# Selenium and Mercury in Pelagic Fish in the Central North Pacific Near Hawaii

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**Abstract** Protective effects of selenium against mercury toxicity have been demonstrated in all animal models evaluated. As interactions between selenium and mercury and their molar ratios in seafood are essential factors in evaluating risks associated with dietary mercury exposure, considering mercury content alone is inadequate. In this study, the absolute and molar concentrations of mercury and selenium were determined in edible portions from 420 individual fish representing 15 species of pelagic fish collected from the central North Pacific Ocean near Hawaii. Selenium was in molar excess of mercury in almost all fish species evaluated. The rank order of mean Se/Hg molar ratios was striped marlin (17.6)>yellowfin tuna (14.1)>mahimahi (13.1)>skipjack tuna (12.8)>spearfish (11.4)>wahoo (10.8)>sickle pomfret (6.7)>albacore tuna (5.3)>bigeye tuna (5.2)>blue marlin (4.1)>escolar (2.4)>opah (2.3)>thresher shark (1.5)>swordfish (1.2)>mako shark (0.5). With a Se/Hg molar ratio of less than 1, mako shark was the only fish containing a net molar excess of mercury. A selenium health benefit value based on the absolute amounts and relative proportions of selenium and mercury in seafood is proposed as a more comprehensive seafood safety criterion.

**Keywords** Mercury · Methylmercury · Selenium · Fish · Seafood · Environmental risk assessment · Seafood safety

# Introduction

The assessment of the potential health risk posed by the presence of mercury (Hg) in pelagic (open ocean) fish is an important and yet unresolved issue [1]. Whereas it is well documented that seafoods contain from nondetectable to low levels of Hg [2–7], the

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potential for adverse health effects from the normal low level Hg exposure from eating ocean fish remains controversial [1, 8–12]. By contrast, nearshore and freshwater fish may pose a greater health risk because of the proximity to and potential for greater Hg accumulation traced to anthropogenic sources. Neurodevelopmental impairments associated with maternal consumption of seafoods containing high levels of Hg in their edible portions [4, 13] have been reported [13–16]. Catastrophic Hg pollution from a chemical plant produced fish with extremely high (~50 ppm) Hg concentrations in Minamata bay in Japan that caused severe toxic effects in the exposed population in the late 1950s and early 1960s [13]. However, for the populations examined in the more recent studies [14–16], the seafood responsible for the majority of dietary Hg exposure was pilot whale meat, not ocean fish. Meanwhile, maternal consumption of ocean fish resulting in similar or higher Hg exposures has not been associated with adverse effects [17, 18]. Most recently, results from the Avon Longitudinal Study of Parents and Children (ALSPAC) performed in the UK [11] found that increasing maternal fish consumption was associated with improved child neurodevelopment. Meanwhile, maternal avoidance of fish during pregnancy not only failed to protect their children's health, instead it actually caused harm. The contrasting findings of these studies appear to reflect real distinctions in the effects of Hg exposure in their respective study groups. However, differences in the elemental composition of the dietary sources of Hg in the populations examined may be responsible for the dramatic contrast in the findings of these studies.

Selenium's (Se) significance in the prevention of Hg toxicity has been recognized for 40 years. The first report on the protective effect of Se against Hg toxicity appeared in 1967 [19]. Since then, numerous studies have shown that Se counteracts the adverse impacts of Hg exposure [20–31] including Se from yellowfin tuna [21, 32], menhaden [33], swordfish [34], and rockfish [35]. The ability of Se to decrease the toxic action of Hg has been established in all species investigated to date [36, 37]. Because of the high binding affinity between Hg and Se [38], the mechanism of Se's protective effect had formerly been thought to involve direct sequestration of Hg by Se, but recent research suggests that supplemental Se protects against Hg toxicity by offsetting the loss and sequestration of Se by Hg [29, 31, 39]. Selenium is highly protective against Hg, preventing lethality and other consequences otherwise associated with Hg toxicity in adults, growing adolescents, and offspring exposed in utero. Therefore, the presence of Se in ocean fish and the significance of its protective effects against Hg are important considerations in the seafood safety issue.

As Hg exposure from fish consumption has been the focus of research in this issue, the molar ratio between Hg and Se has generally been overlooked. The Hg/Se molar ratio tends to be quite low in the edible muscle of most commercially important ocean fish [3, 6, 40]. This may be because ocean fish are among the richest dietary sources of Se, comprising 17 of the top 25 sources in the American diet [41]. However, in certain seafoods, there can be a molar excess of Hg over Se [3, 4], and maternal consumption of such seafoods is uniformly associated with reports of Hg-dependent harm to developing children.

Selenium is a nutritionally essential trace element that is absolutely required for the activity of 25–35 enzymes with important functions [42, 43]. Although normally present in all cells of all higher animal life, the functions of these enzymes are especially important in the brain [44, 45] and endocrine organs [46]. These enzymes perform numerous antioxidant functions [42] and, together with other Se-containing molecular species, appear to be important in Se's roles in the prevention of cancer [47, 48] and in supporting a healthy immune system [49].

Evaluation of the health risk posed by Hg exposure from seafood consumption requires concurrent consideration of the Se content in the particular species. The molar ratio of Hg and Se in the diet appears to be the essential criterion of risk from Hg exposure rather than Hg content alone [31, 37, 39]. For these reasons, a survey was conducted to determine Hg/Se molar ratios of 15 species of pelagic fish, caught in the central North Pacific Ocean in the Hawaii pelagic longline fishery.

### **Materials and Methods**

#### Fish Species Sampled

The 15 pelagic fish species sampled are presented in Table 1. Fish were sampled in 2006 from commercial landings at the Honolulu Fish Auction (operated by the United Fishing Agency) with cooperation of the Hawaii Longline Association. All fish were caught on pelagic longline gear and stored in ice. Onboard fish handling procedures before storage followed standard commercial practices. These included (1) bleeding only (whole fish), (2) evisceration and removal of gills (gilled and gutted), and (3) evisceration and removal of gills and head (headed and gutted), depending on the market specifications for the fish species.

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Fish species	Scientific name	Sample n	Landed form	Weight (kg), mean±SD	Minimum weight	Maximum weight
Bigeye	Thunnus obesus	50	GG	41.2±20.4	11.3	89.8
Yellowfin	Thunnus albacares	50	GG	$41.1 \pm 16.7$	13.2	76.2
Albacore	Thunnus alalunga	20	GG	$22.6 \pm 3.8$	16.3	29.9
Skipjack	Katsuwonus pelamis	10	GG	8.6±1.3	6.4	10.4
Swordfish	Xiphias gladius	50	HG	$78.4 \pm 55.9$	10.4	198.7
Blue Marlin	Makaira mazara	50	GG	97.9±63.8	33.1	318.9
Striped Marlin	Tetrapturus audax	30	GG	32.2±17.1	6.4	69.4
Spearfish	Tetrapturus angustirostris	20	GG	11.9±3.3	7.3	18.6
Mahimahi	Coryphaena hippurus	30	GG	8.2±3.9	1.8	20.4
Wahoo	Acanthocybium solandri	30	GG	10.2±6.3	5.0	29.0
Opah	Lampris guttatus	30	W	45.4±12.4	26.8	64.0
Sickle pomfret	Taractichthys steindachneri	10	W	7.3±1.7	5.0	10.0
Escolar	Lepidocybium flavobrunneum	10	W	12.8±8.0	5.0	37.2
Mako shark	Isurus oxyrinchus	10	HG	57.4±12.5	40.8	80.3
Thresher shark	Alopius vulpinus	10	HG	$71.3 \pm 29.4$	36.7	129.3

Table 1 Pelagic Fish Species Studied, Sample Number, Mean Weights, and Weight Range

Fish were landed in three different forms.

GG Gilled and gutted, HG headed and gutted, W whole

Fish weights (kilograms) were obtained from a State of Hawaii certified platform scale, rounded to the nearest pound and converted to kilogram. A sampling plan was prepared to collect a predetermined number of fish within typical market size classes representing each fish species. Data recorded included fish species, weight and landed form (whole, gilled and gutted or headed and gutted).

# Sampling Site and Sample Handling

All samples were a minimum of 100 g of edible muscle collected from the anterior portion of the dorsal muscle mass ("top quarter or loin"). Muscle samples were placed in new plastic bags labeled with a unique sample number. Samples were frozen immediately and stored at -23°C. A chain of custody form was maintained from the time the samples were collected until they were received at the laboratory for analyses. Frozen samples were shipped with dry ice, by overnight courier to the Energy and Environmental Research Center at the University of North Dakota in Grand Forks, North Dakota, for Hg and Se analysis.

# Laboratory Analyses

Sample aliquots of ~0.4 g accurately weighed to 0.0001 g from each individual fish sample were transferred into single use, trace element-free 50-ml digestion tubes (Environmental Express, Mt. Pleasant, SC 29464) with every tenth fish sample being prepared in duplicate and with elemental spike recovery samples being performed accompanying each batch. Each digestion batch included blank and certified standard reference materials (dogfish muscle certified reference material DORM-2, National Research Council of Canada, Ottawa, Ontario, Canada). Samples were treated with 5 ml of 16 N nitric acid (Fisher Trace Metal Grade, Fisher Scientific, http://www.fishersci.com) and heated at 85°C in deep cell hot blocks (Environmental Express) for 24 h in capped tubes to preserve samples from trace element contamination. Samples were cooled, and 1.5 ml of 30% hydrogen peroxide (Fisher Certified A.C.S., Fisher Scientific) was added, and samples were recapped and returned to heating in the dry block at 85°C. Samples were cooled, and 15 ml of 12 N HCl (Fisher Trace Metal Grade, Fisher Scientific) were added. Samples were heated at 90°C for 90 min to reduce Se-VI to Se-IV. Samples were cooled and diluted to 50 ml with double distilled water. Samples were analyzed for Hg content by cold vapor atomic absorption spectrophotometry using a CETAC M-6000A (CETAC Technologies, Omaha, NE), and Se was analyzed by hydride generation atomic fluorescence spectroscopy using a PS Analytical Dual Millennium Excalibur (PS Analytical, Deerfield Beach, FL).

### Selenium Health Benefit Value

Foods that supply more Se than Hg protect against mercury toxicity and provide Sedependent health benefits, whereas those that contain more Hg than Se are associated with mercury health risks. To better describe and integrate Se-specific nutritional benefits in relation to potential Hg-exposure risks presented by a given type of seafood, the proposed selenium health benefit value (Se HBV) is calculated as follows:

Se HBV = (Se/Hg molar ratio x total Se) - (Hg/Se molar ratio x total Hg)

Elemental analysis results for Hg and Se from individual fish samples were used to calculate selenium health benefit values. The mean of these individual value determinations was used to establish the selenium health benefit values for each of the fish species studied.

### Data Treatment and Statistical Analysis

Before data from sample analysis runs were entered into the database, Hg and Se concentrations in sample batch digestion blanks and elemental recoveries in samples of certified standard reference materials were evaluated to qualify the analysis batch data for inclusion. Total Hg and Se mass concentrations (parts per million) for each individual fish sample were converted to molar concentrations (micromole per kilogram), and molar ratios of Hg and Se in individual samples were calculated. The molar excess of Se (free Se) relative to Hg present in the samples was determined by subtracting the molar Hg concentration from the molar Se concentration for individual samples. Means and standard deviations of elemental mass and molar concentrations, molar ratios, and molar excess were calculated and graphed for evaluation.

#### Results

The common names, scientific names, number, landed form, and weight (mean, standard deviation and range) of fish analyzed are shown in Table 1. The fish selected for analysis in this study are representative of the size range of fish that landed in Hawaii's longline fishery but not necessarily the size frequency. The total elemental distributions of Hg and Se in fish sampled in this study are presented in mass (microgram per gram or parts per million) and molar (micromole per kilogram) concentrations in Table 2. The mean total Hg concentrations of the 15 species sampled ranged from 0.13 ppm for mahimahi to 2.38 ppm for blue marlin. The mean total Hg concentrations were below 1 ppm for all tuna species, striped marlin, spearfish, mahimahi, wahoo, sickle pomfret, opah, escolar, and thresher shark. The mean total Hg concentration for swordfish was close to 1 ppm. The mean total mercury concentrations of the 15 pelagic fish species sampled ranged from 0.32 ppm for mako shark up to 1.59 ppm for blue marlin.

The mean molar concentrations of Se and Hg are compared in Fig. 1. Calculated molar ratios (Se to Hg and Hg to Se) and free Se are presented in Table 2. Although measurable amounts of Hg were present in all fish, in 13 of the 15 pelagic fish species studied, there was a molar excess of Se over Hg. Yellowfin and skipjack tuna were particularly rich sources of Se, containing many moles of Se for every mole of Hg. Although blue marlin Hg concentrations increased with size, so did their Se contents. As a result, blue marlin tissue Se concentrations were uniformly in molar excess of Hg. This was in contrast to swordfish, whose Se contents were independent of size, but because their Hg concentrations increased proportionally with increasing weight, their mean Se and Hg contents were nearly equimolar. The mako shark was the only fish evaluated in this study that was found to have a molar excess of Hg over Se. Although Hg concentrations consistently increased with increasing size, all mako shark samples contained approximately twofold more Hg than Se. The calculated mean Hg to Se molar ratios (Hg/Se) are ranked from low to high from the left to right in Fig. 2. Fish species with Hg to Se molar ratios greater than 1 contain a molar excess of Hg over Se.

Fish type	п	Mercury co	ontent	Selenium content		Molar ratios		Free Se	Se HBV
		µg Hg/g	µmol Hg/kg	µg Se/g	µmol Se/kg	Se/Hg Hg/Se		(Se–Hg)	(calculated)
Yellowfin	50	$0.30 {\pm} 0.18$	$1.51 {\pm} 0.88$	1.25±0.27	$15.80 \pm 3.44$	14.12	0.10	14.29	201.7
Mahimahi	30	$0.13{\pm}0.07$	$0.66 {\pm} 0.32$	$0.53{\pm}0.09$	$6.66 \pm 1.11$	13.07	0.10	6.00	78.4
Skipjack	10	$0.34{\pm}0.10$	$1.68 {\pm} 0.52$	$1.57{\pm}0.92$	$19.83 \pm 11.71$	12.82	0.13	18.16	232.7
Spearfish	20	$0.21{\pm}0.13$	$1.05 {\pm} 0.63$	$0.58 {\pm} 0.12$	$7.30 \pm 1.46$	11.36	0.14	6.25	71.0
Wahoo	30	$0.25{\pm}0.20$	$1.23 \pm 1.01$	$0.65{\pm}0.14$	$8.27 \pm 1.72$	10.82	0.15	7.05	76.2
Albacore	20	$0.50 {\pm} 0.24$	$2.49 \pm 1.18$	$0.88 {\pm} 0.19$	$11.11 \pm 2.40$	5.26	0.24	8.62	45.4
Bigeye	50	$0.60{\pm}0.25$	$3.00 {\pm} 1.23$	$0.99{\pm}0.28$	$12.38 \pm 3.48$	5.17	0.26	9.4	48.6
Sickle pomfret	10	0.47±0.53	2.35±2.65	$0.71 \pm 0.11$	8.99±1.45	6.69	0.28	6.63	44.4
Striped Marlin	30	0.47±0.37	2.34±1.83	0.72±0.20	9.06±2.56	17.61	0.28	6.72	118.3
Blue Marlin	50	2.38±3.00	11.87±14.95	1.59±0.17	20.17±14.78	4.11	0.46	8.29	34.1
Opah	30	$0.56{\pm}0.28$	$2.81 \pm 1.38$	$0.42{\pm}0.06$	$5.38 \pm 0.78$	2.29	0.53	2.58	5.9
Escolar	20	$0.74{\pm}0.29$	$3.68 {\pm} 1.45$	$0.56{\pm}0.14$	$7.12 \pm 1.73$	2.4	0.55	3.44	8.3
Thresher shark	10	$0.98 {\pm} 0.32$	4.86±1.60	0.52±0.12	6.55±1.51	1.49	0.75	1.68	2.5
Swordfish	50	$1.07 {\pm} 0.60$	$5.32{\pm}2.98$	$0.39{\pm}0.07$	$5.43 {\pm} 1.48$	1.16	0.99	0.10	0.1
Mako shark	10	$1.81 \pm 0.40$	9.01±1.99	0.32±0.04	$4.07 {\pm} 0.48$	0.46	2.25	-4.93	-11.1

Table 2 Mass and Molar Concentrations and Molar Ratios of Mercury and Selenium in Pelagic Fish Species

Free Se reports the group mean of free Se estimated by simple subtraction of  $\mu$ mol Hg/kg content from  $\mu$ mol Se/kg for individual fish samples. The Se health benefit value (HBV) was calculated as (Se/Hg molar ratio× total Se)–(Hg/Se molar ratio× total Hg)

### Discussion

The Se contents of most ocean fish (13 of 15) included in this study were in molar excess of the Hg that was also present. These results corroborate previous studies that found that pelagic fish tend to be rich dietary sources of Se. In almost all cases, the molar ratios of Se to Hg were favorable, indicating substantial Se was available to counter the Hg that was also present in these fish. The Se and Hg contents in swordfish were nearly equimolar; thus, the net effect of these elements would be balanced by each other's presence, resulting in little or no net effect of Se contribution or Hg exposure. The only fish in this study that possessed a molar excess of Hg over Se was the mako shark, which contained approximately twice as much Hg as Se.

As a result of their rich Se and low-Hg contents, skipjack and yellowfin tuna have the most favorable selenium health benefit values, followed by striped marlin, mahimahi, wahoo, spearfish, bigeye tuna, albacore tuna, sickle pomfret, and blue marlin. Although not as rich a source of Se, escolar, opah, and thresher shark still provide positive Se health benefits. The equimolar ratios of Hg and Se in swordfish make its selenium health benefit value negligible, but as their average Hg content does do not exceed their Se content, Hg-exposure risks would be similarly negligible. Because of the significantly negative selenium health benefit value of mako shark, consumption of this fish during pregnancy would not be advisable, as it would impair maternal delivery of Se to the developing fetus. Information on pilot whale meat from the Faroe Islands [4] is included in Fig. 3 for comparison

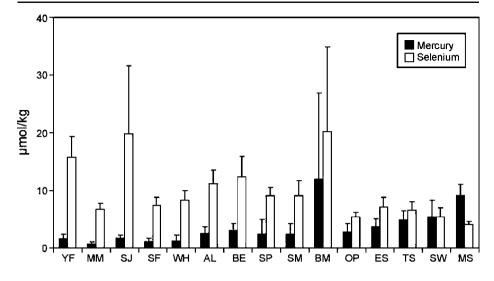


Fig. 1 Molar concentrations of mercury and selenium in fish species. Data are expressed as means  $\pm$  standard deviations. *YF* yellowfin, *MM* mahimahi, *SJ* skipjack, *SF* spearfish, *WH* wahoo, *AL* albacore, *BE* bigeye, *SP* sickle pomfret, *SM* striped marlin, *BM* blue marlin, *OP* opah, *ES* escolar, *TS* thresher shark, *SW* swordfish, *MS* mako shark

purposes. Pilot whale meat contains a substantial molar excess of Hg over Se content, resulting in a net delivery of Hg and a highly negative selenium health benefit value. As the Health Ministry of the Faroe Islands has already recognized, consumption of pilot whale meat during pregnancy is contraindicated and is diminishing in response to health advisories that distinguish pilot whale from ocean fish [50]. However, consumption of Serich open ocean fish that contain a molar excess of Se over Hg would be expected to afford

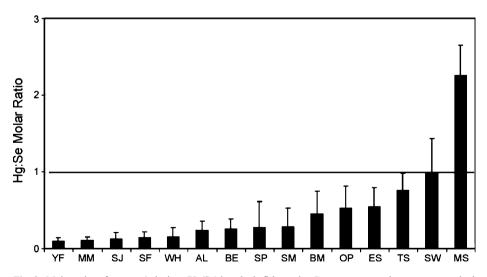
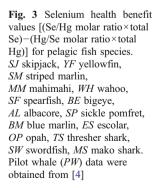


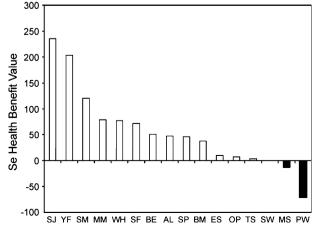
Fig. 2 Molar ratios of mercury/selenium (Hg/Se) in pelagic fish species. Data are expressed as means  $\pm$  standard deviations. *YF* yellowfin, *MM* mahimahi, *SJ* skipjack, *SF* spearfish, *WH* wahoo, *AL* albacore, *BE* bigeye, *SP* sickle pomfret, *SM* striped marlin, *BM* blue marlin, *OP* opah, *ES* escolar, *TS* thresher shark, *SW* swordfish, *MS* mako shark

the numerous health benefits associated with augmented Se status such as enhanced antioxidant tone, improved immune function, and anticancer effects. As Se protects against Hg toxicity, eating fish with rich Se contents would be a particularly beneficial consideration for consumers of potentially hazardous foods, such as make shark, pilot whale, or any other foods with negative selenium health benefit values.

### Conclusions

Major studies performed in the Faroe Islands [14–16], New Zealand [51], Seychelles Islands [17, 18], and in the UK [11] have evaluated the effects of maternal mercury exposure from seafood on subsequent child development. The Faroe Islands study of 917 children found a statistically significant association between mercury exposure in utero and diminished neurodevelopmental test scores. The maternal diet in the Faroe Islands population included codfish known to be low in mercury, but also included periodic feasts on pilot whale, which supplied over 90% of that population's overall mercury exposure. The New Zealand study of 237 children selected from a total cohort of 10,970 new mothers suggested maternal Hg-exposure that may influence child development, but the interpretation of results was highly dependent on whether or not one high achieving child whose mother had by far the highest Hg exposure was included or excluded from the study. In contrast, the Seychelles Islands Child Development Study found no adverse health impacts in 711 children borne to mothers that ate 12 meals per week of ocean fish, but no marine mammal meat or shark during pregnancy. The larger (11,875 mother child pairs) and more comprehensive results from the ALSPAC study in the UK indicate that substantial benefits were associated with increasing maternal fish consumption. In contrast to the findings in the Faroe Islands, this study found no indication of harm accompanied with increasing Hg exposure. Instead, neurodevelopmental impairments were associated with maternal avoidance of fish consumption [11]. There are significant discrepancies between the findings of these three important studies if their results are considered from the perspective of Hg exposure alone. However, the results of these studies exactly coincide with what





would be expected if considered from the more comprehensive perspective of Hg/Se molar ratios.

The interactions of Se and Hg and the differences in the Hg/Se molar ratios in the seafoods consumed by the study populations appear to account for the differences in observed outcomes of these studies. The ~4:1 molar ratio of Hg/Se in the pilot whale meat eaten in the Faroe Islands and its extremely high-Hg concentration [4], 16  $\mu$ mole/kg (3.37 ppm), is reflected in its highly negative selenium health benefit value (-70). The high and disproportionate Hg exposure from pilot whale meat consumption would be expected to substantially diminish Se availability in the maternal blood supply and, therefore, transiently reduce transplacental delivery of Se to the fetus. The neurodevelopmental consequences of Hg exposure from maternal pilot whale meat feasts would have been partially offset by Se intake from maternal consumption of codfish, a rich Se source with relatively little Hg. A recent reanalysis reveals that benefits from fish consumption partially offset the harm from the high-Hg exposure accompanying consumption of pilot whale meat [52].

The Se present in the fish consumed by the New Zealand population during the time of the maternal Hg-exposure study was never assessed. However, it is important to note that the Se status of the New Zealand population was notoriously poor [53], making them more vulnerable to Hg exposure. The take-away fish and chips consumed by New Zealanders in the late 1970s included fish such as sharks [54] with average Hg above 10  $\mu$ mole Hg/kg (2 ppm) with some as high as 22  $\mu$ mole Hg/kg (4.4 ppm). Based on the findings of the current study and previous studies that measured Hg and Se in various shark species [3], with such high-Hg contents, it is likely that the Hg/Se molar ratio in these shark portions was disproportionate and the selenium health benefit values were extremely negative.

The ocean fish diet consumed in the Seychelles Islands is about an order of magnitude lower in Hg content (1.5  $\mu$ mole Hg/kg; 0.3 ppm) than in the other studies [55] and tends to be Se-rich. The calculated selenium health benefit values for 4 of the 16 species assessed are in excess of 400. Because the Se status of the population studied is good [40], no adverse effects would be expected, and none were noted. It is possible that beneficial effects of fish consumption may not have been apparent in this study because the population of the Seychelles Islands was in uniformly good Se status and there were insufficient differences to enable an assessment.

Like most of Northern Europe, the people of the UK have notably poor Se status [56]. As the background Se status of the ALSPAC study population is low [43], increasing maternal consumption of ocean fish would progressively improve Se delivery to the developing fetus. Therefore, the beneficial effects of fish consumption would be especially important for this population. Meanwhile, avoidance of fish consumption during pregnancy not only diminishes Se status, but also limits intake of other nutrients such as omega-3 fatty acids [11]. As these nutrients are important for fetal brain development, it appears likely that increased fish consumption would be similarly beneficial in other populations.

It is important to recognize that, although most ocean fish are Se-rich, the Se status of freshwater fish is highly variable. More Hg accumulates in fish growing in lakes where Se availability is limited, and Se supplementation to normal levels results in Hg diminishing by more than 75% after 3 years [57]. Therefore, fish from low Se lakes would not only have low Se contents, they could also have relatively high-Hg contents, a dangerous combination that would result in highly negative selenium health benefit values. This may account for the adverse cardiovascular effects associated with Hg exposure from consumption of freshwater fish in a recent Finnish study [58]. Finland has historically had notably poor Se status, but has recently recognized cardiovascular health benefits resulting from its

nationwide effort to improve Se availability through use of Se-supplemented fertilizers [59]. Chronic consumption of high-Hg, low-Se freshwater fish with negative selenium health benefit values apparently reverses these benefits and impairs cardiovascular health.

As the availability of Se for biological uptake by lake fish tends to be variable, the same atmospheric Hg deposition that causes little Hg bioaccumulation in fish of some lakes in Se-rich areas may result in far more accumulation in Se-poor regions. These effects may be responsible for Hg "hotspots" that have already been recognized [60], but further study will be needed to establish the extent and effects of Hg/Se molar ratios in environmental Hg assessments. Even in lakes where high-Hg accumulations in fish have not been noted, the unfavorable Hg/Se molar ratios in fish from such "unbalanced" lake ecosystems could result in highly negative selenium health benefit values. Therefore, unrecognized environmental Hg exposure risks may exist in locations where they are not currently expected to occur.

In conclusion, the species, origin and size of fish must be clearly described in studies that report on the health benefits or adverse impacts of seafood consumption. It is essential that the molar ratios of Se and Hg in seafood be determined and integrated into the evaluation of the risk of dietary Hg exposure. The pelagic fish species examined in this study are extremely important food fish. They generally contain a molar excess of Se over Hg (a positive selenium health benefit value) and are therefore more likely to prevent Hg toxicity than contribute to it. This integrated consideration of Se and Hg molar ratios provides an improved safety standard for seafood and environmental risk assessment that appears to be more useful than criteria based on evaluation of Hg concentrations alone. The selenium health benefit value is useful in differentiating between seafood and freshwater fish that provide a rich source of Se from those that are potentially harmful because they contain an excess of Hg. It can also be used to establish a rank order of ocean fish that are rich dietary sources of Se and therefore beneficial to child neurodevelopment and cardiovascular health.

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#### References

- Clarkson TW, Magos L (2006) The toxicology of mercury and its chemical compounds. CRC Crit Rev Toxicol 36(8):609–662
- Jacobs G (1977) Total and organically bound mercury content in fishes from German fishing grounds. Z Lebensm Unters Forsch 164(2):71–76
- Hall RA, Zook EG, Meaburn GM (1978) National Marine Fisheries Service Survey of Trace Elements in the Fishery Resource. NOAA Technical Report NMFS SSRF-721. US Dept of Commerce, National Oceanographic and Atmospheric Administration, National Marine Fisheries Service
- Julshamn K, Anderson A, Ringdal O, Morkore J (1987) Trace elements intake in the Faroe Islands. I. Element levels in edible parts of pilot whales (*Globicephalus meleanus*). Sci Total Environ 65:53–62
- 5. National Research Council (2000) Toxicological effects of methylmercury, National Academy Press, Washington, DC
- Cabanero AI, Carvalho C, Madrid Y, Batoreu C, Camara C (2005) Quantification and speciation of mercury and selenium in fish samples of high consumption in Spain and Portugal. Biol Trace Elem Res 103(1):17–35

- Kojadinovic J, Potier M, Le Corre M, Cosson RP, Bustamante P (2006) Mercury content in commercial pelagic fish and its risk assessment in the Western Indian Ocean. Sci Total Environ 366(2–3):688–700
- Daniels JL, Longnecker MP, Rowland AS, Golding J (2004) Fish intake during pregnancy and early cognitive development of offspring. Epidemiology 15:394–402
- Rasmussen RS, Nettleton J, Morrissey MT (2005) A review of mercury in seafood: special focus on tuna. J Aqua Food Prod Tech 14(4):71–100
- Mozaffarian D, Rimm EB (2006) Fish intake, contaminants, and human health evaluating the risks and the benefits. JAMA 296(15):1885–1899
- Hibbeln JR, Davis JM, Steer C, Emmett P, Rogers I, Williams C, Golding J (2007) Maternal seafood consumption in pregnancy and neurodevelopmental outcomes in childhood (ALSPAC study): an observational cohort study. Lancet 369:578–585
- Institute of Medicine (2007) Seafood choices: balancing the benefits and risks. In: Nesheim MC, Yaktine AL (eds) National Academies Press, Washington, DC
- Harada Y (1968) Congenital (or fetal) Minamata disease. In Study Group of Minamata Disease. Kumamoto University, Kumamoto, pp 93–117
- Grandjean P, Weihe P, Joergensen PJ, Clarkson T, Cernichiari E, Videroe T (1992) Impact of maternal seafood diet on fetal exposure to mercury, selenium, and lead. Arch Environ Health 47(3):185–195
- Grandjean P, Weihe P, White RF, Debes F, Araki S, Murata K (1997) Cognitive deficit in 7-year-old children with prenatal exposure to methylmercury. Neurotoxicol Teratol 19:417–428
- Grandjean P, Weihe P, White RF, Debes F (1998) Cognitive performance of children prenatally exposed to "safe" levels of methylmercury. Environ Res 77:165–172
- 17. Davidson PW, Myers GJ, Cox C, Axtell C, Shamlaye C, Sloane-Reeves J, Cernichiari E, Needham L, Choi A, Wang Y, Berlin M, Clarkson TW (1998) Effects of prenatal and postnatal methylmercury exposure from fish consumption on neurodevelopment: outcomes at 66 months of age in the Seychelles child development study. J Am Med Assoc 280(8):701–707
- Myers GJ, Davidson PW (1998) Prenatal methylmercury exposure and children: neurologic, developmental, and behavioral research. Environ Health Perspect 106(3):841–847
- Parizek J, Ostadalova I (1967) The protective effect of small amounts of selenite in sublimate intoxication. Experiential 23(2):142–143
- Iwata H, Okamoto H, Ohsawa Y (1973) Effect of selenium on methylmercury poisoning. Res Comm Chem Path Pharm 5:673–680
- Ohi G, Nishigaki S, Seki H, Tamura Y, Maki T, Konno H, Ochiai S, Yamada H, Shimamura Y, Mizoguchi I, Yagyu H (1976) Efficacy of selenium in tuna and selenite in modifying methylmercury intoxication. Environ Res 12:49–58
- El-Begearmi MM, Sunde ML, Ganther HE (1977) A mutual protective effect of mercury and selenium in Japanese quail. Poultry Sci 56(1):313–322
- Beijer K, Jernelov A (1978) Ecological aspects of mercury-selenium interaction in the marine environment. Environ Health Perspect 25:43–45
- Sugiura Y, Tamai Y, Tanaka H (1978) Selenium Protection against mercury toxicity: high binding affinity of methylmercury by selenium containing ligands in comparison with sulfur containing ligands. Bioinorg Chem 9:167–180
- El-Begearmi MM, Ganther HE, Sunde ML (1982) Dietary interaction between methylmercury, selenium, arsenic, and sulfur amino acids in Japanese quail. Poultry Sci 61(2):272–279
- Imura N (1986) The role of micronutrient, selenium, in the manifestation of toxicity of heavy metals. Dev Toxicol Environ Sci 12:115–123
- Whanger PD (1992) Selenium in the treatment of heavy metal poisoning and chemical carcinogenesis. J Trace Elem Electrolytes-Health Dis 6(4):209–221
- Suzuki KT (1997) Equimolar Hg–Se complex binds to selenoprotein P. Biochem Biophys Res Commun 231(1):7–11
- Watanabe C, Yin K, Kasanuma Y, Satoh H (1999) In utero exposure to methylmercury and selenium deficiency converge on the neurobehavioral outcome in mice. Neurotoxicol Teratol 21(1):83–88
- Watanabe C (2001) Selenium deficiency and brain functions: the significance for methylmercury toxicity. Nippon Eiseigaku Zasshi 55(4):581–589
- Ralston NVC, Blackwell JL III, Raymond LJ (2007) Dietary selenium-dependent protection against methylmercury toxicity, Biol Trace Elem Res (this issue)
- 32. Ganther HE, Goudie C, Sunde ML, Kopecky MJ, Wagner P, Oh SH, Hoekstra WG (1972) Selenium: relation to decreased toxicity of methylmercury added to diets containing tuna. Science 175:1122
- Stillings BR, Lagally H, Bauersfield P, Soares J (1974) Effect of cystine, selenium, and fish protein on the toxicity and metabolism of methylmercury in rats. Toxicol Appl Pharmacol 30:243–254

- Freidman MA, Eaton LR, Carter WH (1978) Protective effects of freeze-dried swordfish on methylmercury content. J Environ Contam Toxicol 19:436–443
- 35. Ohi G, Nishigaki S, Seki H, Tamura Y, Maki T, Minowa K, Shimamura Y, Mizoguchi I, Inaba Y, Takizawa Y, Kawanishi Y (1980) The protective potency of marine animal meat against the neurotoxicity of methylmercury: its relationship with the organ distribution of mercury and selenium in the rat. Food Cosmet Toxicol 18(2):139–145
- Culvin-Aralar LA, Furness RW (1991) Mercury and selenium interaction: a review. Ecotoxicol Environ Saf 21:348–364
- Ralston CR, Blackwell JL III, Ralston NVC (2006) Effects of dietary selenium and mercury on house crickets (*Acheta domesticus* L.): implications of environmental co-exposures. Environmental Bioindicators 1(1):98–109
- Dyrssen D, Wedborg M (1991) The sulfur-mercury (II) system in natural waters. Water Air Soil Pollut 56:507–519
- Raymond LJ, Ralston NVC (2004) Mercury: selenium interactions and health implications. Seychelles Med Dent J 7(1):72–77 (spec issue)
- 40. Robinson J, Shroff J (2004) Observations on the levels of total mercury (Hg) and selenium (Se) in species common to the artisanal fisheries of Seychelles. Seychelles Med Dent J 7(1):55–60
- USDA National Nutrient Database for Standard Reference, Release 17. Selenium, Se (µg) Content of Selected Foods. http://www.nal.usda.gov/fnic/foodcomp/ (updated: 7/27/2005)
- 42. Behne D, Pfeifer H, Rothlein D, Kyriakopoulos A (2000) Cellular and subcellular distribution of selenium and selenium-containing proteins in the rat. In: Roussel AM, Favier AE, Anderson RA (eds) Trace elements in man and animals 10. Kluwer, New York, pp 29–34
- 43. Rayman M (2000) The importance of selenium to human health. Lancet 356:233-241
- 44. Chen J, Berry MJ (2003) Selenium and selenoproteins in the brain and brain diseases. J Neurochem 86 (1):1–12
- Schweizer U, Bra
  üer AU, Kohrle J., Nitsch R, Savaskan NE (2004) Selenium and brain function: a poorly recognized liaison. Brain Res Rev 45(3):164–178
- Kohrle J, Jakob F, Contempré B, Dumont JE (2005) Selenium, the thyroid, and the endocrine system. Endocr Rev 26(7):944–984
- 47. Clark C, Combs GF Jr, Turnbull BW, Slate EH, Chalker DK, Chow J, Davis LS, Glover RA, Graham GF, Gross EG, Krongrad A, Lesher JL Jr, Park HK, Sanders BB Jr, Smith CL, Taylor JR (1996) Effects of selenium supplementation for cancer prevention in patients with carcinoma of the skin: a randomized controlled trial. JAMA 276(24):1957–1963
- 48. Schrauzer GN (2000) Anticarcinogenic effects of selenium. Cell Mol Life Sci 57(13-14):1864-1873
- Beck MA, Levandert OA, Handy J (2003) Selenium deficiency and viral infection. J Nutr 133(5:2):1463S– 1467S (1 May 2003)
- Weihe P, Grandjean P, Jørgensen PJ (2005) Application of hair mercury analysis to determine the impact of a seafood advisory. Environ Res 97:200–207
- 51. Crump KS, Kjellstrom T, Shipp AM, Silvers A, Stewart A (1998) Influence of prenatal mercury exposure upon scholastic and psychological test performance: benchmark analysis of a New Zealand cohort, risk analysis. Risk Anal 18(6):701–713
- Budtz-Jørgensen E, Grandjean P, Weihe P (2007) Separation of risks and benefits of seafood intake. Environ Health Perspect 115(3):323–327
- Thomson CD, Robinson MF (1980) Selenium in human health and disease with emphasis on those aspects peculiar to New Zealand. Am J Clin Nutr 33(2):303–323
- Mitchell JW, Kjellstrom TE, Reeves RL (1982) Mercury in takeaway fish in New Zealand. N Z Med J 95 (702):112–114
- 55. Myers GJ, Davidson PW, Cox C, Shamlaye C, Palumbo D, Cernichiari E, Sloan-Reeves J, Wilding GE, Kost J, Li-S Huang, Clarkson TW (2003) Prenatal methylmercury exposure from ocean fish consumption in the Seychelles Child Development Study. Lancet 361:1686–1692
- 56. Rayman MP (2002) The argument for increasing selenium intake. Proc Nutr Soc 61(2):203-215
- Paulsson K, Lindbergh K (1989) The selenium method for treatment of lakes for elevated levels of mercury in fish. Sci Total Environ 87–88:495–507
- Salonen JT, Seppanen K, Nyyssonen K, Korpela H, Kauhanen J, Kantola M, Tuomilehto J, Esterbauer H, Tatzber F, Salonen R (1995) Intake of mercury from fish, lipid peroxidation, and the risk of

myocardial infarction and coronary, cardiovascular, and any death in Eastern Finnish men. Circulation 91 (3):645–655

- Pietinen P, Vartiainen E, Seppanen R, Aro A, Puska P (1996) Changes in diet in Finland from 1972 to 1992: Impact on coronary heart disease risk. Prev Med 25(3):243–250
- Evers DC, Han Y-J, Driscoll CT, Kamman NC, Goodale MW, Lambert KF, Holsen TM, Chen CY, Clair TA, Butler T (2007) Biological mercury hotspots in the Northeastern United States and Southeastern Canada. BioScience 57(1):29–43