Estuarine, Coastal and Shelf Science 200 (2018) 116–125

Contents lists available at ScienceDirect

Estuarine, Coastal and Shelf Science

journal homepage: www.elsevier.com/locate/ecss

Mercury biomagnification through food webs along a salinity gradient down-estuary from a biological hotspot

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ARTICLE INFO

Article history:
Received 27 May 2017
Received in revised form
1 September 2017
Accepted 25 October 2017
Available online 28 October 2017

Keywords:
Trophic magnification slope
Basal mercury
Carbon and nitrogen stable isotopes

Regional index terms:
USA
Everglades National park
Shark River Estuary

ABSTRACT

To examine down-estuary effects and how differences in food webs along a salinity gradient might influence mercury (Hg) biomagnification, we conducted a study from 2010 to 2015 in an estuary with a known biological hotspot at its headwaters. Over 907 samples of biota, representing 92 different taxa of fish and invertebrates, seston and sediments were collected from the upper, middle and lower reach for Hg determination and for stable nitrogen and carbon isotope analyses. Trophic magnification slopes (TMS; log Hg versus δ15N), as a measure of biomagnification efficiency, ranged from 0.23 to 0.241 but did not differ statistically among reaches. Hg concentrations were consistently highest, ranging as high as 4.9 mg/kg in top predatory fish, in the upper-reach of the estuary where basal Hg entering the food web was also highest, as evidenced by methylmercury concentrations in suspension feeders. Top predatory fish at the mouth of the estuary contained relatively low [THg], likely due to lower basal Hg. This was nonetheless surprising given the potential for down-estuary biotransport.

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1. Introduction

Mercury (Hg) is a pollutant of global concern due to its potential for long-range atmospheric transport and subsequent microbial methylation (Driscoll et al., 2013). The neurotoxic effects of methylmercury (MeHg), the form of Hg that biomagnifies through food webs, are well documented (NRC, 2000). The principal pathway for MeHg exposure to humans is through the consumption of marine fish (Sunderland, 2007). This is of particular importance in the Gulf of Mexico (GOM) because its seafood is known to have elevated Hg levels prompting consumption advisories by all five U.S. Gulf states (Harris et al., 2012). Recent surveys have identified south Florida's coastal environment as a regional "hotspot" where fish have higher Hg concentrations than surrounding areas (Adams et al., 2003; Evans et al., 2015; Evans and Crumley, 2005; Rumbold et al., 2014). One such hotspot is the headwaters of the Shark River Estuary in Everglades National Park where largemouth bass (*Micropterus salmoides*) have consistently had some of the highest Hg levels in Florida for more than a decade (Gu et al., 2012), invariably exceeding benchmarks for the protection of both piscivorous wildlife and humans (USEPA, 1997; 2001).

Yet, many uncertainties remain regarding the relative importance of the different factors controlling Hg entry and biomagnification through marine food webs (Chen et al., 2008), particularly in the GOM (Harris et al., 2012). Differences in inorganic Hg loading, its bioavailability to Hg methylating bacteria, and rates of bacterial activity are all thought to be sources of the extreme spatial variability that can result in hotspots (Chen et al., 2012; Heyes et al., 2006). Additionally, the natural variation in trophic structure among communities (e.g., food chain length, linkage strength, etc.), degree of openness, and primary and secondary productivity can also influence the rate of MeHg biomagnification (Chen et al., 2008; Kidd et al., 2012a). The effect that community structure has on MeHg biomagnification efficiency in GOM food webs remains poorly characterized.

Recently, several investigations have quantified loads of both total-Hg (THg) and MeHg through estuaries to the GOM (Bergamaschi et al., 2012; Buck et al., 2015; Rumbold et al., 2011). While these studies have revealed important information that will improve and constrain models, they have focused on advection in water or on sediment. Little attention has been given to the

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https://doi.org/10.1016/j.ecss.2017.10.018
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potential importance of biological transport or "bioadvection" of Hg through estuaries (Chen et al., 2008; Harris et al., 2012). There is also growing interest in how differences in food web structure among different habitats, as might be expected along an estuarine salinity gradient (Bulger et al., 1993; Ley et al., 1999; Cloern et al., 2017), influence Hg biomagnification (Jardine et al., 2006; Lavoie et al., 2013).

The purpose of our study was twofold: (1) determine if the Hg hotspot known to occur at the headwaters extends down-estuary and, (2) by using a stable isotope analysis approach, assess the relative importance of basal Hg and food web structure along the salinity gradient as drivers of Hg biomagnification. Additionally, there was also a desire to generate a data set that could be used, in the future, to improve existing food web models (Harris et al., 2012; Evans et al., 2015).

2. Methods

2.1. Study area

The study took place in the Shark River Estuary (SRE) within Everglades National Park (ENP) located in south Florida, USA (Fig. 1). The SRE is located at the terminus of the major historical flow pathway through the Everglades Ecosystem (Childers et al., 2006) and, as part of a national park, remains relatively undisturbed. Yet, since the late 1980s fish and wildlife from ENP and the rest of the Everglades have been known to contain high Hg levels (Ware et al., 1990). More recent surveys of Hg in ospreys (*Pandion haliaetus*) and dolphins (*Tursiops truncatus*) from this area continue to report higher levels than conspecifics from other parts of North America (Rumbold et al., 2016; Damseaux et al., 2017). Multi-agency research indicates the source of the Hg is from high atmospheric deposition, reflecting a combination of high rainfall with high inorganic Hg concentrations (Guentzel et al., 2001; Gu et al., 2012). The upstream slough of the SRE is dominated by freshwater marsh and splits into a wide band of mangrove-dominated creeks within 15 km of the GOM. Although microtidal, a large expanse of mangrove-dominated ecotone is inundated and drained during each tide due to the low topography. Species of fish are known to form distinct assemblages along salinity gradients of southwest Florida estuaries due to taxon-specific (and life stage) salinity tolerances (Ley et al., 1999; Green et al., 2006). Yet mobile animals may move with the natural isohaline movement along the main stem of the river and/or move in and out of marshes and creeks or "backwaters" as water levels and salinity permits (Boucek and Rehage, 2013; Green et al., 2006; Rehage and Loftus, 2007). Boucek and Rehage (2013), for example, have tracked fish and invertebrates that move down into the upper SRE from adjacent drying marshes, returning to the marshes when they flood.

SRE has been characterized as a relatively oligotrophic, upside

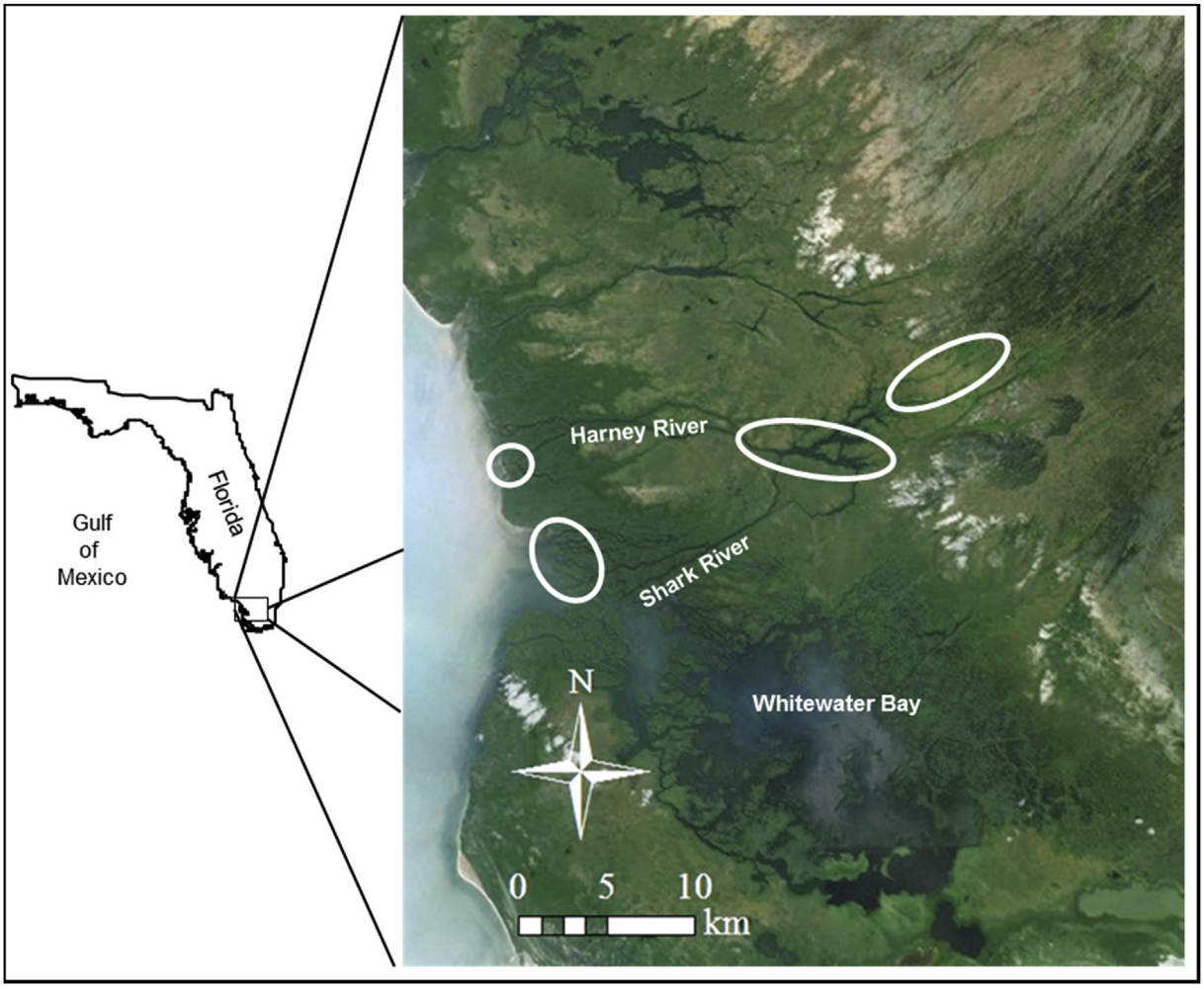

Fig. 1. Map and satellite image showing approximate sampling areas in upper, middle and lower reach of Shark River Estuary (since they share the same watershed, the mouth of the Harney River was included in the lower reach).

down estuary with total phosphorus (as the limiting nutrient) increasing down-estuary (Childers et al., 2006). Salinity, total nitrogen (TN) and total phosphorus (TP) for the different reaches are summarized in Supplemental Table S1.

2.2. Sampling protocol

Sampling took place from September 2010 through May 2015 (semi-annually during the first 4 years and quarterly in the final year) in three reaches (upper-, mid- and lower-estuary). This protracted sampling period was necessary to allow for the collection of sufficient samples from representative trophic levels, particularly invertebrates; while largemouth bass are sampled at the upstream site as part of the state's long-term monitoring program, temporal trend analysis was not a primary objective of this study. Over the study period, over 907 samples were collected for analyses (i.e., small organisms were occasionally composited into a single sample to increase mass); 73% of the samples (including all sediment, seston and invertebrates except oysters) were collected during the final year of the study, when additional funding became available. Of these, 893 samples were biota from 92 different taxa (most identified to species but a few only to genera or family). While the vast majority of the biota were fish, 11% were comprised of 23 different macroinvertebrate taxa; common names and species of collected fish and macroinvertebrates are summarized, by reach, in Supplemental Tables S2–S4.

Fish were collected primarily using hook and line, seine nets, minnow traps and, where conductivity was low (upper- and mid-reach during the wet season), using boat-mounted, pulsed DC electro-fishing equipment. Macroinvertebrates were also collected in seine nets, crab traps and occasionally during electrofishing. Three top predatory fish species: 1) largemouth bass, 2) common snook, and 3) mangrove snapper were more heavily targeted than other fishes for several reasons. First, snook and mangrove snapper were relatively abundant within all three reaches. Second, snook were the focus of other studies in SRE, which should provide additional life-history information. Third, as mentioned above, largemouth bass have been used as sentinels for long-term Hg monitoring throughout south Florida (Gu et al., 2012).

Seston samples ($n = 10$) and sediment samples ($n = 14$) were collected during the final year. Seston was collected using a plankton net at each station (30 cm dia. 5:1, 35 μm mesh net with Stainless Steel ring; SEA-GEAR Corporation, Melbourne, FL). Sediment samples were collected most often using a clean butyrate core tube (from approximately the top 4 cm). Occasionally, where the sediment layer was thin over rock, it was collected by simply scooping the top 4 cm into a clean sampling jar or, on two occasions, by using a plastic bag as a glove. Both seston and sediments were stored frozen (for < 6 months) in glass jars or scintillation vials until processing.

Field collection and sample processing were conducted with great care to minimize sample contamination by exogenous Hg (particularly of inorganic Hg due to its numerous sources) and from sample carryover (i.e., from one sample to the next during processing). This was a particular concern for seston given their low THg concentrations. Accordingly, patterns of Hg in sediments, invertebrates and seston were evaluated based on measured MeHg (which has fewer contamination sources) rather than THg.

2.3. Processing samples

Total length and weight of individual fish were determined using a measuring board or tape (±0.1 cm) and digital balance (±0.1 g) or Pesola spring scale (±10 g). Invertebrates were also weighed and measured prior to soft muscle tissue removal from exoskeleton or shell. All invertebrates and smaller species of prey fish were processed as whole body homogenates using a commercial meat grinder (Hobart Meat Chopper, Beltram South, Fort Myers, FL), laboratory grinder (Grindomix GM200, Retsch, U.S.), hand-held homogenizer (Tissue Master 125, Omni International, Kennesaw, GA) or some combination. Alternatively, due to the impracticalities of homogenizing larger samples, only a portion of the fillet was analyzed for larger sport fish species.

Sediment and seston were oven-dried at 60 °C with percent solids determined. Dried sediments were then combusted at 550 °C for 4 h to determine percent organics.

All equipment (cutting board, knife, grinders) were washed with ambient water (if processed in the field) or tap water between each use to avoid cross-contamination. The vast majority of samples were analyzed individually; however, small organisms (e.g., shrimp, snails, small prey fish) were occasionally composited into a single sample to increase mass; all bivalve species (e.g., oysters, clams, mussels, etc.) were also composite samples of up to 10 individuals. All samples were stored in scintillation vials at −20 °C and analyzed within six months.

Several previous studies have similarly used whole-body concentrations of invertebrates and small prey fish species at the bottom of the food web in combination with muscle tissues of larger organisms at the top of the food web (Chumchal et al., 2011; Riget et al., 2007; Thera and Rumbold, 2014). This introduces uncertainty because, although highly correlated, Hg concentration in whole-body tends to be lower than muscle tissue alone (Bank et al., 2007; Peterson et al., 2005). Nonetheless, in accordance with previous studies, adjustments were not made based on tissue type.

2.4. Analytical methods

Total-Hg concentration (includes all Hg species; hereafter designated as [THg]) was determined in all matrices (e.g., fish, invertebrates, and sediment) at Florida Gulf Coast University via thermal decomposition, gold amalgamation, and atomic absorption spectrometry (EPA method 7473) with a direct Hg analyzer (Nippon Model MA-2000 from 2010 to 2103 and Milestone DMA-80 from 2014 to 2015). Calibration curves were generated using varying masses of Certified Reference Materials (CRMs; National Research Council Canada, Institute for National Measurement Standards, Ottawa, ON, Canada): DOLT-3 (Dogfish Liver), DORM-3 (Fish Protein) or DORM-4 (Fish Protein) and BCR-463 and ERM-CE464 (LGC Standards USA, 276 Abby Road, Manchester, NH). These same CRMs were also used for continuing calibration verification at the start and at the end of every batch of 20 samples; laboratory duplicates were also run during each batch.

Because the vast majority of THg in the muscle tissue of fish is in the form of MeHg (Grieb et al., 1990), the analysis of fish for THg is interpreted as being equivalent to MeHg. Alternatively, the percentage of THg present as MeHg is more variable in invertebrates (Thera and Rumbold, 2014). Accordingly, MeHg was also determined for all sediments, seston, most bivalves and representative samples of other invertebrate taxa (summarized in Table S5, Supplemental Information) by the USGS Wisconsin Mercury Research Laboratory (Middleton, Wisconsin). For those few invertebrates where [MeHg] was not measured, it was estimated based adjusting measured [THg] by %MeHg determined in representative invertebrates. Quality assurance of Hg analysis is reviewed in Supplemental Information.

For determination of stable nitrogen (δ15N) and carbon (δ13C) isotopes, subsamples of tissues (i.e., homogenates or fillets) and seston were dried at 60 °C to constant weight (>48 h), and then ground to a fine powder using a mortar and pestle. Dried samples were later acidified with 10% hydrochloric acid to remove

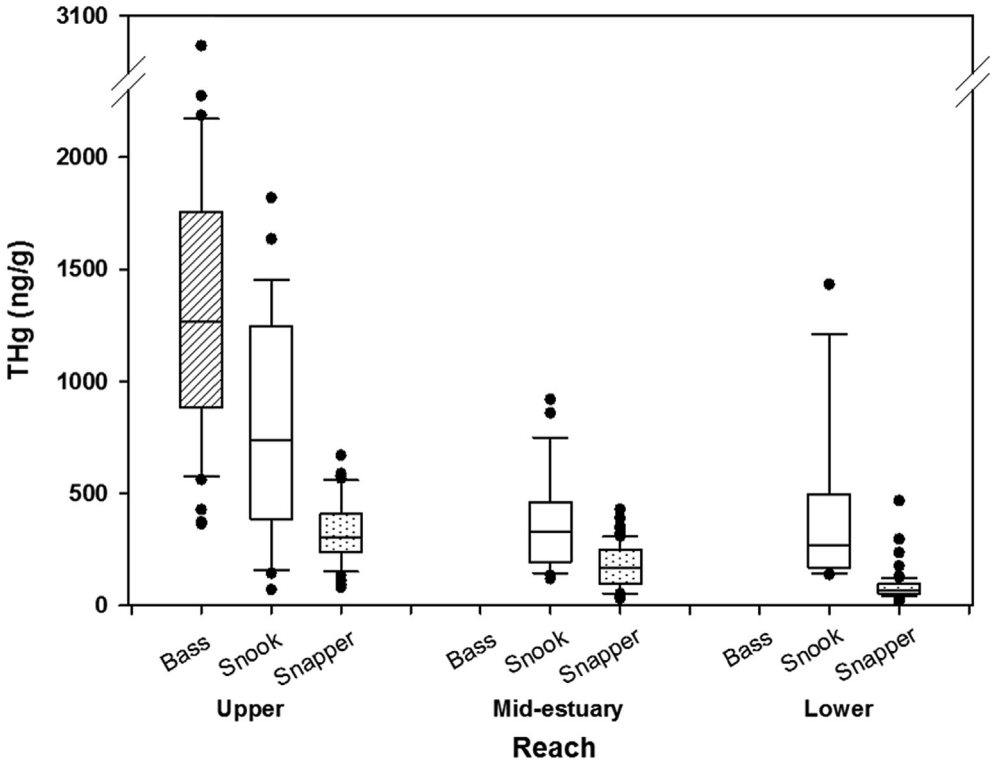

Fig. 2. Boxplot of [THg] (ng/g wet wt.) in predatory fish from different reaches of the estuary. Note, bass were only caught in the upper reach. The boundary of the box closest to zero indicates the 25th percentile, the line within the box, the median, and the boundary of the box farthest from zero, the 75th percentile. Whiskers above and below the box indicate the 90th and 10th percentiles with symbols representing outliers.

carbonates and re-dried; lipids were not removed from samples. Aliquots of the final powder were then weighed into tin capsules (Costech, Valencia, CA) and shipped to a contract laboratory for analyses. The vast majority of samples for isotope analysis were shipped to the University of California Davis' Stable Isotope Facility (Davis, California, USA; 33 samples were sent to the Yale Stable Isotope Facilities in 2012 and 20 samples were sent to University of South Florida Stable Isotope Lab in 2013). Quality assurance of the isotope analysis is reviewed in Supplemental Information.

Stable isotope analysis (SIA) of nitrogen and carbon has been employed to investigate trophic position and community dynamics in a variety of ecosystems including estuaries and, more recently, utilized to assess both the efficiency by which Hg biomagnifies through different food webs (for review, see Jardine et al., 2006). Because δ15N ratios can vary over space or time due to anthropogenic inputs, previous studies have sometimes standardized δ15N based on basal values in a local, primary consumer (for review, see Jardine et al., 2006). These studies often then used an average diet-tissue discrimination factor (or enrichment factor) to translate δ15N to trophic level to estimate a food web magnification factor (FWMF). This allowed for estimation of basal Hg entering the food web based on the y-intercept. There is a growing debate, however, about the choice of an "averaged" diet-tissue discrimination factor (Wolf et al., 2009) and, therefore, some recent studies have explicitly not translated δ15N into trophic level (Kidd et al., 2012b). Standardizing the δ15N changes the intercept but does not change the slopes (i.e., TMS). Accordingly, we did not standardize or adjust δ15N. Rather than estimating basal Hg from the y-intercept of a FWMF, which is very sensitive to the slope, we relied on [MeHg] measured in primary consumers and to a lessor extent seston and sediments.

2.5. Data analysis

Patterns of Hg in sediments, seston and bivalves from different reaches were based on measured [MeHg]. Where comparisons were made to THg in fish (i.e., the relation between log [THg] and δ15N for entire food web), THg in invertebrates were adjusted to estimate MeHg so as to be equivalent to fish (i.e., adjusting THg concentrations determined by the DMA-80 using %MeHg in representative species).

Unless otherwise noted (as above), all [THg] were reported in ng/g on a wet weight basis. [THg] in top predatory fish caught from different reaches were compared using parametric and nonparametric tests. Because [THg] often increases with increasing fish size (within species), analysis of covariance (ANCOVA), with size as a covariate, was used to assess differences in [THg] where average size (i.e., total length) of sampled fish populations differed. However, ANCOVA use is predicated on several critical assumptions including sufficient sample size and range in the covariate, and homogeneity of slopes (Zar, 1999) that was not always achieved in this study. In those cases, we resorted to using a one-way analysis of variance (ANOVA).

Simple linear regression analysis was used to describe the relationship between log[THg] versus δ15N for food webs in each of the reaches. ANCOVA was used to determine if slopes of these relationships (hereafter termed trophic magnification slope or TMS) differed.

3. Results

3.1. [THg] in top predatory fish

[THg] in individual samples of fish collected from 2010 to 2015

spanned four orders of magnitude and ranged from 0.45 ng/g in a mullet to 4946 ng/g in a bowfin. Although temporal trend analysis was not a primary objective, because samples were collected over a 5 year period, possible temporal trends in [THg] were first assessed using the three fish species caught most frequently (results presented in Supplemental Information). Because there was no clear temporal trends, data were pooled across years.

[THg] differed among top predator fish in the upper reach (Fig. 2; Kruskal-Wallis $H = 53.3$, $df = 2$, $p < 0.001$) with [THg] in bass > snook > snapper (all pairwise comparisons were significant, $p < 0.05$). More importantly, [THg] declined markedly in these fish down-estuary (Fig. 2). For instance, [THg] was significantly higher in snook from the upper reach as compared to snook from mid-estuary (2.2x higher) and from the mouth (2.7x higher, Fig. 2; Kruskal-Wallis; $H = 10.6$; $df = 2$; $p = 0.005$ with post-hoc, pairwise comparisons between upper estuary versus both the other reaches significant, Dunn's test, $p < 0.05$); these snook did not differ in size (Kruskal-Wallis; $H = 1.32$; $df = 2$; $p = 0.52$).

Likewise, upper-reach snappers contained significantly higher [THg] than snappers from mid-estuary (1.8x) or the mouth (4.5x, Kruskal-Wallis; $H = 93.9$; $df = 2$; $p < 0.001$; Dunn's post-hoc $p < 0.05$ all pairwise comparisons); size differences among these fish were only minor ($H = 5.9$; $df = 2$; $p = 0.052$) and involved snappers from mid-estuary versus lower estuary.

3.2. Basal Hg: suspension feeders, seston, and sediment

Because the same bivalve species were not found in all three reaches, oysters represented primary consumers in the lower estuary while mussels and clams were utilized in the mid and upper estuary, respectively. [MeHg] measured in dried samples (determined by USGS) were much higher in Carolina marsh clams from the upper reach than bivalves from the other two reaches (Kruskal-Wallis, $H = 7.6$, $df = 2$, $p = 0.006$; Dunn's pairwise comparisons all significant, Table 1). In general, [MeHg] in bivalves exhibited similar spatial patterns. The Carolina marsh clams from the upper estuary contained higher [MeHg] than conspecific clams and Scorched mussels from mid-estuary and oysters from the lower estuary (Fig. 4, Supplemental Tables S2–S4). Alternatively, Ribbed mussels from the mid-estuary had higher [MeHg] than either the clams or oysters (Fig. 4, Supplemental Tables S2–S4).

Similar to the Carolina marsh clams, seston collected in the upper reach contained the highest [MeHg] compared to other areas, although small sample size does not allow for robust statistical analysis. [MeHg] also declined down-estuary in seston (Table 1).

Sediment samples from the upper reach had relatively high [MeHg] but not as high as sediments from the mid-estuary (Table 2). Both were much higher in [MeHg] than sediments from the lower estuary that contained much less organic matter (Table 2).

3.3. Spatial patterns in δ15N and δ13C

Cross-plots of δ15N versus δ13C revealed interesting isotope ratios in biota from the three reaches (Fig. 3). δ15N values generally declined down-estuary (Fig. 3, Table 1 and Supplemental Fig. S2). The δ13C of the food web also shifted down-estuary with biota from the lower estuary less negative as compared to biota from the upper reach (Fig. 3, Supplemental Fig. S2 and Tables S2–S4). While seston and oysters collected from the lower reach had similar δ13C ratios, values differed between seston and clams or mussels from other reaches with clams and mussels less enriched in 13C than corresponding seston (Fig. 3, Table 1). Correlations between δ15N and δ13C, and between stable isotopes and fish size were evaluated only for top predatory fish (e.g., bass, snook and snapper; results reported in Supplemental Information).

It was noteworthy that the range of δ13C values was narrowest in bass (found only in the upper estuary) whereas both snook and snappers showed more overlap in δ13C values from the upper- and mid-reaches than the lower reach (Supplemental Fig. S2).

Table 1
Average [MeHg] (dry wt., measured by USGS), δ15N, and δ13C (mean, ±1SD, n) in seston and suspension feeders from different reaches of Shark River Estuary; range given where n = 2.

Reach	Common name	MeHg (ng/g)	δ15N (‰)	δ13C (‰)
Upper	Seston	14.1 (±3.4, 2)	5.9 (±1.2, 3)	−30.6 (±0.6, 3)
	Carolina Marsh Clam	207.7 (±15.9,3)	10.4 (±0.14, 3)	−33.7 (±0.1, 3)
Mid-estuary	Seston	8.4 (±2.5, 2)	5.5 (±0.6, 3)	−30.1 (±0.6, 3)
	Scorched mussels	29.7 (±8.1, 3)	7.9 (±0.4, 3)	−35.7 (±0.46, 2)
Lower	Seston	4.4 (±2.2, 3)	4.5 (±2.1, 3)	−28.3 (±2.0, 3)
	Eastern Oyster	34.9 (±3.5, 6)	5.9 (±0.7, 13)	−29.1 (±1.2, 13)

Table 2
Average [THg], [MeHg] (dry wt, measured by USGS; ±1SD, n), %MeHg and % organics in sediments (i.e., loss of mass from combusting dried sediment for 4 h at 550 °C) collected from the different reaches of Shark River Estuary.

Reach	n	THg (ng/g)	MeHg (ng/g)	%MeHg	% organics
Upper	6	301.4 (±147.4)	2.55 (±0.83)	1.1% (±0.9)	75.8% (±12.4)
Mid-estuary	4	98.7 (±37.8)	6.11 (±2.37)	6.4% (±1.7%)	77.7% (±4.4)%
Lower	4	23.1 (±22.7)	0.67 (±0.41)	4.6% (±2.4%)	18.0% (±15.9%)

3.4. Trophic magnification slopes

While generally fish had higher average [THg] and were more enriched in 15N than macroinvertebrates (Fig. 4, Supplemental Tables S2–S4), there were some notable exceptions. For example, several of the crab species (particularly blue crabs, Supplemental Tables S2–S4) had [THg] and δ15N values similar or higher than some of the forage fish (Fig. 4). But even more noteworthy was that suspension feeders (e.g., clams and mussels) from the upper- and mid-estuary had δ15N values higher than several fish species (Figs. 3 and 4).

Macroinvertebrates were caught and sampled more heavily in the lower reach (Fig. 4). To avoid any potential bias, regressions of log [THg] versus δ15N were based on species means for fish only (using means rather than individual data points should also avoid bias due to uneven sampling of fish species). The resulting regressions were statistically significant in all three reaches (Fig. 4; upper: $F = 11.5$; $df = 1,24$; $p = 0.002$; mid: $F = 8.8$; $df = 1,17$; $p = 0.009$; lower: $F = 52.8$; $df = 1,37$; $p < 0.001$). While TMS decreased down-estuary (Fig. 4) the differences were not statistically significant (ANCOVA interaction: $F = 0.008$; $df = 2, 78$; $p = 0.9$). To improve statistical power, the regressions were re-calculated and slopes defined for each reach based on raw, individual data points (fish only) rather than means (Supplemental Fig. S1). The resulting slopes based on individual data points were generally similar to those based on means and again did not differ among reaches (ANCOVA interaction: $F = 0.935$; $df = 2, 592$; $p = 0.39$).

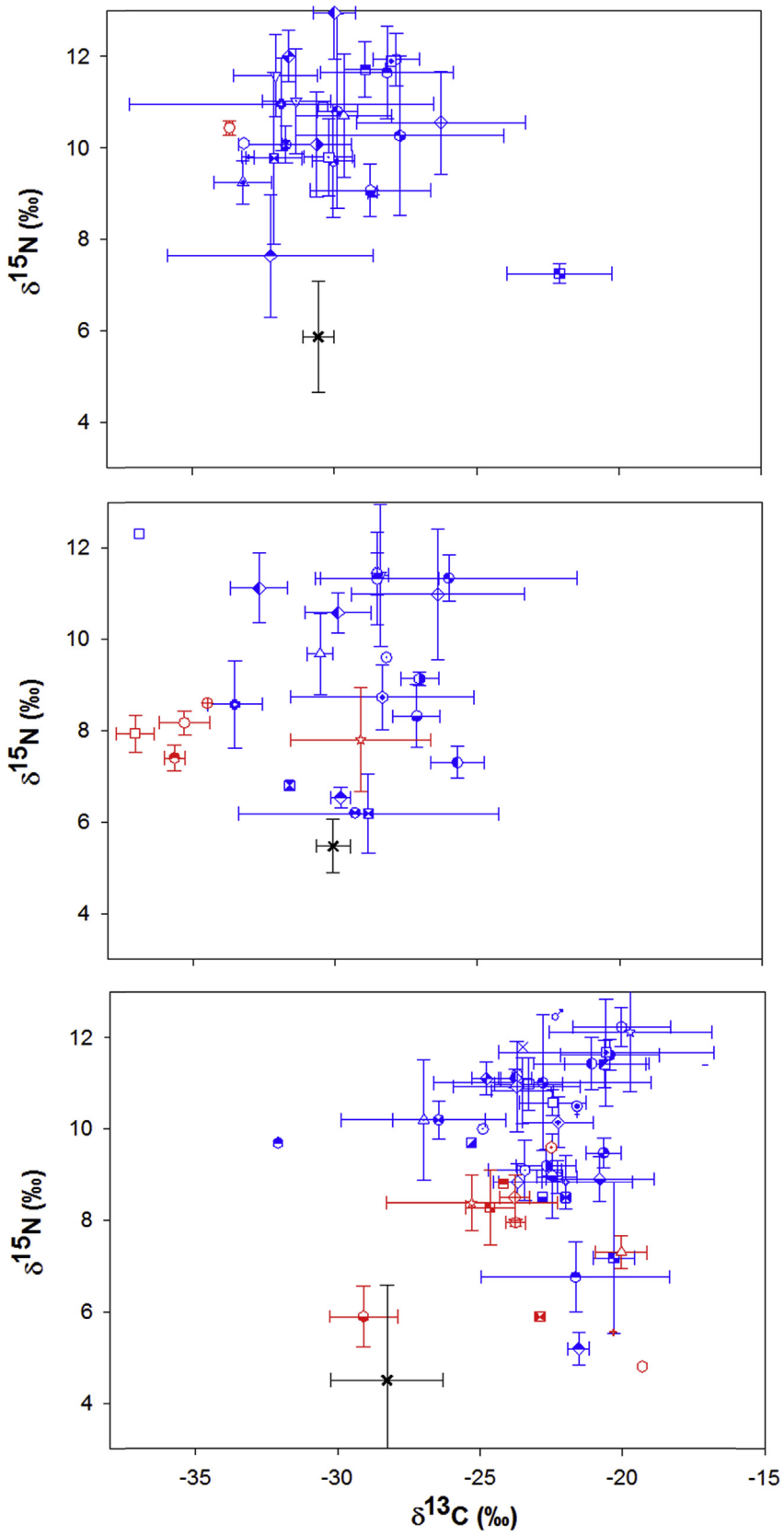

Fig. 3. Cross-plots of δ15N versus δ13C (species means ± 1SD unless n < 3 where range is shown) for upper (top panel), middle and lower reaches (bottom panel) of the Shark River Estuary (for detailed analysis of cross-plots for bass, snook and snapper, see Supplemental Information); blue symbols represent fish, red inverts and black seston. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

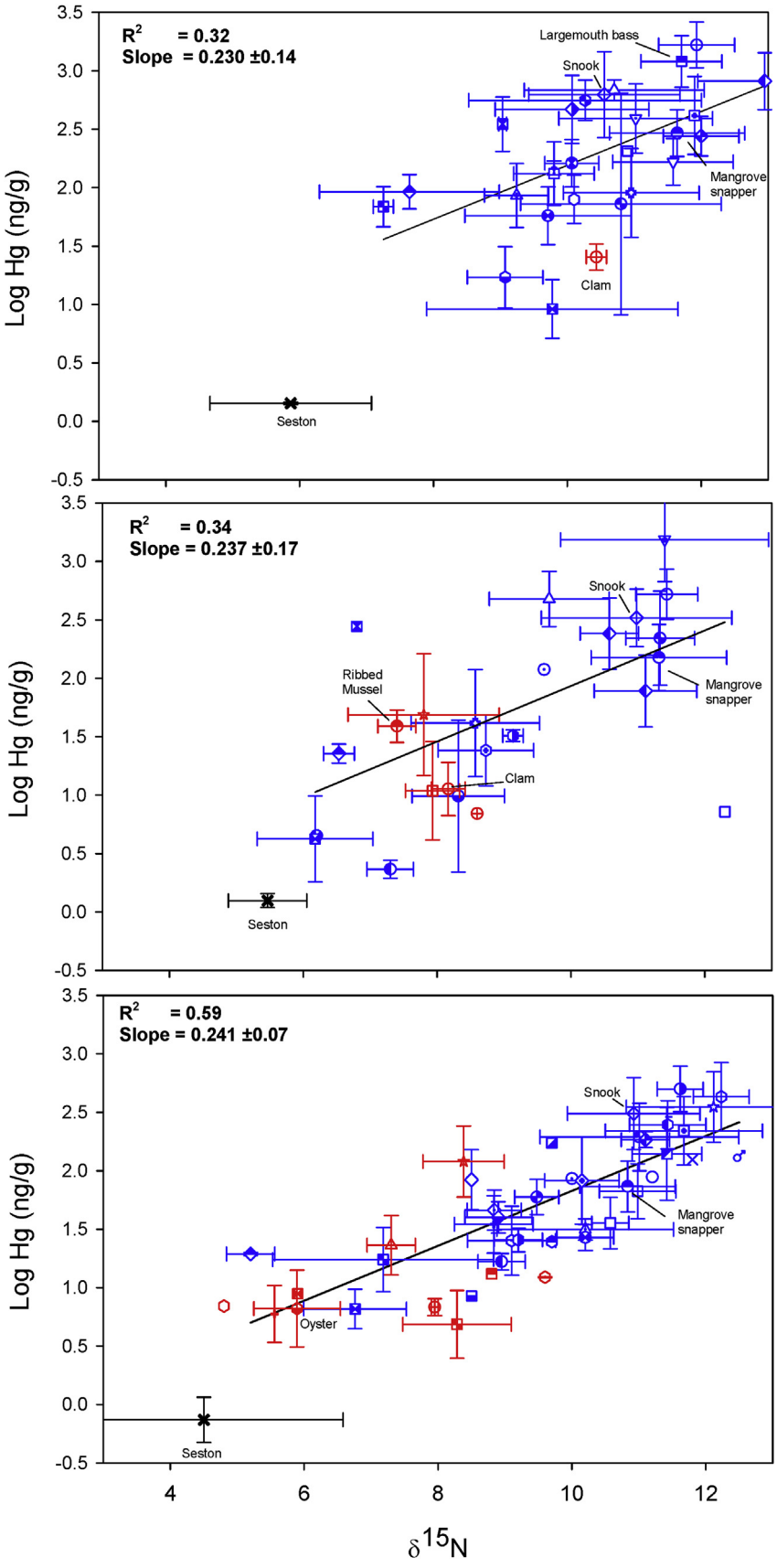

Fig. 4. The relation between Log[THg] and $\delta^{15}N$ (species means ±1SD or range where n < 3; coefficient of determination and TMS ±95%CI based on fish only shown) for upper (top), mid (middle) and lower estuary (bottom) of Shark River Estuary (for summaries for each species by reach and symbol legend, see Supplemental Table S2 – S4).

4. Discussion

4.1. [THg] in top predatory fish

The high [THg] in bass, snook and snapper from the upper reach were consistent with the results of the state's long-term monitoring program of THg in fish at the headwaters of SRE (Gu et al., 2012). The lack of a clear temporal trend (see, Supplemental Information) was also consistent with the results of that monitoring program which has found no significant trend in annual median [Hg] over the past 23 years (Gu et al., 2012). The concentrations in these fish and several others in the upper reach (e.g., bowfin, Florida gar and bluegill) were some of the highest levels recently reported for these species in south Florida (Adams et al., 2003; Evans, 2009; Gu et al., 2012). Levels were much higher than concentrations reported for conspecifics from the upper reaches of other GOM estuaries (Farmer et al., 2010; Fry and Chumchal, 2012).

The inter-specific differences in [THg] among fish species at the upper reach were not surprising given their different life-histories. As previously discussed the bass likely spend much of their time in the marsh but move down to deeper estuarine creeks along with other mobile species when the marsh drys (Boucek and Rehage, 2013). By comparison, while snook in the upper SRE are thought to be almost completely subsidized by freshwater prey that moved down during the dry season (Boucek and Rehage, 2013), they must switch and rely more heavily on estuarine prey during the wet season (either in the upper estuary or by moving down-estuary). Consequently, they are diluting the exposure of Hg bioadvected from the marsh as compared to the species that returned to the freshwater marshes. Considering the lower reach is located down-estuary from this hotspot and, thus, primed to receive biota moving down stream or, as discussed below, receive inputs of high water-column [MeHg], it was expected to have biota with higher [THg]. Yet, the observed trend of decreasing [THg] down-estuary was consistent with previous reports that [THg] in fish communities tend to be lower in the lower reaches of estuaries (Farmer et al., 2010; Fry and Chumchal, 2012).

Although caution must be used when making comparisons to levels reported in the published literature (due to possible differences in size and age), fish from the lower reach had [THg] similar or lower than fish from other Florida estuaries (Adams et al., 2003; Evans, 2009; Evans and Crumley, 2005; Hong et al., 2013) but much higher than conspecifics in other estuaries of the GOM (Farmer et al., 2010; Fry and Chumchal, 2012; Showalter, 2010).

4.2. Basal Hg: suspension feeders, seston and sediment

Basal Hg entering the food web was gauged by [MeHg] measured in the suspension-feeding bivalves as primary consumers. This was further assessed by evaluating [MeHg] measured in seston and sediments. The much higher [MeHg] in the Carolina marsh clams from the upper reach were interpreted has an indication of higher basal Hg entering the food web of the upper reach. The [MeHg] in these clams were among the highest [MeHg] reported in oysters from estuaries around the GOM collected as part of NOAA's Mussel Watch program (i.e., 211 ng MeHg/g dry wt., Apeti et al., 2012). Alternatively, [MeHg] in the oysters from the mouth of SRE were generally similar or lower than concentrations reported in oysters from other areas in southwest Florida, including Charlotte Harbor, Rookery Bay, Faka Union Bay and Florida Bay (Apeti et al., 2012). Still, it is important to note that all of these Florida locations are characterized as hotspots by NOAA (based on comparisons to [MeHg] in oysters from estuaries in the central and western GOM and as compared to the national median concentrations; Apeti et al., 2012).

Seston from the upper reach also contained the highest [MeHg] relative to seston from the other reaches (Table 1). Although comparable data are sparse, the [MeHg] in seston in the present study were markedly lower than concentrations in seston collected along a transect through Guanabara Bay, Brazil and out into the Atlantic (ranging from 22.8 ng/g to 204.9 ng/g; Kehrig et al., 2009). [MeHg] in seston from the lower estuary was only slightly lower than values in zooplankton from nearby Sarasota Bay (3.7 ± 2.2 ng/g wet wt., mean ±1SD, Hong et al., 2013).

Sediment [MeHg] concentrations in the present study were within the range reported from marsh and bay sediments from Florida Bay (Rumbold et al., 2011) and from lower reaches of other estuaries around the GOM (Apeti et al., 2012). The higher [MeHg] in sediments with increased organic content was consistent with previous reports (for review, see Chen et al., 2008). It is noteworthy that organic matter in sediments has also been reported to reduce MeHg bioavailability to benthic fauna (Chen et al., 2008). The mid-estuary maximum in sediment [MeHg] was interesting in light of findings of a study, which was complementary to the present study, that measured [THg] and [MeHg] in the water column along this salinity gradient in 2010 and 2011 (Bergamaschi et al., 2012). That study observed a mid-estuary maximum in water-column concentrations and concluded the mangrove ecotone was a major zone of MeHg production (Bergamaschi et al., 2012), which was also consistent with spatial patterns observed in a previous study in Florida Bay (Rumbold et al., 2011).

The relatively low basal Hg in primary consumers from the lower estuary was surprising given the potential for biotransport from the headwaters and given the previous finding of extraordinarily high water-column [MeHg] (as high as 26.8 ng MeHg/L) in SRE by the complementary study that sampled water (Bergamaschi et al., 2012). However, they also found a very strong relationship between filtered [MeHg] and filtered dissolved organic material (DOM; and specific UV absorbance, SUVA254). That strong statistically relationship may suggest that the DOM was 'complexed' with the MeHg and served as a carrier (i.e., colloidal transport; cf., Stordal et al., 1996). A number of studies have shown that DOM can modify, often reducing, MeHg bioavailability (for review, see Chen et al., 2008; Gorski et al., 2008). Differences in bioavailability of MeHg, as it relates to its carrier, such as DOM, were also invoked to explain differences in bioaccumulation observed in two adjacent watersheds delivering water to northeastern Florida Bay (Rumbold et al., 2011).

4.3. Spatial patterns in δ15N and δ13C

In general, taxon-specific stable isotopes values and their gradients down-estuary (Fig. 3, Supplemental Fig. S2) were in good agreement with results of previous studies done in SRE ((Fry and Smith, 2002; Matich and Heithaus, 2014; Pineda, 2003; Wozniak et al., 2012). Based on samples collected along a transect through SRE, Fry and Smith (2002) reported higher δ15N in mussels collected up estuary as compared to mussels from the lower estuary. Wozniak et al. (2012) investigated nitrogen cycling in ENP, which they characterized as being derived mostly from Lake Okeechobee and the Everglades Agricultural Area, reported higher δ15N values in macrophytes near canals in the ENP than the estuarine ecotone. They speculated the lower δ15N values in the estuary were due to new N from N2-fixation by cyanobacterial periphyton mats (Wozniak et al., 2012).

The down-estuary shift in δ13C observed in the present study was also consistent with the study by Fry and Smith (2002) that found δ13C in mussels from SRE (with values similar to the present study) became less negative from freshwater to marine habitats. Matich et al. (2011) reported a similar δ13C gradient in consumers

down the SRE. Likewise, Pineda (2003) also found macroinvertebrates in upper SRE to have very depleted 13C, particularly if collected within the mangroves. These down-estuary patterns suggest a shift in carbon source from terrestrial vegetation to mangroves and benthic microalgae mid-estuary to a mixture of seagrass and phytoplankton signatures in the lower reaches (Fry and Smith, 2002; Pineda, 2003).

4.4. Trophic magnification slopes

As expected, the higher trophic-level piscivorous fish generally had higher average [THg] and were more enriched in 15N than fish species with an increasing invertebrate diet (e.g., redear sunfish, hogchoker, sheepshead, etc.) and as compared to most macroinvertebrates. The relatively high [THg] and δ15N values in several of the crab species as compared to forage fish is consistent with omnivorous and scavenger diet of crabs (Laughlin, 1982) and was comparable to previous Hg surveys that included crabs (Adams and Engel, 2014; Lewis and Chancy, 2008; Thera and Rumbold, 2014). In general, the [MeHg] and %MeHg (Supplemental Table S5) in macroinvertebrates in the present study were consistent with results of the limited number of previous studies in the GOM that included invertebrates (Cunningham, 2003; Lewis and Chancy, 2008; Farmer et al., 2010; Apeti et al., 2012; Thera and Rumbold, 2014; Adams and Engel, 2014). Lower δ15N values of some of the fish species, particularly mullet, as compared to invertebrates likely demonstrates "telescoping of the food chain" where even large fish can feed directly on first trophic level (Odum, 1970).

The relationship between log [THg] and δ15N, i.e., TMS, was consistent with earlier studies (Fry and Chumchal, 2012; Kidd et al., 1995). As mentioned previously, we had expected that the biological communities to be structured by salinity gradients along the estuarine axis (Bulger et al., 1993; Ley et al., 1999; Green et al., 2006). Further, because many fish species can change their diet in response to prey availability, this was expected to result in variations in food webs. Yeager et al. (2014), for example, analyzed 267 stomachs from mangrove snappers along a salinity gradient in a Florida east-coast estuary and found their diets shifted from a dominance by low quality, intertidal crabs in the upper estuary to an increased reliance on higher quality shrimp, fishes and benthic crabs in the lower estuary. While we observed a shift in δ15N (and δ13C) across the salinity gradient (as discussed above), the TMS, as in indication of biomagnification efficiency, did not statistically differ among the reaches.

The values of the TMS in the present study were slightly steeper than the average TMS reported for tropical marine food webs from a world-wide meta-analysis (i.e., 0.16 ± 0.08; Lavoie et al., 2013) but clearly had overlapping confidence intervals. The slopes from the present study were shallower than a recently reported TMS for a food web from nearby Sarasota Bay (i.e., 0.27, Hong et al., 2013) but were slightly steeper than a TMS reported for a study completed just offshore of southwest Florida (i.e., 0.207, Thera and Rumbold, 2014) and a recent study of tropical coastal food webs in Guanabara Bay and Ilha Grande Bay (ranged as high as 0.22) in Brazil (Bisi et al., 2012).

Early literature reviews also found TMS similar among habitats which led some authors to suggest that spatial variation observed in biomagnified Hg may largely be a result of variability in basal Hg rather than differences in food web structure (Campbell et al., 2005; Chasar et al., 2009; Chumchal et al., 2011; Kidd et al., 2003; Riget et al., 2007). Yet, as pointed out by Ward et al. (2010) and others, this may have been because variation in food webs in those studies were relatively small compared to the differences in basal Hg. This was likely the case in the present study. More recent studies have had the power to discern differences in slopes between habitats (Clayden et al., 2013; Kidd et al., 2012b; Lavoie et al., 2013; Swanson and Kidd, 2010) and attributed those differences to a number of different factors possibly acting individually or in combination.

Acknowledgments

We would like to thank FGCU staff L. Haynes and B. Wasno and all the FGCU students that assisted on this project as paid interns or volunteers, including: H. Benedict, B. Morris, J. Neurohr, M. Nicholson, K. Ribble, J. Schoonover, M. Scroggin, and K. Workinger. Likewise, we thank S. Niemczyk who also helped in the field. Finally, we would also like to thank D. Krabbenhoft (USGS) and J. Ogorek (USGS) who stepped in after the untimely death of D. Evans (NOAA) and ran the MeHg analyses. We would like to thank D. Evans (NOAA) for being a great collaborator. Finally, we thank two anonymous reviewers for their comments that improved this manuscript. Funding for this work was provided by two congressionally-directed grants from the US Department of Education (P116Z090117 and P116Z100149 from 2009 to 2013) and a grant from the Gulf of Mexico Alliance (121128-00 in 2014). However, the contents do not necessarily represent the policy of the US Department of Education or GOMA and do not indicate endorsement by the federal government.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.ecss.2017.10.018.

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