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Estimation of acceptable mercury intake from fish in Taiwan

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Abstract

The US Food and Drug Administration (USFDA) and Environmental Protection Agency (USEPA) are advising women and young children to avoid eating fish that contain high levels of mercury (Hg). However in Taiwan, the annual Black Fin Tuna Festival encourages the public to consume fish. The aim of this study was to assess fish intake in relation to the health risks of mercury exposure and calculate the acceptable and safe intake of fish in children and women of childbearing age. From the Monte Carlo simulation, based on USEPA's reference dose (0.1 μg kg^{-1} d^{-1}), we found that 21.6%–24.3% and 45.6%–57.4% of the daily mercury dose estimates exceeded the reference dose for typical and high-seafood consumers. The acceptable ingestion rates are <50 g d^{-1} (children) and 90.8 ± 15.7 g d^{-1} (women of childbearing). Sensitivity analysis suggests that Hg concentration in fish may be a key parameter to aid governments as they offer guidance for risk management.

Keywords: Mercury; Black Fin Tuna Festival; Monte Carlo simulation; Mercury consumption; Fish intake

1. Introduction

Methylmercury (MeHg) is a well-known neurotoxicant in humans and has also been reported associated with developmental delays in children whose mothers were exposed during pregnancy (Bakir et al., 1973; WHO, 1990; Dolbec et al., 2000). Inorganic mercury can be converted to MeHg by microorganisms. The highest levels of MeHg are found in the predatory fish at the top of food chain due to bioaccumulation. More than 90% of the total mercury in certain fish tissues is in the form of MeHg (Bloom, 1992; Kim, 1995; USEPA, 2001). People obtain mainly through seafood, fishes and shellfish possess relatively high concentrations of mercury ranging from 0.04 to 1.0 μg/g (wet wt.) (NAS, 1978; Lodenius et al., 1983; Hakanson et al., 1990; Barghigiani and De Ranieri, 1992). The range for vegetables is less than 0.05 μg/g (dry wt.) (USPHS, 1994), and the concentration in drink water is less than 2.0 μg/l (Zumbroich, 1997). In Belgium, seafood intake contributes about 20% of bodily accumulation of mercury while in the US, seafood contributes about 85% (Galal-Gorchev, 1991). About 33% of the total daily mercury dose in the UK general population comes from seafood (MAFF, 1999). Populations that consume a large amount of fish regularly and frequently have the highest risk for MeHg toxicity (Klassen, 2001).

In 1997, the US Environmental Protection Agency (USEPA) recommended 0.1 μg kg^{-1} d^{-1} as a safe lifetime daily intake level of MeHg (USEPA, 1997). The action level set by the US Food and Drug Administration (USFDA) to regulate MeHg in commercial fish is 1.0 μg g^{-1}. They also indicated that mercury content is high in shark and swordfish, with the mean values around 1.0 μg g^{-1}. Burger et al. (2001) found there were significant differences in mercury levels in fish species and in human health risk based on ethnicity due to differences in the type

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of fish consumed. Kinetic models suggest that MeHg binds to glutathione in human blood and rat brain and that 3%–7% of the total body burden is found in the brain (Naganuma et al., 1980). One study found that the blood mercury levels of full-term and normal-weight children may be twice that of maternal blood. Such children present cerebral palsy, and delay in speech and motor activity (Amin-Zaki et al., 1979).

In Taiwan, a number of incidents of mercury contamination have been reported, especially in traditional Chinese herbs, in shark fin, and in a manufacturing plant pollution case. Traditional Chinese herbs have been used in Chinese society for thousands of years and many Taiwanese believe in the potency of Chinese herbal medicines to keep the body in good health. A Taiwan EPA follow-up investigation of the manufacturing plant pollution case found abandoned factories were seriously polluted with heavy metals, mercury, pentachlorophenol and dioxin. Soil and underground water in the surrounding areas showed severe pollution. The mercury concentration was 11 600 mg kg^{-1} in the soil, 580 times greater than the limit.

Since 2000, Pingdong Black Tuna Festival is held annually in May at Tungkung, Taiwan. The festival brings in several billion NT dollars of business opportunities to Pingdong each year. Blue fin tuna are very popular and sold at premium prices in Taiwan. Approximately, two-thirds of blue fin tuna caught by Taiwan fishing boats are for local consumption. The consumers love to eat raw fish in sashimi and sushi. Taiwanese eat fish as three or more meals per week, a level of consumption that increases with age (DOH, 1999). Several studies indicate that high fish-consumers eat fish three to four meals per week, while the highest consumers eat fish in six to eight meals per week (Lin and Sung, 2001). Although Taiwan is an island and a high fish-consuming country, very little information on the concentrations of mercury in seafood in Taiwan is available. In this study we investigate the total mercury concentration of different seafood. The purposes of this study were to assess fish intake in relation to the health risks of mercury exposure in typical consumers and high-seafood consumers. Further, to assess the uncertainty in risk assessment and the impact of these uncertainties on the estimation of expected risk of mercury intake from fish in Taiwan using the Monte Carlo technique.

2. Materials and methods

2.1. Sampling and procedures

In order to study the acceptable mercury intake from fish in Taiwan, the popular fishes of households were selected. Samples of seafood were randomly obtained from August 2003 to October 2004 at fish markets, seaports, and marine coastal areas of Taiwan. Samples came from one of three subgroups: fish, bivalve molluscs, and crustaceans. Samples were placed in a refrigerated container and transferred to the laboratory immediately after collection, and flesh of samples were obtained, cleaned, grinded down via a knife and pestle and the goal is to improve sample homogeneity. Approximately 5 g of sample were digested in flasks for 10 h with 5 mL nitric acid at 60 °C water-bath. After cooling, the residue fluid was diluted to 50 mL with distilled water.

2.2. Instrumentation

Mercury concentration was analyzed by a mercury analyzer (HG-200, Hiranuma, Mito, Japan). Certified reference material (CRM) SRM 1573a Oyster Tissue and DORM-2 were used to perform a standard material test to ensure the precision and accuracy of the seafood analyses. The precision was 105.1% and 101.8%, respectively, and accuracy values were 3.5% and 5.6%. The detection limit for Hg analysis was 0.04 μg g^{-1}.

2.3. Daily mercury exposure dose

Information on dietary intake of fish, bivalve molluscs, and crustaceans was obtained in our previous study. We defined typical consumers and high-seafood consumers as dietary intakes below 50 g d^{-1} and over 90 g d^{-1}, respectively (Chien et al., 2003). We modified Eq. (1) (USEPA, 1994) to predict daily mercury exposure dose.

$$E_\mathrm{m} = \frac{C_\mathrm{m} \times \mathrm{IR}}{\mathrm{BW}} \tag{1}$$

where E_m: dietary mercury exposure dose from seafood (μg kg^{-1} d^{-1}), C_m: mercury concentration in seafood (μg g^{-1}), IR: ingestion rate of seafood (g d^{-1}), BW: body weight (kg).

Eq. (2) used for calculating daily mercury exposure dose was:

$$E_\mathrm{m} = \frac{\left(\frac{\sum_{j=1}^{n}(C_{\mathrm{m}j} \times \mathrm{IR}_j)}{\sum_{j=1}^{n}\mathrm{IR}_j}\right)\left(\sum_{j=1}^{n}\mathrm{IR}_j\right)}{\mathrm{BW}} = \frac{\sum_{j=1}^{n}(C_{\mathrm{m}j} \times \mathrm{IR}_j)}{\mathrm{BW}} \tag{2}$$

where E_m: dietary mercury exposure dose from seafood (μg kg^{-1} d^{-1}), $C_{\mathrm{m}j}$: mercury concentration in species j seafood (μg g^{-1}), IR_j: ingestion rate of species j seafood (g d^{-1}), BW: body weight (kg).

2.4. Estimating acceptable daily ingestion rate of fish

We applied the Eq. (3) to predict the acceptable daily ingestion rates of fish for Taiwanese children and women of childbearing age based on geometric mean Hg concentration 0.06 μg/g (wet wt.). The definition for women of childbearing age means the women were 15–49 years old. Typical weights of these subgroups were obtained from DOH (2005).

$$\mathrm{IR}_\mathrm{f} = \frac{C_\mathrm{mf}}{\mathrm{BW} \times \mathrm{RfD}_\mathrm{f}} \tag{3}$$

Table 1
Input variables/parameter values used to define distributions for Monte Carlo simulation

Input variable	Symbol	Distribution
Hg concentration ($\mu g\ g^{-1}$)		
Fish	C_{mf}	Lognormal (Geomean = 0.06, GSD[a] = 2.74)
Bivalve molluscs	C_{mb}	Lognormal (Geomean = 0.06, GSD = 2.37)
Crustaceans	C_{mc}	Lognormal (Geomean = 0.016, GSD = 2.14)
Typical consumers ingestion rates ($g\ d^{-1}$)		
Fish	IR_{f}	Lognormal (mean = 37, 95th = 114)
Bivalve molluscs	IR_{b}	Lognormal (mean = 3.0, 95th = 11.6)
Crustaceans	IR_{c}	Lognormal (mean = 5.0, 95th = 14.5)
High-seafood consumers ingestion rates ($g\ d^{-1}$)		
Fish	$\mathrm{HIR}_{\mathrm{f}}$	Lognormal (mean = 90, 95th = 237)
Bivalve molluscs	$\mathrm{HIR}_{\mathrm{b}}$	Lognormal (mean = 10, 95th = 35)
Crustaceans	$\mathrm{HIR}_{\mathrm{c}}$	Lognormal (mean = 8.0, 95th = 21)
Body weight (kg)		
Male	BW_{m}	Normal (mean = 64.3, SD = 9.5)
Female	BW_{f}	Normal (mean = 54.5, SD = 9.4)

[a] GSD – Geometric standard deviation.

where IR_{f}: ingestion rate of fish ($g\ d^{-1}$), C_{mf}: average mercury concentration in fish ($\mu g\ g^{-1}$), BW: body weight (kg), $\mathrm{RfD}_{\mathrm{f}}$: USEPA's reference dose ($0.1\ \mu g\ kg^{-1}\ d^{-1}$).

2.5. Uncertainty and Monte Carlo analysis

The chi-square and Kolmogorov–Smirnov (K–S) statistics were used to optimize the goodness of fit of the distributions of mercury concentrations in seafood. Distribution of the body weight of male and female was fitted to data obtained from the Department of Health (DOH, 2005). The implemented parameter probability distributions are summarized in Table 1. To assess uncertainty in risk assessment and its impact on the estimation of expected risk, we used the Monte Carlo technique. We input individual distributions of exposure variables to generate output probability distributions of the health risk estimates (USEPA, 1997). The Monte Carlo simulation and estimation of sensitivity were performed using Crystal Ball software (version 2000.2, Decisioneering, Denver, CO, USA). The health risk was calculated from 10000 iterations of the risk model using randomly selected values derived from each probability distribution of the model parameters.

3. Results and discussion

3.1. Mercury concentration in seafood

The mercury concentrations of fish are summarized in Fig. 1. The highest mercury concentrations were in swordfish ($n = 58$, $0.77 \pm 0.83\ \mu g\ g^{-1}$ wet wt.) and shark ($n = 41$, $0.73 \pm 0.54\ \mu g\ g^{-1}$ wet wt.) and the lowest mercury concentration was in ocean sunfish ($n = 8$, $0.02 \pm 0.02\ \mu g\ g^{-1}$ wet wt.). The mercury concentrations of seafood decreased in the following order: fish ($n = 199$, 0.005–$4.69\ \mu g\ g^{-1}$ wet wt.) > bivalve molluscs ($n = 16$, oyster, clam, trochus; 0.021–$0.363\ \mu g\ g^{-1}$ wet wt.) > crustaceans ($n = 17$, shrimp, lobster, crab; 0.004–$0.035\ \mu g\ g^{-1}$ wet wt.). 7.5% of the shark and 34.7% of the swordfish samples mercury concentrations exceeded the Codex guideline level of $1\ mg\ kg^{-1}$ (FAO/WHO, 1991). According to the USFDA data, 36% of swordfish, 33% of shark, and nearly 4% of large tuna

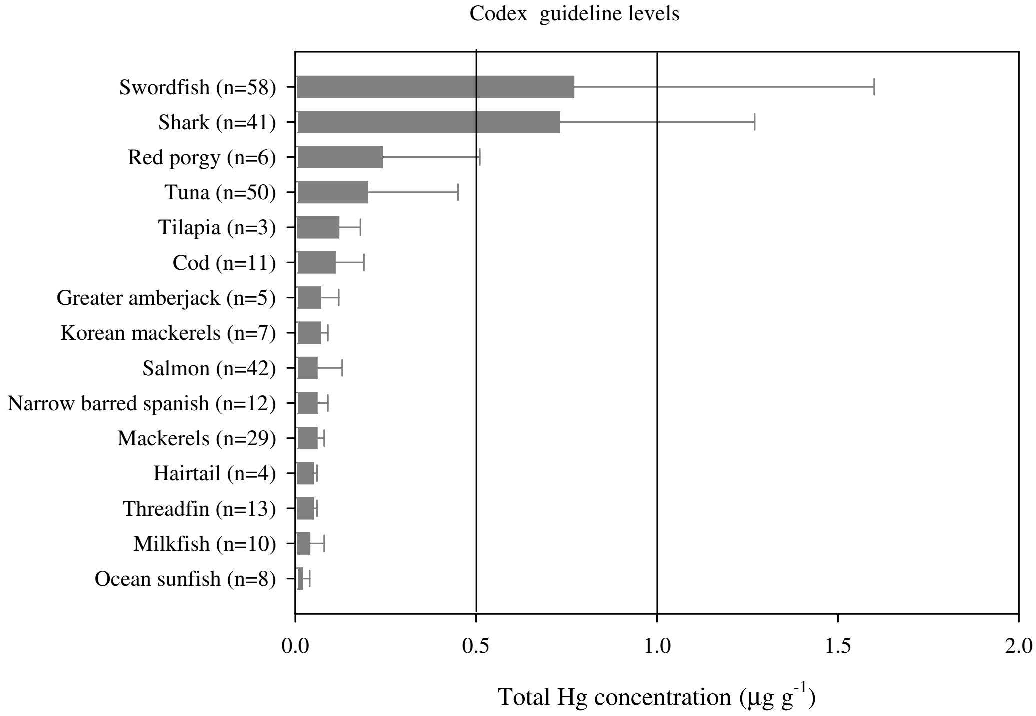

Fig. 1. Average mercury concentrations and standard deviation (error bars) for fish in comparison to FAO/WHO codex guideline levels.

sold commercially had MeHg concentrations exceeding 1 mg kg^{-1} (Bender and Williams, 2000). Because they are predatory fish they may accumulate mercury via bioaccumulation mechanism. In our study, the type of fish not belong to predatory fish are low Hg concentration.

In Japan, the mercury levels in predatory whales was 1.64–46.9 μg g^{-1} wet wt., in dolphins was 4.70–15.0 μg g^{-1} wet wt., and in filter-feeding whales was 0.02–0.10 μg g^{-1} wet wt. (Endo et al., 2003). Shark mercury concentrations in South East Brazil (94.0–17.9 μg g^{-1} wet wt.) and in the Mediterranean Sea (1.30–5.16 μg g^{-1} wet wt.) (Lacerda et al., 2000; Storelli et al., 2002) were higher than in our study (0.73 ± 0.54 μg g^{-1} wet wt.). In England, mercury levels were 1.0–2.2, and 0.15–2.7 μg g^{-1} wet wt. in shark and fresh/frozen swordfish, similar to our study (FSA, 2003).

3.2. Daily mercury exposure dose and acceptable ingestion rates

Fig. 2 shows the probability density distribution of predicted daily mercury exposure dose in male and female typical and high-seafood consumers. Based on the US ATSDR's minimal risk level of 0.5 μg kg^{-1} d^{-1}, the Monte Carlo simulation showed that 1.3%–2.1% and 7.1%–9.1% of the daily mercury exposure dose estimates exceeded the minimal risk level for the typical and the high-seafood consumers respectively. Whereas, based on USEPA's reference dose, 21.6%–24.3% and 45.6%–57.4% of the daily mercury exposure dose exceeded 0.1 μg kg^{-1} d^{-1}. Note that a daily mercury exposure dose exceeding the minimal level or reference dose may indicate that the most sensitive subgroups may suffer harmful effects over a lifetime,by consuming seafood or may be at risk for deleterious non cancerous effects from chronic exposure.

Fetuses, children, and women of childbearing age are more sensitive to the adverse effects of methylmercury toxicity. One study simulated daily mercury exposure dose for American women of childbearing age (15–49 years old) and children (1–4 years old). (Bangerter, 1998) For women of childbearing age, we found that only 3.3% of the daily mercury exposure dose estimates exceeded the USEPA's reference dose and none of the estimates exceeded the ATSDR's minimal risk level. However, for children

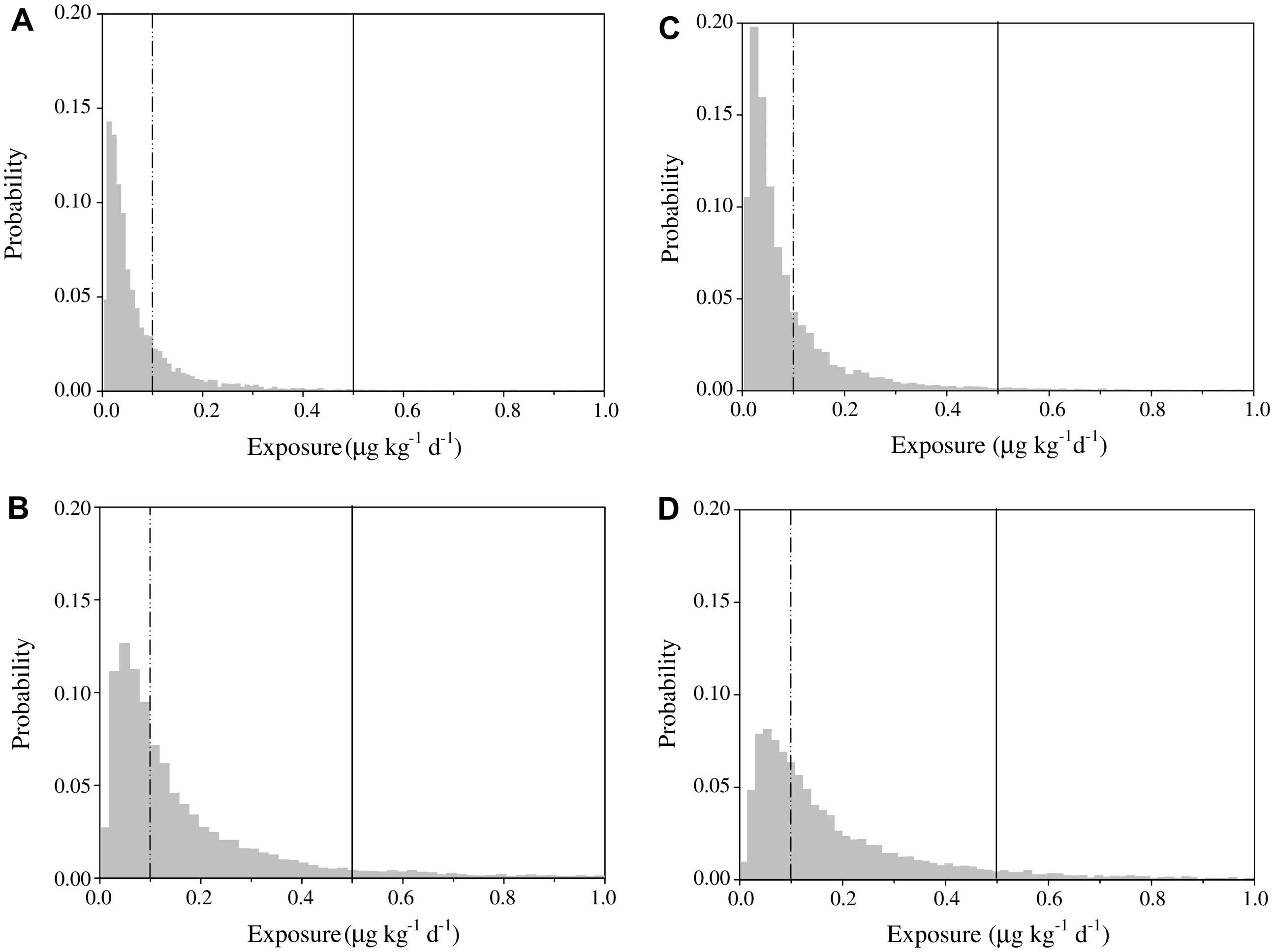

Fig. 2. Probability density distribution of predicted daily exposure dose in the typical and high-seafood consumers for males and females exposed to mercury. (A) Typical consumers, male; (B) high-seafood consumers, male; (C) typical consumers, female and (D) high-seafood consumers, female.

38.2% of estimates exceeded the USEPA's reference dose, and 0.3% exceeded the ATSDR's minimal risk level.

Fig. 3 presents the estimated acceptable ingestion rate of fish for Taiwanese children and women of childbearing age. Based on USEPA's reference dose and Hg concentration of fish, the acceptable ingestion rates are $<50\ \mathrm{g\ d^{-1}}$ and $90.8 \pm 15.7\ \mathrm{g\ d^{-1}}$, respectively. The USFDA and the USEPA advises pregnant women, women of childbearing age, nursing mothers, and young children to avoid eating fishes that contain high levels of mercury such as shark, swordfish, king mackerel, and tilefish. A new draft advisory from the USFDA is the first to specify mercury levels in tuna (Stephenson, 2004). A recent American study found that after the USFDA recommended that pregnant women limit their consumption of certain fish because of concerns about mercury contamination, pregnant women reported a reduction in total fishes consumption of ≈1.4 servings per month (95% confidence interval 0.7, 2.0) (Oken et al., 2003). In Taiwan, however the amount of fish consumed increased from eight meals per week before pregnancy, to eleven meals per week while pregnant (Liu, 2005).

In our study, tuna had the fourth highest mercury level ($0.2 \pm 0.25\ \mu\mathrm{g\ g^{-1}}$ wet wt.) (Fig. 1). Our previous study suggested that breast milk mercury concentrations in the city group did not differ significantly from the fishermen's group ($2.02\ \mu\mathrm{g\ l^{-1}}$ versus $2.04\ \mu\mathrm{g\ l^{-1}}$) likely because some mothers in the city group ate sashimi and sushi of swordfish, tuna, and salmon more frequently (Chien et al., 2006). However, Pingdong County's annual Black Tuna Festival is famous and the Taiwan government encourages the public to consume tuna because it brings in several billion NT dollars of business opportunities each year to Pingdong. The government neglects mercury toxicity in humans at all ages, particularly pregnant women in Taiwan who, because they consume predatory fish, are more likely to put their babies at risk for mercury exposure.

3.3. Sensitivity analysis

Table 2 indicates variables for daily mercury exposure dose for typical and high-seafood consumers. Hg concentration in fish is the key parameter which contributes to ≈65.1%–72.4% of output variances. The secondary parameter is the ingestion rate of fish; the contribution to variance ranged from 22.6% to 30.0%. These results might offer guidance for risk management.

Based on our results, reducing Hg concentration in fish and the ingestion rate of fish may be the most effective methods for decreasing daily mercury exposure dose. Amelioration and eventual elimination of mercury hazards require more effective application of control measures to prevent the disposal of industrial wastes into aquaculture estuarine and coasts may reduce Hg concentration in fish. Several studies have identified health benefits of eating fish such as reduction in various cardiovascular risk factors in adults, fatal and non-fatal coronary heart disease, myocardial infraction, and stroke (Kromhout et al., 1985; Gramenzi et al., 1990; Ascherio et al., 1995; Daviglus et al., 1997; Siscovick et al., 2000; Hu et al., 2002; Iso et al., 2001). The beneficial effects of fish consumption are attributed to omega-3 (n-3) polyunsaturated fatty acids (PUFA) found in fish. Balancing and managing the risk/benefit of fish consumption is now a visible public health issue. One study proposed a compound dose–response curve for the

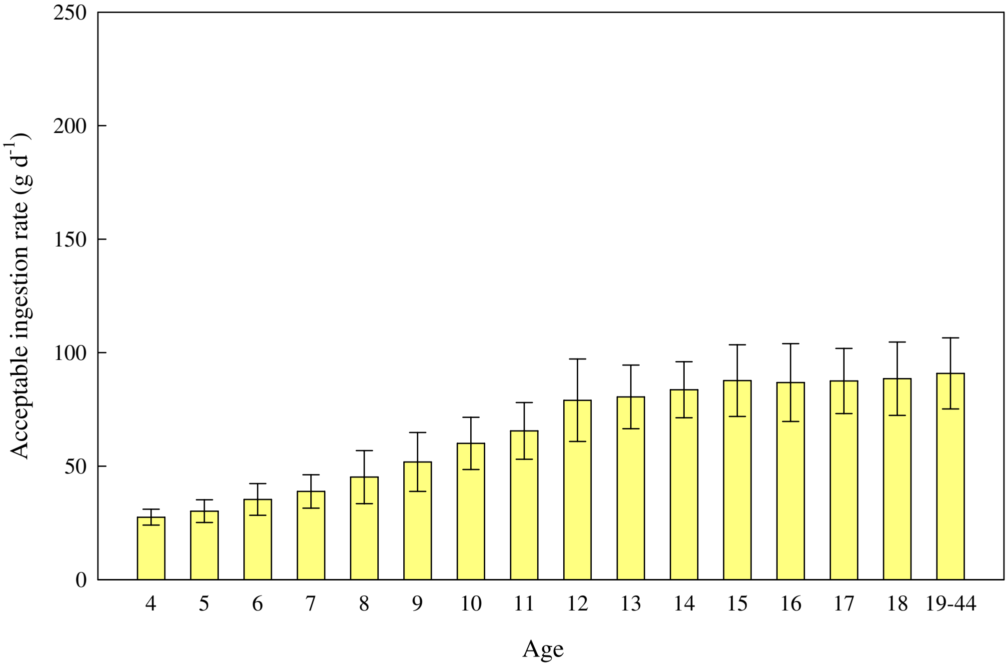

Fig. 3. Acceptable ingestion rates of fish for children and women of childbearing age base on USEPA's reference dose ($0.1\ \mu\mathrm{g\ kg^{-1}\ d^{-1}}$).

Table 2
Sensitivity analyses for daily mercury exposure dose for typical consumers (TC) and high-seafood consumers (HSC) using Monte Carlo simulation

	Rank correlation coefficient		Contribution to variance rank (%)	
	TC	HSC	TC	HSC
Males				
Hg concentration in fish	0.77 (1)	0.80 (1)	65.1 (1)	72.4 (1)
Ingestion rate of fish	0.52 (2)	0.46 (2)	30.0 (2)	22.6 (2)
Hg concentration in bivalve molluscs	0.12 (3)	0.15 (3)	1.6 (3)	1.8 (4)
Body weight	−0.12 (4)	−0.14 (4)	1.5 (4)	2.1 (3)
Females				
Hg concentration in fish	0.77 (1)	0.79 (1)	65.9 (1)	68.6 (1)
Ingestion rate of fish	0.50 (2)	0.47 (2)	27.7 (2)	24.4 (2)
Body weight	−0.16 (3)	−0.17 (3)	2.8 (3)	3.2 (3)
Hg concentration in bivalve molluscs	0.14 (4)	0.14 (4)	2.1 (4)	2.0 (4)

benefits and harm of fish consumption (Gochfeld and Burger, 2005). Based on the USEPA's reference dose $0.1\ \mu g\ kg^{-1}\ d^{-1}$, the fish intake threshold for the worst case of harm converts to $27\ g\ d^{-1}$, and $65\ g\ d^{-1}$ when the average MeHg concentration of fish is $0.23\ \mu g\ g^{-1}$ or $0.1\ \mu g\ g^{-1}$, respectively. It is apparent that the risk/benefits of eating fish are communicated to the public and government should provide appropriate information such choosing fish in low MeHg and high PUFA.

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