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High mercury levels in hair samples from residents of Taiji, a Japanese whaling town

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ABSTRACT

We investigated the mercury concentrations in red meat from pilot whales consumed by some residents of the Japanese whaling town, Taiji, and in hair samples from 50 residents for their maker of mercury burden. The methyl mercury (M-Hg) level in the red meat was 5.9 μg/wet g, markedly higher than the US FDA action level and Cordex Alimentarius guideline level for predatory fish (1.0 μg/wet g). The average level of total mercury (T-Hg) in the hair from residents who ate whale meat more than once a month was 24.6 μg/g, whereas the average from the residents who did not consume any whale meat was 4.3 μg/g. The T-Hg concentrations in the hair from three donors exceeded 50 μg/g, the level for NOAEL set by WHO. The T-Hg level found in the Taiji whale meat consumers was markedly higher than that observed in the Japanese population overall (about 2 μg/g).

1. Introduction

Mercury (Hg) is distributed through the environment by both natural and anthropogenic processes. Inorganic Hg released into the environment is transformed to organic Hg, mainly in methylated form (methyl mercury; M-Hg), resulting in anthropogenic Hg contamination, and there has been global concern about the high toxicity of M-Hg and its bioaccumulation via the food web (WHO, 1990). In humans, the Hg concentration in hair is the preferred marker for evaluating Hg exposure for a period of several weeks or months (JECFA, 2003). World Health Organization (WHO) concluded that a Hg level of 50 μg/g in human hair corresponds to 'no observed adversary effect level' (NOAEL) of M-Hg for adults, determined by neurotoxicological data (WHO, 1990).

Large epidemiological studies to determine the relationship between maternal exposure to M-Hg and impaired neurological development in children are currently underway in two locations: the Faroe Islands and the Seychelles. Faroe Islanders sometimes eat the meat of long-finned pilot whales (*Globicephala meleanus*) contaminated with high levels of M-Hg, while the Seychelles population, which does not eat whale meat, has a larger intake of fish contaminated with low levels of M-Hg. On the basis of preliminary reports of these studies, the Food and Agriculture Organization (FAO)/WHO Joint Expert Committee on Food Additives (JECFA) lowered its guideline value for provisional tolerable weekly intake (PTWI) of M-Hg from 3.3 μg/kg-bw/week to 1.6 μg/kg-bw/week (JECFA, 2003). By comparison, the US Environmental Protection Agency (US EPA) developed a reference dose (RfD) for M-Hg of 0.1 μg/kg-bw/day (US EPA, 1997). The revised PTWI and RfD correspond to a hair Hg level of 2.2 and 1.0 μg/g, respectively (Yasutake et al., 2004). It is worthy of note that in August 2008, the health authorities in the Faroe Islands revised their recommendations to indicate that long-finned pilot whales are no longer considered fit for human consumption, as the meat and blubber from pilot whales was considered to contain high levels of Hg, PCBs and DDT derivatives as to make it unsafe (Weihe and Joensen, 2008). The revised recommendations suggest that, in addition to damage to fetal neural development, exposure to low levels of Hg could be linked to high blood pressure and impaired immunity in children, as well as increased rates of Parkinson's disease, circulatory problems and possibly infertility in adults (Weihe and Joensen, 2008).

Taiji is a small town of 3400 people (as of 2008) in Wakayama Prefecture, Japan, which has suffered severely from emigration, with about one third of its residents now aged 65 or over (Fig. 1). Taiji is famous as the birthplace of traditional whaling in Japan, and continues the practice of commercial hunting for small cetaceans such as short-finned pilot whales (*Globicephala macrorhynchus*), Risso's dolphins (*Grampus griseus*) and striped dolphins (*Stenella coeruleoalba*). High levels of toxic substances such as heavy metals and organohalogen compounds have been found in the food products from toothed whales, dolphins and porpoises sold for human consumption in Japan, reflecting their position at the top of marine food web and their relative longevity (Haraguchi et al., 2000; Simmonds et al., 2002; Endo et al., 2002, 2003, 2004, 2005). The contamination level of Hg in the red meat from short-finned pilot whales sold in Japan (Endo et al., 2003, 2005) has been shown to be several times higher than that of long-finned pilot whales consumed in the Faroe Islands (Dam and Bloch, 2000). 'Whale meat' (including dolphins and porpoises) remains a

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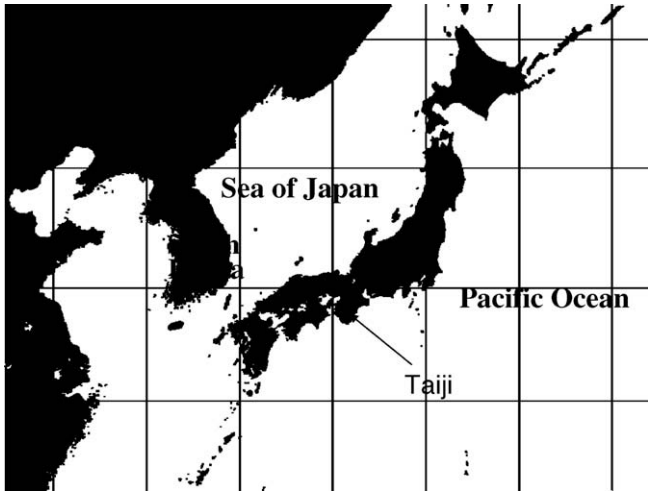

Fig. 1. Map of Japan showing Taiji.

traditional food source enjoyed by many Taiji residents, particularly those residents who making a living from whaling. Furthermore, Taiji residents frequently consume tuna as the town is adjacent to Nachikatsuura, a port famous throughout Japan for its tuna. Although the hazard to the health of Taiji residents from the consumption of whale products is suspicious, no epidemiologic survey has yet been conducted in Taiji.

Here we analyzed the T-Hg and M-Hg in the red meat from small cetaceans (pilot whales and dolphins), slices of fresh tuna (sashimi) and fillets of other fish species marketed in and around Taiji, together with the T-Hg concentrations in the head hair from 50 residents living in Taiji. We then compared the T-Hg concentrations between whale meat consumers and non-consumers, and discussed the possible health problems associated whale meat consumption.

2. Materials and methods

2.1. Sampling of cetacean and fish meats and head hair

The red meat of short-finned pilot whale (*G. macrorhynchus*), striped dolphin (*Stenella coerulecalba*) and Risso's dolphin (*G. griseus*) had purchased from the markets in and around Taiji between 2002 and 2005 (Fig. 1). The sashimi (slices of raw fish) of albacore tuna (*Thunnus alalunga*), yellowfin tuna (*Thunnus albacares*) and skipjack (*Katsuwonus pelamis*), and the fillets of dolphinfish (*Coryphaena hippurus*), ribbonfish (*Trichiurus japonicus*), common headfish (*Mola mola*), swordfish (*Xiphias gladius*) and striped marline (*Tetrapturus audax*) were also purchased from those markets between 2003 and 2008.

Head hair samples from 50 residents living in Taiji (30 men and 20 women) were collected by local collaborators from among their acquaintances between December 2007 and July 2008. At the time of collection, a simple questionnaire, detailing age, the species consumed, and frequencies of consumption of toothed whales and dolphins per month and tunas and other marine products per week, was completed, along with a declaration of informed consent. The average age of the 50 donors who cooperated in this survey was high (more than 50 years) as expected from the Taiji demographics. No whalers or members of their families, who were presumed to consume a comparatively large amount of pilot whale products, were included in the 50 donors. According to the collaborators' observations, none of the donors showed apparent symptoms of Hg poisoning such as tremble. The hair samples were packed in polyethylene bags and stored at room temperature until analysis.

2.2. Chemical analyses

The total mercury (T-Hg) concentrations in the samples were determined using a flameless atomic absorption spectrophotometer (Hiranuma Sangyo Co. Ltd., HG-1) after digestion by a mixture of HNO_3, HCO_4 and H_2SO_4 (Endo et al., 2002). Methyl mercury (M-Hg) concentrations in the samples were determined using a gas chromatograph (Shimazu Co. Ltd., GC-14A) with a ^{63}Ni electron capture detector (ECD) (Haraguchi et al., 2000). DOLT-2 (National Research Council of Canada) and CRB463 (BCR European Commission) were used as analytical quality control sample for the determination of T-Hg and M-Hg (Endo et al., 2003, 2004). Recoveries of T-Hg and M-Hg were 95% and 85%, respectively.

The concentrations of T-Hg and M-Hg in the red meat and filet samples shown in Table 1 and Fig. 2 were the mean of two or three determinations. The concentration of T-Hg in hair sample was expressed not only by arithmetic mean (AM) ± SD but also geometric mean (GM) with range to compare previous reports.

3. Results

Table 1 shows the contamination levels of T-Hg and M-Hg found in red meat from pilot whales and dolphins, slices of tuna and skipjack, and fillets of other fish species marketed in and around Taiji. As expected, high levels of Hg were found in the red meat from the pilot whales and dolphins. In particular, the contamination levels of T-Hg (9.6 ± 5.3 μg/wet g) and M-Hg (5.9 ± 2.9 μg/wet g) in the red meat from pilot whales were extremely high. Contamination levels of T-Hg and M-Hg in the red meat of striped dolphin were 4.0 ± 3.4 μg/wet g and 2.2 ± 0.8 μg/wet g, respectively, and those of Risso's dolphin were 4.4 ± 2.3 μg/wet g and 3.1 ± 1.7 μg/wet g, respectively. Average concentration of T-Hg in albacore was the same to the T-Hg concentration of the Japanese regulation (0.4 μg/wet g), and that of yellowfin tuna and skipjack were slightly lower than the regulation level. Contamination levels of T-Hg found in swordfish and striped marline were markedly higher than that of tuna, while contamination levels of T-Hg found in dolphinfish, ribbonfish and common head-fish were lower.

According to the results of the questionnaire (Table 2), 39 residents ate red meat from small cetaceans at least once every few months, whereas 11 residents did not eat 'whale meat' at all. Among the 39 residents that were red meat consumers, 28 residents usually ate red meat a few times each month, and 11 residents usually ate it once every few months. The most popular small cetacean species eaten was the short-finned pilot whale. All red meat consumers ate short-finned pilot whale, and some consumers occasionally consumed Risso's and striped dolphins. Many residents ate fresh slices of albacore and/or yellowfin tuna more than once each week, but no resident ate bluefin tuna, probably because of higher price of bluefin tuna. Most of residents ate other species of fish and shellfish at least a few times each week. The frequency of whale meat consumption was higher in males than in females, and all residents who ate whale meat also ate tuna.

The analytical results of T-Hg in hair from the 50 residents are summarized in Fig. 2. The T-Hg concentrations in three residents who ate whale meat more than once each month exceeded the NOAEL (50 μg/g). The highest T-Hg concentration was 67.2 μg/g in a male aged in his 50's. The lowest was 0.4 μg/g in two schoolchildren (aged less than 10 years) who did not eat whale meat or slices of tuna. The average T-Hg concentrations for residents who ate pilot whale and/or dolphin meat more than once each month or once every few months were 24.6 ± 15.6 μg/g (GM and range were 20.4 and 6.6–67.2 μg/g, $n = 28$) and 15.5 ± 10.0 μg/g (GM and range were 13.0 and 4.4–40.2 μg/g, $n = 11$), respectively,

Table 1
Mercury contamination levels in red meat of odontocetes, tunas and other fishes marketed in and around Taiji.

	n	Total mercury (μg/wet g)	Methyl mercury (μg/wet g)
Short-finned pilot whale	22	9.6 ± 5.3 (3.1–21.4)	5.9 ± 2.9 (2.1–12.0)
Striped dolphin	13	4.0 ± 3.4 (1.0–15.7)	2.2 ± 0.8 (1.0–4.1)
Risso's dolphin	19	4.4 ± 2.3 (1.7–9.2)	3.1 ± 1.7 (1.3–8.8)
Albacore	34	0.40 ± 0.12 (0.16–0.75)	N.D.
Yellowfine tuna	21	0.26 ± 0.24 (0.1–1.0)	N.D.
Skipjack	9	0.26 ± 0.17 (0.04–0.54)	N.D.
Dolphinfish	7	0.17 ± 0.09 (0.06–0.30)	N.D.
Ribbonfish	3	0.06 ± 0.03 (0.03–0.09)	N.D.
Common head-fish	6	0.05 ± 0.03 (0.03–0.10)	N.D.
Sword fish	5	1.6 ± 0.24 (0.1–1.0)	N.D.
Striped marline	5	0.78 ± 0.39 (0.32–1.35)	N.D.

The data are shown as mean ± SD with range.
N.D., not determined.

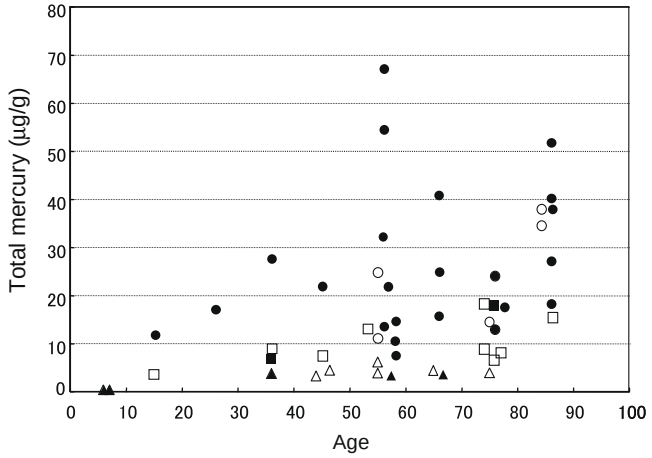

Fig. 2. Mercury concentrations in head hair in the whale meat consumers of Taiji residents. Closed and open symbols represent the mercury concentration in male and female of Taiji residents, respectively. Circle and square represent the whale meat consumers once or more time per month and less than once per month, respectively, and triangle represents the non-consumer.

Table 2
Number and frequency of consumption of red meat from small cetaceans.

Age	Male	Female
0–9	2 (C, C)	0
10–19	1 (A)	1(B)
20–29	1 (A)	0
30–39	3 (A, B,C)	1(B)
40–49	1 (A)	3 (B, C, C)
50–59	9 (A, A, A, A, A, A, A, A, C)	5 (A, A, B, C, C)
60–69	4 (A, A, A, C)	1 (C)
70–79	4 (A, A, A, B)	6 (A, B, B, B, B, C)
80+	5 (A, A, A, A, A)	3 (A, A, B)

A: once or more per month.
B: less than once per month.
C: no consumption.

whereas the average for residents who did not eat pilot whale or dolphin meat at all was 4.3 ± 1.7 μg/g (GM and range were 2.5 and 0.4–6.1 μg/g, $n = 11$). The T-Hg averages of 50 residents, 30 men and 20 women were 17.7 ± 11.5 μg/g (GM was 15.0 μg/g), 21.6 ± 16.7 μg/g (GM was 13.7 μg/g) and 11.9 ± 10.1 μg/g (GM was 9.0 μg/g), respectively. The hair T-Hg concentration tended to increase with age. Among the 50 donors, only one woman was of childbearing age (aged in her 30's, 8.8 μg/g). She ate whale meat once every few months and tuna a few times each week. The hair T-Hg concentrations below 10 μg/g and 20 μg/g were 40% and 70% of 50 donors, respectively.

As data not shown in figure, the hair T-Hg concentration who stopped the eating of pilot whale meat decreased markedly from 33.0 μg/g (December 2007) to 14.7 μg/g (June 2008). This donor was shown in Table 1 as male at 50's with the consumption of "whale meat" once or more time per month.

4. Discussion

4.1. Comparison of hair mercury level of Taiji residents with that of national average

Two research groups had recently conducted the large-scale survey of hair T-Hg concentration in Japan (Yasutake et al., 2004; Yasuda et al., 2005). According to Yasutake et al. (2004), the averages of hair T-Hg concentration (GM) were 2.42 μg/g in male ($n = 4274$) and 1.37 μg/g in female ($n = 4391$) and the total average of male and female was 1.82 μg/g. According to this survey, the percentages below 2.0, 5.0, and 10 μg/g were about 50%, 90%, and 99%, respectively, and the highest concentration was found in a male at 26.76 μg/g. Yasuda et al. (2005) analyzed 5846 hair samples and reported that the GM of hair T-Hg concentration in male adults tended to increase with age from 2.4 μg/g at high-teens up to a peak of 5.9 μg/g at 50's, and then decreased with further aging, and the hair T-Hg level in female was significantly lower than that in male.

In the present survey of hair T-Hg conducted in Taiji, the averages of male and female were 21.6 μg/g (GM was 13.7 μg/g, $n = 30$) and 11.9 μg/g (GM was 9.0 μg/g, $n = 20$), respectively, and the percentage of donors below 10 μg/g was only 40%. These GM values in male and female were about six times higher than those of national average, respectively, and the percentage below 10 μg/g in the Taiji residents (40%) was markedly lower than that in the national average (99%) (Yasutake et al., 2004). The highest concentration of hair T-Hg found in Taiji residents was 67.2 μg/g and the average of hair T-Hg in the male of 50's was 20.3 μg/wet g (GM was 14.0 μg/g, $n = 14$), while corresponding those values reported in the national surveys were 29.37 μg/g (Yasutake et al., 2004) and 5.9 μg/g (Yasuda et al., 2005), respectively. Although the sample size in the present survey was limited ($n = 50$) and the hair samples were not collected using a standardized procedure, the hair T-Hg level in Taiji residents appears to be markedly higher than that in other areas of Japan, and is correlated with frequency of whale meat consumption (Table 2). The hair T-Hg concentration in the Taiji resident who stopped the pilot whale meat consumption was markedly decreased within six months, suggesting that the whale meat is the major intake route for Hg. The half-life of Hg in blood

was reported to be about 70 days (Miettinen et al., 1971). The Taiji residents who make their living from whaling are expected to consume much more whale meat and to have higher hair T-Hg levels, but none of those residents were included among the 50 donors.

4.2. Risk assessment of mercury exposure due to the whale consumption in Taiji residents

Contamination levels of M-Hg in the red meats of short-finned pilot whale, striped and Risso's dolphins were markedly higher than the US FDA action level and Cordex Alimentarius guideline level of M-Hg in predatory fish (1.0 μg/wet g). Based on the present data of M-Hg concentration in the red meat of short-finned pilot whale (5.9 μg/wet g), consumption of only 17 g and 8 g of the red meat per one week exceeds the PTWI and the RfD of M-Hg per 60 kg, respectively. The whale meat consumers in Taiji also appear to preferentially consume tuna and other fish species, and most of whale meat consumers in Taiji appear to intake M-Hg exceed the PTWI and the RfD. The intake of M-Hg at PTWI (1.6 μg/kg/week) set by JECFA (2003) corresponds to a hair T-Hg concentration of 2.2 μg/g (Yasutake et al., 2004). Only two donors in the present survey (school children) were below this T-Hg level. The highest concentration of 67.2 μg/g found in the present survey corresponds to about 31 times of the PTWI. The hair T-Hg concentration of 20.4 μg/g (the average of the pilot whale meat consumers more than once in every month) corresponds to the consumption of 150 g of the pilot whale meat (5.9 μg/wet g) per one week, exceeding 9.3 times the PTWI. According to the questionnaire, however, no donor ate the whale and dolphin meats more than once per week. Therefore, the consumption of tunas and other fish species may contribute to the high concentration of hair T-Hg in the Taiji residents known to consume whale meat. The Hg levels of tunas and other fish species shown in Table 1 are comparable to those in previous reports (Yamashita et al., 2005; Kojadinovic et al., 2006; Kaneko and Ralston, 2007).

4.3. Comparison of mercury levels of hair and whale meat in the Faroe Islands

As mentioned in Introduction, the health authorities in the Faroe Islands revised their recommendations to indicate that long-finned pilot whales are no longer considered fit for human consumption, as the meat and blubber from pilot whales was considered to contain high levels of Hg, PCBs and DDT derivatives (Weihe and Joensen, 2008). Choi et al. (2009) suggested that the increased M-Hg intake from pilot whale meat promoted the development of cardiovascular disease of Faroese whaling men. They reported that the current level of T-Hg (GM) in the hair of Faroese whaling men was 7.31 μg/g (0.92–46.0 μg/g, $n = 42$) and more than half of the men ate the whale meat three or more time each month (Choi et al., 2009). The hair T-Hg level of the Taiji residents (GM was 15.0 μg/g, $n = 50$) was higher than that of the Faroese whaling men. The Taiji residents may be eating the whale red meat more polluted by Hg, because levels of T-Hg and M-Hg found in the short-finned pilot whales sold in and around Taiji (Table 1) were a few times higher than those in the long-finned pilot whales caught off Faroe Islands (Julshamn et al., 1987; Dam and Bloch, 2000). In addition, Taiji residents tend to preferentially eat albacore and yellowfin tunas and other fish species. Furthermore, contamination levels of PCBs and other organohalogen compounds fund in whale products sold in and around Taiji (Haraguchi et al., 2000; Simmonds et al., 2002) were compatible levels to those reported in the Faroe Islands (Dam and Bloch, 2000). The large-scale survey of hair T-Hg concentration in Taiji residents is necessary to prevent the health problems associated with the consumption of whale products.

4.4. Comparison of mercury levels in hair due to the fish consumption

Apart from Japan, hair Hg levels associated with the consumption of marine food have been surveyed in China and Indonesia (Feng et al., 1998), US (McDowell et al., 2004), Cambodia (Agusa et al., 2005), Morocco (Elhamri et al., 2007) and Malaysia (Hajeb et al., 2008). According to those studies, the hair Hg levels in populations (GM) with little or no fish consumption were below 0.5 μg/g (McDowell et al., 2004), and the averages (AM or GM) among fish consumers increased by up to about 15 μg/g in proportion to the level of fish consumption (Hajeb et al., 2008). To our knowledge, no hair Hg exceeding 50 μg/g (NOAEL) has yet been reported in fish consumers. In contrast, the T-Hg concentrations in the hair from three residents who ate the whale meat more than once each month exceeded 50 μg/g (Fig. 2). Choi et al. (2009) reported the hair T-Hg concentration of 54.1 μg/g in a Faroese whaling men. Extremely high level of hair Hg (705 μg/g) was reported in a patient of Minamata disease who ate fish contaminated with anthropogenic origin of Hg (Harada, 1995).

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