

Atlantic Bluefin Tuna (*Thunnus thynnus*) Population Dynamics Delineated by Organochlorine Tracers

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Atlantic bluefin tuna (ABFT) are highly valued and heavily exploited, and critical uncertainties regarding their population structure hinder effective management. Evidence supports the existence of two breeding populations of ABFT; a western population in the Gulf of Mexico and an eastern population in the Mediterranean Sea; both of which migrate and mix in the North Atlantic. Conventional tagging studies suggest low rates of trans-Atlantic migrations; however, electronic tagging and stable isotopes in otoliths indicate stock mixing up to 57% between management zones delineated by 45° W longitude. Here we show that organochlorine pesticides and polychlorinated biphenyls (PCBs) can be used as tracers of bluefin tuna foraging grounds in the North Atlantic and confirm that stock mixing of juvenile tuna within the U.S. Mid Atlantic Bight is indeed high (33–83% eastern origin), and is likely spatially and temporally variable. We further demonstrate that >10% of the Mediterranean population is migratory, that young bluefin tuna migrate from the Mediterranean to western Atlantic foraging grounds as early as age 1, and then return to the Mediterranean Sea as young as age 5, presumably to breed. The tracer method described here provides a novel means for distinguishing bluefin tuna populations and ontogenetic shifts in migration in the North Atlantic.

Introduction

Atlantic bluefin tuna (ABFT), high-valued recreational and commercial fish, are distributed from subtropical to subarctic regions throughout the North Atlantic (1). The member

nations of the International Commission for the Conservation of Atlantic Tunas (ICCAT) currently manage ABFT fisheries assuming two units (a western stock spawning in the Gulf of Mexico, and an eastern stock which spawns in the Mediterranean Sea) ostensibly separated by the 45° W meridian with little intermixing between stocks. However, tagging studies indicate that bluefin tuna undergo extensive and complex migrations, including trans-Atlantic migrations, and that stock mixing could be as high as 30% (2–4). Extensive mixing of eastern and western stocks (35–57% bluefin tuna of eastern origin) within the U.S. Mid Atlantic Bight was also reported recently based on otolith $\delta^{18}\text{O}$ values (5). The uncertainty of stock structures due to mixing makes it difficult for fisheries managers to assess the effectiveness of rebuilding efforts for the dwindling western Atlantic spawning stock of bluefin tuna. Understanding ABFT spatial distributions and dynamics are vital for robust population assessments and the design of effective management strategies, and there is a critical need for improved methods to resolve key attributes of this highly migratory species (1).

Reports of low levels of chlordane compounds (*cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, oxychlordane) relative to polychlorinated biphenyls (PCBs) in marine species from the Mediterranean Sea (MS, 6, 7) compared to the western North Atlantic (WNA, 8–11) led us to propose chlordane/PCB ratios as chemical tags for fish feeding in these geographically distinct ecosystems. PCBs and chlordanes are synthetic chemicals that were released into the environment by human activity, bioaccumulate in organism lipids, and biomagnify, increasing in concentration with trophic level such that top predators attain the highest concentrations (12–15). Nonmetabolizable PCB congeners persist in fish and the environment; likewise, chlordanes, a group of organochlorine pesticides are slowly metabolized, if at all, by fish (16, 17). PCB and chlordane concentrations increase with fork length in Pacific bluefin tuna (*Thunnus orientalis*) from juveniles through adult sized fish (18), indicating that uptake exceeds elimination and that persistent organochlorine compounds are retained in these fish such that they could be useful tracers of bluefin tuna foraging regions over time scales of years.

The objective of our research was to establish the utility of PCBs and organochlorine pesticides as tracers of ABFT natal origin and stock mixing. A tracer technique based on the ratios of chemical markers was proposed as it is advantageous compared to measurement of absolute concentrations of specific markers for various reasons. First, concentrations of persistent organochlorine compounds increase with fish lipid content (10) and size (18), and are also higher in fish that reside in more contaminated habitats (19, 20). These confounding variables can be eliminated by using compound ratios to assign origin to an individual within a mixed population of fish provided that the compound ratios differ significantly between foraging habitats and remain constant in fish residing within a single habitat. Large differences in the relative amounts of chlordanes and PCBs are found in marine organisms from the MS and WNA, as noted above, and data from the Sea of Japan demonstrate linear increases in PCBs and chlordanes with size such that the ratio of these compounds remains constant over the life span of bluefin tuna residing within a specific ecosystem (18). Therefore, chlordane/PCB ratios are likely to be useful for distinguishing ABFT origin and stock mixing. Second, there is often interlaboratory variability in measurement of absolute concentrations of PCBs and pesticides based on extraction techniques, sample recoveries, and instrument

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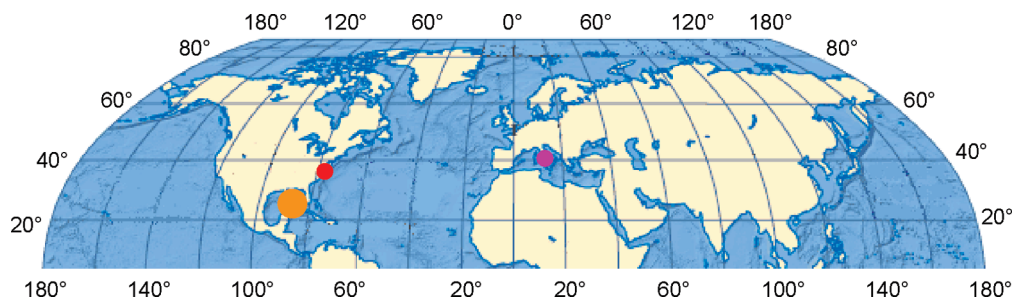


FIGURE 1. Atlantic bluefin tuna sampling locations in the Gulf of Mexico (orange), western North Atlantic (red), and Mediterranean Sea (purple).

TABLE 1. Size Classes, Length/Weight, and Estimated Ages for Atlantic Bluefin Tuna Sampled^a

location	size class	fork length (cm)	weight (kg)	age (y)	<i>n</i>
Western North Atlantic	young-of-the-year	23–38		0	19
	small school	63–91		2	23
	large school	111–139		3–4	15
Gulf of Mexico	medium to giant	159–267		5–19	16
Mediterranean Sea	young-of-the-year	34–50	0.7–2.1	0	6
	large school	96–143	15–50	3–5	20
	medium to giant	160–223	70–189	7–12	7
	unidentified				11

^a Italicized values calculated as described in text; *n* = sample number.

response. Such interlaboratory variability is minimized by evaluating compound ratios. Moreover, using compound ratios obviates the need to measure organism lipids, eliminating another potential source of variability. Finally, distinguishing the food web origin of an animal based on compound ratios versus absolute concentrations means that sample analysis is not limited to a single tissue type provided that there is sufficient chemical signal in the tissue analyzed and there is no tissue-dependent metabolism of the tracer. Consequently, the method presented here using chlordane/PCB ratios to distinguish eastern and western stocks of ABFT is expected to be more robust than one based on measurement of PCBs or chlordanes alone.

Materials and Methods

Sample Collection. Muscle tissue samples were collected from bluefin tuna caught in three separate regions of the North Atlantic (Figure 1). The fish were classified according to size based on U.S. National Marine Fisheries Service size categories for ABFT (21). Western North Atlantic (WNA) bluefin tuna samples were collected within the U.S. Mid Atlantic Bight in coastal waters off Point Pleasant, New Jersey (small school) during Sept.–Nov. 2006 and 2007, the eastern shore of Virginia (small and large school) during June 2006, and off the coast of Virginia Beach, VA Aug.–Sept., 2008 (young-of-the-year (YOY)) (Table 1). Medium to giant bluefin tuna samples from the Gulf of Mexico (Table 1) were obtained from the National Ocean Services Marine Forensics Archive in Charleston, South Carolina and were all from fish caught during the breeding season (April–June (1)) in 2000 and 2002. Mediterranean Sea (MS) tuna samples were collected between May and October 2003 from the southern Tyrrhenian Sea (22) and included YOY and various larger sized bluefin tuna (Table 1). All tissue samples were frozen as soon as possible after collection until analysis, and were extracted and purified as described elsewhere (22, 23).

Analyses. All extracts were analyzed for *trans*-nonachlor, *cis*-nonachlor, PCB153, and PCB187 by gas chromatography/negative chemical ionization mass spectrometry as previously described (23) using a J&W DB-XLB narrow bore capillary

column (30 m length, 0.18 mm diameter, 0.18 μm film thickness), selective ion monitoring, and a slightly modified temperature program to allow for analysis of PCBs and organochlorine pesticides in a single run. Method parameters for analysis were as follows: 70 °C, initial hold time of 1 min; 70–150 °C @ 20 °C min⁻¹; 150–230 °C @ 10 °C min⁻¹, hold for 5 min; 230–300 °C @ 6 °C min⁻¹, hold for 3 min, source temperature 150 °C and a quad temperature of 130 °C. Gulf of Mexico and WNA samples were analyzed at the Virginia Institute of Marine Science, and the MS sample extracts were analyzed at the University of Florence using the same method. All chemical signals exceeded the method detection limit (3 × the average blank level) by >10:1 in all samples except for *cis*-nonachlor in a single sample from the WNA in which it was not detected. Gulf of Mexico and WNA samples were quantified relative to PCB204 added as an internal standard; MS samples were analyzed relative to PCB209 added as an internal standard (22). Recoveries of spiked internal standards averaged 95(±31)% (mean ± standard deviation) for the Gulf of Mexico and WNA samples, and between 86(±19)% and 97(±15)% (mean ± standard deviation) for various PCB congeners in spiked samples run in conjunction with the MS samples (22). Potential interlaboratory bias in the reported marker ratios was assessed by evaluating the differences in the relative response factors (RRFs) of *trans*-nonachlor and *cis*-nonachlor relative to PCB153 and PCB187, respectively, for a series of response factor standard analyses. There was no significant difference (two-tailed *t* test, *P* > 0.05, *df* = 28) in RRF_{*trans*-nonaCl}/RRF_{PCB153} and RRF_{*cis*-nonaCl}/RRF_{PCB187} between laboratories indicating that observed differences in the measured nonachlor/PCB ratios between samples collected from the WNA and MS are entirely due to differences in the mass of the marker compounds in the tissue samples.

Length, Weight, and Age Calculations. Fork length (FL; (cm)) was measured directly or estimated using the following equation: 0.9201 · CFL (CFL = curved fork length, *n* = 3, *r*² = 0.963, *P* = 0.012) for fish collected from the WNA, and estimated for MS tuna using the following equation: 38.707(wt)^{0.334} where wt = weight (kg) (24, 25). Weight (kg) was estimated for fish collected in the WNA and determined

to be of eastern origin using the following equation: $2(10)^{-5} FL^{2.991}$ (24, 25). Fish age was estimated using the following equations: $0.8614e^{0.0115*FL}$ and $0.0009e^{1.7566*FL}$ derived from age-length data for bluefin tuna in the WNA and MS, respectively (26).

Results and Discussion

Selection of Organochlorine Tracers. Two chlordane compounds (*trans*-nonachlor, *cis*-nonachlor) and two recalcitrant PCB congeners (PCB153, PCB187) were selected for use in evaluating ABFT migration and mixing patterns. PCB congeners 153 and 187 are prominent in tuna tissue samples (7, 22) and are essentially nonmetabolizable in many organisms because their molecular structures do not contain vicinal C–H pairs that allow for epoxide formation, a necessary step in PCB metabolism via CYP isoenzymes (27–29). Among the chlordane compounds, accumulation rates in bluefin tuna are highest for *trans*-nonachlor \gg *cis*-chlordane $>$ *cis*-nonachlor $>$ oxychlordane \gg *trans*-chlordane (18), indicating that the nonachlors and *cis*-chlordane are metabolized slowly, if at all, into oxychlordane by bluefin tuna as observed in other fish species (30–32). High levels of Endosulfan I in ABFT, however, caused significant interference with the analysis of *cis*-chlordane, therefore, *trans*- and *cis*-nonachlor were selected for use as tracers in the present study.

Baseline Nonachlor/PCB Ratios in Young-of-the-Year Tuna. YOY bluefin tuna from the MS and WNA were used to determine the expected baseline nonachlor/PCB ratios for ABFT feeding in these food webs as it is expected that YOY fish have not yet undergone a trans-Atlantic migration (33). Ratios of *trans*-nonachlor/PCB153 (mean \pm standard error) were $0.007(\pm 0.006)$ and $0.282(\pm 0.013)$ in YOY tuna from the MS and WNA, respectively, and were significantly different (two-tailed *t* test, $P < 0.0001$) between the two foraging regions. Likewise, ratios of *cis*-nonachlor/PCB187 (mean \pm standard error) were $0.031(\pm 0.022)$ and $0.277(\pm 0.022)$ for YOY tuna from the MS and WNA, respectively, and were significantly different (two-tailed *t* test, $P < 0.0001$) between the MS and WNA. These results are consistent with previous studies that show low levels of chlordanes relative to PCBs in marine organisms in the MS versus WNA (6–11), and further indicate that nonachlor/PCB ratios will be useful tracers of fish that have foraged in these distant food webs.

Nonachlor/PCB Ratios in Gulf of Mexico Tuna. Nonachlor/PCB ratios in medium to giant-sized ABFT (>159 cm FL; Table 1) captured in the Gulf of Mexico during the spawning season were similar to, or slightly greater than, those of YOY tuna from the WNA (Figure 2). This indicates that adult fish entering western breeding grounds have foraged along the WNA coast as opposed to in the MS, consistent with previous studies that indicate little or no stock mixing on known spawning grounds (4, 5). Specifically, electronic tagging studies show that adult ABFT that migrate throughout the North Atlantic and enter western spawning grounds in the Gulf of Mexico do not enter the MS (4). Consistent with this finding we found no evidence of biomass acquisition in the MS (i.e., nonachlor/PCB ratios $<$ WNA YOY) among mature tuna captured in the Gulf of Mexico. Rather, the substantial overlap in nonachlor/PCB ratios for medium to giant-sized bluefin tuna from the Gulf of Mexico and WNA YOY illustrates the importance of WNA foraging grounds for western spawning ABFT.

Nonachlor/PCB Ratios in WNA Juvenile Tuna. Nonachlor/PCB ratios in juvenile ABFT captured within the U.S. Mid Atlantic Bight distinguished these fish as (a) individuals composed of biomass accumulated predominantly ($>90\%$) in the WNA with nonachlor/PCB ratios indistinguishable from WNA YOY, or (b) organisms composed of biomass accumulated in substantial proportions within both of the two

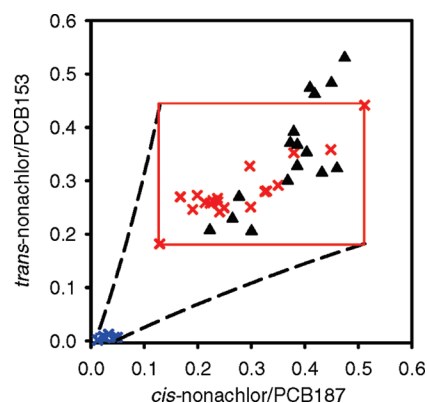


FIGURE 2. Nonachlor/PCB ratios for medium to giant ABFT captured in the Gulf of Mexico (black symbols). Red symbols and square show the values and range for YOY bluefin tuna from the WNA, and blue symbols and square show the values and range for YOY bluefin tuna from the MS. Dashed curves delineate the range in expected values for fish composed of biomass acquired in both the WNA and MS.

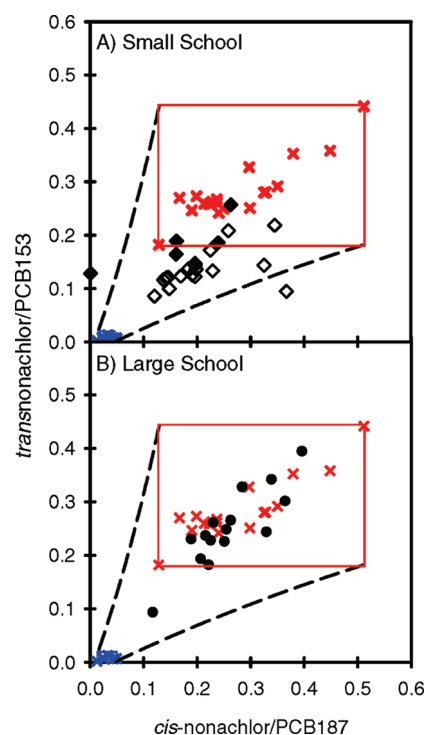


FIGURE 3. Nonachlor/PCB ratios for (A) small and (B) large school-sized bluefin tuna captured in the WNA within the U.S. Mid Atlantic Bight. Closed black symbols correspond to fish captured off the VA coast in June 2006; open black symbols correspond to fish captured off of the NJ coast in fall 2006–2007. Red symbols and squares show the values and range for YOY bluefin tuna from the WNA, and blue symbols and squares show the values and range for YOY bluefin tuna from the MS. Dashed curves delineate the range in expected values for fish composed of biomass acquired in both the WNA and MS.

foraging regions examined with nonachlor/PCB ratios falling in between the range of values measured in YOY from the MS and WNA (Figure 3).

Among the small school size class (63–91 cm FL; Table 1) of ABFT sampled in the WNA, 70% had *trans*-nonachlor/PCB153 ratios indicative of fish that have migrated from the MS (i.e., *trans*-nonachlor/PCB153 ratios $<90\%$ that of the lowest value measured in WNA YOY; Figure 3a). Of these putative migrants from the MS, two were from a total of six

small school fish (33%) captured along the VA coast in June 2006, and 10 were out of a total of 12 small school fish (83%) captured along the NJ coast in Sept.–Nov. 2007, indicating that stock mixing of juvenile bluefin tuna in the WNA is high and may be spatially and temporally variable. Our findings confirm a recent report of extensive stock mixing among school-sized ABFT within the U.S. Mid Atlantic Bight (5).

The putative small school fish of eastern origin in the WNA would have initially had low nonachlor/PCB ratios that turn over (change signal) due to foraging in the WNA coincident with accumulation of biomass tagged with higher nonachlor/PCB ratios. Marker ratio turnover in ABFT muscle tissue appears to be more rapid for *cis*-nonachlor/PCB187, likely due in part to the smaller initial difference in signals between eastern and western fish (Figure 3). Changes in nonachlor/PCB ratios in fish that move from the Mediterranean to WNA foraging grounds can be predicted using a two-component, nonlinear mixing model (34, 35):

$$F_{MS} = \frac{(R_C - R_{WNA})(R_{MS} + 1)}{(R_{MS} - R_{WNA})(R_C + 1)} \quad (1)$$

where F_{MS} is the fraction of body mass gained prior to emigration from the MS, R_{MS} is the upper limit of the nonachlor/PCB ratio for fish feeding exclusively in the MS, R_{WNA} is the lower limit of the nonachlor/PCB ratio for fish feeding exclusively in the WNA, and R_C is the nonachlor/PCB ratio at the time of collection. Solving this equation using the measured *trans*-nonachlor/PCB153 ratios in the small school-sized bluefin tuna captured in the WNA and identified as originating from Mediterranean nursery grounds, indicates that the average fraction of biomass acquired by these fish in the MS prior to emigration was 0.31 (± 0.10) (mean \pm standard deviation). Based on length–age relationships (26), all of the small school-sized bluefin tuna sampled in the WNA were age 2 at the time of collection (Table 1). This implies that small school-sized bluefin tuna captured within the U.S. Mid Atlantic Bight in the present study, and identified as originating from the eastern Atlantic, migrated at age 1 from the Mediterranean to WNA foraging grounds, which is consistent with the migration patterns of young Pacific bluefin tuna (36).

Of 15 large school-sized fish (111–139 cm FL; Table 1) collected in the WNA, only one could be distinguished as originating in the MS (Figure 3b). This apparently low extent of stock mixing compared to other recent estimates for this size class of tuna taken from the U.S. Mid Atlantic Bight (5) is likely due to complete turnover of the nonachlor/PCB ratios in bluefin tuna of eastern origin that migrated to the WNA at age 1 such that these fish can no longer be distinguished from those of western origin. Nonetheless, the single large school fish identified as being of eastern origin indicates that some adolescent bluefin tuna from the MS undergo trans-Atlantic migration after age 1. Using the model equation above, the single large school fish identified as having migrated to the WNA from the east is estimated to have emigrated from the MS at age 2 and gained 51% of its body mass prior to capture while foraging in the WNA.

Nonachlor/PCB Ratio Turnover Times in Juvenile Bluefin Tuna. As noted above, concentrations of PCBs and nonachlors increase with size in juvenile through adult-sized bluefin tuna (18) indicative of consistent uptake with minimal elimination of these highly lipophilic compounds. Moreover, the PCBs and nonachlors selected as tracers are essentially nonmetabolizable in bluefin tuna (27–32). Therefore, turnover of the nonachlor/PCB ratios in ABFT that migrate between food webs is a function solely of the acquisition of biomass by these fish of indeterminate growth, with replacement of the nonachlor/PCB ratios reflective of prey in the initial foraging region with that of the nonachlor/PCB

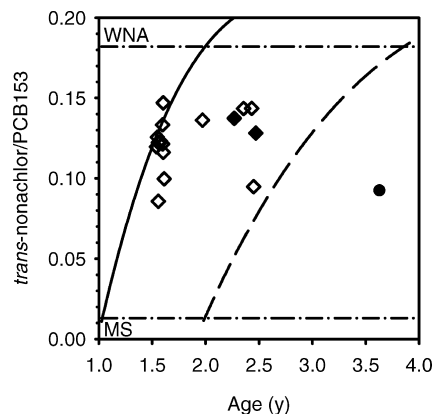


FIGURE 4. *trans*-Nonachlor/PCB153 ratios and modeled turnover times for school-sized bluefin tuna from the MS captured in the WNA. Symbols are the same as in Figure 3. Solid curve depicts the modeled turnover of the *trans*-nonachlor/PCB153 ratio in a MS fish arriving in the WNA at age 1; dashed curve depicts the modeled turnover of the *trans*-nonachlor/PCB153 ratio in a MS fish arriving in the WNA at age 2. Dashed-dot lines correspond to the upper and lower measured values for *trans*-nonachlor/PCB153 ratios in YOY fish from the MS and WNA, respectively.

ratios reflective of prey from the current foraging region, such that the turnover of the nonachlor/PCB signal is predicted based on the fraction of body mass the fish has acquired in each food web. Rearranging eq 1, and using average values for R_{MS} and R_{WNA} from YOY fish, R_C was calculated for 1- and 2-year-old tuna that migrate from the MS to the WNA where they feed and grow, decreasing F_{MS} with age. These calculations indicate that the turnover time for the *trans*-nonachlor/PCB153 ratio in MS bluefin tuna that arrive on WNA feeding grounds at age 1 is 10 months and increases to 1.6 y for MS tuna that arrive in the WNA at age 2 (Figure 4). Plotted along with these curves are the *trans*-nonachlor/PCB153 ratios and estimated ages for school-sized bluefin tuna captured in the WNA and identified as eastern migrants. The data demonstrate that the majority of these migrants arrived in the WNA between age 1 and 2 y.

Nonachlor/PCB Ratios in Mediterranean Bluefin Tuna.

Complete turnover of the nonachlor/PCB ratios in juvenile bluefin tuna of eastern origin while foraging in the WNA suggests that individuals subsequently returning to the MS should be readily identified. Indeed, five of the 38 bluefin tuna > age 1 collected from the MS (13%) were clearly identifiable as having recently returned from foraging outside of the Mediterranean with nonachlor/PCB ratios overlapping those of WNA YOY as opposed to YOY from the MS (Figure 5). These recent migrants ranged in size from 35 to 178 kg (age 5–11 y) and were all captured during the summer fishery, which is suggested to be composed of both resident and migratory fish (37) at a time when Mediterranean bluefin tuna are known to spawn (4). Since western spawning tuna are not known to enter the MS (4) we presume that fish tagged with WNA nonachlor/PCB ratios captured in the Mediterranean are eastern spawning fish that have returned to the MS possibly to breed. Moreover, the small fraction of migrants returning to the MS (13%) compared to the large fraction of eastern emigrants in the WNA (33–83%) is reflective of the different sizes of these fish stocks, with the eastern spawning stock biomass estimated to be 5–10 times larger than that of the western stock (1).

Lastly, several bluefin tuna collected from the MS had nonachlor/PCB ratios much lower than YOY from the WNA, but greater than MS YOY (Figure 5). Because young bluefin tuna are thought to be more likely to undergo trans-Atlantic migration compared to adults (4, 5), these fish may include individuals that migrated to the WNA as juveniles, but which

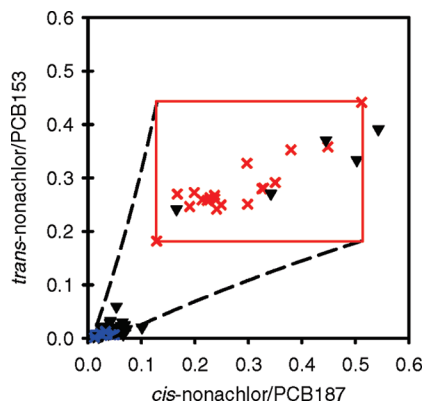


FIGURE 5. Nonachlor/PCB ratios for bluefin tuna (age >1 y) captured in the MS (black symbols). Red symbols and square show the values and range for YOY bluefin tuna from the WNA, and blue symbols and square show the values and range for YOY bluefin tuna from the MS. Dashed curves delineate the range in expected values for fish composed of biomass acquired in both the WNA and MS.

subsequently returned to, and remained in, the MS (e.g., after spawning). The nonachlor/PCB ratios in migrants that have returned to the MS from the WNA would again turn over, changing from a signal reflective of the WNA to that of the MS as they feed and acquire body mass in the MS.

Method Validation and Application. The method described here exploits large differences in levels of persistent organochlorine pollutants in food webs in geographically distant ecosystems to gather insights into the migration patterns and stock mixing of ABFT. This technique or similar methods may likewise be useful for acquiring ecological information on other highly migratory species, particularly when migrants periodically occupy regions with markedly different levels of long-lived chemical tracers in the food web.

Although our results are consistent with current scientific understanding of the population ecology of ABFT these findings should be validated with larger and more representative samples, and by using other techniques including conventional or electronic tags, genetics, or otolith stable isotope measurements. For example, we surmise that tuna captured in the MS with nonachlor/PCB ratios indicative of the WNA (Figure 5) are fish of eastern origin that migrated to the WNA and subsequently returned to the MS to breed. This supposition could be validated by measuring $\delta^{18}\text{O}$ in otoliths to determine natal origin (5) along with nonachlor/PCB ratios in muscle tissue to evaluate recent foraging grounds of ABFT from the MS.

In addition to validation of the technique, it remains to be determined if the chemical signatures we measured are unique to the MS and WNA, or if other regions routinely occupied by ABFT (e.g., Gulf of Maine, Gulf of St. Lawrence, Bay of Biscay, west coast of Morocco; (1)) impart similar nonachlor/PCB ratios to the fish. Moreover, it will be important to determine if there is spatial (e.g., within the MS) and temporal variation in the baseline nonachlor/PCB ratios in YOY bluefin tuna. Nonetheless, our novel application of persistent organochlorines as tracers to evaluate bluefin tuna migration and stock mixing is promising and potentially offers significant advantages particularly when used in combination with more established methods.

As with genetic markers, chemical tags are acquired by all animals within a population or region allowing for sampling of the entire population as compared to a sub-component of the population affixed with physical tags that can potentially be biased by size. Moreover, the large difference in nonachlor/PCB ratios between the WNA and

MS food webs coupled with tissue turnover times of ≥ 1 y for these markers in ABFT allows not only migrants between foraging regions to be identified, but also the ontogeny of migration to be evaluated. Therefore, paradoxically, important ecological information that may help to conserve and manage a highly exploited species may accrue from the inadvertent contamination of its food web with persistent pollutants.

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Supporting Information Available

Tables of the measured nonachlor and PCB masses and mass ratios in ABFT. This information is available free of charge via the Internet at <http://pubs.acs.org>.

Literature Cited

- Fromentin, J. M.; Powers, J. E. Atlantic bluefin tuna: population dynamics, ecology, fisheries and management. *Fish Fish.* **2005**, *6*, 281–306.
- Lutcavage, M. E.; Brill, R. W.; Skomal, G. B.; Chase, B. C.; Howey, P. W. Results of pop-up satellite tagging of spawning size class fish in the Gulf of Maine: Do North Atlantic bluefin tuna spawn in the mid-Atlantic? *Can. J. Fish. Aquat. Sci.* **1999**, *56*, 173–177.
- Block, B. A.; Dewar, H.; Blackwell, S. B.; Williams, T. D.; Prince, E. D.; Farwell, C. J.; Boustany, A.; Teo, S. L. H.; Seitz, A.; Walli, A.; Fudge, D. Migratory movements, depth preferences, and thermal biology of Atlantic bluefin tuna. *Science* **2001**, *293*, 1310–1314.
- Block, B. A.; Teo, S. L. H.; Walli, A.; Boustany, A.; Stokesbury, M. J. W.; Farwell, C. J.; Weng, K. C.; Dewar, H.; Williams, T. D. Electronic tagging and population structure of Atlantic bluefin tuna. *Nature* **2005**, *434*, 1121–1127.
- Rooker, J. R.; Secor, D. H.; De Metrio, G.; Schloesser, R.; Block, B. A.; Neilson, J. D. Natal homing and connectivity in Atlantic bluefin tuna populations. *Science* **2008**, *322*, 742–744.
- Mckenzie, C.; Godley, B. J.; Furness, R. W.; Wells, D. E. Concentrations and patterns of organochlorine contaminants in marine turtles from Mediterranean and Atlantic waters. *Mar. Environ. Res.* **1999**, *47*, 117–135.
- Stefanelli, P.; Ausili, A.; Ciuffa, G.; Colasanti, A.; Di Muccio, S.; Morlino, R. Investigation of polychlorobiphenyls and organochlorine pesticides in tissues of tuna (*Thunnus thynnus*) from the Mediterranean Sea in 1999. *Bull. Environ. Contam. Toxicol.* **2002**, *69*, 800–807.
- Loganathan, B. G.; Sajwan, K. S.; Richardson, J. P.; Chetty, C. S.; Owen, D. A. Persistent organochlorine concentrations in sediment and fish from Atlantic coastal and brackish waters off Savannah, Georgia, USA. *Mar. Pollut. Bull.* **2001**, *42*, 246–250.
- Hobbs, K. E.; Muir, D. C. G.; Mitchell, E. Temporal and biogeographic comparisons of PCBs and persistent organochlorine pollutants in the blubber of fin whales from eastern Canada in 1971–1991. *Environ. Pollut.* **2001**, *114*, 243–254.
- Deshpande, A. D.; Draxler, A. F. J.; Zdanowicz, V. S.; Schrock, M. E.; Paulson, A. J. Contaminant levels in the muscle of four species of fish important to the recreational fishery of the New York Bight Apex. *Mar. Pollut. Bull.* **2002**, *44*, 164–171.
- Turek, K. J. S.; Kucklick, J. R.; McFee, W. E.; Pugh, R. S.; Becker, P. R. Factors influencing persistent organic pollutant concentrations in the Atlantic white-sided dolphin (*Lagenorhynchus acutus*). *Environ. Toxicol. Chem.* **2005**, *24*, 1079–1087.
- Kawano, M.; Inoue, T.; Wada, T.; Hidaka, H.; Tatsukawa, R. Bioconcentration and residue patterns of chlordanes compounds in marine animals: Invertebrates, fish, mammals, and seabirds. *Environ. Sci. Technol.* **1988**, *22*, 792–797.
- Loganathan, B. G.; Kannan, K. Global Organochlorine Contamination Trends: An Overview. *Ambio* **1994**, *23*, 187–191.

- (14) Wiberg, K.; Letcher, R. J.; Sandau, C. D.; Norstrom, R. J.; Tysklind, M.; Bidleman, T. F. The enantioselective bioaccumulation of chiral chlordane and α -HCH contaminants in the polar bear food chain. *Environ. Sci. Technol.* **2000**, *34*, 2668–2674.
- (15) Fisk, A. T.; Hobson, K. A.; Norstrom, R. J. Influence of chemical and biological factors on trophic transfer of persistent organic pollutants in the northwater polynya food web. *Environ. Sci. Technol.* **2001**, *35*, 732–738.
- (16) Dearth, M. A.; Hites, R. A. Chlordane accumulation in people. *Environ. Sci. Technol.* **1991**, *25*, 1279–1285.
- (17) Fisk, A. T.; Tittlemier, S. A.; Pranschke, J. L.; Norstrom, R. J. Using anthropogenic contaminants and stable isotopes to assess the feeding ecology of Greenland shark. *Ecology* **2002**, *83*, 2162–2172.
- (18) Ueno, D.; Iwata, H.; Tanabe, S.; Ikeda, K.; Koyama, J.; Yamada, H. Specific accumulation of persistent organochlorines in bluefin tuna collected from Japanese coastal waters. *Mar. Pollut. Bull.* **2002**, *45*, 254–261.
- (19) Ashley, J. T. F.; Secor, D. H.; Zlokovitz, E.; Wales, S. Q.; Baker, J. E. Linking habitat use of Hudson River striped bass to accumulation of polychlorinated biphenyl congeners. *Environ. Sci. Technol.* **2000**, *34*, 1023–1029.
- (20) Zlokovitz, E. R.; Secor, D. H.; Piccoli, P. M. Patterns of migration in Hudson River striped bass as determined by otolith microchemistry. *Fish. Res.* **2003**, *63*, 245–259.
- (21) U. S. National Marine Fisheries Service. Size class categories of Atlantic bluefin tuna - Table; www.nmfspermits.com/other/BFTsizeclass.doc.
- (22) Corsolini, S.; Sara, G.; Borghesi, N.; Focardi, S. HCB, p,p'-DDE and PCB ontogenetic transfer and magnification in bluefin tuna (*Thunnus thynnus*) from the Mediterranean Sea. *Environ. Sci. Technol.* **2007**, *41*, 4227–4233.
- (23) Geisz, H. N.; Dickhut, R. M.; Cochran, M. A.; Fraser, W. R.; Ducklow, H. W. Melting glaciers: A probable source of DDT to the Antarctic marine ecosystem. *Environ. Sci. Technol.* **2008**, *42*, 3958–3962.
- (24) La Mesa, M.; Sinopoli, M.; Andaloro, F. Age and growth rate of juvenile bluefin tuna *Thunnus thynnus* from the Mediterranean Sea (Sicily, Italy). *Sci. Mar.* **2005**, *69*, 241–249.
- (25) El Tawil, M.; El Kabir, N.; Ortiz de Urbina, J. M.; Valeiras, J.; Abad, E. Length-weight relationships for bluefin tuna (*Thunnus thynnus* L.) caught from the Libyan trap fishery in 1999–2002. *Col. Vol. Sci. Pap. ICCAT* **2004**, *56*, 1192–1195.
- (26) Rooker, J. R.; Alvarado Bremer, J. R.; Block, B. A.; Dewar, H.; De Metrio, G.; Corriero, A.; Kraus, R. T.; Prince, E. D.; Rodríguez-Marín, E.; Secor, D. H. Life history and stock structure of Atlantic bluefin tuna (*Thunnus thynnus*). *Rev. Fish. Sci.* **2007**, *15*, 265–310.
- (27) Nimi, A. J.; Oliver, B. G. Biological half lives of polychlorinated biphenyl congeners (PCB) in whole fish and muscle of rainbow trout (*Salmo gairdneri*). *Can. J. Fish. Aquat. Sci.* **1983**, *40*, 1388–1394.
- (28) Boon, J. P.; van der Meer, J.; Allchin, C. R.; Law, R. J.; Klungsøyr, J.; Leonards, P. E. G.; Spliid, H.; Storr-Hansen, E.; Mckenzie, C.; Wells, D. E. Concentration-Dependent Changes of PCB Patterns in Fish-Eating Mammals: Structural Evidence for Induction of Cytochrome P450. *Arch. Environ. Contam. Toxicol.* **1997**, *33*, 298–311.
- (29) van den Brink, N. W.; de Ruiter-Dijkman, E. M.; Broekhuizen, S.; Reijnders, P. J. H.; Bosveld, A. T. C. Polychlorinated biphenyls pattern analysis: Potential nondestructive biomarker in vertebrates for exposure to cytochrome P450-inducing organochlorines. *Environ. Toxicol. Chem.* **2000**, *19*, 575–581.
- (30) Murphy, D. L.; Gooch, J. W. Accumulation of *cis* and *trans* chlordane by channel catfish during dietary exposure. *Arch. Environ. Contam. Toxicol.* **1995**, *29*, 297–301.
- (31) Pereira, W. E.; Hostettler, F. D.; Cashman, J. R.; Nishioka, R. S. Occurrence and distribution of organochlorine compounds in sediment and livers of striped bass (*Morone saxatilis*) from the San Francisco bay-delta estuary. *Mar. Pollut. Bull.* **1994**, *28*, 434–441.
- (32) Falandysz, J.; Strandberg, L.; Puzyn, T.; Gucia, M.; Rappe, C. Chlorinated cyclodiene pesticide residues in blue mussel, crab, and fish in the Gulf of Gdańsk, Baltic Sea. *Environ. Sci. Technol.* **2001**, *35*, 4163–4169.
- (33) Graves, J. E.; Gold, J. R.; Ely, B.; Quattro, J. M.; Woodley, C.; Dean, J. M. Population genetic structure of bluefin tuna in the North Atlantic Ocean. I. Identification of variable genetic markers. *ICCAT, Col. Vol. Sci. Papers* **1996**, *45*, 155–157.
- (34) Bidleman, T. A.; Falconer, R. L. Enantiomer ratios for apportioning two sources of chiral compounds. *Environ. Sci. Technol.* **1999**, *33*, 2299–2301.
- (35) Dickhut, R. M.; Canuel, E. A.; Gustafson, K. E.; Liu, K.; Arzayus, K. M.; Walker, S. E.; Edgecombe, G.; Gaylor, M. O.; MacDonald, E. H. Automotive sources of carcinogenic polycyclic aromatic hydrocarbons associated with particulate matter in the Chesapeake Bay region. *Environ. Sci. Technol.* **2000**, *34*, 4635–4640.
- (36) Itoh, T.; Tsuji, S.; Nitta, A. Migration patterns of young Pacific bluefin tuna (*Thunnus orientalis*) determined with archival tags. *Fish. Bull.* **2003**, *101*, 514–534.
- (37) Di Natale, A.; Mangano, A.; Piccinetti, C.; Ciavaglia, E.; Celona, A. Bluefin tuna (*Thunnus thynnus* L.) line fisheries in the Italian Seas. Old and recent data. *Col. Vol. Sci. Pap. ICCAT* **2005**, *58* (4), 1285–1295.

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