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# Indicators of sediment and biotic mercury contamination in a southern New England estuary

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ABSTRACT

Total mercury (Hg) and methylmercury (MeHg) were analyzed in near surface sediments (0–2 cm) and biota (zooplankton, macro-invertebrates, finfish) collected from Narragansett Bay (Rhode Island/Massachusetts, USA) and adjacent embayments and tidal rivers. Spatial patterns in sediment contamination were governed by the high affinity of Hg for total organic carbon (TOC). Sediment MeHg and percent MeHg were also inversely related to summer bottom water dissolved oxygen (DO) concentrations, presumably due to the increased activity of methylating bacteria. For biota, Hg accumulation was influenced by inter-specific habitat preferences and trophic structure, and sediments with high TOC and percent silt–clay composition limited mercury bioavailability. Moreover, hypoxic bottom water limited Hg bioaccumulation, which is possibly mediated by a reduction in biotic foraging, and thus, dietary uptake of mercury. Finally, most biota demonstrated a significant positive relationship between tissue and TOC-normalized sediment Hg, but relationships were much weaker or absent for sediment MeHg. These results have important implications for the utility of estuarine biota as subjects for mercury monitoring programs.



## 1. Introduction

Mercury (Hg) is an environmental toxicant of concern because of its pervasiveness and adverse effects on wildlife and human health (US EPA, 1997). Specifically, methylmercury (MeHg) is the organic form of Hg that has toxic effects to the neurological, cardiovascular, and reproductive systems of biota (Wolfe et al., 1998). The severity of these health impacts depends on the concentration and duration of MeHg exposure and the characteristics of the biotic receptors (US EPA, 1997). In aquatic environments, upper trophic level organisms are exposed to MeHg almost exclusively via dietary uptake (Hightower and Moore, 2003). The dominance of this exposure pathway reflects the tendency for MeHg to biomagnify across successive trophic levels, resulting in MeHg concentrations that are potentially toxic to top-level consumers, including humans (US EPA, 1997; Wiener et al., 2003). Accordingly, monitoring mercury contamination in aquatic environments is necessary to properly assess risks to ecological and human health resulting from MeHg exposure (Wiener et al., 2007).

Environmental Hg levels that could compromise the health of aquatic ecosystems have increased substantially during the industrial age (Fitzgerald and Clarkson, 1991; Mason et al., 1994). In estuarine and coastal habitats of the northeastern United States, elevated Hg contamination results from both local point sources and long-range transport processes (Clarkson, 1992; Balcom et al., 2004). A large fraction of Hg that enters a water body is entrapped in sediments (Balcom et al., 2004), and may be subjected to methylation by anaerobic bacteria (Gilmour et al., 1992; Benoit et al., 2003). Methylation rates are likely accelerated in estuarine and coastal sediments because these environments receive high inputs of inorganic Hg from anthropogenic sources (Varekamp et al., 2003; Conaway et al., 2007; Fitzgerald et al., 2007), and are characterized by *in situ* geochemical conditions that promote MeHg production (Compeau and Bartha, 1985; Mason and Lawrence, 1999). Estuaries also serve as critical habitat for a diverse assemblage of invertebrates and finfish, and these biota may experience increased environmental MeHg exposure.

Sediment-derived MeHg is generated and transferred to biotic receptors through several physical and biological processes (Chen et al., 2008). For example, MeHg mobilized from surface sediments to the water column is bioconcentrated in phytoplankton; a functional group that transfers MeHg to either pelagic or benthic trophic assemblages (Mason et al., 1996; Moye et al., 2002; Pickhardt and Fisher, 2007). MeHg enters pelagic food webs via the grazing actions of phytoplanktivorous zooplankton (Watras and Bloom, 1992; Mason et al., 1996) and is biomagnified in secondary consumers (Mathews and Fisher, 2008; Gehrke et al., 2011a). MeHg bioconcentrated in phytoplankton may also be returned to the sediment–water interface by suspension feeding

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invertebrates, serving as a biological conduit to the benthic trophic pathway (Locarnini and Presley, 1996; Chase et al., 2001). Moreover, benthic infauna and epifauna may have secondary exposure to MeHg through superficial contact with contaminated sediments and pore water, as well as the direct ingestion of sediments and detritus (Reynoldson, 1987; Locarnini and Presley, 1996; Lawrence and Mason, 2001).

MeHg mobilized from estuarine and coastal sediments contributes to the contamination of biota across contiguous marine ecoscapes, and may represent a key source of MeHg in global marine biota (Chen et al., 2008). Therefore, the causative factors leading to enhanced environmental Hg in estuaries and its subsequent incorporation into local food webs warrant further investigation. This study sought to: (1) characterize the distribution of mercury (total Hg and MeHg) in surface sediments collected from a southern New England estuary; the Narragansett Bay (Rhode Island and Massachusetts, USA) and adjacent embayments and tidal rivers, (2) describe the natural and anthropogenic factors that are associated with enhanced sediment mercury contamination, (3) examine the effect of spatially-explicit geochemical conditions and trophic processes on the Hg concentration of estuarine biota, i.e., zooplankton, macro-invertebrates, and forage finfish, and (4) assess the utility of focal biota as bio-indicators of sediment total Hg and MeHg contamination in an effort to inform potential monitoring programs.

## 2. Methods

### 2.1. Study site

Narragansett Bay is a temperate estuary that is contiguous with Rhode Island Sound at its mouth and extends northward into Rhode Island (RI) and Massachusetts (MA) (total area ~380 $km^2$; mean depth ~8 m; Chinman and Nixon, 1985; Fig. 1). The geographically-complex system is comprised of several tributary-estuaries and bays, including Greenwich Bay to the west, Providence and Seekonk Rivers to the northwest, and Taunton River and Mt. Hope Bay to the northeast. Moreover, the bay proper is longitudinally divided by islands forming the West Passage, East Passage, and Sakonnet River.

Narragansett Bay serves as a drainage basin for several rivers and streams (watershed area ~4790 $km^2$), of which the Blackstone, Pawtuxet, and Taunton Rivers are important sources of freshwater discharge. The estuary is generally well-mixed with a slight down-bay salinity gradient (range ~24–33 in the bay proper; Kremer and Nixon, 1978) (Table 1). Annual water temperatures typically range from −0.5 to 25 °C (Kremer and Nixon, 1978), and portions of the upper bay, including Greenwich Bay, are prone to episodic hypoxia during summer months (Deacutis et al., 2006; Melrose et al., 2007; Saarman et al., 2008). Bay sediments are comprised mostly of fine-grained silts and clays, with fine sands becoming increasingly abundant in shoal areas of the upper bay and in the lower bay (Murray et al., 2007). Further, relative to the lower bay, the upper bay is subject to greater anthropogenic influences (e.g., nutrient and contaminant loading), and thus, experiences comparatively deteriorated water quality (Desbonnet and Costa-Pierce, 2008).

### 2.2. Sample collection, processing, and preservation

Surface sediments and biota were collected from Narragansett Bay in May–June 2006 and May–September 2006–2010, respectively (Fig. 1). Sediments were collected from 51 locations using a Van Veen benthic grab, from which two replicate cores were taken (5-cm diameter; 10-cm depth) and the top 2 cm were processed. Biota were collected throughout the bay using a combination of plankton nets (100-μm mesh), trawls, seines, and hand collections. Collected biota were identified to the lowest practical taxon and subsequently categorized into eight focal groups (zooplankton, polychaete, gastropod, bivalve, amphipod-isopod, shrimp, crab, and killifish), which included 20 unique species. These particular species were selected as mercury bio-indicators according to the criteria and recommendations presented by Wiener et al. (2003) and Evers et al. (2008); selecting an array of biota that are locally abundant, exhibit high site fidelity, and are accessible using routine sampling techniques. Moreover, candidate bio-indicators were chosen based on their diverse life history characteristics (e.g., longevity, dietary habitats, and foraging ecology) and because they represent important biological vectors for the transfer of MeHg to higher trophic levels.

Sediments and biota collected in the field were placed in sterile polyethylene bags, put on ice for transportation, and frozen at −20 °C after returning to the laboratory. In the laboratory, biota were partially thawed and measured for wet weight (g) and body size (cm): total length (fish and crangonid shrimp), carapace width or length (crabs and palaemonid shrimp, respectively), and shell height (bivalves). Biota were then processed and analyzed as whole-bodies, with the exception of bivalves and gastropods that had their shells removed. As needed, diminutive biota (zooplankton, amphipods-isopods, and gastropods) from the same collection site were combined to ensure adequate tissue mass for subsequent mercury and stable isotope analysis. For final preservation, all sediment and biotic samples were freeze-dried for 48 h, homogenized with a mortar and pestle (biota only), and stored at room temperature in clear borosilicate vials.

### 2.3. Mercury analysis of sediments and biota

Sediment and biota total Hg concentrations (ppm dry wt.) were measured using a direct mercury analyzer (DMA-80, Milestone, Shelton, Connecticut) that utilizes thermal decomposition, amalgamation, and atomic absorption spectrophotometry (EPA method 7473; US EPA, 1998). The DMA-80 has a detection limit of 0.01 ng Hg (typical working range = 0.05–600 ng), and the Hg content of all samples in this study were above the instrument's detection limit. Certified reference materials (CRMs) prepared by the National Research Council Canada, Institute of Environmental Chemistry (Ottawa, Canada) were used to calibrate the DMA-80 and included TORT-1 (lobster hepatopancreas) and DORM-2 (dogfish muscle) (US EPA, 1998). Calibration curves were highly significant (mean $R^2 = 1.00$; range $R^2 = 0.99–1.00$; $p < 0.0001$), and the recovery of the TORT-1, DORM-2, and PACS-2 (marine sediment) CRMs ranged from 92.7% to 106.7% (mean = 95.6%). All samples were analyzed as duplicates, and an acceptance criterion of 10% was implemented. Two concurrent investigations also determined that the DMA-80 used in this study produced statistically comparable results to isotope dilution gas chromatography-inductively coupled plasma mass spectrometry (ICP-MS), with $R^2$ values of 0.902–0.946 between the two methods (Piraino and Taylor, 2009; Payne and Taylor, 2010).

Sediment MeHg concentrations (ppb dry wt.) were measured at the Trace Element Analysis Laboratory, Dartmouth College (Hanover, New Hampshire). Sediment samples (50 mg dry wt.) were weighed into individual polypropylene vials, after which ~150 pg of aqueous $CH_3Hg^{201}$ was added as the enriched isotope spike, followed by 5 mL of 6 mol/L HCl. A 5 mL aliquot of $CH_2Cl_2$ was then added to each sample, after which the $CH_2Cl_2$ fraction containing MeHg was extracted and transferred to a separate polypropylene vial. Ten milliliter of deionized (DI) water was added to each new vial, and placed in a ~50 °C water bath and bubbled with argon until the $CH_2Cl_2$ evaporated. Samples were then transferred to amber glass vials, buffered, ethylated, and filled with DI water prior to analysis. Final sample analysis was performed using a purge and

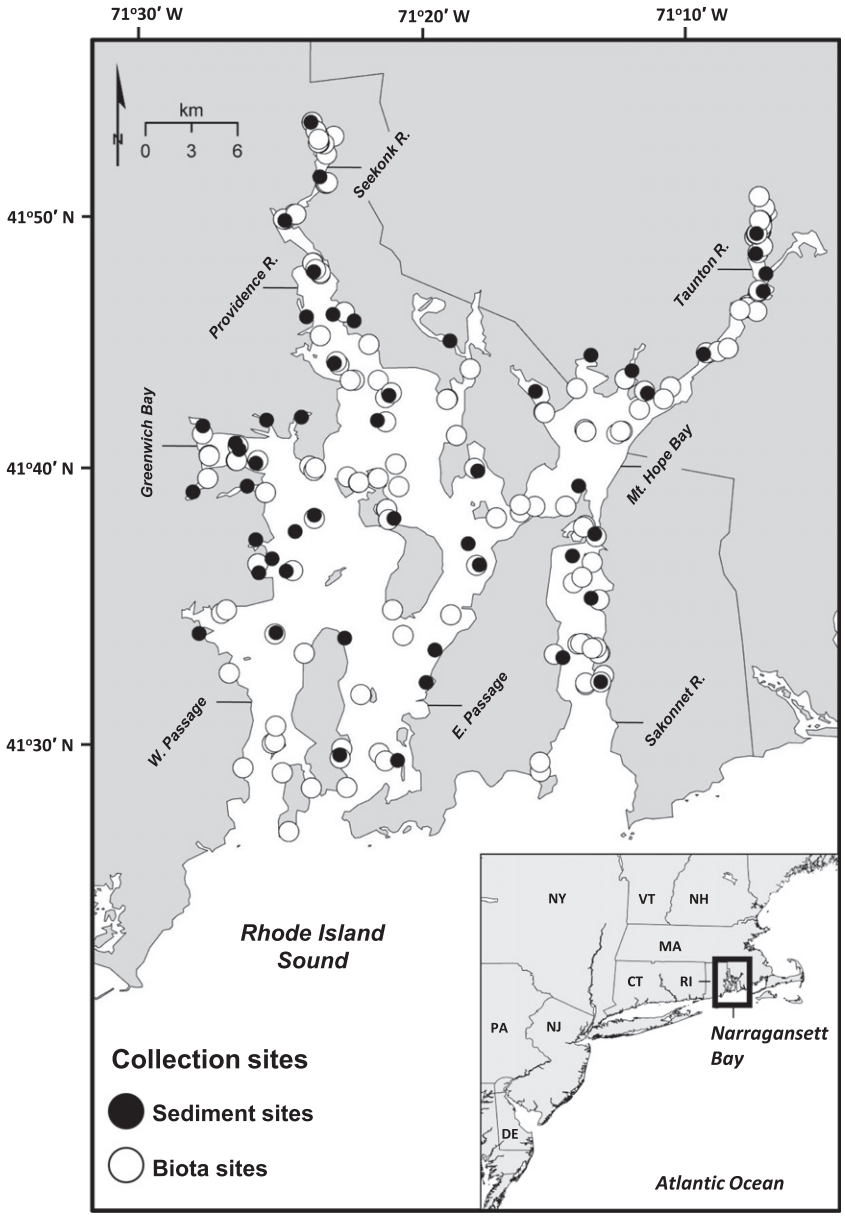


**Fig. 1.** Map of the Narragansett Bay (Rhode Island/Massachusetts, USA) and adjacent embayments and tidal rivers with points denoting sediment and biota collections sites.

trap MERX GC (Brooks Rand, Seattle, Washington) interfaced with an Agilent 7500 ICP-MS (Santa Clara, California). The recovery of MeHg in European Reference Material CC580 (estuarine sediment) was 97.5% (range = 94.1–99.8%), and sample precision for duplicate runs was 91.3% (range = 89.7–94.4%).

### 2.4. Stable isotope analysis of biota

Stable isotope analysis was used to assess the effect of trophic processes on the Hg content of biota (Piraino and Taylor, 2009; Payne and Taylor, 2010; Szczebak and Taylor, 2011). Specifically, nitrogen ($^{15}N/^{14}N$) stable isotope signatures were used to estimate time-integrated feeding history, whereas carbon ($^{13}C/^{12}C$) stable isotopes were used as indicators of the initial carbon source to the estuarine food web (e.g., pelagic and benthic primary production) (Fry, 2006). Isotope measurements of a subsample (~1 mg dry wt.) of biotic tissue ($n$ = 19–38 per species) were performed by the Boston University Stable Isotope Laboratory (Boston, MA) using automated continuous-flow isotope ratio mass spectrometry (CF-IRMS). Ratios of $^{15}N/^{14}N$ and $^{13}C/^{12}C$ are described using the standard delta notation ($\delta$), expressed as the relative per mil (‰) difference between the samples and international standards (atmospheric nitrogen, $^{15}N_{air}$, and Vienna Pee Dee Belemnite, $^{13}C_{V\text{-}PDB}$, respectively), and calculated using the following equation: $\delta X = (R_{sample}/R_{standard} - 1) \times 1000$, where $X = {}^{15}N$ or $^{13}C$ and $R = {}^{15}N/{}^{14}N$ or $^{13}C/^{12}C$. The recovery of internal reference material (peptone and glycine) for the CF-IRMS method was 99.7% and 99.6% for nitrogen and carbon, respectively. Moreover, the mean sample precision determined from duplicate analyses was 97.6% (range = 90.8–100.0%) and 98.6% (range = 91.9–100.0%) for nitrogen and carbon, respectively.

### 2.5. Data and statistical analysis

Site-specific geochemical conditions are potentially relevant to mercury dynamics in Narragansett Bay, and these data were accessed from several sources. The factors of interest included sediment characteristics [grain size < 63 μm (% silt–clay dry wt.) and

**Table 1**
Summary of sediment/bottom water characteristics and land use patterns within a 5-km radius of sample locations in the Narragansett Bay.

| Variable | Mean (SE) | Range |
|---|---|---|
| *Sediment* | | |
| Total mercury (Hg) (ppm dry wt.) | 0.555 (0.082) | 0.035–2.629 |
| Methylmercury (MeHg) (ppb dry wt.) | 1.852 (0.274) | 0.142–8.586 |
| Percent methylmercury (% MeHg) | 0.42 (0.04) | 0.03–1.32 |
| Grain size (% silt–clay) | 48.8 (2.0) | 25.1–83.4 |
| Total organic carbon (TOC; % dry wt.) | 2.74 (0.16) | 0.67–5.47 |
| *Water* | | |
| Bathymetric depth (m) | 4.9 (0.6) | 0.6–15.8 |
| Dissolved oxygen (DO; mg/L) | 6.4 (0.3) | 1.4–10.5 |
| Salinity (Sal) | 25.6 (1.2) | 0.3–32.6 |
| Temperature (Temp; °C) | 17.1 (1.2) | 12.0–27.4 |
| *Land use* | | |
| Human population (total number) | 33,990 (4935) | 234–155,511 |
| Hazardous waste sites (total number) | 14.9 (1.8) | 0–49 |
| Agriculture (ha) | 130.3 (13.6) | 5.7–3228.4 |
| Wetlands (ha) | 322.0 (33.5) | 4.7–525.7 |
| Vegetation (ha) | 1049.2 (95.2) | 332.7–3115.9 |
| Residential (ha) | 1245.4 (77.3) | 148.2–3001.4 |
| Commercial/Industry (Comm./Ind.) (ha) | 526.7 (66.9) | 3.1–2091.9 |

*Note:* Means, standard error of the mean (SE), and ranges are provided.

total organic carbon (TOC, % dry wt.)] and bottom water characteristics [bathymetric depth (m), dissolved oxygen (mg/L), salinity, temperature (°C)] (Table 1). The composition of bay sediments was assessed in concurrent studies between 2000 and 2006 (% silt–clay: $n = 253$; TOC: $n = 232$), and data were accessed from Murray et al. (2007) and US EPA (2011). Measurements of water characteristics were made at each site per sampling effort using a handheld YSI Model 85 meter (YSI Incorporated, Yellow Spring, Ohio), and additional data were obtained from parallel studies conducted from June to August 1999–2006 (dissolved oxygen: $n = 1327$; temperature and salinity: $n = 1354$) (Prell et al., 2004, 2006). Geographically-referenced information on RI and MA human population density (2000 census), location of hazardous waste sites (e.g., combustion and utility boilers, waste incinerators, sewage treatment facilities and outfalls), and land use patterns between 2003 and 2005 were accessed through publicly available sources (Mass GIS, 2011; RIGIS, 2011) (Table 1). Land use patterns followed the Anderson Level I classification scheme (Anderson et al., 1976).

To provide continuous spatial data throughout Narragansett Bay, isopleth maps of the sediment and water characteristics were generated using ArcView geographic information (GIS) software (ESRI, Inc., Redlands, California). Isopleth calculations were conducted using an optimizing approach that interpolates across a surface using the principle of inverse distance weighting procedure. Also using ArcView GIS, estimates of site-specific sediment and water characteristics were interpolated from the isopleth maps using the overlay feature, whereas land use patterns within a 5-km radius of a geographic point of interest were determined from proximity analysis.

Stepwise multiple linear regression analysis was used to identify the influence of geochemical variables on sediment mercury concentrations (total Hg, MeHg, and % MeHg = MeHg/total Hg) and biota-sediment accumulation factors ($BSAF_{(Hg)}$ and $BSAF_{(MeHg)}$ = biota total Hg/sediment total Hg or MeHg). Univariate linear regression models were used to analyze sediment mercury concentrations (normalized by TOC) as a function of surrounding land use patterns, and biota Hg concentrations as a function of TOC-normalized sediment total Hg and MeHg. Also among biota, differences in total Hg, isotopic signatures ($\delta^{15}N$ and $\delta^{13}C$), and BSAFs were analyzed with one-way analysis of variance (ANOVA) models using taxa as a fixed factor. Mean differences in total Hg, $\delta^{15}N$, $\delta^{13}C$, and BSAF values across 8 levels of taxa were contrasted with Ryan-Einot-Gabriel-Welsch (Ryan's Q) multiple comparison tests. Linear regression models were also used to examine the effect of $\delta^{15}N$ and $\delta^{13}C$ on the total Hg content of biota.

## 3. Results and discussion

### 3.1. Sediment mercury analysis

Mean total Hg and MeHg concentrations of surface sediments collected from Narragansett Bay equaled 0.56 ± 0.08 ppm and 1.85 ± 0.27 ppb, respectively, with concentrations greatest in the upper reaches of the estuary and decreasing southward to Rhode Island Sound (Table 1, Fig. 2A and B). Sediment mercury concentrations were particularly elevated in urbanized regions of the bay, including the Taunton River and portions of the Seekonk River, Providence River, Greenwich Bay, and Mt. Hope Bay. Conversely, relatively low levels of mercury contamination were detected in areas further removed from anthropogenic influences, including the East Passage and Sakonnet River. These results are further reflected in the spatial analysis of mercury contamination as a function of surrounding land use patterns. Specifically, sediment total Hg and MeHg concentrations normalized by organic content increased significantly in residential areas, and % MeHg was significantly related to human population density (Table 2). The range of sediment total Hg and MeHg concentrations measured in this study were consistent with levels observed in other urbanized estuaries of the United States (Bloom et al., 1999; Mason and Lawrence, 1999; Varekamp et al., 2000; Conaway et al., 2003; Hammerschmidt and Fitzgerlad, 2004; Gehrke et al., 2011b).

Sediment total Hg was positively related to total organic carbon (TOC) (Table 2, Fig. 3A), indicating that Hg has a high affinity for organic matter, as reported elsewhere (Mason and Lawrence, 1999; Hammerschmidt and Fitzgerlad, 2004; Chen et al., 2009). Moreover, the positive Hg–TOC relationship further suggests that sediments with high organic content may effectively scavenge and retain Hg from the water column, as demonstrated in other estuarine and coastal marine systems (Cossa et al., 1988; Conaway et al., 2003; Sunderland et al., 2006). Sediment total Hg was also inversely related to water depth (Table 2; Fig. 3B), corroborating the finding that Hg contamination is elevated in relatively shallow, near shore areas of the upper Narragansett Bay, and especially in the proximate embayments and rivers (Fig. 2A).

Rates of MeHg production in estuarine and marine sediments are governed by Hg inputs, and *in situ* geochemical conditions that affect the microbial activity of methylating bacteria or the bioavailability of the inorganic Hg substrate (Benoit et al., 2003). In this study, MeHg accounted for a small fraction of total Hg in surface sediments (mean % MeHg = 0.42 ± 0.04%; Table 1, Fig. 2C), and thus, inorganic Hg was the dominant mercury species. Results further revealed a significant positive relationship between sediment total Hg and MeHg concentrations (Table 2, Fig. 3C). Comparable sediment total Hg–MeHg relationships were purported in other estuaries (Benoit et al., 1998; Conaway et al., 2003; Hammerschmidt and Fitzgerlad, 2004; Sunderland et al., 2006), and reflect the role of inorganic Hg as the substrate for methylation. Conversely, several studies reported relatively weak or no association between total Hg and MeHg in estuarine sediments (Lambertsson and Nilsson, 2006 and references therein), thereby emphasizing the additional effect of geochemical factors on MeHg dynamics.

Sulfate-reducing bacteria function optimally at the interface between oxic and anoxic conditions in sediments and the water column, i.e., the redoxcline (Sunderland et al., 2006). In this study, sediment MeHg and % MeHg, (proxy for net MeHg production; Benoit et al., 2003) were inversely related to summer bottom water dissolved oxygen (DO) concentrations (Linear regression: MeHg, $p < 0.01$, $R^2 = 0.141$; % MeHg, $p < 0.05$, $R^2 = 0.118$) (Fig. 3D and E).

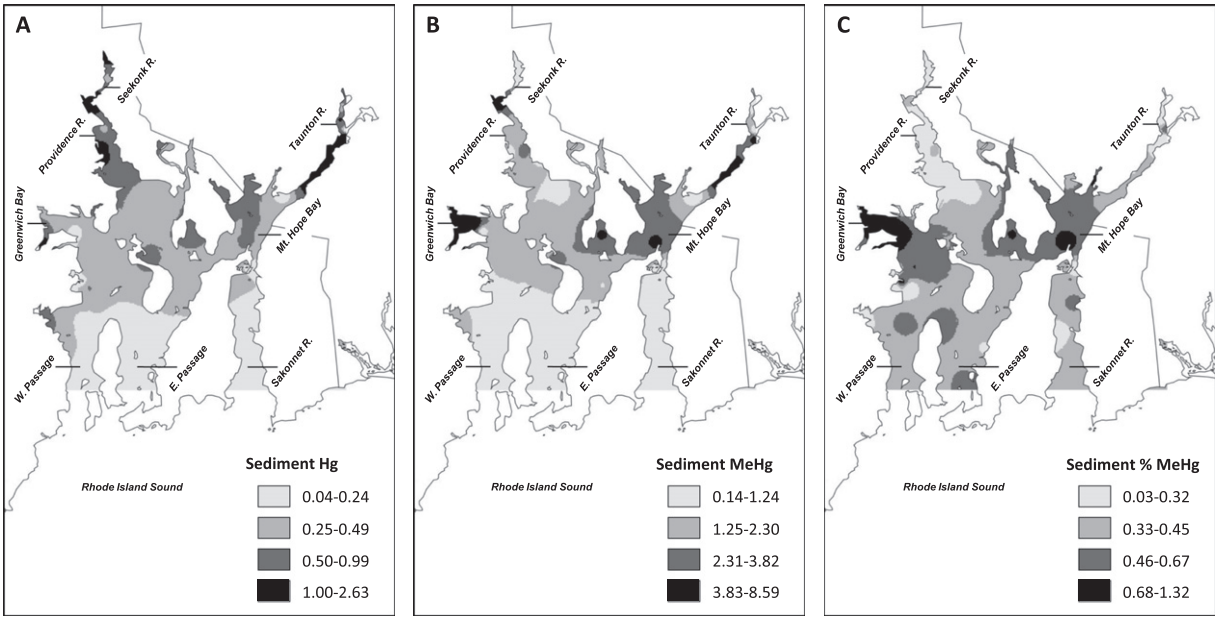


**Fig. 2.** Spatial distribution of total mercury (Hg; ppm dry wt.) (A), methylmercury (MeHg; ppb dry wt.) (B), and percent methylmercury (% MeHg = MeHg/total Hg) (C) in near surface sediments (0–2 cm) collected from the Narragansett Bay ($n = 51$).

**Table 2**
Parameter estimates (standard error; SE) and summary statistics for stepwise multiple linear regression models used to analyze sediment total mercury (Hg), methylmercury (MeHg), percent methylmercury (% MeHg = MeHg/total Hg), and biota-sediment accumulation factors ($BSAF_{(Hg)}$ and $BSAF_{(MeHg)}$ = biota total Hg/sediment Hg or MeHg).

| Relationship | Model parameters | | $F$ | $df$ | $R^2$ | $p$ |
|---|---|---|---|---|---|---|
| | Estimate | SE | | | | |
| *Sediment Hg* | | | | | | |
| TOC | 0.183 | 0.049 | 17.12 | 1, 50 | 0.259 | <0.0001 |
| Depth | −0.027 | 0.013 | 4.20 | 1, 50 | 0.056 | <0.05 |
| Intercept | 0.623 | 0.676 | – | – | – | – |
| *Sediment MeHg* | | | | | | |
| Total Hg | 0.444 | 0.102 | 18.92 | 1, 50 | 0.278 | <0.0001 |
| Intercept | −0.226 | 0.082 | – | – | – | – |
| *Sediment % MeHg* | | | | | | |
| Sal | 0.017 | 0.006 | 8.28 | 1, 50 | 0.145 | <0.01 |
| Intercept | −0.939 | 0.158 | – | – | – | – |
| *Sediment Hg (normalized)* | | | | | | |
| Residential | $0.551E^{-4}$ | $0.177E^{-4}$ | 9.71 | 1, 50 | 0.166 | <0.005 |
| Intercept | 0.030 | 0.059 | – | – | – | – |
| *Sediment MeHg (normalized)* | | | | | | |
| Residential | $0.170E^{-3}$ | $0.070E^{-3}$ | 5.88 | 1, 50 | 0.107 | <0.05 |
| Intercept | 0.168 | 0.236 | – | – | – | – |
| *Sediment % MeHg (normalized)* | | | | | | |
| Population | $0.610E^{-5}$ | $0.272E^{-5}$ | 5.03 | 1, 50 | 0.093 | <0.05 |
| Intercept | 0.320 | 0.132 | – | – | – | – |
| $BSAF_{(Hg)}$ | | | | | | |
| TOC | −0.070 | 0.027 | 86.58 | 1, 217 | 0.290 | <0.0001 |
| DO | 0.116 | 0.022 | 27.67 | 1, 217 | 0.082 | <0.0001 |
| % silt–clay | −0.005 | 0.002 | 6.11 | 1, 217 | 0.018 | <0.05 |
| Sal | 0.012 | 0.005 | 5.82 | 1, 217 | 0.017 | <0.05 |
| Intercept | −1.272 | 0.295 | – | – | – | – |
| $BSAF_{(MeHg)}$ | | | | | | |
| DO | 0.188 | 0.016 | 145.28 | 1, 217 | 0.407 | <0.0001 |
| Intercept | −2.338 | 0.101 | – | – | – | – |

*Note:* Response variables were $\log_{10}$-transformed and analyzed as a function of sediment characteristics [grain size (% silt–clay) and total organic carbon (TOC)] and bottom water characteristics [bathymetric depth, dissolved oxygen (DO), salinity (Sal), and temperature]. Sediment mercury concentrations normalized by TOC were also analyzed relative to land use patterns within a 5-km radius of a sample site (human population, hazardous waste sites, agriculture, wetlands, vegetation, residential, and commercial/industry). Only parameters significant at $p < 0.05$ were included in the model, and variable units are identified in Table 1.

These models suggest that areas of Narragansett Bay that are prone to hypoxia (e.g., Greenwich Bay; Deacutis et al., 2006; Melrose et al., 2007; Saarman et al., 2008) have increased Hg methylation rates because of the accelerated activity of anaerobic bacteria (Fig. 2B and C). However, whether the increase in sediment MeHg during low oxygen events are the result of a shoaling redoxcline and concomitant increase in sediment pore water methylation (Sunderland et al., 2006; Merritt and Amirbahman, 2008), or

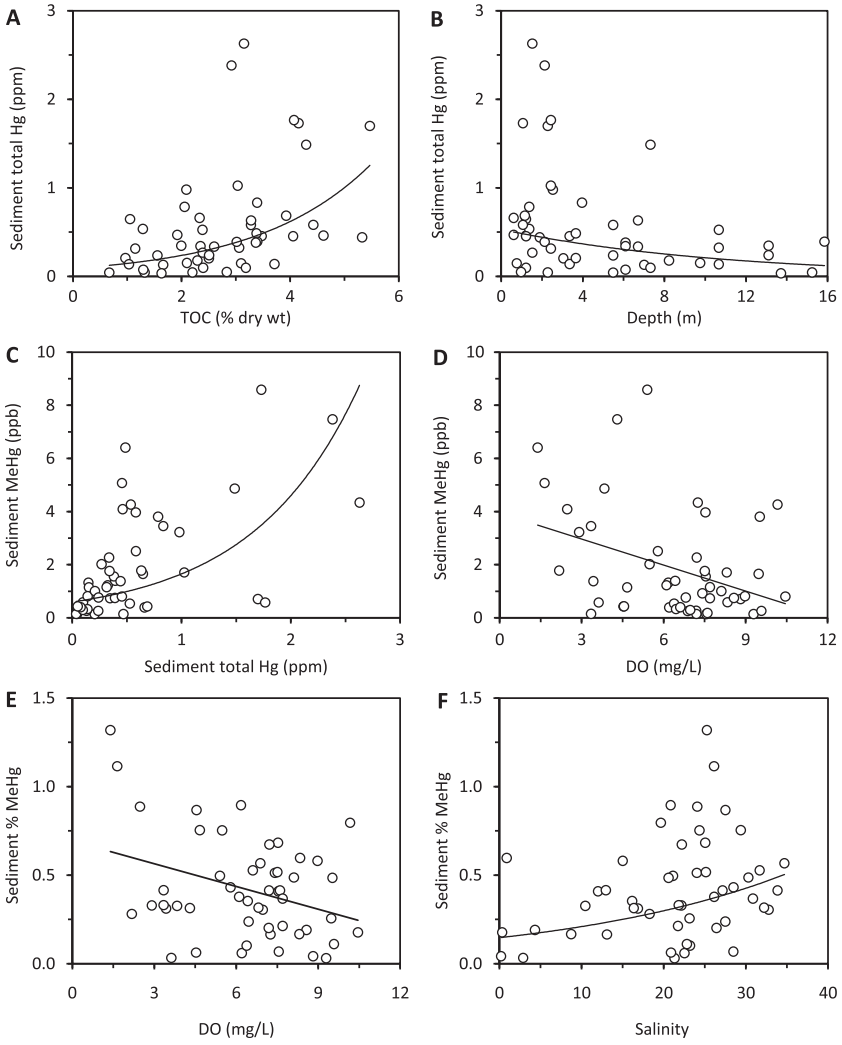


**Fig. 3.** Surface sediment total mercury (Hg; ppm dry wt.) (A, B), methylmercury (MeHg; ppb dry wt.) (C, D), and percent methylmercury (% MeHg = MeHg/total Hg) (E, F) as a function of site-specific geochemical conditions, including total organic carbon (TOC, % dry wt.), water depth (m), dissolved oxygen (DO, mg/L), and salinity. Least-squares exponential or linear regression models were fit to data ($n = 51$).

conversely, MeHg production in the water column (Sunderland et al., 2010) is unknown. For the latter, waterborne MeHg may be scavenged by underlying sediments that are high in organic carbon, as discussed above (Cossa et al., 1988; Conaway et al., 2003; Sunderland et al., 2006). Indeed this posit is substantiated by the positive relationship between sediment MeHg and TOC concentrations (Linear regression: $p < 0.05$, $R^2 = 0.104$). Alternatively, sediment organic matter can directly stimulate microbial activity by serving as a substrate for mineralization (Stoichev et al., 2004; Sunderland et al., 2004; Lambertsson and Nilsson, 2006), and this process would likely increase during episodic anoxia-hypoxia at the sediment–water interface (Sunderland et al., 2006). Importantly, peak methylation rates may occur at sediment depths greater than those examined in this study (~4 cm; Bloom et al., 1999; Merritt and Amirbahman, 2008), and therefore, MeHg in surface sediments (0–2 cm) may be the products of methylation at-depth, followed by the diffusion of MeHg toward the sediment–water interface and its entrapment at the redoxcline (Mason and Lawrence, 1999).

Spatial patterns in net MeHg production in Narragansett Bay, approximated by sediment % MeHg, were positively related to bottom water salinity (Table 2, Fig. 3F). This finding is contrary to other investigations that observed an inverse relationship between salinity and the methylation of inorganic Hg (Blum and Bartha, 1980; Compeau and Bartha, 1984; Benoit et al., 1998). The salinity effect documented in these previous studies was attributed to the reduction of sulfate to sulfide in high salinity (>25), low redox potential sediments (Compeau and Bartha, 1984), and the subsequent formation of charged Hg−S complexes that limit the bioavailability of the inorganic Hg substrate (Benoit et al., 1999). Narragansett Bay, however, is a relatively well-mixed, high salinity estuary with a slight salinity gradient; hence, the potential effects of salinity on MeHg production are expected to be relatively uniform for the majority of the system. This does not include the main freshwater inputs to the bay that have markedly lower saline conditions (<10), yet also experience decreased levels of sediment % MeHg (upper reaches of Seekonk and Taunton Rivers; Fig. 2C). The exact mechanisms explaining the lower rates of net Hg methylation in these systems are unclear, but likely involve the simultaneous influence of demethylation (Merritt and Amirbahman, 2008), as well as the complex interactions among sediment geochemical factors, including organic content, sulfate and sulfide concentrations, and dynamics in redox potentials (Chen et al., 2008).

### 3.2. Biota mercury analysis

Methylmercury mobilized from estuarine and coastal sediments is readily incorporated into the base of food webs, after which the contaminant biomagnifies across successive trophic

levels (Chen et al., 2008). Low trophic level biota are thus critically important in this process because they transfer MeHg through pelagic or benthic trophic pathways. In this study, mean total Hg concentrations among estuarine invertebrate and finfish species ranged from 0.06 to 0.23 ppm (Table 3), and differed significantly across the eight broader taxa (Table 4; Fig. 4A). Killifish had significantly higher total Hg concentrations then the remaining focal taxa (mean total Hg = 0.20 ± 0.01 ppm). In contrast, polychaetes, mollusks, and macro-crustaceans had moderate levels of total Hg that were comparable among each other (mean total Hg = 0.12 ± 0.01 ppm), and zooplankton comprised of copepods and decapod zoeae had the lowest total Hg across all groups (mean total Hg = 0.06 ± 0.01 ppm).

Total Hg concentrations of biota collected from Narragansett Bay were within range or lower than contaminants measured in conspecifics and congeners in other estuaries of the United States. Previous studies in Long Island Sound and Gulf of Maine, for example, reported comparable total Hg concentrations in *Acartia* copepods (~0.03–0.05 ppm), *Littorina littorea* (0.18 ± 0.03 ppm), *Carcinus maenas* (0.11 ± 0.02 ppm), and *Fundulus heteroclitus* (0.15 ± 0.03 ppm) (Watras and Bloom, 1992; Hammerschmidt and Fitzgerald, 2006; Chen et al., 2009). An exception to this observation was *Mytilus edulis*, whereby the total Hg of the blue mussel was ~64–93% higher in the Gulf of Maine relative to Narragansett Bay (Chase et al., 2001; Chen et al., 2009). Substantial differences in biotic total Hg were also observed in estuaries more distantly located from Narragansett Bay. Zooplankton from western Long Island Sound (New York), Chesapeake Bay (Maryland), and Core Sound (North Carolina), for example, had total Hg concentrations of 0.11–0.19 ppm (Cocoros and Chan, 1973). The total Hg of *L. littorea* collected from Georgia salt marshes was also elevated relative to conspecifics in this study, ranging between 0.6 and 33.1 ppm (Horne et al., 1999). Further, in Lavaca Bay, polychaetes (*Nereis occidentalis* and *N. succinea*), bivalves (*Enis minor*), and decapods (*Callinectes sapidus*) had mean total Hg concentrations 34–90% higher than values documented for similar species in Narragansett Bay (Locarnini and Presley, 1996). The observed differences in biotic Hg across ecosystems are attributed to spatial variations in Hg inputs and the *in situ* geochemical conditions that affect MeHg production and mobilization (Benoit et al., 2003). Moreover, physicochemical and ecological processes in estuaries vary over relatively small spatial scales, thus affecting the incorporation of MeHg into the food web and the contamination of resident biota (Chen et al., 2008).

**Table 4**
Summary statistics for one-way analysis of variance models testing for mean differences in total mercury (Hg; ppm dry wt.), stable nitrogen ($\delta^{15}$N) and carbon ($\delta^{13}$C) isotope signatures, and biota-sediment accumulation factors ($BSAF_{(Hg)}$ and $BSAF_{(MeHg)}$ = biota total Hg/sediment Hg or MeHg) among biota.

| Variable | *F* | *df* | *p* |
|---|---|---|---|
| Total Hg | 18.23 | 7, 1865 | <0.0001 |
| $\delta^{15}$N | 73.84 | 7, 458 | <0.0001 |
| $\delta^{13}$C | 52.15 | 7, 458 | <0.0001 |
| $BSAF_{(Hg)}$ | 10.57 | 7, 1865 | <0.0001 |
| $BSAF_{(MeHg)}$ | 8.95 | 7, 1865 | <0.0001 |

*Note:* Response variables were $\log_{10}$-transformed prior to statistical analysis.

### 3.3. Biota isotope analysis

Stable nitrogen ($\delta^{15}$N) and carbon ($\delta^{13}$C) isotope signatures of biota were used to infer relative trophic position and structure (Fry, 2006), which ultimately affect Hg dynamics in estuarine fauna

**Table 3**
Summary of total mercury (Hg; ppm dry wt.), stable nitrogen ($\delta^{15}$N) and carbon ($\delta^{13}$C) isotope signatures, length (mm), and whole-body wet weight (g) of estuarine biota collected from the Narragansett Bay.

| Taxon/species | *n* | Total Hg | $\delta^{15}$N[a] | $\delta^{13}$C[a] | Length[b] | Weight |
|---|---|---|---|---|---|---|
| *Zooplankton* | | | | | | |
| Copepods/decapod zoeae | 131 | 0.056 (0.006) | 11.86 (0.19) | −19.50 (0.14) | – | – |
| *Polychaete* | | | | | | |
| *Nereis* spp. | 83 | 0.139 (0.031) | 13.63 (0.16) | −16.51 (0.26) | – | – |
| *Gastropod* | | | | | | |
| *Littorina littorea* | 37 | 0.090 (0.004) | 11.22 (0.19) | −15.93 (0.33) | – | – |
| *Nassarius obsoletus* | 28 | 0.177 (0.036) | 13.00 (0.42) | −14.11 (0.56) | – | – |
| *Bivalve* | | | | | | |
| *Ensis directus* | 6 | 0.066 (0.007) | – | – | 11.03 (1.05) | 10.25 (2.72) |
| *Mercenaria mercenaria* | 22 | 0.104 (0.010) | 11.53 (0.11) | −18.08 (0.12) | 8.00 (0.51) | 29.81 (3.47) |
| *Modiolus demissus* | 135 | 0.137 (0.007) | 10.65 (0.21) | −20.46 (0.22) | 5.98 (0.14) | 4.46 (0.30) |
| *Mya arenaria* | 86 | 0.148 (0.011) | 11.38 (0.13) | −18.90 (0.24) | 4.51 (0.20) | 7.98 (1.14) |
| *Mytilus edulis* | 193 | 0.095 (0.004) | 10.67 (0.21) | −20.20 (0.18) | 4.01 (0.09) | 2.37 (0.18) |
| *Amphipod-Isopod* | | | | | | |
| Anthurid isopod | 11 | 0.132 (0.022) | – | – | – | – |
| *Gammarus* spp. | 40 | 0.093 (0.013) | 11.63 (0.24) | −17.50 (0.24) | – | – |
| *Shrimp* | | | | | | |
| *Crangon septemspinosa* | 294 | 0.121 (0.005) | 13.67 (0.17) | −16.43 (0.40) | 3.78 (0.05) | 0.49 (0.02) |
| *Palaemonetes pugio* | 189 | 0.128 (0.010) | 13.62 (0.21) | −16.54 (0.37) | 1.02 (0.03) | 0.39 (0.02) |
| *Crab* | | | | | | |
| *Callinectes sapidus* | 28 | 0.230 (0.03) | – | – | 4.81 (0.36) | 11.07 (2.04) |
| *Cancer* sp. | 7 | 0.096 (0.016) | – | – | 3.44 (0.78) | 16.86 (8.38) |
| *Carcinus maenas* | 120 | 0.126 (0.012) | 13.03 (0.14) | −15.30 (0.19) | 4.18 (0.17) | 24.17 (2.50) |
| *Hemigrapsus sanguineus* | 16 | 0.073 (0.012) | – | – | 1.83 (0.32) | 3.79 (1.57) |
| *Panopeus herbstii* | 53 | 0.063 (0.011) | 12.87 (0.23) | −14.58 (0.40) | 1.57 (0.07) | 1.78 (0.24) |
| *Killifish* | | | | | | |
| *Fundulus heteroclitus* | 164 | 0.232 (0.017) | 14.69 (0.23) | −19.19 (0.45) | 6.42 (0.11) | 4.32 (0.26) |
| *Fundulus majalis* | 223 | 0.178 (0.016) | 14.49 (0.25) | −18.53 (0.78) | 6.19 (0.15) | 3.98 (0.26) |

*Notes:* Sample sizes (*n*), mean values, and standard error of the means (in parentheses) are presented. Portions of the mercury and isotope data sets are published in Piraino and Taylor (2009), Payne and Taylor (2010) and Szczebak and Taylor (2011).

[a] Sample size for stable nitrogen and carbon isotope analysis was 19–38 per species.

[b] Length measured as shell height for bivalves, total length for crangonid shrimp and killifish, carapace length for palaemonid shrimp, and carapace width for crabs.

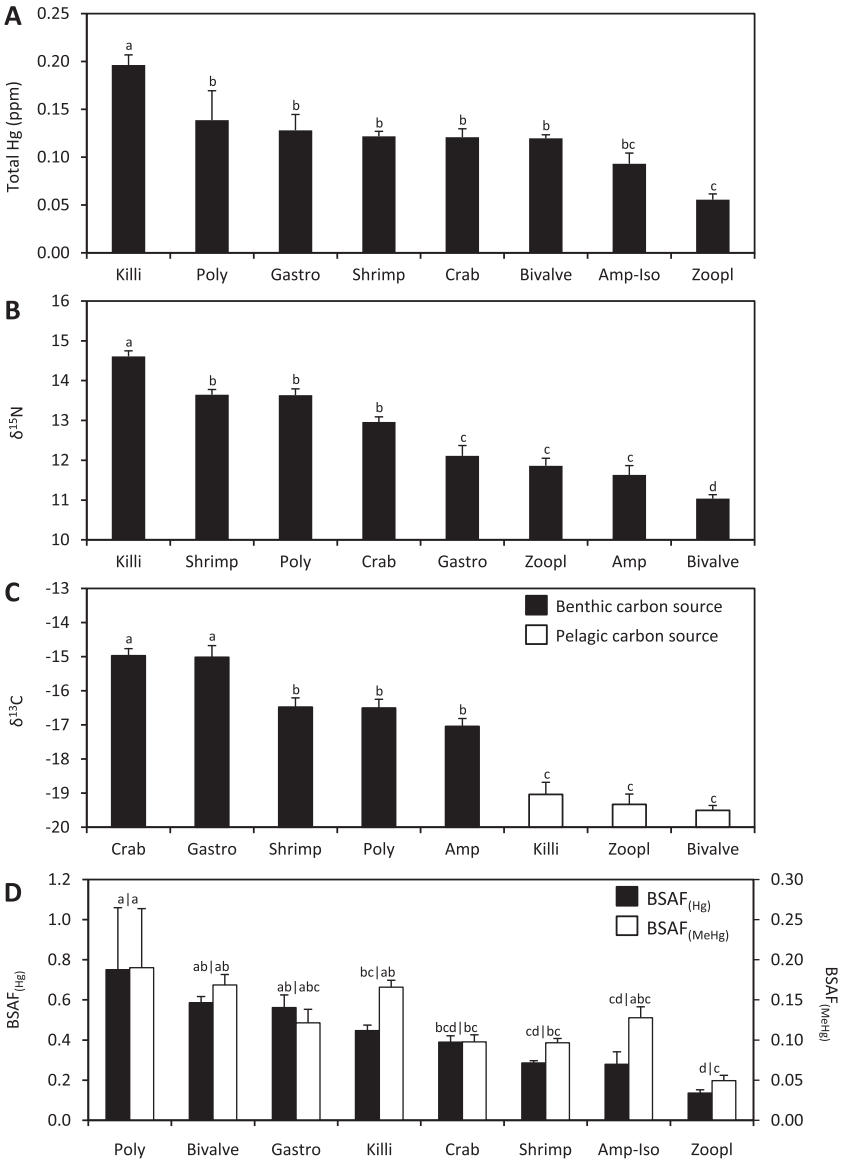


**Fig. 4.** Biotic total mercury concentrations (Hg; ppm dry wt.) (A) stable nitrogen ($\delta^{15}$N) and carbon ($\delta^{13}$C) isotope signatures (B, C), and biota-sediment accumulation factors ($BSAF_{(Hg)}$ and $BSAF_{(MeHg)}$ = biota total Hg/sediment Hg or MeHg, respectively) (D). Biota include killifish (Killi), polychaetes (Poly), gastropods (Gastro), shrimp, crabs, bivalve, amphipods-isopods (Amp-Iso), and zooplankton (Zoopl). Different lowercase letters denote significant differences in mean values, whereas the same lowercase letters indicate non-significant differences (Ryan's Q multiple comparison test). Also, for panel C, interspecies $\delta^{13}$C signatures were used to assess whether biota derived carbon from pelagic or benthic sources (Peterson and Howarth, 1987). All values represent means ±1 standard error.

(Piraino and Taylor, 2009; Payne and Taylor, 2010; Szczebak and Taylor, 2011). Mean $\delta^{15}$N values of biota ranged between 10.65‰ and 14.69‰ and differed significantly among taxa (Tables 3 and 4; Fig. 4B). Killifish, decapod crustaceans, and polychaetes had enriched $\delta^{15}$N signatures ($\delta^{15}$N > 12.9‰), indicating a carnivorous/omnivorous feeding strategy and relatively high trophic level status. Conversely, the $^{15}$N values of mollusks, amphipods, and zooplankton were significantly depleted ($\delta^{15}$N < 12.2‰), reflecting an increased reliance on basal food resources, such as phytoplankton or benthic algae. These results are consistent with previous analyses of $\delta^{15}$N ratios in conspecifics collected from Narragansett Bay (Pruell et al., 2006; Oczkowski et al., 2008).

Carbon isotope signatures were used as biomarkers for the different sources of primary production, and thus, define benthic and pelagic trophic assemblages in Narragansett Bay (Fry, 2006). Mean $\delta^{13}$C values of focal biota ranged from −20.46‰ to −14.11‰, which were further separated into three significantly distinct isotopic groupings (Tables 3 and 4; Fig. 4C). The enriched $\delta^{13}$C signatures of crabs and gastropods ($\delta^{13}$C = −14 to −15‰) and shrimp, polychaetes, and amphipods ($\delta^{13}$C = −16 to −17‰) indicate that these biota derive carbon from a combination of benthic algae, and to a lesser extent, detrital matter from sea grasses. Conversely, killifish, zooplankton, and bivalves had depleted $\delta^{13}$C values ($\delta^{13}$C < −18‰), and a carbon source consistent with a pelagic, phytoplankton-based, food web (Peterson and Howarth, 1987).

### 3.4. Biota-sediment mercury relationships

Biota-sediment accumulation factors, calculated from sediment total Hg and MeHg ($BSAF_{(Hg)}$ and $BSAF_{(MeHg)}$), were used to elucidate the linkages between faunal and environmental mercury contamination. Mean $BSAF_{(Hg)}$ and $BSAF_{(MeHg)}$ differed significantly across taxa, such that polychaetes, mollusks, and killifish generally had elevated BSAFs relative to planktonic and macro-crustaceans (Table 4; Fig. 4D). The increased Hg accumulation factors in these taxa are attributed to their respective habitat preferences and

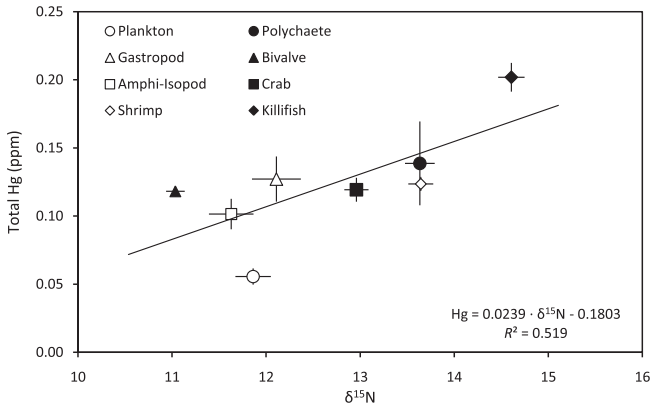


**Fig. 5.** Relationship between total mercury concentration (Hg; ppm dry wt.) and stable nitrogen ($\delta^{15}$N) signatures of biota (means ±1 standard error). A least-squares linear regression model was fit to the full data set, and the equation and $R^2$-value are presented.

foraging ecology. Nereid polychaetes and several species of mollusks examined in this study are classified as benthic infauna, which in turn favors the additional uptake of inorganic Hg and MeHg via direct contact with sediments and pore water (Wang et al., 1998; Coelho et al., 2006). Killifish, in contrast, maintain a relatively high trophic status in the estuary (increased $\delta^{15}$N signature), and therefore experience elevated BSAFs because MeHg biomagnifies in estuarine food webs (Chen et al., 2008). This supposition is supported by the positive relationship between biotic total Hg concentrations and $\delta^{15}$N values (Linear regression: $p < 0.05$, $R^2 = 0.519$) (Fig. 5), indicating that MeHg burdens are increased in upper trophic level organisms in Narragansett Bay. Unlike the positive total Hg–$\delta^{15}$N relationship, there was no relationship between biotic total Hg and $\delta^{13}$C values (Linear regression: $p = 0.933$). This observation differs from previous reports that estuarine biota with depleted $\delta^{13}$C signatures (pelagic-feeding) have higher Hg concentrations relative to organisms that forage benthically (Chen et al., 2009). However, the relatively high BSAF values of killifish and bivalves suggest that some pelagic-feeding fauna in the estuary have increased Hg bioaccumulation rates.

Spatially-explicit sediment and water characteristics significantly affected the relative rate of Hg bioaccumulation in focal biota. Sediment TOC and % silt–clay explained ~30% of the variation in $BSAF_{(Hg)}$ across taxa, and the estimated coefficients for both variables were negative in the regression model (Table 2; Fig. 6A and B). As previously discussed, mercury effectively binds to organic material and fine-grained substrates, thus reducing the bioavailability of mercury for biota (Chen et al., 2008). BSAFs are therefore expected to decrease in environments characterized by substrates with high organic content and small grain sizes, as verified in several field and laboratory investigations (Muhaya et al., 1997; Mason and Lawrence, 1999; Lawrence and Mason, 2001; Chen et al., 2009). Sediments with high TOC are also expected to reduce the DO concentration of overlying water because of increased bacterial respiration of the organic substrate (Diaz and Rosenberg, 1995). This response may explain the positive relationship observed between BSAFs and bottom water DO (Table 2; Fig. 6C and D), i.e., the apparent effect of DO on biotic mercury accumulation is an artifact of sediment TOC simultaneously reducing Hg bioavailability and water oxygen content. Alternatively, the positive BSAF–DO relationships may be caused by the direct response of biota to varying oxygen levels. Most notably, DO concentrations <4 mg/L potentially depresses feeding rates in estuarine biota (Breitburg et al., 1994; Sobral and Widdows, 1997; Rosas et al., 1998; Sagasti et al., 2001), but the concentration at which this sub-lethal effect is realized varies among taxa (Diaz and Rosenberg, 1995). Given that

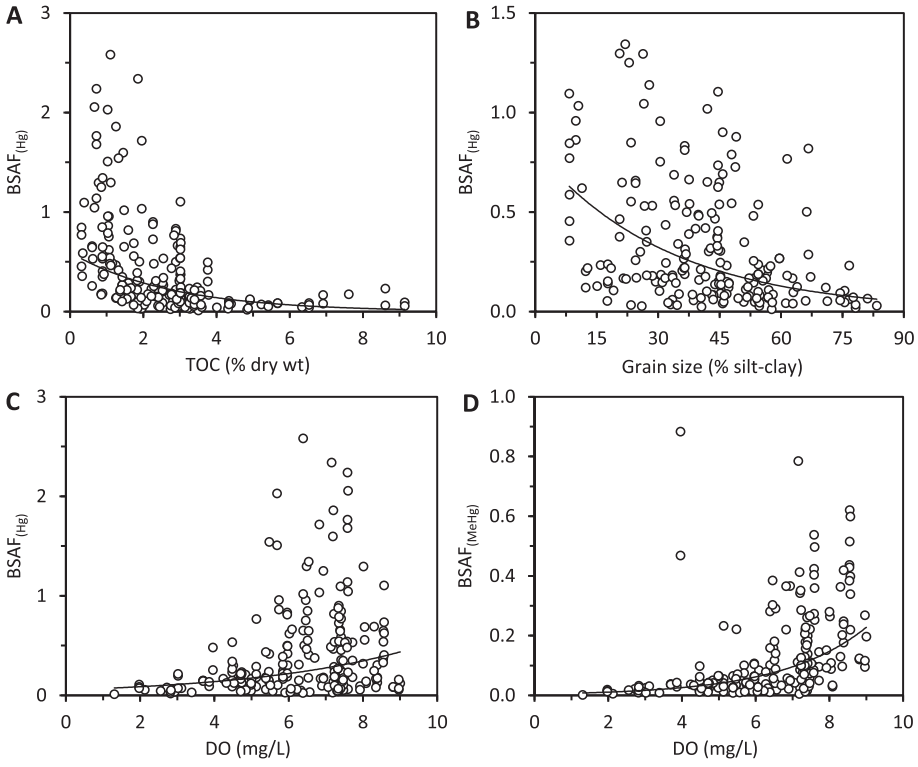


**Fig. 6.** Biota-sediment accumulation factors ($BSAF_{(Hg)}$ and $BSAF_{(MeHg)}$ = biota total Hg/sediment Hg or MeHg) as a function of site-specific geochemical conditions, including total organic carbon (TOC, % dry wt.) (A), grain size (% silt–clay) (B), and bottom water dissolved oxygen (DO, mg/L) (C, D). Least-squares exponential regression models were fit to data ($n = 218$).

**Table 5**
Parameter estimates (standard error; SE) and summary statistics for least-squares linear regression models used to analyze total mercury (Hg) concentrations of biota as a function of normalized sediment total Hg and methylmercury (MeHg) content.

| Relationship | Model parameters | | *F* | *df* | $R^2$ | *p* |
|---|---|---|---|---|---|---|
| | Intercept | Slope | | | | |
| *Zooplankton* | | | | | | |
| Sediment Hg | 0.02 (0.01) | 0.13 (0.05) | 7.56 | 1, 36 | 0.178 | <0.01 |
| Sediment MeHg | 0.04 (0.02) | 0.01 (0.02) | 0.25 | 1, 36 | 0.007 | 0.622 |
| *Polychaete* | | | | | | |
| Sediment Hg | −0.12 (0.10) | 1.68 (0.41) | 16.45 | 1, 14 | 0.559 | <0.005 |
| Sediment MeHg | −0.05 (0.13) | 0.37 (0.15) | 6.42 | 1, 14 | 0.331 | <0.05 |
| *Gastropod* | | | | | | |
| Sediment Hg | −0.05 (0.07) | 1.23 (0.30) | 16.21 | 1, 13 | 0.575 | <0.005 |
| Sediment MeHg | 0.18 (0.15) | −0.01 (0.14) | 0.001 | 1, 13 | 0.0001 | 0.972 |
| *Bivalve* | | | | | | |
| Sediment Hg | 0.07 (0.02) | 0.28 (0.09) | 8.70 | 1, 32 | 0.219 | <0.01 |
| Sediment MeHg | 0.12 (0.03) | 0.003 (0.03) | 0.007 | 1, 32 | 0.0002 | 0.932 |
| *Amphipod-Isopod* | | | | | | |
| Sediment Hg | 0.06 (0.02) | 0.13 (0.06) | 5.61 | 1, 7 | 0.483 | <0.05 |
| Sediment MeHg | 0.11 (0.04) | −0.04 (0.06) | 0.53 | 1, 7 | 0.081 | 0.495 |
| *Shrimp* | | | | | | |
| Sediment Hg | 0.08 (0.02) | 0.17 (0.07) | 6.52 | 1, 39 | 0.147 | <0.05 |
| Sediment MeHg | 0.12 (0.02) | 0.01 (0.02) | 0.25 | 1, 39 | 0.007 | 0.617 |
| *Crab* | | | | | | |
| Sediment Hg | 0.09 (0.03) | 0.12 (0.11) | 1.05 | 1, 34 | 0.031 | 0.313 |
| Sediment MeHg | 0.08 (0.03) | 0.04 (0.03) | 1.81 | 1, 34 | 0.052 | 0.188 |
| *Killifish* | | | | | | |
| Sediment Hg | −0.01 (0.05) | 0.79 (0.13) | 35.38 | 1, 35 | 0.510 | <0.0001 |
| Sediment MeHg | 0.12 (0.06) | 0.13 (0.06) | 4.36 | 1, 35 | 0.114 | <0.05 |

*Note:* Sediment mercury concentrations were normalized by total organic carbon.

biota are exposed to MeHg principally through their diet (Hightower and Moore, 2003), environmental conditions that decrease foraging activity will likely have a concomitant effect on biotic mercury uptake.

### 3.5. Biota as indicators of environmental mercury contamination

Establishing associations between biota and sediment mercury concentrations often requires normalizing data to minimize the effect of confounding factors (Mason and Lawrence, 1999). In this study, the total Hg of focal biota were analyzed relative to sediment total Hg and MeHg that were normalized *a posteriori* by sediment organic content (TOC). Accordingly, the total Hg of most taxa was related to normalized sediment total Hg (Table 5; Fig. 7), indicating that these organisms were suitable indicators of environmental Hg contamination. Importantly, however, the amount of biotic Hg variability explained by sediment Hg differed considerably among taxa, and thus, species selection changes the predicative capability of a given regression model. Research in other estuarine and coastal environments reported similar biota–sediment mercury relationships (Hammerschmidt and Fitzgerlad, 2004), and recent advances in mercury isotope analysis have verified that sediments are a dominant source of Hg to biota (Gehrke et al., 2011a). Other investigations, however, have reported no relationship between sediment and biota Hg, including the whelk *Nassarius reticulates* (Coelho et al., 2006), amphipod *Leptocheirus plumulosus* (Lawrence and Mason, 2001), and polychaete *Nereis diversicolor* (Muhaya et al., 1997). Similarly, in this study, no significant relationship was detected between the total Hg of crabs and sediments (Fig. 7G), and is likely due to species-specific variations in Hg content within this taxon (Table 3). Moreover, in most instances, the total Hg of focal taxa was unrelated to sediment MeHg, with only weak positive relationships detected for killifish and polychaetes (Table 5). MeHg accounts for a relatively small percentage of total Hg in low trophic level organisms, particularly benthic infauna and epifuana (% MeHg typically <40%; Chen et al., 2009), and this may partially explain the general dissociation between biota total Hg and sediment MeHg. Percent MeHg is also highly variable within estuarine species (Chen et al., 2009), thus making it difficult to detect direct relationships between biota and sediment mercury contamination.

## 4. Summary

Spatial patterns in sediment total Hg in Narragansett Bay were governed by the distribution of TOC, indicating that inorganic Hg has a high affinity for organic matter. The methylation of sediment inorganic Hg, in turn, was inversely related to bottom water DO, which is presumably caused by the accelerated activity of sulfate-reducing bacteria in oxygen-depleted waters. Moreover, sediment MeHg was positively related to TOC and further suggests that organic matter in surface sediments can effectively scavenge and retain mercury from the water column, or alternatively, organic carbon stimulates methylating bacteria by acting as a substrate for mineralization.

The efficacy with which mercury was transferred from surface sediments to estuarine biota was assessed through $BSAF_{(Hg)}$ and $BSAF_{(MeHg)}$. Accordingly, BSAFs differed significantly across taxa and were elevated in: (1) benthic infauna that experience additional uptake of mercury via direct contact with sediments and pore water, and (2) biota that maintain a relatively high trophic position in the food web. Among all taxa, BSAFs were negatively related to sediment TOC and % silt–clay composition, indicating that organic matter and fine-grain substrates reduce mercury bioavailability. Moreover, BSAFs were positively related to bottom water DO, and may result from biota decreasing feeding rates in low DO waters, thereby reducing dietary uptake of mercury.

The utility of estuarine biota as bio-indicators of environmental mercury contamination was assessed by regressing taxon-specific Hg concentrations with TOC-normalized sediment total Hg and MeHg. With the exception of crabs, the total Hg of focal taxa was positively related to sediment total Hg, demonstrating that select biota were suitable indicators of environmental Hg contamination. There was a general dissociation between biotic Hg and sediment

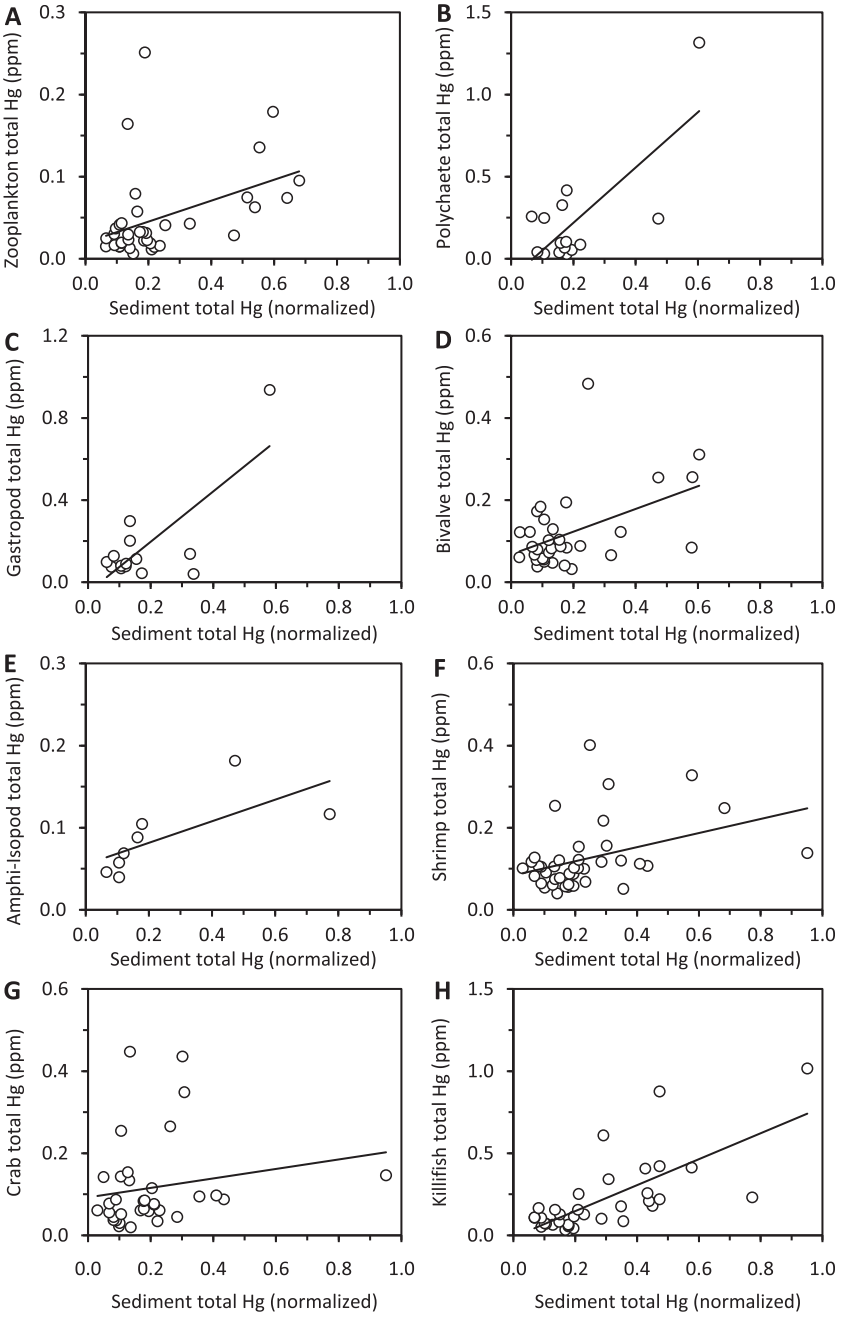


**Fig. 7.** Biotic total mercury concentrations (Hg; ppm dry wt.) as a function of sediment total Hg (Hg; ppm dry wt.) normalized by total organic carbon (% dry wt.). Biota include zooplankton (A; $n = 37$), polychaetes (B; $n = 15$), gastropods (C; $n = 14$), bivalves (D; $n = 33$), amphipods-isopods (E; $n = 8$), shrimp (F; $n = 40$), crabs (G; $n = 35$), and killifish (H; $n = 36$). All data points represent the mean total Hg of individuals collected from a single site (average ~10 individuals per site; range = 2–49 individuals per site). Least-squares linear regression models were fit to the data.

MeHg, however, and is likely due to MeHg comprising a small percentage of the organismal total Hg burden. Estuarine biota may therefore represent important targets for mercury monitoring programs, although the utility of candidate bio-indicators varies according to their inter-specific life history traits, as well as the mercury species of interest.

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