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Declining Mercury Concentrations in Bluefin Tuna Reflect Reduced Emissions to the North Atlantic Ocean

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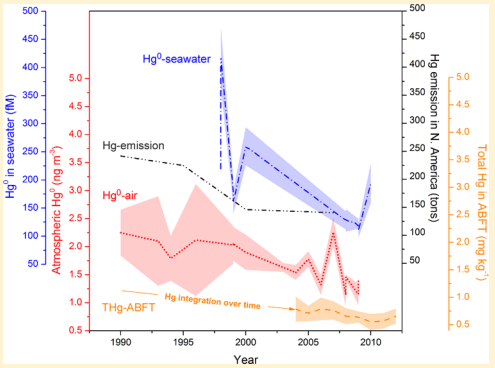
Supporting Information

ABSTRACT: Tunas are apex predators in marine food webs that can accumulate mercury (Hg) to high concentrations and provide more Hg (~40%) to the U.S population than any other source. We measured Hg concentrations in 1292 Atlantic bluefin tuna (ABFT, *Thunnus thynnus*) captured in the Northwest Atlantic from 2004 to 2012. ABFT Hg concentrations and variability increased nonlinearly with length, weight, and age, ranging from 0.25 to 3.15 mg kg^{-1}, and declined significantly at a rate of 0.018 ± 0.003 mg kg^{-1} per year or 19% over an 8-year period from the 1990s to the early 2000s. Notably, this decrease parallels comparably reduced anthropogenic Hg emission rates in North America and North Atlantic atmospheric Hg^0 concentrations during this period, suggesting that recent efforts to decrease atmospheric Hg loading have rapidly propagated up marine food webs to a commercially important species. This is the first evidence to suggest that emission reduction efforts have resulted in lower Hg concentrations in large, long-lived fish.



INTRODUCTION

High human consumption and moderate to high species-dependent Hg concentrations cause tunas to provide more Hg (~40%) to the U.S population than any other source.[1] All tuna species support large international fisheries due to high economic value and popularity as seafood. Relatively high Hg concentrations in larger, long-lived tunas have been posited as a potential health concern for frequent consumers, women of child-bearing age, fetuses, and young children.[2] Unresolved are trade-offs between Hg-dependent impacts on human health and health benefits of omega-3 fatty acids, selenium, and other nutrients in seafood.[3−5] Average Hg concentrations reported for different tuna species vary from 0.118 ppm (0.047−0.400 ppm) in canned skipjack tuna to 0.796 ppm (0.057−3.030 ppm) in wild bluefin tuna.[6] The long lifespan and highly migratory behavior of ABFT may make them valuable bioindicators of changes in Hg over time on basin-wide scales. In addition, bluefin tunas have enormous economic value, are fished globally, and are iconic species in ocean conservation campaigns.

Monomethylmercury (MeHg), a highly toxic and bioaccumulative form of Hg, is mainly formed by diverse bacteria that methylate inorganic Hg both in sediments and in the water column.[7−9] In the open ocean, MeHg levels have recently been reported to be higher in the subsurface oxygen-deficit zone than in deeper waters.[8,10,11] In addition, Lehnherr et al.[12] suggested that in situ methylation was the dominant source of MeHg in the mixed layer. Phytoplankton at the base of the food web concentrate (enriched about $10^5\times$) both inorganic and methylmercury from ambient seawater and serve as highly enriched sources of mercury for marine food chains.[13] Unlike inorganic Hg and most other metals, MeHg is transferred efficiently to subsequent trophic levels through diet and eliminated by aquatic animals at extremely low rates.[14] Thus, MeHg constitutes nearly all Hg in fish,[15,16] and muscle tissue concentrations are highest in upper trophic level animals (e.g., tunas, billfish, sharks).[6,17] Efficient transfer makes MeHg concentrations in marine predators especially sensitive to food web dynamics, including MeHg levels at the base of the food web[18] and in forage fish,[19] and variation in food chain length.[20]

Superimposed on ecological complexities are changing seawater Hg concentrations, caused in part by coal burning, gold mining, and other industrial activities. Recent efforts to reduce atmospheric contamination have resulted in reduced Hg loadings into the atmosphere in North America[21−23] and decreased concentrations in Atlantic Ocean surface waters.[21,24] A recent study reported declining trends of Hg in Atlantic bluefish (*Pomatomus saltatrix*) since the 1970s, reflecting a decline

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in Hg emissions.[25] Bonito et al.[26] synthesized available published data of Hg in numerous species of marine fish across the global ocean since 1969. They averaged all species and sizes from different trophic levels and reported a decline in mean Hg concentrations only in Altantic Ocean fish. However, the extent to which recent reductions have affected Hg concentrations in the largest, highest mercury fish (e.g., long-lived tunas) has not been quantified. Reported Hg concentrations in wild Atlantic bluefin tuna from different locations have varied widely.[27] Few data are available for Hg concentrations in western ABFT, and prior reports have often been based on small sample sizes that cannot represent populations or rigorously identify temporal trends. Relationships of Hg concentrations with ABFT age, length, weight, sex, or habitat have not been comprehensively examined in previous studies, limiting the conclusions that can be drawn from Hg measurements.

We measured total Hg concentrations in 1292 samples of wild-caught western ABFT for which length, weight, gender, year of capture, and location of capture were known. This unique data set allows assessment of Hg concentration changes in western ABFT over a time period in which atmospheric loading of Hg from North America declined. We report a decline in ABFT Hg concentrations over the period 2004–2012 that appears to reflect decreasing atmospheric loading in the North Atlantic Ocean, linking reduced Hg emissions to decreasing Hg concentrations in an apex ocean predator.

MATERIALS AND METHODS

Sample Information. ABFT tissue samples were collected from commercial landings from 2004 to 2012, primarily in the Gulf of Maine (n = 1279) and the Gulf of St. Lawrence (n = 13) by rod and reel and purse seine, for a total of 1292 samples comprising the LPRC (Large Pelagics Research Center) bluefin biosampling archive. Known capture locations are shown in Figure S1. All tissue was subsampled either from caudal white muscle tissue or from cranial muscle and stored frozen. Length (curved fork length or CFL in cm: a line tracing the lateral contour of the body from the tip of the upper jaw to the fork of the tail), weight, gender, year of capture, and approximate location of capture were recorded for each individual sampled fish. Body length and weight ranged from 65 to 326 cm (mean 218 ± 30 cm) and 58 to 347 kg (mean 132 ± 54 kg), respectively. The approximate age of western ABFT was estimated from CFL using an empirical growth curve generated from direct age–length observations from otoliths and modal progression data.[28] Uncertainty of age estimation increases in fish beyond 10 years of age.[29]

Total Hg Analysis. All samples were freeze-dried and homogenized prior to Hg analysis. Total Hg concentrations were determined using a DMA-80 direct Hg analyzer (Milestone, Inc.). Water content was determined in all samples (mean water content, 66 ± 7%), and dry weight concentrations were converted to a wet weight basis based on water content of each sample. Reported Hg concentrations in ABFT are wet weight concentrations unless otherwise noted. Given that >90% of Hg in tuna tissue is in the methylated form MeHg, we assumed that total Hg concentrations represent MeHg concentrations in ABFT. The DMA-80 was calibrated with an Hg standard solution (VHG Labs, Inc.). Certified reference material (DORM-4, NRCC) was measured to validate quality assurance. All analyses of Hg in reference material were within certified ranges (certified value 0.410 ± 0.055 mg kg^{-1}, measured value 0.395 ± 0.031 mg kg^{-1}, n = 43). Calibration checks were performed every 10 samples to monitor analyzer stability. To evaluate precision, duplicate samples were randomly measured in every set of analyses, and duplicate measurements of Hg concentrations varied by <3%.

Statistical Analysis. In order to assess interannual trends of Hg concentrations in ABFT, we grouped samples into age groups (Table S1, age estimated from length, as described above) in 1-year increments and compared mean Hg concentrations in same-aged fish across different years of capture (2004–2012). Age groups with sample size >50 (9–14 years old) were analyzed for temporal trends. Least-squares regressions of same-age ABFT Hg concentration versus year of capture were performed. Calculated slope values of Hg versus time for each age group were compared using an F-test for homogeneity of regression slopes. This was followed by an analysis of covariance (ANCOVA) to test adjusted means for age group effects. To assess the effect of departures from normality and homogeneity of variances in the data, the analysis was repeated on log-transformed Hg data, and the linear and log–linear models were compared using the MacKinnon–White–Davidson PE test.[30] Error structure effects were examined further by comparing linear model parametric results to bias- and skew-corrected bootstrap confidence intervals.[31] Data were resampled 5000 times with replacement within each age group by year of capture combination. The bias- and skew-corrected 95% confidence interval for the slope [−0.022, −0.015] differed only slightly from the parametric interval, indicating that normality and homogeneity assumption violations had little effect on the results.

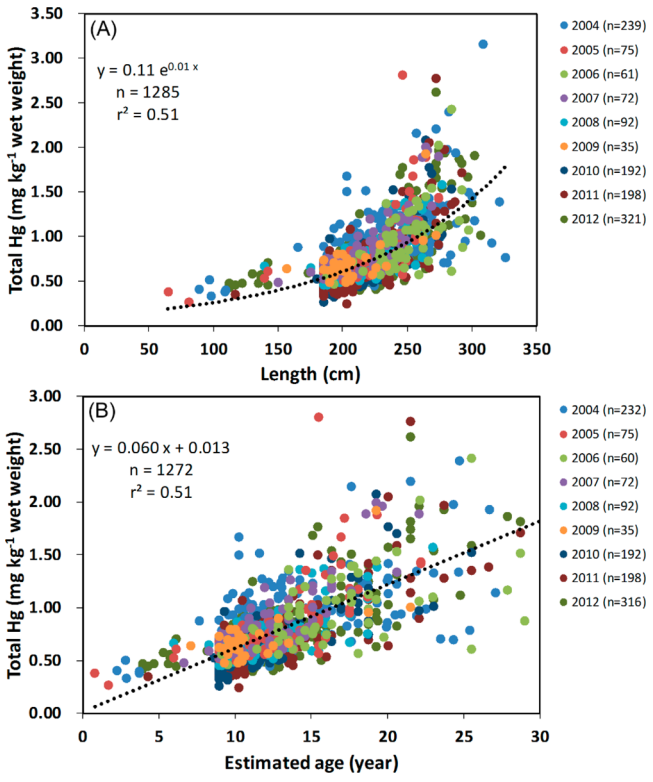

Figure 1. Relationship between total Hg in ABFT and individual ABFT (A) length and (B) estimated age. Circles represent individual Hg measurements with color indicating year of capture (2004–2012). Stippled black lines show (A) exponential and (B) linear fits to data. Estimation of age beyond 30 years using length–age algorithms is highly uncertain; thus, ABFT estimated >30 years old are not shown.

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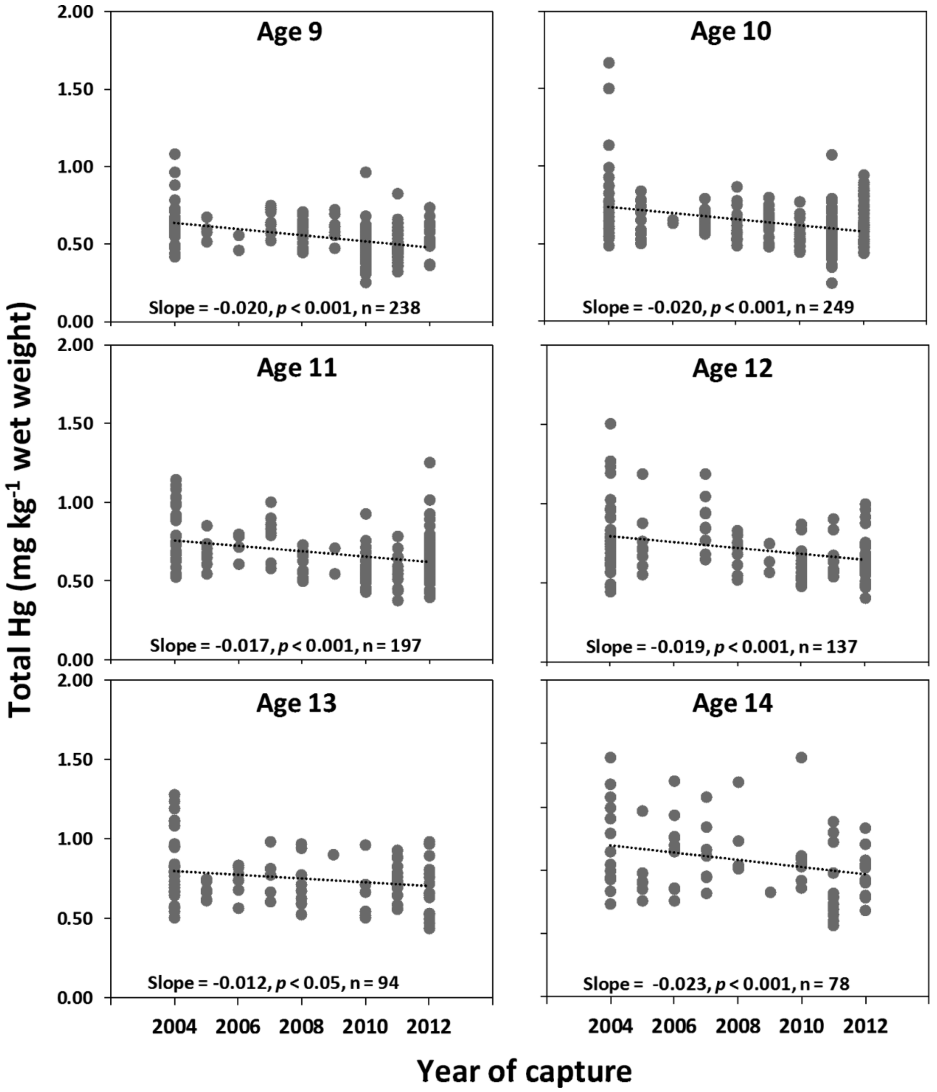

Figure 2. Hg concentrations in same-age ABFT decrease from 2004 to 2012. For each age group, circles represent Hg measurements for individual fish and stippled line shows linear fit to data. All slopes were negative and significant (see individual panels for p value of trend for each age class). Corresponding fish length of each age group is presented in Table S1.

Repeating the analysis on log-transformed Hg data resolved non-normality and heterogeneity of variance problems but altered the functional form of the relationship between Hg and year. PE tests comparing linear and log−linear models for each age group could not distinguish between the two models; thus, the linear model was accepted because of its simplicity.

We also compared ABFT Hg concentrations by year to assess whether changes could be detected on year-to-year time scales. ABFT Hg concentrations were grouped by year (2004−2012) and compared using the nonparametric Kruskal−Wallis test. Analyses were carried out in MATLAB vR2015a and in R v3.2.2.

RESULTS AND DISCUSSION

The mean Hg concentration in ABFT muscle samples was 0.76 ± 0.33 mg kg^{-1} (n = 1292, range 0.25−3.15 mg kg^{-1}). Hg concentrations in ABFT correlated positively with length and age (Figure 1). Hg concentrations did not differ significantly between male and female ABFT (Figure S2) for which sex was identified (n = 609, 47%). Variability of Hg concentrations was lowest among smaller fish (<200 cm) and increased markedly with size, particularly among very large fish (>250 cm) (Figures 1 and S3). Using dissolved MeHg concentrations in surface waters of the Gulf of Maine (3−11 pg L^{-1})[32] and recent data in North Atlantic surface water (12 ± 10 pg L^{-1}),[10] bioconcentration factors of MeHg in ABFT (MeHg in fish divided by MeHg in seawater) were up to 10^8.

Hg in ABFT declined significantly from 2004 to 2012 within each age group analyzed (ages 9−14 years old, see Methods; Figures 2 and S4, Table S2). Slopes relating ABFT Hg concentrations to time did not differ significantly across age groups (F = 0.557, df = [5, 981], p = 0.733; Figure S5). The overall decline in tissue Hg for the pooled data (see Methods) was 0.018 mg kg^{-1} per year (95% CI = [−0.021, −0.015]), corresponding to an average decline of 19% from 2004 to 2012. Comparison of adjusted regression means in ANCOVA indicated a highly significant age group effect (F = 61.2, df = [5, 986], p < 0.001). Our calculated decline rate of Hg in ABFT (~2% year^{-1}) was comparable to that reported for Atlantic bluefish (*Pomatomus saltatrix*)[25] and mean Atlantic fish (many species, trophic levels, ages combined).[26]

Figure 3 shows that the decline in ABFT Hg parallels declining Hg emissions in North America (−2.8% year^{-1} from

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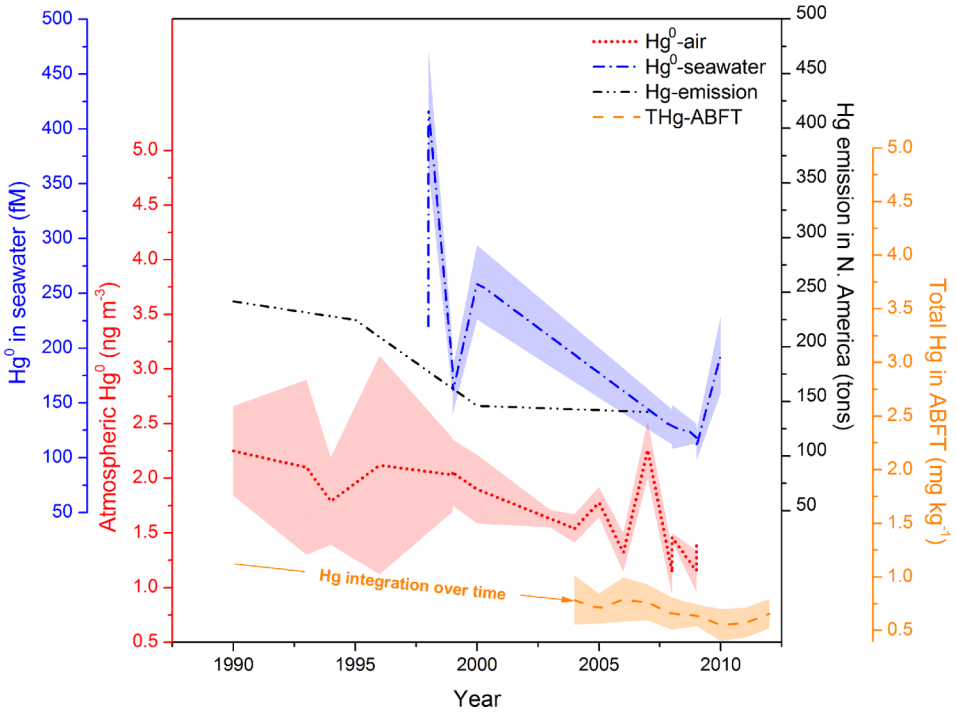

Figure 3. Temporal trends of anthropogenic Hg emission in North America (n = 4),[21,22] atmospheric Hg^0 above the North Atlantic (n = 18),[33] dissolved gaseous Hg^0 in North Atlantic surface water (n = 10),[33,34] and total Hg in ABFT caught in the Gulf of Maine (n = 993, age group 9−14). Shaded areas show variability (1 standard deviation) around measurements. Note that Hg in ABFT was an integration of Hg accumulated over the entire lifetime of the fish.

1990 to 2007),[21,22] Hg(II) wet deposition fluxes in North America (−1.6% $year^{-1}$ from 1996 to 2013),[23] atmospheric Hg^0 concentrations 12 m above sea surface over the North Atlantic (−2.5% $year^{-1}$ from 1990 to 2009),[33] and seawater Hg^0 concentrations (−4.3% $year^{-1}$ from 1998 to 2010).[33,34] As there is no long-term record of dissolved MeHg in the North Atlantic, temporal trends in surface seawater Hg^0 concentrations were compared with ABFT Hg levels, although it is recognized that dissolved Hg^0 concentrations are variable and subject to local conditions at the time of sampling. Over the 8-year period (2004−2012) Hg in ABFT declined by 19%, compared to a 20% decline in Hg^0 in North Atlantic air for the overlapping 8-year period 2001−2009. Thus, an ABFT of a given age, caught in 2004, spent more of its life exposed to higher 1990s oceanic Hg concentrations than one caught in 2012 (Figure S4). Higher Hg concentrations in seawater in the 1990s presumably propagated up food webs to prey (forage fish, squids: Table S3) and ultimately to ABFT, whereas declines in Hg emissions led to reduced Hg at the forage base resulting in gradually declining ABFT Hg concentrations in the early 2000s.

While global Hg emissions to the atmosphere continue to increase due largely to increased coal burning in Asia,[35] regional declines of atmospheric Hg deposition into the North Atlantic Ocean have resulted from reduced North American Hg emissions in the past few decades.[23] This regional decline may explain the 1% per year decrease in Mid-Atlantic Bight bluefish Hg concentrations from 1972 to 2011.[25] Declining bluefish Hg indicates that other, more frequently consumed ABFT prey may have experienced similar declines. This reported decline in bluefish was based on relatively few samples from initial study years (n = 54; 1972−1973) compared with many more samples taken ≥20 years later. Trends potentially were driven by a few endmembers for larger fish or for temporal endpoints.

Rates of Hg decline reported here for ABFT are based on year-to-year measurements of sample sizes high enough to discern reliable trends despite observed variability. Interannual comparisons reinforce the importance of large sample size across long study periods. For example, Hg in 2004 was no different from 2005 to 2007 but was much higher than 2008−2012 (Table S4a). Because within-year ABFT Hg variability was high and the slope of change relatively low, years of data may be required for detection of trends in tunas and other marine species. Slow changes in tuna Hg may be driven by long (>1 year) histories reflected by muscle Hg of large marine predators. Kwon et al.[36] reported a biological half-life of Hg in captive Pacific Bluefin tuna (PBFT) of ∼500 days. Assuming comparable metabolic rates of ABFT and PBFT and given the larger size of ABFT in this study, Hg levels in ABFT likely represent exposure periods of ≥500 days.

Intra-annual variability of ABFT Hg concentrations was very high, representing 69−85% of the total variability in each age group. Pairwise comparisons of ABFT Hg concentrations between consecutive years were often not significant, emphasizing the need to carry out age-group-specific analyses over multiple years to identify temporal trends. Of the consecutive year pairwise comparisons, 63% were nonsignificant at both an individual (Table S4a) and family (Table S4b) error rate of 0.05. Although requiring large samples and a multiyear time span to identify the negative trend in ABFT Hg concentrations, this trend was consistently observed across all age groups and survived all challenges to the form of the regression model and error structure (see Methods and Table S2).

Since Hg accumulates in fish over time and ABFT are long-lived, variability in large ABFT may represent time-integrated diet differences. Large ABFT feed on a larger size range of prey than smaller ABFT and encounter different prey in various

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ocean regions.[37,38] Thus, variability in trophic levels and Hg concentrations of prey for large ABFT are greater than those of smaller ABFT. Similar increasing Hg variability with ABFT size has been observed for other fish species off the east coast of North America.[39]

Shifts in ABFT diet over time provide a putative explanation for observed declines in Hg of western ABFT. Specific prey items are important, prey abundance is not constant, and declines in ABFT Hg could result from shifts to low Hg prey (e.g., menhaden) (Table S3). However, diet studies of ABFT completed over the same spatiotemporal range of this study indicate that herring remained a dominant prey component of adult size classes.[37] It also could be argued that variation of migratory histories within specific ABFT cohorts could affect Hg trends. However, the declining trend in ABFT Hg was consistent across multiple age groups, suggesting that age-specific migratory differences cannot account for this observed decline. Furthermore, Hg concentrations of primary ABFT prey in the east Atlantic (myctophids, anchovy, squids) are comparable to western Atlantic prey (Table S3). Regardless, a decrease in bioavailable Hg in the North Atlantic would presumably decrease Hg in pelagic prey items throughout this ocean basin (as has been suggested for bluefish),[25] causing lower Hg in ABFT and other apex predators in the Atlantic.

Declining Hg in ABFT parallels declines of atmospheric Hg concentrations, suggesting that Hg in ABFT is linked to human activities and that recent efforts to decrease atmospheric Hg loading have propagated up marine food webs to commercially exploited apex predators. It also suggests that marine animals can respond concurrently to changes in ocean Hg deposition and that continued efforts to decrease atmospheric loading of Hg could further lower Hg concentrations in seafood. The declines in ABFT MeHg concentrations observed on a decadal time scale suggest a short residence time for Hg in the mixed layer, which is consistent with water column measurements that suggest a residence time of about 1 year.[40] This study provides the first evidence that efforts to reduce mercury emissions have positively impacted bluefin tuna. It remains to be investigated if other tuna species have responded similarly. Robust analyses of both prey and predator species and their ecological relationships should refine our understanding of regional and temporal benefits of reducing ocean Hg contamination.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b04328.

Additional tables and figures as discussed in the manuscript (PDF)

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Notes

The authors declare no competing financial interest.

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