

Potential Moderating Effects of Selenium on Mercury Uptake and Selenium:Mercury Molar Ratios in Fish From Oak Ridge and Savannah River Site - 12086

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ABSTRACT

Mercury contamination is an important remediation issue at the U.S. Department of Energy's (DOE) Oak Ridge Reservation and to a lesser extent at other DOE sites because of the hazard it presents, potential consequences to humans and eco-receptors, and completed pathways, to offsite receptors. Recent work has emphasized that selenium might ameliorate the toxicity of mercury, and we examine the selenium:mercury (Se:Hg) molar ratios in fish from Oak Ridge, and compare them to Se:Hg molar ratios in fish from the Savannah River. Selenium/mercury molar ratios varied considerably among and within fish species. There was considerable variation in the molar ratios for individual fish (as opposed to mean ratios by species) for freshwater fish from both sites. The inter-individual variation in molar ratios indicates that such that the molar ratios of mean Se and Hg concentrations may not be representative. Even for fish species with relatively low mercury levels, some individual fish have molar ratios less than unity, the value sometime thought to be protective. Selenium levels varied narrowly regardless of fish size, consistent with homeostatic regulation of this essential trace element. The data indicate that considerable attention will need to be directed toward variations and variances, as well as the mechanisms of the interaction of selenium and mercury, before risk assessment and risk management policies can use this information to manage mercury pollution and risk. Even so, if there are high levels of selenium in the fish from Poplar Creek on Oak Ridge, then the potential exists for some amelioration of adverse health effects, on the fish themselves, predators that eat them, and people who consume them. This work will aid DOE because it will allow managers and scientists to understand another aspect that affects fate and transport of mercury, as well as the potential effects of methylmercury in fish for human and ecological receptors. The variability within fish species, however, suggests that the relative Se:Hg molar ratios in fish are not stable enough to be used in risk assessment at this time. Nor is it known how much excess selenium is required to confer any degree of protectiveness. That is, in conducting risk assessments, it is not possible to determine the spread of ratios, which would be needed for probabilistic risk assessment. Significantly more fish samples per species are required to begin to generate data that would allow it use in risk assessment. Adding Se:Hg molar ratios seems to complicate risk assessment for the potential adverse effects of mercury exposure, and using mercury levels at this time remains the most viable option.

INTRODUCTION

Mercury is one of the most important contaminants of concern at contaminated sites, including “Superfund” sites on the National Priorities List [1]. Mercury, derived from industrial processes or from waste generated from coal-fired power plants, is also a significant contaminant at many of the U.S. Department of Energy’s (DOE) sites. The Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA) requires that the National Oil and Hazardous Substances Pollution Contingency Plan develop a National Priorities List (NPL) of sites with known or threatened releases of hazardous substances, pollutants, or contaminants throughout the United States [2]. Further, the top three ranked priority hazardous substances at Superfund Sites in terms of potential toxicity are arsenic, lead, and mercury [1]. Thus there is a regulatory driver, as well as concern related directly to protection of human health and the environment.

The large quantity of mercury contamination, including ongoing release to offsite surface water, providing completed exposure pathways to eco-receptors and humans, makes the issue of mercury particularly critical at DOE’s Oak Ridge Reservation. Mercury contamination in and under the Y-12 buildings [3-4] is a source of ongoing release. Although there has been a substantial decrease in mercury inputs to East Fork Poplar Creek (E FPC) due to active remediation, interdiction, and stream flow augmentation [5], some fish still have levels above the Environmental Protection Agency’s (EPA) freshwater criterion of 0.3 ppm in fish tissue. Similarly, other DOE sites, such as Brookhaven and the Savannah River Site have mercury contamination [6-7].

The relationship between mercury and selenium in selected fish from Oak Ridge and from the Savannah River, adjacent to the Savannah River Site, is examined to determine the selenium:mercury molar ratios. Selenium has a protective effect on mercury toxicity. Although the amount of selenium needed to confer protection is unknown, it has been suggested that a Se:Hg molar ratios a greater than unity, may be sufficient in some cases to prevent mercury toxicity [4, 8-12]. Two questions are addressed: 1) what are the Se:Hg molar ratios in selected fish from Oak Ridge and the Savannah River Site, and 2) what is the variation within and among fish species? Further information on mercury and other metals in fish from Oak Ridge and Savannah River is provided in other papers [6,13].

Background on mercury

Mercury is one of the oldest contaminants known to have a detrimental effect on humans, and methylmercury has the greatest effect on the health of vertebrates, including humans [14]. The main source of exposure to methylmercury in humans and other vertebrates is from fish consumption [14]. Fish provide high quality protein as well as nutrients, yet many Americans are faced with deciding whether the benefits of eating fish outweigh the risks from contaminants [15]. Fish are a low-fat source of protein that contributes to low blood cholesterol [16], positive pregnancy outcomes, better child cognitive test performances [17], incidence of heart disease, blood pressure, stroke, and pre-term delivery [18-19].

Levels of methylmercury (MeHg) and other contaminants in some fish are high enough to cause effects on the fish themselves, and on top-level predators, including humans [20-22]. Effects of high mercury exposure in humans include neurodevelopmental deficits [21], behavioral deficits in infants [23], poorer cognitive test performance [24], promotion of cardiovascular disease [25], and neurological and locomotary deficits [26]. People with high fish intake may be at risk from chronic, high exposure to methylmercury [27]. Wildlife are similarly affected by high levels of mercury [28], especially piscivorous birds [29]. Risk

management of mercury toxicity from fish involves source reduction, physical remediation and restoration, dilution, and blocking the pathway to receptors (through physical or behavioral means), as well as the potential for moderating toxicity because of selenium levels.

Background on selenium and mercury toxicity

Selenium plays a protective role against mercury toxicity with uncertainty about both mechanisms and the protection conferred by different molar ratios [8-9]. Selenium-mercury interaction may reduce the bioavailability or toxicity of methylmercury, and conversely some mercury toxicity may be due to impaired selenium-dependent enzyme synthesis or activity [8-9, 30-31]. Mercury and methylmercury are irreversible selenoenzyme inhibitors [32] that impair selenoprotein form and function. Mercury binds to selenium with a high affinity, and high maternal exposure inhibits selenium-dependent enzyme activity in the brain [33].

The general applicability of using the molar ratio of selenium:mercury, rather than concentrations or intakes of mercury in risk assessment and risk management is an important management and policy issue. Understanding how much selenium is protective for mercury toxicity and their mutual bioavailability, needs further research. Knowledge of the Se:Hg ratios in fish consumed by eco-receptors and people, and understanding the variation within and among fish is critical to evaluating the potential effect of mercury and assessing risk. This is particularly true for DOE sites where mercury contamination is still above the EPA freshwater criterion. Understanding the Se:Hg ratios in fish that are consumed by people will allow risk assessors and DOE to understand whether the high levels in EFPC and elsewhere are less detrimental than thought on the basis of the mercury levels alone. While DOE regularly monitors mercury concentrations in fish at contaminated sites, selenium is not routinely analyzed.

METHODS

Fish were collected with fishing rods and seines in 2001-2002 at Oak Ridge, and by electro shocking methods in 1997 from the Savannah River (Fig. 1). At Oak Ridge, fish were collected from the lower 4-km reach of Poplar Creek and from the 1.6-km reach of the Clinch River below the Melton Hill Dam. Fish were collected at the Savannah River above, below, and along the Savannah River Site. Fish were collected under appropriate state fishing licenses or permits, and with protocol approvals from Rutgers University Animal Review Committee. Collected white bass were large enough to be eaten by people (at least 22 cm in length), while Stonerollers, eaten whole by piscivorous birds and mammals, are used as bioindicators at Oak Ridge [34]. Scientific names for species are given in Table 1. Most of the fish species collected from Savannah River are eaten by people and eco-receptors. Once collected, fish were weighed and their total lengths measured before edible fillets were removed. The fillets were frozen, labeled by fish, date, and collection location, and transported to the Environmental and Occupational Health Sciences Institute (EOHSI) for metals analysis.

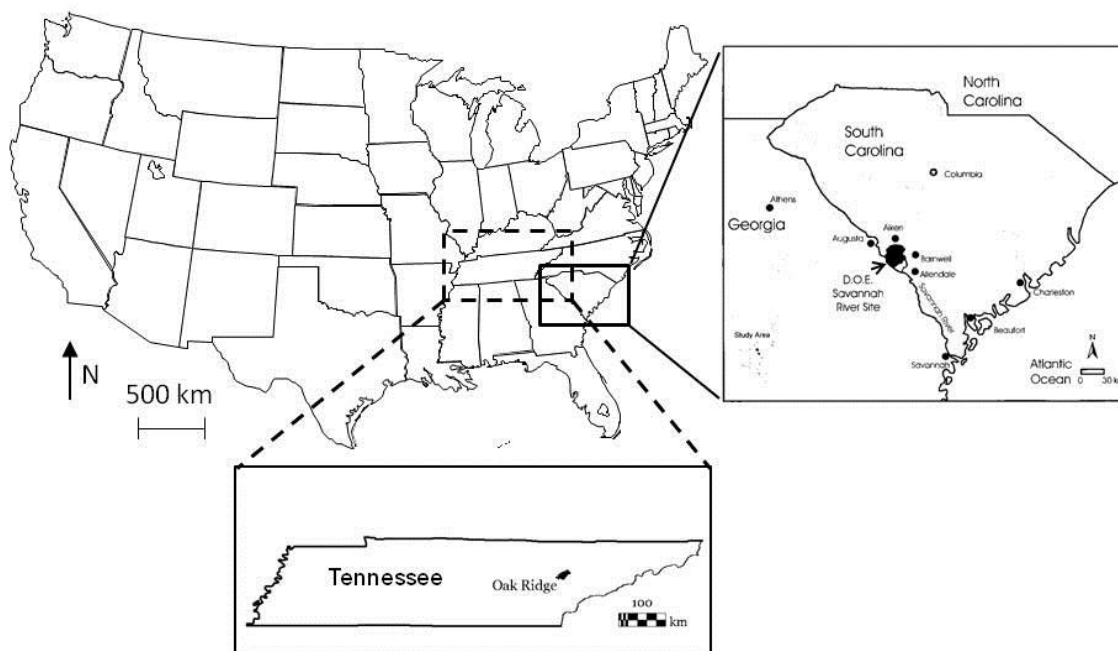


Fig. 1. Map showing Oak Ridge Reservation in eastern Tennessee and Savannah River site location in South Carolina where fish samples were collected .

At EOHSI, tissues were washed vigorously in deionized water alternated with acetone to remove external contamination [35]. All laboratory equipment and containers were washed in 10% HNO₃ solution prior to each use. A 2 gm sample was digested in ultrex ultrapure nitric acid in a microwave (MD 2000 CEM), using a digestion protocol of three stages of ten min each under 50, 100 and 150 pounds per square inch (3.5, 7, and 10.6 kg/cm²) at 70X power. Digested samples were subsequently diluted in 100 ml deionized water. Total selenium was analyzed by graphite furnace atomic absorption, and total mercury was analyzed by the cold vapor technique (HGS-4) [13]. Detection limits in ng/g (ppb) were total Hg=0.2, total Se=0.7. All tissue concentrations are expressed in parts per million (ppm, µg/g on wet weight). All specimens were run in batches that included blanks, a standard calibration curve and spiked specimens. The accepted recoveries for spikes ranged from 85 % to 115%. No batches were outside of these limits. The coefficient of variation (C.V.) on replicate samples ranged 2-9 %. Further descriptions of methods (and data from additional fish and locations) can be found in Burger and Campbell [13], and Burger et al. [36]. In most fish, 90 % or more of total mercury is methylmercury [37].

For each species a mean Se:Hg molar ratio was calculated (Table 1). The average mercury concentration (in µg/g) for the species was divided by 200.59 (molecular weight of mercury) and the average selenium concentration (in µg/g) was divided by 78.96, the molecular weight of selenium). The Se:Hg ratio was computed from these average molar concentrations. This is the usual method used to determine molar ratios. This may give a somewhat different result from calculating a ratio for each individual and then taking the average of the individual ratios. Note that some papers report the Hg:Se ratio rather than the Se:Hg reported in this paper (e.g. Cappon and Smith [38]). Hg:Se is the reciprocal of Se:Hg. The object of using ratios is to examine the molar ratios with respect to their chemical interaction, rather than just emphasizing concentrations in relation to specific location, or as representative of current conditions (as mercury levels have dropped over time at EFPC [5]).

RESULTS

There were significant interspecific differences in mercury and selenium levels, and in the selenium:mercury molar ratios (Table 1). Except for Bowfin, Se:Hg molar ratios were above 1:1 for all species at both sites. For all species the ratio was inversely correlated with mercury levels (Fig. 2).

Table I. Mercury and selenium levels (ppm wet weight) ($\mu\text{g/g}$), selenium-mercury ratios, and relationship of these ratios to fish size for fish from Oak Ridge and the Savannah River. Given are arithmetic means \pm standard error (SE).

Common Name (Scientific Name)	n	Mercury Mean \pm SE	Se Mean \pm SE	Hg nmol/ g wet wt.	Se nmol/ g wet wt.	Se:Hg Ratio (Means) ^a	Se:Hg Ratio Correlation with Hg tau (p)	Se:Hg Ratio Correlation with Length tau (p)
Savannah River Site								
Bowfin (<i>Amia calva</i>)	58	0.94 \pm 0.05	0.25 \pm 0.01	4.69	3.17	0.68	-0.57 (<0.0001)	-0.24 (0.007)
Largemouth Bass (<i>Micropterus salmoides</i>)	48	0.46 \pm 0.04	0.33 \pm 0.02	2.29	4.18	1.82	-0.65 (<0.0001)	-0.39 (0.0001)
Spotted Sucker (<i>Minytrema melanops</i>)	35	0.27 \pm 0.04	0.36 \pm 0.05	1.35	4.56	3.39	-0.57 (<0.0001)	-0.34 (0.005)
Black Crappie (<i>Pomoxis nigromaculatus</i>)	53	0.24 \pm 0.02	0.34 \pm 0.03	1.20	4.31	3.60	-0.53 (<0.0001)	-0.24 (0.01)
Channel Catfish (<i>Ictalurus punctatus</i>)	45	0.20 \pm 0.02	0.21 \pm 0.01	1.00	2.66	2.67	-0.69 (<0.0001)	-0.30 (0.004)
Bluegill Sunfish (<i>Lepomis macrochirus</i>)	30	0.14 \pm 0.02	0.41 \pm 0.03	0.70	5.19	7.44	0.60 (<0.0001)	-0.04 (NS)
Kruskal Wallis X^2 (p)		141 (<0.0001)	56.1 (<0.0001)			120 (<0.0001)		
Oak Ridge Reservation								
Central stonerollers (<i>Camptostoma anomalum</i>)	20	0.37 \pm 0.05	0.61 \pm 0.05	1.84	7.76	4.22	-0.53 (0.001)	0.42 (0.009)
Striped Bass (<i>Morone saxatilis</i>)	15	0.30 \pm 0.04	0.41 \pm 0.03	1.48	5.21	3.53	-0.49 (0.01)	-0.02 (NS)
White Bass (<i>Morone chrysops</i>)	29	0.12 \pm 0.02	0.65 \pm 0.04	0.58	8.23	14.11	-0.69 (<0.0001)	0.26 (0.05)
Crappie (<i>Pomoxis spp.</i>)	14	0.05 \pm 0.02	0.42 \pm 0.03	0.25	5.26	21.09	-0.62 (0.002)	-0.06 (NS)
Kruskal Wallis X^2 (p)		46.8 (<0.0001)	25.6 (<0.0001)			46.1 (<0.0001)		

a. The Se:Hg molar ratios are calculated on unrounded mean Hg and Se values.

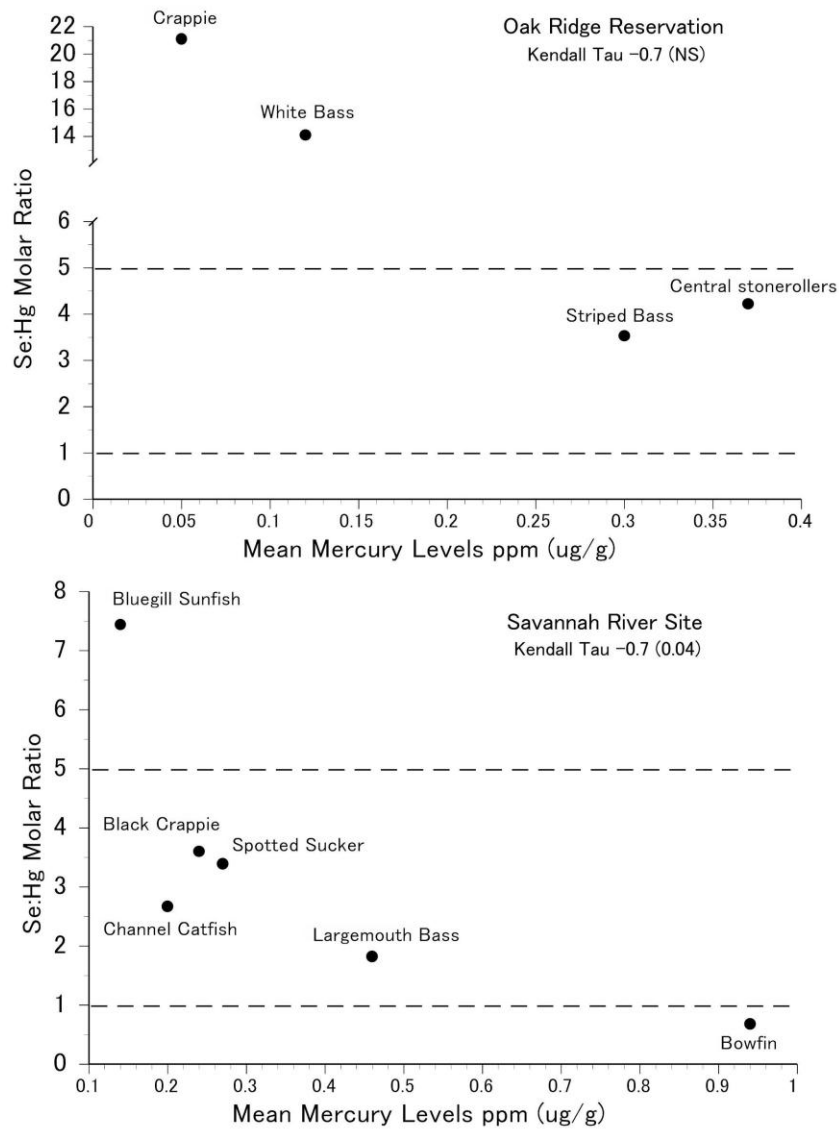


Fig. 2. Selenium:Mercury ratio plotted against mean mercury levels (ppm) for all fish species.

Interspecific differences

The relationship between Se:Hg molar ratios and fish size (e.g. length) varied among fish species. It was negative for most, but positive for Stonerollers and White Bass (Table 1, Fig. 3). The lack of a consistent relationship with fish size complicates the use of molar ratios in risk assessment and its use by the public in making informed decisions regarding what fish to eat.

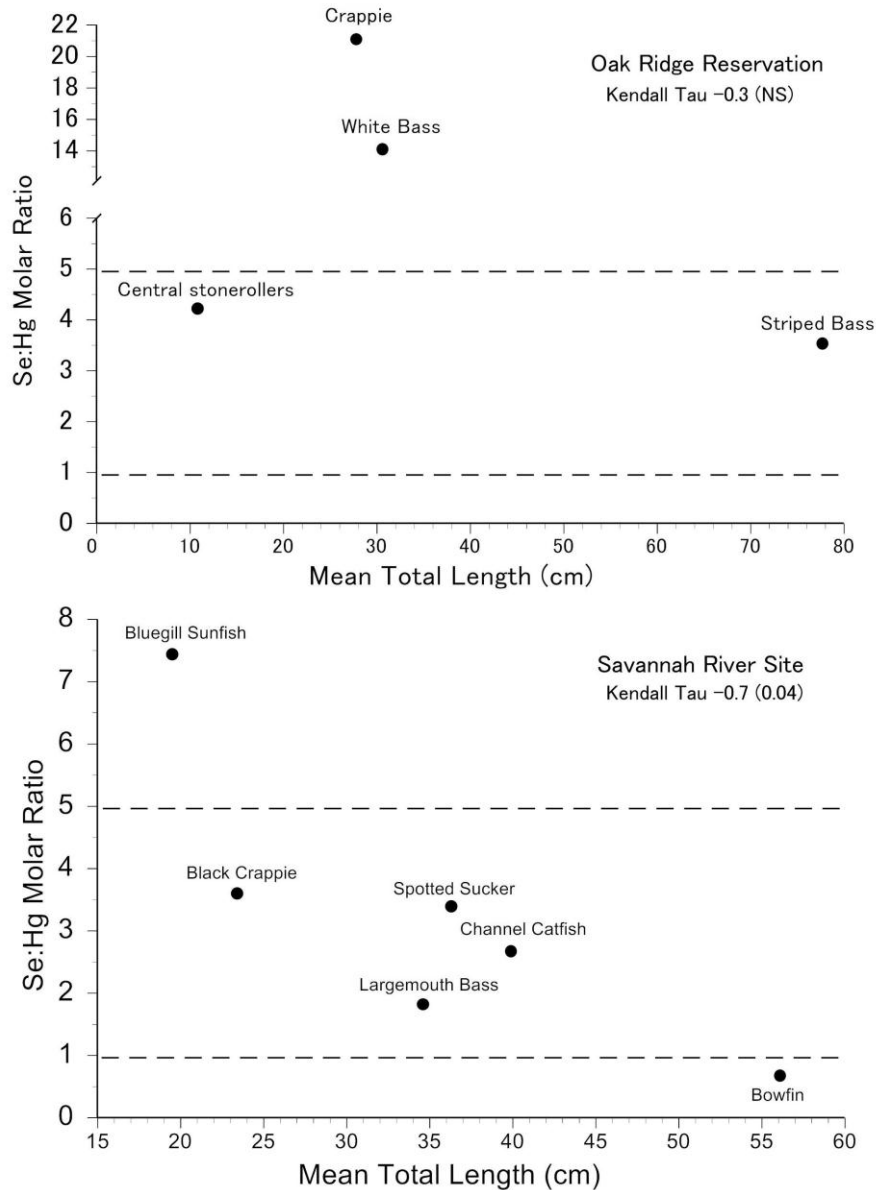


Fig. 3. Se:Hg ratios by mean total length (cm)

Intraspecific differences

To be useful, The Se: Hg ratio would be more useful if there were little variation Se:Hg within a species, allowing for predicting possible benefits of the ratio. Although standard errors give some indication of variance, they are hard to interpret by risk managers or the general public. Figure 4 shows the Se:Hg molar ratios for all individual fish for four species; to illustrate variability. They show the variation within a species, the clear positive size relationship for Stoneroller, the negative size relationship for Largemouth Bass, and no relationship for Spotted Sucker.

A quantitative method of examining this is to look at the percent of fish of each species with a given selenium: molar relationship (Table 2). All the fish at Savannah River had some

fish with ratios below 1:1 (except for bluegill sunfish), and none of the fish at Oak Ridge had ratios below 1. All species, except crappie, had some fish with ratios in the 1 to 5 range.

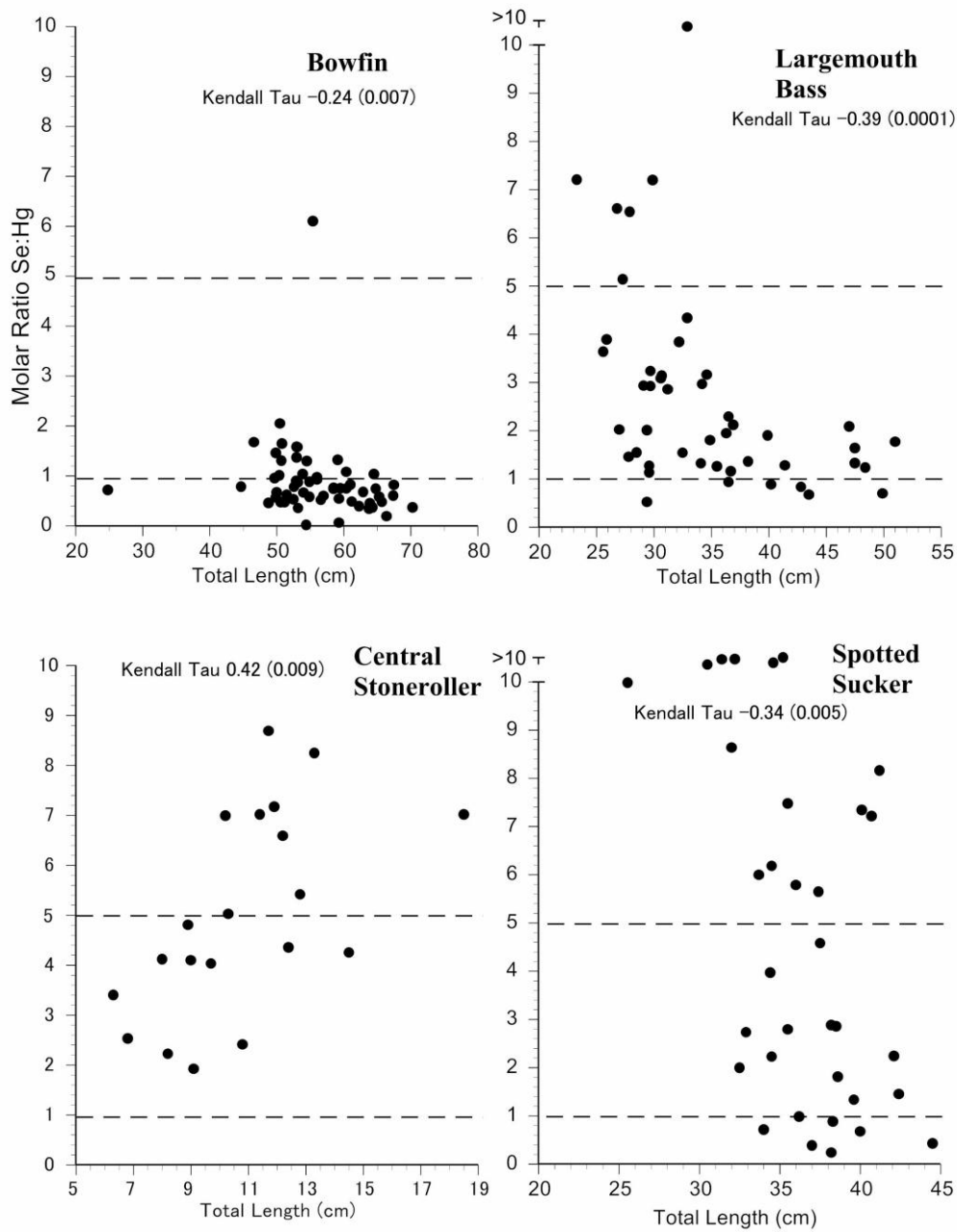


Fig. 4. Selenium: mercury ratio by total length for individual fish of four different species.

DISCUSSION

Table II. Variation in Selenium-Mercury Ratios within fish that might be useful in risk assessment or risk management.

Common Name	Se:Hg Ratio (Means) ^a	Se:Hg Mean ± SE (Individual fish)	% Fish		
			< 0.99	1.0-5.0	>5.0
Savannah River Site					
Bowfin	0.7	0.9 ± 0.1	74.1%	24.2%	1.7%
Largemouth Bass	1.8	2.7 ± 0.4	12.5%	75.0%	12.5%
Spotted Sucker	3.4	9.7 ± 3.2	20.0%	37.1%	42.9%
Black Crappie	3.6	4.3 ± 0.3	7.6%	56.5%	35.9%
Channel Catfish	2.7	4.1 ± 0.6	10.7%	63.8%	25.5%
Bluegill Sunfish	7.4	15.3 ± 3.5	0.0%	30.0%	70.0%
Oak Ridge Reservation					
Central stonerollers	4.2	5.0 ± 0.5	0.0%	55.0%	45.0%
Striped Bass	3.5	4.3 ± 0.5	0.0%	71.4%	28.6%
White Bass	14.1	21.4 ± 2.4	0.0%	10.3%	89.7%
Crappie	21.1	38.4 ± 11.5	0.0%	0.0%	100.0%

a. The Se:Hg molar ratios are calculated on unrounded mean Hg and Se values.

Both the addition of selenium to water, and the natural use of selenium supplementation as a moderator of mercury toxicity to consumers have been advanced as possible methods of reducing and managing risk to humans and eco-receptors [3, 8-9]. Here we examined whether Se:Hg molar ratios were sufficiently high to imply protectiveness against mercury toxicity, and whether the variation in these ratios was low enough to allow predictability of its protectiveness. Most fish from both sites had mean Se:Hg ratios that were above 1 (except for Bowfin), the ratios related to mercury concentrations, and the ratios generally decreased with fish size (except for stonerollers and white bass. These have implications for managing mercury fluxes from the Oak Ridge Reservation, in risk assessment and risk management, and for individual choices of fish species to consume.

Implications for managing mercury fluxes

The Department of Energy devotes considerable time and money to monitoring and managing mercury fluxes from Oak Ridge Reservation to Upper East Fork Poplar Creek (UEFPC), and thus to East Fork Poplar Creek (EFPC) by the Department of Energy [3]. Releases of mercury during operations of the Y-12 complex during the 1950s and 1960s resulted in contamination of soil and groundwater within the facility, and subsequent transport to East Fork Poplar Creek.

The Y-12 National Security Complex buildings and underground soil continue to be a source of offsite mercury transport. The building and drainage environment in the Y-12 area is complex, resulting in a complicated conceptual model requiring understanding of the dynamic nature of the hydrologic, geochemical and physical environment, within a framework of mercury fate and transport. While the Y-12 Complex is the source of mercury, the critical pathway endpoint remains mercury levels in the ecosystem, particularly fish in East Fork Poplar Creek (EFPC). Transport of elemental mercury (Hg⁰) in water and sediment, biomethylation, and bioamplification through the food chain, result in high mercury levels in higher trophic level biota, including humans who consume fish. While the mercury fate, transport and flux has varied significantly in the last 10 years, mercury continues to be a problem in fish in EFPC because some fish do not meet the EPA freshwater criteria for mercury (< 0.3 µg/g=ppm), despite declines in mercury levels in water, sediment and soil [3-4, 39].

Source reduction ultimately will involve decommissioning and decontamination of the relevant buildings in Y-12 Complex, and removal or remediation of contaminated soil underlying the buildings. Data gaps include improving understanding of mercury sources, transport pathways, flux at the Y-12 plant, mercury speciation, relationship between mercury and selenium, pathways near buildings and under storm drains, and mercury concentrations and flux in downstream media and bioavailability [3]. Further, there are no current plans to deal with mercury fluxes on SRS into the Savannah River.

Understanding the relative protective nature of selenium with respect to mercury toxicity in specific fish allows managers and the public to determine whether mercury toxicity should be considered on the basis solely of the levels of mercury (see introduction), or whether selenium is protective. That is, are concentrations of selenium sufficiently high to suggest that the levels of mercury are not as toxic as predicted on the basis of the mercury levels. The data suggest that relying on levels of selenium to reduce the mercury toxicity in organisms themselves (e.g. people or other top-level predators that consume these fish) as a management strategy will not be effective.

Understanding the levels of mercury and selenium in fish from Oak Ridge and Savannah River also requires comparing these levels to those from elsewhere. In a previous study, mercury and selenium levels from salt water fish from New Jersey ranged from 0.10 ppm to 0.52 (most were below 0.20 ppm), except for shark (a much larger fish than found at the sites described in this paper)[43]. Thus overall, several of the fish from Oak Ridge and SRS were higher than the fish from New Jersey. A more detailed comparison of fish from elsewhere can be found in Burger and Gochfeld [43].

Implications for risk assessors

The data presented suggest that the mean Se:Hg molar ratios are below 5 for most fish collected from the Savannah River, although ratios were higher in fish from Oak Ridge. The variability within fish species, however, suggests that the relative Se:Hg molar ratios in fish are not stable enough to be used in risk assessment. That is, in conducting risk assessments, it is not possible to determine the spread of ratios, which would be needed for probabilistic risk assessment. Significantly more fish samples per species are required to begin to generate data that would allow its use in risk assessment. Adding Se:Hg molar ratios seems to complicate risk assessment for the potential adverse effects of mercury exposure, and using mercury levels at this time remains the most viable option. The U.S. usually deals with the risk from mercury in fish by issuing fish consumption advisories [40-42]. However, at present, it seems unlikely and impractical to use the Se:Hg molar ratios in these advisories, even when and if a protective ratio is identified.

Implications for individual fish consumers

One of the objectives of risk assessment and risk management is to develop information for the public that will allow people to make meaningful and rationale decisions about what fish to eat, and how much fish to eat. In general, levels of mercury in fish, and variations in mercury levels as a function of size, are used by health professionals and the public to make decisions about fish consumption. This data from two freshwater systems near DOE sites, indicates that there is both interspecific and intraspecific variation in selenium: mercury ratios, making it impractical at this time for health professionals and the public to use these ratios in making sound decisions.

Further, that the Se:Hg ratios decrease with fish size in most species of fish, but increase for other fish makes interpretation difficult. If ratios decrease with fish size, it means that there is less and less selenium available to ameliorate the adverse effects of mercury, at the same time that the absolute levels of mercury are increasing in the fish. The interaction between these two factors further complicates interpretation.

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