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Metal levels in flathead sole (*Hippoglossoides elassodon*) and great sculpin (*Myoxocephalus polyacanthocephalus*) from Adak Island, Alaska: Potential risk to predators and fishermen

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Abstract

Increasingly there is a need to assess the contaminant levels in fish as indicators of the health and well-being of both the fish and their consumers, including humans. This paper examines the levels of arsenic, cadmium, chromium, lead, manganese, mercury, and selenium in the kidney, liver, and muscle of great sculpin and flathead sole from Adak Island in the Aleutian Islands, Alaska. Both species are consumed by the local Aleuts and others. There were significant differences in the levels of heavy metals as a function of tissue for both fish species; the liver of sculpin and sole generally had the highest levels of most metals, except for arsenic, lead, and selenium. Sole had significantly higher mean levels of arsenic in kidney (32,384 vs. 531 ppb, wet weight), liver (18,954 vs. 2532 ppb), and muscle (19,452 vs. 1343 ppb) than did sculpin. Sole also had higher mean levels of cadmium (230 vs. 63 ppb), lead (1236 vs. 48 ppb), mercury (150 vs. 107 ppb), and selenium (5215 vs. 1861 ppb) in kidney than did sculpin. There were significant correlations among weight and length measurements for both species. However, except for mercury, there were few significant correlations among tissue types for most metals. Only mercury and manganese levels were significantly correlated with size for sculpin (but not for sole). Levels of arsenic, lead, and mercury may pose a risk to predators that consume them, and arsenic and mercury may pose a risk to human consumers.
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1. Introduction

Increasingly, the public, policymakers, and managers require information about contaminants in biota to assess the health and well-being of organisms and the consumers that eat them. In many parts of the world fish provide an important and essential source of protein. Further, fishing is an enjoyable activity. Recreational and subsistence fishing are significant features of rural culture and tradition (Toth and Brown, 1997; Jensen et al., 1997; Fleming et al., 1995; Burger, 2002). High fishing rates occur in a wide range of cultures, including in urban areas (Burger et al., 1999, 2001a–c; Bienenfeld et al., 2003; Ramos and Crain, 2001), among Native Americans (Harris and Harper, 1998; Burger, 1999), and in other parts of the world (Burger et al., 2003). Fish are excellent low-fat sources of protein and provide many benefits, such as contributing to low blood cholesterol (Anderson and Wiener, 1995). Fish provide omega-3 (n-3) fatty acids that reduce cholesterol levels and the incidence of heart disease, stroke, and preterm delivery (Anderson and Wiener, 1995; Daviglus et al., 2002; Patterson, 2002).

However, the levels of contaminants in fish are of considerable interest because of potential effects on the fish themselves and on the top-level predators that consume them, including people. Fish consumption is the only significant source of methylmercury (MeHg) for the public

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(Rice et al., 2000). Levels of MeHg and polychlorinated biphenyls (PCBs) are sufficiently high in some fish to cause adverse human health effects in people consuming large quantities (Stern, 1993; IOM, 1991; Hightower and Moore, 2003; Hites et al., 2004). MeHg counteracts the cardioprotective effects of omega-3 fatty acids (Guallar et al., 2002) and can damage developing fetuses and young children (NRC, 2000). Maternal exposures threaten the fetus because chemicals are transferred across the placenta (Gulson et al., 1997, 1998). There is a positive relationship between mercury and/or PCB levels in fish, fish consumption by pregnant women, and deficits in neurobehavioral development in children (Jacobson and Jacobson, 1996; Lonky et al., 1996; Schantz, 1996; NRC, 2000, Stern et al., 2004, Schantz et al., 2003). There is also a decline in fecundity in women who consume large quantities of contaminated fish from Lake Ontario (Buck et al., 2000).

People who live on isolated oceanic islands often rely rather heavily on marine resources, particularly where fishing is an integral part of their culture and traditions. While considerable attention has been given to the subsistence foods of Native Alaskans (Norman et al., 1992; Egeland and Middaugh, 1997; Duffy et al., 1999; Rothschild and Duffy, 2002; Jewett et al., 2003), less attention has been devoted to the Aleut and Pribilof Islanders because of their relatively small populations and the remoteness of their villages. Some villages are accessible only by infrequent air flights or freight liners. Yet, fish are an important part of their year-around diet.

In this paper, we examine the levels of arsenic, cadmium, chromium, lead, manganese, mercury, and selenium in the liver, kidney, and muscle of flathead sole (*Hippoglossoides elassodon*) and great sculpin (*Myoxocephalus polyacanthocephalus*) from Adak Island in the Aleutians. Fish were collected in the village at the harbor where local anglers regularly fished. We were particularly interested in whether the metal levels posed risks to the fish and the people that consumed them. We also compared the levels of the seven metals among tissues and between species. These fish species are eaten both as whole fish in stews and soups and as fillets and are frozen for later consumption.

2. Methods

2.1. Study site

Adak Island, located in the Aleutian Chain, is approximately 1900 km west of the tip of the Alaskan Peninsula (Fig. 1). The Aleutian Islands were established as a National Wildlife Refuge in 1913 by executive order of President Taft (ATSDR, 2004). A Naval Air Facility occupied the northern portion of the island from 1942 until 1997, when operations closed (ATSDR, 2004). At its peak, over 100,000 military personnel were stationed on the island; thus the potential exists for historic contamination of the marine environment, particularly near the seaport and airport areas of Adak. In 1994 the Naval Air Station at Adak was placed on the National Priority List based on an EPA RCRA inspection (ATSDR, 2004). Most of the population (currently less than 200 people) resides in the town of Adak, which has an airport and a seaport. The island is partly
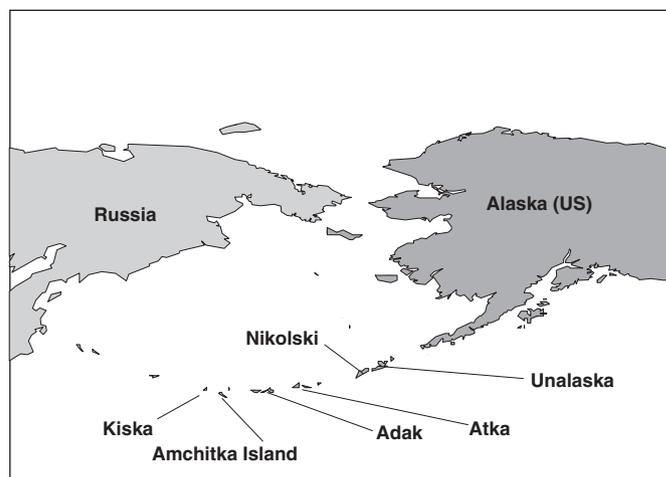

Fig. 1. Map showing the location of Adak Island, along with the major Aleut villages (Nikolski, Atka, Unalaska), military islands (Kiska) and nuclear testing site (Amchitka).

owned by the US Fish and Wildlife Service and partly by the Aleut Corp., which is in the process of developing commercial fisheries.

2.2. Methods

Under appropriate permits from the State of Alaska's Department of Fish and Game (No. CF-04-043), fish were collected in June 2004 from the harbor in Adak, a popular fishing area for the local population, including Aleuts. We fished in the same manner and from the same spots as the local fishermen. Fish were immediately measured, weighed, and dissected, and samples of kidney, liver, and muscle were frozen for later analysis. Liver and kidney were collected as indicators of potential risk to the fish and muscle was taken as an indicator of risk to consumers, including humans.

Fish were shipped frozen to the Environmental and Occupational Health Sciences Institute (EOHSI) of Rutgers University for metal analysis. At EOHSI, a 2-g (wet weight) sample of fish tissue was digested in ultrex ultrapure nitric acid in a microwave (MD 2000 CEM), using a digestion protocol of three stages of 10 min each under 50, 100, and 150 pounds per square inch (3.5, 7, and 10.6 kg/cm^2) at 80 × power. Digested samples were subsequently diluted to 25 ml with deionized water. All laboratory equipment and containers were washed in 10% HNO$_3$ solution and deionized water rinse prior to each use (Burger et al., 2001a).

Mercury was analyzed by the cold vapor technique using the Portable Zeeman Lumex (RA-915) mercury analyzer, with an instrument detection level of 0.2 ng/g and a matrix level of quantification of 0.002 μg/g. All other metals were analyzed with graphite furnace atomic absorption (GFAA), including arsenic, cadmium, chromium, lead, manganese, and selenium. Instrument detection limits on the GFAA were 0.2 ppb for arsenic, 0.1 ppb for cadmium, 1.0 ppb for chromium, 2.0 ppb for lead, 1.0 ppb for manganese, and 0.5 ppb for selenium. Matrix detection levels were about an order of magnitude higher. All concentrations are expressed in parts per billion (ppb = ng/g) on a wet weight basis. In another study (Burger et al., 2001b) we found that the dry weight ranged from 23% to 33% of the corresponding wet weight (i.e., water content of 67–77%) for 11 types of fish from the Savannah River. Many studies have shown that almost all of the mercury in fish tissue is MeHg, and 90% is a reasonable approximation of this proportion, which does vary somewhat among fish types and laboratories.

A DORM-2 Certified dogfish tissue was used as the calibration verification standard. Recoveries between 90% and 110% were accepted to validate the calibration. All specimens were run in batches that included blanks, a standard calibration curve, two spiked specimens, and one duplicate. The accepted recoveries for spikes ranged from 85% to 115%

and no batches were outside of these limits; 10% of samples were digested twice and analyzed as blind replicates (with agreement within 15%). For further quality control on mercury, our laboratory periodically runs a random subset of samples in the Quebec Laboratory of Public Health; the correlation between the two laboratories is over 0.90 ($P < 0.0001$; see Burger and Gochfeld, 2004).

We used Kruskal–Wallis nonparametric one-way analysis of variance (generating a χ^2 statistic) to examine differences among tissues and between fish species. We also used ANOVA with Duncan multiple range test on log-transformed data to identify the significant differences (SAS, 1999). Kendall correlations were used to examine relationships among metals. The level for significance was set at $P < 0.05$.

3. Results

There were significant differences in the levels of heavy metals as a function of tissue for both fish species (except for arsenic in sole, Tables 1 and 2). In great sculpin, the liver generally had the highest levels of most metals (Table 1). In contrast, in flathead sole the liver had the highest levels of some metals, but not arsenic, chromium, lead, or selenium (Table 2). Muscle tissue of sculpin had significantly lower levels of only lead and selenium than the liver and kidney, while the muscle tissue of sole was the lowest only for selenium.

Sole had significantly higher levels of arsenic in all its tissues than did sculpin (kidney: $\chi^2 = 33.4$, $P < 0.0001$; liver: $\chi^2 = 34.3$. $P < 0.0001$; muscle: $\chi^2 = 36.3$, $P < 0.0001$).

Sole also had higher levels of cadmium, lead, mercury, and selenium in kidney than did sculpin (cadmium: $\chi^2 = 20.6$, $P < 0.0001$; lead: $\chi^2 = 31.9$, $P < 0.0001$; mercury: $\chi^2 = 4.3$, $P < 0.04$; selenium: $\chi^2 = 25.8$, $P < 0.0001$).

There were significant correlations among weight and length for both species (Table 3). However, except for mercury, there were few significant correlations among tissue types for most metals (Table 3). There were also few significant correlations among metals in muscle for either sole or sculpin. There were only 3 of 28 possible correlations for sculpin, and 6 for sole (Table 3). Further, only mercury and manganese levels were significantly correlated with size for sculpin (but not sole).

4. Discussion

4.1. Interspecific differences

There were interspecific differences in metal levels for arsenic (all tissues) and for cadmium, lead, and selenium in the kidney. Some differences might be expected due to the different ecological niches of sole and sculpin. While both species are mainly benthic/nektobenthic feeders, great sculpin occasionally take pelagic prey (Mito, 1974). Sole live in the silt or mud bottoms from nearshore to less than 366 m in depth (Mecklenburg et al., 2002). They rarely

Table 1

Contaminant levels in three organs (kidney, liver, and muscle) of 18 great sculpin specimens collected from Adak during the summer of 2004 (given are ppb, wet weight)

Metals	Kidney mean ± std. err geometric mean ($n = 16$)	Liver mean ± std. err geometric mean ($n = 18$)	Muscle mean ± std. err geometric mean ($n = 18$)	χ^2 (P)
Arsenic	531 ± 29 520 (C)	2532 ± 402 1958 (A)	1343 ± 216 1108 (B)	21 (0.0001)
Cadmium	63 ± 21 30 (B)	1261 ± 279 648 (A)	4 ± 1 2 (B)	38 (0.0001)
Chromium	169 ± 10 163 (A, B)	241 ± 69 144 (A)	90 ± 16 73 (B)	14 (0.0007)
Lead	48 ± 3 46 (A)	35 ± 8 14 (A)	14 ± 4 2 (B)	18 (0.0001)
Manganese	405 ± 40 383 (B)	978 ± 96 869 (A)	472 ± 64 425 (B)	19 (0.0001)
Mercury	107 ± 22 77 (B)	286 ± 52 222 (A)	323 ± 58 245 (A)	15 (0.0006)
Selenium	1861 ± 172 1718 (A)	2075 ± 242 1886 (A)	609 ± 34 592 (B)	32 (0.0001)

Table 2
Contamininant levels in three organs (kidney, liver, and muscle) of 39 flathead sole specimens collected from Adak during the summer of 2004 (given are ppb, wet weight)

Metals	Kidney mean ± std. err geometric mean ($n = 39$)	Liver mean ± std. err geometric mean ($n = 38$)	Muscle mean ± std. err geometric mean ($n = 39$)	χ^2 (P)
Arsenic	32,384 ± 6071 20,740 (A)	18,954 ± 1959 15,895 (B)	19,452 ± 1013 18,505 (B)	NS
Cadmium	230 ± 31 184 (B)	4948 ± 491 4118 (A)	4 ± 1 3 (B)	100 (0.0001)
Chromium	276 ± 65 179 (B)	107 ± 36 62 (A)	95 ± 12 75 (B)	37 (0.0001)
Lead	1236 ± 89 1118 (A)	123 ± 7 116 (B)	50 ± 8 40 (B)	96 (0.0001)
Manganese	344 ± 29 308 (B)	892 ± 57 845 (A)	355 ± 31 285 (B)	64 (0.0001)
Mercury	150 ± 22 122 (B)	243 ± 17 222 (A)	276 ± 16 265 (A)	46 (0.0001)
Selenium	5215 ± 545 4442 (A)	2603 ± 118 2510 (B)	398 ± 37 338 (C)	90 (0.0001)

relinquish contact with the ocean floor except to make rare short forays to capture small fish (Simenstad et al., 1977; Johnson, 2003). Flathead sole eat mainly clams, worms, and crustaceans, including euphausiids (Mito, 1974; Smith et al., 1978; Chuchukalo et al., 1994; Pacunski et al., 1998). They shift diet from mainly crustaceans to ophiuroids with increasing size (Mito, 1974; Smith et al. 1978; Pacunski et al., 1998). Their diet also varies greatly with sediment type (McConnaughey and Smith, 2000). The maximum age for flathead sole from the Bering Sea is 27 years (Munk, 2001), but age data are not available for the sculpin (McConnaughey and Smith, 2000). Fish in this study were presumably adults.

Great sculpin live on sand and mud bottoms around rocks and are usually found at less than 200-m depths (Simenstad et al., 1977; Mecklenburg et al., 2002). Great sculpin eat mainly decapods offshore, but they also eat fish and amphipods inshore (Mito, 1974; Simenstad et al., 1977; Jewett and Powell, 1979), where we caught them. Tokranov (1992) also reported that they eat mainly fish (flatfish were a major prey item). During the day sculpin often rest near the bottom in the littoral community, supported by their broad pectoral fins (Simenstad et al., 1977).

There are almost no data on metal levels in these two species, from here or elsewhere, and few data on metals levels in other fish from the Aleutians. Ben-David et al. (2001) found mean mercury levels of 53 ppb for sculpin from Prince William Sound and 63 ppb for coast range sculpin in Alaska. Further, Crayton (2000) published levels of heavy metals (but not mercury) in livers of Pacific cod (*Gadus macrocephalus*) and rock greenling (*Hexagrammos lagochephalus*) from the marine environment of Amchitka Island. Levels of cadmium were lower and those of chromium and manganese were higher than those for the liver of the fish that we collected. However, the area around Amchitka Island is relatively uncontaminated with metals because of relatively low human activity, while Adak was a large military base with an active harbor and airport.

The general lack of correlation of metal levels with size for both species may have been related to the small range of sizes of fish examined in this study.

4.2. Risk to the fish and predators that consume them

Contaminants in fish can pose health risks to the fish and their predators. Arsenic poisoning is relatively rare in wildlife (Eisler, 1994). Most arsenic in fish is organic arsenic, which is less toxic than inorganic arsenic species (Eisler, 1994; ATSDR, 2000). Arsenic is interesting because most studies investigate inorganic arsenic, yet the arsenic in

Table 3

Correlation among metals for muscle and size measurements for great sculpin and flathead sole from Adak in the Aleutian Chain (given are the Kendal tau correlations (and significance))

	Great sculpin	Flathead sole
Sample size	18	39
Size		
Weight (g)	1786 ± 71	587 ± 14
Total length (cm)	45 ± 1	39 ± 0.27
Standard length (cm)	41 ± 1	33 ± 0.25
Snout gill (cm)	16 ± 0.3	10 ± 0.09
Size relationships		
Weight and total length (cm)	0.43 (0.01)	0.79 (0.0001)
Weight and standard length	0.51 (0.004)	0.75 (0.0001)
Weight and snout gill length	0.64 (0.0004)	0.59 (0.0001)
Correlations among tissues		
Arsenic for muscle and liver	NS	NS
for muscle and kidney	NS	NS
for liver and kidney	NS	NS
Cadmium for muscle and liver	NS	NS
for muscle and kidney	NS	NS
for liver and kidney	0.38 (0.04)	0.45 (0.0001)
Chromium for muscle and liver	NS	NS
for muscle and kidney	NS	NS
for liver and kidney	NS	NS
Lead for muscle and liver	NS	NS
for muscle and kidney	NS	NS
for liver and kidney	NS	0.25 (0.03)
Manganese for muscle and liver	NS	NS
for muscle and kidney	NS	NS
for liver and kidney	−0.41 (0.03)	NS
Mercury for muscle and liver	0.53 (0.002)	0.31 (0.007)
for muscle and kidney	0.76 (0.0001)	0.2 (0.08)
for liver and kidney	0.44 (0.02)	0.37 (0.001)
Selenium for muscle and liver	NS	NS
for muscle and kidney	NS	NS
for liver and kidney	NS	0.33 (0.004)
Correlation among metals for muscle		
arsenic and cadmium	−0.42 (0.01)	NS
arsenic and manganese	−0.32 (0.01)	NS
arsenic and mercury	NS	0.25 (0.02)
chromium and lead	NS	0.34 (0.003)
chromium and selenium	NS	−0.28 (0.01)
lead and manganese	0.47 (0.02)	0.23 (0.04)
lead and selenium	NS	−0.21 (0.06)
manganese and selenium	NS	−0.41 (0.0003)
Among metals and size		
Total weight and mercury	0.49 (0.005)	NS
Snout gill length and mercury	0.32 (0.07)	NS
Standard length and manganese	−0.37 (0.04)	NS

NS, not significant.

fish is mainly organic (Eisler, 1994), making it difficult to examine effects. However, the levels in flathead sole were significantly higher than those in sculpin from Adak; arsenic levels in liver of sole were seven times higher than those in sculpin (for muscle it was 14 times higher). The high level of arsenic in flathead sole from Alaska has been noted previously (Meador et al., 2004).

Adverse effects from cadmium can occur in fish with dietary levels of 0.1 ppm (Eisler, 1985). Whole-body burdens of cadmium in fish from the US overall average 0.03 ppm (wet weight), with the maximum being 0.22 ppm (Schmitt and Brumbaugh, 1990), Cadmium levels can range as high as 0.54 ppm in free ranging fish (Burger et al., 2002). In sole and sculpin from Adak, cadmium levels were comparatively low. These species can be eaten by other fish, birds, or marine mammals. Birds may be less sensitive to cadmium in their diet than mammals but are adversely affected at levels of 1.0 ppm (Eisler, 1994). In nearby arctic regions, there is some evidence that cadmium levels in some seabirds are high enough to cause kidney damage (AMAP, 2002). Thus, there may be some cause for concern for top-level avian predators that eat some of these fish, particularly if they eat the liver of sole.

Levels of 10 ppm of chromium in the diets of birds and mammals are considered to cause adverse effects in some wildlife species (Eisler, 1986). Levels of chromium in sole and sculpin from Adak were well below 10 ppm, suggesting that predators or scavengers would not be at risk from chromium if they ate them in the wild.

Lead is a neurotoxin that causes behavioral deficits in vertebrates (Weber and Dingel, 1997) and can cause decreases in survival, growth rates, learning, and metabolism (Eisler, 1988; Burger and Gochfeld, 2000). Levels of 50 ppm in the diet can cause reproductive effects in some predators, and dietary levels as low as 0.1–0.5 ppm are associated with learning deficits in some vertebrates (Eisler, 1988). In this study, the levels of lead in liver, kidney, and muscle of sculpin were well below the adverse effects level, suggesting that predators would not be adversely affected because of lead. However, levels of lead in the kidney of flathead sole averaged 1.2 ppm, well above the effects levels for some predators.

There are remarkably few studies on the dietary effects of manganese on predators or on the adverse effects associated with particular tissue levels on organisms. Manganese is in most need of extensive laboratory and field studies, particularly with fish that are consumed by sensitive or endangered/threatened predators. Manganese, selenium, and chromium are essential trace elements, although all can cause toxicity at high doses (Burger and Gochfeld, 1995).

Mercury concentrations of 5 ppm (wet weight) in fish muscle can be associated with emaciation, decreased coordination, loss of appetite, and mortality in fish (Eisler, 1987), while concentrations of 15 ppm are required for adverse effects in predators that eat the fish (Spry and Wiener, 1991; Wiener and Spry, 1996). Sensitive birds that consume fish can exhibit effects at dietary mercury concentrations of 0.05–0.5 ppm for sensitive mammals, harmful effects occur at dietary levels of 1.1 ppm (Eisler, 1987; WHO, 1990, 1991). In this study, mercury levels in muscle averaged 0.3 ppm, suggesting that some sensitive species may be adversely affected, although seabirds are generally less sensitive to high levels than other birds

(Furness, 1996). Although mercury is the heavy metal found in greatest abundance in animal tissue collected from the Bering Sea (Johnson, 2003), we did not find this to be the case at Adak.

Selenium can be toxic at high levels, even though it is an essential micronutrient (Coyle et al., 1993). A concentration of about 1 ppm (wet weight) in prey is the threshold for selenium toxicity in some fish, while muscle concentrations of 2.6 ppm are associated with adverse effects in fish (Lemly, 1993a, b). Further, selenium concentrations of 1 ppm in food are toxic to other wildlife that consume them (Lemly, 1993a), suggesting that selenium may be a problem for predators of sole and sculpin only if the liver and kidney are eaten.

4.3. Risk to humans

There is a lively national discussion about the health benefits and risks from consumption of fish, mainly focused on recreational and subsistence fish. While the definition of subsistence is arguable generally, fish comprise an important part of the diet of the people living in the Aleutian and Pribilof Islands in the northern Pacific/Bering Sea region (Patrick, 2002). Providing these people with information about contaminants in the fish that they consume is thus an important public health mandate.

The metal that was the highest relative to averages for the US generally was arsenic in sole. The EPA has set arsenic tissue residues of 1.3 ppm fresh weight in freshwater fish as the criterion for human health protection (Eisler, 1994). In this study, arsenic in muscle averaged 1.3 ppm in sculpin and 19.5 in sole. This suggests that some sculpin may pose a human health risk for arsenic, but sole surely does.

The USFDA has an action level for MeHg in fish (FDA, 2001); the level of 1.0 mg/kg (ppm w/w) is a regulatory action level rather than a risk level. Originally the FDA had set 0.5 ppm as the action level, comparable to many other nations reviewed in Burger and Gochfeld (2004). The UK and the European Union have established criteria for certain metals in fish (e.g., the level for mercury is 0.5 ppm in edible fish, with up to 1 ppm allowed for certain 'exempt' predatory fish species). China has set standards for MeHg in canned fish (ppm wet weight) of 0.5 ppm (except 1 ppm is allowed in shark, sailfish, tuna, pike, and other high-mercury fish). In 1982 the European Commission set an Environmental Quality Standard for mercury; the mean concentration in mercury of a representative sample of fish shall not exceed 0.3 mg/kg (wet weight). The US EPA promulgated this value as an ambient freshwater quality standard in 2001 (http://www.epa.gov/fedrgstr/EPA-WATER/2001/January/Day-08/w217.htm). Muscle in the sculpin and sole in this study averaged about 0.3 ppm, suggesting that some fish were above the ambient freshwater quality standard and above the health standards set for some countries.

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