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Mercury and Methylmercury Levels in Marine Fish Species from Korean Retail Markets

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Abstract Mercury and methylmercury concentrations were measured in 225 marine fish samples purchased from retail markets in 5 major Korean cities in 2005. The results showed that the concentrations of mercury and methylmercury ranged from 0.006-0.455 (mean=0.054±0.025) mg/kg and 0.004-0.120 (mean=0.021±0.008) mg/kg, respectively, in fish caught from the coastal waters of the Korean Peninsula. The concentrations in deep-sea fish including tuna ranged from 0.066-2.315 (mean=0.095±0.640) mg/kg for mercury and 0.027-0.897 (mean=0.281±0.182) mg/kg for methylmercury. The estimated weekly intakes (EWIs) of mercury and methylmercury reached 3.92 and 4.70% of the provisional tolerable weekly intakes (PTWI), respectively. Consequently, the levels of mercury and methylmercury in marine fish from current retail markets do not pose any significant health risks for Koreans.

Keywords: mercury, methylmercury, fish, estimated weekly intake (EWI), provisional tolerable weekly intake (PTWI)

Introduction

Mercury is deposited widely in the environment through both natural and human activities and can be found in three forms: elemental, inorganic, and organic. In aquatic environments, inorganic mercury is converted into methylmercury, the most common form of organic mercury, by anaerobic bacteria present in sediment (1-10). Methylmercury tends to accumulate in fish according to a number of key factors, including the age and weight of the fish, environmental conditions, and food sources. Predatory fish are likely to contain higher levels of methylmercury (1,2) and fish and seafood are considered to be the main source of methylmercury in the human diet (3).

Methylmercury can induce toxic effects in several organ systems including the nervous system, kidney, liver, and reproductive organs (4). The Food and Agriculture Organization (FAO) of the United Nations and World Health Organization (WHO) joint expert committee on food additives (JECFA) has recently revised the provisional tolerable weekly intake (PTWI) of methylmercury, which is the tolerable intake established for the population, to protect the most sensitive subgroups of the population from toxic effects (5,6). Because the developing fetus is considered to be the most sensitive subgroup, in the case of methylmercury the current PTWI of 1.6 μg/kg body weight (BW) is considered sufficient to protect this subgroup.

National food safety agencies have issued guidelines regarding reductions in the consumption of predatory fish by more vulnerable groups such as children, pregnant women, and women of childbearing age. Australia has advised eating one or two fish meals per week, with a serving size between 80 and 120 g. The Food and Drug Administration (FDA) of the United States and other food safety authorities have also advised women who may become pregnant, pregnant women, nursing mothers, and young children may reduce methylmercury exposure from high levels of mercury in fish and shellfish (2-10).

Codex (11) and the US FDA (12) set a maximum level of 1 mg/kg methylmercury in predatory fish, and the European Committee (13) set maximum levels of mercury (0.5 mg/kg) and methylmercury (1.0 mg/kg) depending on the fish species. Japan set maximum levels of 0.4 mg/kg mercury and 0.3 mg/kg methylmercury in all fish excluding deep-sea fish such as tuna (14). Recently, South Korea has revised the maximum level for non-deep-sea fish to 0.5 mg/kg (15) of mercury, and for deep-sea fish including shark, swordfish, and tuna to 1.0 mg/kg of methylmercury. However, this regulation will be reprieved till 2009 (16).

In this study, the methylmercury and total mercury levels in marine fish from Korean retail markets were investigated and the average intakes were estimated to support a scientific risk analysis.

Materials and Methods

Materials A total of 255 samples from 15 species of marine fish were collected in the forms sold in the following 5 major Korean cities: Seoul, Busan, Gwangju, Daejeon, and Gangneung in 2005. After discarding the inedible components, the fish were ground using a stainless steel blender in order to obtain a consistent homogeneity and the ground samples were stored in a freezer (–20°C) until the time of analysis.

Reagents All chemicals were purchased as reagent grade from Sigma-Aldrich Co. (St. Louis, MO, USA). Standard solutions were prepared according to Kim *et al.* (17).

Mercury and methylmercury content determination Both mercury (Hg) and methylmercury (MeHg) analyses

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were conducted as described in Kim *et al.* (17). A mercury analyzer (model SP-3D; Nippon Instrument Co., Osaka, Japan) and a gas chromatograph with an electron capture detector (ECD, model 6890A; Agilent, Palo Alto, CA, USA) were used.

Estimated weekly intake (EWI) of mercury and methylmercury EWIs of mercury and methylmercury were calculated based on mercury or methylmercury levels from 15 species of marine fish and their consumption amounts reported in National Health and Nutrition Survey (18). A weight of 60 kg was assumed for Korean adults. The mean intake and 95th percentile intake of daily exposure were obtained using Crystall ball® 7 (Version 7.3, Oracle crystal ball business unit; Denver, CO, USA), purchased from the Eretec Company (Seoul, Korea). The PTWI of mercury and methylmercury, which are safety threshold limits of 5 and 1.6 μg/kg BW/week set by JECFA, were compared with the obtained EWIs.

Results and Discussion

Mercury contents in fish Table 1 indicates the levels of mercury and methylmercury and the ratios of methylmercury to mercury in each fish species examined. One-hundred ninety-five marine fish caught off the coast of South Korea contained from 0.006 to 0.455 mg/kg of mercury and none of the fish groups examined exceeded the mercury limit of 0.5 mg/kg. However, Japanese red sea bream contained a rather wide range of methylmercury levels (0.011-0.120 mg/kg), though the proportion of methylmercury to mercury was rather low, ranging from 18.9 to 31.9%, as compared to other fish species in which the proportion of methylmercury to mercury reached 85.4%. Considerably high levels of mercury were detected from 20 deep-sea fish including tuna, which exceeded the mercury limit of 0.5 mg/kg, but none of these species exceeded the methylmercury limit of 1.0 mg/kg (Table 2). Mercury and methylmercury levels in deep-sea fish including tuna ranged from 0.066 to 2.312 and 0.037 to 0.897 mg/kg, respectively, while the proportions of methylmercury to mercury ranged from 11.0 to 98.7%.

It is known that pacific mackerel, currently marketed in South Korea, reaches a length of about 25 cm within 1 year and grows 40-50 cm in 5 years, but the lengths of marketed deep-sea fish species such as tuna are much longer and

Table 1. Mercury (Hg) and methylmercury (MeHg) levels in marine fish species (excluding deep-sea fish)

Species		No. of samples	Hg (mg/kg)[1)]	MeHg (mg/kg)	% Hg as MeHg
Scientific name	Common name				
Todarodes pacificus	Japanese common flying squid	15	0.038-0.074 (0.058±0.011)	0.018-0.034 (0.024±0.004)	34.0-52.4 (42.6±5.5)
Scomber japonicus	Pacific mackerel	15	0.020-0.183 (0.059±0.052)	0.014-0.070 (0.028±0.021)	37.7-72.0 (52.5±9.6)
Larimichthys polyactis	Small yellow croaker	15	0.021-0.081 (0.040±0.016)	0.012-0.030 (0.019±0.005)	29.6-68.0 (50.3±10.5)
Trichiurus lepturus	Hair-tail	15	0.017-0.145 (0.066±0.035)	0.011-0.062 (0.026±0.013)	30.4-69.6 (43.0±11.0)
Paralichthys olivaceus	Bastard halibut	15	0.023-0.081 (0.044±0.015)	0.012-0.038 (0.020±0.007)	34.2-68.9 (45.7±9.8)
Scomberomorus niphonius	Japanese spanish mackerel	15	0.013-0.053 (0.027±0.012)	0.009-0.015 (0.013±0.002)	28.5-81.8 (53.3±15.9)
Octopus minor	Minor octopus	15	0.006-0.048 (0.022±0.013)	0.004-0.017 (0.008±0.004)	28.4-67.1 (41.5±11.5)
Cololabis saira	Pacific saury	15	0.047-0.082 (0.061±0.010)	0.014-0.051 (0.033±0.011)	25.8-85.4 (54.4±14.5)
Sebastes hubbsi	Black rockfish	15	0.036-0.104 (0.060±0.019)	0.014-0.043 (0.020±0.008)	23.6-63.7 (34.4±9.3)
Pleuronectes herzenstein	Brown sole	15	0.015-0.113 (0.045±0.024)	0.009-0.031 (0.013±0.006)	20.0-60.0 (33.1±11.1)
Lophiomus setigerus	Angler fish	15	0.017-0.068 (0.032±0.013)	0.010-0.021 (0.012±0.003)	28.6-59.5 (41.3±9.5)
Pagrus major	Japanese red sea bream	15	0.047-0.455 (0.121±0.116)	0.011-0.120 (0.033±0.031)	18.9-31.9 (27.5±3.5)
Gadus macrocephalus	Pacific cod	15	0.016-0.127 (0.072±0.032)	0.008-0.030 (0.019±0.008)	13.1-49.2 (28.8±10.1)
Total		195	0.006-0.455 (0.067±0.067)	0.004-0.120 (0.021±0.008)	13.1-85.4 (44.3±10.1)

[1)]The values are expressed as min-max (mean±SD) of data.

Table 2. Mercury (Hg) and methylmercury (MeHg) levels in deep-sea fish species (bigeye tuna and swordfish)

Species		No. of samples	Hg (mg/kg)[1)]	MeHg (mg/kg)	% Hg as MeHg
Scientific name	Common name				
Deep-sea fish	Bigeye tuna	15	0.066-1.565 (0.493±0.453)	0.027-0.420 (0.152±0.097)	19.7-~98.7 (42.9±20.6)
	Swordfish	15	0.567-2.315 (1.407±0.480)	0.122-0.897 (0.410±0.217)	11.0-53.4 (28.9±11.0)
Total		30	0.066-2.315 (0.950±0.640)	0.027-0.897 (0.281±0.182)	11.0-98.7 (35.9±17.8)

[1)]Values are expressed as min-max (mean±SD) of data.

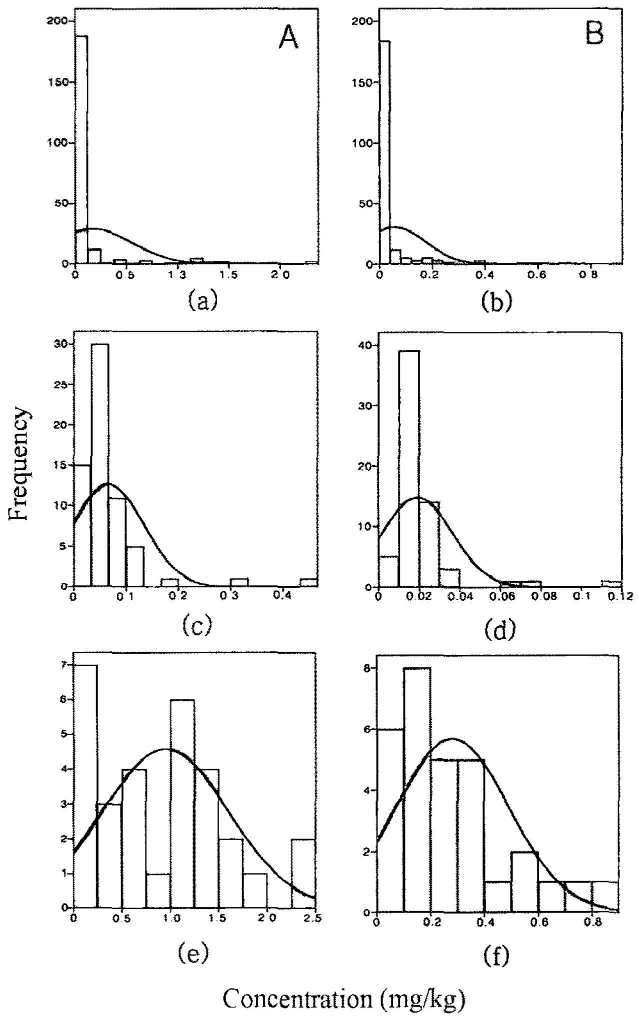

Fig. 1. Levels of mercury (A) and methylmercury (B) in marine fish. (a) (b), All fish; (c) (d), fish excluding deep-sea fish; (e) (f), deep-sea fish (bigeye tuna and swordfish).

vary from 1-4 m (19). A number of studies have reported a positive correlation between fish size and mercury levels indicating the bioaccumulation of mercury with age (4). Though regulatory limits for mercury in fish are set in terms of total mercury, there are a number of reports in the literature including a recent JECFA meeting report demonstrating that methylmercury in fish has toxicological effects (4).

Crépet *et al.* (20) had suggested that the concentrations of methylmercury might be assumed from corresponding mercury concentrations by applying the following conversion factors to analytical data on the mercury content in different foods: 0.84 for fish, 0.43 for mollusks, and 0.36 for shellfish. On the basis of our results, it is logical that longer fish seem to build up higher levels of mercury in their tissues but it is difficult to extrapolate the concentration of methylmercury from the concentration of mercury due to the inconsistencies and variabilities that exist in fish lengths and weights.

Many food safety authorities have established that a 2-tiered approach in management policy may be applied to reduce public exposure to methylmercury. Figure 1 indicates that the levels of mercury and methylmercury in marine fish excluding deep-sea fish showed a normal distribution curve with a long tailing. However, the levels of mercury in deep-sea fish species exhibited small bumps above 0.5 mg/kg (Fig. 1a).

Therefore, it may be reasonable to have 2 separate mercury maximum limits for general marine fish and deep-sea fish. In addition, the long term monitoring of commonly consumed fish, and monitoring of imported and exported fish and seafood for mercury content in order to generate comparisons with the maximum limits of methylmercury should be carefully planned to keep highly contaminated products out of the food supply.

Assessment of EWI of mercury and methylmercury in fish The main source of human exposure to methylmercury is through the consumption of fish and the exposure can be estimated from fish consumption habits and estimates of the average portions of fish usually consumed coupled with determinations of the mercury content in fish consumed by the population (4). Recently, a Korean total diet study has reported that mercury intake in the fish and seafood group was higher than in other commodities and that it was higher in the summer and autumn seasons (21).

Distributions for the daily intake of mercury and methylmercury in fish were plotted and Fig. 2 shows marine fish species that were reported to have a higher rate of consumption according to the National Health and Nutrition Survey (18). Based on the high mercury contents detected in bigeye tuna in the 95th percentile intake, the highest intake was estimated to be 0.46 μg/day, however, methylmercury levels of the 95th percentile intake were estimated to be similar to those of the Japanese common flying squid.

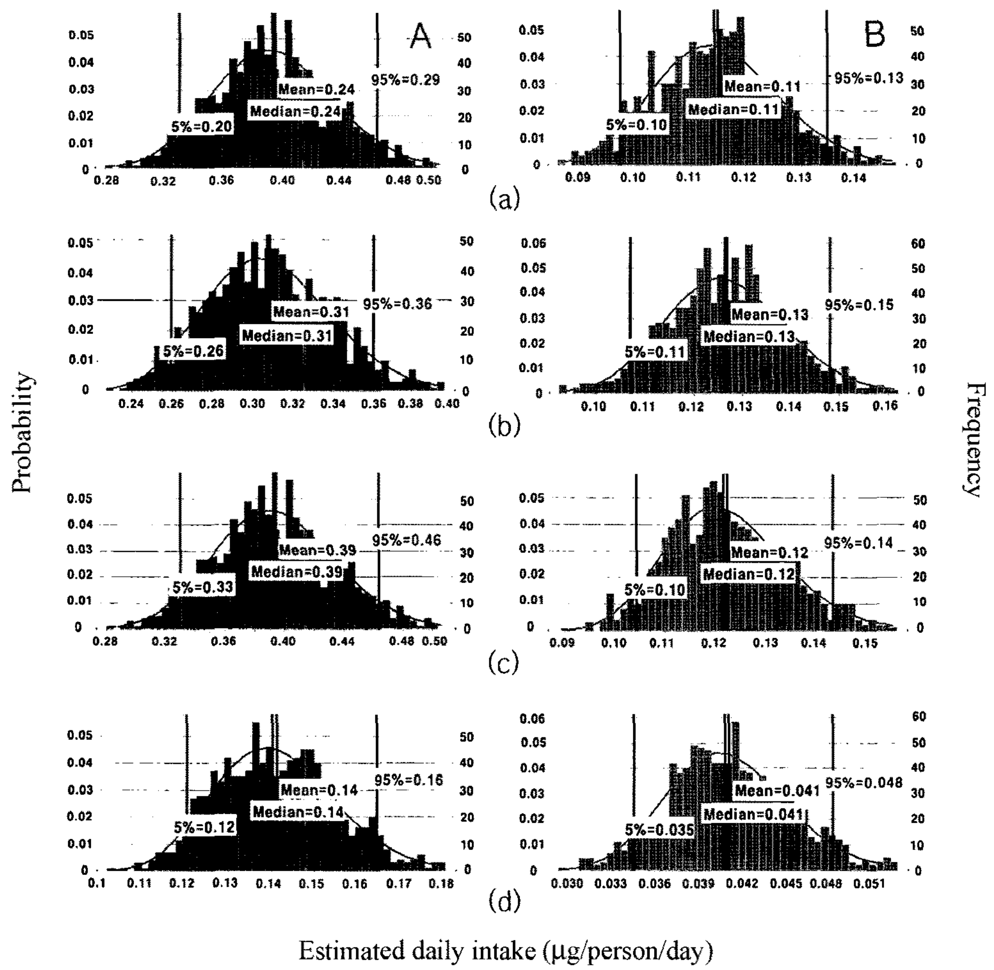

Fig. 2. Distribution for daily exposures of mercury (A) and methylmercury (B) in various fish species estimated using Crystall ball® 7. (a) Pacific mackerel; (b) Japanese common flying squid; (c) bigeye tuna; (d) swordfish.

Table 3. Estimated weekly intakes of mercury (Hg) and methylmercury (MeHg) from marine fish compared with the provisional tolerable weekly intake (PTWI) determined by JECFA

Samples	Mean content (mg/kg)		Food intake (g/day)	Estimated daily intake (μg/person/day)		Estimated weekly intake (μg/kg BW[1]/week)		% of PTWI[2]	
	Hg	MeHg		Hg	MeHg	Hg	MeHg	Hg	MeHg
Japanese common flying squid	0.058	0.024	5.3	0.308	0.127	0.036	0.018	0.72	1.13
Pacific mackerel	0.059	0.028	4.1	0.241	0.115	0.028	0.016	0.56	1.00
Small yellow croaker	0.040	0.019	2.7	0.108	0.051	0.013	0.007	0.25	0.44
Hair-tail	0.066	0.026	2.4	0.158	0.062	0.018	0.009	0.37	0.56
Bastard halibut	0.044	0.020	1.2	0.053	0.024	0.006	0.003	0.12	0.19
Japanese spanish mackerel	0.027	0.013	1.0	0.027	0.013	0.003	0.002	0.06	0.13
Minor octopus	0.022	0.008	0.9	0.020	0.007	0.002	0.001	0.05	0.06
Pacific saury	0.061	0.033	0.8	0.048	0.026	0.006	0.004	0.11	0.25
Black rockfish	0.060	0.020	0.7	0.042	0.014	0.005	0.002	0.10	0.13
Brown sole	0.045	0.013	0.6	0.027	0.008	0.003	0.001	0.06	0.06
Angler fish	0.032	0.012	0.6	0.019	0.007	0.002	0.001	0.04	0.06
Japanese red sea bream	0.121	0.033	0.5	0.061	0.017	0.007	0.002	0.14	0.13
Pacific cod	0.072	0.019	0.5	0.036	0.010	0.004	0.001	0.08	0.06
Bigeye tuna	0.493	0.152	0.8	0.394	0.122	0.046	0.017	0.92	1.06
Swordfish	1.407	0.410	0.1	0.14	0.041	0.016	0.006	0.33	0.38

1)Body weight (mean) of adults (60 kg).
2)Established by JECFA (Hg=5 μg/kg BW, MeHg=1.6 μg/kg BW).

The average estimated weekly intake of mercury and methylmercury for marine fish including tuna are described in Table 3. While the contribution of mercury intake from tuna was still recognized as the largest, Japanese common flying squid showed a relatively larger contribution than other fish groups for mercury and methylmercury intake in the Korean diet, which were estimated at 0.72% of the PTWI for mercury and 1.13% of the PTWI for methylmercury. Based on these results, other risk management options may not be considered since deep-sea fish species did not show levels exceeding 10% of the PTWI, which the FAO/WHO consultation has recommended (5).

Consequently, the results showed that the mercury and methylmercury levels in marine fish from retail markets do not pose any significant health risks for Koreans. Several national food safety agencies have made recommendations regarding fish consumption limits, particularly for predatory fish consumption but the US FDA and Environmental Protection Agency (EPA) of the United States still emphasize the benefits of eating fish; consumers should know that fish and shellfish can be important parts of a healthy and balanced diet. Therefore, it is necessary that approaches to reduce the methylmercury exposure from fish focus on reducing emissions and other human activities that contribute to mercury levels in the environment and on developing appropriate consumer policies and communication tools to influence fish consumption.

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