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Contamination by mercury and cadmium in the cetacean products from Japanese market

T. Endo [a,*], K. Haraguchi [b], F. Cipriano [c], M.P. Simmonds [d], Y. Hotta [a], M. Sakata [a]

[a] Faculty of Pharmaceutical Sciences, Health Sciences University of Hokkaido, 1757, Ishikari-Tobetsu, Hokkaido 061-0293, Japan

[b] Daiichi College of Pharmaceutical Sciences, 22-1 Tamagawa-Cho, Minami-Ku, Fukuoka 815-8511, Japan

[c] Conservation Genetics Laboratory, San Francisco State University, San Francisco, CA 94123, USA

[d] Natural Resources Institute, University of Greenwich, Chatham, Maritime, Kent, ME4 4TB, UK

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Abstract

Cetaceans hunted coastally in Japan include several species of odontocete (dolphins, porpoises and beaked whales), and fresh and frozen red meat and blubber, as well as boiled internal organs, such as liver, lung, kidney and small intestine, are still sold for human consumption. Furthermore, red meat and blubber products originating from mysticete minke whales caught in the Antarctic and Northern Pacific are also sold for human consumption. We surveyed mercury and cadmium contamination levels in boiled liver, lung, kidney and red meat products being marketed in Japanese retail outlets. We also analyzed the DNA of these products to obtain information concerning gender and species. Total mercury (T-Hg) and methyl mercury (M-Hg) contamination levels in all the cetacean products were markedly higher in odontocete species than in mysticete species, and slightly higher in females than in males. T-Hg contamination in the organs was seen in the following order: boiled liver > boiled kidney = boiled lung > red meat. In particular, T-Hg concentrations in the boiled liver were high enough to cause acute intoxication even from a single ingestion: the mean ±SD (range) of T-Hg was 388 ± 543 (0.12–1980) μg/wet g. In contrast, although M-Hg contamination in the liver was not markedly higher than that in other organs, M-Hg contamination was in the following order: boiled liver > odontocete red meat > boiled kidney > boiled lung. The contamination levels of T-Hg and M-Hg in odontocete red meat, the most popular whale product, were 8.94 ± 13.3 and 5.44 ± 5.72 μg/wet g, respectively. These averages exceeded the provisional permitted levels of T-Hg (0.4 μg/wet g) and M-Hg (0.3 μg/wet g) in marine foods set by the Japanese Ministry of Health, Labor and Welfare by 22 and 18 times, respectively, suggesting the possibility of chronic intoxication by T-Hg and M-Hg with frequent consumption of odontocete red meat. Cadmium contamination levels in boiled liver, kidney and lung were 8.59 ± 12.0, 10.4 ± 8.6 and 1.66 ± 1.27 (μg/wet g), respectively.

Keywords: Mercury; Cadmium; Liver; Kidney; Dolphin; Whale

1. Introduction

Cetaceans hunted coastally in Japan include several species of odontocete (toothed whales) such as Risso's dolphin (*Grampus griseus*), striped dolphin (*Stenella coeruleoalba*), bottlenose dolphin (*Tursiops truncatus*),

* Corresponding author. Tel./fax: +81-1332-3-3902.
E-mail address: endotty@hoku-iryo-u.ac.jp (T. Endo).

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pilot whale (*Globicephala macrorhynchus*), and Baird's beaked whale (*Berardius bairdii*). They are located at the top of marine food chain and feed mainly on fish and squid, making them a good indicator of marine pollution. As they live in different areas and eat different foods, their level of chemical contamination naturally varies considerably. Every year a total of about 20 000 dolphins, porpoises and beaked whales are caught off the coast of Japan. Cetaceans are consumed locally in coastal whaling areas and are also marketed across Japan. On the other hand, over 500 minke whales are caught annually in the Antarctic (*Balaenoptera bonaerensis*) and Northern Pacific (*Balaenoptera acuturostrate*) under the "research whaling program" and their products are also sold across Japan.

Heavy metals and organochlorines are well known environmental pollutants that accumulate in the bodies of odontocete (Haraguchi et al., 2000; Simmonds et al., 2002). Among these pollutants, contamination with mercury (Hg) is prominent. High levels of Hg are known to accumulate in the internal organs, especially in the liver (Honda et al., 1983; Itano et al., 1984; André et al., 1991; Leonzio et al., 1992; Holsbeek et al., 1998; Meador et al., 1999; Zhou et al., 2001). To our knowledge, the maximal concentration of total mercury (T-Hg) so far reported in whale liver was 1500 μg/wet g in a striped dolphin (André et al., 1991) and 13 156 μg/dry g in a bottlenose dolphin (Leonzio et al., 1992). Marine mammals are principally exposed to methyl mercury (M-Hg), because almost all of the Hg present in fish and squid is methylated (Caurant et al., 1996; Das et al., 2000). Nevertheless, the major part of Hg accumulated in marine mammal internal organs is inorganic mercury (I-Hg). The demethylation of M-Hg, followed by the formation of a less toxic Hg-Se complex, is thought to occur mainly in cetacean livers (Caurant et al., 1996; Holsbeek et al., 1998; Wagemann et al., 1998; Meador et al., 1999). High levels of cadmium (Cd) are also known to accumulate in the internal organs of odontocete (Honda et al., 1983; Leonzio et al., 1992; Holsbeek et al., 1998) and bind to metallothioneins (Das et al., 2000).

Red meat (muscle) from cetaceans is the most popular of the whale products marketed in Japan. However, some people living in Wakayama Prefecture eat not only the red meat and blubber but also the internal organs such as the liver, kidney, lung, stomach and small intestine of cetaceans. In spite of very high levels of Hg contamination, a mixture of boiled internal organs is still sold for human consumption at supermarkets, fishmongers, cetacean product shops and souvenir shops. The Hg contamination level in odontocetes caught off the coast of Japan has been reported to vary according to species, location (Honda et al., 1990) and age (Honda et al., 1983). Furthermore, levels of Hg contamination may vary between males and females (André et al., 1990; Beck et al., 1997; Meador et al., 1999; Zhou et al., 2001). However, no detailed survey of Hg contamination levels in the whale products in Japan has yet been undertaken, and little attention has been paid to the potential human health problems associated with this source of Hg contaminants.

The aim of the present study was to survey the levels of T-Hg, M-Hg and Cd contamination in the boiled internal organs and red meat from cetaceans marketed for human consumption in Japan. We also contrast the contamination levels of T-Hg and M-Hg among organ types, among species, and between males and females, and discuss the potential for human health problems related to the consumption of whale products.

2. Materials and methods

Sampling: Samples of a mixture of boiled internal organs marketed for human consumption were purchased between 1999 and 2001 from retail outlets in Taiji, Katsuura and Kushimoto in Wakayama Prefecture, Japan. The mixtures had been vacuum packed and sold as frozen food. Most of these products were simply labeled as "whale" or "pilot whale". The boiled liver, kidney and lung were submitted for the determination of total mercury (T-Hg), methyl mercury (M-Hg) and cadmium (Cd), and for DNA analysis to determine their gender and species. Samples of uncooked red meats (fresh and frozen) were purchased between 1999 and 2001 from retail outlets across Japan, and the concentrations of T-Hg and M-Hg and the gender and species of the whales from which the samples were taken were analyzed. To minimize duplicate sampling of the same animal, we purchased only one package from each vendor or a few packages labeled at different dates of manufacture, and all samples were stored at −20 °C until analysis (Simmonds et al., 2002).

Chemical analyses: T-Hg in the internal organs and red meat was analyzed using a flameless atomic absorption spectrophotometer (Hiranuma Sungyo Co. Ltd., HG-1) after digestion by a mixture of HNO_3, $HClO_4$ and H_2SO_4 (Endo et al., 2002). Cd was determined using a Polarized Zeeman flame atomic absorption spectrophotometer (Hitachi Co. Ltd., Z-8100) after digestion by a HNO_3–$HClO_4$ mixture (Haraguchi et al., 2000). M-Hg was determined using a gas chromatograph (Shimazu Co. Ltd., GC-14A) with a ^{63}Ni electron capture detector (ECD) (Haraguchi et al., 2000). M-Hg concentration was expressed as Hg concentration basis. Metal concentrations in the whale products were expressed as wet weight. DOLT-2 (National Research Council of Canada) and CRB 463 (BCR, European Commission) were used as analytical quality control samples for the determination of T-Hg, M-Hg and Cd. The recoveries of these metals were 85–96%. DNA analysis of whale products was undertaken to elucidate their species origins according to the method of Cipriano and Palumbi (1999).

3. Results

T-Hg, M-Hg and Cd concentrations in the boiled livers, lungs and kidneys purchased from Wakayama Prefecture, together with their species and sex as determined from DNA analyses are shown in Tables 1–3, respectively. Further, T-Hg and M-Hg concentrations and the DNA analyses of the red meat products purchased across Japan are shown in Tables 4 and 5. In spite of much effort, we could not determine the species of all the odontocete products. In particular, genetic discrimination among striped dolphin (*Stenella coeruleoalba*), bottlenose dolphin (*Tursiops truncatus*) and common dolphin (*Delphinus delphis*) is difficult (Dizon et al., 2000). Therefore, these samples are noted as "S/T/D dolphin". Only a few samples could be identified specifically as bottlenose dolphin (indicated as T. dolphin in the tables). From DNA analyses, the boiled internal organs purchased from Wakayama Prefecture were shown to originate from odontocetes (pilot whale, Risso's dolphin and S/T/D dolphin), except for one sample originating from a mysticete (minke whale). Unfortunately, the randomly chosen boiled kidney samples did not include any kidney originating from a pilot whale. The red meat products purchased across Japan originated from not only the odontocete species mentioned above together with Baird's beaked whale, but also from the mysticete minke and Bryde's whales (*Balaenoptera edeni*). Figs. 1 and 2 show species differences in T-Hg, M-Hg and Cd concentrations in the boiled organs and red meats, respectively.

T-Hg contamination levels in the boiled liver samples were extremely high. The mean concentration of T-Hg was 388 μg/wet g with a large standard deviation of ±543 (Table 1). The three highest individual T-Hg concentrations (914, 1970 and 1980 μg/wet g) were found in female S/T/D dolphins. Compared with T-Hg, the mean ± SD concentration of M-Hg was markedly lower (11.6 ± 6.2 μg/wet g). Of 21 liver samples genetically analyzed, one only originated from a mysticete (minke whale), and this sample contained the lowest concentrations of T-Hg and M-Hg.

Table 1
Total and methyl mercury and cadmium concentrations in the boiled whale livers

Species	Gender	T-Hg (μg/wet g)	M-Hg (μg/wet g)	Cadmium (μg/wet g)
Pilot whale (JH00-34)	M	422	9.83	35.9
Pilot whale (JH00-35)	M	390 (406)	14.8 (12.3)	48.6 (42.3)
Risso's dolphin (JH00-9)	M	286	15.3	3.63
Risso's dolphin (JH00-15)	M	30.2	12.4	1.1
Risso's dolphin (JH00-25)	M	504	12.6	26.1
Risso's dolphin (JH00-48)	M	41.2	7.95	3.21
Risso's dolphin (JH00-59)	M	47.4	7.43	11.2
Risso's dolphin (JH00-12)	F	645	14.0	3.15
Risso's dolphin (JH00-56)	F	72.6 (232)	8.73 (11.2)	8.45 (8.12)
S/T/D dolphin (JH00-5)	M	352	8.02	0.76
S/T/D dolphin (JH00-52)	M	55.9	8.44	6.21
S/T/D dolphin (JH00-68)	M	393	8.69	5.97
S/T/D dolphin (JH00-76)	M	134	8.82	7.94
S/T/D dolphin (JH00-29)	F	1980	23.5	8.07
S/T/D dolphin (JH00-30)	F	914	22.7	2.02
S/T/D dolphin (JH00-47)	F	22.7	8.41	4.22
S/T/D dolphin (JH00-62)	F	174	9.72	3.82
S/T/D dolphin (JH00-65)	F	7.60	3.01	1.36
S/T/D dolphin (JH00-71)	F	1970	25.7	4.21
T. dolphin (JH00-17)	F	36.8 (549)	9.19 (12.4)	0.30 (4.08)
Minke whale (JH00-21)	M	0.12	0.12	1.00
n.d. (JH00-41)	M	39.0	7.84	5.91
n.d. (JH00-79)	M	368	20.3	4.38
n.d. (JH00-1)	n.d.	413	11.3	n.d.
Mean ± SD		388 ± 543, $n = 24$	11.6 ± 6.2, $n = 24$	8.59 ± 12.0, $n = 23$

Parentheses indicate the discrimination number of sample and the mean of species.
n.d.: Not determined.

Table 2
Total and methyl mercury and cadmium concentrations in the boiled whale lungs

Species	Gender	T-Hg (μg/wet g)	M-Hg(μg/wet g)	Cd (μg/wet g)
Pilot whale (JH00-44)	M	19.1	1.89	1.21
Pilot whale (JH00-37)	M	12.5	2.08	5.12
Pilot whale (JH00-55)	F	52.9	0.71	3.45
Pilot whale (JH00-54)	F	63.3 (37.0)	0.50 (1.30)	2.40 (3.05)
Risso's dolphin (JH00-60)	M	1.54	0.20	2.88
Risso's dolphin (JH00-50)	M	1.00	0.68	1.72
Risso's dolphin (JH00-16)	M	2.38	2.43	1.88
Risso's dolphin (JH00-13)	M	145	0.78	4.42
Risso's dolphin (JH00-2)	F	20.5	1.66	0.83
Risso's dolphin (JH00-26)	F	76.6 (41.2)	0.65 (1.07)	2.11 (2.31)
S/T/D dolphin (JH00-78)	M	15.2	0.61	1.63
S/T/D dolphin (JH00-70)	M	79.6	0.23	1.76
S/T/D dolphin (JH00-67)	M	39.4	0.46	1.31
S/T/D dolphin (JH00-64)	M	14.3	0.73	1.35
S/T/D dolphin (JH00-6)	M	11.5	3.33	0.57
S/T/D dolphin (JH00-49)	M	4.70	2.41	1.41
T. dolphin (JH00-38)	M	12.2	1.60	0.66
S/T/D dolphin (JH00-75)	F	21.2	1.60	0.94
S/T/D dolphin (JH00-74)	F	70.9	1.70	1.10
S/T/D dolphin (JH00-39)	F	2.60	1.80	0.14
S/T/D dolphin (JH00-32)	F	63.0	5.53	1.00
T. dolphin (JH00-81)	F	164	5.72	0.40
T. dolphin (JH00-18)	F	23.6 (40.2)	9.93 (2.74)	0.92 (1.01)
Minke whale (JH00-22)	M	0.16	0.12	0.10
n.d. (JH00-45)	M	21.9	1.40	2.08
n.d.(JH00-43)	M	2.10	0.87	3.76
n.d. (JH00-83)	F	62.1	1.80	0.25
n.d.(JH00-84)	n.d.	63.0	1.47	1.00
Mean ± SD		38.1 ± 42.1, $n = 28$	1.88 ± 2.09, $n = 28$	1.66 ± 1.27, $n = 28$

Parentheses indicate the discrimination number of sample and the mean of species.
n.d.: Not determined.

T-Hg concentrations in the boiled lungs and kidneys from odontocetes were one order of magnitude smaller than those in the livers (Fig. 1), and there was less variance in M-Hg concentrations among these organs. Like the boiled liver, T-Hg concentrations in the boiled lung and kidney samples tended to be higher in females, and T-Hg and M-Hg concentrations in kidney and lung samples were the lowest in those from minke whales (Tables 2 and 3).

The T-Hg contamination level in the red meat was markedly higher in odontocetes (Table 4) than in mysticetes (Table 5): the T-Hg concentrations in some mysticetes were close to the detection level (0.03 μg/wet g). The three highest individual T-Hg concentrations (13.8, 22.5 and 63.4 μg/wet g) were found in the red meat of female odontocetes, but no gender differences in T-Hg or M-Hg were observed in the red meat originating from mysticetes. The T-Hg contamination level in red meat originating from Baird's beaked whale was the lowest among the odontocete species analyzed. Similar species and gender differences were observed in M-Hg concentrations.

The hepatic Cd concentrations were higher in the two pilot whales analyzed (35.9 and 48.6 μg/wet g) than in the Risso's dolphins (8.12 ± 8.67 μg/wet g, n = 7) or S/T/D dolphins (4.08±2.76 μg/wet g, $n = 11$), and the renal Cd concentration was higher in the Risso's dolphins (17.5 ± 10.9 μg/wet g, $n = 4$) than in the S/T/D dolphins (6.46 ± 3.84 μg/wet g, $n = 10$) (Fig. 1). The Cd contamination level was markedly lower in the boiled lungs than in the boiled livers and kidneys. The Cd concentrations in the red meat were below detection level (less than 0.1 μg/wet g).

In Figs. 1 and 2, no consistent pattern of species differences in T-Hg and M-Hg concentrations among pilot whales, Risso's dolphins and S/T/D dolphins was observed. The variances in T-Hg and M-Hg levels in S/T/D dolphins were considerably higher than those for other individual species analyzed, but the variance in Cd levels was similar.

Table 3
Total and methyl mercury and cadmium concentrations in the boiled whale kidneys

Species	Gender	T-Hg (μg/wet g)	M-Hg (μg/wet g)	Cd (μg/wet g)
Risso's dolphin (JH00-14)	M	28.8	3.87	4.42
Risso's dolphin (JH00-27)	M	24.1	1.59	24.6
Risso's dolphin (JH00-61)	M	8.67	1.70	12.7
Risso's dolphin (JH00-42)	F	7.30 (17.2)	2.76 (2.48)	28.1 (17.5)
S/T/D dolphin (JH00-7)	M	23.9	1.03	13.3
S/T/D dolphin (JH00-63)	M	20.8	1.61	10.8
S/T/D dolphin (JH00-69)	M	27.5	0.67	10.6
S/T/D dolphin (JH00-77)	M	7.85	0.62	7.21
S/T/D dolphin (JH00-80)	M	29.2	21.0	3.45
T. dolphin (JH00-3)	M	24.0	3.79	3.90
S/T/D dolphin (JH00-31)	F	142	8.15	3.04
S/T/D dolphin (JH00-66)	F	2.28	0.75	5.95
S/T/D dolphin (JH00-73)	F	153	6.20	3.30
T. dolphin (JH00-19)	F	95.1 (52.6)	7.07 (5.09)	3.04 (6.46)
Minke whale (JH00-23)	M	0.01	0.01	0.01
n.d. (JH00-10)	M	13.7	1.54	21.1
Mean ± SD		36.6 ± 46.8, $n = 16$	3.76 ± 5.05, $n = 16$	10.4 ± 8.6, $n = 16$

Parentheses indicate the discrimination number of sample and the mean of species.
n.d.: Not determined.

Table 4
Total and methyl mercury concentrations in the red meat originating from odontocetes

Species	Gender	T-Hg (μg/wet g)	M-Hg (μg/wet g)
Pilot whale (JH00-88)	M	5.38	4.31
Pilot whale (JH00-91)	M	12.3	12.1
Pilot whale (JH00-104)	F	13.8	7.72
Pilot whale (JH00-4)	F	7.49 (9.74)	4.48 (7.15)
Risso's dolphin (JH00-92)	M	9.21	8.78
Risso's dolphin (JH00-95)	M	9.09 (9.15)	4.34 (6.56)
S/T/D dolphins (JH00-86)	M	1.43	1.40
S/T/D dolphin (JH00-87)	M	3.47	3.35
S/T/D dolphin (JH00118)	M	8.96	8.72
S/T/D dolphin (J06)	M	2.19	1.72
S/T/D dolphin (J12)	M	2.28	2.25
S/T/D dolphin (J13)	M	2.24	2.16
T. dolphin (J00-127)	M	2.36	1.24
S/T/D dolphin (J00-126)	F	2.74	1.51
S/T/D dolphin (J00-128)	F	63.4	26.2
S/T/D dolphin (J7-1)	F	2.17	2.02
S/T/D dolphin (J00-108)	n.d.	14.2	7.29
T. dolphin (J15)	F	22.5 (10.7)	10.9 (5.73)
Baird's beaked whale (J00-105)	M	2.97	1.81
Baird's beaked whale (J00-85)	F	1.48	0.95
Baird's beaked whale (A2)	n.d.	5.30	4.76
Baird's beaked whale (A20)	n.d.	1.71 (2.87)	1.58 (2.28)
Mean ± SD		8.94 ± 13.3, $n = 22$	5.44 ± 5.72, $n = 22$

Parentheses indicate the discrimination number of sample and the mean of species.
n.d.: Not determined.

T-Hg versus the percentage of M-Hg/T-Hg in the boiled livers, lungs, kidneys and red meat originating from both odontocetes and mysticetes was plotted (Fig. 3), according to previous studies (Caurant et al., 1996;

Table 5
Total and methyl mercury concentrations in the red meat originating from mysticetes

Species	Gender	T-Hg (μg/wet g)	M-Hg (μg/wet g)
Mink whale (JH00-107)	M	0.09	0.06
Mink whale (JH00-113)	M	0.05	0.04
Mink whale (JH00-119)	M	0.10	0.04
Mink whale(JH00-123)	M	0.07	0.03
Mink whale (J1)	M	0.07	0.02
Mink whale (J6)	M	0.11	0.02
Mink whale (J10-1)	M	0.04	0.01
Mink whale (J00-102)	F	0.17	0.07
Mink whale (J00-103)	F	0.11	0.06
Mink whale (J00-112)	F	0.03	0.04
Mink whale (J00-116)	F	0.09	0.03
Mink whale (J00-117)	F	0.07	0.01
Mink whale (J17)	F	0.04	0.01
Mink whale (JH00-121)	n.d	0.12 (0.08)	0.05 (0.04)
Bryde's whale (J8)	F	0.09	0.02
Mean ± SD		0.08 ± 0.04, $n = 15$	0.03 ± 0.02, $n = 15$

Parentheses indicate the discrimination number of sample and the mean of species.
n.d.: Not determined.

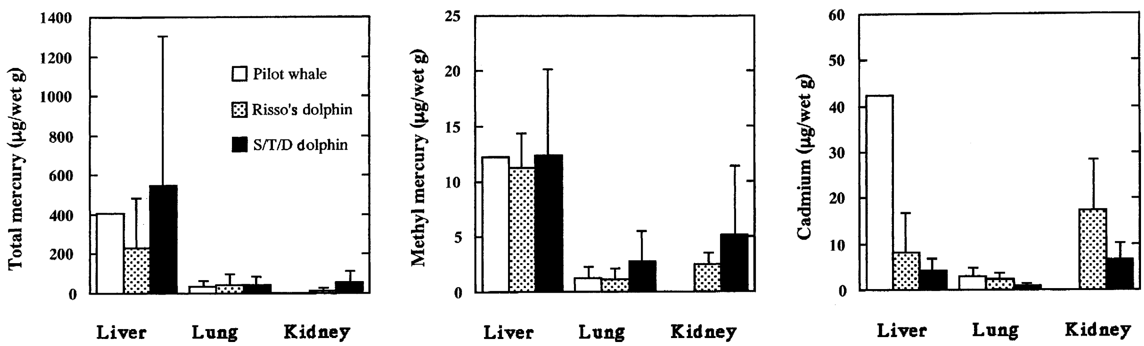

Fig. 1. Differences in the distribution of total mercury, methyl mercury and cadmium concentrations among whale species. Each bar represents the mean with SD. Numbers of boiled liver, lung and kidney samples from pilot whales were 2, 4, and 0, respectively, those from Risso's dolphins were 7, 6, and 4, respectively, and those from S/T/D dolphins were 11, 13, and 10, respectively.

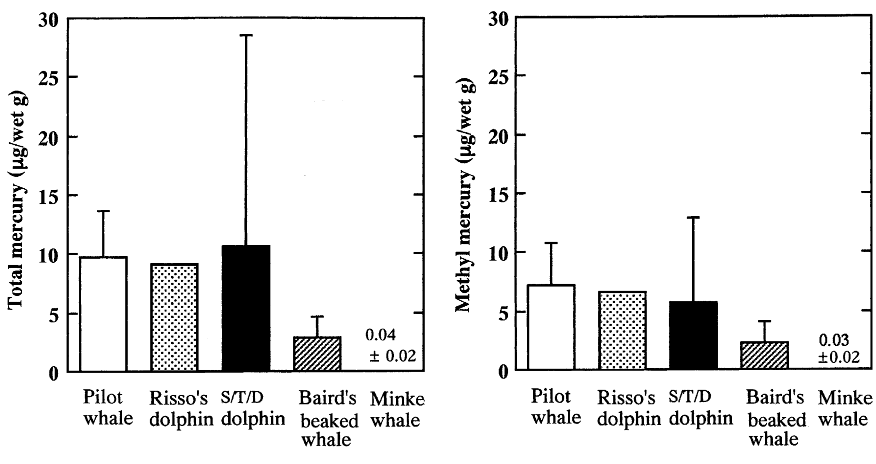

Fig. 2. Species differences in total mercury and methyl mercury concentrations in red meat. Each bar represents the mean with SD for 4 pilot whales, 2 Risso's dolphins, 12 S/T/D dolphins, 4 Baird's beaked whales, or 14 minke whales.

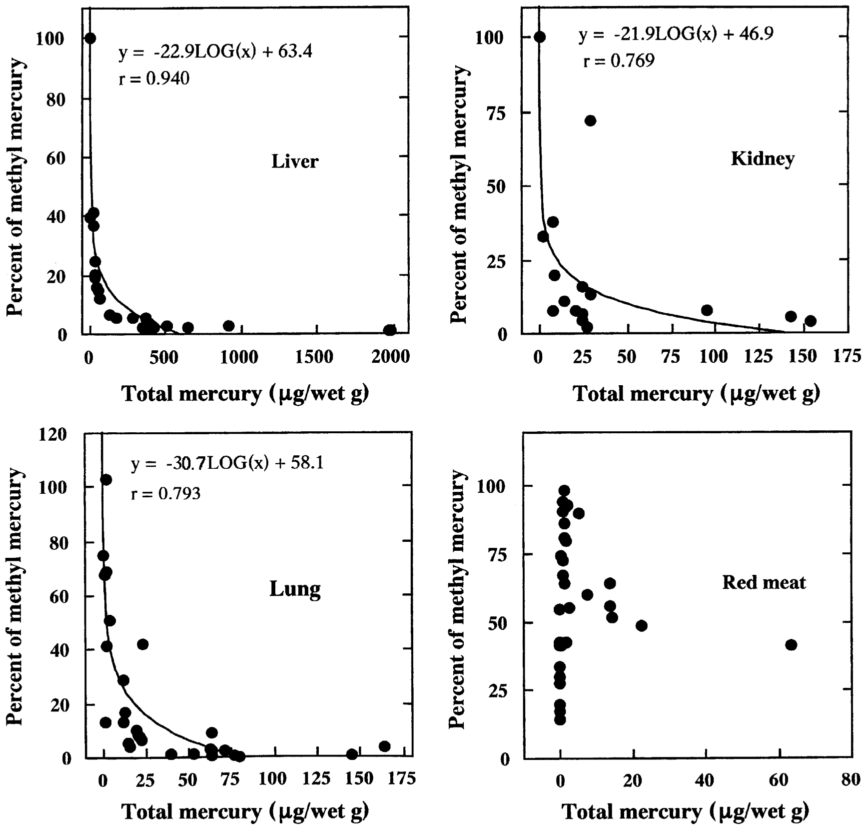

Fig. 3. Total mercury versus the percentage of methyl mercury to total mercury in different tissue types. Methyl mercury is plotted as a percentage of total mercury in the boiled livers, lungs, kidneys and red meat. Regression equations were generated using a personal computer and CA-Cricket Graph III.

Holsbeek et al., 1998). The plot of data obtained from the internal organs was well fitted by a logarithmic equation, and the regression lines were found to be similar. In contrast, the plot for the red meat was not fitted by any regression function. The mean ± SD of M-Hg/T-Hg was 62.7 ± 21.1% ($n = 37$).

4. Discussion

The T-Hg contamination levels in the boiled liver products were markedly higher than in the boiled kidney, lung, or red meat, but no predominant accumulation of M-Hg was observed in the liver. Similar distributions of T-Hg and M-Hg in uncooked tissues of cetaceans have been reported by many researchers (Honda et al., 1983; Itano et al., 1984; André et al., 1991; Leonzio et al., 1992; Holsbeek et al., 1998; Meador et al., 1999; Zhou et al., 2001). In agreement with our survey of T-Hg, Honda et al. (1983) reported that the hepatic concentration of T-Hg in striped dolphins caught off Taiji, Wakayama Prefecture, was 205 ± 139 μg/wet g (1.7–485 μg/wet g, $n = 59$). As far as we know, the maximal concentrations of T-Hg and M-Hg reported in the cetacean livers were 1500 μg/wet g (André et al., 1991) and 30.4 μg/wet g (Storelli et al., 1998) in striped dolphins, respectively. Although the T-Hg concentrations in the liver were expressed on a dry weight basis, Leonzio et al. (1992) reported T-Hg concentrations of 13 156 μg/dry g in a bottlenose dolphin and 4400 μg/dry g in a striped dolphin.

The T-Hg contamination level in the red meat originating from pilot whales, Risso's dolphins and S/T/D dolphins was markedly lower than that in the boiled lung and kidney, but the M-Hg contamination level in the former was higher than that in the latter. In our survey (Table 4), the maximal concentration of T-Hg was 63.4 μg/wet g in the red meat from either a striped, bottlenose or common (S/T/D) dolphin. Arima and Nagakura (1979) reported 51.8 μg T-Hg/wet g in the muscle of a bottlenose dolphin caught off Japan, and

André et al. (1991) reported 81.2 μg T-Hg/wet g in the muscle of a striped dolphin caught in the Mediterranean.

Many researchers have postulated the demethylation of M-Hg, followed by the formation of Hg–Se complex in the internal organs, especially in liver (Caurant et al., 1996; Holsbeek et al., 1998; Wagemann et al., 1998; Meador et al., 1999; Endo et al., 2002). In agreement with Caurant et al. (1996) and Holsbeek et al. (1998), T-Hg versus M-Hg/T-Hg in the boiled liver, lung and kidney samples decreased with increases in T-Hg (Fig. 3), suggesting the demethylation of M-Hg in cetacean organs. Interestingly, all regression lines for the internal organs were similar. In contrast, the data for T-Hg and M-Hg in the red meat samples could not be fitted to a logarithmic equation. Although detailed survey is under investigation, low concentrations of T-Hg and M-Hg in the red meats or low activity of demethylation of M-Hg in muscle may be related to this difference between the internal organs and the red meat (Fig. 3). Meador et al. (1999) reported the exponential fitting when the percentage of M-Hg/T-Hg of a combined fresh sample of liver, kidney and brain of bottlenose dolphins was plotted as the ordinate and log T-Hg as the abscissa ($Y = 99.6e^{-1.45X}$, $\gamma^2 = 0.92$). Our data, especially for boiled liver samples (Table 1), well fitted their fitting, resulting in a similar equation ($Y = 107e^{-1.29X}$, $\gamma^2 = 0.86$). The similarity may indicate that boiling has no marked effect on T-Hg and M-Hg concentrations in cetacean internal organs.

The Cd contamination levels in the boiled liver samples were similar to those in the boiled kidney samples (Tables 1 and 3). Conversely, many researchers have shown higher contamination levels of Cd in the kidney than in the liver (Honda et al., 1983; Leonzio et al., 1992; Dietz et al., 1996; Holsbeek et al., 1998). This discrepancy may be related to the small number of boiled kidney samples in our study (Table 3). As stated below, the Cd contamination level appears to be higher in pilot whales than in the other species (Fig. 1), but unfortunately no boiled kidney samples from pilot whales were used in this study. As far as we know, the highest reported concentrations of Cd in the kidney and liver of cetaceans have been 134 and 125 μg/wet g in pilot whales, respectively (Julshamn et al., 1987). The levels of Cd contamination in odontocete caught off the coast of Japan are not so remarkable.

Levels of T-Hg and M-Hg contamination in all of the whale products tended to be higher in females than in males. Higher contamination levels of T-Hg and M-Hg in female odontocetes have been reported by many researchers (André et al., 1990; Beck et al., 1997; Meador et al., 1999; Zhou et al., 2001), but no particular gender difference was reported by Honda et al. (1983). Differences in feeding rate and metabolism (Zhou et al., 2001) or in lipid cycling related to gestation and lactation (Meador et al., 1999) may cause the observed gender differences in T-Hg and M-Hg distributions. Gender differences in M-Hg after oral administration have been shown in experiments with mice, where the M-Hg concentrations in the organs except for kidneys were higher and the excretion of M-Hg was slower in females than in males (Nielsen and Andersen, 1991). Beside gender and species differences, T-Hg and Cd contamination increases with age (Honda et al., 1983; Itano et al., 1984; Leonzio et al., 1992; Dietz et al., 1996; Beck et al., 1997; Holsbeek et al., 1998; Meador et al., 1999), and varies according to habitat (André et al., 1990; Meador et al., 1999) and stage of lactation (Caurant et al., 1996).

The species differences in T-Hg and M-Hg concentrations in the internal organs were unclear (Fig. 3), probably because of the small number of samples and unresolved species identification of striped, bottlenose and common (S/T/D) dolphins. The data for S/T/D dolphins showed high variances (Figs. 1 and 2), as would be expected if there are species- specific differences. As stated above, the maximal values of T-Hg and M-Hg concentrations have been reported in striped or bottlenose dolphins, but not in common dolphins, and T-Hg concentrations in liver, kidney and muscle of striped dolphins caught off Taiji were markedly higher than those of common dolphins (Honda, 1990). Discrimination among these dolphins is necessary to better describe the species differences in T-Hg and M-Hg contamination. Although there were no kidney samples from pilot whales in the present survey, the level of Cd concentration in each boiled organ increased according to species: S/T/D dolphins < Risso's dolphins < pilot whales (Fig. 1). This order of Cd contamination is consistent with the data for heavy metals in cetaceans caught off the coast of Japan (Honda et al., 1990). The levels of T-Hg and M-Hg contamination in the red meat as well as the boiled internal organs originating from the mysticete species (minke and Bryde's whales) were much lower than those from odontocetes, probably reflecting differences in their trophic levels. We previously reported similar differences in organochlorine levels in the red meat and bacon from odontocete and mysticete (Haraguchi et al., 2000).

In response to the Minamata disease tragedy, the Japanese Ministry of Health, Labor and Welfare (JMHLW) set provisional permitted levels of T-Hg and M-Hg in marine foods at 0.4 and 0.3 μg Hg/wet g, respectively, to prevent chronic intoxication by M-Hg (JMHLW, 1973). WHO set the provisional tolerable weekly intakes (PTWI) of T-Hg and M-Hg at 300 μg/60 kg and 200 μg/60 kg bw/week, respectively (WHO, 1972). Notice that the maximal values of T-Hg and M-Hg detected in the boiled liver (1980 and 25.7 μg/wet g) exceed these limits by about 5000 and 85 times, respectively. According to Bidstrup (1964), the ingestion of 500 mg of mercuric chloride causes severe poisoning and sometimes death in humans. Although most of the

Hg in the boiled liver appears to exist as the less toxic compound HgSe (Endo et al., 2002), the consumption of boiled liver still provides enough Hg to cause acute renal intoxication (Endo et al., 2003). In the present survey, the maximal values of T-Hg and M-Hg found in red meat products, the most popular whale products in Japan, were 63.4 and 26.2 μg Hg/wet g, respectively. The consumption of only 4.7 and 7.6 g, respectively, of this red meat exceeds the PTWI of T-Hg and M-Hg for a 60-kg consumer. On the other hand, WHO set the PTWI of Cd at 6.7–8.3 μg/kg bw/week (WHO, 1972). As the level of Cd contamination in the odontocetes caught off the coast of Japan is markedly lower than the levels of T-Hg and M-Hg contamination, Cd intoxication due to the consumption of whale products may not be as serious a problem as T-Hg and M-Hg intoxication.

Having recognized that environmental pollutants such as Hg and PCBs enter the body of pregnant women via pilot whale products, causing potentially serious developmental damage to their infants, the government of the Faroes Island, Denmark, issued the following recommendations to the public in 1998 (Anonymous, 1998).

- Adults should only eat blubber and meat once or twice a month;
- Girls and women should not eat blubber until they have given birth to all their children;
- Meat should not be eaten within three months of planned pregnancy and not eaten at all by pregnant and nursing women; and
- Organs (e.g. liver and kidney) should not be eaten at all.

In 2001, the US Food and Drug Administration (FDA, 2001) called attention to the fact that pregnant women and women of childbearing age should not eat longer-living and larger fish that feed on other fish such as shark, swordfish, mackerel and tilefish (the average of M-Hg of each species is above 0.7 μg/wet g) because they may contain enough M-Hg to damage the fetus's nervous system.

In conclusion, T-Hg concentration in the liver of odontocetes was extremely high. The levels of T-Hg and M-Hg contamination in red meat originating from odontocetes were markedly higher than the provisional permitted levels set by JMHLW. More attention must be paid to the need for recommendations like those made by the Faroes Island and the US FDA. Although there was no information on age of cetacean, levels of T-Hg and M-Hg contamination were slightly higher in females than in males. Species differences in Hg levels were unclear because of the small number of samples, the incomplete identification of striped, bottlenose and common dolphins, and lack of information concerning age. A larger scale survey and fully resolved identification of all species involved are necessary to elucidate the actual contamination levels of Hg and Cd in the cetacean products in Japan.

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