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TOTAL MERCURY INTAKE FROM FISH AND SHELLFISH BY JAPANESE PEOPLE

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ABSTRACT

Elevated mercury concentrations have been reported in fish in recent years. Japanese people eat a great deal of raw fishes and shellfishes as "Sashimi" and "Sushi". The action level of large predatory fish such as tuna with total mercury levels exceeding the Japanese maximum permitted limit of 0.4 ppm is exempted from regulation in Japan. Therefore, current total mercury intake from fish and shellfish of Japanese people is unknown. The purpose of this investigation was to estimate the total mercury intake from fish and shellfish. It was found that the mean total mercury concentration of 1.11 ppm in tuna of eatable base as Sashimi or Sushi was clearly higher than the normal level. The mean total mercury intake from fish and shellfish was 0.17mg per capita per week . According to the hypothesis that 75% of total mercury in fish and shellfish is methylmercury, the weekly intake of 0.13 mg as methylmercury was corresponding amount to about 74% of provisional tolerable weekly intake 0.17 mg of methylmercury set by the Welfare Ministry of Japan. ©1997 Elsevier Science Ltd

INTRODUCTION

In recent years, elevated mercury concentrations have been reported in fish in several countries (1-3). Mercury in fish occurs almost entirely as methylmercury in muscle tissue, where it is associated with protein sulfhydryl groups. Fish consumption is the primary pathway of human exposure to methylmercury. Methylmercury is highly toxic. The Japanese mean consumption of fish and shellfish is about 100g (eatable base) per capita per day. In particular, Japanese people eat a great deal of raw fishes and shellfishes as "Sashimi" and "Sushi". Slices of raw fishes are called Sashimi in Japanese. Sushi is rice cakes covering raw fish or rolled in seaweed and

sprinkled with vinegar. The mean fish consumption is over 5 meals(about 100 g as main dish at a meal) of sliced raw fishes per week among Japanese people living near the sea. The U.S.Food and Drug Administration(FDA) has set an Action Level of 1ppm(wet weight) for concentration of mercury in fish. Fish containing concentrations of mercury above this level are considered to be hazardous for human consumption and cannot be sold in interstate commerce. In Japan, fish containing total mercury concentrations exceeding the Japanese maximum permitted limit of 0.4 ppm (wet base, as Hg) is commonly considered unsuitable for human consumption. Large predatory long-lived marine species such as tuna and swordfish show frequently mercury concentrations higher than 1 ppm (wet base, as Hg) and almost all of it is methylmercury (4, 5). However, large predatory fishes (*Thunnidae, Xiphias gladius, Katsuwonus pelamis, et al.)* with total mercury levels exceeding the Japanese Provisional Action Level of 0.4 ppm for fish and shellfish were exempted from regulation providing the Action Level in July,1973, in Japan. Therefore, from 1976 to this day, the official inspection on mercury concentration in fish and shellfish has not been reported in the public research institutes. Therefore, current total mercury intake of Japanese people from fish and shellfish is unknown. The purpose of this investigation is to estimate the total mercury intake from fish and shellfish.

SAMPLING AND ANALYTICAL METHODS

Raw fish and shellfish of eatable base available in the market at Tokyo and its surrounding areas was used as samples for mercury investigation. Samples were collected during the one-year period from April,1996 to March,1997.

Total mercury contents in fish and shellfish samples were determined by using a cold flameless atomic absorption method. Namely, one sample of about 10 mg was placed on a boat in an electric furnace pre-heated at 700°C and was heated at 700°C for 6 minutes to vaporize mercury. Vaporized mercury was collected in a trap tube. The trap tube was rapidly heated for 5 minutes at 650°C to release mercury and to carry released mercury from the tube to a cold flameless atomic absorption spectrophotometer. Detection limit by this analytical method is 0.01 ng and reproducibility in 1-10 ng contents is ±1%.

RESULTS AND DISCUSSION

Total mercury concentrations in fish and shellfish of eatable base

The total mercury concentrations in fish and shellfish of 28 species, 360 samples(eatable base) collected from markets during the one-year are showed in Table 1. Mercury concentrations were quite variable, depending on fish species and sizes. Large fishes such as tuna and swordfish presented the highest mercury concentrations among the fish and shellfish collected, with 93% of tuna, with 100% of swordfish and with 33%(120 samples) of all samples presented concentrations higher than the Japanese maximum permissible total mercury limit of 0.4 ppm set by the Health Organization of Japanese Welfare Ministry for human consumption. In 1975, the mean

total mercury concentrations in tuna and swordfish by the statistical report of the Japanese Prime Minister's Office were 0.60 ppm and 0.80 ppm respectively. A comparison of the results, obtained in 1975 and 1996, suggests that total mercury concentrations in these fishes are increasing, particularly in large predatory species.

Table 1 Total mercury concentrations in fish and shellfish of eatable base

Fish and shellfish species: *Scientific name*	Number of samples	Range (ppm)	Mean (ppm)	Eatable condition	Cases exceeding 0.4ppm (%)
Tuna: *Thunnus thynnus*	58	0.36–5.25	1.11	Raw	93
Swordfish: *Xiphias gladius*	34	1.15–3.01	1.82	Roast	100
Bonito: *katsuwonus pelamis*	18	0.12–0.41	0.25	Raw	11
Yellow tail: *Seriola dorsalis*	6	0.20–0.22	0.21	Boiled	0
Young yellow tail	8	0.06–0.76	0.26	Raw	25
Seabass: *Seriola purpurascens*	6	0.04–0.37	0.20	Raw	0
Salmon: *Onchorhynchus*	32	<0.01–1.26	0.19	Roast	13
Salmon roe	4	1.30–1.97	1.64	Salted raw	100
Mackerel: *Scomber scombrus*	4	0.11–0.43	0.27	Roast	50
Herring: *Clupea pallasi*	2	0.42–0.66	0.54	Roast	100
Saurel: *Trachurus symmetricus*	6	0.01–0.72	0.32	Roast	33
Conger: *Conger conger*	6	0.07–0.42	0.21	Boiled	33
Cod: *Gadus callarias*	8	0.01–0.07	0.04	Boiled	0
Cod roe	4	0.10–1.14	0.62	Salted raw	50
Flatfish: *Heterosomata*	8	0.07–0.26	0.20	Roast	0
Smelt: *Osmerus eperlanus*	8	<0.01–1.25	0.51	Roast	50
Sardine: *Sardina pilchardus*	20	<0.01–0.10	0.01	Roast	0
Cutlassfish: *Trichiurus lepturus*	12	0.20–0.33	0.25	Roast	0
Seabream: *Archosagus rhomboidalis*	8	0.08–0.37	0.17	Roast	0
Pacific saury: *Cololabis saira*	10	<0.01–0.07	0.04	Roast	0
Atka mackerel: *Pleurogrammus azonus*	4	<0.01–0.16	0.08	Roast	0
Cuttlefish: *Sepia officinialis*	20	<0.01–0.88	0.15	Raw	10
Octopus: *Octopoda*	10	<0.01–0.20	0.04	Raw	0
Prawn: *Palaemon serratus*	22	<0.01–1.31	0.23	Raw	9
Scallop: *Pecten irradians*	20	<0.01–0.46	0.09	Raw	10
Turban shell: *Turbo cornutus*	6	0.01–0.06	0.03	Roast	0
Ark shell: *Arcidae*	8	<0.01–0.04	0.03	Raw	0
Abalone: *Haliotidae*	6	0.01–0.04	0.03	Raw	0
King crab: *Paralithodes camtschatica*	2	0.01	0.01	Raw	0

Total mercury intake from foods by Japanese people

Changes of total mercury intake from food are shown in Table 2. The data in 1975 were quoted from an estimate by Fujii (6). The data in 1996 represent the results of our analyses. A comparison of the results obtained in 1975 and 1996, although the intakes of total food were nearly constant, the intake of fish and shellfish in 1996 increased by 1.7 times greater than that of 1975, and the mean total mercury concentrations in fish and shellfish also increased by 1.5 times. Therefore, the total mercury intakes from food and fish and

shellfish increased by 1.14 and 2.48 times respectively. The ratio of total mercury intake from fish and shellfish in 1975 was about 45% of total mercury intake from all foods, but the ratio in 1996 is over 97%.

Table 2. Change of mean total mercury intake from food of Japanese people per capita per day

	1975			1996		
	A daily mean intake of food (g)	Mean mercury content (ppm)	Mean total mercury intake (μg)	A daily mean intake of food (g)	Mean mercury content (ppm)	Mean total mercury intake (μg)
Total fishes and shellfishes	**95.6**	**0.101**	**9.70**	**163.3**	**0. 148**	**24.09**
Tuna,Swordfish,Bonito	5.7	0.600	3.42	8.8	1.19	10.46
Other fishes	34.5	0.079	2.71	33.1	0.256	8.49
Cuttlefish,Octopus,Crustacea	16.7	0.037	0.61	22.6	0.152	3.44
Other marine products	38.7	0.076	2.96	98.8	0.050	1. 70
Cereals	**395**	**0.0097**	**3.85**	**359**	**0.0008**	**0.28**
Vegetables and fruits	**621**	**0.0088**	**5.44**	**576**	**0.0004**	**0.21**
Meats,Eggs, Dairy products	**200**	**0.011**	**2.10**	**247**	**0.0006**	**0.16**
Others	**65**	**0.0078**	**0.51**	**45**	**0.00004**	**0.002**
Total intake of food and mercury per capita per day	**1,377**	**0.016**	**21.65**	**1,326**	**0. 019**	**24.74**

Methylmercury intake from food

In the present study, methylmercury in food was not determined. However, from the aspects of environmental toxicology and food chain impacts, methylmercury is important. Fish consumption is the primary of human exposure to methylmercury. Mercury in fish occurs almost entirely as methylmercury in muscle tissue, where it is associated with protein sulfhydryl groups. Ingestion of fish muscle is an important exposure pathway of mercury to humans. Methylmercury is highly toxic. It is thought to inhibit enzyme activity in the cerebellum, which is responsible for neuron growth in early developmental stages. Chronic exposure to organomercurials can result in mental retardation (7). As a result, the Japanese Welfare Ministry has set an Action Level of 0.3 ppm for concentration of methylmercury in fish and shellfish, that is 75 % of the Japanese maximum permitted limit of total mercury 0.4 ppm. According to the hypothesis that 75 % of total mercury in fish and shellfish and 10 % of total mercury in other food groups are methylmercury, total methylmercury intakes from food and, fish and shellfish were estimated at 18.13μg (as Hg) and 18.07μg (as Hg, correspond to 99.6 % in total methylmercury in food) per capita per day respectively.

On the other hand, the Health Organization of Japanese Welfare Ministry has set intake limit of total mercury as 0.25 mg and methylmercury as 0.17mg per an adult weighing 50 kg. Namely, the weekly intake of 0.126 mg

(0.018mg×7day) as methylmercury per capita is corresponding amount to about 74 % of the provisional tolerable weekly intake 0.17 mg.

Also, the administration limit of methylmercury can be showed by the following experimental equation (6): Acceptable daily intake×biological half time(BHT)×1.44=0.17(mg)÷7(day)×70(day)×1.44=2.5(mg). Accordingly, the mean administration value of methylmercury of Japanese people is 0.018×70×1.44=1.8mg This value corresponds to about 72 % of the administration limit of methylmercury.

If one consumes 100 g (the normal diet at a meal) of tuna as Sashimi containing a total of 1.11 ppm mercury which is 2.8 times the Japanese maximum permitted limit of total mercury 0.4 ppm for human consumption, then this 111 μg Hg (almost all of it is methylmercury) can be estimated at 11 mg as administration value in the equation described above. If one consumes 200 g of tuna, then this 22 mg of methylmercury containing in tuna is very close to the administration limit 25 mg of minimum threshold value in methylmercury poisoning (8). It may be a significant body load and an important human health risk. Because this high level from human consumption of large fish is a daily occurrence in Japan, and is not subject to any effective means of reduction other than eliminating favorite foods from national diets. Further studies are needed to elucidate these phenomena, that is, detailed epidemiological information such as eating habits and amounts of fish and their species commonly consumed should be more deeply investigated in Japanese people.

REFERENCES

(1) Mason,R.P. and Sullivan, K.A., *Environ.Sci.Technol.* **31**, 942-947(1997).

(2) Science News reports in *Environ.Sci.Technol.***30**,154A,193A,and 331A(1996).

(3) Driscoll,C. T., Yan, C., Schofield,C.L., Munson, R.,and Holsapple,J.,*Environ.Sci.Technol.* **28**,,136A(1994).

(4) Silver,S., Endo, G.,and Nakamura, K., *Mizu Kankyo Gakukaisi(Tokyo)*,**17**,235-243(1994).

(5) Nippon Kagaku Kai ed. *Mercury*, Maruzen, Japan, 192(1977).

(6) Kitamura,S., Kondo,M., Takizawa,T., Fujii,M., *Mercury*, Kodansha, Japan, 267-273,(1976).

(7) Kurzel R. B.,and Cetrulo C.L., *Environ.Sci.Technol.***15**, 626-640(1981).

(8) Saito, K., ed., *Health and Environment*, Asakura, Japan, 94, (1984).