Bioaccumulation of mercury in pelagic fishes from the northern Gulf of Mexico

Yan Cai, Jay R. Rooker, Gary A. Gill, and Jason P. Turner

Abstract: Total mercury (Hg) concentration was determined in the tissues of 10 pelagic fishes in the northern Gulf of Mexico, and dietary tracers (stable isotopes and fatty acids) were used to evaluate the relationship between Hg and feeding history. Highest Hg levels were observed in blue marlin (*Makaira nigricans*), carcharhinid sharks (*Carcharhinus* spp.), and little tunny (*Euthynnus alletteratus*), ranging from 1.08 to 10.52 ppm. Moderate to low concentrations (<1.0 ppm) were observed in blackfin tuna (*Thunnus atlanticus*), cobia (*Rachycentron canadum*), dolphinfish (*Coryphaena hippurus*), greater amberjack (*Seriola dumerili*), king mackerel (*Scomberomorus cavalla*), wahoo (*Acanthocybium solandri*), and yellowfin tuna (*Thunnus albacares*). For the majority of species examined, Hg concentrations did not vary significantly between location (Texas vs. Louisiana) or collection period (2002 and 2003). Significant positive relationships between Hg concentration and body size and (or) weight were detected for 6 of the 10 taxa examined. Hg concentration was also positively associated with trophic position. Three natural associations were identified using stable isotope and fatty acid signatures. Still, no connection between these natural trophic associations and Hg concentration was observed, suggesting that Hg concentration in pelagic fishes was more closely linked to trophic position and size than feeding history.

Résumé : Nous avons déterminé les concentrations de mercure total (Hg) dans les tissus de 10 poissons pélagiques du nord du golfe du Mexique et utilisé des traceurs alimentaires (isotopes stables et acides gras) pour évaluer les relations entre Hg et l'histoire alimentaire. Les concentrations d'Hg les plus fortes s'observent chez le makaire bleu (*Makaira nigricans*), les requins carcharinidés (*Carcharhinus* spp.) et la thonine commune (*Euthynnus alletteratus*) et elles varient de 1,08 à 10,52 ppm. Les concentrations moyennes à faibles (<1,0 ppm) se retrouvent chez le thon à nageoires noires (*Thunnus atlanticus*), le cobia (*Rachycentron canadum*), la coryphène (*Coryphaena hippurus*), la sériole couronnée (*Seriola dumerili*), le tassard royal (*Scomberomorus cavalla*), le tassard bâtard (*Acanthocybium solandri*) et l'albacore à nageoires jaunes (*Thunnus albacares*). Chez la plupart des espèces examinées, les concentrations d'Hg ne varient pas significativement d'un endroit à une autre (Texas et Louisiane) ou d'une année à l'autre (2002 et 2003). Nous trouvons des relations positives significatives entre la concentration d'Hg et la taille et (ou) la masse du corps chez 6 des 10 taxons étudiés. Il y a aussi une corrélation positive entre la concentration d'Hg et le niveau trophique. Les signatures d'isotopes stables et d'acides gras révèlent trois associations naturelles. Néanmoins, nous ne trouvons aucun lien entre ces associations trophiques naturelles et les concentrations d'Hg, ce qui laisse croire que les concentrations d'Hg chez les poissons pélagiques sont plus intimement reliées à la position trophique et à la taille qu'à l'histoire alimentaire.

[Traduit par la Rédaction]

Introduction

Many biotic, ecological, and environmental factors contribute to the uptake of methylmercury (MeHg) in fishes (Wiener et al. 2003), with dietary uptake accounting for more than 90% of the total uptake of MeHg in wild fishes (Hall et al. 1997). When elimination rates of Hg are lower than the uptake rate, concentration of MeHg increases with increasing age or body size of the fish (Wiener and Spry 1996). The uptake of MeHg is also influenced by diet history and trophic position, with rates of Hg accumulation higher for fishes feeding at a higher trophic position (Cabana and Rasmussen 1994; Cabana et al. 1994; Wiener et al. 2003). Moreover, other studies have shown the spatial variation in fish Hg concentrations was attributed to environmental factors like pH, water temperature, lake size, and dissolved organic carbon (DOC) concentrations that control the biogeochemical processes and transformation of MeHg in the ecosystem (Häkanson et al. 1988; Bodaly et al. 1993).

Because most MeHg in fish is obtained through the food chain, information on the feeding ecology of marine consumers is needed to determine the source(s) of MeHg and

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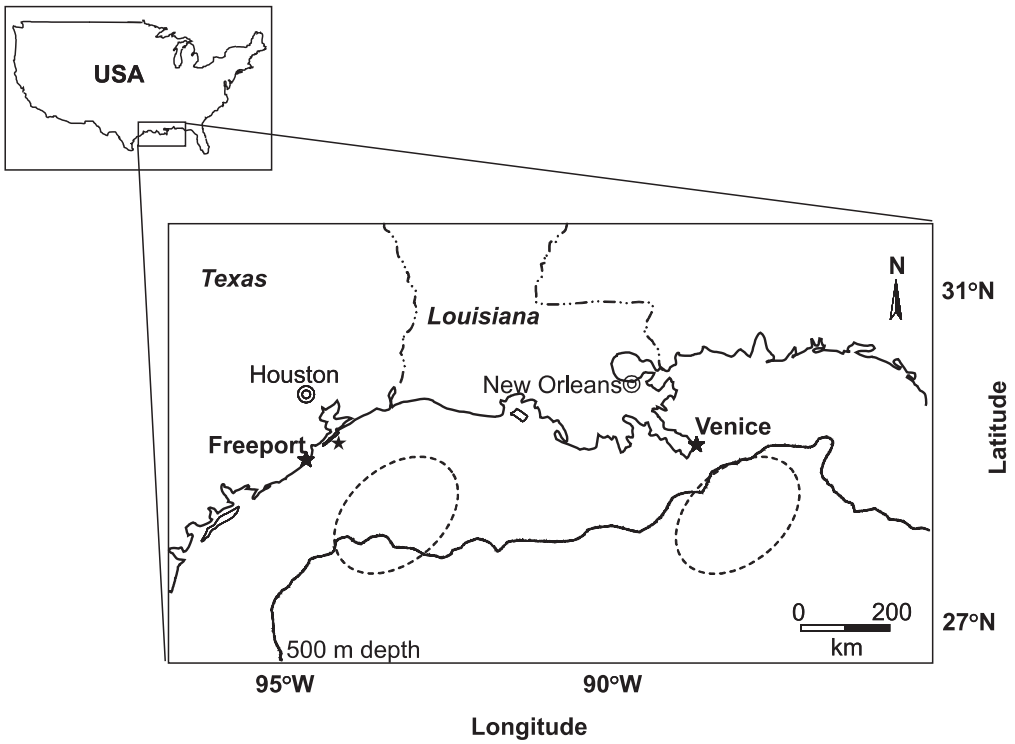
Fig. 1. Map of sampling locations: offshore area near Galveston–Freeport in Texas, USA, and Venice in Louisiana, USA, in the northern Gulf of Mexico.



examine the bioaccumulation of this stressor in marine consumers. Because conventional gut analysis has a number of inherent constraints (e.g., reflects only short-term information on diet; provides limited information on source(s) of organic matter), stable isotope analysis and fatty acid analysis are increasingly used to reconstruct feeding histories and improve our understanding of trophic relationships within aquatic systems (e.g., Kharlamenko et al. 2001; Rooker et al. 2006; Turner and Rooker 2006). Recently, dietary tracer analysis has been coupled with contaminant analysis to determine the pattern and extent of biomagnification of Hg and other contaminants in aquatic food webs (Cook et al. 2004). However, most of these studies have been located in freshwater ecosystems. The purpose of the present study was to examine the trophic structure and contaminant bioaccumulation of Hg in pelagic fishes in the Gulf of Mexico. Efforts were focused on determining body burdens of Hg across several pelagic taxa targeted by recreational and commercial anglers in this region. In addition, stable isotopes and fatty acids were also used to relate feeding history and trophic position to the observed patterns of MeHg in the tissue of top-level predators.

Materials and methods

Sample collection

Pelagic fishes were sampled at docks from two regions of the northern Gulf of Mexico: Galveston–Freeport, Texas, and Venice, Louisiana (Fig. 1). In addition, samples were collected with hook-and-line to complement dock sampling efforts. To assess annual variation, samples were collected during the summer in 2002 and 2003. For each fish sample, ~20 g of muscle tissue were removed from the dorsal region behind the head. Samples were individually bagged and labeled with collection date, species name, and fish length (total length in cm). Samples were stored on ice in a cooler before being transported back to the laboratory and put into a freezer (–20 °C). Ten taxa–species were targeted: blackfin tuna (*Thunnus atlanticus*), blue marlin (*Makaira nigricans*), carcharhinid sharks (*Carcharhinus* spp.), cobia (*Rachycentron canadum*), dolphinfish (*Coryphaena hippurus*), greater amberjack (*Seriola dumerili*), king mackerel (*Scomberomorus cavalla*), little tunny (*Euthynnus alletteratus*), wahoo (*Acanthocybium solandri*), and yellowfin tuna (*Thunnus albacares*).

Determination of total Hg in fish tissues

Measurements of total Hg in fish tissue were conducted using a Milestone DMA-80 direct Hg analyzer (Cizdziel et al. 2002). Fish muscle samples were taken directly from the freezer, cut into 0.1–0.3 g pieces, weighed, and placed directly into sample boats for analysis. The instrument was calibrated each day using standard reference materials (SRM) prepared by the National Research Council of Canada (i.e., TORT-2 and DORM-2). Blanks and a separate standard reference material (DORM-1 or 1566*a*) were always analyzed at the beginning of every batch of 10 samples to assess accuracy. Blanks consisted of an empty boat. Three replicate analyses were conducted for the first three samples of each batch. If the relative percent difference was within 10%, the rest of the batch was analyzed once only. Otherwise, beginning samples of the batch were run again. Fish samples that did not fall within the concentration range of the standards were reanalyzed with a more appropriate amount of wet tissue.

To evaluate the effect of dehydration on samples with different storage times, a second piece of each sample was weighed when the first piece was introduced into the instrument for Hg analysis. After drying in the oven at 50 °C for 48 h, they were weighed again to determine the water concentration of the tissue at the time of analysis and then introduced into the instrument for Hg analysis. We observed no significant dehydration effect (y = mean dry Hg concentration, x = mean wet Hg concentration, $y = 3.80x + 0.04$, $R^2 \approx 1$); therefore, only wet-weight Hg concentrations are reported.

Calibration curves were linear (mean: $R^2 = 1.00$; range: $R^2 = 0.97–1.00$; $n = 68$), and regression equations were used to correct the wet-weight Hg concentration data directly measured by the DMA-80 Hg analyzer. Recovery of the SRM (DORM-2 or DOLT-1) ranged from 90.6% to 112.2% and averaged 97.8% (standard deviation, SD = 5.2%). Mean detection limit, based on three times the SD of blanks, was 0.2 ng Hg·g^{-1} of wet tissue. The coefficient of variation, based on triplicate measurements of the same samples, ranged from 0.3% to 7.1% and averaged 2.9%. Because MeHg comprises an average of more than 95% of the total Hg in fish tissue (Bloom 1998), total Hg was measured to represent the MeHg level in fish in the present study. To simplify the presentation, the term Hg concentration is used from this point on to represent total Hg wet-weight concentration in muscle tissue.

Stable isotope analysis

Tissue samples from randomly selected individuals (~five per species) of nine species (blackfin tuna, blue marlin, cobia, dolphinfish, greater amberjack, king mackerel, little tunny, wahoo, yellowfin tuna) were subject to stable isotope analysis. For each sample, ~0.6 mg of the fish muscle tissue was ground and used for isotopic determination. Isotope ratios ($^{13}C/^{12}C$ and $^{15}N/^{14}N$) and total carbon and nitrogen content were determined using a Finnigan MAT DeltaPlus continuous-flow stable isotope mass spectrometer attached to a Carlo Erba elemental analyzer at the University of Texas at Austin Marine Science Institute. Isotope ratios were reported in parts per thousand (relative to standards (PeeDee Belemnite for carbon and atmospheric N for nitrogen)) and defined in delta (δ) notation as

$$\delta^{15}\text{N or } \delta^{13}\text{C} = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 10^3$$

where $R = {}^{15}N/{}^{14}N$ or $^{13}C/^{12}C$. A secondary standard reference material (chitin of marine origin, Sigma Aldrich Co., No. C-8908) was used to verify the accuracy of isotopic measurements (Herzka and Holt 2000).

Fatty acid analysis

Tissue samples from randomly selected individuals of all 10 taxa were subject to fatty acid analysis. Lipid was extracted in duplicate aliquots using techniques from Folch et al. (1957) as modified by Iverson et al. (2001). The Hilditch procedure (Iverson et al. 1992) was used to prepare fatty acid methyl esters (FAME). Analysis of FAME was run in duplicate using temperature-programmed gas chromatography on a Perkin-Elmer Autosystem II Capillary FID gas chromatograph. Separation of FAME was performed on a 30 m × 0.25 mm internal diameter column coated with 50% cyanopropyl polysilohexane (0.25 μm film thickness; J&W DB-23, Folsom, California, USA). Helium was used as a carrier gas. Individual peaks of FAME were identified by comparing retention times with known composition and commercially available standards (Nu Check Prep., Elysian, Minnesota, USA). A computerized integration system (Turbochrome 4 software, PE Nelson) was used to calculate chromatographic data (Iverson et al. 1997*a*, 1997*b*). Individual fatty acids were converted to mass percentage of total fatty acids using conversion factors from Ackman (1972, 1991). The fatty acid nomenclature used here is of the form 18:2(*n*-3), where 8 designates the total number of carbon atoms, 2 is the number of double bonds, and (*n*-3) is the position of the first double bond from the methyl end of the fatty acid. Average data of the individual fatty acids are expressed as a mass percentage of total fatty acids.

Statistical analyses

To assess regional and interannual variability in Hg concentrations, analysis of covariance (ANCOVA) was performed for each species or taxon, setting location and year as main factors, with log Hg as the dependent variable. The covariate (size) was used to compensate for size-related differences between regions or years. To explore the relationship between Hg concentration and fish body size, linear regression analysis was conducted for each species, setting size as the independent variable and Hg concentration as the dependent variable. Linear regression analysis was also conducted to test for the effect of trophic position (expressed as $\delta^{15}N$) on Hg concentrations in the tissue of pelagic fishes. Hierarchical cluster analysis was used to identify natural associations or groupings with similar dietary histories using fatty acid signatures (based on percent composition). Euclidean distance and the average linkage-joining algorithm were used to produce hierarchical trees for fatty acid data. After observing that fishes from the same location, rather than fishes of the same species, were grouped together, one-way analysis of variance (ANOVA) tests were performed for each taxon to contrast fatty acid profiles of collections from Texas and Louisiana. Before all parametric testing, assumptions of normality (error terms) and homogeneity of variances were examined using Kolmogorov–Smirnov (K–S) test and Levene's test. Distributions of Hg were not always normal between years and locations for each taxon because of the lack of samples for those taxa in a certain year or location. To improve normality and homogeneity of variance, all Hg concentrations were $\log_{10}$-transformed. The α value was set as 0.05 for all statistical tests, and analyses were performed using SPSS software (version 12.0, SPSS Inc., Chicago, Illinois).

Results

Hg survey of pelagic fishes

Mean Hg concentrations across the 10 taxa ranged from 0.07 to 10.52 ppm (n = 387) (Table 1), and concentrations of three taxa were higher than the Food and Drug Administration (FDA) (2001) criterion value of 1.0 ppm wet weight: blue marlin, carcharhinid sharks, and little tunny (10.52, 1.61, and 1.08 ppm, respectively). In addition to these taxa, five others were above a reduced advisory level of 0.3 ppm

Table 1. Hg level of 10 taxa of pelagic fishes from the northern Gulf of Mexico.

		[Hg] (total ‰ wet weight)			Total length (cm)		
Species	*N*	Mean	SD	Range	Mean	SD	Range
AJ*	44	0.60	±0.23	0.24–1.07	84	±10	69–112
BM**	9	10.52	±5.03	4.95–18.72	285	±23	256–311
BX*	48	0.64	±0.31	0.00–1.41	73	±6	22–87
CO*	17	0.89	±0.52	0.20–2.40	97	±16	76–142
CS**	9	1.61	±0.45	0.46–4.08	69	±6	15–96
DF	57	0.07	±0.09	0.01–0.49	79	±30	38–135
KM*	39	0.96	±0.27	0.37–1.46	84	±9	64–104
LT**	9	1.08	±0.72	0.24–2.52	56	±4	52–66
WA*	52	0.78	±0.87	0.01–3.31	133	±16	103–175
YT	103	0.18	±0.15	0.07–0.87	112	±17	54–159

Note: Fish abbreviations are as follows: AJ, greater amberjack (*Seriola dumerili*); BM, blue marlin (*Makaira nigricans*); BX, blackfin tuna (*Thunnus atlanticus*); CO, cobia (*Rachycentron canadum*); CS, carcharhinid sharks; DF, dolphinfish (*Coryphaena hippurus*); KM, king mackerel (*Scomberomorus cavalla*); LT, little tunny (*Euthynnus alletteratus*); WA, wahoo (*Acanthocybium solandri*); YT, yellowfin tuna (*Thunnus albacares*). *N*, number of individuals; SD, standard deviation.

*Above US Environmental Protection Agency 2002 recommended criteria level (0.3 $\mu g \cdot g^{-1}$ wet weight).

**Above FDA 2001 recommended criteria level (1.0 $\mu g \cdot g^{-1}$ wet weight).

wet weight set by the US Environmental Protection Agency (EPA) (US Environmental Protection Agency 2002): king mackerel, mean = 0.96 ppm; cobia, mean = 0.89 ppm; wahoo, mean = 0.78 ppm; blackfin tuna, mean = 0.64 ppm; greater amberjack, mean = 0.60 ppm. Mean Hg concentrations in yellowfin tuna and dolphinfish were below all consumption advisory levels (0.18 ppm and 0.07 ppm, respectively).

Interannual and regional variation in Hg concentration was investigated, and the majority of taxa examined showed no effect for either factor. Hg levels of only one species, yellowfin tuna, varied significantly between years (P = 0.041), with lower levels in 2003 (0.10 ppm wet weight) compared with 2002 (0.25 ppm wet weight); no interannual effect was observed for blackfin tuna (P = 0.267), dolphinfish (P = 0.574), wahoo (P = 0.296), king mackerel (P = 0.506), or cobia (P = 0.230). Differences in Hg concentration between Texas and Louisiana were not statistically significant for blackfin tuna (P = 0.111), dolphinfish (P = 0.457), and wahoo (P = 0.234); however, Hg concentrations in yellowfin tuna were significantly higher (P = 0.009) in Texas (0.20 ppm) than in Louisiana (0.18 ppm). Sample size limitations did not warrant interannual and (or) regional comparisons for blue marlin, greater amberjack, little tunny, or carcharhinid sharks.

Relationships between length and Hg concentration, as well as between body weight and Hg concentration, were examined for six taxa, and all regressions were significant with positive slopes, indicating that Hg concentration increases with increasing length or weight (Table 2). Blackfin tuna, carcharhinid sharks, and yellowfin tuna showed highest coefficients of determination: blackfin tuna (R^2 = 0.73, n = 48, $P < 0.001$), carcharhinid sharks (R^2 = 0.69, n = 9, $P < 0.001$), yellowfin tuna (R^2 = 0.64, n = 103, $P < 0.001$). Linear regression analysis of Hg concentration by weight indicated that relationships were significant for the same six taxa. Highest R^2 values were observed in blackfin tuna (R^2 = 0.79, n = 48, $P < 0.001$), dolphinfish (R^2 = 0.56, n = 57, $P < 0.001$), and yellowfin tuna (R^2 = 0.55, n = 103, $P < 0.001$). For both length and weight regressions, slope values were significantly higher for blackfin tuna and wahoo than for other taxa examined (Fig 2).

Relationship between Hg tissue concentration and stable isotopes

Stable nitrogen and carbon isotopes ($\delta^{15}N$ and $\delta^{13}C$) were quantified for all species except carcharhinid sharks. $\delta^{15}N$ values ranged from 6.7‰ to 16.2‰, and the highest $\delta^{15}N$ values were observed for little tunny ($\delta^{15}N$ = 13.9‰–16.2‰) and king mackerel ($\delta^{15}N$ = 12.9‰–15.5‰) (Fig. 3). Although blue marlin had the highest Hg level (mean = 10.52 ppm), predicted trophic position of this consumer was intermediate ($\delta^{15}N$ = 10.0‰–11.2‰). The lowest $\delta^{15}N$ values were observed for dolphinfish (6.7‰–9.3‰). Excluding blue marlin, there was a significant positive relationship (R^2 = 0.63, P = 0.011) between Hg level and $\delta^{15}N$ (Hg = 0.004exp(0.38 ($\delta^{15}N$)) (Fig. 3). $\delta^{13}C$ values varied from –17.43‰ to –15.00‰ across species, and signatures of amberjack, yellowfin tuna, and blackfin tuna (range: –17.43‰ to –17.08‰) were depleted relative to other taxa (Fig. 4): cobia (–15.00‰), dolphinfish (–16.00‰), blue marlin (–16.12‰), little tunny (–16.41‰), and king mackerel (–16.59‰). No significant relationship was detected between total Hg and $\delta^{13}C$.

Relationship between Hg tissue concentration and fatty acids

The fatty acid composition of consumer pelagic fish tissue was analyzed, and ~70 fatty acids and their isomers were routinely identified. Only those found at levels ≥0.5% of the total fatty acids were presented here to simplify the presentation (Table 3). Fatty acid signatures of pelagic fishes were characterized by high levels of polyunsaturated fatty acids (PUFAs), which constituted between 41.99% (little tunny) and 57.87% (dolphinfish) of the total fatty acid content. Five major PUFAs (22:6(n-3) (docosahexaenoic acid), 22:5(n-3) (docosapentaenoic acid), 20:5(n-3) (eicosapentaenoic acid, EPA), 20:4(n-6) (arachidonic acid), and 18:2(n-6) (linoleic

Table 2. Regression analysis of Hg tissue concentrations as a function of size and body weight for apex predator species in northern Gulf of Mexico.

	[Hg] and size				[Hg] and weight			
Species	*a*	*b*	R^2	*P*	*a*	*b*	R^2	*P*
BX	–5.043	4.193	0.79	<0.001	–2.579	1.361	0.79	<0.001
CS	–1.478	2.463	0.35	<0.001	0.172	0.838	0.35	<0.001
DF	–1.821	1.857	0.56	<0.001	–0.497	0.720	0.56	<0.001
KM	–0.863	1.991	0.49	<0.001	0.389	0.688	0.49	<0.001
WA	–10.358	6.097	0.35	<0.001	–5.389	1.911	0.35	<0.001
YT*	–2.548	2.354	0.59	<0.001	–1.536	0.841	0.59	<0.001

Note: Significant linear functions ($Y = a + bX$) were fitted. Y (= $\log_{10}$[Hg]) is log-transformed Hg level (ng^{-1}); X is $\log_{10}$ total length (cm) or $\log_{10}$ total body weight (g). The estimated intercept (*a*), slope (*b*), R^2 value, and *P* value are listed by taxa–species: BX, blackfin tuna (*Thunnus atlanticus*); CS, carcharhinid sharks; DF, dolphinfish (*Coryphaena hippurus*); KM, king mackerel (*Scomberomorus cavalla*); WA, wahoo (*Acanthocybium solandri*); YT, yellowfin tuna (*Thunnus albacares*).

*Equation based on samples from Louisiana 2002 only.

Fig. 2. Comparison of regression of Hg tissue concentrations (wet weight) as a function of body weight for six pelagic taxa. BX, blackfin tuna (*Thunnus atlanticus*), $y = 1.36x - 2.6$, $R^2 = 0.79$; CS, carcharhinid sharks (*Carcharhinus* spp.), $y = 0.84x + 0.17$, $R^2 = 0.35$; DF, dolphinfish (*Coryphaena hippurus*), $y = 0.72x - 0.5$, $R^2 = 0.56$; KM, king mackerel (*Scomberomorus cavalla*), $y = 0.69x + 0.39$, $R^2 = 0.49$; WA, wahoo (*Acanthocybium solandri*), $y = 1.91x - 5.4$, $R^2 = 0.35$; YT, yellowfin tuna (*Thunnus albacares*), $y = 0.79x - 1.23$, $R^2 = 0.59$.

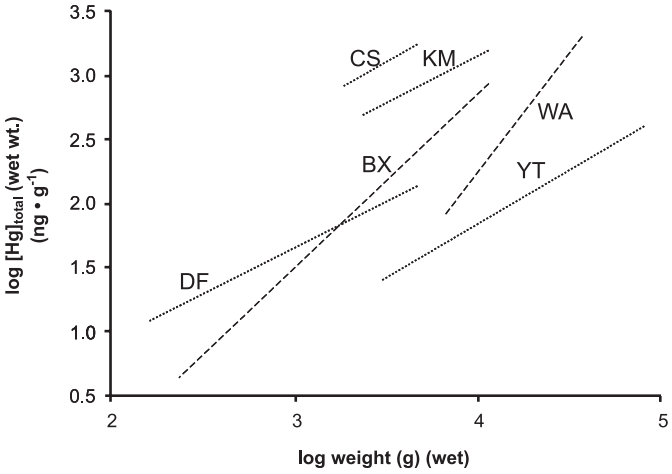
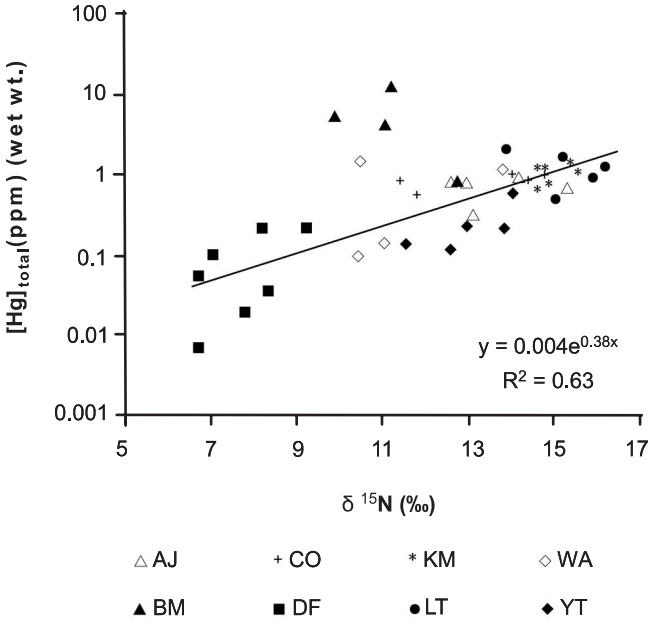


Fig. 3. Linear regression of Hg tissue concentrations (wet weight) as a function of trophic position for pelagic fishes collected in the northern Gulf of Mexico. An exponential equation was fitted to data from eight species. AJ, greater amberjack (*Seriola dumerili*); CO, cobia (*Rachycentron canadum*); DF, dolphinfish (*Coryphaena hippurus*); KM, king mackerel (*Scomberomorus cavalla*); LT, little tunny (*Euthynnus alletteratus*); WA, wahoo (*Acanthocybium solandri*); YT, yellowfin tuna (*Thunnus albacares*). BM blue marlin (*Makaira nigricans*) was not used in the regression.



acid)) were present, and the relative quantities of each PUFA were relatively similar among the taxa examined (Fig. 5). 22:6(*n*-3) was the dominant PUFA, comprising more than 50% of the PUFA content of each taxon. Saturated fatty acids (SAT) were the second largest fatty acid group present in the muscle tissue of pelagic fishes, comprising an average of 32.55% of the total fatty acid content.

Hierarchical cluster analysis based on fatty acid profiles of 10 pelagic taxa showed that most species were grouped together by location first, then by species (Fig. 6). Year and size effect were not observed to have an impact on the natural associations, and measures of dissimilarity were highest between taxa from Texas and Louisiana (Fig. 6*a*). A classification tree based on fatty acid components was built for pelagic fishes from each region (Figs. 6*b*, 6*c*), and natural breaks in the fatty-acid-based trees were linked to natural breaks in stable carbon ($\delta^{13}C$) isotope values of these consumers. In Texas, three groups were detected basing on a rescaled squared Euclidean distance, and these groups could also be distinguished by their $\delta^{13}C$ value: cobia (–15‰ < $\delta^{13}C$); blue marlin, king mackerel, and dolphinfish (–17‰ < $\delta^{13}C$ < –15‰); and greater amberjack, yellowfin tuna, and blackfin tuna ($\delta^{13}C$ < –17‰). Similarly, the two groups based on fatty acid signatures in Louisiana could also be separated to a high degree by their $\delta^{13}C$ value: little tunny and wahoo (–17‰ < $\delta^{13}C$ < –15‰); yellowfin tuna and

Table 3. Fatty acid composition (percent weight of total fatty acids) of pelagic fish from the northern Gulf of Mexico.

Fatty acid	AJ (n = 18)	BM (n = 4)	BX (n = 12)	CO (n = 6)	CS (n = 4)	DF (n = 17)	KM (n = 11)	LT (n = 3)	WA (n = 1)	YT (n = 7)
14:0	1.10	1.58	2.58	1.15	0.44	0.36	0.43	3.03	4.27	1.23
15:0	0.39	0.48	0.48	0.33	0.20	0.36	0.24	0.67	0.67	0.43
ISO16	0.70	1.88	1.16	3.54	3.22	4.62	0.85	0.34	0.19	1.59
16:0	21.14	18.30	19.31	15.25	18.89	15.76	19.74	22.75	21.45	18.36
16:1(n-9)	0.56	0.37	0.29	0.42	0.22	0.45	0.34	0.33	0.67	0.27
16:1(n-7)	2.22	2.26	4.55	2.14	1.54	0.89	1.09	5.07	6.76	2.30
16:2 (n-4)	0.96	1.03	0.87	0.53	0.18	1.25	0.67	0.72	0.63	0.96
16:4(n-1)	1.09	1.99	1.19	1.62	1.70	1.12	1.59	1.33	1.73	1.10
17:0	0.68	0.59	0.75	0.95	0.38	0.88	0.73	1.12	1.38	0.98
17:1	0.32	0.28	0.34	0.49	0.26	0.29	0.30	0.25	0.12	0.28
18:0	7.93	9.27	7.72	10.45	10.77	9.53	9.45	8.57	5.72	8.16
18:1(n-9)	8.41	9.51	10.11	10.96	4.99	7.74	7.68	7.44	10.13	8.31
18:1(n-7)	2.04	1.80	2.51	2.13	9.03	1.37	2.00	7.82	2.90	1.83
18:2(n-6)	0.53	0.87	0.97	0.83	0.51	0.43	0.58	0.88	0.96	0.89
20:1(n-7)	0.52	0.50	0.83	0.38	1.04	0.18	0.47	0.87	0.92	0.49
20:4(n-6)	3.49	4.70	2.71	9.49	5.16	7.09	5.50	2.19	2.41	3.44
20:5(n-3)	3.25	2.99	6.35	4.24	1.55	3.49	4.43	4.50	7.40	5.02
22:5(n-6)	2.92	3.51	2.00	3.33	7.23	3.76	3.69	1.93	1.55	2.95
22:5(n-3)	2.99	2.71	2.63	3.43	4.56	2.18	2.12	2.29	3.31	2.05
22:6(n-3)	33.57	29.36	23.63	22.70	22.80	34.26	32.27	20.02	20.38	33.45
SAT	31.94	32.10	32.00	31.67	33.89	31.50	31.44	36.48	33.68	30.76
MONO	13.76	14.43	18.28	16.02	16.82	10.62	11.58	21.53	21.38	13.19
PUFA	54.31	53.47	49.71	52.31	49.29	57.87	56.98	41.99	44.94	56.05

Note: AJ, greater amberjack (*Seriola dumerili*); BM, blue marlin (*Makaira nigricans*); BX, blackfin tuna (*Thunnus atlanticus*); CO, cobia (*Rachycentron canadum*); CS, carcharhinid sharks; DF, dolphinfish (*Coryphaena hippurus*); KM, king mackerel (*Scomberomorus cavalla*); LT, little tunny (*Euthynnus alletteratus*); WA, wahoo (*Acanthocybium solandri*); YT, yellowfin tuna (*Thunnus albacares*). SAT, saturated fatty acid; MONO, monounsaturated fatty acid; PUFA, polyunsaturated fatty acid.

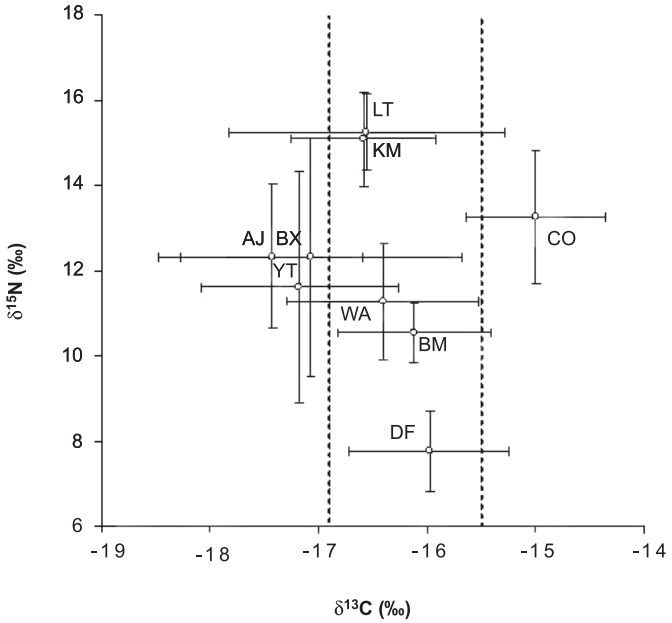
blackfin tuna ($\delta^{13}C < -17‰$). Greater amberjack in Louisiana was an exception from the groupings.

Discussion

Hg tissue concentrations differed between species, and three general categories of Hg bioaccumulation were identified: high range (>1.0 ppm), middle range (between 0.3 and 1.0 ppm), and low range (<0.3 ppm). High range taxa included blue marlin, carcharhinid sharks, and little tunny; middle range included blackfin tuna, cobia, greater amberjack, king mackerel, and wahoo; low range included dolphinfish and yellowfin tuna. Hg tissue concentrations observed in this study for certain taxa were similar to results from other studies conducted in the Gulf of Mexico (Adams et al. 2003; FDA 2004; US Environmental Protection Agency 2004*a*). For example, blue marlin and little tunny were two of the three species observed in the high range category, and these two species had the highest Hg concentrations of pelagic fishes collected from a recent Hg survey completed in Florida (Adams et al. 2003). Moreover, carcharhinid sharks, the second highest Hg concentration taxa in this study, were also the second highest Hg fish on the EPA not-to-eat fish list (US Environmental Protection Agency 2004*b*). Even though taxa in the highest Hg category were similar to other studies, both blue marlin and carcharhinid sharks showed considerably higher Hg concentrations (up to two–three times higher) than previously reported (Adams et al. 2003; FDA 2004). Similarly, except for blackfin tuna, observed Hg levels of all taxa in the middle range (king mackerel, wahoo, greater amberjack, cobia) were 0.3–0.5 ppm higher than previous reports (Adams et al. 2003; Watanabe et al. 2003). Observed patterns suggest that regional variation in fish tissue Hg occurs within the Gulf of Mexico; however, differences in sample size and fish length–weight preclude further interpretation of these data.

MeHg accumulates from one trophic level to the next, and Hg concentrations in large, apex predators are often above advisory levels set by FDA and EPA (Andersen and Depledge 1997). Compared with most estuarine species, pelagic fishes attain larger body sizes and feed at higher trophic levels (Rooker et al. 2006), which likely increases the bioaccumulation process and may result in higher Hg concentrations. Sager (2004) measured the total Hg tissue concentrations of three estuarine fishes (southern flounder, *Paralichthys lethostigma*; spotted seatrout, *Cynoscion nebulosus*; and red drum, *Sciaenops ocellatus*) from the same area (NW Gulf of Mexico), and all three species showed lower Hg concentrations (range: 0.06–0.10 ppm) than the EPA 0.3 ppm threshold. Similarly, a study in Galveston Bay, Texas, examined the same species and indicated that Hg concentrations of these species were normally below 0.25 ppm, with an average near 0.10 ppm (GBNEP 1992). In contrast, 8 out of 10 pelagic taxa examined in the present study had higher Hg concentrations than the EPA 0.3 ppm threshold, with three species above the FDA 1.0 ppm level. Considering that all the pelagic fishes in this study were apex predators with either long life spans (>10 years) and

Fig. 4. Stable-carbon and stable-nitrogen isotope values (mean ± standard deviation) of nine pelagic fishes from northern Gulf of Mexico. AJ (n = 5), greater amberjack (*Seriola dumerili*); BX (n = 5), blackfin tuna (*Thunnus atlanticus*); BM (n = 4), blue marlin (*Makaira nigricans*); CO (n = 5), cobia (*Rachycentron canadum*); DF (n = 7), dolphinfish (*Coryphaena hippurus*); KM (n = 6), king mackerel (*Scomberomorus cavalla*); LT (n = 5), little tunny (*Euthynnus alletteratus*); WA (n = 5), wahoo (*Acanthocybium solandri*); YT (n = 5), yellowfin tuna (*Thunnus albacares*). Broken lines indicated groupings following fatty acid classification tree results.



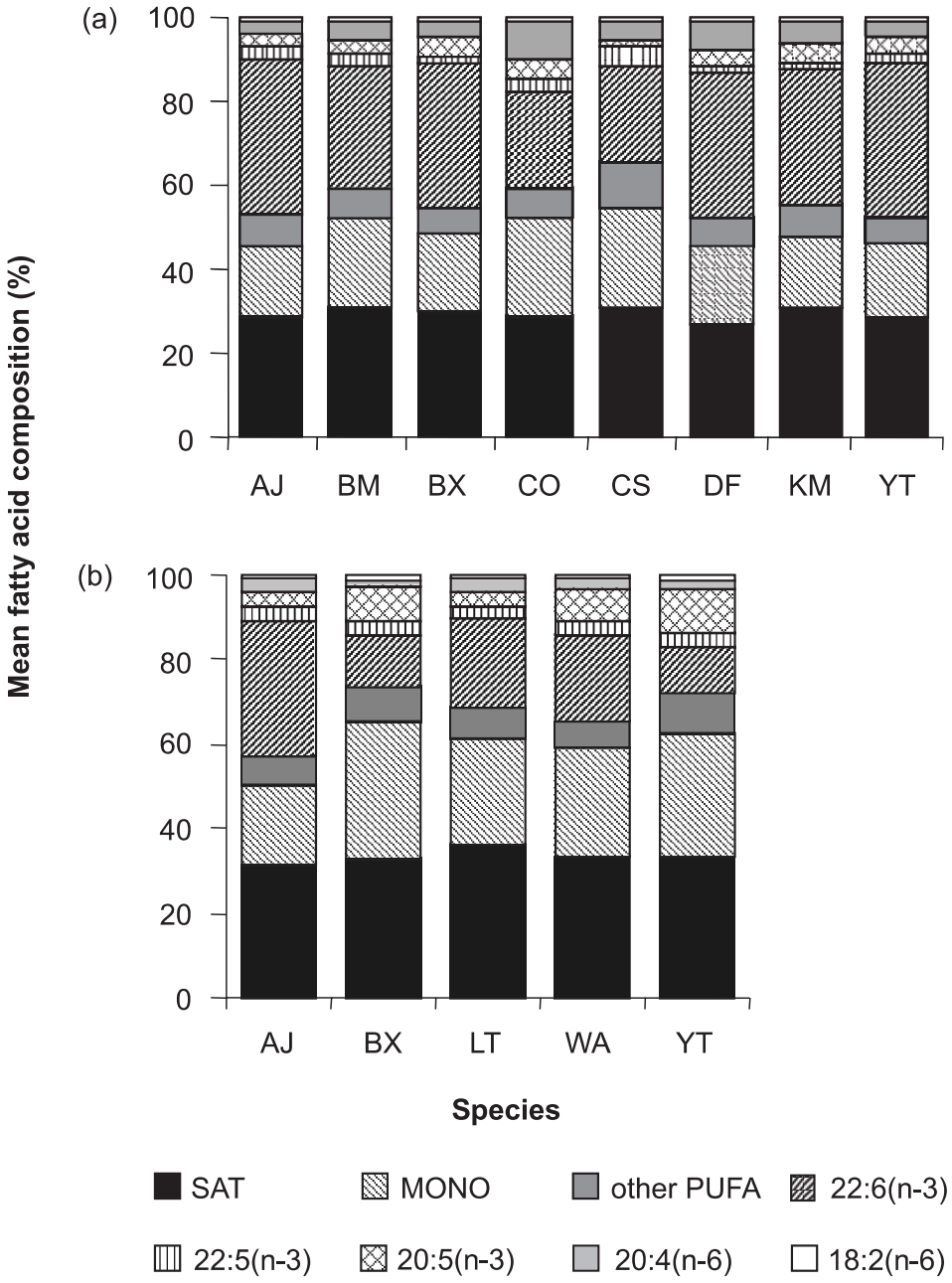
(or) large body size (>100 cm) or both (e.g., Collette and Nauen 1983; Bauchot 1987), elevated Hg concentrations detected in these fish were not surprising. Moreover, the species with the highest Hg concentration in this study, blue marlin, was the largest species examined (mean = 285 cm lower jaw fork length), and this species is very long-lived (>20 years) (Wilson et al. 1991). In contrast, several of the other species examined (e.g., dolphinfish, cobia, greater amberjack, yellowfin tuna) typically live 4–10 years (Manooch and Potts 1997; Franks et al. 1999; Massuti et al. 1999), and thus even with a relatively large body size, they may not live long enough to accumulate large amounts of Hg. The life span effect seems to apply particularly well to dolphinfish, since this species, which has the shortest lifespan of any of the taxa examined (3–5 years), also had the lowest Hg concentration in its tissue.

No regional or annual differences in Hg concentration were detected for the majority of taxa examined. The major reason for the lack of regional differences could be that many of the pelagic fishes surveyed here (e.g., yellowfin tuna, little tunny, blue marlin) are known to be highly migratory species and may travel hundreds to thousands of kilometres in short periods of time (e.g., Deriso et al. 1991; FAO Fisheries Department 1994; Kerstetter et al. 2003). As a result, individuals caught in Texas or Louisiana waters may have spent a portion of their life in the other region or other parts of the Gulf of Mexico. Electronic tagging studies are currently underway in the northern Gulf of Mexico, and migration between the two regions has been documented for billfishes (J. Rooker, unpublished data). With respect to temporal differences, the 2-year period was a relatively short time interval, and environmental changes or tissue turnover during this period may have been insufficient to produce notable changes in Hg tissue concentrations.

Significant positive relationships between Hg concentration and size (length and (or) weight) were detected for all species examined, and similar relationships have been documented previously (e.g., Monteiro and Lopes 1990; Wiener and Spry 1996; Sager 2002). Moreover, recent work by Adams (2004) on pelagic species (yellowfin tuna and little tunny) showed positive relationships between Hg concentration and fish length and (or) weight. In the present study, slope values from linear regressions of Hg concentration and size were similar (0.010–0.011) for several species (carcharhinid sharks, yellowfin tuna, dolphinfish, and king mackerel), suggesting that the rate of Hg accumulation may be closely tied to growth. To verify this theory, Hg was plotted against body weight. All six species not only showed significant positive relationships between their Hg concentration and body weight, but the same four species showed similar Hg accumulation rates (slope values). The positive relationship between Hg concentration and size is likely a function of a slow rate of elimination of MeHg relative to its rapid rate of uptake (Trudel and Rasmussen 1997). Given the fact that MeHg forms covalent bonds with proteins in muscle after it is transported through the blood (Carty and Malone 1979; Georgieva et al. 2004), and also that in carnivorous fish, protein assimilation is generally equal to 80% of total assimilation from food consumption (Brett and Groves 1979), the positive relationship between Hg concentration and body mass observed here is in accord with expected patterns of accumulation.

A positive exponential relationship between Hg concentration and trophic position (expressed as $\delta^{15}N$) was observed here, and a positive relationship between these two parameters has been observed in other studies (Lyle 1984; Cabana et al. 1994). Besides Hg, concentrations of other contaminants in the tissue of fishes also increase with increasing trophic position. For example, Hobson et al. (2002) reported a linear relationship between PCB concentration and $\delta^{15}N$ values for organisms in the North Water food web. The positive relationships between Hg level (and other contaminants) and trophic position in fishes suggests that biomagnification of Hg takes place in marine ecosystems. This process occurs because consumers feeding at higher trophic positions consume larger prey with higher body burdens than smaller prey (Muir et al. 1988; Watras et al. 1998; Bowles et al. 2001). Although blue marlin did not occupy the highest trophic level, their Hg levels were well above the Hg–$\delta^{15}N$ regression line. This could be explained in part by the fact that blue marlin are relatively long-lived species (>20 years) (Wilson et al. 1991), which would allow them to accumulate more Hg relative to other shorter-lived taxa. Overlap of $\delta^{15}N$ values was observed for many pelagic fish species in this study because of the relatively large intraspecific variation of $\delta^{15}N$ values. The broad range of observed $\delta^{15}N$ values for

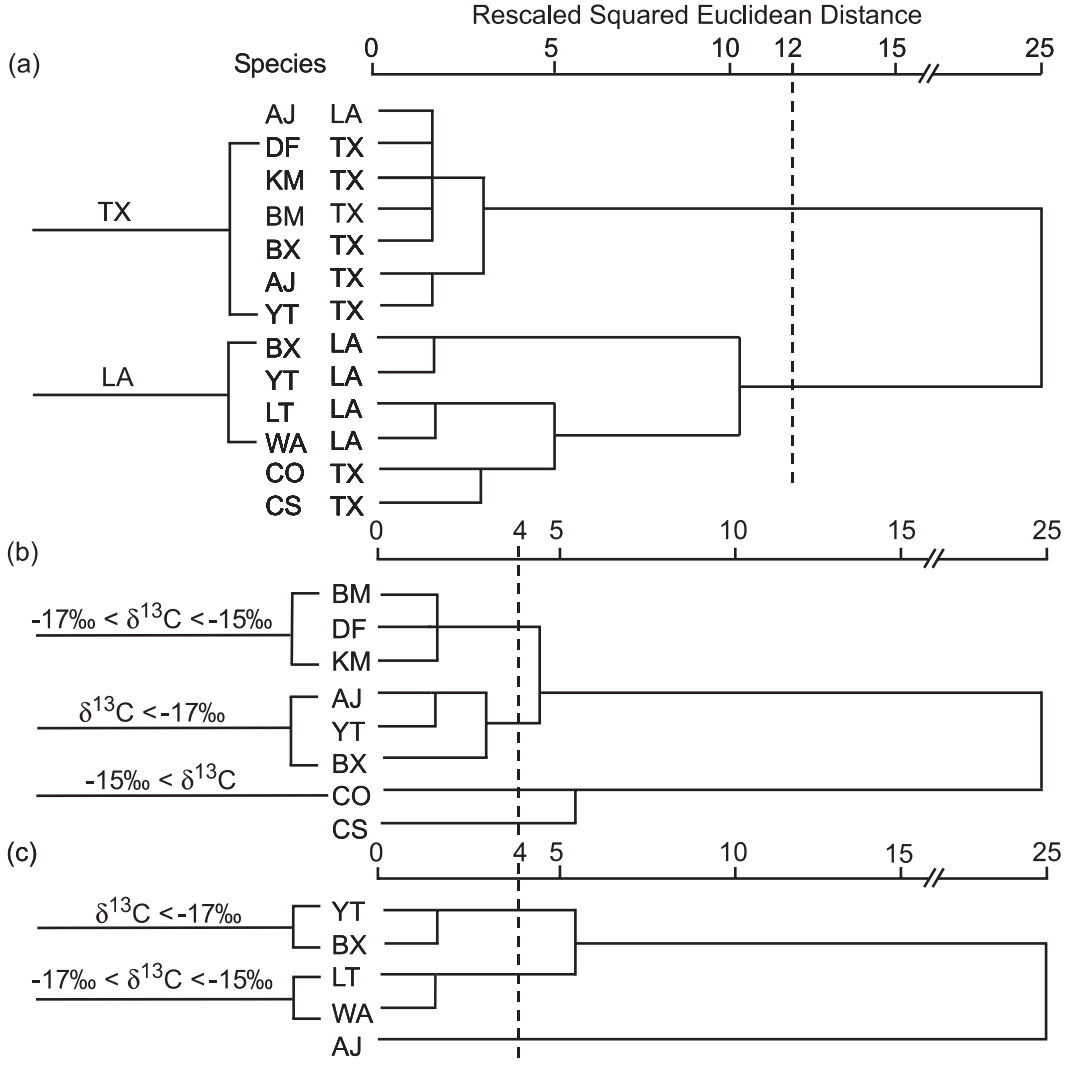
Fig. 5. Mean percent fatty acid composition of 10 pelagic fishes from (*a*) Texas and (*b*) Louisiana. AJ, greater amberjack (*Seriola dumerili*); BM, blue marlin (*Makaira nigricans*); BX, blackfin tuna (*Thunnus atlanticus*); CO, cobia (*Rachycentron canadum*); CS, carcharhinid sharks (*Carcharhinus* spp.); DF, dolphinfish (*Coryphaena hippurus*); KM, king mackerel (*Scomberomorus cavalla*); LT, little tunny (*Euthynnus alletteratus*); WA, wahoo (*Acanthocybium solandri*); YT, yellowfin tuna (*Thunnus albacares*). SAT, saturated fatty acid; MONO, monounsaturated fatty acid; PUFA, polyunsaturated fatty acid.



conspecifics may be related to individuals feeding on prey from different trophic levels (i.e., generalist feeding strategy) or ontogenetic shifts in diet (Kidd et al. 1995; Herzka and Holt 2000). Moreover, $\delta^{15}N$ values vary as a function of the isotopic composition of the inorganic nitrogen at the base of the food web. Thus, precaution must be exercised here because signatures of consumers will not be linked directly to trophic position when multiple producers (pathways) are present.

Three natural groupings of pelagic fishes were identified using either $\delta^{13}C$ values or fatty acid profiles, suggestive of common carbon sources or prey resources among members of each association. Rooker et al. (2006) examined the trophic ecology of pelagic fishes from the same region using stable isotopes and fatty acids. Findings from this study indicated that the largest fraction of organic matter used by pelagic consumers was derived from phytoplankton production; there did not appear to be multiple trophic pathways within this pelagic ecosystem. Stable isotope and fatty acid signatures of the majority of consumers in the present study also suggested a phytoplankton-based food web, and observed groupings may be linked to regional differences in phyto-

Fig. 6. Results of hierarchical cluster analysis on fatty acid profiles of pelagic fishes from Texas (TX) and Louisiana (LA). AJ, greater amberjack (*Seriola dumerili*); BX, blackfin tuna (*Thunnus atlanticus*); BM, blue marlin (*Makaira nigricans*); CO, cobia (*Rachycentron canadum*); CS, carcharhinid sharks (*Carcharhinus* spp.); DF, dolphinfish (*Coryphaena hippurus*); KM, king mackerel (*Scomberomorus cavalla*); LT, little tunny (*Euthynnus alletteratus*); WA, wahoo (*Acanthocybium solandri*); YT, yellowfin tuna (*Thunnus albacares*). (*a*) Fishes from the same location were grouped together according to a squared Euclidean distance value of 12. (*b*) Fishes from Texas were clustered into three groups according to a squared Euclidean distance value of 4. (*c*) Fishes from Louisiana were clustered into two groups according to a squared Euclidean distance value of 4. Stable isotope categories used in (*b*) and (*c*) are mean values; the range of observed values for each taxon may be outside the category containing the mean value.



plankton signatures or other factors linked to foraging (i.e., shifts in trophic position). Regardless of the actual cause of these grouping, there did not appear to be a link to Hg bioaccumulation. For example, blue marlin and little tunny were in the high Hg group and dolphinfish was in the low Hg group. Nevertheless, all three species had similar $\delta^{13}C$ values, which may indicate the source of organic matter for these species was the same. The lack of connection between these natural groupings and the Hg pattern observed in these fishes indicated that the distinguishing factors for the groupings were not Hg accumulation in these fishes. Since the same natural groupings were obtained for both dietary markers, it not only confirmed the existence of these natural groupings but also indicated that the distinguishing factors were somewhat connected with the dietary history of these fishes. One possible explanation for the observed patterns could be linked to pelagic or benthic foraging strategies of the taxa examined. In a marine ecosystem, $\delta^{13}C$ values of benthic and pelagic prey differ; thus, consumers feeding on or near the bottom can be distinguished from consumers feeding in the water column (Hatase et al. 2002; Hobson et al. 2002). Moreover, $\delta^{13}C$ values of benthic prey of large consumers in the Gulf of Mexico are more enriched compared with their pelagic counterparts (J. Rooker, unpublished data). In the present study, cobia had the most enriched $\delta^{13}C$ value and thus appeared to feed on more benthic prey than all the other pelagic fishes in this study. Moreover, levels of one PUFA, 20:4(*n*-6), were noticeably higher in cobia than in other species examined, which may also be linked to benthic food source(s).

In this study, the classification tree based on the fatty acid profiles of pelagic fishes separated fishes from different regions (Texas vs. Louisiana). Because the fatty acid composition of the diet influences the lipid composition of tissues in fishes (Turner and Rooker 2005*a*, 2005*b*; Rooker et al. 2006), it appears that pelagic taxa within the same region share similar prey resources or a common source of organic matter in their food web. Since many of the pelagic taxa examined here are opportunistic feeders (e.g., Manooch et al. 1984; Meyer and Franks 1996; Abitia-Cardenas et al. 1999), their diets may be composed of a few dominant prey species, which vary from one region to another. Also, the primary source of organic matter supplied to primary, secondary, and tertiary consumers may vary spatially (Iverson et al. 1997*b*; Cook et al. 2000), leading to regional differences in fatty acid signatures. Although regional variability in fatty acid signatures was present, certain taxa were aligned with members of the other region. Although carcharhinid sharks and cobia from Texas were present in the Louisiana grouping, it is difficult to interpret this result, since samples of both taxa were only available from Texas. Moreover, most taxa in our assessment are highly migratory, and therefore they may utilize feeding areas in both regions. Species-specific variation in Hg levels between the two regions was negligible. The lack of agreement between fatty acid and Hg measurements (e.g., fatty acid group contained both high and low Hg taxa) appears to indicate that Hg accumulation is not tied directly to prey selection patterns. If this is true, other factors such as trophic position, age, and Hg concentrations of the entire prey community are likely the primary determinants of Hg loading. Moreover, Hg concentration is the result of a lifelong Hg accumulation process, and thus our dietary markers may not represent the cumulative lifetime feeding patterns of the consumers.

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