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Mercury and selenium levels in 19 species of saltwater fish from New Jersey as a function of species, size, and season

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ABSTRACT

There are few data on risks to biota and humans from mercury levels in saltwater fish. This paper examines mercury and selenium levels in muscle of 19 species of fish caught by recreational fisherfolk off the New Jersey shore, as a function of species of fish, size, and season, and risk of mercury to consumers. Average mercury levels ranged from 0.01 ppm (wet weight) (Menhaden *Brevoortia tyrannus*) to 1.83 ppm (Mako Shark *Isurus oxyrinchus*). There were four categories of mercury levels: very high (only Mako), high (averaging 0.3–0.5 ppm, 3 species), medium (0.14–0.20 ppm, 10 species), and low (below 0.13 ppm, 5 species). Average selenium levels for the fish species ranged from 0.18 ppm to 0.58 ppm, and had lower variability than mercury (coefficient of variation = 38.3 vs 69.1%), consistent with homeostatic regulation of this essential element. The correlation between mercury and selenium was significantly positive for five and negative for two species. Mercury levels showed significant positive correlations with fish size for ten species. Size was the best predictor of mercury levels. Selenium showed no consistent relationship to fish length. Over half of the fish species had some individual fish with mercury levels over 0.3 ppm, and a third had fish with levels over 0.5 ppm, levels that pose a human health risk for high end consumers. Conversely several fish species had no individuals above 0.5 ppm, and few above 0.3 ppm, suggesting that people who eat fish frequently, can reduce their risk from mercury by selecting which species (and which size) to consume. Overall, with the exception of shark, Bluefin Tuna (*Thunnus thynnus*), Bluefish (*Pomatomus saltatrix*) and Striped Bass (*Morone saxatilis*), the species sampled are generally medium to low in mercury concentration. Selenium:mercury molar ratios were generally above 1:1, except for the Mako shark.

1. Introduction

For many coastal states and countries, saltwater fishing is an important commercial, recreational and subsistence activity. High fishing rates (days per year) occur in a wide range of cultures, including in rural and urban areas (Burger et al., 1999, 2001a, b; Bienenfeld et al., 2003), among Native Americans (Burger et al., 2007; Harper and Harris, 2008), and in other regions of the world (Burger et al., 2003). Fish provide fishmeal for human and aquaculture use (Brunner et al., 2009), and recreational, cultural and aesthetic pleasures (Toth and Brown, 1997; Burger, 2000, 2002). They also contain protein and valuable nutrients including polyunsaturated fatty acids and selenium.

However, levels of methylmercury (MeHg) and other contaminants in some fish are high enough to potentially cause effects on the fish themselves, on top-level predators, and on people (WHO, 1989; EPA, 1997; NRC, 2000; Consumer Reports, 2003). Consumption of mercury-contaminated fish came to attention after the outbreaks at Minamata and Niigata, Japan in the 1950s and 1960s (Harada, 1995). Fish consumption is the only significant source of methylmercury exposure for the public today (Rice et al., 2000), although historic epidemics attributed to grain seed treated with organomercurial fungicides occurred, most notably in Iraq in 1973 (Amin-zaki et al., 1978), and some mercury enters the food chain from mining (Qiu et al., 2009). Mercury occurs naturally in seawater, and coastal waters receive mercury runoff from land, input from rivers, and airborne deposition. Biomethylation of mercury occurs in sediment, allowing for food chain biomagnifications (Downs et al., 1998; Morel et al., 1998). Mercury in fish tissue may be six orders of magnitude higher than the mercury concentration in the water column (Scudder et al., 2009).

Levels of methylmercury are sufficiently high in some fish to cause adverse health effects in people consuming large quantities (Institute of Medicine, 1991, 2006; Grandjean et al., 1997; Gochfeld, 2003; Hightower and Moore, 2003; Hites et al., 2004), with neurodevelopmental effects from fetal exposure the most prominent effect

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(Amin-zaki et al., 1978; Crump et al., 1998; Steuerwald et al., 2000; NRC, 2000). Prenatal methylmercury has led to behavioral deficits in infants (JECFA, 2003) and to poorer cognitive test performance (Oken et al., 2008). Methylmercury can counteract the cardioprotective effects of fish consumption (Guallar et al., 2002; Rissanen et al., 2000; Salonen et al., 1995). Thus, communities that rely on fish intake for daily nutrient sustenance may be at risk from chronic, high exposure to methylmercury (Grandjean et al., 1997), as well as other persistent organic pollutants. Hughner et al. (2008) estimated that 250,000 women may be exposing their fetuses to levels of methylmercury above federal health guidelines. Similarly, high-end fish consumers, whether recreational or subsistence, are at risk from mercury exposure (Hightower and Moore, 2003; Lowenstein et al., 2010).

The U.S. Food and Drug Administration (USFDA, 2001) issued consumption advisories based on methylmercury that suggested that pregnant women and women of childbearing age who may become pregnant should limit their fish consumption, should avoid eating four types of marine fish (shark, swordfish, King Mackerel, and Tilefish), should also limit their consumption of all other fish to just 12 oz (=342 g) per week (USFDA, 2001), and there are recent warnings about canned white tuna (USFDA/USEPA, 2004a). These are all saltwater fish, while most studies of mercury levels have focused on freshwater fish (Legrand et al., 2005).

In freshwater fish, variations in water pH can account for up to 70% of the variation in mercury levels (Watras et al., 1998). Microbial methylation of mercury is favored by anaerobic conditions and low dissolved oxygen (DOC, Regnell, 1994). Much of the data dealing with the effects of fish size on mercury levels comes from freshwater fish (Simonin et al., 2008). Yet for many coastal states, consumption of saltwater fish is an important potential source of mercury exposure that has been largely ignored until recently. Fish are an important dietary item of the people living along coastal New Jersey, and recreational fishers often freeze fish for consumption at all times of the year (Pottern et al., 1989; Burger, 2005; Gobeille et al., 2005). It is therefore important to understand how to reduce the risk from mercury, and to provide the public with information on fish that are low in mercury (as well as high). Although Burger et al. (2009) examined mercury in flatfish that had relatively low levels, there is a need for a broader spectrum analysis of marine fish from one general geographical area.

Fish are an excellent, low-fat source of protein that contributes to low blood cholesterol, to positive pregnancy outcomes, and to better child cognitive test performances (Oken et al., 2008). Fish contain omega-3 (n-3) fatty acids that reduce cholesterol levels and the incidence of heart disease, stroke, and pre-term delivery (Daviglus et al., 2002; Patterson, 2002; Virtanen et al., 2008). Further, fish, particularly oceanic fish, are relatively rich in selenium, necessary for seleno-enzyme functions, and selenium has long been known to offer some protection against mercury toxicity. The public thus must choose whether to eat fish, what species to eat, as well as what size fish, what size portions, and how often. Sound choices require adequate information about a range of fish. There is some indication that the recent FDA warnings about fish consumption (USFDA, 2006), focusing on species high in mercury, have resulted in a reduction in the consumption of fish generally, and of canned fish specifically (Shimshack et al., 2007), while some authors argue that the advantages of fish consumption outweigh the mercury risk (Mozaffarian, 2009). Information on species low in mercury would be advantageous.

In this paper we examine levels of mercury in a wide range of fish species from coastal New Jersey to provide information that can be used to evaluate the potential risk to the fish themselves, to their predators, and to humans who consume them. Unlike many studies, we did not focus only on those species expected to have high levels (and thus pose the greatest risk), but examined levels in the wide range of fish caught by local recreational fishermen. Too often levels of mercury are provided for fish that people should avoid, without providing information on species that are low in mercury (and thus provide little risk). Risk balancing by the public is possible when mercury levels are available for a range of fish. The fishers requested this information after media coverage of mercury in fish, and worked with us on providing the fish samples.

Levels of selenium were analyzed because selenium offers some protection against mercury exposure (Satoh et al., 1985; Ralston, 2009; Lémire et al., 2010), lower levels of nonfatal heart attacks have been associated with higher levels of selenium (Mozaffarian, 2009), and some recent studies with animal models have suggested that some (if not most) of the adverse impacts of high methylmercury exposure occur as a result of mercury's impairment of selenium-dependent enzyme activities (Watanabe et al., 1999a; Ralston, 2008, 2009; Ralston et al., 2008). Park and Mozaffarian (2010) reported evidence that although fish consumption substantially reduced cardiovascular risk, clinical trials demonstrated mixed and inconclusive results for cardiovascular effects of methylmercury and selenium. Ralston and others (Ralston, 2008; Peterson et al., 2009) have argued that selenium:mercury molar ratios above 1 are protective for adverse mercury affects. However, the interaction between selenium and mercury is complex and warrants continued examination. There are several issues that need further examination, but are not within the scope of this paper, including whether selenium merely chelates mercury keeping it from attacking disulfide bonds, whether mercury creates a relative selenium deficiency or inactivates essential seleno-proteins, and what other endogenous and exogenous factors influence the interaction. Ralston (2008, 2009) suggests that the molar ratio is the key value (rather than the level of methylmercury) for risk assessment.

2. Methods

Fish of 19 species were collected (2003–2008) from several sites along the New Jersey shore (Fig. 1; scientific names found in Table 1), mainly from recreational fisherfolk, who were either fishing individually or were taking part in fishing tournaments. Most of the actual sampling, however, was done by our personnel who went to local docks and fishing sites to meet fisherfolk. The 19 species are the fish most often caught by N.J. fishermen, and were selected because they are most relevant to recreational fishermen in the region. The project was a collaboration with local fishing clubs (Jersey Coast Anglers Association, Jersey Shore Shark Fishermen) and others, who greatly influenced the species collected. In many coastal regions there are a number of fishing tournaments that focus on Bluefish, Striped Bass, and Mako (all Shortfin Mako). Fish from tournaments were either taken home for consumption by the families of the fishermen, or were donated to orphanages or other facilities. We obtained either whole fish, or took an approximately 50 g sample plug biopsy from the side of the fish, over the lateral line just anterior to the tail. In addition, we obtained small individuals (below the recreational size limits) of some species (bluefish and striped bass) collected by the NJ Department of Environmental Protection trawls. Data on the entire size range are provided for comparison with other studies that concentrated on fish biology, rather than risk to fish consumers.

Fish or samples were kept in coolers and brought to the Environmental and Occupational Health Sciences Institute (EOHSI) of Rutgers University for element analysis. However, all samples were run with standard calibration curves by the same laboratory chemist to avoid any variations. All fish were analyzed individually for total mercury during the last two years of the study. At EOHSI, a 2 g (wet weight) sample of skinless fish muscle was digested in 4 ml of Fisher Scientific Trace metal grade nitric acid and 2 ml deionized water in a microwave (MD 2000 CEM), using a digestion protocol of three stages of 10 min each under 50, 100 and 150 lbs/in.^{2} (3.5, 7, and 10.6 kg/cm^2) at 80% of total power. Digested samples were subsequently diluted to 25 ml with deionized water. The same digestion methods were used for both mercury and selenium. All laboratory