fig. 1. Subecoregions of Massachusetts and study lake locations.

degree of development on or near the lakes. Eight lakes within each of three ecological subregions [11], representing contrasting environmental settings in Massachusetts, were selected (Fig. 1).

The Green Mountain/Berkshire Highlands subregion, located in northwestern Massachusetts, is characterized by relatively high elevations, which reach approximately 305 to 762 m above mean sea level. Metamorphic geology composed of schists, gneiss, and marbles creates a steep terrain that is overlaid by thin deposits of glacial till. Forest types include northern hardwoods (maple, beech, birch), spruce, and fir. Surface waters are generally low in phosphorus, with alkalinity under 200 μg/L [11].

The Worcester/Monadnock Plateau is located in the north-central part of the state at 152 to 457 m above sea level. The monadnocks are formed of granite plutons that dominate the surrounding geology of metamorphic schists and gneiss. Forest types include transition hardwoods (maple, beech, birch; oak, hickory) and some northern hardwoods. Surface waters are poorly buffered and acidic, with alkalinities generally between 50 and 100 μg/L [11]. Some surface waters exhibit moderate to high concentrations of dissolved organic compounds.

The Narragansett/Bristol Lowland is located in the southeastern part of the state. The landscape of this region consists of flat to rolling plains that seldom exceed 61 m above mean sea level, with numerous wetlands and bogs. Extensive deposits of glacial till and outwash material make up the soils and sediments. Central hardwoods (oak, hickory) are common, as are elm, ash, red maple, cottonwood, white pine, and red pine. Phosphorus in surface waters ranges widely, and alkalinities are in the 50 to 400 μg/L range [11].

The suitability of each lake identified in each ecosubregion on U.S. Geological Survey (USGS), 7.5′ series topographical maps was assessed using the following exclusion criteria in order to identify 24 lakes for study: surface area less than four hectares; proximity to concentrated urban, agricultural, or industrial areas; evidence of impact from human activities based on prior studies [12,13]; potential point or nonpoint sources of pollution.

Lake watershed areas were delineated based on USGS topographic quadrangles. Wetlands within the watersheds were delineated from U.S. Fish and Wildlife Service National Wetlands Inventory maps (1:24,000) and from stereoscopic analysis of high-altitude aerial photographs. Lake areas were calculated from digitized 1:25,000 coverages or from USGS topographic quadrangles.

Fish, water, and sediment sampling

The test species were selected principally because they encompass a range of fish trophic levels. Largemouth bass are fish-eating predators, although their diet also includes invertebrates and amphibians. The species did not occur in all of the study lakes. Yellow perch are omnivorous, consuming insects, invertebrates, and other fish, and brown bullhead are bottom-feeding omnivores [14].

Nine individuals of each species were targeted for collection from each lake. Fish sampling was conducted in the early fall after summer spawning. Total length criteria of 20 to 25 cm for yellow perch and brown bullhead and 30 to 36 cm for largemouth bass were established. The larger size was selected for largemouth bass because 30.5 cm is the legal minimum size limit for this species and may be representative of fish retained for consumption. Fish obtained by electrofishing, gill netting, and trot lines were rinsed in ambient water, chilled on ice, wrapped individually in aluminum foil, placed inside polyethylene zip-lock bags, and delivered to the laboratory on ice within 24 h of collection.

Water-quality sampling was conducted during midsummer, not coincident with fish sampling but during the period when lakes would be thermally stratified and when measures of degree of eutrophy might be strongest. In stratified lakes, a composite sample of water taken from the deepest part of the lake at 1.5 m below the surface, taken at mid-thermocline, and taken at 1.5 m above the bottom was prepared. The composite was then divided into three precleaned glass containers for chemical analyses. Single samples were taken from mixed lakes (non–thermally stratified) at 1.5 m below the surface. All water-quality sampling and handling was performed in accor-

dance with U.S. Environmental Protection Agency (U.S. EPA) protocols [15]. The following parameters were measured in the field using a Datasonde® Hydrolab (Hydrolab, Austin, TX, USA): pH, dissolved oxygen (DO), temperature, depth, and conductivity. Water clarity was measured using a Secchi disk. Chlorophyll *a* samples were taken at the deepest part of the lake, 1.5 m below the surface. The samples were filtered in the field following U.S. Environmental Monitoring and Assessment Program (EMAP) protocols [15].

Sediments were sampled using an Ekman dredge (GENEQ, Montreal, QC, Canada) at two locations in each body of water—at the deep hole and halfway to a shore. These samples were combined. In addition, a replicate sample was taken at the deep hole. Precleaned, wide-mouthed glass jars were inverted and pushed into the portion of sediment sample away from the sides of the dredge and were then capped with Teflon®-lined caps (VWR, Canlab, Mississauga, ON, Canada) and placed on ice for shipment to the lab. All sediment sampling and handling was performed in accordance with U.S. EPA protocols [15].

Laboratory methods

Fish specimens were processed for analysis in accordance with U.S. EPA procedures [16]. Dissection and tissue homogenization were conducted in a small, clean laboratory (not class 100) dedicated for fish processing.

Individual fish homogenates were analyzed for total mercury by cold-vapor atomic absorption spectrometry, using U.S. EPA method 245.6 [17], within their recommended holding-time limit for mercury (28 d) [16]. All handling of fish homogenates prior to analysis was conducted in a laminar airflow polypropylene fume hood for trace metal analysis that exceeds federal standard 209B for class-100 clean benches. Trace metal–grade sulfuric and nitric acids were used for fish sample digestions. The method detection limit (MDL) for mercury analysis in fish tissue of 0.020 mg/kg was experimentally determined using the conventional U.S. EPA procedure [18]. Accuracy for spiked fish samples and precision of the analyses were 104 ± 12.4 and 12.1 ± 9.7% (means ± 1 SD). The reference standard for mercury in fish tissue was freeze-dried oyster tissue (NBS 1566A). The accuracy of analyses of that standard was 101 ± 14.1%. Mercury in all laboratory reagent blanks was less than the MDL of 0.0002 mg/L.

Water-column samples were analyzed for chloride, using the argentometric method [19]; for calcium, using inductively coupled plasma–atomic emission spectrometry (ICP-AES) using U.S. EPA method 200.7 [20]; for sulfate, using turbidimetric nephelometry using U.S. EPA method 375.4 [21]; for ammonia-N (MDL = 0.02 mg/L), nitrate-N (MDL = 0.02 mg/L), and total phosphorus (MDL = 0.01 mg/L), using automated colorimetry on an autoanalyzer using U.S. EPA methods 350.1 [22], 353.1 [17], and 365.4 [17], respectively; and for dissolved organic carbon on glass-fiber–filtered samples, using ultraviolet (254-nm) absorbence, with potassium biphthalate as the standard [19].

Sediment samples were analyzed for total mercury and selenium. Sample aliquots for mercury analysis were digested in concentrated nitric and sulfuric acids and analyzed by cold-vapor atomic absorption spectrometry using U.S. EPA method 7471A [23]. For total selenium, the sediment samples were digested according to U.S. EPA method 3050A [23] and were analyzed by graphite furnace atomic absorption spectrometry using U.S. EPA methods 7060A and 7740 [23]. Accuracy for field sediments and precision for mercury determinations were 104 and 0.8%, respectively, and for selenium, they were 80.5 and 5.8%, respectively,. All reagent blanks were less than mercury and selenium MDLs of 0.0002 and 0.002 mg/L. The reference standard for sediments was dry river sediment (NBS 1645). Accuracies of analyses of that standard were 98 and 82% for mercury and selenium, respectively. Trace metal–grade acids were used for these analyses. Analyte concentrations were expressed as μg/g (dry weight).

Statistical methods

The number of each species of fish to be sampled in each lake in order to provide adequate statistical validity to the results was determined using fish mercury-concentration sampling variance from 10 years of monitoring in Massachusetts [8] and following consideration of available resources for fish collection and analysis.

Bivariate plots of all pairs of variables were also visually examined for outliers. Prior to statistical analyses of the raw tissue concentration data, the data were examined with linear regression analysis for correlations between mercury content and fish size (length or weight).

Lake trophic states were characterized with Carlson's Trophic State Index (TSI)[24], which gives a scaled measurement of water quality. Chlorophyll *a* measurements were used to calculate TSIs using the formula TSI = 30.6 ± 9.81 ln Chlorophyll *a* (mg/m^3) [25]. The TSIs of water bodies are scaled from 0 to 110, with oligotrophic lakes between 0 and 39, mesotrophic lakes between 40 and 50, and eutrophic lakes between 51 and 110. Lakes were grouped into these three categories. Because of their small number, mesotrophic lakes were grouped with eutrophic lakes for analyses of variance (ANOVA). The oligotrophic and eutrophic or mesotrophic categories were coded as 5 and 4 for statistical analyses. Subecoregions were numerically coded for analyses as follows: 1, Green Mountain/Berkshire Highlands; 2, Narragansett/Bristol Lowland; 3, Worcester/Monadnock Plateau.

The relative importances of the geographical locations of lakes (three subecoregion levels) and of their trophic states (two levels) were assessed with fixed-constants Model I ANOVA of mean lake tissue mercury concentrations, with replication for both yellow perch and brown bullhead. A separate analysis of covariance (ANCOVA) of mean lake tissue mercury concentrations across ecoregions and trophic states, using fish weight as a covariate, was performed for largemouth bass because of an observed relationship between weight (or total length) and mercury concentrations in this species [26]. Lake mean mercury values for each species were normally distributed (Kolmogorov–Smirnov test statistic with an α of 0.05 [26]); therefore, no data transformation was necessary to satisfy normality assumptions. We found unequal regression slopes [26] of tissue mercury on weight between lakes for largemouth bass and brown bullhead and equal slopes for yellow perch. These results were interpreted to mean that fish weight may have a differential effect on fish mercury concentrations between lakes for brown bullhead and largemouth bass. The effect of size may not have been removed from the data set for these two species even if ANCOVA was used to standardize mercury concentrations to a standard-sized fish [27]. Consequently, we chose to treat fish weight as an independent variable in all of our subsequent statistical procedures.

The multivariate data set was analyzed by factor analysis

[28] to assess which environmental parameters might influence regional differences associated with the bioaccumulation of mercury. Pearson's product moment correlation matrices for each species's mercury concentrations and environmental data were calculated. A varimax normalized rotation strategy was needed only with the bullhead data set to improve the separation of variables on factors. In factor analysis, the number of variables analyzed is limited to the number of cases. All species of fish were not available in every lake. We collected brown bullhead in 22 lakes, largemouth bass in 19 lakes, and yellow perch in 22 lakes. Stepwise multiple regressions were used to eliminate poorly correlated variables. Initially, the factor analysis was computed for two factors. The number of factors was increased iteratively until mercury in the species being analyzed scored high on only one factor. All statistical evaluations were performed with the Statistica/W, Version 5.0 software package (StatSoft, Tulsa, OK, USA).

RESULTS

Summary statistics for mercury concentrations in each species in the 24 lakes are presented in Table 1. Nine individuals of each species were not obtained in all water bodies. The results of physical and chemical sampling and measurement are contained in Tables 2 and 3. The pH value of 10.5 for Prospect Hill Pond (Table 2) was eliminated from further analysis as an outlier, since other chemical values for this pond suggested inconsistencies. Results for water ammonia-N, nitrate-N, and total phosphorus are not shown, as the majority of results were below method-detection limits.

Brown bullhead generally had the lowest muscle mercury concentrations, with mean tissue concentrations of 0.15 mg/kg wet weight (range = 0.01–0.79 mg/kg, 95th percentile concentration 0.32 mg/kg); yellow perch were intermediate, with 0.31 mg/kg (range = 0.01–0.75 mg/kg, 95th percentile concentration 0.57 mg/kg); and largemouth bass were highest, with 0.39 mg/kg (range = 0.05–1.1 mg/kg, 95th percentile concentration 0.91 mg/kg) (Table 1). The distribution of individual values of largemouth bass tissue mercury concentrations was somewhat similar to the log-normally distributed mercury concentration values in yellow perch and brown bullhead, in the concentration range of 0.2 to 0.6 mg/kg, but the bass distribution had a tail to the right beyond 0.6 mg/kg, with upper concentrations up to 1.1 mg/kg (plots not shown).

Largemouth bass are the only one of the three species in this study that exhibited a significant correlation ($r = 0.72$; p for H_0; $\rho = 0$ was 0.01) between fish length and mercury content for the combined data set. Similar relationships existed for weight (not shown). Correlation coefficients for regression equations of mercury on length for each species for individual lakes also generally exhibited the same pattern. The slopes of these regression lines were not equal among lakes for largemouth bass ($F_{16,116} = 4.74$; $p \leq 0.01$) and brown bullhead ($F_{17,125} \leq 3.59$; $p \leq 0.01$). They were equal for yellow perch ($F_{20,147} = 1.44$; $p = 0.11$).

The lake trophic-state indicator values ranged from 19 to 75, with 13 lakes falling in the oligotrophic range, 7 lakes in the mesotrophic range, and 4 lakes in the eutrophic range (Table 3). Analyses of variance showed no significant differences in tissue mercury concentrations ($p > 0.05$) between lakes of different trophic states for any of the three species. The ANOVA also determined that significant differences in fish mercury concentrations between subecoregions existed only in yellow perch ($p = 0.05$; $F_{2,16} = 3.62$) (Fig. 2). Regionally, the Narragansett/Bristol Lowlands subecoregion and the Green Mountain/Berkshire Highlands subecoregion have somewhat lower mercury in all species than does the Worcester/Monadnock Plateau.

Mercury concentrations in bass (Fig. 3a) were most strongly positively associated with the weight of the fish, lake size, and variables representing potential source area–contribution sizes (wetlands and watersheds). Mercury concentrations in this species did not correlate with either subecoregion or lake trophic state. Sediment mercury and selenium score high on an independent factor that also correlates with low DO. These two factors explained 46% of the variance in the data set. Mercury concentrations in yellow perch have a high negative correlation with factor 1 (Fig. 3b), while at the opposite end of the factor are high positive correlations for pH, conductance, and calcium, indicating inverse correlations between the tissue mercury and these lake chemistry variables. Variables loading orthogonal to this factor on factor 2 are primarily indicators of lake trophic status and are independent of the species' mercury concentrations. These two factors explained 43% of the total variance in the yellow perch data set. Mercury concentrations in brown bullhead tissue and pH had high opposite sign-factor scores on factor 6 (Fig. 3c). Trophic state indicator variables (DO, chlorophyll *a*) were independent of tissue mercury patterns, having high absolute value scores on factor 1. These two factors explained approximately 29% of the variance in the data set.

DISCUSSION

This study of the variation and possible determinants of fish tissue mercury in relatively non–source affected fresh water lakes in Massachusetts revealed that the order of species mercury concentrations, within the size ranges of fish sampled, was largemouth bass > yellow perch > brown bullhead. The largemouth bass sampled were primarily in the 4+ and 5+ year classes. Comparable mean concentrations to the 0.39 mg/kg for this data set for similarly aged fish in Michigan and Wisconsin data sets were 0.43 and 0.33 mg/kg [6], and they were 0.59 and 0.65 mg/kg in Lake Tohopekaliga, Florida [29]. The mean yellow perch mercury concentration of 0.31 mg/kg primarily represented fish in the 4+ and 5+ year classes. Comparable means for these year classes of yellow perch from other studies were 0.36 and 0.43 mg/kg in the Adirondacks of New York State [30] and 0.25 and 0.27 mg/kg in the Upper Peninsula of Michigan and in Wisconsin [6]. The majority of brown bullhead represented the 2+ through 4+ year classes. The interspecific differences in tissue mercury concentrations recorded in this study were consistent with observations from other studies using the same species or species representing the same trophic level [6,31,32]. They are also consistent with a priori considerations of the trophic level at which each species functions.

Variation in fish muscle mercury concentrations may be the result, in varying degrees, of biological variability associated with the species themselves (age, size, physiology, diet), of geological influences (bedrock and sediments), of chemical variability (water quality and mercury biogeochemistry), of physical variability (e.g., water temperature, lake and watershed size), and of other influences, such as climate and atmospheric deposition [33].

In our study design, we sought to control several sources of potential variation in tissue mercury concentrations. Seasonal influences on fish physiology and subsequently on fish

Table 1. Summary statistics for mercury concentrations in brown bullhead, largemouth bass, and yellow perch in Massachusetts lakes

Species	Region	Lake	*n*	Mercury mean ± 1 SD (mg/kg)	Mean weight (g)
Brown Bullhead					
	Green Mountain/Berkshire	Plainfield Pond	9	0.182 ± 0.069	97.11
		Ashfield Pond	9	0.083 ± 0.029	144.89
		Yokum Pond	6	0.050 ± 0.014	225.89
		Buckley Dunton Reservoir	9	0.168 ± 0.138	185.56
		Center Pond	9	0.123 ± 0.051	195.67
		Ashley Lake	10	0.099 ± 0.029	175.70
		Bog Pond	9	0.149 ± 0.056	72.67
		Crooked Pond	9	0.115 ± 0.046	136.94
	Narragansett/Bristol	Elders Pond	6	0.279 ± 0.265	466.00
		West Meadow Pond	8	0.074 ± 0.019	515.00
		Little Quitticas Pond	4	0.225 ± 0.152	470.75
		Prospect Hill Pond	0	—	—
		North Watuppa	2	0.100 ± 0.002	563.50
		Somerset Reservoir	2	0.187 ± 0.028	733.50
		Middle Pond	3	0.026 ± 0.014	416.00
		Watson Pond	9	0.069 ± 0.025	460.33
	Worcester/Monadnock	Wampanoag Lake	9	0.214 ± 0.123	105.67
		Upper Naukeag	0	—	—
		Hilchey Pond	9	0.186 ± 0.062	205.62
		Sheomet Pond	9	0.097 ± 0.037	66.67
		Upper Reservoir	2	0.260 ± 0.018	224.50
		Laurel Lake	9	0.116 ± 0.054	329.00
		Gales Pond	9	0.322 ± 0.127	142.44
		Fitchburg Reservoir	8	0.107 ± 0.058	172.00
				Species mean = 0.147 ± 0.078	
Largemouth bass					
	Green Mountain/Berkshire	Plainfield Pond	9	0.626 ± 0.281	767.75
		Ashfield Pond	9	0.468 ± 0.315	419.11
		Yokum Pond	9	0.188 ± 0.081	374.50
		Buckley Dunton Reservoir	11	0.426 ± 0.233	572.00
		Center Pond	9	0.323 ± 0.139	729.10
		Ashley Lake	0	—	—
		Bog Pond	9	0.413 ± 0.192	794.44
		Crooked Pond	0	—	—
	Narragansett/Bristol	Elders Pond	9	0.250 ± 0.075	555.78
		West Meadow Pond	9	0.144 ± 0.050	298.33
		Little Quitticas Pond	5	0.280 ± 0.110	272.60
		Prospect Hill Pond	9	0.199 ± 0.049	541.44
		North Watuppa	9	0.724 ± 0.198	1150.56
		Somerset Reservoir	9	0.668 ± 0.298	713.50
		Middle Pond	10	0.330 ± 0.188	556.80
		Watson Pond	9	0.309 ± 0.057	581.22
	Worcester/Monadnock	Wampanoag Lake	9	0.439 ± 0.148	475.11
		Upper Naukeag	1	0.366	328.00
		Hilchey Pond	0	—	—
		Sheomet Pond	0	—	—
		Upper Reservoir	9	0.551 ± 0.107	488.89
		Laurel Lake	9	0.392 ± 0.100	619.11
		Gales Pond	0	—	—
		Fitchburg Reservoir	0	—	—
				Species mean = 0.394 ± 0.165	
Yellow perch					
	Green Mountain/Berkshire	Plainfield Pond	9	0.342 ± 0.126	80.78
		Ashfield Pond	9	0.330 ± 0.085	75.67
		Yokum Pond	9	0.105 ± 0.046	118.11
		Buckley Dunton Reservoir	9	0.272 ± 0.145	96.33
		Center Pond	9	0.181 ± 0.079	121.44
		Ashley Lake	10	0.380 ± 0.176	104.80
		Bog Pond	10	0.284 ± 0.071	133.11
		Crooked Pond	9	0.46 ± 0.076	139.70

mercury concentrations were reduced by the choice of sampling time. Control of the influence of fish size and age on tissue mercury was accomplished by confining our sampling to restricted size ranges of fish. In practice, a wider size range of fish than intended was obtained. However, the lack of correlation, over all samples, between mercury concentration and size in yellow perch or brown bullhead suggests that our attempt to control for fish size by limiting the size range during

Table 1. Continued

Species	Region	Lake	*n*	Mercury mean ± 1 SD (mg/kg)	Mean weight (g)
	Narragansett/Bristol	Elders Pond	9	0.273 ± 0.062	124.56
		West Meadow Pond	0	—	—
		Little Quitticas Pond	9	0.272 ± 0.139	113.89
		Prospect Hill Pond	9	0.106 ± 0.063	122.78
		North Watuppa	9	0.338 ± 0.163	170.88
		Somerset Reservoir	9	0.203 ± 0.054	32.44
		Middle Pond	9	0.155 ± 0.052	258.00
		Watson Pond	9	0.195 ± 0.065	87.89
	Worcester/Monadnock	Wampanoag Lake	9	0.439 ± 0.067	74.88
		Upper Naukeag	9	0.547 ± 0.091	94.67
		Hilchey Pond	9	0.314 ± 0.090	142.67
		Sheomet Pond	0	—	—
		Upper Reservoir	9	0.465 ± 0.148	103.56
		Laurel Lake	9	0.219 ± 0.056	97.56
		Gales Pond	9	0.514 ± 0.073	91.00
		Fitchburg Reservoir	9	0.326 ± 0.088	112.22
				Species mean = 0.305 ± 0.125	

capture was successful. The observed relationship with size and mercury in bass may be related to interspecific variation in the kinetics of mercury bioaccumulation [32]. Largemouth bass are long-lived and have the largest body sizes and probably the lowest rates of growth and metabolism at older ages [14]. They are also the only species studied here that had a positive, significant correlation between mercury and weight. Yellow perch and brown bullhead have smaller body sizes, shorter lifespans (in the case of perch), and, presumably, higher rates of growth and metabolism. The older, slower growing fish had longer times to accumulate and concentrate mercury (as a result of more uptake than excretion), because growth dilution of methylmercury is not sufficiently rapid to offset this effect. In the other two species, the higher growth rates may have resulted in growth dilution of their body burdens of mercury, thereby offsetting possible accelerated mercury uptake due to higher metabolic rates and age-dependent bioaccumulation.

A slight geographic gradient of fish mercury concentration for yellow perch was detectable in our analyses, even across the relatively narrowly defined differences between ecological subregions. Allen-Gil et al. [34] did not note spatial differences in fish species mercury concentrations across geographic regions delineated on the basis of ecological, geological, and climatic factors. Lathrop et al. [35] noted a west to east increase in walleye mercury concentrations across northeastern Minnesota, northern Wisconsin, and southeastern Ontario, Canada, which is possibly related to acidic deposition patterns. Ecoregional differences in Massachusetts are associated with pH differences and may also be overshadowed by other lake-specific factors.

Mercury concentrations in sediment samples ranged from 0.008 mg/kg to 0.425 mg/kg (Table 3). None of the species studied in this project showed a relationship between tissue mercury and sediment mercury or selenium concentrations. Figure 3a shows that sediment mercury and selenium vary independently from fish mercury. Selenium can form highly insoluble complexes with mercury and thereby reduce its biological availability [36]. Under low pH conditions, leaching of sedimentary metals into surface waters and subsequent availability of these metals for bioaccumulation may be facilitated in a complex relationship modulated by the amount and types of particulate and organic matter in the water column and by the pH and Eh of the sediment [37]. In brown bullhead, the source of mercury may not be confined to diet, given the bottom-dwelling habitat of the species and its scaleless, permeable skin. Underlying relationships between sedimentary mercury and selenium may have been obscured with our bulk mercury concentration determination, since mercury is probably preferentially associated with silts and clays, and a normalization to the mass of this size fraction might have been more useful.

Our analyses indicated a clear link between certain environmental characteristics and elevated mercury concentrations in fish. Low pH of the water body was a major correlate to tissue mercury concentrations in brown bullhead and yellow perch (Fig. 3b, c) but not in largemouth bass (Fig. 3a). The association between high mercury concentrations and low pH is clearly delineated by factor analysis. This environmental variable would also seem the most likely to represent subecoregion variability in our analyses. Some of the continuous variables measured in the field (e.g., Secchi disk depth, chlorophyll *a,* DO) represent measures of trophic status that are perhaps better suited for use in the correlation and other association tests than as a coded variable. The factor analysis provided complementary information, scoring mercury in perch highly negative on the same factor as pH. Suns and Hitchin [38] observed a similar relationship in yellow perch from 16 lakes situated on the Precambrian Canadian Shield north of Toronto, Canada.

Low pH has been most consistently documented as being responsible for elevated tissue mercury concentrations in freshwater fish in uncontaminated lakes [2,6]. Possible mechanisms associated with this relationship include [37] (1) mercury entering watersheds with atmospheric deposition; (2) mobilization of existing sediment-bound mercury and mercury present in the surrounding watershed by acidification of surface water runoff and lake water, leading to increases in the amount of mercury available for methylation and bioaccumulation; (3) differential production of the more bioavailable monomethylmercury form of mercury at lower pH; and (4) alteration of rates of mercury methylation and demethylation by microorganisms by acidic conditions. Having reviewed evidence for each of these mechanisms, Richman et al. [37] concluded that they were not mutually exclusive processes and that mercury cycling and uptake into fish tissues was governed

Table 2. Lake water quality characteristics[a]

Lake	pH	Chlorophyll *a* (mg/m^3)	Secchi depth (m)	Depth (m)	Conductivity (mS)	DO (mg/L)	DOC (mg/L)	Cl^- (mg/L)	Ca (mg/L)	SO_4 (mg/L)
Plainfield Pond	7.5	1.2	2.75	2.75	37	8.6	≤MDL	4	2.3	≤MDL
Ashfield Pond	8.5	.5	3.1	5	178	8.87	0.7	28	16	4
Yokum Pond	7.2	.8	2.75	2.9	50.8	8	0.5	1	20	4
Buckley Dunton Reservoir	5.7	5.1	1.2	3.25	29.1	6.36	9.7	2	1.8	2
Center Pond	7.5	2.1	2.75	4.8	114	8.34	0.5	19	28	4
Ashley Lake	7.9	1.9	4	13.8	47.9	8.09	3.1	2	3.8	≤MDL
Bog Pond	6.5	3.7	1.5	2	19.2	7.21	12.1	1	2.7	≤MDL
Crooked Pond	6.7	2.8	2.25	2.75	23	7.13	2.2	≤MDL	1.4	≤MDL
Elders Pond	7.1	14.3	2.9	13.8	117.4	7.85	3.4	21	3	8
West Meadow Pond	7.6	90.8	0.04	1.5	209	2.54	23.3	35	8.3	6
Little Quitticas Pond	7.1	1.5	2.5	3.75	102.8	7.54	8	18	3.4	6
Prospect Hill Pond	10.5	1.9	1.25	2	135.7	9.92	6.1	11	0.8	17
North Watuppa	6.1	1.1	2.75	4.75	93.3	7	4	17	2.9	8
Somerset Reservoir	7.3	2.9	2.5	9.5	101.7	7.39	6	13	6.6	9
Middle Pond	8.9	2.5	2.2	4.5	152.6	7.9	2.5	22	7.7	4
Watson Pond	8.3	40.2	0.6	2.9	101.3	7.32	13	16	5.6	≤MDL
Wampanoag Lake	5.4	1.1	2.5	3.75	79.2	7.84	6.1	18	2.2	2
Upper Naukeag	5.6	.4	7.5	13.75	47.8	7.64	0.1	9	1	≤MDL
Hilchey Pond	7.3	13.2	0.07	2.7	152	7.13	40	14	5.5	4
Sheomet Pond	6.8	1.8	2.25	3.2	37	7.92	4.2	3	2.2	3
Upper Reservoir	4.9	2.3	0.75	1.1	45.8	7.15	58.8	6	1.8	≤MDL
Laurel Lake	6.4	.3	6	7.3	24.7	7.87	5	≤MDL	5.5	2
Gales Pond	6.1	4.4	0.75	1.3	36.7	7.19	37	5	2.6	2
Fitchburg Reservoir	6.3	4.1	5.25	6	73.9	7.99	0.8	14	1.7	4

[a] DO = dissolved oxygen; DOC = dissolved organic carbon; MDL = method detection limit.

Table 3. Lake characteristics

Lake	Sediment		Trophic state[a]	Watershed area (hectares)	Pond area (hectares)	Wetland area (hectares)
	Mercury (mg/kg)	Selenium (mg/kg)				
Plainfield Pond	0.200	1.80	o	170	25.5	9.5
Ashfield Pond	0.172	1.10	o	287	15.8	1.7
Yokum Pond	0.030	0.32	o	161	38.4	2.8
Buckley Dunton Reservoir	0.290	1.34	m	581	58.7	17.1
Center Pond	0.008	0.29	o	256	41.3	9.5
Ashley Lake	0.222	1.26	o	173	44.9	4.2
Bog Pond	0.133	1.27	m	353	15.0	13.8
Crooked Pond	0.250	1.92	m	96	13.8	8.3
Elders Pond	0.029	0.11	e	232	55.4	5.9
West Meadow Pond	0.366	2.81	e	1196	29.1	88.5
Little Quitticas Pond	0.279	1.54	o	417	112.5	52.8
Prospect Hill Pond	0.213	1.62	o	124	17.0	8.7
North Watuppa	0.149	≤MDL	o	2935	700.1	304.5
Somerset Reservoir	0.215	0.76	m	374	66.4	36.7
Middle Pond	0.128	0.76	m	416	9.7	39.9
Watson Pond	0.425	1.98	e	157	29.1	25.7
Wampanoag Lake	0.301	1.14	o	773	90.7	110.3
Upper Naukeag	0.148	2.31	o	495	123.0	27.2
Hilchey Pond	0.282	0.69	e	823	4.9	144.2
Sheomet Pond	0.266	0.95	o	1382	12.5	19.8
Upper Reservoir	0.215	2.05	o	445	16.6	85.9
Laurel Lake	0.274	1.45	o	219	16.6	3.1
Gales Pond	0.356	1.85	m	828	4.5	78.1
Fitchburg Reservoir	0.260	1.06	m	554	60.7	19.1

[a] o = oligotrophic; e = eutrophic; m = mesotrophic; MDL = method detection limit.

by an array of interrelated, variables, the relative importance of which can differ from lake to lake.

Our analyses did not show an association between fish tissue mercury concentrations and the lake TSI. Trophic status and variables associated with it are relatively independent of both fish mercury and pH (Fig. 3). For example, chlorophyll *a* and Secchi disk depth, both associated with lake trophic status, did not partition onto the same factor as species mercury values, indicating that variance in trophic state variables was independent of mercury concentrations in most fish tissue. Other reviews [37] and studies [9,39,40] on this specific relationship have noted that, while the general availability of mercury within aquatic ecosystems may be affected by trophic status, other abiotic factors interfere with and confound the issue.

The ratio of basin area to pond area was not a strong correlate of fish mercury concentrations in any of the species we studied in Massachusetts. The absence of such a relationship does not support the logic that where the basin (watershed) is much larger than the pond, there should be a tendency to have higher mercury concentration in fish tissue, reflective of mercury transport from the watershed [41]. In largemouth bass, however, we did find significant correlations between tissue mercury concentrations and the size of the watershed and the lake area as well as the area in the watershed occupied by wetlands (Fig. 3a). The relative importance of watershed-derived mercury to fish mercury is not consistent in various studies [2,6] and sometimes appears to be a function of the types of water inputs to the lakes. In cases where there has been little surface-water inflow into lakes [6,41], no relationship has been seen between fish mercury concentrations and watershed area to lake volume ratios, whereas positive relationships have been seen in lakes with greater surface-water inputs from drainage basins [38].

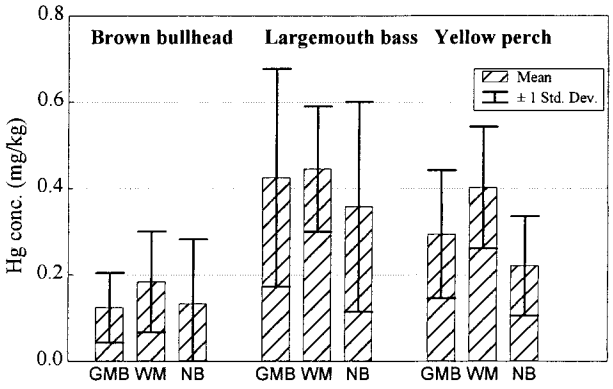

Fig. 2. Mean species mercury concentrations (mg/kg) in Massachusetts subecoregions. GMB = Green Mountain/Berkshire Highlands; WM = Worcester/Monadnock Plateau; NB = Narragansett/Bristol Lowland.

In addition to substantiating the recognized association between fish tissue mercury and acid waters, the principal contributions of the present study include insight into the relative importance of the various biologic, chemical, and geologic factors that may influence fish mercury bioconcentration patterns. Specifically, given that significant ecoregional differences in tissue mercury concentrations only existed in one species, the properties of individual lakes within these narrowly defined regions are more important than are regional variations in determining fish mercury concentrations. The results for largemouth bass, contrasted with those of yellow perch and brown bullhead, suggest that species whose mercury concentrations exhibit major variation associated with size or food-chain position may not be good choices for evaluating the effects of environmental variables. The additional variability introduced by using such species tends to obscure other

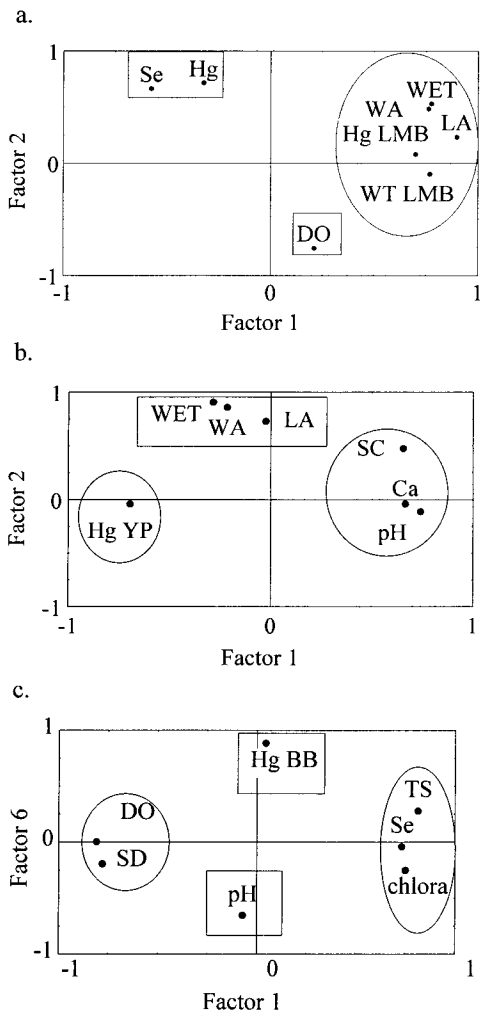

Fig. 3. Rotated factor score plots for: (**a**) largemouth bass, (**b**) yellow perch, and (**c**) brown bullhead. Only variables with scores >|0.65| are shown. Elipses and squares highlight variables grouping on factor 1 or on other factors, respectively. Key: Ca = calcium; chlora = chlorophyll *a*; DO = dissolved oxygen; Hg = sediment mercury; HgBB = mercury in bullhead; HgLMB = mercury in bass; HgYP = mercury in yellow perch; LA = lake area; SC = specific conductance; SD = Secchi disk depth; TS = trophic status; WA = watershed area; WET = wetland area; WTLMB = wet weight of bass.

relationships. This study clearly shows the value of using a specified size range of species that exhibit little size to mercury ratio covariance.

Studies such as this, in which fine-scale ecoregional differences are not usually significant, do not indicate that ecoregional differences are not meaningfully related to fish mercury on larger geographic scales. Indeed, the variables measured in this study may well be important on larger geographic scales and may be beneficially examined in that context. Literature on mercury bioaccumulation is generally dominated by data from waters in regions where bioaccumulation has reached levels of concern, whereas data from areas where bioaccumulation has not been a concern has not been published in the open literature as frequently. Inclusion of this type of data in regional analyses would provide a broader spectrum of conditions for evaluating the importance of ecoregional differences.

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