Total Mercury, Methyl Mercury, and Selenium Levels in the Red Meat of Small Cetaceans Sold for Human Consumption in Japan

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We surveyed the total mercury (T-Hg) and methyl mercury (M-Hg) levels in red meat products ($n = 160$) from small cetacean species sold for human consumption in markets throughout Japan from 2000 to 2003. Genetic identification showed that the red meat products originated from nine species: false killer whale, bottlenose dolphin, short-finned pilot whale, striped dolphin, rough-toothed dolphin, Risso's dolphin, pantropical spotted dolphin, Baird's beaked whale, and Dall's porpoise. T-Hg and M-Hg concentrations in all red meat products exceeded the provisional permitted levels of T-Hg (0.4 μg/wet g) and M-Hg (0.3 μg/wet g) in fish and shellfish set by the Japanese government, respectively. The average M-Hg level in the most contaminated species (false killer whale) was 11.5 μg/wet g, and that in the least contaminated species (Dall's porpoise) was about 1.0 μg/wet g, exceeding or equaling the Codex guideline of M-Hg in predatory fishes (1.0 μg/wet g). Contamination levels of T-Hg and M-Hg differed considerably among samples of the nine species and among individuals of a particular species. The highest M-Hg was about 26 μg/wet g in a sample from a striped dolphin, 87-times higher than the permitted level. The consumption of only 4 g of this product would exceed the provisional tolerable weekly intake of M-Hg for someone of 60 kg body weight (1.6 μg/kg-bw/week). Although a high correlation between T-Hg and selenium (Se) was observed in these products, the molar ratio of T-Hg to Se was substantially higher than 1. The consumption of red meat from small cetaceans, therefore, could pose a health problem for not only pregnant women but also for the general population.

Introduction

The consumption of whale products in Japan has decreased since 1986, due to a significant reduction in Japanese whaling as a consequence of the International Whaling Commission moratorium on commercial whaling. Thirteen species of great whales (baleen whales and the sperm whale, the largest toothed whale) are covered by this moratorium. However, the hunting of smaller odontocetes or toothed whales (small cetaceans), such as Baird's beaked whale (*Berardius bairdii*), the short-finned pilot whale (*Globicephala macrorhynchus*), and other dolphins and porpoises, is not covered by the moratorium and is generally regulated by national governments. The halt in the commercial whaling of larger types of whales has resulted in an increased demand and increased price of small cetacean products (*1*). To supply this market, approximately 17,000 small cetaceans are caught annually in Japanese coastal waters through drive fisheries, hand-harpoon hunting, and small-type whaling. The main species of small cetaceans taken in these regulated hunts are Dall's porpoise (*Phocoenoides dalli*), Baird's beaked whale, short-finned pilot whale, pantropical spotted dolphin (*Stenella attenuata*), Risso's dolphin (*Grampus griseus*), rough-toothed dolphin (*Steno bredanensis*), striped dolphin (*Stenella coeruleoalba*), bottlenose dolphin (*Tursiops truncatus*), and false killer whale (*Pseudorca crassidens*) (*2*).

As small cetaceans are long-lived and occupy the top of the marine food web, they accumulate or biomagnify marine pollutants such as heavy metals and organochlorine compounds (*3*, *4*). Among these pollutants, contamination with mercury (Hg) is prominent. A high level of Hg is known to accumulate not only in the internal organs of small cetaceans but also in the muscle (*5*−*11*). Marine mammals are principally exposed to methyl mercury (M-Hg), because almost all of the Hg present in fish and squid is methylated (*12*, *13*). 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 complex of I-Hg and selenium (Se), is thought to occur mainly in cetacean livers (*8*, *12*, *14*−*16*). In contrast to the internal organ samples (liver, lung, and kidney), the demethylation of M-Hg has not been clearly observed in samples of muscle (red meat), probably because of low concentration of Hg, low activity of the demethylation, and/or small number of red meat samples (*11*).

In response to the Minamata tragedy, where the local human population was poisoned by eating fishes contaminated with M-Hg from industrial waste, the Japanese Ministry of Health and Welfare (JMHW) set the provisional levels of total mercury (T-Hg) and M-Hg in fishes and shellfishes at 0.4 and 0.3 μg/wet g, respectively (*17*). These levels were based on the provisional tolerable weekly intake (PTWI) of M-Hg set at 3.3 μg/kg-body weight (bw) by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) in 1972. Only recently, however, has there been public concern about Hg contamination of some cetacean products. In a previous related survey, we reported on levels of T-Hg in red meat of large and small cetacean sold for human consumption in Japanese market (*10*). Although this survey included market products not genetically analyzed, all red meat assumed to have derived small cetaceans in this survey ($n = 137$) contained high levels of T-Hg, exceeding the provisional permitted level (0.4 μg/wet g), with the highest concentration being 81 μg/wet g in a false killer whale. In parallel with these data, the Japanese Ministry of Health, Labor, and Welfare (JMHLW, JMHW expanded to JMHLW in 2001) investigated contamination levels of T-Hg and M-Hg in five species of toothed whales, and advised pregnant women to limit consumption of whale meats originating from sperm whale, short-finned pilot whale, bottlenose dolphin, and Baird's

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beaked whale, based on the PTWI of M-Hg (3.3 μg/kg-bw) (*18*). However, the size and range of the JMHLW survey was limited, and soon after this advice the PTWI of M-Hg was revised from 3.3 μg/kg-bw to 1.6 μg/kg-bw to protect against neurodevelopment disorders in the fetus (*19*).

Here, we report on a large-scale survey of red meat products from small cetaceans purchased in Japanese markets from 2000 to 2003. Red meat is the most popular cetacean product in Japan and originates from a wide range of whale, dolphin, and porpoise species. Although the Hg contamination levels vary considerably according to species origin (*10*), the labeling of some whale products is sometimes incomplete or incorrect (*1*). For this reason, we used genetic identification to verify the species of origin for most market products (*20*). Furthermore, we include data on M-Hg and Se, the most important toxicological data for the consumption of marine products. These measurements were not included in the previous related survey of red meat products (*10*).

Materials and Methods

Sampling of Market Products. As reported previously (*10*), fresh and frozen red meats from cetaceans were purchased in Japan between 2000 and 2003, in and around Abashiri and Hakodate (Hokkaido Prefecture), Otsuchi (Iwate Prefecture), Ayukawa (Miyagi Prefecture), Tokyo Metropolitan, Wada (Chiba Prefecture), Yaidu and Ito (Shizuoka Prefecture), Taiji (Wakayama Prefecture), Ohsaka (Osaka Prefecture), Saga (Saga Prefecture), and Nago (Okinawa Prefecture). Most of the red meat samples purchased from Nago were boiled products sold as frozen food. These samples were stored at −20 °C until analysis.

Chemical Analyses. T-Hg in the red meat was analyzed using a flameless atomic absorption spectrophotometer (Hiranuma Sangyo Co. Ltd., HG-1) after digestion by a mixture of HNO_3, $HClO_4$, and H_2SO_4 (*9*). M-Hg was determined using a gas chromatograph (Shimazu Co. Ltd., GC-14A) with a ^{63}Ni electron capture detector (*3*). M-Hg concentration was expressed on the basis of Hg concentration. Selenium (Se) was determined by the method of Watkinson (*21*) using a Hitachi fluorescence spectrophotometer F-450 after digestion in a HNO_3-HClO_4 mixture. As reported previously (*9*−*11*), 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 Se. All metals concentrations in the red meats presented were based on wet weight and were expressed as the mean ± standard deviation (SD).

Species Identification. As reported in detail previously (*20*), the species origin of most read meat products was identified by comparison of mitochondrial (mt) DNA sequences amplified from the products. Identification of species origin by comparison of mtDNA sequences was unambiguous except for some species of the genus Stenella. These species are difficult to distinguish from each other with statistical confidence because of intraspecific diversity and low intraspecific divergence (*22*). However, given the reported pattern of small cetacean hunting around Japan (*23*), it is likely that these products originated from the striped dolphin. The southern and northern "forms" of the short-finned pilot whales were discriminated here by the location of the purchases relative to their assumed geographic ranges (*2*).

Results

A total of 160 products of cetacean red meat intended for human consumption were purchased in Japanese markets. Genetic identification confirmed that the products originated from nine species of small cetaceans: false killer whale, bottlenose dolphin, short-finned pilot whales, striped dolphin, rough-toothed dolphin, pantropical spotted dolphin, Risso's dolphin, Baird's beaked whale, and Dall's porpoise. Furthermore, we discriminated the southern and northern forms of short-finned pilot whales by the location of purchased areas (Table 1).

All T-Hg and M-Hg concentrations in the red meat samples (n = 160) exceeded the permitted level of T-Hg (0.4 μg/wet g) and M-Hg (0.3 μg/wet g) in fish and shellfish set by JMHW (*17*). The red meat products originating from false killer whales contained the highest average levels of T-Hg (39.5 ± 28.4 μg/wet g) and M-Hg (11.2 ± 1.9 μg/wet g), although the sample size was limited in number (n = 4). One product containing 81.0 μg/wet g of T-Hg and 13.3 μg/wet g of M-Hg was purchased from Nago in 2001, and the others were purchased from Taiji and nearby area in 2002 (n = 2) and Nago in 2003 (n = 1). The second most contaminated species samples originated from bottlenose dolphins (T-Hg = 17.8 ± 19.3 μg/wet g, M-Hg = 6.83 ± 3.99 μg/wet g, n = 37) was purchased in and around Nago (n = 32) and Taiji (n = 5). One product containing 98.9 μg/wet g of T-Hg and 15.4 μg/wet g of M-Hg was purchased from Nago in 2003.

The third most contaminated samples (T-Hg = 11.6 ± 8.2 μg/wet g, M-Hg = 6.45 ± 3.53 μg/wet g, n = 34), originating from the presumed southern short-finned pilot whale products, were purchased in and around Nago (n = 16) and Taiji (n = 18). These levels were markedly higher than the products from the presumed northern short-finned pilot whale products purchased around Ayukawa (n = 8), which ranked eighth among the 10 species. The fourth most contaminated species samples (T-Hg = 8.55 ± 14.2 μg/wet g, M-Hg = 3.74 ± 5.44 μg/wet g, n = 20) were striped dolphin products purchased in and around Nago (n = 4), Taiji (n = 14), Wada (n = 1), and Tokyo Metropolitan (n = 1). One striped dolphin product containing 63.4 μg/wet g of T-Hg and 26.2 μg/wet g of M-Hg was purchased in metropolitan Tokyo in 2001.

Three species showed moderate levels of contamination (T-Hg ≈ 4−5 μg/wet g): rough-toothed, Risso's, and pantropical spotted dolphins. The samples of rough-toothed dolphins (n = 5) and pantropical spotted dolphins (n = 4) were purchased from Nago, and the samples of Risso's dolphin were purchased from Taiji (n = 15), Ohsaka (n = 1), and Saga (n = 1). The lowest average levels of contamination (T-Hg ≈ 1.5 μg/wet g) were found in the Baird's beaked whale, the northern short-finned pilot whale, and the Dall's porpoise. The samples of Baird's beaked whales were purchased from the Abashiri (n = 3), Hakodate (n = 12), Ayukawa area (n = 5), and Wada area (n = 2). The samples of Dall's porpoises were purchased in and around Otsuchi (n = 4), Yaidu (n = 1), Ito (n = 1), and Nago (n = 3), and their contamination levels of T-Hg and M-Hg were the lowest among the 10 species.

The relationship between T-Hg and M-Hg is shown in Figure 1. The M-Hg concentration appears to reach the plateau level (about 15 μg/wet g) with an increase in the T-Hg concentration, with one outlying sample exception. The inset shows the relationship between the average levels of T-Hg and M-Hg from 10 species (Table 1). These were well fitted to two straight lines with the intersection point being 6.5 μg/wet g. The data for T-Hg and percentage of M-Hg/T-Hg showed a logarithmic relationship ($y = -29.4 \log x + 96.1$, $R^2 = 0.532$, $n = 160$), similar to that found previously for the internal organs of small cetaceans (*11*). As expected from this relationship, the percentage of M-Hg in T-Hg found in Dall's porpoise samples was the highest and that in false killer whale samples was the lowest.

Averages of molar ratio of T-Hg to Se in the red meat products originating from eight species were between 1.3 and 2.4 (Table 1). A high correlation was observed between T-Hg and Se in those products (Figure 2, $n = 129$, $R^2 = 0.822$), and the molar ratio of T-Hg to Se calculated from the

TABLE 1. Total Mercury, Methyl Mercury, and Selenium Concentrations in Odontocete Red Meats (Muscles) Caught around Japan

species	n	total mercury (T) (μg/wet g)	methyl mercury (M) (μg/wet g)	M/T (%)	selenium (Se) (μg/wet g)	molar ratio of T to Se ((T-M) to Se)	ref
Dall's porpoise	9	1.23 ± 0.49 (0.83−2.39)	1.02 ± 0.38 (0.68−1.95)	84 ± 10	N.D.		this study
	4	1.0 ± 0.20[a] (0.74−1.2)	0.37 ± 0.33[a] (0.02−0.67)	33 ± 28[a]	N.D.		JMHLW (*18*)
	47	0.90 ± 0.56	N.D.		N.D.		Honda et al. (*6*)
short-finned pilot whale	8	1.50 ± 0.48 (0.79−2.24)	1.25 ± 0.50 (0.50−1.88)	81 ± 11	0.51 ± 0.16 (0.50−1.88)	1.3 ± 0.8 (0.20 ± 0.08)	this study
(northern form)	31	2.78 ± 0.80					Honda et al. (*6*)
Baird's beaked whale	22	1.77 ± 1.43 (0.75−6.46)	1.25 ± 0.78 (0.56−3.47)	78 ± 14	N.D.		this study
	5	1.2 ± 0.9[a] (0.44−2.6)	0.70 ± 0.43 (0.37−1.3)	67 ± 14[a]	N.D.		JMHLW (*18*)
	37	1.56 ± 1.11	N.D.		N.D.		Honda et al. (*6*)
pantropical spotted dolphin	4	4.87 ± 0.44 (4.28−5.32)	2.62 ± 0.49 (2.01−3.16)	54 ± 7	0.95 ± 0.21 (0.77−1.21)	2.1 ± 0.3 (0.97 ± 0.15)	this study
	53	3.64 ± 2.21 (1.05−12.0)	N.D.		1.36 ± 0.52 (0.76−4.19)		Chen et al. (*25*)
Risso's dolphin	17	4.46 ± 2.30 (1.71−9.21)	3.15 ± 1.76 (1.33−8.78)	74 ± 20	0.93 ± 0.42 (0.63−2.37)	1.9 ± 0.8 (0.52 ± 0.48)	this study
	4	3.17 ± 4.36 (0.71−9.70)	N.D.		1.77 ± 1.29 (0.78−3.63)		Chen et al. (*25*)
rough-toothed dolphin	5	5.02 ± 3.63 (1.22−9.98)	3.51 ± 2.34 (1.11−6.06)	74 ± 15	1.04 ± 0.40 (0.69−1.71)	1.8 ± 0.9 (0.49 ± 0.35)	this study
	13	1.02 ± 0.48	N.D.		N.D.		Honda et al. (*6*)
striped dolphin	20	8.55 ± 14.2 (1.04−63.4)	3.74 ± 5.44 (0.97−26.2)	63 ± 23	2.54 ± 3.50 (0.58−16.2)	1.3 ± 0.7 (0.59 ± 0.60)	this study
	11	4.63 ± 2.57[a] (0.95−9.43)	3.00 ± 1.49[a] (0.31−5.88)	67 ± 19[a]	1.03 ± 0.70[a] (0.22−2.48)	1.9 ± 0.4[a]	Arima and Nagakura (*24*)
	26	15.2 ± 8.0	5.3 ± 0.8	35	2.8 ± 2.2		Itano et al. (*5*)
	59	7.02 ± 4.07	N.D.		N.D.		Honda et al. (*6*)
short-finned pilot whale	34	11.6 ± 8.2 (1.21−37.6)	6.45 ± 3.53 (0.93−17.2)	64 ± 20	1.97 ± 1.54 (0.82−7.91)	2.6 ± 1.3 (0.99 ± 0.90)	this study
(southern form)	4	7.1 ± 1.9[a] (4.71−8.9)	1.5 ± 0.9[a] (0.45−2.1)	22 ± 12[a]	N.D.		JMHLW (*18*)
	12	4.16 ± 0.72[a] (3.01−5.18)	N.D.		0.87 ± 0.22[a] (0.61−1.30)	2.0 ± 0.5[a]	Arima and Nagakura (*24*)
bottlenose dolphin	37	17.8 ± 19.3 (0.59−98.9)	6.83 ± 3.99 (0.58−15.4)	54 ± 23	3.79 ± 5.09 (0.88−26.7)	2.3 ± 1.2 (1.1 ± 0.9)	this study
	5	21 ± 16[a] (1.0−37)	6.6 ± 3.9[a] (0.61−9.7)	42 ± 17[a]	N.D.		JMHLW (*18*)
	1	51.8	9.00		13.9		Arima and Nagakura (*24*)
false killer whale	4	39.5 ± 28.4 (17.4−81.0)	11.2 ± 1.9 (9.02−13.3)	36 ± 15	7.16 ± 5.43 (3.11−15.2)	2.2 ± 0.1 (1.4 ± 0.3)	this study

[a] Calculated values shown as mean ± SD (minimum−maximum) where available from the reference. N.D.: not determined.

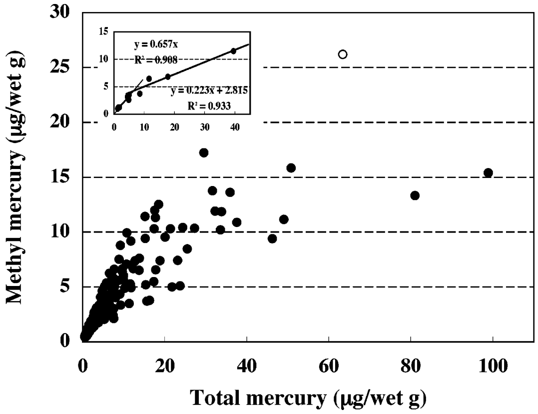

FIGURE 1. Relationship between total and methyl mercury concentrations in odontocete red meat products sold for human consumption in Japan (n = 160). One striped dolphin sample containing T-Hg = 63.4 μg/wet g and M-Hg = 26.2 μg/wet g is shown by ○. The inset shows the relationship between the mean concentrations of total and methyl mercury from the 10 species (see Table 1).

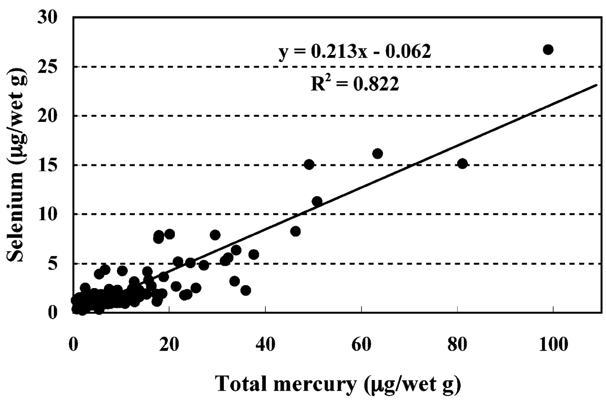

FIGURE 2. Relationship between total mercury and selenium concentrations in odontocete red meat products sold for human consumption in Japan (n = 129; see Table 1).

regression line of slope was 1.9. We estimated the amount of inorganic mercury (I-Hg) by the difference between T-Hg and M-Hg, and calculated the molar ratio of estimated I-Hg to Se. Averages of this molar ratio shown in the parentheses of Table 1 were between 0.2 and 1.4.

Discussion

Government Advisories and Human Health Risks. All T-Hg and M-Hg concentrations in red meat originating from the 10 species of small cetaceans in our survey (includes two "forms" of short-finned pilot whales, $n = 160$) exceeded the provisional permitted level of T-Hg (0.4 μg/wet g) and M-Hg (0.3 μg/wet g) in marine foods set by Japanese government (*17*). The highest concentration of M-Hg was 26.2 μg/wet g in a sample from a striped dolphin. This is 87 times higher than the permitted level. Many products from the southern form of short-finned pilot whales, bottlenose dolphins, and false killer whales exceeded 10 μg/wet g of M-Hg. Consumption of only a small portion of these products (4−10 g) would exceed the revised PTWI of M-Hg for someone of 60 kg-bw (1.6 μg/kg-bw/week). Even in the least contaminated species (the Dall's porpoise), the average level of M-Hg was about 1.0 μg/wet g, equal to the Codex guideline of M-Hg in predatory fishes such as shark, swordfish, tuna, and pike. To our knowledge, 26.2 μg/wet g of M-Hg found in a striped dolphin sample and 98.9 μg/wet g of T-Hg found in a bottlenose dolphin sample (Table 1) are the highest values reported in the muscles (red meats) of small cetaceans to date. At these levels, consumption of red meat from small cetaceans could pose health problems for the general population as well as high risk segments of the population such as pregnant women.

The Japanese government has conducted its own survey of Hg contamination in fishes and red meat of small cetaceans and, as a result, issued specific advice for pregnant women (*18*). According to Government's data on odontocetes, the means of T-Hg and M-Hg concentrations (μg/wet g) in Dall's porpoises ($n = 4$) were 1.0 and 0.37, those in Baird's beaked whales ($n = 5$) were 1.2 and 0.70, those in short-finned pilot whales ($n = 4$) were 7.1 and 1.5 (no special mention of southern or northern form), and those in bottlenose dolphins ($n = 5$) were 21.0 and 6.6, respectively (Table 1). The average levels of T-Hg, M-Hg, and the percentage of M-Hg in bottlenose dolphin samples calculated from Government's survey were similar to those found in our survey. For Dall's porpoise samples, however, averages contamination of M-Hg and percentage of M-Hg in the Government's survey (0.37 ± 0.33 and 33 ± 28) were markedly lower than in ours (1.02 ± 0.38 and 84 ± 10), although the averages of T-Hg were similar. The percentage of M-Hg tends to decrease with an increase in T-Hg concentration (Table 1), and our percentage of M-Hg in Dall's porpoise samples is consistent with this tendency.

The Japanese Government advised pregnant women to limit consumption of red meats from Baird's beaked whale and pilot whale to no more than once a week and bottlenose dolphin meat to no more than once every 2 months, according to their own data (*18*). However, this advisory was based on the previously accepted PTWI of 3.3 μg/kg-bw/week for M-Hg and did not consider other species of small cetaceans available on the market. Soon after this JMHLW advisory, the JECFA revised the PTWI down to 1.6 μg/kg-bw/week (*19*). Based on the revised PTWI and the results of the our more extensive survey, there is now an urgent need to revise the JMHLW advisory to pregnant women and extend it to other species of small cetaceans.

Comparison to Other Hg Contamination Surveys. *Honda* (*6*) analyzed the distribution of T-Hg in several species of whales, dolphins, and porpoises caught off Japan. According to his data, T-Hg concentration levels in the muscles of Dall's porpoises caught in the Bering Sea and N. Pacific Ocean, short-finned pilot whales caught off Ayukawa (northern form), Baird's beaked whales caught off Wada, rough-toothed dolphins caught off Taiji, and striped dolphins caught off Taiji generally agree with those we found in the red meats marketed for human consumption, except for the rough-toothed dolphin samples (Table 1). In our sample of rough-toothed dolphin from Nago, the average of T-Hg was 5 times higher than reported for this species by Honda (*6*) 15 years ago.

Many research groups have investigated Hg contamination levels in the red meat of striped dolphin. Arima and Nagakura (*24*) reported relatively low concentrations of T-Hg and M-Hg in the muscle of striped dolphin samples caught off Japan. The mean ± SD of T-Hg, M-Hg, and the percentage of M-Hg/T-Hg, calculated from their data, were 4.63 ± 2.57 (μg/wet g), 3.00 ± 1.49 (μg/wet g), and 67 ± 19%, respectively. In contrast, Itano et al. (*5*) reported high contamination levels of T-Hg and M-Hg and a low percentage of M-Hg/T-Hg in the muscle of striped dolphin samples caught off Shizuoka and Wakayama Prefectures, Japan: 15.2 ± 8.0 and 5.3 ± 0.8 (μg/wet g) and 35%, respectively. They noted that T-Hg level increased with age, and 70−100% of T-Hg was found as M-Hg in the muscle of striped dolphin samples below 10 years of age. Our data of T-Hg, M-Hg, and the percentage of M-Hg are intermediated values reported between those reports. The highest concentration of T-Hg found in the striped dolphin samples (Table 1) was 63.4 μg/wet g. Andre et al. (*8*) reported high concentrations of T-Hg in the muscle of striped dolphin samples caught along the Mediterranean coasts with the three highest values being 60.0, 68.1, and 81.2 μg/wet g.

Prior to the results presented here, little information was available on Hg contamination of short-finned pilot whales (southern and northern forms), bottlenose dolphins, Risso's dolphin, pantropical spotted dolphin, and false killer whale. Our averages of T-Hg in the southern and northern forms of short-finned pilot whale samples were somewhat lower and higher than those by Arima and Nagakura (*24*) and Honda (*6*), respectively. Arima and Nagakura (*24*) also reported the T-Hg, M-Hg, and Se concentrations in the muscle from a bottlenose dolphin samples. Chen et al. (*25*) reported the contamination levels of T-Hg in the red meats samples of Risso's dolphin and pantropical spotted dolphin caught off Taiwanese waters, southern area of Okinawa Prefecture. Our results for bottlenose, Risso's, and pantropical spotted dolphins roughly agree with these previous reports. To our knowledge, there is no report concerning the contamination level of Hg in false killer whale, except for the previous report (*10*). False killer whales are presumably located at the highest position of the marine food web and accumulate Hg by eating large predatory fish, such as tuna, as well as other species of dolphins (*26*).

As stated above, contamination levels of T-Hg and M-Hg in cetaceans are markedly different among species (Table 1) and increased with age (*5*, *14*, *15*). Furthermore, contamination may vary according to living area. Previously, we reported that contamination levels of T-Hg in Baird's beaked whales and short-finned pilot whales (southern and northern forms) caught off southern areas were higher than those caught off northern areas although most of samples from the previous related survey were not genetically analyzed (*10*). In the present survey, contamination levels of T-Hg (11.5 ± 10.6 μg/wet g) and M-Hg (5.92 ± 4.38 μg/wet g) in southern form of short-finned pilot whales purchased in and around Nago (south, $n = 16$) were similar to T-Hg (11.2 ± 5.7 μg/wet g) and M-Hg (6.68 ± 2.74 μg/wet g) of those purchased in and around Taiji (north, $n = 18$). In contrast, the levels of T-Hg (13.4 ± 7.1 μg/wet g) and M-Hg (4.40 ± 0.62 μg/wet g) in striped dolphins purchased from Nago area ($n = 4$) were higher than T-Hg (8.15 ± 15.8 μg/wet g) and M-Hg (3.92 ± 6.31 μg/wet g) of those from Taiji area ($n = 14$), and the levels of T-Hg (19.2 ± 19.4 μg/wet g) and M-Hg (7.27 ± 3.92 μg/wet g) in bottlenose dolphins from Nago area ($n = 32$) were higher than T-Hg (6.67 ± 6.94 μg/wet g) and M-Hg (3.60 ± 2.66 μg/wet g) of those from Taiji area ($n = 5$), although sample sizes of striped dolphin from Nago area and bottlenose dolphin from Taiji area were limited. Our survey is now in progress to clarify the regional difference in Hg contamination of cetaceans.

Relationship of T-Hg, M-Hg, and Se. Many researchers have postulated the demethylation of M-Hg, followed by the formation of an Hg−Se complex in the internal organs of cetaceans, especially in liver (*8*, *12*, *14*−*16*). We previously reported the contamination levels of T-Hg and M-Hg in the internal organ samples originating from cetaceans: Contamination levels of T-Hg and M-Hg in the odontocete livers were 424 ± 583 and 12.0 ± 5.9 μg/wet g ($n = 20$), in the odontocete lungs were 39.9 ± 44.5 and 2.05 ± 2.25 μg/wet g ($n = 23$), and in the odontocete kidney were 42.5 ± 49.7 and 4.34 ± 5.40 μg/wet g ($n = 14$), respectively (*11*). The contamination level of T-Hg in the liver samples was about 10 times higher than that in red meat samples from false killer whale, although contamination levels of M-Hg were similar. In addition, contamination levels of T-Hg in the lung and kidney samples were similar to that in the red meat samples of false killer whale, although the contamination levels of M-Hg in the formers were markedly lower that in the latter. Thus, the percentage of M-Hg/T-Hg can be higher in the red meat samples than in the internal organs, especially in liver.

We previously reported the increase in T-Hg contamination levels in boiled liver, lung, and kidney samples of small cetaceans tended to decrease the percentage of M-Hg/T-Hg, and these were well fitted to a logarithmic equation (*11*). However, such a tendency was not observed in the mixed samples of red meats originating from odontocetes ($n = 22$) and mystecetes ($n = 15$) (*11*). In the larger sample size of the present study of small cetaceans ($n = 160$), however, the increase in T-Hg contamination level showed a logarithmic relationship with a decrease in percentage M-Hg/T-Hg (figure not shown). The more limited sample of red meat products, as well as the inclusion of mysticete red meat products and the absence of highly contaminated samples, may have obscured this tendency in the previous analysis.

Chen et al. (*25*) fitted their data of T-Hg and organic mercury concentrations from the muscle of pantropical spotted dolphin samples ($n = 53$) to two straight lines and suggested that the demethylation is activated at above the intersection point of the two lines (4 μg/wet g). Itano et al. (*5*) reported that M-Hg concentration in the muscle of striped dolphin samples increased with their ages and reached a constant level at about 6 μg/wet g. Similar phenomena were observed in the present data (Figure 1): The plot of mean T-Hg and M-Hg for the 10 species (includes two forms) in our survey showed a good fit to two straight lines with an intersection point of about 6.5 μg/wet g, and a plateau level at about 15 μg/wet g for M-Hg. This intersection point and plateau level could reflect the demethylation activity of M-Hg mainly in cetacean liver.

The one-to-one molar association of Hg and Se, tiemannite HgSe, in the liver of many mammals is well documented (*13*, *15*, *16*, *27*). We previously found a high correlation between T-Hg and Se concentrations in boiled liver, kidney, and lung samples, and we reported the molar ratios of T-Hg to Se in those samples as 1.18, 1.11, and 1.43, respectively (*9*). In contrast, the molar ratios of T-Hg to Se in the muscle (red meat) have been reported to be substantially higher than those in the liver (*27*). Averages of molar ratio of T-Hg to Se found in eight species were between 1.3 and 2.4 (Table 1), and the molar ratio of T-Hg to Se in these products calculated from the slope of regression lines was 1.9, substantially higher than 1 (Figure 2). The molar ratios of estimated I-Hg to Se found in the northern form of short-finned pilot whale, Risso's dolphin, rough-toothed dolphin, and striped dolphin (low and moderate levels of Hg contamination) were markedly lower than 1 (Table 1). Se is an essential metal, and its endogenous level in these red meats would result in the low molar ratio of I-Hg to Se.

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