

Environmental Toxicology

TEMPORAL AND GEOGRAPHIC TRENDS IN MERCURY CONCENTRATIONS IN MUSCLE TISSUE IN FIVE SPECIES OF HUDSON RIVER, USA, FISH

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(Received 7 August 2007; Accepted 7 January 2008)

Abstract—We analyzed a New York (USA) state database of mercury concentrations in muscle tissue for five species of fish (striped bass, yellow perch, largemouth bass, smallmouth bass, and carp) over a range of locations in the Hudson River (USA) between 1970 and 2004. We used regression models to discern temporal and geographic change in the fish while controlling for a positive correlation between mercury concentration and body mass. Mercury concentrations significantly increased in fish from New York Harbor waters to the mid-Hudson River. Striped bass and yellow perch showed a shallower increase in mercury concentration with river mile than did carp, largemouth bass, and smallmouth bass. Mercury concentrations declined over the 34-year period. These results imply that a geographically restricted source of mercury may be spread throughout the watershed by toxin-laden dispersing species. The increase of mercury toward the north may relate to a point source in the mid-Hudson River, or it may indicate mercury released from the Adirondack watershed. The decline of mercury over three decades corresponds to a reduction of various inputs in the region. The temporal and geographic pattern of mercury in sediments corresponds to the geographic trend of mercury in fish.

Keywords—Mercury Fish Hudson River Geographic trend Temporal trend

INTRODUCTION

Methylmercury is the primary form of mercury found in fish [1], and mercury toxicity causes neurological disorders [2], particularly in the human fetus [3]. Mercury also causes myocardial infarction in humans [4]. Mercury concentrations in lake and river fish often exceed guidelines for human consumption, and this has caused advisories against consumption of fish in Sweden, Canada, and the northern and northeastern United States [5–7]. Certain human populations are especially at risk [8]. For example, anglers on lower Hudson River (USA) who consume their catch have significantly higher concentrations of mercury compared with nonanglers [9]. Mercury forms vary in their toxicities. For instance, mercury in fish may exist as methylmercury cysteine, and cells might convert this to methylmercury chloride, which is more toxic [10].

Human intake of mercury mainly takes the following pathway: Atmospheric mercury deposition on watersheds, entry into watershed soil and sediments, dissolved methylmercury in water, methylmercury in fish through a bioaccumulation factor from prey in water, and then human consumption of fish [11]. Atmospheric deposition of mercury produces the inorganic form [12], so microbial activity is necessary for conversion to methylmercury. Mercury enters aquatic systems, such as rivers, and conversion to methylmercury usually occurs in sediments, facilitated by microbial processes, such as sulfate reduction [13,14]. Acid deposition, which is typical of the Adirondack watershed, may enhance methylmercury for-

mation [15]. As long as no strong proximal inorganic mercury source is present, methylmercury is the most common form of mercury in fish, especially carnivorous species [16].

Point sources of mercury (e.g., factory outfalls into rivers) that enter sediments also may enter the pathway [17]. In industrialized rivers, such as the Penobscot River in Maine (USA) mercury in sediment originates mainly from a factory point source [18]. By burning less refuse and eliminating other point sources, mercury concentrations have declined in the sediments of New York–New Jersey Harbor (USA) over the past few decades [17].

Our objective was to use a large database of mercury measures from five common species of Hudson River estuarine fish to determine temporal and spatial trends in the mid and lower Hudson River. We tested two null hypotheses. The first was that mercury has not changed in concentration in common fish species over the past few decades; the second was that mercury concentrations do not significantly change with river mile in the Hudson River, suggesting that no specific, geographically localized source of contamination exists within the Hudson River. We hoped that trends would be sufficiently repeatable among species to provide a robust test of these hypotheses. We also tested the hypothesis that no differences among species existed in the variation of mercury with Hudson River location or with time.

The bivariate nature of time and space is complicated by the common, strongly positive relationship between mercury concentration and body mass (BM) [19]. In some cases, within a single year, growth rate (and, therefore, size) may decrease with increasing mercury concentrations, particularly within rapidly growing individual fish [20]. We therefore adopted a multivariate statistical approach to test these hypotheses, especially to correct for the effect of body size.

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Contribution 1160 from the Graduate Program of Ecology and Evolution, Stony Brook University.

Published on the Web 2/11/2008.

MATERIALS AND METHODS

Data collection

All data were obtained from the New York State Department of Environmental Conservation's Toxic Materials Database. Data was provided in the form of a Microsoft (Redmond, WA, USA) FoxPro® file, which included data for total mercury concentrations in fish fillets, body length, BM, year of collection (YR), and exact location of collection (Hudson River mile [HRM]), defined as the number of miles north of The Battery, at the southern tip of Manhattan Island, New York City, NY, USA (Fig. 1). Mercury was measured in fillets of five species of fish: Striped bass (SB; *Morone saxatilis*), yellow perch (YP; *Perca flavescens*), largemouth bass (LMB; *Micropterus salmoides*), smallmouth bass (SMB; *Micropterus dolomieu*), and carp (CP; *Cyprinus carpio*). The database also included other information, such as indicators of the laboratories performing mercury analyses. Data for the five species were available in varying degrees from 1970 to 2005 (SB, 1970–1998; YP, 1983–2005; LMB, 1970–2004; SMB, 1970–2004; CP, 1970–1998). Varying ranges of HRMs also were available (SB, 10–52; YP, 150–220; LMB, 50–310; SMB, 90–275; CP, 50–220). The raw data are available online at <http://life.bio.sunysb.edu/marinebio/hudsonfishhg.html>.

Data analysis

We used multiple-regression techniques [21] in which total mercury concentration (C_{Hg}) in fillets was the dependent variable and species identity (SPP), BM, YR, and HRM were the independent variables. A total analysis was done for all species using the following general model, which calculates regression coefficients (B_i) and an error term (e):

$$C_{\text{Hg}} = B_0 + B_1(\text{SPP}) + B_2(\text{YR}) + B_3(\text{BM}) + B_4(\text{HRM}) + e$$

We then used a forward stepwise-regression model [21] to analyze the relationship between mercury concentration and YR, BM, and HRM for each species individually. In this approach, independent variables (YR, BM, and HRM) were added sequentially and incorporated into the final model if they were significant ($p < 0.05$). This approach gives us an order of importance for the independent variables and also allows the calculation of a partial r^2 , which tells us how much of the total variance each variable explains.

Most of the present paper involves analyses of the entire data set, including all localities and years. In a few cases, we had sufficient data for a single species in a given year to show spatial trends in mercury concentration. These are presented by plotting the residuals from the Hg concentration expected for a given BM as a function of HRM.

All calculations were done with the aid of JMP software (Ver 5.0; SAS Institute, Research Triangle, NC, USA).

RESULTS

General model

The full multiple-regression model explained 34.9% of the variance, with $p < 0.0001$ (degrees of freedom = 7,819, $n = 826$). Each of the variables contributed a significant amount to the explained variation (SPP, $p < 0.0001$; YR, $p = 0.004$; BM, $p < 0.0001$; HRM, $p < 0.0001$). It therefore made sense to examine the species individually.

Analyses by species

Regression equations for individual species are shown in Table 1. For all species, the relationship between the inde-

pendent variables and mercury concentrations was highly significant. Depending on the species, 15 to 48% of the variance in mercury concentration was explained by a combination of YR, BM, and HRM. Table 2 shows the relative importance of dependent variables, using stepwise regression. All variables were significant except HRM for YP and YR for LMB.

When examined at the species level, BM explained a significant amount of variation in mercury concentrations in all species (Fig. 2). Body size in the LMB collections also was strongly correlated with increasing YR ($r^2 = 0.47$, $n = 89$, $p < 0.0001$), and the same held true for SMB ($r^2 = 0.05$, $n = 130$, $p \approx 0.01$). These relationships demonstrate that it is necessary to take the approach of discerning the independent effects of time and space by subtracting such correlation components in the multiple-regression analysis (Table 1).

The most striking finding seen in Table 1 concerns the directions of the relationships. In every case, BM and HRM were positively related to mercury concentrations, and YR was negatively related to mercury concentrations. The striking BM effect can be seen in Figure 2, in which separate analyses of the relationship of BM to Hg concentration are performed for the five species.

Changes in mercury concentrations over time

The full regression model and the species-level models showed that mercury concentrations declined significantly over time for four of the five fish species (Table 2). To show this relationship graphically, we inserted the average BM for each species and calculations of Hg concentrations at HRM 100 into the equation shown above. Figure 3 shows the predicted change in mercury concentrations over time. For all five species, mercury concentrations declined on the order of 0.5 to 1% per year over the three-decade study period. For fish of average mass at HRM 100, the drop over the entire period sampled ranged from 31 to 62%.

Changes in mercury concentrations over space

The full regression model and the species-level stepwise-regression analyses showed that mercury concentrations increased with HRM for four of the five fish species. To show this relationship graphically, we inserted the average BM for each species and year 1970 into the equation shown above. Figure 4 shows the predicted change in mercury concentrations over HRM. For fish of average mass in the year 1970, mercury concentrations increased by approximately 0.001 to 0.002% per HRM over the 250-mile (≈ 417 -km) study area.

The combined multivariate and residual analyses were consistent with those few cases that had adequate data to see geographic trends for a given year. Figure 5 shows mercury BM residuals for the five species in two years, 1992 and 1998, when the sample sizes were large enough for such an analysis to be performed. In four of the five cases, a significant positive relationship was found between mercury BM residual and HRM.

There appears to be a difference among species in the relationship of the mercury concentration of average individuals for a given species as a function of HRM (Fig. 4). Striped bass and YP appear to have similarly low slopes relative to LMB, SMB, and CP. We speculate that the lower slopes for STB and YP reflect their greater dispersal abilities compared with the other species. We tested the possibility that these two groups differed in change of mercury concentration with HRM by using the following multiple-regression model, which in-

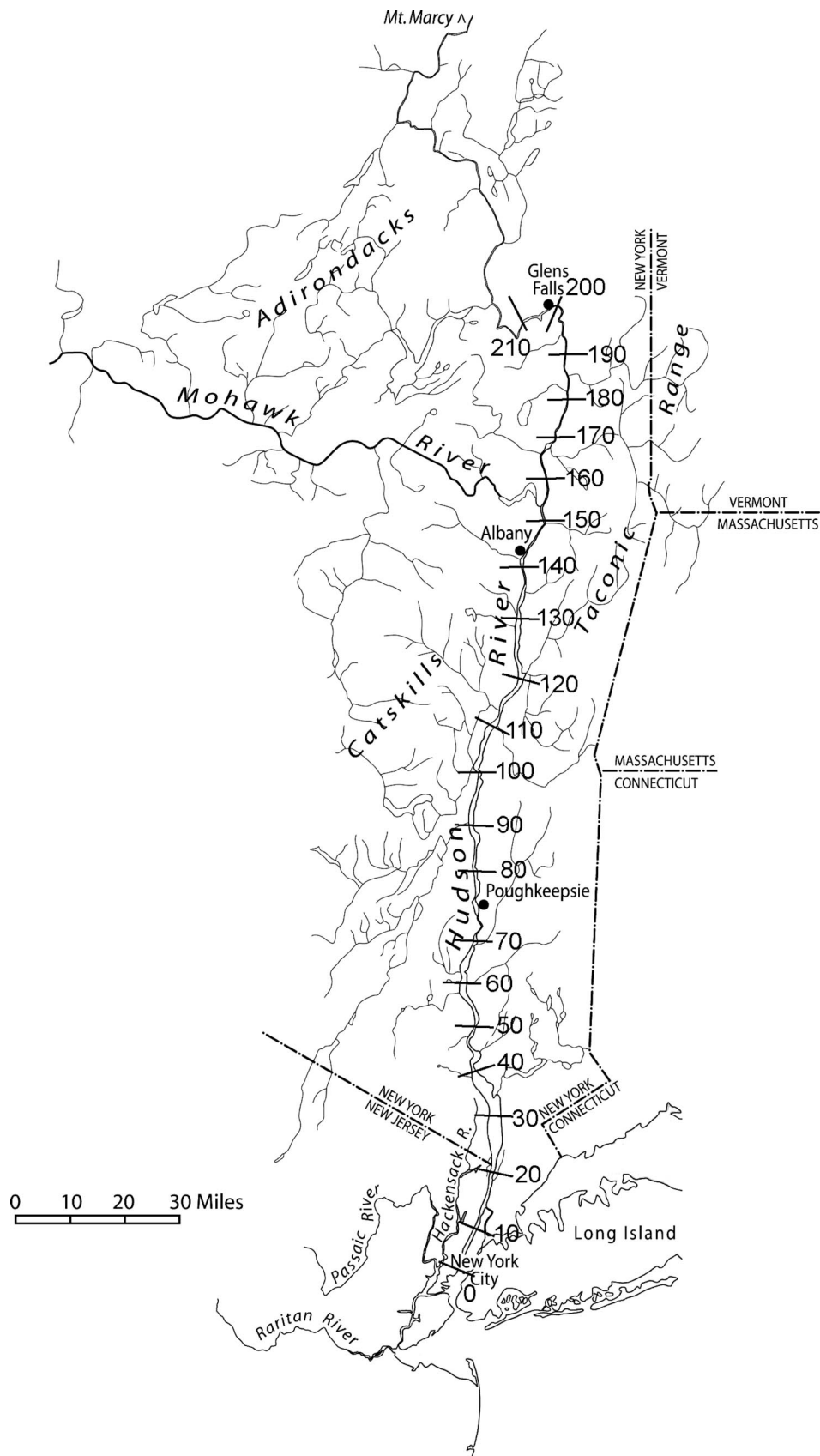


Fig. 1. Map of the Hudson River (NY, USA), showing river mile locations. Possible mercury sources include the Ciba-Geigy chemical plant near Glens Falls (NY, USA) and watershed sources from the Adirondacks, above Hudson River mile 200.

Table 1. Regression equations for individual species^a

Species	Equation	r ²	p	n
STB	Hg = 11.787658 - 0.005809(YR) + 0.0000248(BM) + 0.0012002(HRM) + e	0.15	<0.0001	291
YP	Hg = 12.575151 - 0.006331(YR) + 0.0009343(BM) + 0.0008919(HRM) + e	0.40	<0.0001	93
LMB	Hg = 11.234311 - 0.005709(YR) + 0.0003748(BM) + 0.0021669(HRM) + e	0.49	<0.0001	89
SMB	Hg = 22.505552 - 0.011342(YR) + 0.0005419(BM) + 0.0022387(HRM) + e	0.20	<0.0001	130
CP	Hg = 11.796458 - 0.005953(YR) + 0.0000099(BM) + 0.0017346(HRM) + e	0.48	<0.0001	224

^a Italicized regression coefficients are not significant at $p < 0.05$; all others are significant ($p < 0.05$). BM = body mass; CP = carp; Hg = mercury concentration; LMB = largemouth bass; SMB = smallmouth bass; STB = striped bass; YP = yellow perch; YR = Hudson River (USA) mile.

cludes parameters for mobility of the species (postulated mobility), YR (year), specimen weight (weight), and river mile of collection of fish (rmile):

$$C_{\text{Hg}} = B_0 + B_1(\text{postulated mobility}) + B_2(\text{year}) \\ + B_3(\text{weight}) + B_4(\text{rmile}) \\ + B_5(\text{mobility} \times \text{rmile}) + e$$

We now have two states, zero for lack of postulated mobility (applies to LMB, SMB, and CP) and unity for mobility (applies to SB and YP). We are testing the hypothesis that $B_5 = 0$, which implies that no difference in slope exists for the two kinds of hypothesized fish mobility. The multiple regression of the model above explains 16.4% of the variance and is significant at $p < 0.0002$ (degrees of freedom = 5,821, $n = 827$). Table 3 shows that B_5 is significantly different from zero—that is, the slope for mobile fish differs significantly from the slope for nonmobile fish when HRM is used to predict mercury concentration.

DISCUSSION

Mercury contamination in New York fish is of importance as a source of toxicity second only to polychlorinated biphenyls [22]. Our analysis shows that mercury concentrations of Hudson River fish have significantly declined over the past three decades in SB, YP, SMB, and CP. Mercury concentrations also declined over time in LMB, but in this case, the rela-

tionship is not significant. Mercury concentrations have declined as much as approximately 60% in some species.

Factories have been releasing less mercury into the Hudson watershed, less mercury-laden waste has been burned in the area, and atmospheric deposition of mercury has decreased [17,23–25]. These reductions are having a measurable effect on our environment. The mercury concentrations in Hudson River fish have significantly declined since 1970.

Mercury concentrations in Hudson River fish show a geographic pattern. As HRM increases—that is, as we travel north from New York City—mercury concentrations increase. This implies that the source of mercury is sufficiently strong, and the depuration rate is sufficiently slow, to prevent homogenization of concentrations by fish dispersal and population mixing. Fish retain a fingerprint of the location of mercury uptake, despite the facts that resuspension and sedimentary particle transport probably distribute mercury in sediment throughout the estuary [26,27] and that fish must swim, which also should reduce the geographic gradient in mercury concentration. The geographic pattern is not restricted to any one species of fish; in fact, it applies to highly dispersing species, such as SB.

Three potential sources of mercury exist in the Hudson River. First is the Hercules/Ciba-Geigy chemical plant, located near Glens Falls (NY, USA). Releases of mercury and other metals from this plant were reflected in high mercury concentrations in sediments, which have declined from the 1960s to the 1990s. High mercury concentrations in sediments also are found south of the site [17].

The Adirondack watershed also may release mercury from the north that originates from atmospheric deposition [12,28]. Atmospheric deposition in the Hudson watershed has declined, resulting in reduced acidification and, perhaps, reduced mercury supply to the watershed and, eventually, the mid-Hudson River [23]. Methylmercury deposited from the atmosphere may be held in forest soils, but runoff would result in release to the watershed [29]. Both the Mohawk and the upper Hudson rivers contribute substantial amounts of mercury to the Hudson watershed [29]. These sources of mercury might reach sediments and might be methylated by microbial processes, eventually reaching the fish by consumption of methylmercury-laden prey [30]. In an analysis of Wisconsin lakes, Greenfield et al. [31] demonstrated a relationship between mercury concentrations in YP and the amount of wetlands in the watershed. Growth factors and local lake chemistry were much more important in explaining mercury variation. Lake acidity can be inversely related to Hg concentrations in fish of adjacent lakes [32].

Whatever the source, mercury in the Hudson River comes from the north. As evidenced by the geographic trends in the

Table 2. Relative importance of variables (year [YR], Hudson River [USA] mile [HRM], and body mass [BM]) as determined from stepwise regression for individual species^a

Species	Variable	Partial r ²	r ²
SB	HRM	0.0665	0.0665
	BM	0.0524	0.1189
	YR	0.0311	0.1500
YP	BM	0.3656	0.3656
	YR	0.0284	0.3940
	HRM	0.0099	0.4039
LMB	BM	0.4191	0.4191
	HRM	0.0516	0.4707
	YR	0.0177	0.4884
SMB	BM	0.1347	0.1347
	YR	0.0123	0.1470
	HRM	0.0490	0.1960
CP	HRM	0.2959	0.2959
	YR	0.1441	0.4400
	BM	0.0397	0.4797

^a Italicized numbers refer to variables that were not significant at $p < 0.05$. CP = carp; LMB = largemouth bass; SMB = smallmouth bass; STB = striped bass; YP = yellow perch.

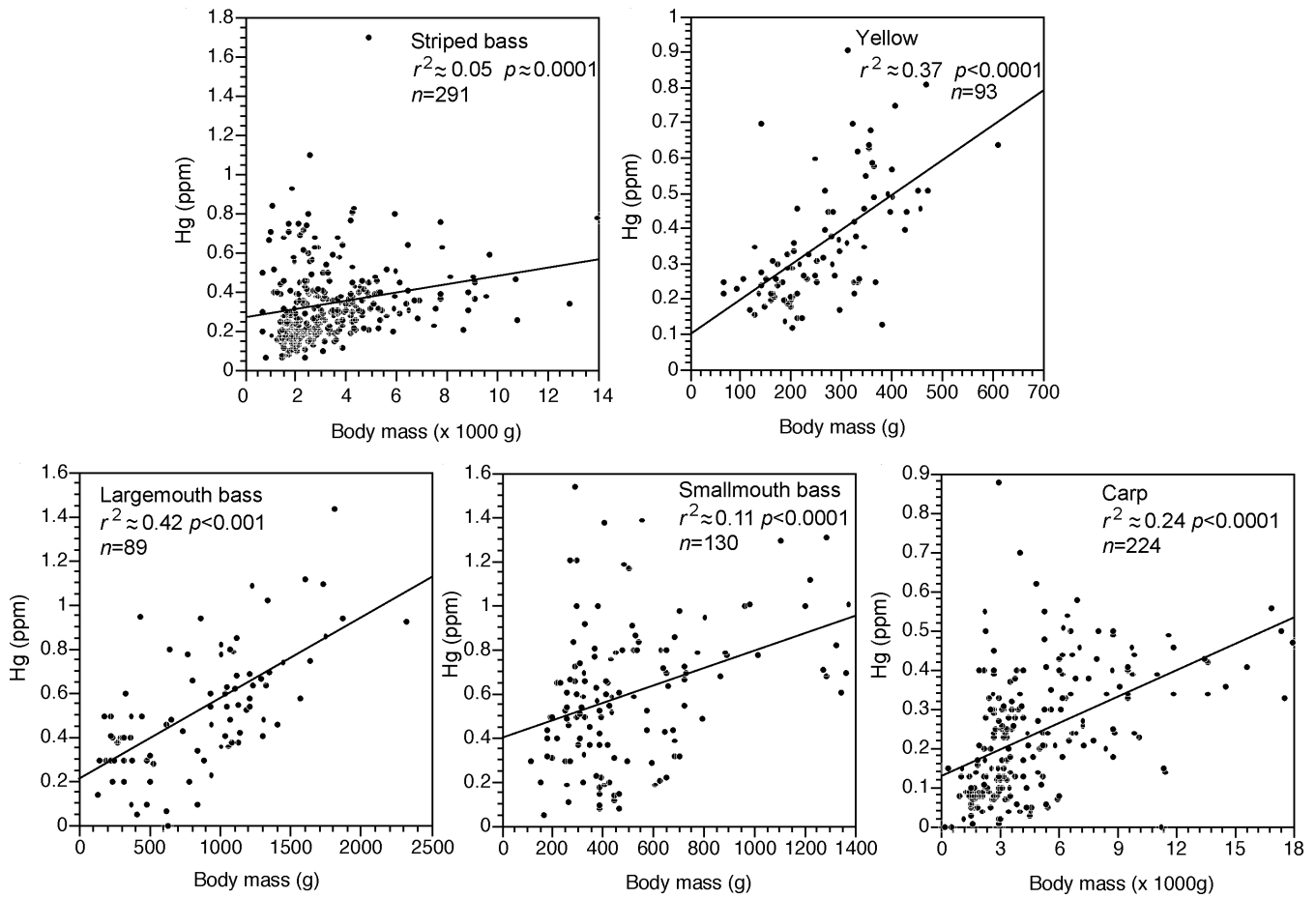


Fig. 2. Relationship between mercury concentration and body mass for all five species. For each species, all fish are combined over all years and localities.

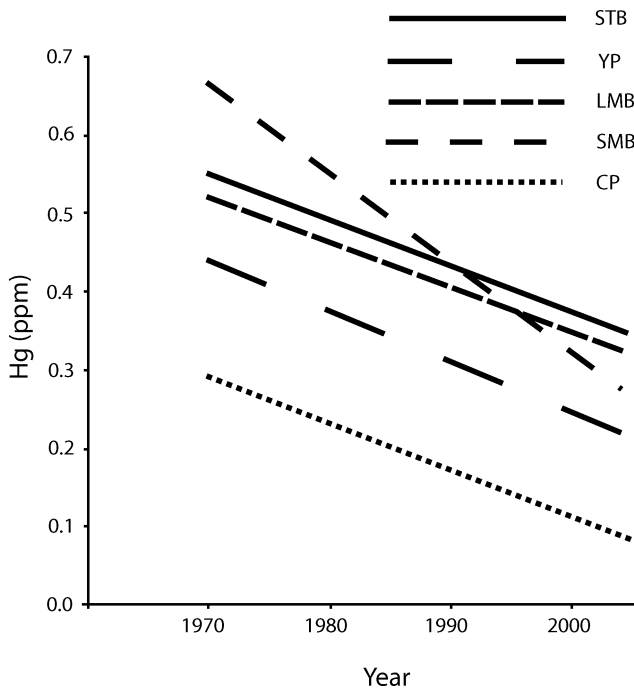


Fig. 3. Prediction from the multiple regression of change in mercury concentrations on year of collection for fish of average mass for each of the five species calculated at Hudson River (USA) mile 100. CP = carp; LMB = largemouth bass; SMB = smallmouth bass; STB = striped bass; YP = yellow perch.

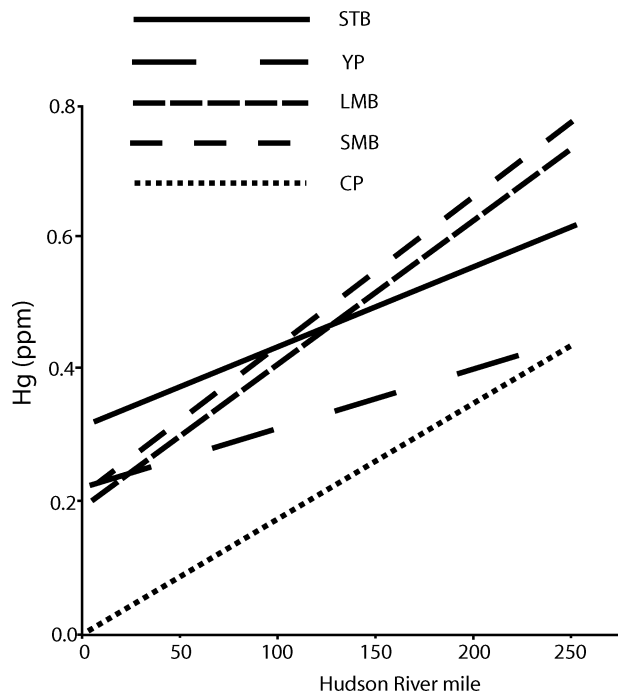


Fig. 4. Predictions for regression of mercury to Hudson River (USA) mile for fish of average weight for each species calculated for the year 1970. CP = carp; LMB = largemouth bass; SMB = smallmouth bass; STB = striped bass; YP = yellow perch.

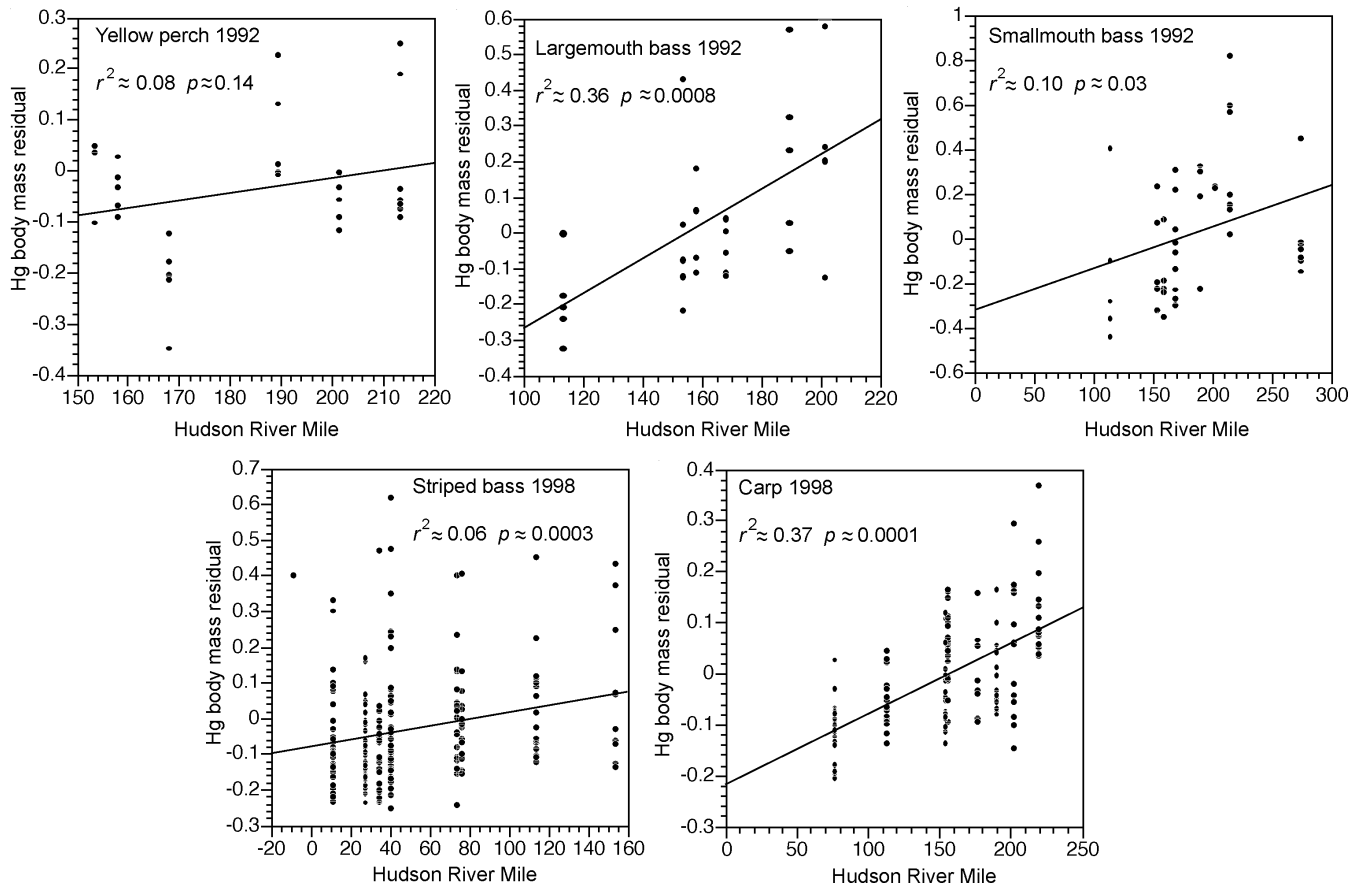


Fig. 5. Relationship of residuals from regression of mercury on body mass to Hudson River (USA) mile for the five species in cases where enough data existed to examine data for single years.

fish, the signal declines toward the south, and the mobility of fish species might increase the exposure of mercury to anglers farther south. Our data suggest that geographic gradients of mercury concentration in the Hudson River may be strongly affected by dispersal distances of the species bearing the toxic substances. Thus, the pattern of geographic change of a toxic substance with distance might be an indicator of dispersal ability. A geographically restricted source of a toxic substance therefore may be spread widely throughout a watershed by toxin-laden dispersing species [33]. These results also may apply to polychlorinated biphenyls in Hudson River fish [34] and to many other watersheds and coastal areas.

Acknowledgement—The present study was supported by grant 00705A from the Hudson River Foundation. We are grateful to Michael Kane, who provided the data from the New York State Department of Environmental Conservation database and helped with explanations of the data and its sources.

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Table 3. Estimates of parameters of multiple regression with mobility hypothesis^a

Term	Parameter	Estimate	Standard error	<i>t</i> Ratio	Prob. > <i>t</i>
Intercept	B_0	12.125179	2.032527	5.97	<0.0001
Mobility	B_1	0.0700394	0.02021	3.47	0.0006
Year	B_2	-0.006017	0.001026	-5.86	<0.0001
Weight	B_3	-0.000009	0.000003	-3.07	0.0022
River mile	B_4	0.0015569	0.000146	10.70	<0.0001
Mobility \times river mile	B_5	-0.00245	0.000292	-8.38	<0.0001

^a Value of *t* from *t* test of regression slopes and respective statistical probabilities (Prob.) are given.

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