

Probabilistic Risk Assessment for High-End Consumers of Seafood on the Northeastern Gulf Coast

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Abstract

The Deepwater Horizon oil spill (April 20, 2010) caused concern regarding Gulf seafood safety. Communities were skeptical of governmental risk assessments because they did not take into account the higher consumption of seafood along coastal areas. The objective of this study was to perform a probabilistic risk assessment based on the consumption rates of high-end consumers of Gulf seafood. We utilized seafood consumption data from five communities across the northeastern Gulf of Mexico. This study collected finfish, shrimp, blue crab, and oysters from these communities and analyzed their tissues for polynuclear aromatic hydrocarbons (PAHs). A probabilistic risk assessment was performed using population-specific seafood consumption rates and body weights for commercial fishers, recreational fishers, and a Filipino-American community. For non-cancer effects, 95th percentile hazard quotients for these targeted populations ranged between 1.84E-04 to 5.39E-03 for individual seafood types. The 95th percentile hazard indices for total seafood consumption ranged from 3.45E-03 to 8.41E-03. Based on total seafood consumption, highest hazard indices were modeled for the Filipino-American community followed by commercial and recreational fishers. Despite higher consumption rates, hazard indices for the high-end consumers targeted in this study were two to three orders of magnitude below the regulatory limit of 1.

Keywords

probabilistic risk assessment; seafood consumption; polynuclear aromatic hydrocarbons

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Introduction:

On April 20, 2010, an explosion on the Deepwater Horizon (DWH) oil drilling rig caused oil to discharge from the Macondo well head into the Gulf of Mexico (Gulf). The oil flowed continuously for three months, releasing a total of approximately 4.9 million barrels (205 million gallons) of oil (McNutt et al. 2012; USCG 2011). Deepwater Horizon is recognized as one of the largest oil spills in U.S. history. The visible effects of the oil to the water and wildlife affected the perception of seafood safety in the Gulf (Greiner et al., 2013; McKendree et al., 2013; Sathiakumar et al., 2017; Simon-Friedt et al., 2016). In an effort to relieve concern and protect public health, the U.S. Food and Drug Administration (FDA) and the National Oceanic and Atmospheric Association (NOAA) assessed the safety of commercially-harvested Gulf seafood. Efforts were made by federal and state agencies to assess the various exposures to oil spill related chemicals and their associated health risks (FDA 2010; FDOH 2010; Gohlke et al., 2011; Ylitalo et al., 2012). These assessments used national statistics to represent body weight and seafood consumption for Gulf residents. The assessments were not accepted by many residents of the coastal communities along the Gulf (Gohlke, 2011; Rotkin-Ellman et al. 2012, Wilson et al. 2015). Distrust stemmed in part from the belief that the agencies' risk assessment did not account for coastal populations who eat more fish and shellfish than populations in non-coastal areas (Mahaffey et al. 2009). The FDA rationale for using 90th percentile national consumption data reported in the National Health and Nutrition Examination Survey (NHANES) was to ensure protection for high-end consumers nationwide (Dickey and Huettel, 2016; Ylitalo et al., 2012). Additionally, the United States Environmental Protection Agency (USEPA) reported that FDA methodologies used in the risk assessment (particularly body weight and seafood consumption) were "never intended to be protective of recreational, tribal, ethnic, and subsistence fishers who typically consume larger quantities of fish than the general population and often harvest the fish and shellfish they consume from the same local water bodies" (USEPA 2000).

Along the Gulf coast, the fishing season lasts for most of the year and, in many communities, fishing is an important dietary constituent and a significant part of the culture. Historical data suggests that commercial fishers, recreational fishers, and Asian American subsets of coastal populations can be considered higher end consumers of seafood (Harris et al.,2009; Polissar et al. 2012; Sechena et al. 2003; Smith et al. 2003; Wilson et al. 2015). To address the concerns of residents in Gulf coastal communities, we focused on assessing risk for these subsets of the population. Commercial seafood production is a way of life for many Gulf coast residents (Smith et al. 2003). Although there are no studies which analyze the consumption rate of finfish of commercial fishers along the Gulf coast, commercial fishers in Chesapeake Bay were shown to consume more than twice as many fish meals as the average person in the U.S. population (Harris et al. 2009). Recreational fishers in this study are defined as persons who engage in the act of fishing for enjoyment, fun, sport as well as a food source. Recreational fishers may enjoy increased access to seafood resources and as such may consume larger amounts of fish and other seafood compared to the general population. A study of recreational fishers at the 2005 Alabama Deep Sea Fishing Rodeo in the Northern Gulf reported that these fishers consumed three to four times as much seafood

compared to national surveys of the U.S. population. Northern Gulf participants consumed an average of 55 g/d compared to the national average of 13–20 g/d (Warner 2007). Members of Asian-American communities often consume greater quantities of both finfish and shellfish than the average American (Polissar et al. 2012; Sechena et al. 2003; WDOE 2012). In a study by Wilson et al. (2015), the average shrimp consumption rate of Vietnamese shrimpers of southeast Louisiana was determined to be 45.2 g/d based on a community survey. This consumption rate is three times greater than the 13 g/d used to develop the FDA level of concern for shrimp and crab (FDA, 2010).

Because these subpopulations are likely to ingest larger quantities of seafood and/or shellfish than the average American, they are more vulnerable to seafood contamination. To address the risk of high-end consumers of Gulf seafood, we performed probabilistic risk assessments for commercial and recreational fishers on the Gulf Coast of Florida and Alabama. We also performed a risk assessment for a Filipino-American community in the Pensacola area. The risk assessments were performed using exposure data from household consumption surveys and chemical analysis of locally caught Gulf seafood after the DWH oil spill (November 2010 to November 2013). This study addressed risk from consumption of Gulf seafood after the DWH oil spill for Gulf commercial and recreational fishers. Additionally, it determined consumption rates for commercial fishers beyond a single ethnic group. Therefore, the commercial fisher consumption rates in this study should be more generally applicable to commercial fishers on the Gulf Coast. It is the first study to show Gulf commercial fishers are high-end consumers of fish and shellfish, regardless of ethnicity.

Methods:

Community survey

To determine the potential health risks to these specific populations, we utilized seafood consumption surveys from five communities across the northeastern Gulf of Mexico: Cedar Key, FL, Steinhatchee, FL, Apalachicola, FL, Pensacola, FL, and Mobile Bay, AL (Figure 1). This allowed us to capture seafood consumption data relevant to the target populations. Seafood consumption data were obtained for consenting participants and their household members (n=914), including spouses, children, and roommates (Kane AS, -submitted). The target population was defined by subpopulations that would provide a cross-section of "highend consumers of inshore-harvested seafood" in these coastal communities. These included participants who self-identified as a commercial fisher (including seafood harvesters, charter boat captains, boat workers or deck hands), a recreational fisher, or a member of the Filipino-American community. This assessment calculates risk for the adult consumers of Gulf seafood (n=484). Data for youth consumers (less than 21 years old) were excluded from the analysis.

Data were collected during the period of September 2013 to June 2014. Since the northeastern Gulf coast communities have 2 primary seasons. i.e., hot and cool, the survey tool was implemented during each of the 6-month "seasons" in each participating community. Warmer months were defined as April through October, and cooler months defined as October through March. Participants (n = 324) were read the informed consent (or read it themselves) in accordance with institutional review board (IRB) regulations and

provided their signature on duplicate documents to confirm their understanding of the study and their voluntary participation. Inclusion criteria for participating in the survey included: 1) 18 years or older, 2) participants consider themselves a gulf coast resident 6 months or more in the last year, and 3) willingness to participant in the study, regardless of whether or not they consume seafood. Procedures for data collection were conducted according to the protocol approved by University of Florida (UF) IRB 333–2011.

Household consumption parameters for the 4 seafood types discerned in this study (finfish, shrimp, oyster and blue crab) were based on self-reported data from study participants. Participants used a picture guide to report the portion sizes of respective seafood types they would typically consume, based on photographs of cooked, plated seafood portions (Mathews et al. 2017). The survey also included questions about respondents and household members' gender, age, and weight. Data were analyzed using USEPA's ProUCL, version 5.1 and Microsoft Excel software packages to provide descriptive statistics and percentiles for the region relative to specific seafood consumption rates.

Finfish and Shellfish Sample collection

All samples from the study region were collected from near shore locations in both oiled and non-oiled regions, representing commonly caught near shore species from November 2010 to November 2013. Sample collection sites were classified as oiled (Pass A Loutre, LA, Mobile Bay, AL, Pensacola, FL) or non-oiled (Cedar Key, FL, Steinhatchee, FL, and Apalachicola, FL) based on the recorded presence of oil in that region by NOAA (Figure 1). Seafood sampled included fish, shrimp, blue crab, and oyster. Fish were collected by hook and line, with a few specimens from Cedar Key, FL, collected by seine net. Shrimp were collected by trawler and dip nets. Blue crab were collected by cage traps, and oysters were collected by hand tonging. All specimens were immediately wrapped in clean aluminum foil, sealed in pouches, and placed on clean ice for transport to the laboratory, where, within 24 hours, further processing for chemical analysis took place. In the laboratory, a transverse section of muscle tissue (minimum 5 grams) with skin, including both dorsal and ventral portions, was excised from the anterior region of the fish's flank, immediately behind the operculum. For fish, 2 sections were taken for polynuclear aromatic hydrocarbon (PAH) analysis. The PAH replicates were wrapped in aluminum foil and placed in plastic bags. Shrimp were speciated, measured, and placed whole in foil. Blue crab were measured and sexed, the carapace was removed, and muscle tissue carefully separated from other viscera. For blue crab, 5-gram samples of muscle tissue were taken for PAH analysis. For oyster, individuals were shucked and placed in foil. All samples were then frozen at -20° C.

Chemical analysis

Fish, shrimp, blue crab, and oyster samples were analyzed individually with the exception of shrimp less than 5 grams, which were pooled together with other small shrimp to ensure a large enough sample for chemical analysis. Therefore, chemical analyses were for individual animals, not composites. Samples were homogenized using dry ice and a cryogenic tissue grinder (BioSpec Products - CTG111). The still frozen homogenate was then placed in scintillation vials with the cap loosely attached and placed in the freezer. Caps were tightened after sublimation of any remaining dry ice.

Extraction of PAHs from seafood samples was carried out using a QuEChERS method (Quick, Easy, Cheap, Effective, Rugged, and Safe; Ramalhosa et al. 2009), modified from the PAH extraction method of Smith and Lynam (2011). After centrifugation for 15 minutes at $500 \times g$ at ambient temperature, the supernatant extract was pipetted into clean 50 mL round-bottom centrifuge tubes. Lipid clean-up powders containing 400 mg end-capped C18, 400 mg primary- secondary amine, and 1,400 mg magnesium sulfate were added to the samples, shaken vigorously for 1 minute, and centrifuged for 15 minutes at $500 \times g$. The supernatant was collected into 15 mL conical tubes and concentrated to 0.5 mL for analysis. A total of 8 calibration standards containing US-106N (16 PAHs), C1 naphthalene, C1 fluorene, C1 phenanthrene, C1 pyrene, and C1 chrysene were prepared in concentrations between 1–100 ng/mL in ACN. The analyte list is provided in Table 1.

PAH analysis was performed using a gas chromatograph-mass spectrometer (GCMS; Agilent Technologies, Santa Clara, CA) equipped with a pre-column (HP-5MS; 5 m \times 0.25 mm \times 0.25 µm, from Agilent) and an analytical column (25 m \times 0.25 mm \times 0.25 µm, from Agilent) in selected ion mode (SIM). During the course of sample analysis, method blanks and positive controls in seafood were included, interspersed with every 25–30 unknowns. Positive controls contained nominal PAH concentrations of 10, 50, or 100 ng/mL in extract, corresponding to nominal concentrations of either 1, 5 or 10 ng/g. QuEChERS extraction efficiency was generally acceptable, with most recoveries between 60–120% of expected concentration. Outlying values >100% of nominal PAH concentration are believed to be a consequence of matrix interference, such as lipid or other components that were not completely removed by QuEChERS extraction.

Probabilistic Risk Analysis

We used the Microsoft Excel Add-in @Risk, version 7.5.0 (Palisade Corporation, Ithaca, NY) for fitting data to distributions, assigning correlations, and probabilistic risk assessment. Distributions were created for body weight and seafood consumption using survey data for adult consumers (21 years old and older). The shape of the distribution was chosen by goodness-of-fit testing and the Akaike Information Criterion. Distributions were truncated at 0 and the 99.9th percentile to ensure unrealistic values were not included in the assessment (Cullen et al. 1995; USEPA 2001). If there were less than 50 data points (or greater than 85% non-detects), a custom distribution was used. Because these data had too few data points to assign a distribution with confidence, the actual data were fit to a risk cumulative distribution. In this distribution, consumption was represented in the same proportion as was seen in the actual populations. The distributions and their graphs are presented as supplemental material. @Risk was run using 10,000 iterations. The sampling type chosen for the assessment was Latin Hypercube with a Mersenne Twister generator. The analysis was run using a fixed seed of 123457.

Body weight: Body weight distributions were obtained using self-reported body weights from the community surveys. Separate distributions were created for each population of interest: commercial fishers, recreational fishers, and Filipino-Americans.

Intake rate: Seafood consumption distributions (in grams per day) were derived for each population of high-end consumers included in the study (commercial fishers, recreational

fishers, and Filipino-Americans). Separate distributions were derived for each of the 4 types of seafood included in the survey: fish, shrimp, blue crab, and oyster. This resulted in 12 intake rate distributions.

PAH concentration: A distribution was derived for each contaminant with at least one detected concentration in oiled or non-oiled fish, shrimp, oyster, or crabs. Detected chemicals included acenaphthene, acenaphthylene, anthracene, fluoranthene, fluorene, naphthalene, C1 naphthalene, phenanthrene, C1 phenanthrene, pyrene, and C1 pyrene (Table 1). Detection limits for each analyte are listed in Table 1. Up to 8 concentration distributions were created for each chemical (1 for each seafood type in both oiled and non-oiled areas) depending on whether it was detected. Based on USEPA risk assessment methodology (USEPA, 1989), if a contaminant is not detected in any sample and the detection limit is below levels of concern (LOCs), the contaminant is not brought forward into the risk assessment. We utilized this convention for the probabilistic risk assessment. All of the detection limits achieved in this analysis were well below LOCs. Therefore, if the contaminant did not have any detected concentrations in a particular group (oiled or nonoiled for each seafood type), it was considered absent from the seafood and a distribution was not created. If a chemical had at least 1 detected concentration it was included in the risk assessment. Data were then fit to a distribution as described above. If the data did not fit a distribution, the detection limit was used for all non-detects. This occurred in datasets with a limited number of detects where the non-detected concentrations could not be inferred by the known detected concentrations. State and federal regulatory agencies often express a preference whether the limit of detection (LOD), ½ the LOD, or the LOD/2^0.5 should be used to replace non-detected values. In this assessment, the detection limit was chosen over one-half the detection limit and other non-detect estimation methods to represent a conservative scenario in addressing non-detects. Other methods of estimating non-detects would result in lower hazard quotients.

Correlations: Three different types of correlations were checked: 1) correlation between body weight and intake rate, 2) correlation between consumption of each of the four types of seafood, and 3) correlation between different PAH congener concentrations in the same animal tissue. Correlations were run using the NMLE procedure in SAS version 9.3 (SAS Institute Inc., Cary, NC, USA). Correlations were included in the @Risk software and incorporated as part of the probabilistic risk assessment.

Hazard quotient: A hazard quotient was calculated for each PAH analyte, seafood type, and population of interest using the equation:

$$\text{Hazard quotient } = \left(C_i \times IR_s \right) / \left(RfD_i \times BW_p \times CF \right)$$

where C_i is the distribution of concentrations (mg chemical/kg tissue) of the PAH analyte i, IR_s is the distribution of intake rates (grams per day) for seafood type s, RfD_i is the reference dose for PAH analyte i (Table 2), BW_p is the distribution of body weights (kg) for the population of interest p, and CF is a conversion factor (1000 g/kg).

Reference doses were not available from the USEPA for acenaphthylene and phenanthrene. Therefore, a surrogate was used to represent the toxicity of these chemicals. We chose naphthalene as the surrogate for these chemicals because it has the highest reference dose of the listed PAHs (it has the most sensitive non-carcinogenic critical effect). Choosing the highest reference dose is a conservative method for estimating hazards from exposure to PAH tainted seafood.

Hazard index: Hazard quotients for the PAH analytes were summed for each seafood type (fish, shrimp, blue crab, oyster) to obtain 4 separate hazard indices for seafood consumption for each population of interest (commercial fisher, recreational fisher, Filipino-American), and location (oiled or non-oiled). This resulted in 24 separate hazard indices (4 seafood types × 3 populations × 2 locations). A screening level risk assessment approach was taken in calculating the hazard indices in which hazard quotients were added for each PAH regardless of the toxic endpoint on which their reference doses were based. This very conservative approach assumes that the effects of all of the PAHs are additive regardless where they occur. Further, to simulate a conservative scenario where Gulf coast residents are high end consumers of all surveyed seafood types, 95th percentile hazard indices for fish, shrimp, blue crab, and oyster were summed for each population and location. With this conservative screening approach, if the hazard indices are below 1, there is little doubt that the combined exposures are safe. If hazard indices above 1 are calculated, a more refined assessment of potential additive effects may be warranted to determine whether the combined exposure presents unacceptable risk.

Statistics

SAS 9.4 software (SAS Institute Inc., Cary, NC, USA) was used to perform the statistical analyses. The number of seafood samples with detected concentrations in the oiled area seafood was compared to the number of seafood samples with detected concentrations in non-oiled area seafood for each PAH. Pearson's chi-square test was used to determine significance for analytes with 5 or more detects in both oiled and non-oiled areas and Fisher's exact test was used to determine significance for analytes with less than 5 detects in either the oiled and non-oiled areas.

Results:

Survey

Consumption data for 489 adult household members were reported by 235 participants in the study across the five Gulf coast communities. Commercial workers comprised 31.3% of the sample size, while recreational fishers represented 61.1%, and the Filipino-American community represented 7.6% of the sample. The study population was 50.9% male and 49.1% female. The average body weight for commercial and recreational fishers were equivalent at 83 kg. The average body weight for Filipino-American adults was 68 kg.

The survey showed increased consumption of fish and shellfish among the targeted Gulf coast populations compared with national consumption rates. The 90th percentile consumption rates for commercial fishers, recreational fishers, and Filipino-Americans were

greater than 90th percentile NHANES consumption estimates used to derive the LOCs (Table 3). The 90th percentile fish consumption rates for the targeted Gulf coast populations were 1.6–2.5 times greater than the nationwide 90th percentile. The 90th percentile shellfish consumption rates were even higher. Oyster consumption in the target Gulf coast populations was 1.3–5.7 times the nationwide 90th percentile and combined shrimp and crab consumption was 3.6–6.5 times the nationwide 90th percentile. The 50th percentile consumption rates for commercial fishers, recreational fishers, and Filipino-Americans were less than or equal to the 90th percentile NHANES nationwide consumption estimates (Table 4). The 50th percentile shrimp and crab consumption rates for the high-end consumers were 1.5–2.2 times the nationwide 90th percentile.

Analytical

Carcinogenic PAHs were not detected in any of the seafood samples analyzed (detection limits = 0.1–2.5 ng/g). Because carcinogenic PAHs were not detected, they were not brought forward into the risk assessment (USEPA, 1989). The primary concern from exposure to PAH is the cancer risk. Therefore, the cancer risk from exposure to the Gulf seafood sampled in this study was de minimis. Eleven non-carcinogenic PAHs were detected in seafood harvested from both oiled areas and non-oiled areas (Table 1). Therefore, this assessment focuses on a hazard assessment of exposure to PAHs. There were differences in detected PAHs between oiled and non-oiled area seafood. Acenaphthylene, anthracene, and C1 phenanthrene were only detected in oiled area seafood while C1 pyrene was only detected in non-oiled area seafood. The most commonly detected PAHs were phenanthrene (51% of all seafood samples), naphthalene (31%), fluoranthene (15%), and fluorene (14%). At least one PAH was detected in 72% of fish samples, 54% of shrimp, 48% of blue crab, and 59% of oysters. The remaining seafood did not have any detections of PAHs. For shrimp, crab, and oyster, seafood collected from the oiled areas had a larger percentage of samples with at least one PAH detection than those collected from the non-oiled areas (Table 5). For fish, non-oiled areas had a larger percentage of samples with at least one PAH detection (67% of fish in oiled areas versus 79% in non-oiled areas; Table 5). Acenaphthene, acenaphthylene, C1 phenanthrene, and C1 pyrene were detected in less than five seafood samples (Table 6). Anthracene and phenanthrene had a greater number of detected concentrations in oiled area seafood than non-oiled area seafood (p<0.05). C1 Naphthalene was detected less in oiled area seafood than non-oiled area seafood (p<0.05; Table 6). Generally (excluding the three exceptions listed above), the percentage of PAH detections between oiled and non-oiled area seafood is similar.

Correlations

The goal of the probabilistic risk assessment is to simulate a study population that accurately reflects the magnitude and variability in the targeted populations along the northeastern Gulf Coast. Therefore, correlations were run between body weight and consumption, consumption of different seafood types, and chemical analytes present to determine whether the assumption of independence is correct for any of these variables. Correlation coefficients between consumption in grams per day for each of the four seafood types and body weight ranged from 0.06 and 0.15 for our study population. These are considered very weak correlations. Based on these correlations, the risk assessment assumed consumption was

independent of body weight. Correlation coefficients were also calculated between seafood types. This correlation determined whether the quantity consumed of one type of seafood was a predictor of the quantity consumed for any other type of seafood. Correlations of consumption between seafood types ranged from 0.06 to 0.55 and are considered weak to moderate (Table 7). Because some moderate correlations were present, correlations between consumption rates of different seafood types were added to the probabilistic risk assessment model.

Correlation coefficients were also calculated between all detected contaminants in each type of seafood for both oiled and non-oiled areas. This resulted in eight different sets of correlation coefficients for the probabilistic risk assessment. These coefficients ranged from 0.07 to 0.55 indicating a weak to moderate correlation between chemical analytes in seafood tissue. Although these correlations did not have a strong effect on the hazard calculation, they were included in the probabilistic risk assessment for completeness.

Risk Assessment

The modeled 95th percentile hazard quotients for commercial fishers, recreational fishers, and a Filipino-American community are presented in Table 8. Hazard quotients are presented for both oiled and non-oiled areas of the Gulf of Mexico by seafood type. The 95th percentile hazard quotient for individual seafood types ranged from 1.84E-04 to 5.39E-03. The largest modeled 95th percentile hazard quotient for individual seafood type of 5.39E-03 was for the Filipino- American community consumption of fish harvested in non-oiled areas. The largest modeled 95th percentile hazard quotient for shrimp (2.16E-03) was also modeled for the Filipino-American community. The largest modeled 95th percentile hazard quotient for oyster (1.52E-03) and blue crab (1.88E-03) were calculated for commercial fisher consumption of these seafood items in oiled areas. The lowest modeled hazard quotients (1.84E-04 to 2.43E-04) are for the consumption of crab harvested from non-oiled areas for all targeted populations. Hazard quotients for consumption of individual seafood types for all targeted populations in both oiled and non-oiled areas are three to four orders of magnitude below the regulatory limit and effect threshold of 1.

The 95th percentile hazard indices for total seafood consumption (sum of the individual seafood 95th percentile hazard indices) ranged from 3.45E-03 to 8.41E-03. Hazard index is presented for seafood consumption from both oiled and non-oiled areas. The largest hazard index for total seafood consumption (8.41E-03) was modeled for the Filipino-American community consumption of seafood harvested in oiled areas (Figure 2a). Hazard indices for total seafood consumption were highest for Filipino-Americans (Figures 2a, b), followed by commercial fishers (Figures 3a, b), followed by recreational fishers (Figures 4a, b). For all three studied populations, hazard indices were higher in oiled versus non-oiled areas. The 95th percentile hazard indices for total seafood are approximately three orders of magnitude below a hazard index of 1. The mean, 95th percentile, and 99th percentile hazard indices for total seafood consumption are presented in Table 9. Mean hazard indices ranged from 1.03E-03 to 2.67E-03. The 99th percentile hazard indices ranged from 8.33E-03 to 1.81E-02. For the high-end consumers of seafood included in this study, all modeled hazard indices are well below the regulatory limit and effect threshold of 1. Based on total seafood

consumption, the Filipino-American community shows the highest modeled hazards indices with a 99th percentile hazard index of 1.81E-02 in oiled areas. Commercial fishers had lower hazard indices than the Filipino-American community but were closer to this community in modeled hazards than to recreational fishers. Recreational fishers show the lowest modeled hazards for total seafood consumption (Table 9).

Discussion:

Risk assessments performed by governmental agencies after the Deepwater Horizon oil spill were not targeted to address the higher seafood consumption rates of coastal populations. Additionally, these assessments were not intended to address ethnic and subsistence fishers who typically consume larger quantities of fish and shellfish than the general population. This study demonstrated increased consumption of fish and shellfish among the targeted Gulf coast populations. Commercial fishers and the Filipino-American participants in this study consumed over twice the amount of fish and greater than six times the amount of shrimp and crab assumed in the FDA risk assessment.

Practical considerations precluded taking a truly random sample of the populations of interest and the sampling strategy instead was opportunistic. As such, the possibility exists that the consumption rates may be biased in some way. However, the higher consumption rates for the Gulf coast participants in this study were consistent with other surveys of highend consumers of seafood. Native Americans in the state of Washington have a 90th percentile fish ingestion rate of 114–397 g/d (WDOE 2012), depending on tribal affiliation. A study in King County Washington characterized the seafood consumption of 10 Asian American and Pacific Islander ethnic groups. The 90th percentile finfish consumption rate was 102 g/d (Sechena et al. 2003). Commercial fishers (106 g/d) and the Filipino-American (121 g/d) 90th percentile consumption rates in this study are similar to other high-end consumers of finfish. This study reported an average finfish consumption rate for recreational fishers of 37 g/d and a 90th percentile consumption rate of 76 g/d, which are similar to consumption rates for recreational fishers in other parts of the United States. Studies that examined fish consumption rates of recreational fisherman in coastal New Jersey and Bar Reservoir in Tennessee reported an average fish consumption rate of 52.8 and 37.4 g/d, respectively (Burger and Campbell 2008; May and Burger 1996). The 90th percentile consumption rates were not reported for these studies. Our survey found 90th percentile shrimp consumption rates of 45, 61, and 71 g/d for recreational fishers, commercial fishers, and the Filipino-American community, respectively. Wilson et al. (2015) reported a mean consumption rate of 45.2 g/d for a Vietnamese-American community in eastern New Orleans, LA. A survey of Louisiana residents reported an upper-end estimate of 65.1 g/d shrimp (Anderson and Rice 1993). Commercial fishers in this study also consumed more than five times the quantity of oysters used in the risk assessment (68 g/d versus 12 g/d assumed by the FDA).

The results this study and others cited above suggest that using national finfish and shellfish consumption rates may underestimate contaminant intake for high-end consumers of locally harvested seafood. Because the populations surveyed in this study are high-end consumers, we also compared the nationwide 90th percentile consumption rates to 50th percentile

consumption rates for commercial fishers, recreational fishers, and the Filipino-American participants. These data show the percent of the high-end consumer population protected under the nationwide consumption estimates. While the increased consumption rates in this study are supported by fish and shellfish consumption surveys performed on other targeted populations, there are differences in the type and amount of seafood consumed between commercial fishers, recreational fishers, and the Filipino-American community. This demonstrates that high-end consumer populations can vary in their seafood consumption patterns, and that these differences should be taken into consideration when assessing risk to these populations and developing health protective LOCs.

Body weight is another important variable that merits attention when considering risk to high-end consumer populations. The Filipino-American community on the Florida Gulf coast had a lower average body weight (68 kg) than the 80 kg assumption used for the FDA risk assessment method. Our finding of a lower average body weight for Asian-American communities has been observed in other surveys. Wilson et al. (2015) reported an average body weight of 63 kg for a Vietnamese-American community in Louisiana and Sechena et al. (2003) reported an average body weight of 62 kg for a group of Asian-Americans and Pacific Islanders in Washington. A lower body weight results in a higher dose (mg/kg-d) of PAHs and therefore higher hazard indices. In this probabilistic risk assessment, the Filipino-American community consumed only 14% more finfish per day than commercial fishers. However, their hazard estimate based on consumption was more than double that of commercial fishers (even though both hazard indices remained well below the threshold of 1). The lower body weight of the Filipino-American community caused their hazard estimate to increase disproportionally due to the difference in consumption rates. Similar to consumption rates, body weights used in the risk assessment should be tailored to the population of concern. Otherwise, risk from exposure to PAHs after future coastal oil spills may be underestimated for some individuals.

LOCs developed following the Deepwater Horizon oil spill were based on exposure to carcinogenic PAHs. This study did not detect carcinogenic PAHs in any seafood sampled from near shore along the northeastern Gulf of Mexico after the Deepwater Horizon oil spill. This is in agreement with a study by Wilson et al. (2015) which also did not detect carcinogenic PAHs in shrimp sampled inshore and offshore in an area of Chandeleur Sound near Louisiana after the Deepwater Horizon oil spill in November 2010. In 2018, Wickliffe et al. reported detections of carcinogenic PAHs in shrimp and finfish consumed by members of the participant household. However, it is unclear what percentage of these samples were obtained locally from the Gulf.

To provide a conservative estimate of risk, the detection limit was used for non-detects in the creation of concentration distributions for the risk assessment. Additionally, the total hazard index was calculated by summing the hazard quotients for all PAHs regardless of their adverse effect, and summing the hazard index for all four seafood types at the same percentiles. This approach assumes additive effects for all PAHs and that an individual is an upper-end consumer of all four seafood types included in this study (fish, shrimp, oysters, and blue crabs), which is unlikely based on the weak correlations between consumption of different seafood types. Despite the increased fish and shellfish consumption rates, lower

body weight for the Filipino-American community, and assumption that participants were high end consumers of all four seafood types included in this study, 99th percentile hazard quotients for commercial fishers, recreational fishers, and the Filipino-American community did not exceed 1. The 99th percentile hazard quotients ranged from 8.33E-03 to 1.81E-02, which is approximately 100 times below the regulatory limit of 1. The risk assessment shows there is no risk from consumption of Gulf seafood in both oiled and non-oiled areas. These small hazard quotients are orders of magnitude below the effect threshold and likely due to ambient concentrations of non-carcinogenic PAHs in fish and shellfish tissue.

We hypothesized that consumption of seafood in grams per day would be correlated to kilograms of body weight. However, there was almost no correlation between these two factors in our study population. This is consistent with the absence of a correlation between fish ingestion rates and body weight from a large (ca. 8,000 household) study of fish consumption in the general population in Florida (CEHT, 2008). It is unclear why body weight was not a predictor of seafood consumption for adults in these studies. Because seafood only makes up a portion of the total diet, it is likely that the seafood consumption rates are driven by other factors (such as availability or cost) besides body weight.

This risk assessment was performed for adult high-end consumers of Gulf seafood. Children were not included in the study because there were not enough surveys for children in each of the targeted populations (commercial fishers, recreational fishers, and the community of Filipino-Americans) to derive distributions with any confidence. Because children consume more food per kilogram of body weight than adults, they represent a sensitive lifestage that is more susceptible to contaminants. The lack of child consumption data for each targeted population could be addressed by combining surveys across target populations to create a high-end consumer distribution for children. Additionally, although this study measured C1 PAH concentrations in seafood, limited toxicity data are available for these compounds. Therefore, the non-methylated versions of these compounds were used as surrogates in the risk assessment. Because the actual toxicity of these methylated PAHs is unknown, use of the non-methylated compound as a surrogate may under- or overestimate risk.

This study demonstrates default national fish consumption rates used in current risk assessments do not accurately represent high-end consumers along the northeast Gulf coast. Gulf of Mexico commercial fishers, recreational fishers, and a coastal Filipino-American community had 90th percentile consumption rates for fish and shellfish up to six times greater than national 90th percentile consumption rates. Although our study was limited to these targeted populations, it is likely that other groups of coastal residents also consume seafood in excess of the national estimates. Although these high-end consumer groups are subpopulations, in some locations, particularly small coastal communities, they can comprise a substantial percentage of the local population. As such, they need to be explicitly considered when developing risk estimates for oil spills and other coastal contamination events, as well as in developing LOCs for protection of public health.

This study provides estimates of risk from Gulf seafood consumption by high-end consumers in the Northeast region of the Gulf of Mexico following the Deepwater Horizon spill, and includes seafood from areas that received oil contamination and those that did not.

Studies by Wilson et al. (2015) and Wickliff et al. (2018) describe risks for high-end consumers in oiled regions of the Western Gulf. Together they provide information on high-end consumer risks along a gradient that spans most of the Gulf coast, ranging from proximity to the spill to what are essentially background levels. Although the populations and technical details of the studies vary somewhat, they are consistent in showing very low risks from PAHs in seafood throughout the Gulf following the spill. The results from these studies collectively should provide useful information to assist the rapid assessment of risk to high-end consumers during future oil spills in the Gulf and perhaps elsewhere.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1 –.

Map of the five communities across the northeastern Gulf of Mexico used in this study:
Cedar Key, FL, Steinhatchee, FL, Apalachicola, FL, Pensacola, FL, and Mobile Bay, AL.
Satellite image ©2018 TerraMetrics.

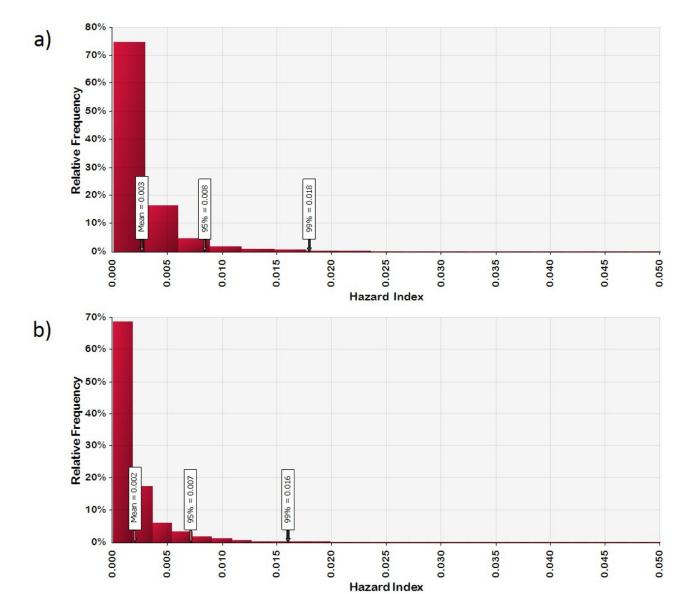


Figure 2 -.

Modeled total seafood consumption hazard distributions for a Filipino-American community in (A) oiled and (B) non-oiled areas along the northern Gulf coast.

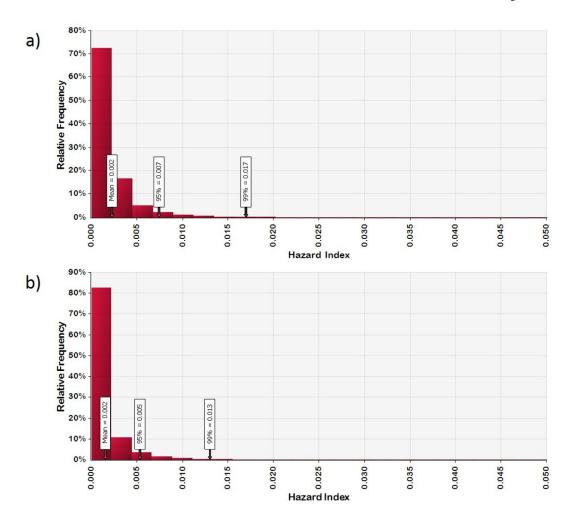
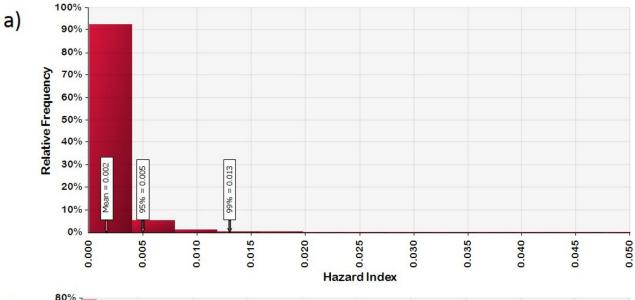


Figure 3 –.

Modeled total seafood consumption hazard distributions for commercial fishers in (A) oiled and (B) non-oiled areas along the northern Gulf coast.



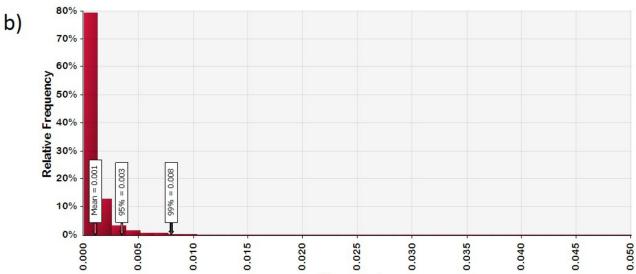


Figure 4 –.Modeled total seafood consumption hazard distributions for recreational fishers in (A) oiled and (B) non-oiled areas along the northern Gulf coast.

Hazard Index

Table 1 – Polynuclear aromatic hydrocarbon analyte list for Gulf seafood.

Chemical	Detection Limits (ng/g) ^a	Detected in non-oiled area seafood	Detected in oiled area seafood
Acenaphthene	0.1 – 0.25	X	X
Acenaphthylene	0.1 – 0.25		X
Anthracene	0.1 – 2		X
Benz[a]anthracene	0.1 – 1		
Benzo[a]pyrene	0.1 – 2		
Benzo[b]fluoranthene	0.1 – 2		
Benzo[g,h,i]perylene	0.1 – 1		
Benzo[k]fluoranthene	0.1 – 1		
Chrysene	0.1 – 1		
C1 Chrysene	0.1 – 2		
Dibenz[a,h]anthracene	0.1 – 2.5		
Fluoranthene	0.1 – 0.5	X	X
Fluorene	0.1 – 1	X	X
C1 Fluorene	0.1 – 1		
Indeno[1,2,3-cd]pyrene	0.1 – 2		
Naphthalene	0.1 – 0.25	X	X
C1 Naphthalene	0.1 – 0.25	X	X
Phenanthrene	0.1 – 0.5	X	X
C1 Phenanthrene	0.1 – 0.5		X
Pyrene	0.1 – 0.5	X	X
C1 Pyrene	0.1 – 0.5	X	

 $[\]stackrel{a}{-}$ Detection limits varied within the range depending upon tissue type

Table 2 -

Reference doses for detected PAH analytes obtained from the November 2017 USEPA Regional Screening Level Table.

Analyte	Reference dose (mg/kg-d)
Acenaphthene	6.0E-02
Acenaphthylene a	2.0E-02
Anthracene	3.0E-01
Fluoranthene	4.0E-02
Fluorene	4.0E-02
Naphthalene	2.0E-02
C1 Naphthalene ^b	2.0E-02
Phenanthrene a	2.0E-02
C1 Phenanthrene b	2.0E-02
Pyrene	3.0E-02
C1 Pyrene ^b	3.0E-02

 $[\]stackrel{a}{\mbox{-}}$ No toxicity value. Toxicity value based on naphthalene (most toxic non-carcinogen)

 $^{{\}color{blue}b}$ - Parent compound toxicity was used for C1 homologues

Table 3 -

90th percentile seafood consumption estimates for commercial fishers, recreational fishers, and Filipino-Americans on the Gulf coast of Florida and Alabama compared with national estimates.

Population	Number	Fish (g/d)	Oyster (g/d)	Shrimp (g/d)	Blue Crab (g/d)	Shrimp and Crab (g/d)
National ^a	NA	49	12	NA	NA	13
Commercial Fishers	153	106	68	61	16	81
Recreational Fishers	294	76	19	45	16	47
Filipino-Americans	37	121	16	71	11	85

^aFDA, 2010, NA – Not available

Table 4 -

 90^{th} percentile national seafood consumption estimates compared to 50^{th} percentile estimates for commercial fishers, recreational fishers, and Filipino-Americans on the Gulf coast of Florida and Alabama

Population	Number	Fish (g/d)	Oyster (g/d)	Shrimp (g/d)	Blue Crab (g/d)	Shrimp and Crab (g/d)
National ^a	NA	49	12	NA	NA	13
Commercial Fishers	153	34	8	24	5	29
Recreational Fishers	294	24	5	16	3	19
Filipino-Americans	37	50	4	25	3	28

^aFDA, 2010, NA – Not available

Table 5 -

Number of samples obtained for each type of seafood and the percentage of samples that had a detectable concentration of at least one non-carcinogenic PAH.

Area	Seafood	Number sampled	Number with PAH detections	Percent detects
	Fish	167	132	79%
Non-oiled	Shrimp	49	17	35%
Non-oned	Oyster	76	34	45%
Crab	Crab	43	14	41%
	Fish	288	194	67%
0:1-4	Shrimp	56	40	71%
Oiled	Oyster	23	13	57%
	Crab	19	17	90%

Table 6 – Number of seafood samples with detected concentrations of non-carcinogenic PAHs

	Non-oiled a	rea seafood	Oiled are	a seafood
Chemical	Number of samples Number of detects		Number of samples	Number of detects
Acenaphthene	335	1	388	4
Acenaphthylene	335	0	388	1
Anthracene	335	0	388	24*
Fluoranthene	335	50	388	62
Fluorene	335	41	388	57
Naphthalene	335	113	388	108
C1 Naphthalene	210	26	307	9*
Phenanthrene	335	146	388	224*
C1 Phenanthrene	210	0	307	4
Pyrene	335	11	388	19
C1 Pyrene	210	1	307	0

 $[\]sp{*}$ - significantly different from control (non-oiled area seafood) with p<0.05

Table 7 -

Correlation coefficients for consumption (in grams per day) between seafood types for (A) commercial fishers, (B) recreational fishers, and (C) a Filipino-American community.

(A) Commercial fisher correlation coefficients						
	Fish Shrimp Oyster Blue Crab					
Fish	1					
Shrimp	0.43	1				
Oyster	0.23	0.19	1			
Blue Crab	0.43	0.33	0.23	1		

(B) Recreational fisher correlation coefficients						
	Fish Shrimp Oyster Blue Crab					
Fish	1					
Shrimp	0.42	1				
Oyster	0.38	0.10	1			
Blue Crab	0.19	0.14	0.35	1		

(C) Filipino-American community correlation coefficients							
	Fish	Fish Shrimp Oyster Blue Crab					
Fish	1						
Shrimp	0.52	1					
Oyster	0.17	0.06	1				
Blue Crab	0.25	0.55	0.31	1			

Table 8 -

95th Percentile hazard quotients for the consumption of fish, shrimp, oyster, and crab in both non-oiled and oiled areas.

Population	Area	Fish hazard quotient	Shrimp hazard quotient	Oyster hazard quotient	Crab hazard quotient	Total hazard index
Commercial Fishers	Non-oiled	3.73E-03	1.55E-03	7.12E-04	2.43E-04	5.33E-03
Commercial Fishers	Oiled	3.28E-03	1.55E-03	1.52E-03	1.88E-03	7.46E-03
D 1 Fish	Non-oiled	2.25E-03	1.04E-03	3.60E-04	2.08E-04	3.45E-03
Recreational Fishers	Oiled	2.18E-03	1.12E-03	7.51E-04	1.49E-03	5.02E-03
Trilinia American	Non-oiled	5.39E-03	2.16E-03	4.35E-04	1.84E-04	7.09E-03
Filipino- Americans	Oiled	5.01E-03	2.07E-03	9.14E-04	1.38E-03	8.41E-03

Non-oiled area - Cedar Key, FL, Steinhatchee, FL, and Apalachicola, FL; Oiled area - Pass A Loutre, Louisiana, Mobile Bay, AL, Pensacola, FL

 $\label{eq:Table 9-Mean} \textbf{Mean}, 95^{th} \text{ percentile, and } 99^{th} \text{ percentile hazard indices for the consumption of all four seafood types.}$

Area	Population	Population Mean hazard index 95th Percentile hazard		99 th Percentile hazard index
Commercial Fishers	Non-oiled	1.56E-03	5.33E-03	1.27E-02
Commercial Fishers	Oiled	2.27E-03	7.46E-03	1.69E-02
Decreational Eighan	Non-oiled	1.03E-03	3.45E-03	8.33E-03
Recreational Fishers Oile		1.61E-03	5.02E-03	1.29E-02
Elining American	Non-oiled	2.05E-03	7.09E-03	1.55E-02
Filipino- Americans	Oiled	2.67E-03	8.41E-03	1.81E-02

Non-oiled area - Cedar Key, FL, Steinhatchee, FL, and Apalachicola, FL; Oiled area - Pass A Loutre, Louisiana, Mobile Bay, AL, Pensacola, FL