The geography of mercury and PCBs in North Carolina’s local seafood

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A B S T R A C T

Mercury and PCBs are used by non-governmental organizations and federal agencies to inform seafood safety recommendations. Pollution dynamics suggest recommendations on the national scale may be too large to be accurate. We tested softshell and hardshell blue crab, white and pink shrimp, oysters, clams, spot, and mullet from fishers in each of the three North Carolina fishery districts. We measured mercury using EPA method 7473 and PCBs using a commercially available ELISA kit. Over 97% of samples were below the Environmental Protection Agency levels of concern for both mercury and PCBs. Mercury and PCBs have different spatial dynamics, but both differ significantly by water body, suggesting that seafood safety recommendations should occur by water body instead of at the national scale. This finding supports previous research suggesting that differences in water chemistry, terrestrial influence, and flushing time in a particular water body control the contaminant load in locally resident species.

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1. Introduction

1.1. Seafood safety recommendations

Seafood safety recommendations by both the federal government and non-governmental organizations (NGOs) for commercial fisheries occur at a national scale. EPA and FDA recommendations are based on data sets collected from monitoring programs all around the country; the recommendation whether or not to eat seafood is based on a national average for contaminants (EPA, 2009). Currently EPA recommendations instruct pregnant women to avoid tilefish, shark, king mackerel, and swordfish (http://www.epa.gov/mercury/advisories.htm), and any other seafood that crosses the threshold of concern of 0.3 mg/kg of mercury or 50 μg/kg PCBs. Though based on the same data set, EPA and FDA recommendations often do not match because of different thresholds of concern.

Information on the toxicity of seafood is also communicated by seafood pocket guides issued by NGOs including the Monterey Bay Aquarium and Food and Water Watch. Aiming to protect consumer health through education, these guides flag seafood types with high levels of mercury and PCBs as health concerns regardless of their stock status or fishing methods. At the start of this research (summer 2009), these cards listed two of North Carolina's largest commercial fisheries – blue crabs and oysters – as having unsafe levels of both mercury and PCBs (above 0.3 mg/kg mercury or 12 μg/kg PCBs for Food and Water Watch). These cards were based upon data from EPA and individual research reports gathered by Environmental Defense that had few sampling sites within North Carolina estuaries (Fitzgerald, pers. comm.).

NGO pocket guides have been criticized for lack of attention to “traceability” back to the origin of the seafood (Jacquet and Pauly, 2007). In addition, the guides emphasize single-species traits while wider ecosystem concerns are not addressed (Jacquet and Pauly, 2007). Environmental toxins are indicated on the cards but are not factored into overall ranking. The cards reflect little on the ecosystems and communities providing the seafood. However, these guides affect consumer purchasing patterns, causing decreased demand for foods deemed unsustainable (Roheim, 2009).

Consumer awareness of seafood toxins is less clear and there is confusion by consumers in how to respond to mercury and other contaminant data (Hicks et al., 2008). Two-thirds of women of childbearing age were found to be unaware of local mercury advisories (Knobeloch et al., 2005), but pregnant women have decreased their consumption of all fish, not just those in the EPA advisory (Oken et al., 2003). Some of this confusion may come from the fact that the science is relatively new, conflicting, and recommendations may differ by region.

Boundaries of both federal and NGO recommendations are based on political jurisdictions rather than environmental variability at the watershed, water body, or stream scale. North Carolina watersheds, such as the Albemarle-Pamlico, each contain a large portion of the coast and are composed of smaller water bodies (sounds or river deltas) that are each fed by multiple tributaries. Recent marketing efforts from fishermen in conjunction with the growing local food movement also function at these smaller scales.
these programs were concerned that the national recommendations may not apply to their catch (Mosher, NCSG, pers. comm.).

1.2. Why mercury and PCBs?

Mercury and PCB levels represent a direct link between source and exposure that simplifies risk assessment for consumption recommendations. Mercury and PCBs often co-occur (Wehie et al., 1996), compounding toxicity and complicating measures of human health effects (Grandjean et al., 2001). In Faroe Islanders, daily average mercury consumption by mothers of 36 µg and PCB consumption of 200 µg led to neurological defects in children, mediated primarily by a PCB-induced decline in dopamine levels (Wehie et al., 1996). Mercury impairs neurological development and causes impaired vision, muscle impairment and lack of coordination, and ‘pins and needles’ feeling in hands, feet, and mouth (EPA, 2010a). PCBs are primarily carcinogens and environmental steroids that cause immune system suppression, decreased fertility, reduced birth weight, learning deficits, and decreased thyroid function (EPA, 2010b).

Seafood safety recommendations, especially seafood cards produced by nongovernmental organizations, focus on mercury and PCB content in edible tissue as an indicator of overall contamination (Lazorchak et al., 2003). Mercury and PCBs are especially important because seafood represents the sole path of exposure for most consumers (Grandjean et al., 1995). Studies of mercury and PCBs in seafood from specific locations focus on recreational catches, though the majority of seafood eaten comes from commercial fisheries (Burger et al., 2005).

Though less studied, effects on wildlife or ecology may be equally or more profound than human health impacts. Wall et al. (2001) found decreased seagrass biomass, fungal biomass, and grass shrimp fecundity in a site contaminated with mercury and PCBs. When combined with documented shifts in trophic structure, these ecosystem changes facilitate trophic transfer of contaminants into fishery species (Wall et al., 2001). Sediments contaminated with a mix of toxins including PCBs decrease macrobenthic diversity and shift trophic structure, decreasing overall productivity (Brown et al., 2000). PCBs also cause acute damage to fauna, such as atrophied digestive epithelium and degenerated connective tissue in oysters (Lowe et al., 1972). Mercury causes acute mortality in wetland organisms, and residue-effect relationships are good indicators of ecological damage (Zillieux et al., 1993). Mercury, PCBs, and DDT are the most common contaminants, explaining most of the observed toxicity in the nation’s estuaries (Kamman et al., 2005). Therefore, levels of these chemicals in small mid-trophic level fish serve as a good indication of overall contaminant effects (Lazorchak et al., 2003).

1.3. Hypothesis

We use mercury and PCBs to inform the spatial scales of seafood consumption advisories in NC. Given coastal North Carolina’s few point sources and primarily rural population, contaminant levels are expected to be low compared with other inshore fisheries. We chose softshell and hardshell crab, pink shrimp, white shrimp, clams, oysters, mussels, and spot as study tissue types because they are relatively sedentary and their toxin loads should reflect levels in a given location. We hypothesize that national predictions are not reliable predictors of contaminant level in localized fisheries. Since both mercury and PCB levels vary by water body due primarily to water chemistry and upstream inputs (Hillery et al., 1997; Kamman et al., 2005), we expect that there will be spatial differences between water bodies within the state.

2. Methods

2.1. Sampling and tissue preparation

Sampling occurred within the commercial fishing season for each species from July 2010 to June 2011. Five individuals of each of 8 types of edible seafood tissue were collected at 5 locations in each of the 3 fishing regions of North Carolina (North, Central, and South) for a total of 600. Edible tissue refers to meat taken from the seafood as if prepared for cooking – fillets for the finfish, whole but shelled bodies for the bivalves and shrimp, whole cleaned bodies for softshell crabs, and picked backfin and leg meat for hardshell crabs. Seafood was donated or purchased from retailers who could identify the fishing location and were therefore within the range of market sizes for each species.

Individual sampling locations identified in Figs. 3a and 3b represent a fisher’s trawl, trap line, or collection site where the samples were collected as marked by GPS or marked by hand on a printed map. Fishers were instructed to provide samples from their usual fishing locations where they consistently have good fishing conditions. Some fishers provided multiple species for study from the same location, depending on gear types available. For example, shrimpers often caught blue crabs and spot in their nets simultaneously; these were all recorded as one sampling location when the net was pulled up. In order to test differences between larger-scale variables, other location information was recorded. Port town, where fishers dock at the end of the day to sell or transport their product, is an approximation of the socioeconomic community associated with each sample location. Water body is recorded as an approximation of the biogeographic community, referring to the larger sound or river delta from which the sample was sourced. Both of these metrics served to investigate an intermediate scale between individual sampling location and the state’s three designated fishery regions.

Samples were immediately frozen at −20 °C. Frozen samples were later thawed and shelled or filleted to separate the edible parts. Tissue from spot, mullet, blue crabs, and softshell crabs were homogenized in a small electric chopper (Black & Decker #HC306). Shrimp, clam, and oyster tissues were too small to be homogenized in the electric chopper, so were minced with an acid-washed scalpel and homogenized by hand. About 10 g of each homogenized tissue was placed in a glass vial (Fisherbrand #03-339-26G), frozen at −80 °C for at least an hour, and lyophilized for 48 h. Wet: dry ratios were determined by weighing 5–10 of these samples of each type of tissue before and after drying. Freeze-dried samples were ground to a fine powder with an acid-washed 3 mm diameter glass stir-rod prior to determination of mercury and PCB levels.

2.2. Mercury

Mercury content was determined by thermal decomposition, amalgamation, and atomic absorption spectroscopy on a Milestone DMA-80 analyzer according to EPA method 7473 (USEPA, 1998) using 0.04–0.12 g of freeze-dried and powdered sample. A standards curve was created by adding 0, 25, 50, 100, 150, 200, and 400 ng of mercury in the form of HgNO₃ to approximately 0.1 g soy flour. Certified reference standards TORT-2 (National Research Council – Canada), DOLT-3 (National Research Council – Canada), and soy flour with 200 ng mercury as HgNO₃ were used to confirm standards curve. Duplicate samples varied by an average of 5%.

2.3. PCBs

PCBs were extracted from 0.09 to 12 g of freeze-dried seafood tissue by column extraction (Lassado et al., 2003). Columns were
made from Pasteur pipets plugged with glass wool and then layered with 0.4 mL of 60/40 silica gel/sulfuric acid followed by 0.1 mL of 70/30 silica gel/sulfuric acid, each dried overnight at 180 °C. Tissue was then placed in the column and extracted with 2 mL hexane. Eluent was then evaporated with nitrogen gas until dry and frozen at −20 °C until Enzyme-Linked Immunosorbent Assay (ELISA) could be performed.

Total PCB content was determined by ELISA using kits obtained from Sdix (Newark, DE). One Hundred microlitres of methanol and 100 μL of diluent from the kit were used to dilute the extracts before analysis. A standard curve was created for analysis using 2.5, 1, and 5 ppm standard solutions of PCBs dissolved in methanol. Samples with concentrations exceeding the range of the standard curve were re-run using 0.3–0.5 g of dry tissue. Duplicate samples varied by an average of 21%.

2.4. Lipid determination

Lipid concentration was determined gravimetrically for a subset of the tissue samples. 0.2 mL, or 10% by volume, of the eluent was taken before evaporation by nitrogen and weighed on a tared aluminum weighing boat. Following a 30 min evaporation period, dried lipid was reweighed to calculate lipid content.

2.5. Estimates of mercury and PCB recovery

Spike-recoveries for PCBs were performed on 45 paired tissue samples, 5–6 samples per tissue type. One nanogram of PCBs dissolved in 200 μL methanol was added to freeze-dried seafood tissue prior to extraction alongside paired non-spiked samples. Spike recoveries for mercury were performed on 40 paired tissue samples, 5 per tissue type. Two hundred nanograms of mercury in the form of HgNO3 was added to freeze-dried tissue prior to analysis. Duplicate samples for both the mercury and PCB analysis were run with each batch of samples analyzed. Spike-recovery averaged 95% for mercury and 120% for PCBs.

2.6. Statistics

Mercury and PCB data were normally distributed and tests of significance were determined by one-way ANOVA followed by Tukey’s multiple comparisons test using the R statistics package (R Foundation for Statistical Computing, Vienna, Austria 2001). Differences were considered significant at the p = 0.05 level.

3. Results

Since the motivation for this study focused on the geography at which seafood safety recommendations can be made, we tested the significance of various spatial scales. Individual fishing routes or shellfish locations were significantly different from one another (p < 0.001 for both mercury and PCBs). Port town, where the fishers docked to sell their catch at the end of each day, was also significantly different from one another for mercury (p < 0.001) but not for PCBs (p = .2116). Water body, which references the various Sounds that punctuate the North Carolina coast and a few river deltas, was the most significantly different (p < 0.001 for mercury and p = .0055 for PCBs, see levels by water body for each species in Figs. 1a and 1b. Notably, the state’s three designated fishing regions (north, central, and south) were also different from one another for mercury (p < 0.001) but not for PCBs (p = .1909) (see Fig. 2). Overall the only spatial scale that elucidated the variation in a statistically significant way for both contaminants was water body. In addition, as shown in Figs. 3a and 3b, mercury had higher spatial variation overall. All but 14 samples were below the EPA recommendations for both mercury and PCB content.

The state averages for mercury in all tissue types ranged from 0.016 mg/kg in pink shrimp to 0.056 mg/kg in spot, placing them far below the state and national thresholds of concern at 0.3 mg/kg (EPA), 0.4 mg/kg (NC DHHS), or 1 mg/kg (FDA). The tissue types were significantly different from one another (p < 0.001) (see Figs. 1a and 1b).

State averages for PCBs ranged from 1.45 μg/kg in pink shrimp to 10.55 μg/kg in spot, far below state and federal thresholds of health concerns at 50 μg/kg (EPA, NCDHHS) or 2000 μg/kg (FDA). Like mercury, the tissue types differed significantly from one another (p = .008), with wider variation both within species and within individual sample locations (see Fig. 4b). Mercury and PCB concentrations also covaried (p = .0062), indicating that locations that are high in mercury tend to be high in PCBs and vice versa. Mercury and PCBs also did not correlate with lipid content (see Fig. 5).

4. Discussion

4.1. Spatial scale considerations

The spatial dynamics of mercury and PCBs are different. At all spatial scales, mercury levels differed, suggesting extremely localized movement and methylation of mercury that could be differentiated in individual fishing locations. Levels of PCBs were more uniform across the entire state, but still differed by water body likely due to upstream contributions. The differences in dynamics between mercury and PCBs suggest there are many processes at work determining contaminant levels that may not yet be fully described but that water body is the best spatial scale to identify differences.

Although mercury and PCB levels co-occur, there were sites that had high mercury and low PCBs and vice versa. Such cases can be explained from known local pollution events, such as the Hewlett’s Creek sewage spill. Since sewage treatment effluent is known to be a significant source of PCBs but not mercury (Samara et al., 2006), elevated levels of PCBs but not mercury are expected. Similarly, areas with high mercury and low PCBs are near areas of disturbed peat land or chloralkali production, both of which are expected to increase mercury in the surrounding environment. Because the association of the two chemicals is uncoupled by localized sources, use of additional indicators for seafood safety would improve assessment of overall contamination.

Previous studies of the spatial heterogeneity of mercury contamination between Newfoundland and New York revealed that mercury varied by water body, dependent on water body acidity and watershed size (Kamman et al., 2005). Similar studies of lakes in Wisconsin show that water chemistry in lakes was a stronger determinant of mercury concentration in perch than many other commonly used variables such as trophic position and latitude (Greenfield et al., 2011). PCBs also are primarily related to distance to original source (Meijer et al., 2003) and areas of high atmospheric deposition (Dierking et al., 2009). Patchy PCB distribution depends on proximity to urban development and meteorological conditions (Hillery et al., 1997).

The developed Southern region of the state (around the city of Wilmington) had the highest levels of both mercury and PCBs due to historic contamination, though all regions were on average below federal health concern thresholds. Other individually elevated samples were proximate to historic pollution events or industrial sites upstream. Therefore, our North Carolina data add to a growing body of evidence (Hillery et al., 1997; Kamman et al., 2005) that water body should be the spatial scale of concern.
due to chemical variables and that these ecological boundaries should be used to create seafood safety guides. A spatially-explicit approach should be replicated in other locations to determine if this is a national or global trend. For North Carolina, scaling down recommendations can occur through tailoring recommendations to local food marketing efforts (such as Outer Banks Catch, Carteret Catch, and Brunswick Catch) or to state fishery management plans that address each water body individually.
4.2. NC seafood is safe to eat

Despite low trophic-level, small body size, and relatively short life cycles (all of which are associated with low toxin levels), blue crabs and oysters were flagged for high toxin levels on national and regional seafood pocket guides. In North Carolina, however, average mercury and PCB levels for these and other commercially important species were ten times lower than the federal and state

Fig. 1b. PCB levels by water body for each tissue type. Graphs are ordered by increasing mercury content and bars within each graph from North to South. The asterisk indicates that the bar for clams in Masonboro Inlet extends to 84 μg/kg but was truncated to have consistent axes. Bars represent standard error and letters above the bars represent statistical significance; graphs without letters above the bars were not statistically significant for the water body scale.

4.2. NC seafood is safe to eat

Despite low trophic-level, small body size, and relatively short life cycles (all of which are associated with low toxin levels), blue crabs and oysters were flagged for high toxin levels on national and regional seafood pocket guides. In North Carolina, however, average mercury and PCB levels for these and other commercially important species were ten times lower than the federal and state
thresholds of concern. This means that at least for oysters and blue crabs, the North Carolina catch is cleaner than the national average. The pattern of contamination found in this study (Figs. 4a and 4b) is likely most related to trophic position, as levels of mercury and PCBs did not correlate to differences in lifespan or lipid content (Fig. 5).

Eastern North Carolina is largely a sparsely populated, rural area, so contaminant loads lower than the national average are not surprising (Street et al., 2005). Impacts from development and industry are not contributing to contaminant load as much as in other regions of the country supplying the same seafood. The presence of low-level mercury and PCB contamination is likely from diluted upstream contributions and aerial deposition from distant sources such as coal-fired power plants; this represents no health concern to humans eating the tested seafood types but is a vector for bioaccumulation in higher trophic level fish. Spot

Fig. 2. State fisheries regions. Bars represent standard error. Letters above the bars represent statistical significance; bars sharing a letter are not significantly different from one another.

Fig. 3a. Geographic variation in mercury content in estuarine commercial fisheries. Darker points indicate more mercury (black over the EPA limit of 0.3 mg/kg), lighter less.
and mullet also migrate offshore, serving as a trophic base elsewhere and another vector for bioaccumulation.

In North Carolina, natural sources contribute to mercury load. The peat bogs that dominate the landscape sequester mercury then release it during disturbance such as fire or land clearing for agriculture; leaching rate is correlated to riverine dissolved organic carbon (Zillioux et al., 1993). Two-thirds of the ambient bioavailable mercury is emitted from coal-fired power production, most

Fig. 3b. Geographic variation in PCB content in estuarine commercial fisheries. Darker points indicate more PCBs (black above the EPA limit of 50 μg/kg), lighter less.

Fig. 4a. State averages for mercury. Bars represent standard error and letters above the bars indicate statistical significance; bars sharing a letter are not significantly different. Recommendations are not within the scale of this chart: 0.3 mg/kg for EPA and EDF, 1 mg/kg for FDA.
notably in China, which is atmospherically upwind of the United States (Lindberg et al., 2007). Global and natural sources of mercury make contamination levels in a particular location difficult to predict (Lindberg et al., 2007), making control of mercury pollution dependent on controlling distant anthropogenic sources (Evans, 1986).

Similar challenges arise when investigating PCB levels. Before the 1979 ban on PCB production, PCBs were ubiquitous, found in almost all electronics and transformers. Modern sources stem largely from former productions sites, disposal sites, waste incineration (Murphy et al., 1985), and wastewater treatment plants (Samara et al., 2006). For North Carolina, a controversial landfill placed in upstream Warren County leaches PCBs over time (Bullard, 1992) into the northern region, the wastewater treatment plant in Morehead City contributes to downstream PCB contamination, and a spill in Hewlett's Creek (southernmost red point, Figs. 3a and 3b) in the Southern region continues to deliver PCBs into the nearby Masonboro Sound (Alphin, UNCW, pers. comm.); these cases explain the 8 of the 14 instances exceeding EPA level of concern. Three others exceeding mercury standards originated near a peatland drainage documented to leach mercury; there are no documented local pollution sources for the final 3 samples exceeding EPA standards.

North Carolina is committed to protecting the natural shoreline through the Coastal Habitat Protection Plan and commercial fisheries play a prominent role in state policy, led by powerful coastal representatives in the state's general assembly. Their work likely also contributes to the comparatively low contaminant levels by favoring more environmentally-friendly development and industrial regulation. The seafood we tested (all short-lived, low-trophic level), with the exception of 14 samples of the total 600, is safe to eat according to both federal and non-governmental organization recommendations. These findings parallel similar work in offshore species in North Carolina (Petre et al., 2012) showing the state may be a more accurate level of analysis than the region or nation. In this case, national recommendations falsely identified two of North Carolina's biggest fisheries as contaminated and likely had a negative impact economically since consumers have been shown to alter their purchasing based on these recommendations (Roheim, 2009). This could easily be avoided in the future.

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Fig. 4b. State averages for PCBs, ordered by increasing mercury concentration for comparison. Bars represent standard error and letters above the bars indicate statistical significance; bars sharing a letter are not significantly different. Recommendations are not within the scale of this chart: 12 µg/kg for EDF, 50 µg/kg for EPA, 2000 µg/kg for FDA.

Fig. 5. Lipid content in the tissue types, shown in the order of increasing mercury content. Bars indicate standard error and letters above the bars indicate statistical significance. Bars sharing a letter are not significantly different from one another.
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