# **Chapter 15 Polycyclic Aromatic Hydrocarbon Baselines in Gulf of Mexico Fishes**



# **Erin L. Pulster, Adolfo Gracia, Susan M. Snyder, Isabel C. Romero, Brigid Carr, Gerardo Toro-Farmer, and Steven A. Murawski**

**Abstract** The lack of baseline data has hindered the assessment of impacts from large-scale oil spills throughout their history. Baseline data collected before an adverse event such as an oil spill are critical for quantifying impacts and understanding recovery rates to pre-spill levels. In the case of the two largest oil spills in the Gulf of Mexico (GoM), *Deepwater Horizon* and Ixtoc 1, the lack of comprehensive contaminant baselines limits our ability to project when the ecosystem will return to pre-spill conditions and assess the short- and long-term impacts of contamination on ecosystems. Beginning in 2011, we initiated comprehensive sampling in the GoM to develop broad-scale and Gulf-wide hydrocarbon contaminant baselines primarily targeting continental shelf fishes in the USA, Mexico, and Cuba. We also developed a time series of collections over 7 years from the region in which DWH occurred. In the event there is another oil spill in the GoM, the samples from these baselines will provide broad-scale but not installation-specific baseline information for the assessment of impact and recovery. This chapter provides a summary of historical sampling and current baseline data for pelagic, mesopelagic, and demersal fish in the GoM. Further, we outline the importance of ongoing and more specific collection of monitoring data for hydrocarbon pollution.

**Keywords** PAHs · Polycyclic aromatic hydrocarbons · Gulf of Mexico · Ixtoc 1 · *Deepwater Horizon* · Oil spill baselines

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E. L. Pulster  $(\boxtimes) \cdot S$ . M. Snyder  $\cdot$  I. C. Romero  $\cdot$  B. Carr  $\cdot$  S. A. Murawski University of South Florida, College of Marine Science, St. Petersburg, FL, USA e-mail: [epulster@mail.usf.edu](mailto:epulster@mail.usf.edu)[; ssnyder4@mail.usf.edu](mailto:ssnyder4@mail.usf.edu); [isabelromero@mail.usf.edu](mailto:isabelromero@mail.usf.edu)[;](mailto:becarr@mail.usf.edu) [becarr@mail.usf.edu](mailto:becarr@mail.usf.edu)[; smurawski@usf.edu](mailto:smurawski@usf.edu)

A. Gracia

Universidad Nacional Autónoma de México, Instituto de Ciencias del Mar y Limnología, Ciudad de México, CDMX, Mexico e-mail: [gracia@unam.mx](mailto:gracia@unam.mx)

G. Toro-Farmer New College of Florida, Sarasota, FL, USA

University of South Florida, College of Marine Science, St. Petersburg, FL, USA e-mail: [gtoro-farmer@ncf.edu](mailto:gtoro-farmer@ncf.edu)

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# **15.1 Introduction**

Assessments evaluating the extent, duration of elevated concentrations, and impacts of oil spills are heavily reliant on data collected prior to, during, and after such events. A chronological sedimentary history of oil deposition can be recorded in cores (e.g., dated through radioisotopes or other means), assuming oil is deposited proportionally to the quantity released, bioturbation does not disturb the chronological order, and bacterial degradation is relatively slow. However, for living resources, depending upon how long after the event sampling occurs, contaminants within the animals may be reduced over time (via decontamination) and transformed into other compounds (via metabolism), some of which may be more toxic than the original exposure. For long-duration, post-spill monitoring assessments (e.g., several decades post-oil spill), few animals will record such events owing to their life spans. The most direct way to assess the effects and impacts of such spills is to have obtained pre-spill baselines beforehand with which to compare contamination levels and toxic biomarkers post-spill. Assessments of the impacts of the subsurface well blowout of the Mexican oil well Ixtoc 1 in 1979 were inconclusive due in part to the lack of pre-spill contaminant data (Amezcua-Linares et al. [2015](#page-14-0)). Ten years later, pre-spill data was dubbed one of the "rarest of all commodities" for researchers following the 1989 *Exxon Valdez* oil spill (Shigenaka [2014](#page-17-0)). Furthermore, 21 years subsequent to EVOS, researchers assessing the *Deepwater Horizon* oil spill (DWH, NAS [2013](#page-16-0)) lamented the virtual lack of relevant comprehensive pre-spill baselines, especially for living resources (Murawski and Hogarth [2013;](#page-16-1) Murawski et al. [2014](#page-16-2)). For decades, the unavailability of baseline data and reference sites in the Gulf of Mexico (GoM) has hindered the evaluation and health assessments of both the biota and the environment (Kennicutt et al. [1988](#page-15-0); Lewis et al. [2002;](#page-16-3) Ward and Tunnell Jr. [2017\)](#page-17-1). This chapter summarizes the availability and adequacy of PAH contamination data from fish tissues collected before and after the two largest spills in the GoM, the DWH and Ixtoc 1.

# **15.2 Pre-***DWH* **Polycyclic Aromatic Hydrocarbon Baselines in Fish**

In the USA, all 50 US states, the District of Columbia, the US territories of American Samos and Guam, and 5 Native American tribes issue fish consumption guidelines for potential health risks from consuming contaminated fish caught in their waters (US EPA [2009\)](#page-17-2). Only five bioaccumulative chemicals (mercury, PCBs, chlordane, dioxins, and DDT) constitute 97% of all advisories in effect. Additionally, all five states bordering the GoM have coastal fish consumption guidelines, yet given the amount of oil and gas activity in the GoM, there are no active advisories or closures for petroleum-derived compounds. Fishery closures were in place during and just after the DWH accident, but most closures were lifted following seafood safety surveillance testing (Ylitalo et al. [2012](#page-17-3)). Importantly, while most fish tested postspill were below levels of concern (LOC), there were little data with which to judge if contamination levels had risen beyond pre-spill levels after the spill or to assess species most at risk.

Several programs have collected PAH contaminant data prior to DWH either as part of surveillance programs or in response to specific events. The National Oceanographic and Atmospheric Administration's (NOAA) National Status and Trends (NS&T) program undertakes nationwide sampling (i.e., Mussel Watch) to describe the current status and trends in the environmental quality of our estuarine and near-coastal waters (Farrington et al. [2016\)](#page-15-1). Sampling included biliary PAH data for six inshore fish species (hardhead catfish, *Ariopsis felis*; Atlantic croaker, *Micropogonias undulatus*; red drum, *Sciaenops ocellatus*; spot, *Leiostomus xanthurus*; black drum, *Pogonias cromis*; and sand seatrout, *Cynoscion arenarius*) collected from 1985 to 1991 from along the US Gulf coast. Total biliary PAHs ranged from 7,000 to 1,000,000 ng FACs/g bile for both low- and high-molecular-weight fluorescent aromatic compounds (FACs; NCCOS [2017\)](#page-16-4). The highest concentrations were found in Atlantic croaker collected in 1989 from Barataria Bay, Louisiana. In general, the samples collected across all years from the northwest region of the GoM, along the Texas coast, had significantly higher biliary PAHs  $(300,000 \pm 240,000 \text{ ng/g}$  bile) than those collected in the north central region  $(p = 0.0046;$  Louisiana, Mississippi, Alabama:  $160,000 \pm 220,000$  ng/g bile) and west Florida ( $p = 0.0054$ ; 94,000 ± 100,000 ng/g bile).

In a separate study, hardhead catfish, Gulf killifish (*Fundulus grandis*), longnose killifish (*F. majalis*), and red drum were collected during 1990–1991 from Tampa and Sarasota bays to compare levels of organic contaminants between industrialized (Tampa Bay) and nonindustrialized areas (Sarasota Bay) (McCain et al. [1996\)](#page-16-5). Mean biliary naphthalene equivalents ranged from 11,000 to 120,000 ng FAC/mg of biliary protein and 6,800 to 150,000 ng FAC/mg of biliary protein in the Tampa Bay and Sarasota Bay, respectively. Mean concentrations of the biliary naphthalene equivalents in hardhead catfish collected in Tampa Bay were all significantly higher than those collected from within Sarasota Bay. In addition, the biliary PAHs measured in fish collected within the more industrialized areas of Tampa Bay (i.e., Hillsborough Bay) were significantly higher than nearby nonindustrialized sites.

Edible muscle samples from hardhead catfish, channel catfish (*Ictalurus punctatus*), and largemouth bass (*Micropterus salmoides*) were surveyed in 1996 to compare the tissue quality between fish collected from wastewater-impacted areas and reference locations in northwestern Florida and southwestern Alabama (Lewis et al. [2002\)](#page-16-3). Average muscle concentrations of PAHs ranged from 0.6 to 3.5 ng/g wet weight (w.w.). The results from this study indicated that total PAH concentrations averaged across fish collected from the wastewater-impacted areas ( $2.2 \pm 3.3$  ng/g w.w.) were similar to those collected at the reference sites  $(1.8 \pm 1.6 \text{ ng/g w.w.})$ . A regional assessment of whole-body concentrations in fish and shellfish across the west, northeast, southeast, and Gulf coasts were analyzed for chemical contaminants to evaluate compliance with the EPA fish meal recommended guidelines (Harvey et al. [2008\)](#page-15-2). In general, PCBs accounted for the greatest percentage (31%) of exceedances across all regions, followed by mercury (29%), PAHs (21%), and total DDTs (11%). In the Gulf region, mercury accounted for the highest percentage of exceedances (19%), followed by PCBs (11%), total DDTs (7%), PAHs (6%), toxaphene (2%), and cadmium (1%).

In the early 1990s, a study supported by the Minerals Management Service (MMS) evaluated biliary PAHs at several offshore gas rigs in the western GoM (McDonald et al. [1996\)](#page-16-6). This study evaluated the impacts of oil contamination from specific rigs by evaluating fish collected near the rigs and those collected from several kilometers away. Biliary PAHs were evaluated for a variety of bottom-dwelling fishes, generally showing weak negative correlation with distances from the rigs (McDonald et al. [1996](#page-16-6)).

In the aftermath of Hurricane Katrina in 2005, Hom et al. [\(2008](#page-15-3)) evaluated the safety of seafood primarily in Mississippi Sound and offshore waters, examining Atlantic croaker, shrimps, and a few other species. No samples were found to exceed LOCs despite the strong storm surge from that hurricane.

Mesopelagic species were collected in 2007 over the slope of the north central GoM near cold-seep habitats to characterize species composition as well as distributions and determine PAH content in muscle tissues (Ross et al. [2010](#page-17-4); Romero et al. [2018\)](#page-17-5). Because of their proximity in time and space, PAH determinations from this dataset represent a serendipitous PAH baseline for DWH-based contamination of mesopelagic fishes in the northern GoM. Pre-spill PAH values ranged from 24 to 555 ng/g w.w., containing a few PAH compounds (mostly naphthalene). These values are relatively high compared to other more shallow-water communities, this trend of organic pollutants accumulating in the deep ocean (Froescheis et al. [2000;](#page-15-4) Koenig et al. [2013](#page-15-5)) is a pattern potentially explained by the natural seeps in the study area and likely due to a combination of biological factors intrinsic to mesopelagic communities (e.g., vertical migration, PAH metabolism).

#### **15.3 Post-DWH and Ixtoc 1 Baselines in Fish**

# *15.3.1 Seafood Safety*

With the release of more than 200 million gallons of crude oil from DWH, there were concerns regarding the integrity and safety of the seafood supply for human consumption, resulting in intensive testing, federal and state fishery closures, risk assessments, and the inclusion of petroleum-related compounds in the 2011 National Listing of Fish Advisories (Ylitalo et al. [2012](#page-17-3); US EPA [2013](#page-17-6)). Between April 28, 2010, and March 31, 2011, more than 8,000 seafood samples were collected and analyzed from within and around closure areas across the GoM for PAHs and the dispersant component dioctyl sodium sulfosuccinate (DOSS). Primarily edible tissues from fish (snappers, groupers, porgies, tuna, etc.) and shellfish contained low concentrations of PAH (<1 ng/g w.w.) and DOSS (0.05–0.29  $\mu$ g/g w.w.) with both at least two orders of magnitude lower than the LOCs for human health risk (Ylitalo et al. [2012](#page-17-3)).

An independent assessment of seafood safety evaluated fishes obtained from within fishing closures along the Mississippi coast (Xia et al. [2012](#page-17-7)). Samples were collected and analyzed weekly from May 27, 2010, until October 2010 and then monthly until August 2011. Eleven fish species consisting of Atlantic croaker, black drum, red drum, cobia (*Rachycentron canadum*), Gulf menhaden (*Brevoortia patronus*), red snapper (*Lutjanus campechanus*), southern flounder (*Paralichthys lethostigma*), spotted seatrout (*Cynoscion nebulosus*), striped mullet (*Mugil cephalus*), tripletail (*Lobotes surinamensis*), and sand seatrout were analyzed for PAHs*.* Edible muscle composites were analyzed for 25 PAHs for each species. Overall, all PAHs were at least three orders of magnitude below the LOCs established by NOAA, the FDA, and the Gulf states. LOCs ranged from 35 ng/g for benzo[a] pyrene and dibenzo[a,h]anthracene to  $490,000$  ng/g w.w. for anthracene and phenanthrene. Maximum concentrations detected in fillets (muscle) ranged from nondetect to 20.1 ng/g for benzo[a]pyrene and anthracene/phenanthrene, respectively. The levels of total PAHs (average,  $16 \text{ ng/g w.w.}$ ) were consistent until January 2011, and thereafter concentrations significantly declined ~44% to an average of 9 ng/g (w.w.).

#### *15.3.2 Hepatobiliary and Extrahepatic PAH Levels in Fish*

The most comprehensive Gulf-wide survey of the health of GoM fishes conducted to date is that by the Center for Integrated Modeling and Analysis of Gulf Ecosystems (C-IMAGE) wherein ~15,000 fishes were evaluated for disease frequencies and a subset sampled for PAH levels. The C-IMAGE sampling campaign focused on reef and deep-dwelling shelf species (e.g., snappers, tilefishes, groupers) collected from 2011 to 2017 (Fig. [15.1](#page-5-0); Murawski et al. [2014](#page-16-2), [2018](#page-16-7)). To date, over 1800 bile samples from 75 offshore species of finfish and sharks have been analyzed for fluorescent aromatic compounds (FACs) in fish bile (Snyder et al., [2015;](#page-17-8) Pulster et al. [2017a](#page-16-8)). These samples were collected between 2011 and 2017 from coastal regions from southwest Florida to the Yucatán Peninsula and Cuba. Preliminary biliary PAHs ranged from <1 to 1,900,000 ng/g bile for naphthalene and benzo[a]pyrene FACs (Pulster et al. [2017a\)](#page-16-8). This represents a 90% increase in the maximum total biliary PAHs compared to those measured by the earlier studies conducted in the northern GoM between 1985 and 1991 (Pulster et al. [2017a;](#page-16-8) NCCOS [2017\)](#page-16-4). Overall for the years 2011–2016, the north central region (39–1,900,000 ng FACs/g bile;  $125,000 \pm 190,000$  ng FAC/g bile) of the GoM remained significantly higher in average total biliary FACs than the southwest region (<1–960,000 ng FAC/g; 73,000  $\pm$  97,000 ng FAC/g bile;  $p < 0.0001$ ), the west Florida shelf (1,500– 360,000 ng FAC/g; 37,000 ± 59,000 ng FAC/g bile; *p* < 0.0001), and the northwest region (130–370,000 ng FAC/g; 90,000 ± 79,000 ng FAC/g bile; Fig. [15.2](#page-5-0); Pulster et al. [2017a](#page-16-8)). The 2011–2016 preliminary biliary naphthalene and benzo[a]pyrene concentrations ranged from 410 to 1,900,000 ng/g and 0.073 to 9,600 ng/g, respectively (Fig. [15.2](#page-5-1)). The highest concentrations of biliary naphthalene and benzo[a]

<span id="page-5-0"></span>

**Fig. 15.1** Standardized catch rates (catch in numbers per 1000 hook-hours) of fishes sampled with demersal longlines, 2011–2017 (Murawski et al. [2018](#page-16-7))

<span id="page-5-1"></span>

**Fig. 15.2** Mean 2011–2016 biliary naphthalene (Nap ng/g) and benzo[a]pyrene (BaP ng/g) in fish collected in the Gulf of Mexico by the C-IMAGE Consortium (Pulster et al. [2017a](#page-16-8)). Black dots indicate station locations where fish were collected

pyrene were found in the north central region of the GoM (where DWH occurred). Additional "hot spots" of elevated levels of biliary FACs occurred near major shipping ports (i.e., Brownsville, Corpus Christi, Louisiana, Tampa), vessel shipping lanes, and rivers (i.e., Rio Grande, Mississippi, Coatzacoalcos).

Following the 2010 DWH oil spill, skin lesion frequencies and biliary FACs declined in a number of species collected from the north central region of the GoM (where DWH occurred), and this decline provides compelling evidence of the acute and episodic nature of PAH exposures from the DWH oil spill (Murawski et al. [2014;](#page-16-2) Snyder et al. [2015](#page-17-8); Pulster et al. [2017a,](#page-16-8) [2020\)](#page-17-9) and the impacts such exposures may have on fish health. Lesion frequencies in red snapper (*Lutjanus campechanus*) declined 53% between 2011 and 2012 (Murawski et al. [2014](#page-16-2)). In addition, 12 species that were repeatedly collected in the north central region of the GoM also demonstrated declines over time in total biliary PAHs in the years following DWH (2011– 2015, Fig. [15.3](#page-6-0), Pulster et al. [2017a\)](#page-16-8). Total biliary PAHs declined by 15% in little gulpers (*Centrophorus uyato*) (2014–2015) to 90% in yellow conger eels (*Rhynchoconger flavus*) (2011–2015). As of 2015, 5 years after DWH, biliary PAH levels had continued to decline suggesting that levels were not yet reduced back to baseline or pre-spill levels.

Golden tilefish (*Lopholatilus chamaeleonticeps*) and a number of other species, including Gulf hake (*Urophycis cirrata*), snowy grouper (*Epinephelus niveatus*), yellowedge grouper (*Hyporthodus flavolimbatus*), red snapper, and red grouper (*Epinephelus morio*), all exhibited an increase in biliary PAHs between 2012/2013 and 2014 (Struch et al. unpublished; Pulster et al. [2017a](#page-16-8), [2018](#page-16-9); Snyder et al. [2015\)](#page-17-8). This pattern of increasing PAH levels during the same time period (2012–2014) has also been observed in common loons, seaside sparrows, and sediments (Perez-Umphrey et al. [2018;](#page-16-10) Paruk et al. [2016](#page-16-11); Turner et al. [2014](#page-17-10)). In Gulf menhaden, body burdens of PAH concentrations decreased, while benzo[a]pyrene toxic equivalents increased between 2012 and 2013 suggesting resuspension of oil residues rather than a new source (Olson et al. [2016\)](#page-16-12). Changes in redox-sensitive elements,

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**Fig. 15.3** Trends (2011–2015) in preliminary mean biliary PAHs (ng FACs/g bile) for multiple fish species collected from the north central Gulf of Mexico. Percent declines in biliary PAHs were calculated between the first and last year sampled for each species. Mean biliary PAHs are the sum of the naphthalene and benzo[a]pyrene fluorescent aromatic compounds (FACs). Error bars are  $\pm 1$ standard error of the mean

radioisotope analyses, and in situ data from the GoM also provide supporting evidence for bioturbation, sedimentation pulses, and resuspension events during a similar time frame (Brooks et al. [2015](#page-14-1); Hastings et al. [2016;](#page-15-6) Diercks et al. [2018\)](#page-15-7). Resuspension events can be caused by a number of physical processes such as variation in subsurface currents, surface and internal waves, and storm-driven surges. An episodic large-scale resuspension event as the result of Hurricane Isaac in 2012 was identified from current meter data and sediment records in the northern GoM (Diercks et al. [2018\)](#page-15-7). This is not surprising considering the GoM experiences frequent tropical storms, for instance, Tropical Storm Debby and Hurricane Isaac in 2012 and Tropical Storm Andrea and Tropical Storm Karen in 2013. The resuspension of sediments reintroduces contaminants into the water column (e.g., freely dissolved or sediment bound), thereby increasing bioavailability to biota. Suspended particles with associated sediment-bound contaminants, including PAHs, enhance the uptake rates and bioaccumulation in fish and other organisms (Zhai et al. [2018;](#page-18-0) Peterson et al. [1996;](#page-16-13) Leppänen and Kukkonen [1998](#page-15-8); Menon and Menon [1999;](#page-16-14) Zhang et al. [2015\)](#page-18-1).

Post-DWH, tissues from a number of fish species were also analyzed for PAHs from multiple regions within the GoM as part of the C-IMAGE Gulf-wide comprehensive fish survey (Murawski et al. [2014;](#page-16-2) Snyder et al. [2015,](#page-17-8) [2017;](#page-17-11) Carr et al. [2018;](#page-15-9) Pulster et al. [2018](#page-16-9); Murawski et al. [2018](#page-16-7)). In 2011, concentrations of muscle and liver PAHs in red snapper collected in the north central region of the GoM were low (<35 ng/g w.w.) compared to the relatively high concentrations of PAH equivalents measured in the bile (41,000–470,000 ng FAC/g bile, Murawski et al. [2014\)](#page-16-2). Despite the ability of fishes to quickly metabolize most PAHs, the PAH composition profiles in the liver samples had a significantly strong positive relationship ( $r^2$  = 0.82,  $p < 0.001$ ) with the profiles of the crude oil from the DWH wellhead (Fig. [15.4](#page-8-0), Murawski et al. [2014\)](#page-16-2).

Muscle tissues collected in 2011 from mesopelagic fish species in the north central region of the GoM contained PAH levels (330–2,350 ng/g w.w.; Romero et al. [2018\)](#page-17-5) more than an order of magnitude higher than other species during the same time period (Murawski et al. [2014;](#page-16-2) Romero et al. [2018](#page-17-5); Ylitalo et al. [2012;](#page-17-3) Xia et al. [2012\)](#page-17-7). Similar to other species, the muscle concentrations in mesopelagic fish demonstrated significant increases between pre- and post-DWH levels, followed by a significant decline by 2015 (Romero et al. [2018](#page-17-5)). Romero et al. suggest the higher tissue concentrations in mesopelagic fish than shallower water species may be due to the increased frequency and duration of exposure periods due to their vertical migratory behaviors throughout the water column (Romero et al. [2018\)](#page-17-5). However, in addition to species-specific differences in metabolic capacity, a number of additional critical environmental and physiochemical factors may partially explain the increased concentrations observed in mesopelagic fish. First, the fish in this study were collected (50–1,000 m) within similar depths of the continuous subsurface plume of oil residues (~1,000–1,200 m) that was identified post-DWH (Camilli et al. [2010\)](#page-14-2). Although there is evidence that chemical cues can elicit habitat avoidance in fish (Pulster et al. [2020](#page-17-9)), the potential for increased and extended periods of exposure in 2010 exists due to the subsurface plume which persisted for

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**Fig. 15.4** Concentrations of some PAH parent compounds and alkylated homologues sampled from the Macondo (*Deepwater Horizon*) wellhead (Reddy et al. [2012](#page-17-12); gray bars) and 2011 composite samples of red snapper livers collected from the Gulf of Mexico (red bars). (Reprinted with permission from Murawski et al. [2014](#page-16-2))

months and continued water column contamination persisted in 2010, likely due to resuspended sediments. Furthermore, water temperatures decrease significantly with depth and are  $15-20^{\circ}$  lower at 1,000 m than at the surface, increasing the persistence and bioavailability of contaminants at deep pelagic depths. Specifically

to PAHs, low water temperatures promote their persistence in water due to lower volatilization and biodegradation which consequently decreases elimination rates further resulting in the retention of PAHs in fish tissues (Varanasi et al. [1981;](#page-17-13) Egaas and Varanasi [1982](#page-15-10); Varanasi [1989](#page-17-14)). For example, starry flounders (*Platichthys stellatus*) held at temperatures (4 °C) similar to the temperatures at the depth of DWH in the GoM were found to have 26–34 times higher naphthalene concentrations than those held at 12 °C (Varanasi et al. [1981](#page-17-13)). The preliminary total PAHs in liver tissues collected ranged from 7.7 to 407 ng/g w.w. for hakes (*Urophycis* sp., 2012– 2015; Struch et al. unpublished), 1200 to 195,000 ng/g w.w. for golden tilefish (2012–2016; Snyder et al. [2017,](#page-17-11) [2018\)](#page-17-15), and 67.6 to 17,300 ng/g w.w. for groupers (*Epinephelus* sp., 2012–2016; Pulster et al. [2018\)](#page-16-9). While there were a few species and site differences, in general the groupers collected within the north central region  $(2.350 \pm 2.800 \text{ ng/g w.w.})$  had similar levels of total PAHs to those collected in the northwest region (1580  $\pm$  2470 ng/g w.w.) yet significantly higher than those collected in the southwest region (983  $\pm$  2,260 ng/g w.w.;  $p < 0.0001$ ). In contrast to biliary PAHs, there appears to be an increasing trend over time in the liver concentrations of PAHs for a number of species. For instance, between 2012 and 2015, liver concentrations in yellowedge groupers from the north central region increased to  $362\%$  ( $p = 0.036$ ) (Fig. [15.5\)](#page-9-0). Similar trends were also observed in the southwest region for both snowy (237% increase) and yellowedge groupers (177% increase) from 2015 to 2016. In addition, PAH levels increased to 102% in hake livers between 2012 and 2015.

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**Fig. 15.5** Trends in preliminary mean liver concentrations of PAHs (ng/g wet weight) in grouper species collected from the southwest (blue bars), north central (red bars), and northwest (green bars) regions of the Gulf of Mexico (2011–2016). Percent increases in liver PAHs were calculated between the first and last year sampled for each species. The ∑PAHs is the sum of 46 parental PAHs and their alkylated homologues. Error bars are  $\pm 1$  standard error of the mean

Preliminary concentrations of total PAHs in liver tissues from golden tilefish sampled between 2015 and 2016 were similar among regions but significantly higher in the northwest region offshore of Texas (31,000  $\pm$  25,000 ng/g w.w.) than the southwest  $(21,000 \pm 28,000 \text{ ng/g w.w.})$  and north central  $(16,000 \pm 11,000 \text{ ng/g}$ w.w.) regions (Snyder et al. [2017](#page-17-11), [2018\)](#page-17-15). The opposite pattern occurred for biliary PAHs. Biliary PAHs are higher in the north central region  $(470,000 \pm 400,000 \text{ ng/g})$ than the southwest  $(170,000 \pm 140,000 \text{ ng/g})$  and northwest regions  $(230,000 \pm 88,000 \text{ ng/g})$ . These results suggest the highest recent exposure to PAHs in the north central region, perhaps more efficient metabolism and elimination, and higher accumulation in the liver tissue in the southwest and northwest regions. As mentioned above, many species, including golden tilefish from the north central GoM, demonstrated increased biliary naphthalene metabolite concentrations between 2012 and 2015, possibly related to the resuspension of sedimented oil as discussed above. In contrast, total liver PAHs have remained relatively constant for golden tilefish sampled in 2012–2015.

In the southwest region, hardhead catfish collected in 2008 from along the Tabasco coast in Mexico had liver concentrations of PAHs ranging from 7,340 to 119,000 ng/g w.w., 1 year after the Kab-121 oil spill and 28 years post-Ixtoc 1 (Gold-Bouchot et al. [2014\)](#page-15-11). Liver concentrations significantly declined to 1390–9470 ng/g w.w. within 3 months of the first sampling event. In 2010, Gracia [\(2010\)](#page-15-12) analyzed the muscle content from 29 fish species collected at 30 sampling locations in the southwestern GoM near Ixtoc I spill area (off Veracruz, Tabasco, and Campeche states, Gracia [2010\)](#page-15-12). The most frequent fish species sampled included hardhead catfish, Mexican flounder, sheepshead (*Archosargus probatocephalus*), gray triggerfish (*Balistes capriscus*), Atlantic croaker, and southern kingfish (*Menticirrhus americanus*). The highest value of PAH muscle concentrations (474 ng/g d.w.) was found in goliath grouper (*Epinephelus itajara*); however, relatively high concentrations of PAHs (100–474 ng/g d.w.) were also found in spotted weakfish (*Cynoscion nebulosus*), sand seatrout, sheepshead (*Archosargus probatocephalus*), and goliath grouper which were mainly found in coastal areas (Fig. [15.6](#page-11-0); Gracia [2010](#page-15-12)). Anthracene (99 ng/g d.w.), fluoranthene (100 ng/g d.w.), and phenanthrene (98 ng/g d.w.) were the dominant PAHs in tissue content.

Fish surveys conducted in 2015 and 2016 by C-IMAGE in the southern GoM evaluated muscle tissues for PAH concentrations in 107 organisms from 16 species. The muscle concentrations of PAHs in red snapper and yellowedge grouper ranged from 10 to 493 ng/g d.w. (Gracia et al. [2018\)](#page-15-13). Red snapper (59–83.4 ng/g d.w.) had higher mean muscle PAH concentrations than yellowedge grouper (53–37 ng/g d.w.), although not significantly so (Fig. [15.7](#page-12-0)). Muscle concentrations of PAHs in red snapper collected within the exclusion zone near where the Ixtoc 1 wellhead was located were relatively high; however, the highest levels were located off the Yucatán Peninsula where there are no known oil rigs or natural oil seeps (Fig. [15.8\)](#page-13-0). The high levels of PAHs observed in multiple species in this location may be the result of proximity to major shipping channels and strong current systems (Snyder et al. [2017;](#page-17-11) Gracia et al. [2018](#page-15-13)). Yellowedge grouper showed high PAH muscle

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**Fig. 15.6** Mean concentrations of PAHs ( $\mu$ g/g d.w.) in muscle tissues from fish (29 species) collected in 2010 from the southern Gulf of Mexico near oil infrastructure within the Ixtoc 1 zone

values in three main areas; however, similar to the red snapper, the concentrations off the Yucatán Peninsula were higher than those samples near the exclusion zone (Fig. [15.9;](#page-14-3) Gracia et al. [2018\)](#page-15-13). However, the mean PAH muscle concentration of both species was very low with respect to US LOCs.

# **15.4 Conclusions**

A complex interaction of physiochemical properties (e.g., molecular weight and octanol-water partition coefficients), species-specific life histories (e.g., habitat, diet), metabolic capacities, and environmental factors (e.g., temperature, salinity) regulates the chemical bioavailability, uptake, metabolic rates, and excretion rates of oil contaminant dynamics in fishes. Although fish have a relatively high capacity to rapidly metabolize PAHs, there is a potential for increased toxicity and bioaccumulation during the metabolic process as PAHs are hydrolyzed, reabsorbed, and returned through circulation (Lech and Vodicnik [1985;](#page-15-14) Kleinow et al. [1987;](#page-15-15) Varanasi et al. [1989\)](#page-17-16).

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**Fig. 15.7** Muscle concentrations of PAHs (μg/kg d.w.) measured in red snapper and yellowedge grouper collected in the southwestern Gulf of Mexico, 2015–2016

Studies have demonstrated that exposure routes have little effect on the biotransformation products of some compounds and the distribution of PAHs within tissues (Stein et al. [1984](#page-17-17); Varanasi [1989;](#page-17-14) Pulster et al. [2017b](#page-16-15)). Rather, differences observed between muscle and liver concentrations are more likely due to chronic exposures and accumulation rates that are faster than the fishes' ability to metabolize and excrete xenobiotics. Importantly, chronically exposed fish have slower elimination and increased PAH levels than those exposed acutely (Varanasi [1989](#page-17-14)). This is evident in the increasing trend in baseline levels of biliary and tissue concentrations of PAHs in a number of fish species between pre-2000 and post-DWH data in the GoM that raises concern for the overall health of the Gulf ecosystem and its fish populations. The increasing oil production (especially in the northern GoM) and the higher fraction of oil derived from ultra-deep waters (>1500 m) mean that fish communities not traditionally exposed to oil contamination from oil extraction activities might now be at a higher risk (Romero et al. [2018\)](#page-17-5). The increasing trend in oil accumulation in deepwater fishes and expansion of drilling into deeper waters warrants increased monitoring and fish health assessments and intensifies the need for more extensive and comprehensive offshore baseline data, since the majority of all baseline studies in the GoM prior to the DWH were for nearshore and shelf fishes.

Given the importance of the GoM to energy production in both the USA and Mexico (Murawski et al. [2020](#page-16-16)), the lack of coordinated, comprehensive, and

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**Fig. 15.8** Muscle concentrations of PAHs (μg/kg d.w.) in red snapper collected in the southwestern Gulf of Mexico, 2015–2016

continuous monitoring of oil-related pollution in sediments, fishes, and waters from these facilities is remarkable. Other than the post-DWH assessment activities, primarily supported by private foundation funding, there is no specific program in the USA to understand and assess pollution related to oil production and transportation activities. The advantages of developing such a program are numerous. Assessing the impacts of pollution from a specific infrastructure failure is the obvious primary advantage, but such data can also identify the interactions between pollution from natural sources (e.g., cold seeps) in relation to the entirety of the marine oil budget.

Pre-Ixtoc 1 data from Mexican waters are virtually nonexistent, and there is at least a 10-year data gap between the pre- and post-DWH data for PAH concentrations in fish from US waters. Given that the GoM is a highly dynamic system with numerous and expanding sources of anthropogenically derived pollution, collection of baseline data at shorter time intervals is critical for assessing and evaluating sources of pollution. Baseline data and long-term trends are essential to monitor the health of the GoM, changing environmental conditions, and the interactions of natural systems with anthropogenic activities. In the context of a future oil spill and changing environmental conditions in the GoM, continued monitoring is required for the assessment of impact, resilience, and recovery from future events.

<span id="page-14-3"></span>

**Fig. 15.9** Distributions of muscle concentrations of PAHs (μg/kg d.w.) in yellowedge grouper collected in the southwestern Gulf of Mexico, 2015–2016

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