Assessing Fisheries as Vectors for Toxic Materials from the Environment to Humans.

An assessment of potential health risks posed by shellfish collected in estuarine waters near Pensacola, Florida.

Natalie K. Karouna-Renier, Richard A. Snyder, K. Ranga Rao

Center for Environmental Diagnostics and Bioremediation, University of West Florida, 11000 University Parkway Pensacola, FL 32514

ABSTRACT

As part of an environmental health study of northwest Florida, we conducted an initial screening level assessment of contaminants in blue crabs (*Callinectes sapidus*) and oysters (*Crassostrea virginica*) collected in bays and bayous in the Pensacola, FL area. Tissue samples were analyzed for mercury, arsenic, cadmium, chromium, copper, lead, nickel, selenium, tin, zinc, 17 dioxin/furan compounds, and 12 dioxin-like PCB congeners (PCB-77, PCB-81, PCB-105, PCB-114, PCB-118, PCB-123, PCB-126, PCB-156, PCB-157, PCB-167, PCB-169, and PCB-189). Contaminant levels were compared to Screening Values (SV) calculated using the U.S. EPA recommendations for establishing consumption advisories. Four different consumption rates were used in the derivation of the SVs.

We identified five chemicals of concern (dioxins/furans/PCBs, arsenic, mercury, cadmium, and zinc) in either crab muscle, crab hepatopancreas, total crab tissue, or oysters based

on exceedence of one or more SVs. We also assessed health risks (non-carcinogenic and carcinogenic) that may arise as a result of consumption of these shellfish species. Dioxins/PCBs accounted for 85-99%, 60-90%, 27-94%, and 53-99% of the total excess cancer risks for crab hepatopancreas, total edible crab tissue, crab muscle, and oysters, respectively. The relative contributions of dioxins/furans and dioxin-like PCBs to the TEQs and resultant risks varied with location, as evident from analysis of the crab hepatopancreas samples. Dioxins/furans were a greater contributor in samples from Bayou Chico and Perdido Bay, whereas PCBs were dominant in Bayou Grande and Western Escambia Bay. The locations that exceeded SVs and had the highest carcinogenic or non-carcinogenic health risks were generally located in urbanized waterbodies (Bayou Texar, Bayou Grande, and Bayou Chico) or downstream of known contaminated areas (Western Escambia Bay). Oysters collected from commercial oyster beds in Escambia and East Bays, and crabs collected from East, Blackwater and Perdido Bays generally had the lowest levels of contaminants. Despite accounting for only 15% of the total tissue, inclusion of hepatopancreas in a crab meal increased contamination to levels above many SVs, and therefore, direct or indirect consumption of hepatopancreas from crabs in the Pensacola Bay system should be discouraged. Further investigation is warranted to determine whether consumption advisories should be issued for shellfish from specific locations in the Pensacola Bay system.

Keywords: blue crabs, oysters, PCBs, dioxins, metals

INTRODUCTION

The U.S. Environmental Protection Agency reported that, for 2002, 32.9% of the nation's lake acreage, 15.3% of the total river miles, 100% of the Great Lakes and their connecting water bodies, and 71% of coastal waters, including 100% of the Gulf Coast were under consumption advisories for fish (U.S. EPA, 2003). Although 39 chemicals were responsible for the advisories, mercury, PCBs, chlordane, dioxins, and DDT accounted for the majority (96%) of consumption restrictions. These chemicals are bioaccumulated in the tissues of aquatic organisms at concentrations many times higher than concentrations in the water, and are passed up the food chain to fish where they may be concentrated to levels that cause physiological impairment in human consumers. Although a number of the monitored chemicals are no longer used or manufactured in the United States, studies have shown that they continue to accumulate in a variety of foods, including shellfish (Jensen and Bolger, 2001). For example, over 90% of human exposure to organochorine compounds occurs through diet, primarily through seafood and meat (Smith and Gangolli, 2002). Segments of the human population with increased toxic exposure risk include consumers of commercially harvested seafood, recreational fishers, and citizens that rely on harvestable species for subsistence.

The Pensacola Bay region is located at the tip of the Florida Panhandle near the Florida-Alabama border. Although historically the area supported a rich and diverse ecology and productive fishery, the effects of many decades of point and nonpoint source pollution, habitat destruction, industrial activities, and development have impaired the health and productivity of the estuarine waters in the region (Thorpe et al., 1997). The area is home to a number of historical and potential contaminant sources including paper mills, a coal-burning power plant,

industrial complexes, military facilities, multiple Superfund sites, sewage treatment plant outfalls, storm water discharges, atmospheric deposition, septic tanks, golf courses and agriculture in the watershed. Significant quantities of fish and shellfish are harvested both commercially and recreationally in Northwest Florida. Although mercury levels have been routinely monitored by the State of Florida, especially for freshwater fisheries, screening of seafood for other contaminants within the state, especially along the Gulf of Mexico has been limited.

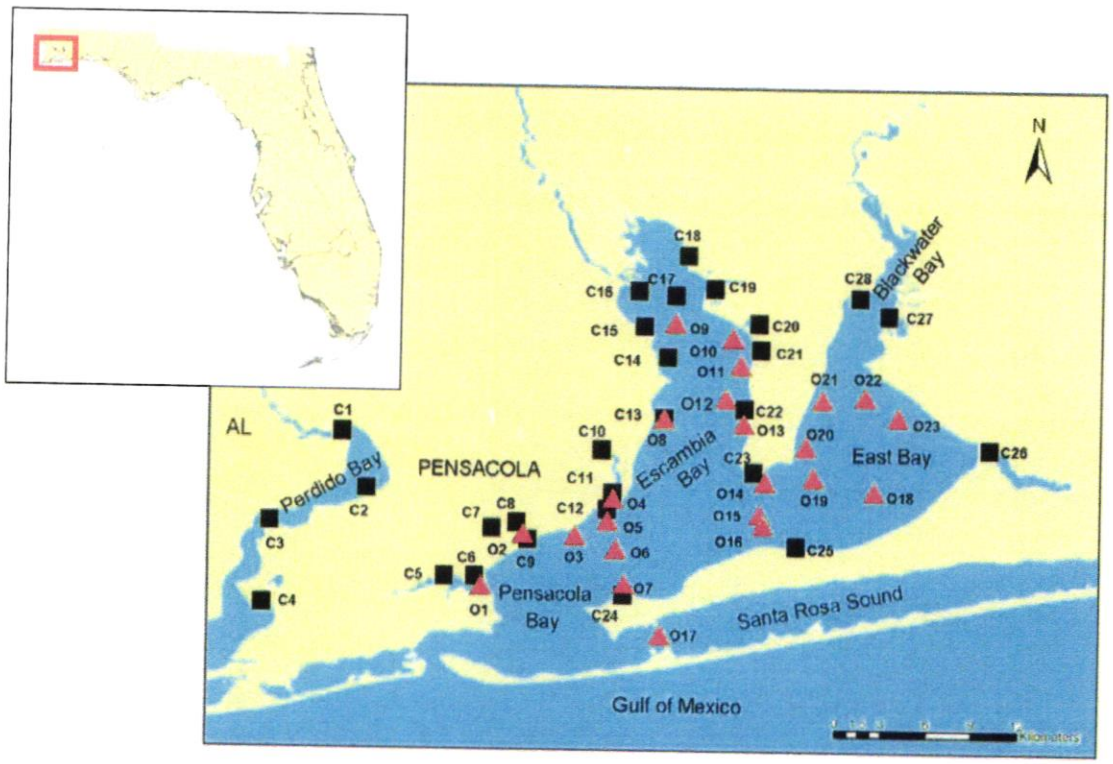

Figure 1. Pensacola Bay area blue crab and oyster sampling locations.
■ = crab sampling station, ▲ = oyster sampling station.

In the present study, we report the results of a screening survey of contaminants in eastern oyster (*Crassostrea virginica*) and blue crab (*Callinectes sapidus*) samples collected at multiple sites in the Pensacola Bay area. These two species are widely consumed along coastal regions of the United States. In 2003, commercial harvest levels for oysters and blue crabs in the two Florida counties (Escambia and Santa Rosa) that make up the Pensacola region totaled 9,140.5 and 60,956.6 kg, respectively (FWCC, 2004). Because they are sessile filter-feeders, oysters bioconcentrate chemicals from the water column and thus are indicative of the types and concentrations of contaminants in the surrounding water. In contrast, blue crabs spend much of their lives in and on the sediments, where they feed predominantly on other benthic organisms and thus are more indicative of sediment contamination levels. The objectives of this study were

to determine whether these shellfish in the Pensacola Bay region carry significant body burdens of toxic chemicals, identify chemicals of concern exceeding screening values, and identify "hotspot" locations of concern where elevated levels of contamination are found. Following the model established by the U.S. EPA in 2000, we analyzed the oyster and crab tissues for metals, including mercury, polychlorinated dibenzo-p-dioxins (PCDD)/ dibenzofurans (PCDF), and dioxin-like polychlorinated biphenyl (PCB) congeners, and used these data to characterize the degree of health risk associated with consuming oysters and crabs from the Pensacola Bay system.

METHODS

Study Area

The Pensacola Bay system and its smaller neighbor, Perdido Bay, are located at the far western tip of the Florida Panhandle, on the border with Alabama (Figure 1). The Pensacola Bay watershed drains 18,100 km^2 of Florida and southern Alabama, and is comprised of five major estuaries - Pensacola, Escambia, Blackwater and East Bays, and Santa Rosa Sound (Thorpe et al., 1997). The estuaries receive drainage from four major rivers (Escambia, Yellow, Blackwater and East Rivers) and many smaller tributaries and bayous. Perdido Bay is a small estuarine system that is fed by freshwater from the Perdido River and several smaller tributaries, and has a drainage area of 3,100 km^2 (U.S. EPA, 1999).

Sample Collection

Sampling locations were spread throughout the various estuaries and bayous draining into Pensacola Bay and on the Florida (east) side of Perdido Bay (Figure 1). Samples were collected from locations identified by biologists and local crabbers/oystermen. In general, based on the EPA guidance for a screening study, we analyzed one composite sample for each target organism at each location.

We collected oysters (*C. virginica*) by using tongs or by hand between March 2003 and July 2004 from 23 locations (Table 1, Figure 1). Sampling locations were classified into three groups - bridges that span the major bays (locations O5-7, O9, and O14-17), commercial oyster beds in Escambia and East Bays (locations O10-13 and O18-23, respectively), and urbanized waterbodies (locations O1-4). Oysters were not collected from Perdido Bay because harvestable populations could not be identified. Upon collection, oysters shell length was measured and the samples were placed on wet ice for transport to the laboratory. Oyster tissues were prepared by severing the adductor muscle, prying open the shell, and removing the soft tissue. A minimum of 10 oysters was composited for each location and shipped to the analytical facilities for homogenization and analyses.

Blue crabs (*C. sapidus*) were collected between June 2003 and June 2004 from 28 locations, using baited commercial crab traps deployed for 24-72 hours. Sampling locations were grouped as follows: Perdido Bay (locations C1-4), urbanized bayous (locations C5-12 and C24), western Escambia Bay (locations C13-17), eastern Escambia Bay (locations C18-22), and East/Blackwater Bay (locations C23 and C25-28). Between 10 and 15 traps were deployed at each location. Crabs over 10.2 cm in carapace width were selected and the sex of the collected crabs was determined. Although blue crabs are known to migrate within an estuary, male

migration areas are generally smaller than those of females (Ju and Harvey, 2002) and therefore, where possible, females were not included in the analyses. Samples were transported to the laboratory on wet ice. Tissue from seven to 15 crabs was composited for each location. Crab muscle and hepatopancreas were analyzed separately. Crabs were prepared by separating the carapace from the body and removing the hepatopancreas using forceps. All other internal organs were also removed and discarded. Edible muscle tissue, including claw meat, was extracted by splitting each thorax in half and processing through a cleaned compression device (Crab Master™). The extracted tissues were then homogenized using a stainless steel hand-held homogenizer and shipped to the analytical facilities for further homogenization and analyses.

Contaminant analysis

All tissue samples were analyzed for the following contaminants: mercury, arsenic (total), cadmium, chromium, copper, lead, nickel, selenium, tin, zinc, 17 dioxin/furan compounds, and 12 dioxin-like PCB congeners (PCB-77, PCB-81, PCB-105, PCB-114, PCB-118, PCB-123, PCB-126, PCB-156, PCB-157, PCB-167, PCB-169, and PCB-189). All metal and mercury analyses were performed by the Florida Department of Health, Bureau of Laboratory Services (Jacksonville, FL) using Inductively Coupled Plasma - Mass Spectrometry (ICPMS; EPA method SW 846-6020) and cold vapor atomic absorption (CVAA; EPA method 245.6), respectively. The majority of oyster samples was analyzed for dioxins/furans and PCBs by Triangle Laboratories, Inc. (Durham, NC). Crab samples and four oyster samples were analyzed for dioxins/furans and PCBs by Alta Analytical Perspectives (Wilmington, NC). Dioxins/furans and PCBs in all samples were analyzed by high-resolution gas chromatography coupled with high-resolution mass spectrometry (HRGC-HRMS) using U.S. EPA methods 1668

and 8290B, respectively. Quality assurance/quality control (QA/QC) measures included analysis of method blanks, duplicate samples, matrix spikes, and laboratory control samples or standard reference materials.

All chemical concentrations are reported on a wet weight basis. A conversion factor of 15% was used to compare concentrations to dry-weight values reported in the literature. For comparison, the concentration of a chemical found below the detection limit was analyzed as both zero (ND=0) and one-half the detection limit (ND=DL/2). Risk calculations were made using ND=DL/2 for contaminants in a sample that were below the detection limit. However, if a chemical was not detected in any sample for a given target species, it was assumed to not be present and thus was not evaluated. Duplicate samples from a specific location were averaged to obtain one concentration for that site.

Assessment of Tissue Contaminant Levels

Tissue contaminant levels were assessed using Screening Values (SV) based on the U.S. Environmental Protection Agency's (EPA) *Guidance For Assessing Chemical Contaminant Data For Use In Fish Advisories: Fish Sampling and Analysis* (U.S. EPA, 2000). The SVs are concentrations of chemicals in fish and shellfish tissue that are of potential public health concern and that are used as threshold values against which tissue levels of the contaminants can be compared (U.S. EPA, 2000). Exceedance of these SVs is an indication that more intensive site-specific monitoring and/or evaluation of human health risk should be conducted. Because a quantitative fish consumption survey has not been previously conducted in the Pensacola area, four different fish consumption rates (CR) were compared in the present study for the adult population. The CR values were based on EPA estimates of the average consumption of fish by

recreational fishers (17.5 g·day^{-1} uncooked weight) and subsistence fishers (142.4 g·day^{-1}), and of the average adult meal size (8 oz) if it was consumed weekly (32 g·day^{-1}). We also evaluated a CR estimate of 46 g·day^{-1}, which is based on the results of a fish and shellfish consumption study conducted throughout Florida by Degner et al. (1994).

Because of the unlikely scenario that an entire meal would consist of crab hepatopancreas alone, we also provide calculations that are based on the consumption of all edible tissues (body muscle, claw, and hepatopancreas). These estimates of contaminant levels in total edible crab tissue were based on the assumption that hepatopancreas accounts for less than 20% of total edible tissue in blue crabs (Tsai et al., 1984; NJDEP, 2002). To account for the proportions of tissue types, we used an estimate of 15% of total edible mass for hepatopancreas and 85% for muscle/claw. Therefore, estimates for whole crab were calculated as follows: $(C_{hep} * 0.15) + (C_{mus} * 0.85)$, where C_{hep} = concentration in hepatopancreas and C_{mus} = concentration in crab muscle.

Based on the EPA guidelines, a 70 kg body weight for adults, a 10^{-5} risk level for carcinogens, and a 70-year exposure duration were used (U.S. EPA, 2000). Cancer Slope Factors (CSF) and oral reference doses (RfD) used in the calculation of SVs were obtained from the U.S. EPA (U.S. EPA, 2000; U.S. EPA, 2002). Table 1 summarizes the SVs used in the present assessment and delineates the respective CSFs and RfDs used in the calculation. The EPA has suggested that in cases where both a carcinogenic and non-carcinogenic SV is available (e.g. arsenic), the lower of the two SVs (generally, the SV for carcinogenic effects) should be used for screening. We assessed the hazards posed by dioxins/furans and dioxin-like PCBs using World Health Organization Toxic Equivalency Factors (TEFs) (Van den Berg et al., 1998), which were summed for each sample to give a Toxic Equivalency Quotient (TEQ) and compared against the

derived SVs. In the present report, we use TEQ_{DFP} to refer to TEQs calculated using concentrations of both dioxins/furans and dioxin-like PCBs, TEQ_{DF} to refer to TEQs calculated for dioxins/furans only, and TEQ_P to refer to TEQs calculated for dioxin-like PCBs only.

Table 1. Tissue contaminant screening values (SV) based on a 70 kg body weight for adults, a 10^{-5} risk level for carcinogens, and 70-year exposure duration.

Chemical	RfD (mg/kg-day)	CSF (mg/kg-d)	SV Units	SV - Recreational[a]		SV - Subsistence[b]		SV - Florida[c]		SV - 8 oz.[d]	
				Non-Carcin.	Carcin.	Non-Carcin.	Carcin.	Non-Carcin.	Carcin.	Non-Carcin.	Carcin.
Arsenic (inorganic)	3.00E-04	1.5	$mg\,kg^{-1}$	1.2	0.027	0.15	0.003	0.46	0.010	0.66	0.015
Cadmium	1.00E-03	-	$mg\,kg^{-1}$	4	-	0.49	-	1.52	-	2.19	-
Chromium	3.00E-03	-	$mg\,kg^{-1}$	12	-	1.47	-	4.56	-	6.56	-
Mercury	1.00E-04	-	$mg\,kg^{-1}$	0.4	-	0.05	-	0.15	-	0.22	-
Nickel	2.00E-02	-	$mg\,kg^{-1}$	80	-	9.83	-	30.42	-	43.75	-
Selenium	5.00E-03	-	$mg\,kg^{-1}$	20	-	2.46	-	7.61	-	10.94	-
Tributyltin	3.00E-04	-	$mg\,kg^{-1}$	1.2	-	0.15	-	0.46	-	0.66	-
Zinc	3.00E-01	-	$mg\,kg^{-1}$	1200	-	147.47	-	456.32	-	656.25	-
Dioxins/Furans *	-	1.56E+05	$pg\,g^{-1}$	-	0.256	-	0.032	-	0.098	-	0.140

* Based on WHO-TEQ values and includes 12 dioxin-like PCBs.
[a] Based on consumption rate of 17.5 $g\,day^{-1}$.
[b] Based on consumption rate of 142.4 $g\,day^{-1}$.
[c] Based on consumption rate of 46 $g\,day^{-1}$.
[d] Based on consumption rate of 32 $g\,day^{-1}$.

Risk Calculations

To further evaluate the human health risks associated with consumption of shellfish species in the Pensacola Bay region, we determined the excess cancer risk (ECR) over a lifetime for each sample using the methods described by the U.S. EPA (2002). The potential cancer risk is estimated as an incremental increase in the probability of an individual developing cancer over a lifetime as a result of exposure to a carcinogen (U.S. EPA, 2002). The excess cancer risk for all carcinogens was summed to provide an estimate of total risk posed by exposure to multiple carcinogens. We also determined species-specific non-cancer hazard risks for each contaminant at each location (U.S. EPA, 2002). In these analyses, an exposure threshold is assumed to exist

below which adverse effects are unlikely to occur, and thus the average daily dose is compared to the reference dose to obtain a hazard quotient (HQ). A total Hazard Index (HI) is calculated by summing all HQ for a particular location across all health effects. If the HI exceeds 1, there is an indication of potential non-carcinogenic health effects, and the greater the magnitude above 1, the greater the level of concern (U.S. EPA, 2002). If the total HI was greater than 1.0, HQs for chemicals with similar target organs or health endpoints (mechanisms of toxicity) were summed to identify potential non-cancer effects (U.S. EPA, 2002). For this risk assessment, consumption rate and exposure duration were varied to estimate exposure under various scenarios. Consumption rates were as described above. Body weight for adults was assumed to be 70 kg, the average body weight for all adults in the general public (U.S. EPA, 2000; U.S. EPA, 2002). Exposure duration, the length of time over which exposure occurs, was assumed to be nine years (the median number of years individuals remain at one residence), 30 years (the national 90th percentile for the length of time an individual stays at one residence), or 70 years (lifetime exposure duration) for adults (U.S. EPA, 2000; U.S. EPA, 2002).