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Freshwater Fish Mercury Concentrations in a Regionally High Mercury Deposition Area

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Abstract We sampled and analyzed individually, edible dorsal muscle from largemouth bass (LMB), *Micropterus salmoides* (n=138) and yellow perch (YP), *Perca flavescens* (n=97) from 15 lakes to investigate potential local impacts of mercury emission point sources in northeastern Massachusetts (MA), USA. This area was identified in three separate modeling exercises as a mercury deposition hotspot. In 1995, 55% of mercury emissions to the environment from all MA sources came from three municipal solid waste combustors (trash incinerators) and one large regional medical waste incinerator in the study area. We determined the mercury accumulation history in sediments of a lake centrally located in the study area. Recent maximum mercury accumulation rates in the sediment of the lake of ~ 88 μg/m^2/year were highly elevated on a watershed area adjusted basis compared to other lakes in the Northeast and Minnesota. Fish from the study area lakes had significantly (p=0.05) greater total mercury concentrations than fish from 24 more rural, non-source-impacted lakes in other regions of the state (LMB n=238, YP n=381) (LMB: 1.5–2.5 x; YP: 1.5 x). The integration of this extensive fish tissue data set, depositional modeling projections, historical record of mercury accumulation in sediments of a lake in the area, and knowledge of substantial mercury emissions to the atmosphere in the area support designation of this area as a mercury depositional and biological concentration hotspot in the late 1990s, and provides further evidence that major mercury point sources may be associated with significant local impacts on fisheries resources.

Keywords Accumulation · Deposition · Fish · Hotspot · Incinerator · Lake · Largemouth bass · Mercury · Muscle · Sediment core · Yellow perch

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1 Introduction

Fish can reflect elevated mercury inputs to the environment and are used as monitoring sentinels (e.g., Riisgard and Famme 1988; Olivero and Solano 1998;

and Haines et al. 2003). Mercury in fish flesh can represent an ecological and human health hazard to those ingesting the fish (Boening 2000; Henny et al. 2002; and Mergler et al. 2007). Lake bottom sediments are also used as sentinels for recent inputs of mercury and, when sampled and analyzed vertically, provide historical records of net mercury deposition to lake bottoms from direct atmospheric deposition and surrounding watershed inputs (Frazier et al. 2000; Kamman and Engstrom 2002).

A statewide advisory is in effect in Massachusetts (MA) warning sensitive human populations to avoid consuming any native freshwater fish caught in the state due to unsafe levels of mercury (MA DPH 2001). Approximately 52% of the rivers and lakes in MA sampled since 1983 are also subject to fish consumption advisories for the rest of the population as a result of mercury contamination (MA DPH 2007).

Many of these MA water bodies do not have water discharge sources of mercury but are instead likely to be primarily impacted by atmospheric mercury deposition. Mercury deposited from the atmosphere is thought to come from long-range transport and near-field point sources (Dvonch et al. 2005). These sources can be anthropogenic, which are likely to predominate in this area, or natural, such as volcanoes and earth crustal off-gassing. Long-range transport-derived deposition should be relatively uniform across a region in the absence of weather-influencing topographic features. Zones downwind from major point sources (e.g., smelters, tailings piles, and power stations (Goodman and Roberts 1971)) or urban areas may be subject to increased atmospheric deposition and subsequent inputs to aquatic sediments of contaminants (Engstrom and Swain 1997). High ambient atmospheric concentrations of Hg(II), which typically occur near large emission sources, may significantly increase overall mercury deposition (US EPA 1997; Bullock and Brehme 2002).

An area encompassing one half degree longitude by one third degree latitude (nominally 36 km) including portions of northeast Massachusetts (NE MA) and southeast New Hampshire in the northeastern continental US was identified through air deposition modeling using the Regional Lagrangian Model of Air Pollution (RELMAP) as having the highest predicted annual levels of atmospheric mercury deposition in New England based on 1989 meteorology and emissions data for the mid 1990s (NESCAUM et al. 1998). In that assessment, performed by the US Environmental Protection Agency (EPA) National Exposure Research Laboratory, mercury wet deposition attributable to regional municipal solid waste combustors was estimated to be in excess of 30 $\mu g/m^2$/year, and total wet and dry deposition from all sources was estimated to be in excess of 100 $\mu g/m^2$/year in the study area. More recent modeling results using the industrial source complex short-term model (ISCST3) also identified this area as a mercury deposition hotspot with predicted deposition rates, based on 5 km grid resolution, ranging from 17–804 $\mu g/m^2$/year in 1996 and 7–76 $\mu g/m^3$/year in 2002 (Evers et al. 2007). Lastly, unpublished results derived using the Regional Modeling System for Aerosols and Deposition (REMSAD) with 36 km grid resolution and 1996 meteorology also predicted this area to have had the highest mercury wet deposition rate in New England in the mid 1990s (Graham et al. 2007). These model-predicted rates of deposition are far in excess of measured wet deposition rates from the Mercury Deposition Network (MDN) sites in the northeast states (VanArsdale et al. 2005). Notably, none of the MDN sites are located within the "hotspot" area predicted by the models. Although the accuracy of modeled deposition estimates for any individual grid are uncertain due to model limitations, these consistent results suggest that this area likely experienced significantly elevated mercury deposition.

Preliminary muscle sampling of fish in NE MA in 1994 also suggested high fish muscle mercury concentrations in the area (Massachusetts Department of Environmental Protection (MassDEP), unpublished data). A northeast United States (US) regional yellow perch (YP) (*Perca flavescens*) mercury hotspot was identified in southern New Hampshire and northeastern Massachusetts by Evers et al. (2007) based, in part, on portions of the data described in this study.

This putative northeastern MA mercury deposition and fish hotspot area, the focus of the present study, had four significant point sources of atmospheric mercury emissions in the last two decades of the twentieth century: three municipal solid waste combustors (MSWC) (Fig. 1) having a combined annual throughput in the middle to late 1990s of approximately 1×10^6 metric tons per year based on facility permits and reporting required under state and federal regulations (MassDEP, unpublished data) and a medical waste incinerator (MWI). The three MSWCs collectively

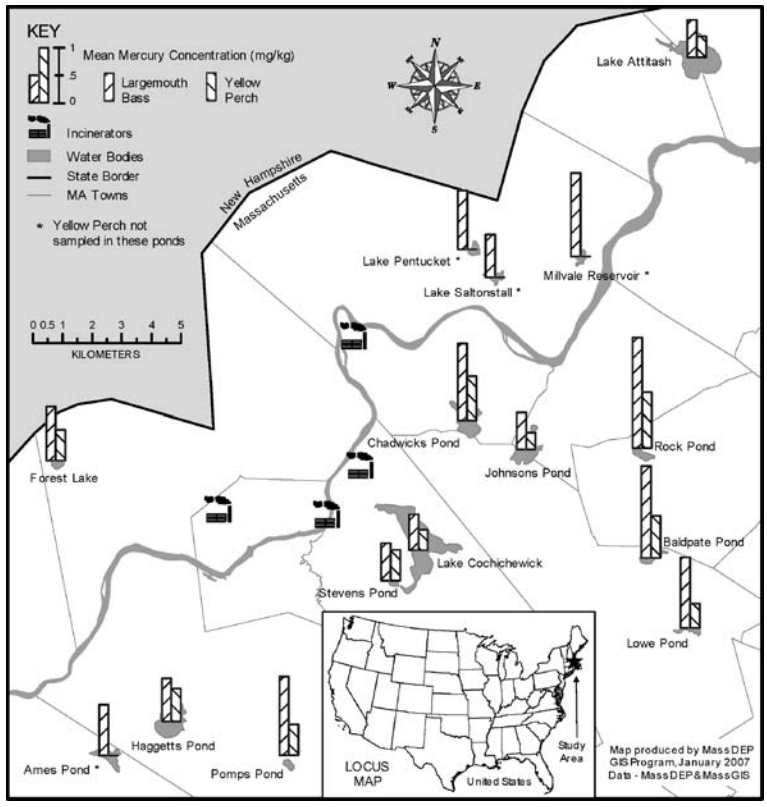

Fig. 1 Incinerator locations and mean muscle mercury concentrations for YP and size-standardized LMB in northeast Massachusetts study lakes

accounted for approximately 62% (~1,700 kg/year) of the statewide stack emissions of mercury from MSWC, and 55% of the total in-state mercury releases to the environment in 1995 (Smith and Rowan West 1996). Prior to 2000 when MSWCs were required to significantly reduce mercury emissions under stringent state and federal regulations, these types of facilities were recognized to be among the largest contributors of mercury emissions in the US (US EPA 1997) and Massachusetts (Smith and Rowan West 1996).

The first objective of this study was to evaluate the historical and recent magnitude of mercury deposition to lake bottom sediments in this targeted geographic area in comparison to published data on other water bodies and to results from atmospheric mercury deposition modeling. This was accomplished using sediment cores from a lake centrally located in the study area. The second objective was to determine if the area was a fish mercury hotspot. This was assessed by comparing the levels of edible fish muscle mercury concentrations in the study area with other regions of the state and country.

2 Materials and Methods

2.1 Study Design

The study area (~20×26 km, bounded by latitudes 42°38′ and 42°51′N, and 70°59′ and 71°15′W longitude) represented a large part of the high mercury deposition zone originally delineated in the 1998 regional deposition modeling project (Fig. 1). We sampled lake bottom sediment from a representative lake centrally located in the study area (Lake Cochichewick) using a sediment corer. Sedimentary layers were analyzed for mercury and other metals using trace metal clean techniques, and ^{210}Pb and ^{137}Cs using established geochronological dating techniques (Appleby and Oldfield 1992) to determine the historical record of mercury deposition to the lake beds and to more specifically provide data on the magnitude of recent mercury accumulation in the sediments.

We also sampled fish from 15 lakes from that area in April–May 1999. Lakes located elsewhere in Massachusetts were used for comparison. These

included 24 lakes that we sampled in the fall of 1994 (Rose et al. 1999), and an additional nine lakes sampled in the springs of 1999, 2001, and 2002 (Table 1). Surface and watershed areas of lakes and ponds were obtained from GIS data layers "Hydrography (1:25,000), 2005", and "Drainage Sub-basins, 2005", developed by the MassDEP and the Office of Geographic and Environmental Information (MassGIS), Commonwealth of Massachusetts, Executive Office of Energy and Environmental Affairs.

Largemouth bass (LMB, *Micropterus salmoides*) and YP were obtained from lakes chosen on the basis of: size of lake (4 ha minimum size), availability of fish species, availability of access, distance from other previously sampled lakes, and absence of any known point source inputs of mercury. Target sample sizes were nine fish of each species from each lake in 1994 and 1999, and 12 LMB and 30 YP in later years. These two species were used because LMB are known to bioaccumulate mercury to relatively high levels in the freshwater food chain (Cizdziel et al. 2002; Cizdziel et al. 2003; Saiki et al. 2005; and Paller and Littrell 2007), they are representative of an upper level trophic group (Scott and Crossman 1973), and are very common throughout Massachusetts (Hartel et al. 2002). YP are ubiquitous introduced omnivores (Scott and Crossman 1973; Hartel et al. 2002) and have been used in other studies as sentinel species (Ion et al. 1997; Rencz et al. 2003; Kamman et al. 2005). Both species are also popular recreational fisheries species in MA (R. Hartley, Massachusetts Department of Fish and Game, Division of Fisheries and Wildlife, personal communication).

2.2 Field Methods

Two sediment cores were taken in May 2001 from Lake Cochichewick, North Andover, MA. This is a 233 ha glacial lake (~14 m maximum depth) with a mixed forest/residential land use watershed of 1,236 ha (Table 1). Cores were obtained from the deeper regions of the lake with a hand-deployed custom-made 15×15 cm box corer with polycarbonate liners, designed to obtain undisturbed cores from soft sediments (Pedersen et al. 1985) from a small boat. After penetration, a lid capping the top of the box corer is activated, the bottom sealed by closure of two clamshell type spades upon retrieval, and the corer brought to the surface with minimal disturbance of the surface layers of the core. Once on board, any surface water remaining on top of the core was carefully removed using a siphon, the core in its polycarbonate liner capped and placed vertically in a cooler with ice, and then returned to the lab where it was sectioned.

Fish collection and handling procedures through laboratory delivery were as described in Rose et al. (1999). Water quality was assessed with depth profiles of water temperature, dissolved oxygen concentration, pH, and conductivity at one-meter depth intervals throughout the water column from one station in each lake located over the deepest portion of the lake.

2.3 Laboratory Procedures

Sediment cores were sectioned at 1 cm intervals using a custom designed PVC extruder. The extruder jammed during sectioning of the first Lake Cochichewick core and prohibited sectioning of this core below the first two centimeters. Lake Cochichewick Core #2 was then sectioned at 1 cm intervals except for the 0–2 cm interval, which was collected as one sample.

Each core section was homogenized using non-metallic trace-metal-clean implements before drying in plastic jars and then weighing. Approximately 100-g wet weight of the homogenized wet sample was placed in Teflon-lined cans and counted directly using two different low-level intrinsic germanium (Ge) detectors. The remainder of the homogenate from each section was dried at 60°C to constant weight and used for chemical analysis, and determination of water content.

All samples were counted for sufficient time to acquire net counts of at least 1000 for the ^{210}Pb (46 keV γ, $t_{1/2}$=22.26 years) isotope. Samples were counted using one of two planar intrinsic Ge detectors, either a Canberra GL2020R or Canberra BE5030. ^{137}Cs (662 keV γ, $t_{1/2}$=30.2 years) data were also used to assist in the dating analysis. Gamma spectra were recorded using a Genie 2000 MCA and software. Excess ^{210}Pb was determined by correction using supported ^{210}Pb counts averaged over the 23–30 cm depth intervals (0.0604±0.0016 Bq/g dry weight). All sample counts were appropriately corrected for background and efficiencies established using an interlaboratory standard ("D" Standard made by combining Hudson River surface sediment with NBS river sediment standard 4350b) provided by the Lamont Doherty Earth Observatory's Isotope

Table 1 Properties of lakes and central tendency fish muscle mercury concentration estimates (mg total Hg/kg wet wt)

Area, date(s) sampled	Lake	Latitude N, longitude W	Surface area (ha)	Watershed area (ha)	Max. depth (m)	pH	LMB $\bar{x}$±1 s		*n*	YP $\bar{x}$±1 s		*n*
							raw	Size-standardized		Raw	Size-standardized	
Northeastern MA., 1999	Ames Pond	42° 38′ 18″–71° 13′ 30″	31	395	3	7.8	0.80±0.16	0.78±0.13	9			0
	Baldpate Pond	42° 41′ 55″–71° 00′ 06″	24	1037	12	8.2	1.33±0.16	1.40±0.11	9	0.61±0.23	0.64±0.22	9
	Chadwicks Pond	42° 44′ 31″–71° 04′ 49″	70	416	8	7.3	1.17±0.29	1.17±0.29	12	0.66±0.21	0.67±0.21	9
	Lake Cochichewick	42° 42′ 16″–71°0 5′ 50″	233	1236	14	7.4	0.58±0.19	0.55±0.16	9	0.32±0.09	0.32±0.09	9
	Forest Lake	42° 43′ 44″–71° 14′ 49″	19	602	7	7.8	0.71±0.07	0.82±0.06	9	0.46±0.14	0.47±0.12	9
	Haggetts Pond	42° 38′ 54″–71° 11′ 55″	85	561	14	8.5	0.89±0.54	0.66±0.26	8	0.38±0.14	0.50±0.14	9
	Johnson Pond	42° 43′ 58″–71° 03′ 06″	78	399	7	6.7	0.61±0.15	0.56±0.07	9	0.30±0.06	0.26±0.06	9
	Lake Attitash	42° 51′ 03″–70° 58′ 57″	149	997	7	7.0	1.01±0.25	0.57±0.15	9	0.29±0.09	0.32±0.09	9
	Pentucket Pond	42° 47′ 29″–71° 04′ 24″	15	50	8	8.0	1.30±0.76	0.90±0.25	10			0
	Lake Saltonstall	42° 47′ 00″–71° 03′ 59″	18	5850	9	7.8	0.51±0.19	0.65±0.06	9			0
	Lowe Pond	42° 40′ 35″–70° 59′ 07″	14	1725	2	8.1	1.11±0.28	1.08±0.23	9	0.43±0.15	0.37±0.14	9
	Millvale Reservoir	42° 47′ 22″–71° 01′ 49″	18	509	3	8.0	1.12±0.18	1.28±0.17	9			0
	Pomps Pond	42° 38′ 09″–71° 09′ 07″	10	691	3	8.0	1.32±0.50	1.20±0.28	9	0.54±0.18	0.47±0.18	7
	Rock Pond	42° 43′ 47″–71°00′ 23″	20	911	6	6.5	1.63±0.21	1.68±0.17	9	0.86±0.18	0.85±0.18	9
	Stevens Pond	42° 41′ 29″–71° 06′ 30″	9	473	3	8.1	0.61±0.17	0.57±0.12	9	0.46±0.09	0.47±0.08	9
Rest of State, 1999, 2000, 2001	Bare Hill Pond	42° 29′ 24″–71° 35′ 54″	126	1976	5.5	7.1	0.55±0.13	0.55±0.1	9	0.34±0.11	0.33±0.11	9
	Fort Pond	42° 31′ 29″–71° 41′ 13″	31	739	4	6.8	0.29±0.07	0.32±0.07	9	0.34±0.13	0.13±0.12	9
	Hickory Hills Pond	42° 36′ 47″–71° 42′ 39″	125	549	4	6.7	0.95±0.19	1.00±0.12	9	0.36±0.10	0.36±0.09	9
	Long Pond	42° 41′ 48″–71° 22′ 9″	68	1912	8	6.7	0.65±0.11	0.66±0.09	9	0.39±0.20	0.55±0.14	9
	Massapoag Pond	42° 38′ 55″–71° 29′ 42″	45	2529	10	7.4	0.78±0.08	0.74±0.06	9	0.43±0.16	0.42±0.12	9
	Newfield Pond	42° 38′ 0″–71° 23′ 21″	31	519	7	8.0	0.66±0.10	1.21±0.08	9	0.28±0.09	0.33±0.06	9
	North Watuppa Pond	41° 43′ 6″–71° 6′ 7″	700	2992	8	7.0	0.72±0.20	0.49±0.11	9	0.34±0.16	0.36±0.11	8
	Onota Lake	42° 28′ 27″–73° 16′ 43″	262	899	16	8.2	0.24±0.11	0.30±0.46	21	0.23±0.08	0.27±0.07	30
	Wequaquet Lake	41° 40′ 22″–70° 20′ 30″	232	54373	9	6.9	0.55±0.3	0.61±0.13	30	0.49±0.13	0.41±0.09	30

LMB Largemouth bass, *YP* yellow perch.

Research Laboratory and NBS river sediment standard 4350b. All standards and samples were decay-corrected as appropriate.

Samples for total mercury and other metal concentration determinations in the dried sediment obtained for each core section were prepared using a microwave-assisted digestion technique (Wallace et al. 1991), validated using appropriate reference standards and subsequent analysis by cold vapor atomic absorbance (CETAC M-6000) or ICP-MS (Perkin Elmer 6100DRC). Detailed methods and results for this portion of the core analysis are not reported here but are available in Wallace et al. (2004). Metal concentration results are expressed on a dry weight basis. Mercury analytical procedural blanks (n=4) averaged 11.5±0.8 ng Hg for the Cochichewick core. The limit of detection (given as 3s of the mean of the procedural blanks) was equivalent to 11.9 ng/g dry weight respectively for a 0.2 g digestion weight. Our digestion blank is typically an order of magnitude lower (<1 ng) for sediments but with similar uncertainty. The higher but consistent blank for the Cochichewick core was attributed to a high mercury concentration in one of the digestion acids used for those samples. Six replicate samples of the PACS-1 sediment reference standards were run with an average recovery of 101% and precision of 2.4%.

Fish were processed for analysis of mercury in lateral muscle in accordance with U.S. EPA procedures (US EPA 1993). Total fish lengths and wet weights were recorded. Scales were removed from the fish for age analysis. Other details of handling and sample preparation are identical to those described in Rose et al. (1999). A Perkin Elmer Flow Injection Mercury System (FIMS 100) consisting of a Perkin Elmer FIAS 100 flow injection platform interfaced to a mercury measurement system (i.e., mercury cold vapor generator and atomic absorption spectrometer) was used for total mercury analysis and results were expressed on a wet weight concentration basis. Accuracy (i.e., Hg percent recovery from Hg-spiked fish samples) and precision (i.e., Hg relative percent difference among duplicate fish samples) in the analyses of fish samples were 103±9.1% and 4.0±3.8% (means±1 s) respectively. The accuracy of analyses of a mercury fish tissue reference standard consisting of freeze-dried tuna tissue (BCR ref. std #463) was 103±4.7% recovery. Mercury in all laboratory reagent blanks was less than the method detection limit (MDL) of 0.02 mg/kg.

2.4 Data Analysis Methods

Mass accumulation rates in the sediment core were determined using a constant flux: constant sedimentation model to establish ^{210}Pb geochronology of the core (Appleby and Oldfield 1992). Ln excess-^{210}Pb counts were regressed against cumulative mass to derive a mass accumulation rate for the core. Temporal variations in mercury fluxes were calculated from mass accumulation rates and section-specific sediment mercury concentrations.

Bivariate plots of individual fish mercury concentrations versus length for each species for each lake were examined to determine if there was a relationship between these two variables. Tests of parallelism of regression line slopes (Sokal and Rohlf 1995) of muscle mercury concentration versus length were performed on the data for individual lakes.

The recognized confounding effect of size on muscle mercury concentration was controlled for by deriving predicted mercury concentrations for a "standard-sized fish" of each individual of each species. The standard size represents the arithmetic mean fish length over all fish sampled (33.9 cm for LMB and 24.3 cm for YP) in our 1994 state-wide study (Rose et al. 1999). 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 a regression of individual fish mercury concentrations on body lengths for fish from the lake, and then solving the regression equation for the predicted muscle mercury associated with the length of the standard-sized fish. In order to retain individually-based fish data in analyses, thereby getting maximal statistical benefit out of the sample size "n" for the lake, individual fish mercury concentrations were also size-adjusted to the mercury concentration of a standard-sized fish along a line with the same slope as the regression line.

The species size-standardized mercury concentrations were $\log_{10}$-transformed because they did not meet the underlying assumptions for analyses of variance (Sokal and Rohlf 1995). The size-standardized mercury concentrations for YP and LMB for lakes in NE MA were compared against the data for these species from our earlier study of the edible muscle mercury concentrations in LMB and YP in 24 rural, non-source-impacted lakes throughout MA (Rose et al.

1999). Four of the 24 lakes reported in that study (Upper Naukeg, Upper Reservoir, Lake Wampanoag, and Gales Pond) were omitted from this analysis because they were from an area having poorly buffered, low pH (<6) lakes containing fish with high mercury concentrations. We (Rose et al. 1999) and others (Lathrop et al. 1991; Qian et al. 2001) have identified lake water pH as an important predictor variable for fish mercury, with mercury fish concentrations being significantly higher in low pH water bodies. Since none of the lakes in the NE data set were low pH lakes, low pH lakes were omitted from the comparison group.

The species-specific mercury concentration data for each of the lakes in the Rose et al. (1999) study were also size-standardized as described above to facilitate comparison, and lake mean species mercury concentrations calculated. The frequency distribution of these statewide means was then used to identify the 25th and 75th percentile concentrations. These points defined three ranges (<25th percentile, 25–75th percentile, and >75th percentile). For each species, the numbers of lakes from the current study falling into each of the three ranges based on sampling from the rural, non-source-impacted lakes were then tabulated using the means of the species-specific size-standardized mercury values determined for each lake in this study. For YP and LMB, lake mean muscle mercury concentrations for the statewide study were compared against those of the NE MA study using a two-sample *t*-test.

The 24 comparison lakes included in the Rose et al. (1999) paper were sampled in the fall of 1994. The deposition hotspot study area sampling was conducted in the spring of 1999. As mercury concentrations in fish may vary by season (Staveland et al. 1993; Farkas et al. 2003), the data from lakes sampled around the rest of the state in the springs of 1999, 2001 and 2002 have also been compared with the NE lakes data. There were no significant (p=0.01) correlations between mean lake species size-standardized mercury concentrations and pH for the lakes used in these comparisons (r=0.03 for LMB; −0.33 for YP), indicating that pH was not a confounder of fish mercury levels in these data sets.

All statistical evaluations in this study were performed with the Statistica/W, Version 7.0 software package (StatSoft, Tulsa, OK, USA).

3 Results

3.1 Sediment Cores

Both cores were considered dateable based on the depth of penetration, degree of disturbance during collection, appropriate grain size and texture, and the absence of any benthic organisms. The top 2 cm of the Lake Cochichewick cores were non-cohesive and of high porosity. There was no obvious evidence (odor, color change) of a change in redox conditions with depth in either core and both lacked the presence of an obvious oxic layer. The sediment was uniform in color (dark gray) and texture below the unconsolidated surface layer. Small leaves were observed in the 7–10 cm depth sections. Agreement in property concentrations between the mean of the 0–1 and 1–2 cm sections from the first core and the 0–2 cm section of the second core was excellent.

Both the dates of the ^{137}Cs peak and maximum Pb concentration (data not shown) in this core were consistent with those expected from the history of ^{137}Cs bomb fall-out and the time of maximum leaded gas use. A mass sedimentation rate of 6.0±0.8 mg/cm^2/year was determined from the ln excess ^{210}Pb regression with cumulative mass (r^2 of 0.98).

The ^{210}Pb inventory of ~5,800 Bq/m^2 for this core is consistent with the regional mean of 5,700 Bq/m^2 reported by Appleby and Oldfield (1992), and suggests the absence of significant sediment focusing at this coring location. In total, the radioisotope data support the conclusion that the core represents a steady- state sedimentation rate, at least over the last 100 years and perhaps longer. The mass accumulation rate established for this core allows calculation of the flux of mercury and other metals to the sediments over this time period.

The mercury concentration–date profile from the core sections is shown in Fig. 2 and resultant mercury sediment accumulation rates versus time in Fig. 3. Note that the concentrations in the bottom sections of the core, below cumulative mass of ~2 g/cm^2 dry weight, are well above the limit of detection but slightly below or close to the limit of quantitation (10 s of the procedural blank) for the analytical method used in their determination. These concentrations are similar to or lower than concentrations in pre-industrial sections near the bottom of cores from Vermont and New Hampshire described by Kamman and Engstrom (2002). The data suggest a low and

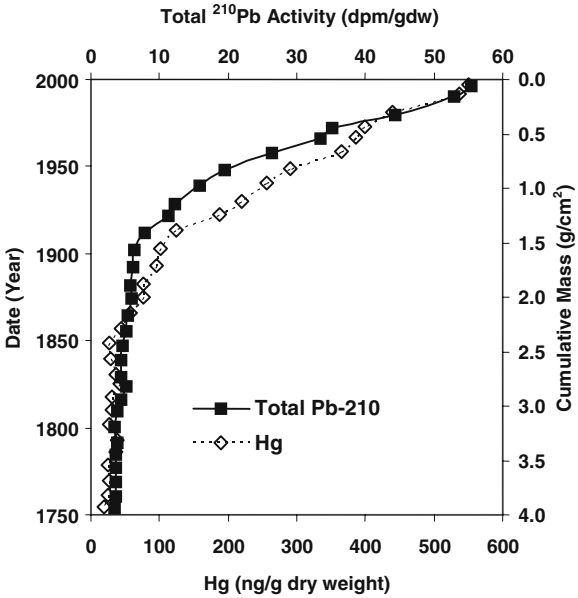

Fig. 2 Sediment core mercury concentrations versus cumulative mass and date as determined from ^{210}Pb geochronology

slowly increasing concentration of mercury before 1900 and then a clear and rapid increase in concentration after that.

Concentrations at the top of the core are over an order of magnitude higher than those observed in the deeper part of the core. The contemporary flux of mercury determined from this core is consistent with an accumulation rate of ~88 μg Hg/m^2/year. The uppermost section of the core analyzed in this work represents a time period of about 4 years or the period from 1997 to date of collection in May 2001. Although there is no evidence for a decrease in mercury concentrations in these recent sections, the temporal resolution at the surface of the core is limited and may mask very recent changes.

3.2 Fish Mercury

YP mercury concentrations for all lakes used in this analysis were not consistently related within lakes to fish length (see composite plot over all lakes, Fig. 4a), with Pearson correlation coefficients between these variables ranging from 0.1 to 0.92 (mean 0.91). We generated basic descriptive statistics for untransformed and size-standardized YP fish muscle mercury concentrations and found that they did not differ appreciably. We therefore chose to use the size-standardized values in our analyses to facilitate comparison with size-standardized values from the Rose et al. (1999) data. LMB mercury concentrations were positively correlated with fish length (Fig. 4b) (correlation coefficients between these two variables for individual species and lakes range from 0.03–0.95, mean 0.70). Slopes of individual lake regression lines of mercury versus length were significantly different between lakes (p=0.05), therefore these data were size-standardized before further analysis. Summary statistics for fish sizes and overall mercury concentrations for each group of fish being compared (NE versus rest of state (1994 from Rose et al. (1999), and 1999–2002) are shown in Table 2.

The mercury concentrations of fish from the NE MA study area were generally greater than those from the rest of the state sampled in 1994 (Fig. 5). This relationship was not confounded by pH differences between size-standardized lake mean mercury concentrations and pH for LMB or YP (r=0.01 and −0.21 respectively, p>0.05). The 25th percentile and 75th percentile size-standardized concentrations for the statewide lakes sampled in 1994 were 0.24 and 0.48 mg/kg for YP and 0.28 and 0.49 mg/kg for standard-sized LMB. The mean size-standardized YP mercury concentrations from eight of the NE MA lakes (Table 1) were in the interquartile range of the rural lake values from 1994; those from the remaining three NE MA lakes were in the upper quartile. None

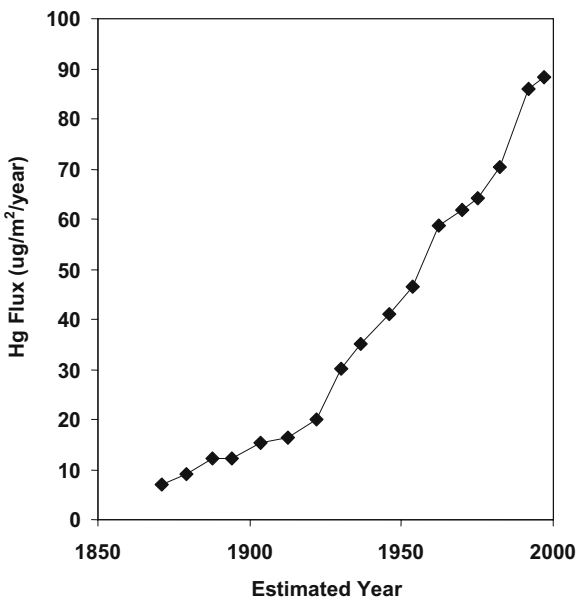

Fig. 3 Mercury fluxes into sediments of Lake Cochichewick over the last 120 years

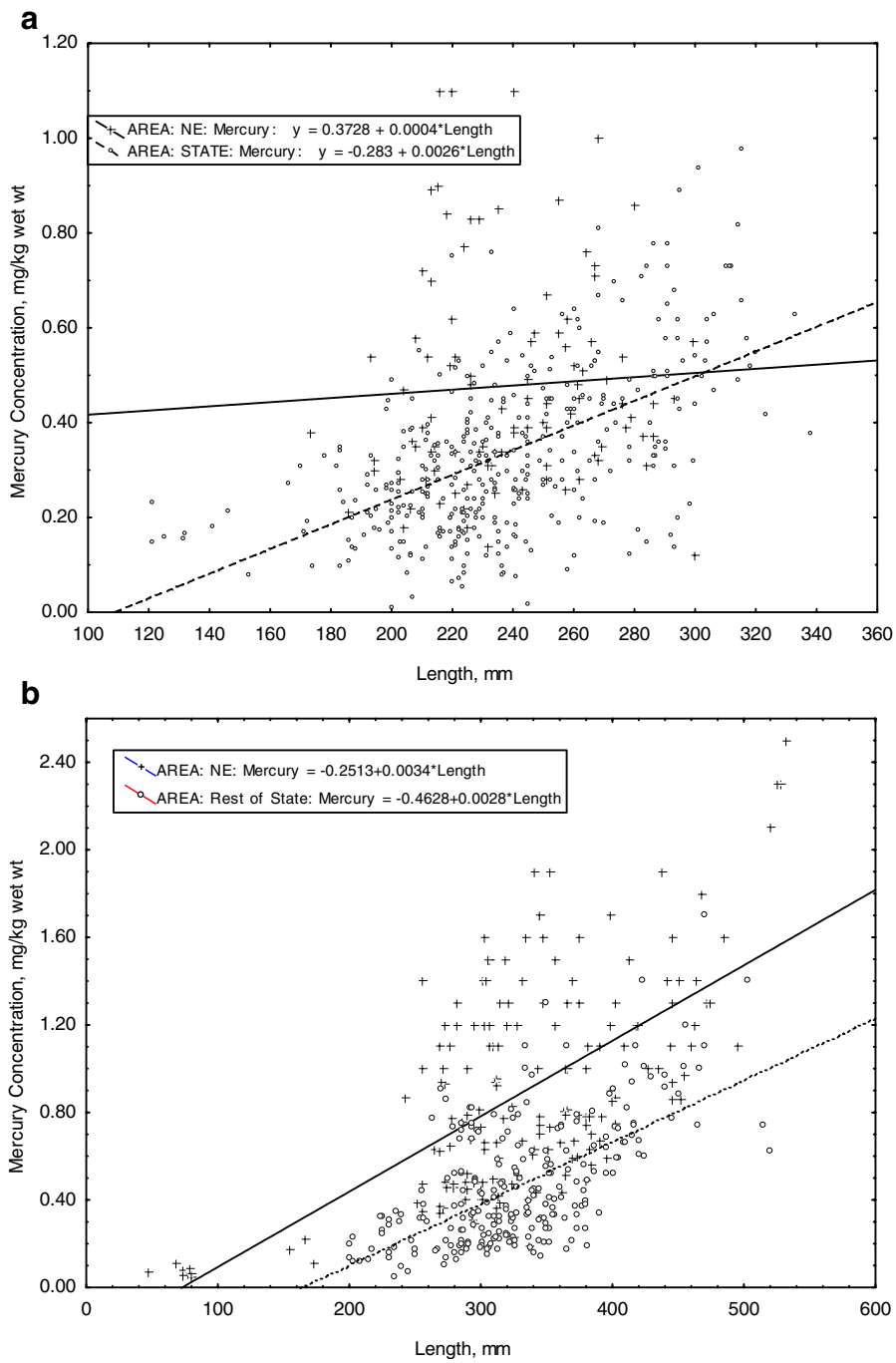

Fig. 4 Collective individual species mercury concentrations versus total length plotted by study area. **a** Yellow perch, **b** largemouth bass

of the northeastern MA lake values were in the lower quartile. The NE MA YP lake mercury concentrations as a group were significantly greater than those of lakes from the rest of the state sampled in 1994 (t= 6.9, 265 df, p=0.01) and in 1999–2002 (t=6.6, 314 df, p=0.01) (Table 2 and Fig. 4a). The overall NE mean was 151 and 52% greater respectively than the 1994 and 1999–2002 means for lakes around the rest of the state.

All of the size-standardized LMB lake mean muscle mercury concentrations from the NE study area lakes were greater than the 75th percentile value

Table 2 Summary statistics for fish populations studied in northeast Massachusetts and the rest of the state

Fish characteristics		LMB			YP		
		NE MA	Rest of state		NE MA	Rest of state	
		1999	1994	1999–2002	1999	1994	1999–2002
N		138	133	105	97	162	219
Total length (cm):	range	24.2–53.2	20.1–51.5	20.3–52.0	17.3–29.9	12.1–30.0	15.3–33.8
	$\bar{x}$±1 s	34.6±6.3	33.8±5.9	31.8±6.3	23.8±2.7	21.9±3.1	25.1±35.7
Total wet wt. (g):	range	152–2392	57–1844	109–2634	52–327	17–348	43–409
	$\bar{x}$±1 s	646±440	608±335	520±446	169±68	118±59	196±85
Raw Hg conc. (mg/kg wet wt.):							
	range	0.34–2.5	0.05–1.10	0.12–1.70	0.14–1.1	0.01–0.75	0.08–0.98
	$\bar{x}$±1 s	0.99±0.45	0.39±0.24	0.55±0.31	0.48±0.22	0.27±0.13	0.39±0.18
Size-standardized Hg conc. (mg/kg wet wt.):							
	$\bar{x}$±1 s	0.93±0.39	0.37±0.14	0.61±0.29	0.49±0.22	0.32±0.15	0.35±0.14

of 0.49 mg/kg from the rural lake values of 1994. As a group, their values were significantly greater than those of fish from lakes from the rest of the state sampled in 1994 (t=16.0, 278 *df*, p=0.01) or 1999–2002 (t=7.5, 250 *df*, p=0.01) (Table 2 and Fig. 4b). The overall NE mean was 53 and 40% greater respectively than the 1994 and 1999–2002 means for lakes around the rest of the state.

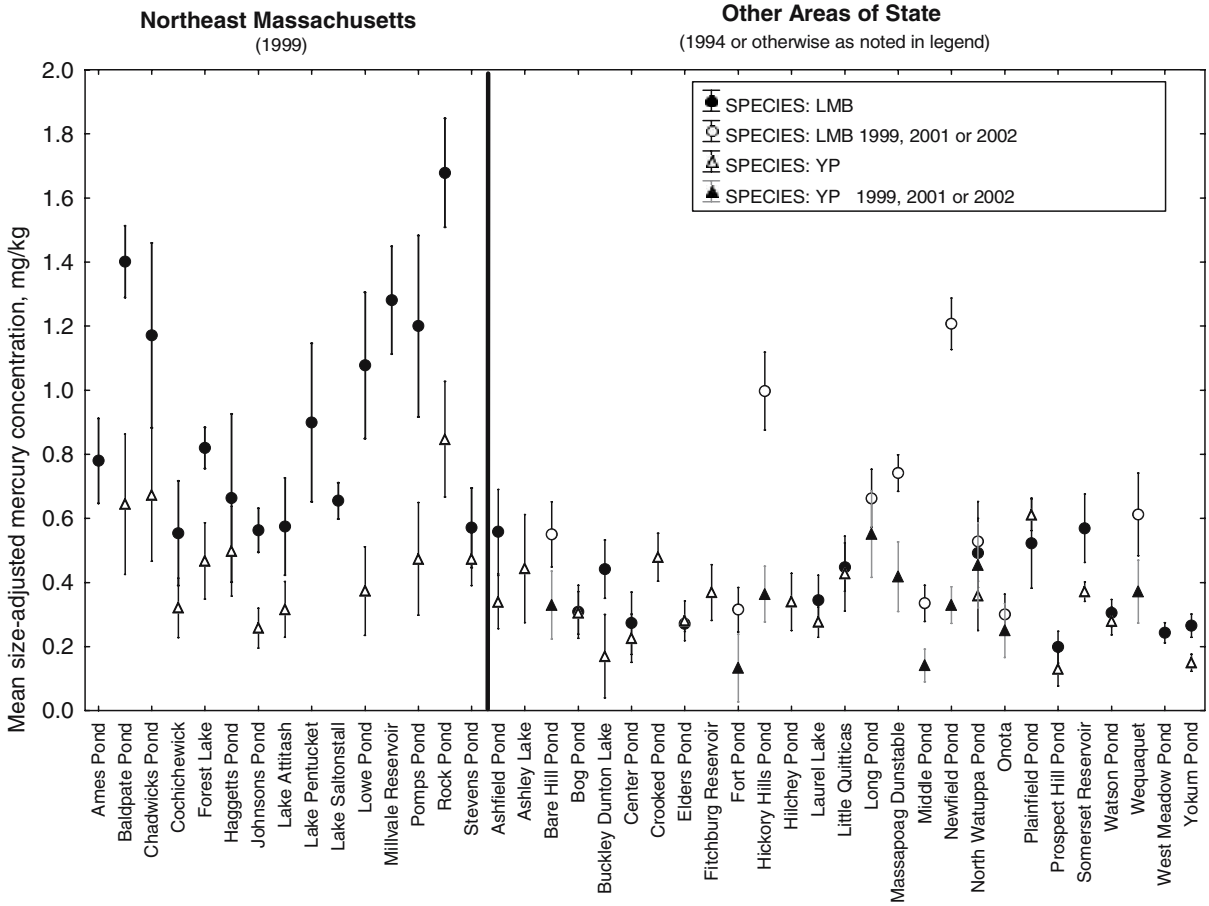

Fig. 5 Mean fish species mercury concentrations (±1s) by location

4 Discussion

This study documents a mercury deposition and fish hotspot area located in NE MA. The designation of this area as a hotspot is supported by four independent lines of evidence: (1) high mercury emissions from local point sources; (2) high predicted atmospheric mercury deposition based on outputs from three deposition-modeling exercises (NESCAUM et al. 1998; Evers et al. 2007; and Graham et al. 2007); (3) elevated mercury accumulation rates in the sediments of Lake Cochichewick, centrally located in the predicted high deposition area; and (4) significantly elevated concentrations of mercury in two species of fish from water bodies in the area.

The Lake Cochichewick sediment core provides a temporally detailed picture of historical mercury deposition to the bottom sediments of one lake in the study area, congruent with the model-predicted high atmospheric mercury deposition in the region. Increases in mercury sediment accumulation rates from pre-industrialization to recent times likely reflect the area's history of industrialization and urbanization dating back to 1835 with the burgeoning of textile mills and associated cities along the Merrimack River (Weible 1991). Potential sources of mercury releases in the area over this period include manufacturing activities, domestic and industrial wastes, combustion of coal for a variety of purposes in the late nineteenth and first half of the twentieth centuries (Smith and Rowan West 1996), and more recently, municipal-level solid waste combustion (Smith and Rowan West 1996; NESCAUM et al. 1998).

The contemporary mercury flux (88 $\mu g/m^2$/year) to the bottom of Lake Cochichewick is consistent with the elevated atmospheric deposition rates predicted for this deposition hotspot area from three models (NESCAUM et al. 1998; Evers et al. 2007; and Graham et al. 2007). This rate is close to the higher range of measured deposition rates between 21 and 83 $\mu g/m^2$/year (mean 42.5) in ten Vermont and New Hampshire lake sediment cores reported by Kamman and Engstrom (2002). However, the two lakes with the highest deposition rates in that study have watershed to lake surface area ratios approximately an order of magnitude greater than that of Lake Cochichewick.

Changes in the mercury accumulation rate in the sediments reflect net changes in the supply of mercury from both atmospheric deposition and runoff from the watershed (Engstrom et al. 1994; Lorey and Driscoll 1999; and Kamman and Engstrom 2002). Highly significant relationships between mercury accumulation rates in sediments and lake watershed areas (WSA) to lake surface areas (LSA) ratios have been reported. The slopes of regression lines fit to accumulation versus area ratios reflect mercury loading rate as a function of watershed area, the intercepts the ambient atmospheric deposition rate, and the ratio of the slope to intercept the fraction of mercury entering the sediments derived from watershed transport (Fig. 6). Engstrom et al. (1994) found a slope of 3.27 for Minnesota and Wisconsin lakes and ponds, with an intercept of 12.5 for the post-industrial period. Lorey and Driscoll (1999) found a slope of 1.93 for Adirondack lakes and ponds and an intercept of 6.9. Kamman and Engstrom (2002) documented a slope of 1.2 for the period 1980 to 1990 and 0.86 for the period 1990–1998 in lakes and ponds sampled in Vermont and New Hampshire, with intercepts of 19 and 30 $\mu g/m^2$/year, respectively.

Using the watershed to surface area ratio for Lake Cochichewick of 5.3 and applying the above range of slope factors produces contemporary (1997) watershed fluxes ranging from 5–17 $\mu g/m^2$/year for this lake and would require a direct atmospheric flux of 71–83 $\mu g/m^2$/year to the lake to sustain the total Hg sediment accumulation rate.

Differences in the slope factors such as those noted above reflect changes in regional source strength along with potential variations in biogeochemical processes influencing transport through the watershed. Much higher slope factors would result in a much stronger influence of watershed contributions. For example, a slope factor of 10 would result in a direct atmospheric deposition of 27 $\mu g/m^2$/year to Lake Cochichewick and a watershed contribution of 61 $\mu g/m^2$/year or ~70% of the total Hg flux to the sediments. Under these conditions, the response of lake sediment accumulation Hg fluxes to decreasing atmospheric fluxes would be potentially buffered by ongoing watershed contributions.

Assessment of the slope factor using lakes with different WSA:LSA ratios in the same region may be useful for determining the relative contributions of the two sources. Even then, the use of this approach requires relatively uniform deposition in a region, and locally influenced lakes would appear as outliers. The mercury flux associated with the watershed/lake area

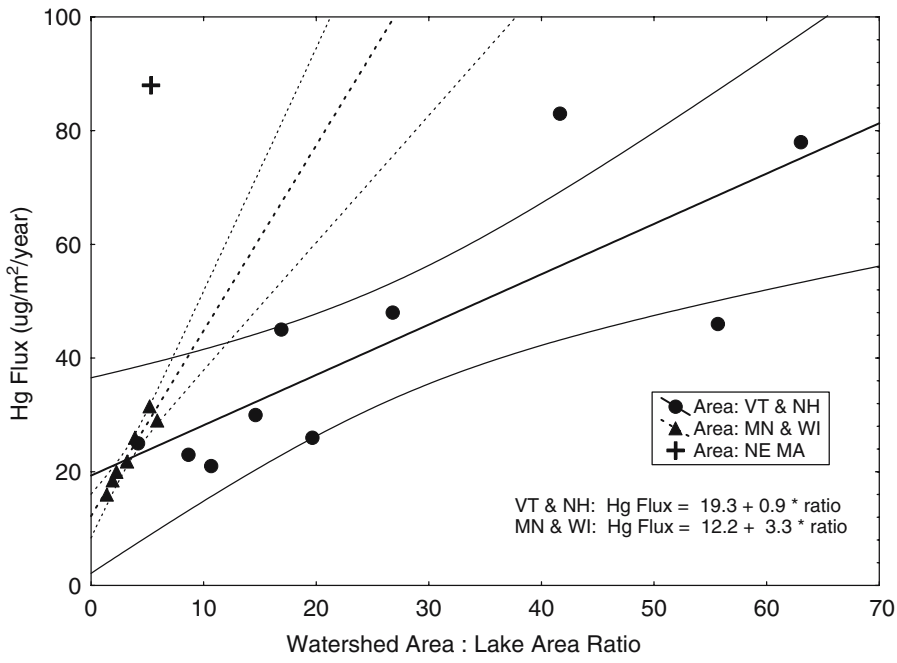

Fig. 6 Total mercury flux of individual lakes versus watershed/lake area ratios. Data from Lake Cochichewick, Engstrom et al. 1994 and Kamman and Engstrom (2002). *VT* Vermont, *NH* New Hampshire, *MN* Minnesota, *WI* Wisconsin

ratio for Lake Cochichewick is well above the upper 95% confidence bounds of the linear regression lines fit to the two data sets of Engstrom et al. (1994) and Kamman and Engstrom (2002) (Fig. 6). This reinforces the point that the high mercury flux calculated for recent years for this sediment core from NE MA is not just a reflection of a larger watershed area in relation to the lake surface area. As there are no known direct mercury sources within the watershed, we thus interpret the high mercury fluxes in the Lake Cochichewick core to reflect local emission source inputs superimposed on a broader regional atmospheric deposition flux of mercury, as observed in other areas in proximity to known emission sources (Lindberg and Stratton 1998; Chillrud et al. 1999; Driscoll et al. 2007; Evers et al. 2007).

Other sediment cores from lakes in the northeastern US (Lorey and Driscoll 1999; Kamman and Engstrom 2002; and Varekamp et al. 2003) indicate mercury fluxes to those lakes decreased beginning in the 1980s to1990s. The lack of discernable decreases in mercury concentrations and flux in the Lake Cochichewick core during this period is consistent with locally elevated atmospheric emissions from nearby emissions sources, which would serve to mask any more regional decrease in atmospheric fluxes as deduced from these and other core studies.

High concentrations of mercury were also observed in fish muscle from lakes in the study area. LMB and YP from the study area had muscle mercury concentrations (size-standardized) on average from 1.5- to 2.5-fold and approximately 1.5 fold, respectively, greater (p=0.01) than values from more rural, non-source-impacted regions of the state sampled in 1994, 1999, 2001, and 2002 (Table 2).

The results from other studies on LMB and YP, summarized in Fig. 7, further support the conclusion that LMB muscle mercury concentrations in northeast MA are high. The levels of mercury in LMB muscle in Maryland lakes were less than those seen in NE MA (Pinkney et al. 1997). In a nationwide dataset including 20 U.S watershed basins (Brumbaugh et al. 2001), the mean muscle mercury concentration in LMB was 0.51 mg/kg in year class-3 fish (n=50). The corresponding mean (±1 s) value for this study's year class-3 fish was 0.84±0.35 (n=33) mg/kg. The YP mean muscle mercury concentrations in non-source impacted lakes in Michigan and Wisconsin (Grieb et al. 1990) and New York State (Simonin et al. 1994) were 0.25 and 0.36 mg/kg, respectively for year class-4 fish. The mean (±1 s) mercury muscle concentration of year class-4 YP in our NE MA data set was 0.47±0.23 mg/kg, considerably higher than the levels reported in these other studies.

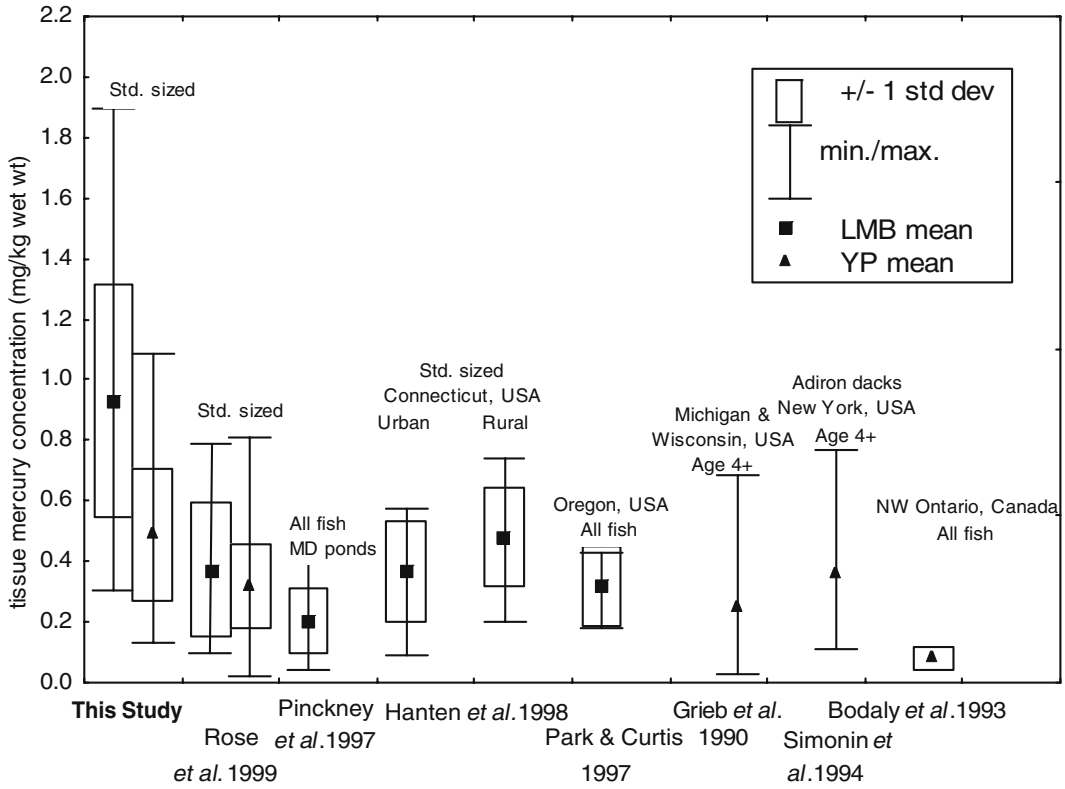

Fig. 7 Comparative northeast Massachusetts mean species muscle mercury concentrations versus regional and national LMB and YP muscle mercury concentrations. Means±1 s, ranges

Fish size and age, inter-lake differences, year-to-year variation, and seasonal variation could potentially influence the levels of mercury in fish muscle in this and other comparative studies, and it is important to control for these where possible and consider their possible influences on the data. In addition, the complex chemistry of mercury in such systems is not yet fully understood, but may lead to distinctly different availability of mercury in otherwise similar lakes.

Older, larger, predatory fish such as LMB tend to accumulate more mercury as they age (Rose et al. 1999). Data from the present study indicate that mercury concentrations in the smallest and largest fish from the same location at the same time may span up to one order of magnitude (Table 2; Fig. 8). The data in this study were normalized to the length of a standard fish size to control for this source of variance.

Although it is not possible to fully account for variability attributable to inter-lake differences (e.g., potentially due to differences in food chain length, pH, productivity, etc.), one significant variable was addressed in this study through the exclusion of lakes that had unusually low pH levels from the in-state data sets (no significant (p=0.05) correlations between lake mean mercury concentrations and pH for the remainder of lakes). Additionally, the use of multiple lakes and multiple comparative data sets minimizes the probability that observed geographic differences in fish mercury concentrations are simply due to unique inter-lake differences.

Inter-annual variation may also impact dataset comparisons and can result from changes in internal process rates such as mercury methylation rates, as well as biological and statistical variation. Thus, using data from different years may introduce uncertainty into geographic comparisons of fish mercury levels. The degree of inter-annual variation observed in other studies varies. Little year-to-year variation was seen in LMB, northern pike, walleye and cisco muscle mercury concentrations over a three year study period in remote, northwestern Ontario lakes (Bodaly et al. 1993). Park and Curtis (1997) recorded substantial inter-annual variation, but some of their variation could have been due to seasonal differences in mercury levels because

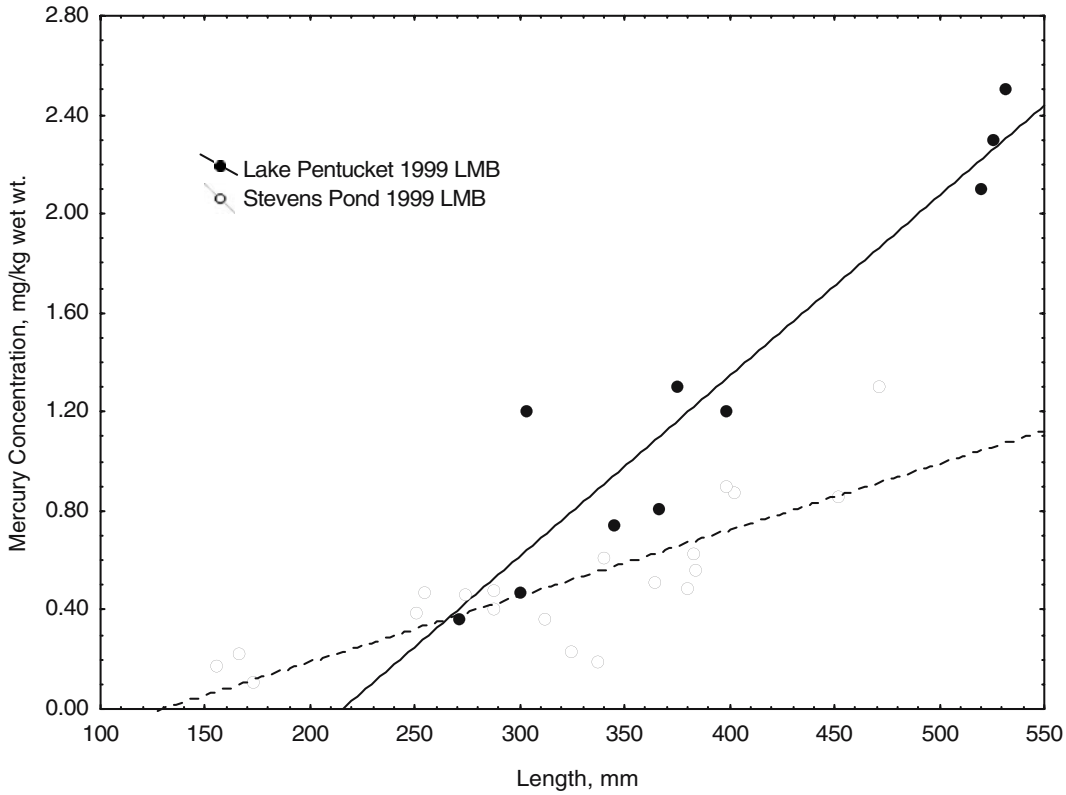

Fig. 8 Tissue mercury concentrations for LMB from NE MA Lakes Pentucket and Stevens Pond, 1999

fish were sampled at different times (June–November) in the different years. Although we have no estimate of the degree of inter-annual variation in our analysis, the consistent finding of elevated fish mercury levels in the study area compared to the two other in-state sets of data (one collected in 1994, and the other between 1999 and 2002) and for LMB in the out-of-state data sets (collected during different years), suggest that the higher mercury levels in the fish from the study area are unlikely to be attributable to inter-annual variation.

Seasonal variation in fish tissue mercury concentrations is a potentially significant component of the variance in the comparison between the study area (April–May sampling) and one of the primary data sets (October, (Rose et al. 1999)) being compared in this study. The physiological and reproductive status of fish are closely tied to annual temperature and photoperiod changes. These status changes with respect to interpreting muscle mercury concentration data may be important (Slotton et al. 1995; Cizdziel et al. 2002; Farkas et al. 2003). Seasonal differences in fish muscle mercury concentrations have been documented by Staveland et al. (1993), Cizdziel et al. (2002), Farkas et al. (2003), and Paller et al. (2004). In contrast, no seasonal changes in fish muscle mercury concentrations were observed by Bidwell and Heath (1993), Park and Curtis (1997) or Farkas et al. (2000).

In order to make an appropriate comparison without this source of potential confounding, the deposition hotspot results were also compared with data from MA water bodies in non-source impacted areas sampled in the spring over the period of 1999–2002 (Tables 1 and 2). Mercury concentrations in fish from the hotspot lakes are elevated when compared to those from lakes located elsewhere in MA even when sampling was conducted in the same season.

These fish tissue results, when viewed collectively with the mercury emissions, deposition modeling, and sediment core data discussed above, provide a strong case that the study region constitutes a mercury deposition and biological hotspot, associated with local mercury emission sources. This more detailed examination of a smaller geographic area with an additional important species of fish (LMB) supports the broader regional conclusions of Evers et al. (2007) based on YP and common loons (*Gavia immer*) as

indicator species. The analyses herein extend their observation by demonstrating a statistically significant difference in YP and LMB fish muscle mercury concentrations between lakes in a mercury emissions and depositional hotspot area compared to similarly sampled lakes and ponds elsewhere in MA. In addition the findings are consistent with those reported for the unique Everglades ecosystem in Southern Florida (Atkeson et al. 2003), where elevated fish tissue mercury levels were also associated with local point source emissions, and they extend concern over mercury emission point source impacts to temperate water bodies.

5 Conclusions

The study assembled several pieces of information supporting designation of a ~20×26 km area in northeastern Massachusetts as a mercury atmospheric deposition and fish tissue hotspot likely attributable in significant part to local emissions sources:

- Sediment coring from one lake in the study area showed that during the twentieth century there was a rapid increase in mercury concentrations in the core with maximum mercury accumulation rate of ~88 μg Hg/m^2/year in the late 1990s. This level was consistent with projected atmospheric deposition rates from three independent models and is significantly elevated when compared to accumulation rates in other lakes and ponds reported in the literature.
- Mercury concentrations in YP and LMB from 15 lakes in the study area were significantly greater (~1.5- to 2.5-fold) than those of fish from the rest of the state, after controlling for the potential confounding effects of fish size and low pH waters in some lakes in the state. Concentrations were also notably high when compared to other data sets from across the nation.
- Notably, the results also have important policy implications for controlling mercury emissions in that they represent an extension of similar findings about the impacts of local emissions sources from subtropical systems to temperate water bodies. These findings also raise important national questions in the United States with respect to proposed pollution trading schemes targeting mercury emission sources, since local sources may contribute to or perpetuate "hotspots".

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References

Appleby, P. G., & Oldfield, F. (1992). Application of lead-210 to sedimentation studies. In M. Ivanovich, & R. S. Harmon (Eds.), *Uranium-series disequilibrium: Applications to earth, marine, and environmental sciences* (pp. 731–778). New York: Oxford University Press.

Atkeson, T., Axelrad, D., Pollma, C., & Keeler, G. (2003). *Integrating atmospheric mercury deposition and aquatic cycling in the Florida Everglades: An approach for conducting a total maximum daily load analysis for an atmospherically derived pollutant*. Report Prepared for Florida Department of Environmental Protection.

Bidwell, J. R., & Heath, A. G. (1993). An *in situ* study of rock bass (*Ambloplites rupestris*) physiology: Effect of season and mercury contamination. *Hydrobiologia*, *264*, 137–152.

Bodaly, R. A., Rudd, J. W. M., Fudge, R. J. P., & Kelly, C. A. (1993). Mercury concentrations in fish related to size of remote Canadian Shield lakes. *Canadian Journal of Fisheries and Aquatic Sciences*, *50*, 980–987.

Boening, D. W. (2000). Ecological effects, transport, and fate of mercury: A general review. *Chemosphere*, *40*, 1335–1351.

Brumbaugh, W. G., Krabbenhoft, D. P., Helsel, D. R., Wiener, J. G., & Echols, K. R. (2001). A national pilot study of mercury contamination of aquatic ecosystems along muliple gradients: Bioaccumulation in fishes. Biological Science Report USGS/BRD/BSR-2001-0009. U.S. Department of the Interior, U.S. Geological Survey.

Bullock, O. R., & Brehme, K. A. (2002). Atmospheric mercury simulation using the CMAQ Model: Formulation description and analysis of wet deposition results. *Atmospheric Environment*, *36*, 2135–2146.

Chillrud, S. N., Bopp, R. F., Simpson, H. J., Ross, J. M., Shuster, E. L., Chaky, D. A., et al. (1999). Twentieth century atmospheric metal fluxes into Central Park Lake, New York City. *Environmental Science & Technology*, *33*, 657–662.

Cizdziel, J., Hinners, T., Cross, C., & Pollard, J. (2003). Distribution of mercury in the tissues of five species of freshwater fish from Lake Mead, USA. *Journal of Environmental Monitoring*, *5*, 802–807.

Cizdziel, J. V., Hinners, T. A., Pollard, J. E., Heithmar, E. M., & Cross, C. L. (2002). Mercury concentrations in fish from Lake Mead, USA, related to fish size, condition, trophic

level, location, and consumption risk. *Archives of Environmental Contamination and Toxicology*, *43*, 309–317.

Driscoll, C. T., Han, Y.-J., Chen, C. Y., Evers, D. C., Lambert, K. F., Holsen, T. M., et al. (2007). Mercury contamination in forest and freshwater ecosystems in the northeastern United States. *Bioscience*, *57*, 17–28.

Dvonch, J. T., Keeler, G. J., & Marsik, F. J. (2005). The influence of meteorological conditions on the wet deposition of mercury in southern Florida. *Journal of Applied Meteorology*, *44*, 1421–1435.

Engstrom, D. R., & Swain, E. B. (1997). Recent declines in atmospheric mercury deposition in the upper Midwest. *Environmental Science & Technology*, *31*, 960–967.

Engstrom, D. R., Swain, E. B., Henning, T. A., Brigham, M. E., & Brezonik, P. L. (1994). Atmospheric mercury deposition to lakes and watersheds—a quantitative reconstruction from multiple sediment cores. In L. A. Baker (Ed.), *Environmental chemistry of lakes and reservoirs* (pp. 33–66). Washington, DC: American Chemical Society.

Evers, D. C., Han, Y.-J., Driscoll, C. T., Kamman, N. C., Goodale, M. W., Lambert, K. F., et al. (2007). Identification and evaluation of biological hotspots of mercury in the northeastern U.S. and eastern Canada. *Bioscience*, *57*, 1–15.

Farkas, A., Salanki, J., & Specziar, A. (2003). Age- and size-specific patterns of heavy metals in the organs of freshwater fish *Abramis brama* L. populating a low-contaminated site. *Water Research*, *37*, 959–964.

Farkas, A., Salánki, J., & Varanka, I. (2000). Heavy metal concentrations in fish of Lake Balaton. *Lakes & Reservoirs: Research & Management*, *5*, 271–280.

Frazier, B., Wiener, J., & Engstrom, D. (2000). Stratigraphy and historic accumulation of mercury in recent depositional sediments in the Sudbury River, Massachusetts, U.S.A. *Canadian Journal of Fisheries and Aquatic Sciences*, *57*, 1062–1072.

Goodman, G. T., & Roberts, T. M. (1971). Plants and soils as indicators of metals in the air. *Nature*, *231*, 287–292.

Graham, J., Miller, P., Savelli, E., & Woo, J.-H. (2007). Modeling mercury in the northeast United States (draft technical report), Northeast States for Coordinated Air Use Management (NESCAUM) Boston, MA.

Grieb, T. M., Driscoll, C. T., Gloss, S. P., Schofield, C. L., Bowie, G. L., & Porcella, D. B. (1990). Factors affecting mercury accumulation in fish in the upper Michigan peninsula. *Environmental Toxicology and Chemistry*, *9*, 919–930.

Haines, T., May, T., Finlayson, R., & Mierzykowski, S. (2003). Factors affecting food chain transfer of mercury in the vicinity of the Nyanza site, Sudbury River, Massachusetts. *Environmental Monitoring and Assessment*, *86*, 211–232.

Hanten, R. P., Neumann, R. M., & Ward, S. M. (1998). Relationships between concentrations of mercury in large-mouth bass and physical and chemical characteristics of Connecticut lakes. *Transactions of the American Fisheries Society*, *127*, 807–818.

Hartel, K. E., Halliwell, D. B., & Launer, A. E. (2002). *Inland fishes of Massachusetts*. Lincoln, MA: Massachusetts Audubon Society.

Henny, C. J., Hill, E. F., Hoffman, D. J., Spalding, M. G., & Grove, R. A. (2002). Nineteenth Century mercury: Hazard to wading birds and cormorants of the Carson River, Nevada. *Ecotoxicology*, *11*, 213–231.

Ion, J., Delafontaine, Y., Dumont, P., & Lapierre, L. (1997). Contaminant levels in St. Lawrence River yellow perch (*Perca flavescens*)—spatial variation and implications for monitoring. *Canadian Journal of Fisheries and Aquatic Sciences*, *54*, 2930–2946.

Kamman, N. C., Burgess, N. M., Driscoll, C. T., Simonin, H. A., Goodale, W., Linehan, J., et al. (2005). Mercury in freshwater fish of northeast North America—a geographic perspective based on fish tissue monitoring databases. *Ecotoxicology*, *14*, 163–180.

Kamman, N. C., & Engstrom, D. R. (2002). Historical and present fluxes of mercury to Vermont and New Hampshire lakes inferred from ^{210}Pb dated sediment cores. *Atmospheric Environment*, *36*, 1599–1609.

Lathrop, R. C., Rasmussen, P. W., & Knauer, D. R. (1991). Mercury concentrations in walleyes from Wisconsin (USA) lakes. *Water Air and Soil Pollution*, *56*, 295–307.

Lindberg, S. E., & Stratton, W. J. (1998). Atmospheric mercury speciation: Concentrations and behavior of reactive gaseous mercury in ambient air. *Environmental Science & Technology*, *32*, 49–57.

Lorey, P., & Driscoll, C. T. (1999). Historical trends of mercury deposition in Adirondack Lakes. *Environmental Science & Technology*, *33*, 718–722.

MA DPH. (2001). Consumer Advisories on Fish Consumption and Mercury Contamination. Massachusetts Department of Public Health. Center for Environmental Health. Boston, MA. Retrieved November 20, 2007 from MA DPH Web site: http://www.mass.gov/?pageID=eohhs2terminal&L=7&L0=Home&L1=Consumer&L2=Community+Health+and+Safety&L3=Environmental+Health&L4=Environmental+Exposure+Topics&L5=Fish+and+Wildlife&L6=Fish+Consumption+Advisories&sid=Eeohhs2&b=terminalcontent&f=dph_environmental_c_fish_consumption_advisory&csid=Eeohhs2.

MA DPH. (2007). Public Health Fish Consumption Advisory. Massachusetts Department of Public Health. Boston. Retrieved November 20, 2007 from MA DPH Web site: http://db.state.ma.us/dph/fishadvisory/.

Mergler, D., Anderson, H. A., Chan, L. H. M., Mahaffey, K. R., Murray, M., Sakamoto, M., et al. (2007). Methylmercury exposure and health effects in humans: A worldwide concern. *Ambio*, *36*, 3–11.

NESCAUM, NEWMOA, NEIWPCC, EMAN (1998). Mercury study. A framework for action. (Boston, MA: Northeast States for Coordinated Air Use Management (NESCAUM), Northeast Waste Management Officials Association (NEWMOA), New England Interstate Water Pollution Control Commission (NEIWPCC), Canadian Ecological Monitoring and Assessment Network(EMAN)).

Olivero, J., & Solano, B. (1998). Mercury in environmental samples from a waterbody contaminated by gold mining in Colombia, South America. *Science of the Total Environment*, *217*, 83–89.

Paller, M. H., Bowers, J. A., Littrell, J. W., & Guanlao, A. V. (2004). Influences on mercury bioaccumulation factors for the Savannah River. *Archives of Environmental Contamination*, *46*, 236–243.

Paller, M. H., & Littrell, J. W. (2007). Long-term changes in mercury concentrations in fish from the middle Savannah River. *Science of the Total Environment*, *382*, 375–382.

Park, J. G., & Curtis, L. R. (1997). Mercury distribution in sediments and bioaccumulation by fish in 2 Oregon reservoirs—point-source and nonpoint-source impacted systems. *Archives of Environmental Contamination*, *33*, 423–429.

Pedersen, T. F., Malcolm, S. J., & Sholkovitz, E. R. (1985). A lightweight gravity corer for undisturbed sampling of soft sediments. *Canadian Journal of Earth Sciences*, *22*, 133–135.

Pinkney, A. E., Logan, D. T., & Wilson, H. T. (1997). Mercury concentrations in pond fish in relation to a coal-fired power-plant. *Archives of Environmental Contamination*, *33*, 222–229.

Qian, S. S., Warren-Hicks, W., Keating, J., Moore, D. R. J., & Teed, R. S. (2001). A predictive model of mercury fish tissue concentrations for the southeastern United States. *Environmental Science & Technology*, *35*, 941–947.

Rencz, A. N., O'Driscoll, N. J., Hall, G. E., Peron, T., Telmer, K., & Burgess, N. M. (2003). Spatial variation and correlations of mercury levels in the terrestrial and aquatic components of a wetland dominated ecosystem: Kejimkujik Park, Nova Scotia, Canada. *Water, Air and Soil Pollution*, *143*, 271–288.

Riisgard, H. U., & Famme, P. B. (1988). Distribution and mobility of organic and inorganic mercury in flounder, *Platichthys flesus*, from a chronically polluted area. *Toxicology and Environmental Chemistry*, *16*, 219–228.

Rose, J., Hutcheson, M. S., West, C. R., Pancorbo, O., Hulme, K., Cooperman, A., et al. (1999). Fish mercury distribution in Massachusetts, USA lakes. *Environmental Toxicology and Chemistry*, *18*, 1370–1379.

Saiki, M. K., Martin, B. A., May, T. W., & Alpers, C. N. (2005). Total mercury concentrations in fillets of bluegill, redear sunfish, largemouth bass, and other fishes from Lake Natoma, Sacramento County, California. *California Fish and Game*, *91*, 193–206.

Scott, W. B. C. E. J., & Crossman, E. J. (1973). *Freshwater fishes of Canada. Bulletin 184*. Ottawa: Fisheries Research Board of Canada.

Simonin, H. A., Gloss, S. P., Driscoll, C. T., Schofield, C. L., Kretser, W. A., Karcher, R. W., et al. (1994). Mercury in yellow perch from Adirondack drainage lakes. In C. J. Watras, & J. W. Huckabee (Eds.), *Mercury pollution: Integration and synthesis* (pp. 457–468). Boca Raton, FL: Lewis Publishers.

Slotton, D. G., Reuter, J. E., & Goldman, C. R. (1995). Mercury uptake patterns of biota in a seasonally anoxic northern California reservoir. *Water Air and Soil Pollution*, *80*, 841–850.

Smith, C. M., & Rowan West, C. (1996). *Mercury in Massachusetts: An evaluation of sources, emissions, impacts and controls*. Boston, MA: Massachusetts Department of Environmental Protection, Office of Research and Standards.

Sokal, R. R., & Rohlf, F. J. (1995). *Biometry: The principles and practice of statistics in biological research*. New York: Freeman.

Staveland, G., Marthinsen, I., Norheim, G., & Julshamn, K. (1993). Levels of environmental pollutants in flounder (*Platichthys flesus* L) and cod (*Gadus morhua* L) caught in the waterway of Glomma, Norway. 2. Mercury and arsenic. *Archives of Environmental Contamination and Toxicology*, *24*, 187–193.

US EPA. (1993). *Methods for the determination of inorganic substances in environmental samples. EPA 600/R-93–100*. Washington, D.C.: US Environmental Protection Agency, Office of Research and Development.

US EPA. (1997). *Mercury study report to congress. Vol. 1–8 EPA-452/R-97–005*. Washington, DC: US Environmental Protection Agency, Office of Air Quality Planning and Standards and Office of Research and Development.

VanArsdale, A., Weiss, J., Keeler, G., Miller, E., Boulet, G., Brulotte, R., & Poissant, L. (2005). Patterns of mercury deposition and concentration in northeastern North America (1996–2002). *Ecotoxicology*, *14*, 37–52.

Varekamp, J. C., Kreulen, B., ten Brink, B., & Mecray, E. L. (2003). Mercury contamination chronologies from Connecticut wetlands and Long Island Sound sediments. *Environmental Geology*, *43*, 268–282.

Wallace, G. T., Krahforst, C., Pitts, L., Shine, J., Studer, M., & Bollinger, C. (1991). *Assessment of the chemical composition of the Fox Point CSO effluent and associated subtidal and intertidal environments: Analysis of CSO effluents, surface sediments and water column samples for trace metals prior to CSO modification*. Final Report to Massachusetts Department of Environmental Protection, Executive Office of Environmental Affairs, Commonwealth of Massachusetts. Boston, MA: Environmental Sciences Program, University of Massachusetts Boston.

Wallace, G. T., Oktay, S., Pala, F., Ferraro, M., Gnatek, M., Luce, D., et al. (2004). *Determination of recent inputs of mercury to lakes/ponds in the Merrimack Valley using sediment cores: A feasibility study*. Final report for Massachusetts Department of Environmental Protection. Boston, MA from University of Massachusetts Boston, Department of Coastal and Ocean Sciences. Boston, MA.

Weible, R. (1991). *The continuing revolution: A history of Lowell, Massachusetts*. Lowell, MA: Lowell Historical Society.