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REGULAR ARTICLE

Preliminary assessment of ecological exposure of adult residents in Fukushima Prefecture to radioactive cesium through ingestion and inhalation

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Abstract

Objective This study aims to estimate the ecological exposure of adult residents of Fukushima Prefecture to ^{134}cesium (Cs) and ^{137}Cs through ingestion and inhalation between July 2 and July 8, 2011.

Methods Fifty-five sets of meals with tap water, each representing one person's daily intake, were purchased in local towns in Fukushima Prefecture. Locally produced cow's milk (21 samples) and vegetables (43 samples) were also purchased. In parallel, air sampling was conducted at 12 different sites using a high-volume sampler. Nineteen sets of control meals were collected in Kyoto in July 2011. ^{134}Cs and ^{137}Cs levels in the samples were measured using a germanium detector.

Results Radioactivity was detected in 36 of the 55 sample meals from Fukushima, compared with one of 19 controls from Kyoto. The median estimated dose level (μSv/year) was 3.0, ranging from not detectable to 83.1. None of the cow's milk (21) or vegetable (49) samples showed levels of contamination above the current recommended limits (Bq/kg) of 200 for milk and 500 for vegetables. The total effective dose levels by inhalation were estimated to be <3 μSv/year at nine locations, but samples at three other locations close to the edge of the 20-km radius from the crippled nuclear power plant showed higher levels of contamination (μSv/year): 14.7 at Iitate, 76.9 at Namie, and 27.7 at Katsurao.

Conclusions Levels of exposure to ^{134}Cs and ^{137}Cs in Fukushima by ingestion and inhalation are discernible, but generally within recommended limits.

Keywords ^{134}Cs · ^{137}Cs · Exposure assessment · Fukushima Daiichi nuclear power plant accident · Ingestion · Inhalation

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Introduction

Following the Tohoku earthquake and tsunami on March 11, 2011, the Fukushima Daiichi nuclear power plant exploded on March 15, 2011, releasing massive amounts of radionuclides, including iodine, cesium (Cs), strontium, and plutonium into the northern part of Japan and the Pacific Ocean, being the second largest nuclear accident, after the Chernobyl disaster [1, 2]. The total amount of ^{137}Cs released into the environment by the Fukushima Daiichi nuclear plant from March 11 to April 15

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(1.3×10^{16} Bq) [3] has been estimated to be 10% of that emitted by the Chernobyl disaster in 1986 [1, 2].

Residents living within a 20-km radius of the nuclear power plant were evacuated soon after the disaster, but people in Fukushima Prefecture have continued to live outside this evacuation zone. Although the direct threat from the radioactive plume is over, it is important to continuously assess the exposure doses due to deposited radioactivity. Contamination with ^{137}Cs has been reported in residential areas in Fukushima Prefecture [4], and the internal doses resulting from inhalation of resuspended deposits [5] and ingestion of contaminated foods need to be monitored.

Residents in particular, but also people in remote areas, are seriously concerned about their levels of internal exposure to radionuclides through ingestion of contaminated food and drink. The ingested dose should be evaluated on the basis of the level of radioactivity contained in complete meals consumed (Bq/day/person), rather than on the radioactive content of an individual item (Bq/kg).

To evaluate potential post-accident internal doses, we conducted a field survey in July 2011, focusing on estimated exposures of adult residents of Fukushima Prefecture to ^{134}Cs and ^{137}Cs through ingestion and inhalation.

Materials and methods

Field survey

We tested whole-day meals, vegetables from local food venders, tap water, and air samples from cities at various distances from the nuclear power plant between July 2 and July 8, 2011 (Fig. 1). In the cities denoted as "M" and "V" in Fig. 1, we purchased whole-day meals and vegetables from local food venders, respectively. Tap water was also collected in the same towns or cities. In the cities denoted by "A," we conducted air sampling using a high-volume sampler (HV-1000F; Sibata, Saitama, Japan) and soil sampling (mixed soil samples from depth of 0–5 cm). We also collected continuous air samples at a fixed point in Fukushima City using a low-volume sampler (SL-30; Sibata, Saitama, Japan) with an eight-stage Andersen cascade impactor sampler (AN-200; Tokyo Dylec Co., Tokyo, Japan).

Food collection and processing for radioactivity determinations

Five male researchers (aged 32–68 years) visited one of the most popular local grocery stores in each city or town and purchased several sets of whole-day meals, according to their personal preferences, as reported previously [6]. A set of whole-day meals comprised prepackaged breakfast, lunch, and dinner, as well as desserts, snacks, and

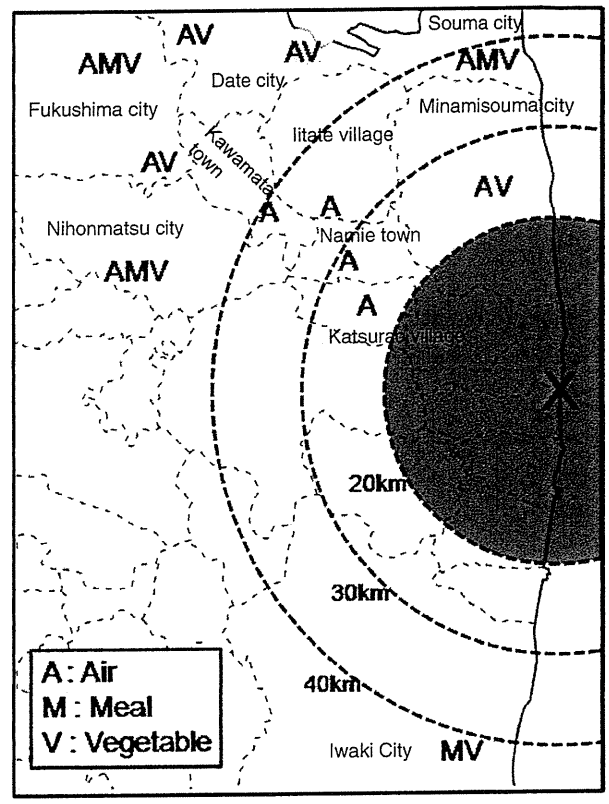

Fig. 1 Geographical locations of the field study areas. "A" represents sites where air sampling was conducted. "M" represents grocery stores where meals were purchased. Tap water (12 L) was collected in the same towns where meals were purchased. "V" represents commercial vender where vegetables were purchased. "X" represents the Fukushima Daiichi nuclear power plant. The *symbols* approximately represent actual geographical positions

beverages. A total of 12 L geographically matched tap water per town was donated by residents of the towns where the grocery stores were located. Locally produced vegetables and cow's milk were also purchased in the same towns. All items were transported daily to Kyoto University at 4°C for processing and analysis.

Daily whole-day meal sets were homogenized with locally collected tap water (approximately 1 L), together with desserts and snacks. The final volumes were recorded, and approximately 1 L of each homogenate was processed for freeze-drying. Vegetables and cow's milk were also freeze-dried. Control meals consisted of whole-day meals collected by 19 females using the food duplicate method, as previously reported [6]. Control meals were collected in July 2011 in Uji, Kyoto, which is located from 540 km to the southwest of the Fukushima nuclear power plant.

Air sampling and determination of radioactivities

A high-volume air sampler was used to collect dust in the air on a quartz membrane filter. A minimum of 50 m^3 was

inspired at all sampling sites at a height of 1.5 m above ground. An Andersen low-volume sampler was used to collect dust of various aerodynamic diameters to estimate the respirable portion of dust in Fukushima Prefecture. This sampler was fixed at a sampling site in Fukushima City. Dust samples were weighed, and their radioactivities were measured.

Determination of ^{137}Cs and ^{134}Cs

Aliquots of 100–200 g from each sample of food and cow's milk (dry weight), and soil (fresh weight) were weighed and sealed in cylindrical plastic containers. Filters from aerosol sampling were pressed into small cylindrical plastic containers. Radiometric determinations were performed using a high-purity, low-background, high-resolution germanium detector (0.7 keV). The detector was protected by a lead shield, 10 cm thick internally, covered with 0.5 mm electrolytic copper. A multichannel analyzer (4,096 channels, range 0–3,000 keV, MCA8000; Princeton Gamma Technologies, NJ, USA) was used for gamma-spectrum acquisition and processing. Characteristic gamma-ray energies were monitored to identify and quantify the radionuclides (^{134}Cs 604.7 and 795.9 keV, ^{137}Cs 661.7 keV). The detector was calibrated using a gamma-ray reference source from the Japan Radioisotope Association (Tokyo, Japan). The gamma spectrum of each sample was measured for >20,000 s for food and dust samples and for >2,000 s for soil samples. The lower limits of detection were 0.05 Bq/kg, 0.2 Bq/kg, 0.2 Bq/kg, 0.2 mBq/m^3, and 1 Bq/kg for food, vegetable, milk, dust, and soil samples, respectively. All samples were assumed to be in radioactive equilibrium. All activities were corrected to March 15, 2011 using physical half-lives (^{134}Cs 2.06 years, ^{137}Cs 30.1 years).

Effective dose coefficients for exposures by ingestion and inhalation

Radioactivities were converted into effective doses using effective dose coefficients of 0.019 μSv/Bq for ^{134}Cs and 0.013 μSv/Bq for ^{137}Cs by ingestion, respectively [7]. For inhalation, we assumed that a standard adult resident inhaled 20 m^3 air per day and used the effective dose coefficients of 0.02 μSv/Bq for ^{134}Cs and 0.039 μSv/Bq for ^{137}Cs for inhalation [7]. For the two routes of exposure, we postulated conservatively that all the radionuclides were retained in the body or in the lung, with no elimination.

Results and discussion

A total of 74 sets of whole-day meals were collected and analyzed. Their menus and components are presented in

Table 1 Dietary intake of radioactive cesium in Fukushima Prefecture

Sampling site	*n*		Food volume (g/day)	Water content (%)	Daily intake (Bq/day)		Estimated dose (μSv/year)
					^{134}Cs	^{137}Cs	
Fukushima total	55	*n* > MDL (%)	–	–	36 (65.5)	35 (63.6)	
		Median (range)	2,053 (1,100–3,145)	80.8 (73.3–97.6)	0.2 (ND–7.2)	0.3 (ND–7.0)	3.0 (ND–83.1)
		Mean ± SD	2,178 ± 400	81.9 ± 4.5	0.5 ± 1.1	0.6 ± 1.0	6.4 ± 12.5
Iwaki	10	*n* > MDL (%)	–	–	9 (90.0)	9 (90.0)	
		Median (range)	2,241 (1,879–2,690)	82.1 (76.8–86.1)	0.4 (ND–2.5)	0.7 (ND–1.6)	6.5 (ND–24.7)
		Mean ± SD	2,238 ± 272	81.5 ± 3.3	0.7 ± 0.8	0.7 ± 0.5	8.6 ± 7.8
Souma	10	*n* > MDL (%)	–	–	7 (70.0)	8 (80.0)	
		Median (range)	2,451 (2,044–2,795)	80.5 (73.3–87.1)	0.6 (ND–7.2)	0.9 (ND–7.0)	8.2 (ND–83.1)
		Mean ± SD	2,395 ± 293	80.1 ± 4.2	1.4 ± 2.2	1.6 ± 2.2	17.4 ± 25.3
Nihonmatsu	10	*n* > MDL (%)	–	–	5 (50.0)	4 (40.0)	
		Median (range)	2,611 (1,964–3,145)	79.4 (75.1–82.6)	0.1 (ND–0.9)	ND (ND–0.9)	1.7 (ND–10.4)
		Mean ± SD	2,529 ± 423	78.9 ± 2.3	0.3 ± 0.4	0.2 ± 0.3	2.9 ± 3.6
Fukushima	25	*n* > MDL (%)	–	–	15 (60.0)	14 (56.0)	
		Median (range)	1,954 (1,100–3,051)	83.7 (77.9–97.6)	0.1 (ND–0.8)	0.2 (ND–1.3)	1.3 (ND–11.3)
		Mean ± SD	1,927 ± 308	84.1 ± 4.8	0.2 ± 0.2	0.2 ± 0.3	2.6 ± 3.1
Kyoto (Uji)	19	*n* > MDL (%)	–	–	1 (5.3)	1 (5.3)	–
		Maximum	–	–	0.4	0.5	5.3
		Mean ± SD	2,955 ± 652	87.2 ± 2.5	–	–	–

Estimated dose is the total for doses attributable to exposure to ^{134}Cs and ^{137}Cs. The effective dose coefficients for ^{134}Cs and ^{137}Cs by oral route were 0.019 and 0.013 μSv/Bq, respectively

MDL method detection limit, *ND* less than MDL

Table 2 Radioactive cesium in local commercial products purchased in Fukushima Prefecture

Sampling site	*n*		Weight (g)	Radioactivity (Bq/kg)			Recommended standard[a] (Bq/kg)
				^{134}Cs	^{137}Cs	Total	
Milk							200
Fukushima total	21	$n >$ MDL (%)	–	20 (95.2)	19 (90.5)	–	
		Median (range)	–	1.8 (ND–4.9)	1.9 (ND–5.5)	4.1 (ND–10.1)	
		Mean ± SD	985 ± 119	2.1 ± 1.7	2.4 ± 1.9	4.5 ± 3.6	
Iwaki	3	$n >$ MDL (%)	–	3 (100.0)	3 (100)		
		Median (range)	–	0.9 (0.6–1.2)	1.2 (1.1–1.3)	2.0 (1.9–2.3)	
		Mean ± SD	752 ± 202	0.9 ± 0.3	1.2 ± 1.1	2.1 ± 0.2	
Souma	6	$n >$ MDL (%)	–	6 (100.0)	6 (100.0)	–	
		Median (range)	–	3.1 (1.4–3.8)	3.1 (1.9–4.4)	6.1 (3.3–8.2)	
		Mean ± SD	1,019 ± 29	2.8 ± 1.0	3.1 ± 1.0	5.9 ± 1.9	
Nihonmatsu	3	$n >$ MDL (%)	–	3 (100.0)	1 (33.3)		
		Median (range)	–	0.2 (0.2–1.3)	ND (ND–1.1)	0.2 (0.2–2.4)	
		Mean ± SD	1,047 ± 15	0.5 ± 0.7	0.4 ± 0.6	0.9 ± 1.3	
Fukushima	9	$n >$ MDL (%)	–	8 (88.9)	8 (88.9)	–	
		Median (range)	–	3.4 (ND–4.9)	3.9 (ND–5.5)	7.3 (0.2–10.1)	
		Mean ± SD	1,021 ± 18	2.6 ± 2.0	2.3 ± 4.4	5.6 ± 4.4	
Kyoto (Uji)	3	$n >$ MDL (%)	–	1 (33.3)	1 (33.3)	–	
		Median (range)	–	ND (ND–0.7)	ND (ND–0.7)	ND (ND–1.4)	
		Mean ± SD	1,037 ± 21	0.2 ± 0.4	0.2 ± 0.4	0.5 ± 0.8	

	Weight (g)	Radioactivity (Bq/kg weight)			Recommended standard[a] (Bq/kg)
		^{134}Cs	^{137}Cs	Total	
Vegetable/fruit					500
Kyoto (Uji)					
Spinach	1,249	ND	ND	ND	
Japanese mustard spinach	3,044	ND	ND	ND	
Fukushima ($n = 43$)					
Date					
Japanese mustard spinach	1,828	2.6	2.2	4.8	
Spinach	1,677	0.2	0.3	0.5	
New Zealand spinach	1,097	29.9	32.7	62.6	
Ceylon spinach	826	2.1	3.1	5.2	
Cucumber	1,643	3.4	4.5	7.9	
Welsh onion	1,770	3.3	2.8	6.1	
Kawamata					
Mizuna	504	5.9	7.7	13.7	
Shiitake	1,012	140.4	164.2	304.6	
Ceylon spinach	503	4.4	3.0	7.4	
Cucumber	1,007	1.3	1.6	2.8	
Broccoli	831	6.4	6.6	12.9	
Chinese chives	704	7.2	4.5	11.7	
Partially dried Japanese persimmon	332	1.8	1.7	3.5	
Welsh onion	1,455	5.7	6.6	12.3	
Fukushima					
Chinese chives	436	1.9	2.0	3.9	
Cucumber	493	2.9	3.9	6.8	

Table 2 continued

	Weight (g)	Radioactivity (Bq/kg weight)			Recommended standard[a] (Bq/kg)
		^{134}Cs	^{137}Cs	Total	
Iwaki					
Spinach	1,903	0.5	0.9	1.4	
Snap bean	860	3.5	3.6	7.1	
Shiitake	89	ND	ND	ND	
Green onion	571	7.3	8.5	15.8	
Chinese chives	615	2.8	3.5	6.3	
Broccoli	1,479	0.9	1.1	2.0	
Ceylon spinach	1,079	1.5	2.6	4.0	
Garlic	691	0.8	0.5	1.3	
Souma					
Welsh onion	1,543	4.1	2.6	6.7	
Peach	794	9.3	7.9	17.2	
Cherry	244	29.3	37.3	66.6	
Broad beans	418	4.9	6.0	10.9	
Onion (large)	835	0.5	0.6	1.1	
Onion (small)	430	9.1	9.2	18.3	
Red onion (large)	589	3.3	5.0	8.3	
Red onion (small)	524	9.6	11.6	21.3	
Garlic	256	9.4	7.2	16.6	
Potato	1,258	1.0	0.8	1.8	
Minamisouma					
Carrot	1,271	1.4	2.1	3.5	
Shiitake	417	127.1	154.7	281.8	
Bell pepper	502	ND	ND	ND	
Nihonmatsu					
Asparagus	637	1.3	1.5	2.8	
Bell pepper	390	12.0	10.7	22.7	
Ceylon spinach	1,533	1.7	3.2	4.9	
Cucumber	2,064	3.6	4.3	7.9	
Welsh onion	1,309	5.4	5.0	10.5	
Cherry	352	24.5	28.5	52.9	

MDL method detection limit, *ND* less than MDL

[a] Recommended by Ministry of Health, Labor, and Welfare of Japan [8]

Table S1. Radioactivity per daily intake (Bq/day) is also summarized in Table 1. ^{134}Cs or ^{137}Cs was detected in 36 of 55 whole-day meal samples from Fukushima Prefecture, compared with only one of 19 from Kyoto. The estimated median dose levels was 3.0 μSv/year, ranging from not detectable (ND) to 83.1 μSv/year in Fukushima, while the maximum dose level in Kyoto was 5.3 μSv/year.

The levels of ^{134}Cs and ^{137}Cs in cow's milk and vegetables were also determined (Table 2). The median total activity in milk from Fukushima Prefecture was 4.1 Bq/kg, ranging from ND to 10.1, which was an order of magnitude lower than the recommended limit set by the Ministry of Health, Labor, and Welfare of Japan [8]. Trace radioactivity was detected in only one sample from Kyoto. No vegetables in Fukushima Prefecture exceeded 100 Bq/kg, except for shiitake mushrooms (*Lentinula edodes*), which contained relatively high levels of radioactivity, up to 60% of the recommended limit (Table 2). Radioactivities in shiitake at Kawamata or Minamisouma were larger than at Iwaki, indicating that a radioactive plume was transferred by northeasterly winds from the nuclear plant. No radioactivity was detected in vegetables from Kyoto. These results indicate that the levels of radioactive Cs ingested were well below the recommended limits [8] in various towns in Fukushima Prefecture, except in the case of shiitake.

Table 3 Particle size distribution and respiratory deposition estimate for radioactive cesium in Fukushima Prefecture

Sampling site		Date (2011)	Andersen low-volume sampler, 224 m^3			
			Fraction (μm)	Dust amount (mg)	Radioactivity (mBq/m^3-air)	
					^{134}Cs	^{137}Cs
Fukushima	37°45′42″N 140°28′18″E	7/2–7/8	100–11.4	0.7	0.4	0.3
			11.4–7.4	1.1	0.3	0.3
			7.4–4.9	1	1.0	0.4
			4.9–3.3	0.9	0.5	0.6
			3.3–2.2	0.6	0.3	0.2
			2.2–1.1	0.8	0.3	0.2
			1.1–0.7	1.3	0.8	0.4
			0.7–0.46	1.3	1.5	1.1
			<0.46	0.9	1.5	1.3
		Total		8.6	6.5	4.7
		Respirable	<4.9	5.8	4.8	3.8

Sampling site		Date (2011) (weather)	High-volume air sampler							Ambient dose rate	Radioactivity in soil (Bq/kg)		
			Air volume sampled (m^3)	Dust amount (mg)	Radioactivity in air (mBq/m^3-air)		Estimated dose[a] (μSv/year)						
					^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs	Total	(μSv/h)	^{134}Cs	^{137}Cs	*n*
Fukushima	37°45′42″N 140°28′18″E	2011/7/2 (F)	473	6.8	1.9	3.0	0.3	0.8	1.1	1.2	NA	NA	
Date	37°47′10″N 140°33′26″E	2011/7/3 (CL)	94	3.5	7.9	6.4	1.1	1.8	3.0	0.9	3,232 ± 2,666	3,855 ± 3,047	5
Fukushima	37°39′26″N 140°32′11″E	2011/7/3 (CL)	83	1.9	4.7	1.5	0.7	0.4	1.1	1.0	2,515 ± 859	3,059 ± 1,077	5
Fukushima	37°45′42″N 140°28′18″E	2011/7/4 (R)	450	8	1.6	1.5	0.2	0.4	0.6	1.2	NA	NA	
Souma	37°46′1″N 140°57′2″E	2011/7/5 (F)	88	0.7	0.6	0.2	0.1	0.1	0.1	0.5	1,710 ± 2,365	2,116 ± 2,976	5
Minami-Souma	37°38′29″N 140°55′30″E	2011/7/5 (F)	84	2.4	0.7	1.1	0.1	0.3	0.4	0.9	1,772 ± 411	2,151 ± 546	5
Souma	37°46′8″N 140°43′1″E	2011/7/5 (F)	84	1.3	1.1	2.3	0.2	0.7	0.8	1.6	1,723 ± 1,792	2,047 ± 2,174	5
Fukushima	37°45′42″N 140°28′18″E	2011/7/5 (F)	220	4	2.9	3.4	0.4	1.0	1.4	1.2	NA	NA	
Nihonmatsu	37°33′21″N 140°27′34″E	2011/7/6 (F)	93	0.1	0.6	0.6	0.1	0.2	0.3	1.2	12,184 ± 12,170	14,202 ± 14,025	5
Nihonmatsu	37°33′21″N 140°30′43″E	2011/7/6 (F)	53	0.3	4.2	7.3	0.6	2.1	2.7	1.9	1,895 ± 674	2,244 ± 755	5
Kawamata	37°36′14″N 140°38′49″E	2011/7/6 (CL)	72	0.4	6.3	6.1	0.9	1.7	2.7	2.0	3,931 ± 4,856	4,741 ± 5,929	5
Fukushima	37°45′42″N 140°28′18″E	2011/7/6 (CL)	246	4	5.3	7.6	0.8	2.2	2.9	1.2	NA	NA	
Fukushima	37°45′42″N 140°28′18″E	2011/7/7 (CL)	259	5.3	1.9	2.5	0.3	0.7	1.0	1.2	NA	NA	
Iitate	37°36′44″N 140°44′52″E	2011/7/7 (CL)	84	1.7	24.6	38.9	3.6	11.1	14.7	9.0	18,531 ± 11,235	23,185 ± 15,664	5
Namie	37°33′38″N 140°45′39″E	2011/7/7 (CL)	84	1.7	148.2	194.2	21.6	55.3	76.9	13.0	13,548 ± 10,469	16,216 ± 12,653	5
Katsurao	37°31′33″N 140°48′21″E	2011/7/7 (CL)	84	1.5	65.0	64.0	9.5	18.2	27.7	10.0	16,332 ± 11,170	16,799 ± 10,058	5

CL cloudy, *F* fine, *R* rainy, *NA* not available

[a] It was assumed that radioactive cesium was in respirable fraction and that a standard human inhales 20 m^3 air

We collected 16 dust samples using the high-volume sampler (Table 3; Fig. 1). Data obtained with the low-flow-volume sampler suggested that a large proportion of the radionuclides from the crippled Fukushima nuclear power plant was in the respirable fraction: 74% (4.8/6.5) of the total ^{134}Cs and 81% (3.8/4.7) of the total ^{137}Cs (Table 3). To estimate the exposure doses for humans, we therefore selected a conservative scenario whereby all ^{134}Cs and ^{137}Cs activities in the dust samples collected using the high-volume sampler were allocated to the respirable fraction (aerodynamic diameter <4.9 μm). The highest dose level of 76.9 μSv/year was recorded in a sample collected at Namie. However, this value was still less than one-tenth of the permissible dose level of 1 mSv/year [8]. The estimated dose levels for ^{137}Cs were significantly correlated with ambient dose rate (μSv/h) ($n = 10$, $r^2 = 0.79$, $p < 0.05$) but not with mean radioactivity levels in soil (Bq/kg) ($n = 11$, $r^2 = 0.32$, $p > 0.05$).

Given that the samples in this study were obtained in early July, about 4 months after the major release of radioactivity, airborne radioactivity was likely to represent resuspended deposited radioactivity, rather than direct transport from the source. Several studies have investigated resuspension from a flat surface [5], but information on resuspension from ecological systems including forests and paddy fields is scant.

We demonstrated the radioactivity levels due to ^{134}Cs and ^{137}Cs in Fukushima Prefecture in July 2011. The maximum total exposure dose through inhalation and ingestion was estimated to be 160 μSv/year (83.1 by ingestion and 76.9 by inhalation) in zones outside a 20-km radius of the crippled Fukushima nuclear power plant.

The amounts of radioactivity in the daily meals consumed by residents of the study regions were well below the regulation limit. However, many food items are now imported globally, such that a high portion of foodstuffs comes from uncontaminated areas. It is possible that the radioactivity in some highly contaminated foodstuffs may be diluted by other "clean" foods. However, the ingested doses estimated in the present study would underestimate the exposure of residents whose daily foods are mostly supplied locally from within the contaminated areas. The conclusions of this study may therefore not be applicable to people in such a situation. Furthermore, the current study only utilized air monitoring in a few, geographically limited areas. All meal samples were obtained from outside a 30-km radius of the nuclear power plant, because no commercial venders were present between 20 and 30 km from the power plant, which had been defined as the planned emergency evacuation zone. In addition to the small number of air samples collected, the survey was conducted in the rainy season when "resuspension" is relatively low. The current study is thus subject to the above limitations and biases. However, the conservative approach adopted in this study maximized the estimated dose levels and would thus partially mitigate the effects of any biases and limitations. In conclusion, the estimated dose levels in residents of Fukushima Prefecture as a result of ingestion and inhalation were much lower than the 1 mSv/year, recognized as a publicly permissible dose [8]. Further studies are needed to perform qualitative risk assessments based on more accurate exposure estimates.

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Conflicts of interest The authors declare that there are no conflicts of interest.

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INTERNATIONAL FORUM

Asian forum on environmental health policy: challenges and perspectives of environmental health problems in the region in the next 30 years

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Introduction

Asia is the world's most populous continent, accomodating circa 3.9 billion people in 48 countries. It is an important player in terms of the environment and world economy, with the second largest gross domestic product of all continents, after Europe. This geographical entity includes many developed countries, such as Japan, South Korea, and Singapore, and a number of very high growth nations, such as China and India. Its climate is very diverse, ranging from the very moist (southeastern parts) to dry (interior), and very cold (e.g., Siberia) to tropical (southern regions). The countries of this continent not only share—to varying degrees—historical experiences but also the effects of environmental pollution, such as Asian dust storms (ADS). The recent Fukushima Daiichi nuclear disaster in Japan has delivered the clear message that although an environmental challenge may be initiated in one specific location, many countries in the region may suffer from the consequences. Global climate change also underlines the fact that environmental issues are often global and, consequently, multinational collaboration is not an option but a necessity. Environmental health problems cannot be properly resolved if the counter-measures are restricted only to within the political borders. Accurate assessment of the problems and the development of appropriate solutions can best be achieved on a broader, regional scale, by communication and collaboration among those scientists and policy-makers facing the same issues.

The past of one country can mirror the present or future situation of another country. Japan, one of the most developed countries in the world, and South Korea, now considered to be a developed country, have experienced very fast economic growth, which has been accompanied by worsening health of its population due to environmental pollution. Ageing has for some time been an important issue in Japanese society and is now also a challenging problem in South Korea. China is the largest economy in Asia and the second largest economy in the world. However, it is now recognizing the adverse consequences of rapid industrialization and development in terms of environmental health. The experiences of South Korea or Japan could help China better prepare for the potential challenges to be faced in terms of environmental health.

Such recognition has led to the organization of a long-term platform for communication and collaboration among environmental health experts representing the diverse geographical areas of Asia. The first International Forum for Environmental Health Policy and Science was held in

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Seoul in November 25, 2011, and delegates from leading academic institutes in Bangkok, Kyoto, Seoul, Taipei, and Wuhan participated. The editorial presents a summary of the motivation for holding this forum, the perspectives of each participant, and future directions that were agreed on, with the intention to encourage other scientists around the region to join in and contribute.

Issues and agendas by delegation

Japan

First of all, we deeply express our sincere thanks for the warm support we have received from Asian countries that may have been directly or indirectly affected by the consequences of the East Japan Earthquake. We also have to admit that serious environmental contamination did take place due to the escape of radionuclides from the crippled Fukushima Daiichi nuclear power plant. International environmental monitoring for radionuclides will be needed for the next 30 years. In addition, we have urged the Japanese government to make information on the crippled plant accessible to the international community.

In the current, rather complex global circumstances, policy-making by any one nation on trans-boundary environmental problems cannot be independent of national interests in making profits because the latter are closely linked to the national economy. However, it is obvious that too much emphasis on the national economy when challenged with trans-boundary environmental problems will only increase conflicts among neighboring countries. Thus, we need to find a way to harmonize policy-making among countries and should establish clear and simple mechanisms to delineate the responsibility of stakeholder countries. One such a way is to visualize the trans-boundary flows of environmental contaminants among Asian countries. Such visualization will increase the transparency to the general populations of Asian countries, facilitating decision-making on the basis of national consensus. The Asian platform is expected to function as a task force for the visualization.

Korea

Demands for a cleaner and healthier environment have become very high in South Korea after several decades of rapid industrialization and development. These demands reflect the increased awareness among the general public of the potential deleterious health effects from exposure to environmental contamination. While serious environmental pollution events due to industrial incidents or by accidental release have since the mid-1990s no longer been frequently occurred, recent experiences, such as the Hebei-Spirit oil spill of 2007, still demonstrate the potential for such events to occur at any time. Therefore, the importance of emergency preparedness cannot be emphasized enough within the framework of environmental health management. In addition to such accidental episodes, the most important health issues related to environmental problems include the consequences of long-term low-dose exposures to multiple environmental contaminants, and environmental health inequity among susceptible populations, such as children and the elderly. Emerging environmental issues associated with global climate change and new technologies (e.g., nanomaterials) also deserve special attention. The Center for Disease Control of South Korea recently reported that the epidemic of acute interstitial pneumonia and the several resulting casualties were likely the result of exposure to a number of disinfectants used in humidifiers. This incidence increased public skepticism on the safety of chemicals that are used in normal daily activities. However, it also provides a chance to critically review the systems for ensuring chemical safety, not only before the release of such chemicals onto the market, but also during the marketing period.

China

China is the world's fastest growing major economy, with annual growth rates of approximately 10% for almost three decades. Since the 1980s China has witnessed increased pollution and degradation of natural resources and now recognizes increasing public health problems due to such pollution. Chronic, non-communicable diseases account for about 80% of deaths and 70% of disability-adjusted life-years lost in China, which is in part related to changes in the environment, lifestyle and diet, as well as an increased life expectancy. Air pollution has emerged as the most important environmental cause of cardiopulmonary diseases and adverse health risks. Lifestyle and diet changes, environmental pollution, and their interaction with genetics or epigenetic factors are involved in endemic, chronic non-communicable diseases. One of the major scientific challenges for the next few decades will be to gain an understanding of the interaction between genetic susceptibility and environmental factors on the etiology of not only cardiopulmonary diseases, such as heart disease, asthma, lung cancer, and chronic obstructive pulmonary diseases, but also on early damage, such as genetic damage, decreased lung function, and heart rate variability. Environment-wide and genome-wide association studies on chronic non-communicable diseases can best be carried out by collaborations and through the exchange of ideas within the region that shares common environmental problems.

Therefore, we are looking forward to future cooperation in Asia.

Taiwan

Asian dust storms occur in the winter and spring, especially from March to May, and mainly originate in the Gobi and Takla Makan deserts of Mongolia and western China. They can move eastward to China, Japan, South Korea, Taiwan, and sometimes to northern Pacific Ocean areas. Particulate matter (PM), especially PM with aerodynamic diameters of <10 μm (PM_{10}), have been recorded at concentrations of >500 μg/m^3, occasionally even exceeding 1,000 μg/m^3, during several ADS in many cities located downwind of these deserts, including Shanghai, Hong Kong, and Taipei. Overall findings of epidemiological studies show that long-range transported Asian dust can increase mortality among residents in downwind areas. Dust derived from mineral soil in deserts and air pollutants from biomass burning are major contributors of long-range transported air pollution across many countries around the world. The negative effects of trans-boundary air pollution on public health are emerging as an important global health issue which needs to be further researched by academics and governments. Global efforts, including alleviating desertification speed in dust-originating areas and reducing industrial emissions along the dust-transporting paths, must be made to tackle the root-causes of trans-boundary air pollution in order to protect the global environment and public health. Collaboration among Asian public health researchers to tackle this trans-boundary pollution problem is recommended as a major step towards protecting public health and environmental quality in this fast growing continent.

Thailand

Thailand is a newly industrialized country and one of the fastest growing economies, ranked 24th on the global market. Thailand is now experiencing environmental issues as a downside of economic growth, which include deforestation and air pollution, among many others. Assessment of health damage due to industrialization and associated remediation approaches are important in Thailand. Climate change, which may affect human health through a range of mechanisms, including the relatively direct risks of floods and storms and the more complex pathways of altered patterns of infectious disease outbreak, is a growing concern of Thailand, and one that might be appropriately addressed by collaborative efforts within Asian countries. Scientific research on common environmental issues, such as trans-boundary air pollution and climate change, is essential given the nature of the problems. It is also important to encourage community empowerment by, for example, organizing practical workshops aimed at distributing research findings to the community or by initiating local-based research activities in accordance with their most critical environmental problems.

Consensus and future directions

We have reached a consensus on the long- and short-term objectives of the forum. As the long-term objective, we should collaborate more closely on Asian environmental health problems at several levels—individual scientist level, academic society level, and governmental level. We believe that collaborations among academic societies can provide the greatest impulse. In pursuing the short-term goals, we agreed it would be important to develop or foster a high-quality scientific journal in environmental health. *Environmental Health and Preventive Medicine* (EHPM) is obviously one of the most potential candidate journals for this role.

To identify priority environmental health issues in the region and to develop a collaborative research network to resolve the challenges, we agreed to organize a steering committee for the Asian Environmental Health Forum, the members of which will be chosen from among the delegates from Asian countries. The committee will support policy-makers in each country by providing knowledge and sharing experiences from other countries. It will also play a pivotal role in identifying those areas warranting the attention of policy-makers and provide channels of communication among policy-makers of countries that share or have shared similar challenges.

To form a steering committee for the forum and to identify environmental health issues in the region that should be given priority in terms of policy, the Korean Society of Environmental Health will take the initiative to organize a series of annual workshops in collaboration with Ministry of Environment of Korea. We hope that the initiative will promote academic communication and identify ideas for synergistic collaborations on environmental health issues during the next 30 years.

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Highlighted Article

Stable isotope ratios and mercury levels in red meat products from baleen whales sold in Japanese markets

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ABSTRACT

We analyzed the $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ values and Hg concentration in red meat products originating from the predominant types sold in Japan for human consumption: two populations of common minke (J- and O-types), Bryde's and sei whales in the western North Pacific Ocean, and fin and Antarctic minke whales in the Southern Ocean. The order of the trophic positions, evaluated by $\delta^{15}N$ values and Hg concentrations, coincided with their known feeding habits: common minke (J-type)=common minke (O-type) > Bryde's ≥ sei ≥ Antarctic minke ≥ fin. The Hg concentrations in the combined samples from the six samples were significantly correlated with their $\delta^{15}N$ values ($\gamma=0.455$, $n=66$, $p<0.05$), reflecting overall differences in the trophic level. This correlation was not significant for within-species comparison for the common minke (J- and O-types) or the Bryde's whale, probably reflecting the higher $\delta^{15}N$ value and lower Hg concentration in the North Pacific Ocean around Japan. Determination of $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ could be used to discriminate between the red meat products originating from the whale species in the North Pacific and Southern Oceans. However, the four whale species or populations in the Pacific Ocean could not be discriminated on basis of these values, nor could the two species in the Southern Ocean. Positive correlations between the $\delta^{13}C$ and $\delta^{15}N$ values and negative correlations between the $\delta^{15}N$ and $\delta^{18}O$ values and the $\delta^{13}C$ and $\delta^{18}O$ values, probably reflecting migration patterns, were found in some whale species in the North Pacific and Southern Oceans.

1. Introduction

Products from whales, dolphins and porpoises (Suborder Cetacea) are sold in Japan for human consumption. Currently, most whale products for human consumption are supplied from the scientific whaling of baleen whales, small-type coastal whaling of toothed whales, and the drive and hand-harpoon fishing of small whales, dolphins and porpoises as well as incidental catch by set nets (Endo et al., 2003). Red meat (muscle) products are the most popular whale products sold in Japan, and most Japanese consumers prefer the red meat originating from mysticetes (baleen whales) to that from odontocetes (toothed whales, dolphins and porpoises). Most red meats from mysticetes sold in Japan originate from the Antarctic minke whale (*Balaenoptera bonaerensis*) and fin whale (*Balaenoptera physalus*) taken in the Southern Ocean and the common minke whale (*Balaenoptera acutorostrata*), Bryde's whale (*Balaenoptera edeni*) and sei whale (*Balaenoptera borealis*) taken in the western North Pacific Ocean. Common minke whales can be categorized into at least two types: the "O type", found primarily in the offshore Pacific Ocean, and the "J type", found primarily in the Sea of Japan and nearshore waters along Japan's Pacific coast (Wade et al., 2010). O-type minke whales are the primary target of Japanese scientific whaling in both coastal and offshore waters of the Pacific, while J-type minke whales are primarily taken as bycatch in coastal set nets around the entire Japanese coastline. Although most baleen whales are assumed to migrate annually between feeding habitat in high latitudes and breeding habitat in low latitudes, the pattern of migration is poorly known for some of the species sampled here.

As odontocetes are long-lived and occupy the top levels of the marine food web, feeding mainly on fish and squid, they biomagnify marine pollutants such as heavy metals and organochlorine compounds (Haraguchi et al., 2000). Among these pollutants,

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contamination with mercury (Hg) is prominent (Endo et al., 2003, 2004, 2005). The contamination levels of pollutants in mysticetes are lower than those in odontocetes, reflecting their preference for plankton and small fish species (i.e., their lower trophic positions). Among the baleen whale species sold in Japan, common minke whales are opportunistic and omnivorous feeders that change their prey temporally and regionally. Compared with common minke whales, Bryde's and sei whales are only moderately omnivorous feeders (Mitani and Bando, 2008; Yasunaga and Fujise, 2009a, b), and Antarctic minke and fin whales are generally zooplankton feeders. In our previous survey of Hg levels (Endo et al., 2003), only one of the 62 red meat products originating from mysticetes showed a Hg concentration exceeding the Japanese permitted level for fish and shellfish (0.4 μg/wet g), whereas all red meat products originating from odontocetes (n=137) exceeded the permitted level.

Stable isotope analysis has been used as a tool to obtain information on the feeding ecology of marine species. The $\delta^{15}N$ value shows a stepwise increase in the trophic level of a food chain (Kelly, 2000), and a positive correlation between the $\delta^{15}N$ value and the Hg concentration in biota has been reported (Yoshinaga et al., 1992; Kidd et al., 1995). On the other hand, the $\delta^{13}C$ value is used to indicate the relative contribution to the diet of potential primary sources, and can demonstrate differences between species taking coastal and offshore prey or between those taking pelagic and benthic prey (Kelly, 2000). A significant increase in $\delta^{15}N$ of 3.4 ± 1.1‰ has been shown to occur between consumer and prey (Minagawa and Wada, 1984), whereas only a small enrichment of about 1‰ is found in the $\delta^{13}C$ value (DeNiro and Epstein, 1981). We recently reported that the $\delta^{15}N$ and $\delta^{13}C$ values in odontocetes caught off or stranded on the coast of northern Japan were higher and lower, respectively, than those in whales in the southern area, probably reflecting the variations in marine environment around Japan (Endo et al., 2010). Mitani et al. (2006) analyzed the $\delta^{13}C$ and $\delta^{15}N$ values in the baleen plates of common minke whales caught during scientific research whaling, and tried to elucidate the migration pattern in relation to dietary shift. However, little information is available about the $\delta^{13}C$ and $\delta^{15}N$ values in the muscle of baleen whales, including common minke, Bryde's and sei whales, caught in the western North Pacific Ocean and Antarctic minke and fin whales caught in the Southern Ocean.

Recently, the $\delta^{18}O$ value, in addition to the $\delta^{13}C$ and $\delta^{15}N$ values, has been used to discriminate, verify and identify the habitat of plants and animals, as the $\delta^{18}O$ value reflects the water environment, temperature and humidity of their habitats. For instance, the $\delta^{18}O$ values in beef oil (Heaton et al., 2008), underground water (Mizota and Kusakabe, 1994) and cultured rice (Suzuki et al., 2009) all tend to decrease with latitude (temperature). To our knowledge, however, the $\delta^{18}O$ values in cetacean species have not yet been reported. According to the above latitude-dependent changes, we speculated that the $\delta^{18}O$ value would be lower in cetaceans caught off the northern areas than off the southern areas of Japan, and that $\delta^{13}C$ and $\delta^{15}N$ values would be negatively correlated with the $\delta^{18}O$ value in the whale products sold in Japan.

The purpose of the present study was to analyze the $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ and the Hg concentration in red meat products originating from the common minke (J- and O- types), Bryde's and sei whales caught in the western North Pacific Ocean and Antarctic minke and fin whales caught in the Southern Ocean. We discuss the correlations between the trophic level, as evaluated by $\delta^{15}N$ value, and the Hg contamination and among the $\delta^{15}N$, $\delta^{13}C$ and $\delta^{18}O$ values, and the possibility of verifying the species origins of red meat products sold in Japan using these stable isotope ratios.

2. Materials and methods

2.1. Sampling of red meat products and genetic analysis for species origin

Red meat products originating from common minke whale (J- and O-types), Bryde's and sei whales caught in the Northwest Pacific Ocean and Antarctic minke and fin whales caught in the Antarctic Ocean were purchased from retail outlets in Japan between 2000 and 2006, as described previously (Endo et al., 2003, 2005). Samples were stored at −20 °C until analysis.

As reported elsewhere (Baker et al., 1996, 2006), the species origin of cetacean products was identified by mitochondrial DNA sequences (control region and cytochrome b) amplified from the products via PCR. The population origin of the common minke whale products (i.e., J- or O-type) was inferred from sequence variation in the mtDNA control region, as described in Baker et al. (2000).

2.2. Chemical analyses

The total mercury (Hg) concentration in the red meat products was determined by a Mercury Analyzer SP-2 (Nippon Instruments Corporation, Tokyo, Japan), as reported previously (Endo et al., 2007). DOLT-2 (National Research Council of Canada) was used as an analytical quality control for Hg. The recovery of Hg was $94 \pm 3\%$ (n=5). The Hg concentration in the red meat products was expressed on a wet weight basis.

Dried subsamples of red meat products were analyzed for stable isotopes (^{13}C, ^{15}N and ^{18}O) after the removal of lipids by chloroform/methanol extraction (Logan and Lutcavage, 2008). The $\delta^{13}C$ and $\delta^{15}N$ analyses were performed using a mass spectrometer (Delta S, Finnigan MAT, Bremen, Germany) coupled with an elemental analyzer (EA1108, Fisons, Rodano, Milan, Italy) held in the Center for Ecological Research (CER), Kyoto University (Kyoto, Japan), as reported previously (Endo et al., 2009, 2010). The $\delta^{18}O$ analysis was performed using a mass spectrometer (Delta V PLUS, Thermo Fisher Scientific, Tokyo, Japan) coupled with an elemental analyzer (TC/EA, Thermo Fisher Scientific, Tokyo, Japan) held in the SI Science Co. Ltd. (Saitama, Japan). The natural abundances of ^{13}C, ^{15}N and ^{18}O are expressed as per mil (‰) deviation from the standards as defined by the following equation:

$$\delta^{13}C, \delta^{15}N \text{ or } \delta^{18}O = (R_{sample}/R_{standard} - 1) \times 1000(‰),$$

where $R = {}^{13}C/{}^{12}C$, ${}^{15}N/{}^{14}N$ or ${}^{18}O/{}^{16}O$. CERKU-1, 2 and 5, certified by CER, were used as $\delta^{13}C$ and $\delta^{15}N$ reference materials (Tayasu et al., 2011), and benzoic acid (A and B), certified by Indiana University (IN, USA), was used as the $\delta^{18}O$ reference material.

2.3. Statistical analyses

The data are shown as mean ± S.D., and were analyzed by Turkey–Kramer multiple comparison test and Pearson's correlation coefficient test, using the Statcell program. The level of significance was set at $p < 0.05$.

3. Results and discussion

The stable isotope ratios of $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ and the Hg concentration in sixty-six red meat product samples from baleen whale species and populations were analyzed (Table 1), and the analytical results are summarized in Table 2. Fig. 1 shows the relationship between the $\delta^{15}N$ value and the Hg concentration in the combined products from the six samples (n=66), and Fig. 2 shows the relationships among the $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ values. Table 3 shows a summary of relationships among the Hg concentration, $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ values for each baleen whale species or population.

In agreement with previously published results (Endo et al., 2003), the contamination levels of Hg in the red meat products were in the following order: common minke whales (J-type)= common minke whales (O-type) > Bryde's whale=sei whale=fin whale ≥ Antarctic minke whale (Table 2). A similar order was found in the $\delta^{15}N$ values for these species (Table 2). The Hg concentrations in the combined products of the six samples were significantly correlated with their $\delta^{15}N$ values (Fig. 1, r=0.455, n=66, $p < 0.05$). As data not shown in Figure, significant correlations were found between the $\delta^{15}N$ values and the Hg concentrations in the combined samples from the North Pacific Ocean (r=0.418, n=46, $p < 0.05$) and from the Southern Ocean (r=0.541, n=20, $p < 0.05$). These correlations between the Hg

Table 1
Analytical results of mercury and stable isotope ratios in red meat products originating from baleen whales sold in Japanese markets.

Species origin	Sample code	Hg (μg/wet g)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	$\delta^{18}O$ (‰)
Common minke whale, J-type	1	0.050	−18.4	11.5	11.9
	2	0.125	−19.3	11.6	10.6
	3	0.074	−17.2	15.8	9.7
	4	0.070	−19.1	11.4	11.6
	5	0.136	−18.0	15.0	10.5
	6	0.239	−17.5	12.6	11.9
	7	0.180	−18.6	10.3	13.3
	8	0.119	−18.7	11.0	12.8
	9	0.031	−19.1	12.1	12.2
	10	0.027	−18.6	12.7	11.8
	11	0.041	−18.3	11.5	12.9
	12	0.061	−17.5	9.6	13.9
	13	0.029	−19.0	11.2	13.4
Common minke whale, O-type	1	0.053	−19.0	12.1	13.0
	2	0.053	−18.3	12.1	13.9
	3	0.056	−18.5	11.6	13.8
	4	0.044	−19.2	11.5	13.1
	5	0.121	−18.0	11.5	13.6
	6	0.053	−20.3	11.4	12.0
	7	0.160	−17.9	12.0	13.8
	8	0.014	−19.1	9.7	13.7
	9	0.174	−17.6	11.1	12.3
	10	0.176	−17.6	12.0	12.2
	11	0.254	−18.9	10.2	12.9
	12	0.027	−18.9	10.9	12.2
Bryde's whale	1	0.037	−17.2	8.6	15.5
	2	0.090	−17.5	9.3	15.5
	3	0.063	−15.9	11.9	13.7
	4	0.053	−17.6	10.1	16.0
	5	0.070	−16.3	11.6	13.9
	6	0.027	−17.2	9.7	16.7
	7	0.055	−15.9	11.2	14.0
	8	0.056	−16.9	9.5	16.1
	9	0.045	−17.2	9.5	15.0
	10	0.055	−17.1	9.8	14.6
	11	0.067	−16.9	8.4	14.9
Sei whale	1	0.026	−23.1	6.3	15.3
	2	0.082	−18.3	9.6	15.8
	3	0.079	−19.1	8.5	13.7
	4	0.028	−21.7	7.1	16.5
	5	0.045	−19.4	7.6	15.6
	6	0.090	−18.9	8.7	15.4
	7	0.046	−18.7	7.6	14.6
	8	0.054	−18.6	10.3	13.1
	9	0.033	−18.1	8.1	14.9
	10	0.061	−18.7	9.5	15.0
Fin whale	1	0.047	−23.0	5.7	15.2
	2	0.050	−21.2	6.1	15.6
	3	0.026	−23.9	6.0	16.5
	4	0.042	−20.8	6.2	15.4
	5	0.031	−23.0	5.7	15.4
	6	0.052	−22.8	5.9	15.6
	7	0.041	−21.9	5.6	15.3
	8	0.090	−21.9	6.3	13.5
	9	0.026	−23.6	5.4	14.4
	10	0.031	−23.1	4.9	14.4
Antarctic minke whale	1	0.027	−24.3	6.1	14.9
	2	0.051	−24.8	6.0	14.2
	3	0.013	−24.2	5.7	13.8
	4	0.013	−25.1	5.9	14.9
	5	0.077	−24.7	6.0	15.2
	6	0.018	−23.9	6.1	13.2
	7	0.014	−24.7	6.3	14.5
	8	0.027	−25.1	6.4	15.1
	9	0.013	−24.7	6.7	15.5
	10	0.014	−24.7	7.2	14.3

level and the trophic level, as evaluated by $\delta^{15}N$ value, were firstly reported in the food products from Papuan New Guinea (Yoshinaga et al., 1992) and from the freshwater biota in Ontario, Canada (Kidd et al., 1995). Although there was an overall correlation in the combined sample from the six whale species or populations, only the sei whale had a significant within-species correlation between the Hg concentration and the $\delta^{15}N$ value ($\gamma=0.651$, $n=10$, $p<0.05$) (Table 3). This probably reflects the marine environment around Japan (higher $\delta^{15}N$ value and lower Hg concentration in the northern area of Japan; Endo et al., 2010). The fin whale in the Antarctic Ocean had high but non-significant correlation between the Hg concentration and the $\delta^{15}N$ value ($\gamma=0.618$, $n=10$, $p>0.05$), while the Antarctic minke whale had negative correlation. The reason for this negative correlation remains unknown.

According to latest reports (Mitani et al., 2006; Yasunaga and Fujise, 2009a, b), O-type common minke whales may be categorized into coastal and offshore whales. The Hg concentration is lower in the coastal whales (about 0.22 ± 0.07 μg/wet g) than in the offshore whales (about 0.3 μg/wet g) as the coastal whales feed on zooplankton, saury and anchovies (the Hg concentrations in these species were below 0.05 μg/wet g) while the offshore whales feed on these three species as well as on pomfret (0.232 ± 0.027 μg/wet g) (Yasunaga and Fujise, 2009a, b). The present Hg value in the O-type whales (0.099 ± 0.076 μg/wet g) is closer to the Hg value in the coastal whales than to that in the offshore O-type whales. The determination of $\delta^{13}C$ and $\delta^{15}N$ may also allow for the discrimination between the coastal and offshore species (Kelly, 2000). However, there has not yet been any comparison of these values between the coastal and offshore populations of common minke whales. We previously analyzed the Hg levels in cetacean products sold in South Korean markets (Endo et al., 2007), and the Hg concentration in the common minke whale (most of the whales were speculated to be J-type from coastal waters) was 0.22 ± 0.11 μg/wet g (0.03–0.43 μg/wet g, $n=30$), which is higher than the present data for the J-type whale (0.091 ± 0.065 μg/wet g). The difference in Hg concentrations in the common minke whale between the previous and present studies may be due to differences in their diet and habitat.

In the present study (Table 2), no differences were found in the results for Hg concentration, or $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ values between the J- and O-types of common minke whales. Compared with the J- and O-types of common minke whales (0.091 ± 0.065 and 0.099 ± 0.076 μg/wet g, respectively), Bryde's and sei whales are only moderately omnivorous feeders and their Hg concentrations were lower (0.056 ± 0.017 and 0.054 ± 0.023 μg/wet g, respectively); Yasunaga and Fujise (2009a) reported similar Hg concentrations in the muscle of Bryde's whales (0.046 ± 0.008 μg/wet g) and sei whales (0.052 ± 0.009 μg/wet g) caught in the western North Pacific Ocean. The Antarctic minke and fin whales in the Southern Ocean are plankton feeders and their Hg levels were slightly lower than those of Bryde's and sei whales in the western North Pacific Ocean (Table 2).

Gendron et al. (2001) analyzed the $\delta^{13}C$ and $\delta^{15}N$ values in skin samples from Bryde's, fin and blue (*Balaenoptera musculus*, a plankton-feeder) whales in the Gulf of California, Mexico. The mean values of $\delta^{13}C$ and $\delta^{15}N$ in the skin samples of the Bryde's, fin and blue whales were −18.1 and 15.8‰ ($n=2$), −16.0 and 15.4‰ ($n=2$), and −18.2 and 12.9‰ ($n=2$), respectively. This order of $\delta^{15}N$ values is consistent with our knowledge of the feeding habits of those whale species, although the $\delta^{15}N$ values in the Bryde's and fin whales are higher than those in the present study (10.0 ± 1.2‰ and 5.8 ± 0.4‰, respectively, Table 2). The variation in $\delta^{15}N$ at the base of the food web is considered to be an important factor in the $\delta^{15}N$ values observed in the upper trophic levels. The $\delta^{15}N$ value in euphausiids (krill) along the west coast of the Gulf of California was 11.0 ± 1.2‰ (Gendron et al., 2001),

Table 2
Summary of analytical results for mercury and stable isotope ratios in red meat products originating from baleen whales sold in Japanese markets.

	Hg (μg/wet g)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)	$\delta^{18}O$ (‰)
Common minke whale (J-type), $n=13$	0.091 ± 0.065^{a}	-18.4 ± 0.7^{a}	12.0 ± 1.7^{a}	12.0 ± 1.2^{a}
Common minke whale (O-type), $n=12$	0.099 ± 0.076^{a}	-18.6 ± 0.8^{a}	11.4 ± 0.7^{a}	13.0 ± 0.7^{a}
Bryde's whale, $n=11$	0.056 ± 0.017^{ab}	-16.9 ± 0.6^{b}	10.0 ± 1.2^{b}	15.1 ± 1.0^{b}
Sei whale, $n=10$	0.054 ± 0.023^{ab}	-19.5 ± 1.6^{c}	8.3 ± 1.3^{c}	15.0 ± 1.0^{b}
Fin whale, $n=10$	0.044 ± 0.019^{ab}	-22.5 ± 1.0^{d}	5.8 ± 0.4^{d}	15.1 ± 0.8^{b}
Antarctic minke whale, $n=10$	0.027 ± 0.021^{b}	-24.6 ± 0.4^{e}	6.2 ± 0.4^{d}	14.6 ± 0.7^{b}

See Table 1.
Different superscripts indicate significant differences ($p < 0.05$).

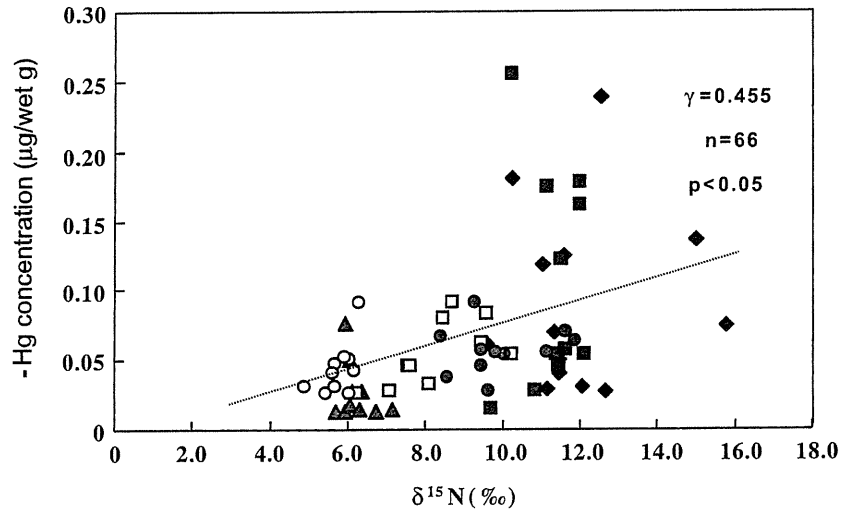

Fig. 1. Relationship between the $\delta^{15}N$ value and the Hg concentration in red meat products originating from baleen whale species or population. See Table 1. J-type common minke whale (◆), O-type common minke whale (■), Bryde's whale (●), sei whale (□), Antractic minke whale (▲), fin whale (○). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

while that in krill found in the stomach of common minke whales caught in the western North Pacific Ocean was 7.2 ± 0.5‰ (Mitani and Bando, 2008). Thus, the trophic positions of Bryde's and fin whales in the western North Pacific Ocean appear to be similar to those in the Gulf of California, respectively.

The $\delta^{13}C$ values in common minke (J- and O-types), Bryde's and sei whales caught in the western North Pacific Ocean were significantly different from those in fin and Antarctic minke whales caught in the Southern Ocean (Table 2), probably reflecting differences in their habitats. Krahn et al. (2008) reported the $\delta^{13}C$ and $\delta^{15}N$ values in the biota of Antarctica: the $\delta^{13}C$ and $\delta^{15}N$ values in the skin of an Antarctic minke whale ($n=1$) were -24.3 and 7.6‰, respectively, and those in the serum of crabeater seals (krill feeders) and in krill were -26.5 ± 1.0 and 8.4 ± 1.6‰ ($n=30$), and -29.8 ± 0.6 and 3.6 ± 0.2‰ ($n=12$), respectively. These $\delta^{13}C$ and $\delta^{15}N$ values in the Antarctic minke whale are in agreement with the present values from the muscle (red meat product) of Antarctic minke and fin whales caught in the Southern Ocean (Table 2). The $\delta^{15}N$ value in krill in the Antarctic Ocean (3.6 ± 0.2‰) was markedly lower than that in the stomach of common minke whales caught in Pacific Ocean (7.2 ± 0.5‰; Mitani and Bando, 2008). Lower $\delta^{15}N$ values in Antarctic minke and fin whales than common minke whale (Table 2) may reflect lower trophic levels of Antarctic minke and fin whales as well as lower $\delta^{15}N$ at the base of Southern food web.

The $\delta^{18}O$ values in common minke whales (J- and O-types) were significantly lower than those in the other whale species (Table 2), whereas the $\delta^{18}O$ values in Bryde's and sei whales caught in the western North Pacific Ocean and those in fin and Antarctic minke whales caught in the Southern Ocean were similar. As far as we know, no information on $\delta^{18}O$ values in cetaceans is available. As the temperature of the Antarctic feeding habitat is lower than that of the temperate North Pacific Ocean habitat, we expected to observe lower $\delta^{18}O$ values in whales in the Antarctic. However, the $\delta^{18}O$ values in the fin and Antarctic minke whales caught in the Antarctic were similar to those of Bryde's and sei whales caught in the North Pacific Ocean. Further study is necessary to explain these unexpected data.

We previously reported the $\delta^{13}C$ and $\delta^{15}N$ values and the Hg concentration in the toothed whale species hunted or stranded along the coast of Japan. The $\delta^{15}N$ values and the Hg concentrations in the toothed whale species (Endo et al., 2005) were markedly higher than those in the baleen whale species in this study (Table 2), reflecting their higher trophic positions. Further determination of $\delta^{18}O$ in the toothed whales from a broad latitudinal range is needed to elucidate whether $\delta^{18}O$ is higher in the toothed whales inhabiting the northern area and whether ^{18}O is bioaccumulated via the food web.

Significant positive correlations ($p < 0.05$) were found between the $\delta^{13}C$ and $\delta^{15}N$ values for Bryde's and sei whales (Table 3), and non-significant positive correlations ($p > 0.05$) were found in the other species caught in the western North Pacific Ocean and the Antarctic Ocean. We previously reported a positive correlation between $\delta^{13}C$ and $\delta^{15}N$ values in wild bluefin tuna taken from different areas around Japan (both values were lower in fish from the northern area), probably reflecting the change in diet due to the wide ranging annual migration from the southern to the northern areas (Hisamichi et al., 2010). Baleen whales, such as the

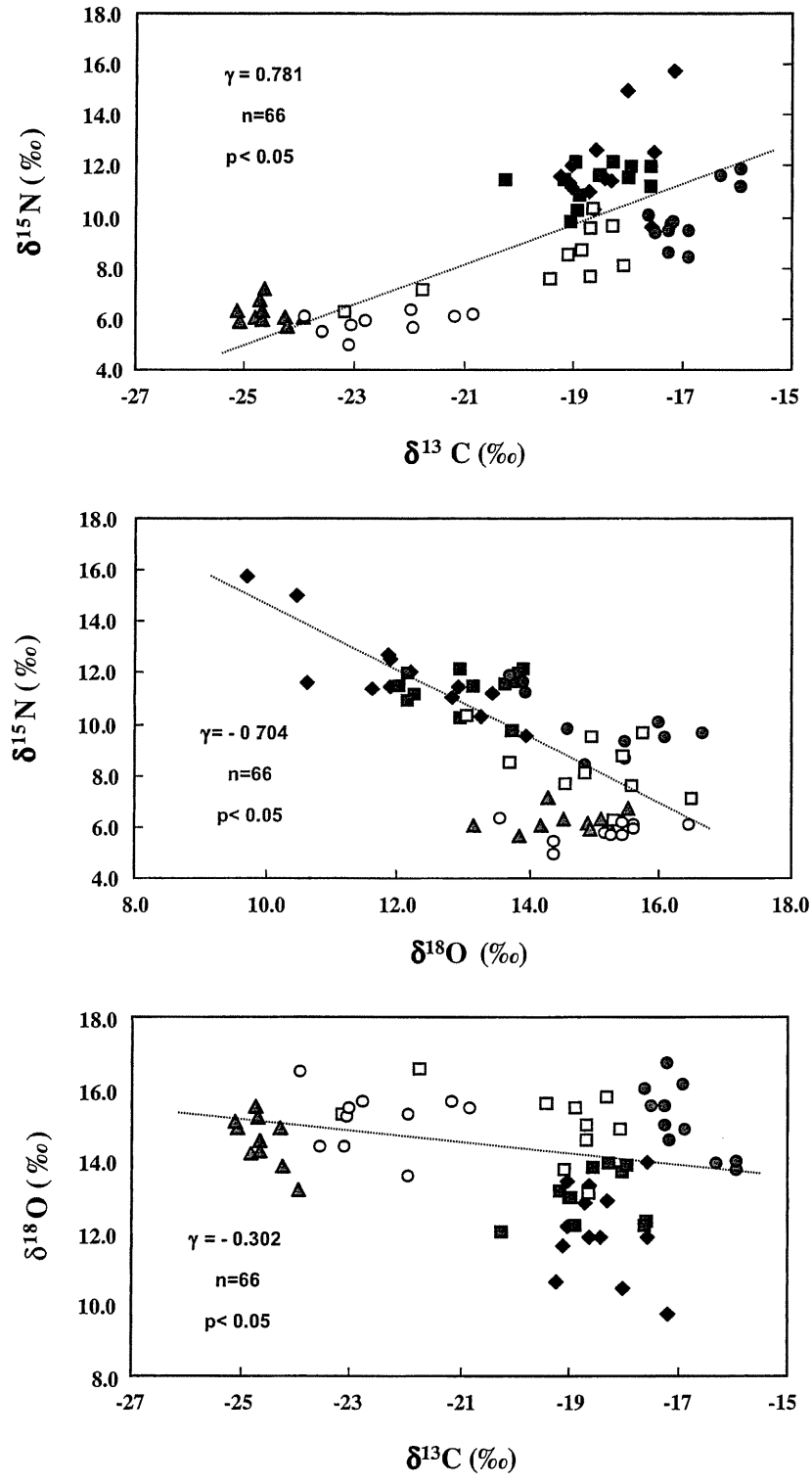

Fig. 2. Relationship among values of $\delta^{13}C$ and the $\delta^{18}O$ in red meat products originating from baleen whale species or population. See Table 1. J-type common minke whale (◆), O-type common minke whale (■), Bryde's whale (●), sei whale (□), Antractic minke whale (▲), fin whale (○). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

common minke (J- and O-types) and Antarctic minke whales, migrate over wide ranges in the North Pacific Ocean and the Southern Ocean, respectively (Kasamatsu et al., 1995; Wade et al., 2010). The positive correlations between the $\delta^{13}C$ and $\delta^{15}N$ values found in the baleen whale species could reflect their migration.

Unfortunately, we do not have any information on whale products with regard to location or date that each whale was killed or the age of the whale. Consequently, it is unclear whether the higher $\delta^{13}C$ and $\delta^{15}N$ values found in the red meat product samples come from whales in the southern or northern areas off

Table 3
Correlation coefficients (γ) of mercury and natural isotopes for within-species samples and overall samples for species or populations of baleen whales.

	Hg vs. $\delta^{15}N$	$\delta^{13}C$ vs. $\delta^{15}N$	$\delta^{15}N$ vs. $\delta^{18}O$	$\delta^{13}C$ vs. $\delta^{18}O$
Common minke whale (J-type), $n=13$	0.074	0.435	−0.852*	−0.185
Common minke whale (O-type), $n=12$	−0.077	0.315	0.199	0.182
Bryde's whale, $n=11$	0.188	0.740*	−0.640*	−0.774*
Sei whale, $n=10$	0.651*	0.751*	−0.484	−0.398
Fin whale, $n=10$	0.618	0.527	0.190	−0.250
Antarctic minke whale, $n=10$	−0.303	0.229	0.229	−0.672*
Overall, $n=66$	0.455*	0.781*	−0.704*	−0.302*

See Table 1.

* $p < 0.05$.

Japan. Based on the negative correlation between the $\delta^{13}C$ and $\delta^{18}O$ values and the $\delta^{15}N$ and $\delta^{18}O$ values (Table 3), it is assumed that the lower $\delta^{18}O$ values in whales in the northern areas result in the higher $\delta^{13}C$ and $\delta^{15}N$ values in whales in the northern area of Japan. However, this hypothesis is not supported by the lower $\delta^{13}C$ and $\delta^{15}N$ values found in wild bluefin tuna in the northern area of Japan (Hisamichi et al., 2010). Further study is necessary to confirm our assumption of spatial variations in $\delta^{18}O$, $\delta^{15}N$ and $\delta^{13}C$ values.

The $\delta^{13}C$–$\delta^{15}N$ plots and the $\delta^{13}C$–$\delta^{18}O$ plots can be discriminated into two groups (Table 2 and Fig. 2): the red meat products originating from the western North Pacific Ocean (J- and O-type common minke whales, Bryde's and sei whales) and the Antarctic Ocean (fin and Antarctic minke whales). We previously analyzed organohalogen compounds such as PCBs and DDTs and reported that the levels were markedly lower in the red meat products originating from the Southern Ocean than in products from the western North Pacific Ocean (Haraguchi et al., 2000). Thus, discrimination between the red meat products originating from the western North Pacific Ocean and the Antarctic Ocean could be achieved by the chemical analysis of stable isotope ratios and the pollutants without the need for genetic analysis. However, Antarctic minke and fin whales, J- and O-type common minke whales and Bryde's and sei whales could not be discriminated on the basis of chemical analysis. On the other hand, the $\delta^{13}C$ and $\delta^{15}N$ values in the red meat products originating from baleen whales sold in Japan were markedly different from those in products originating from toothed whales (Endo et al., 2010). Furthermore, contamination levels of Hg as well as organohalogens found in the baleen whales were markedly lower than those in toothed whales. Thus, the red meat originating from mysticetes and odonotocetes sold in Japan can be discriminated through chemical analysis.

In conclusion, we analyzed the Hg concentration and the $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ values in red meat products originating from common minke (J- and O-types), Bryde's and sei whales in the western North Pacific Ocean and fin and Antarctic minke whales in the Southern Ocean. The range of Hg concentrations and the $\delta^{15}N$ values in the baleen species and populations were in agreement with the known feeding habits of those. The $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ values could be used to discriminate between the red meat products originating from the mysticetes in the western North Pacific Ocean and those from the Southern Ocean. However, the four mysticetes in the western North Pacific Ocean and the two mysticetes in the Southern Ocean could not be identified on the basis of these data alone. A positive correlation between the $\delta^{13}C$ and $\delta^{15}N$ values and negative correlations between the $\delta^{13}C$ and $\delta^{18}O$ values and the $\delta^{15}N$ and $\delta^{18}O$ values, probably reflecting migration, were found in some species in the western North Pacific Ocean and the Southern Ocean.

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Levels of Mercury and Organohalogen Compounds in Pacific Bluefin Tuna (*Thunnus orientalis*) Cultured in Different Regions of Japan

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Abstract Contamination levels of total mercury (T-Hg), *p,p'*-DDE, and polychlorinated biphenyls (PCBs) in *akami* (leaner meat) and *toro* (fatty meat) samples from Pacific bluefin tuna cultured in the southern (four locations) and central (three locations) regions of Japan were analyzed. The contamination level of T-Hg in the *akami* and *toro* samples from the southern region tended to decrease with an increase in latitude, whereas those of *p,p'*-DDE and PCBs tended to increase. These spatial trends in contaminants were similar to those reported previously in wild tuna caught off the coast of Japan (Hisamichi et al., in Environ Sci Technol 44:5971–5978, 2010). However, the contamination level of T-Hg in *akami* and *toro* samples from one location in the central region was the highest among all seven locations, whereas the contamination level of *p,p'*-DDE was lower than that from any location studied in the southern region. Thus, contamination levels of T-Hg, *p,p'*-DDE, and PCBs in the cultured tuna may reflect contamination levels not only in the marine environment but also in prey fish used as bait.

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Large predatory fishes, such as tuna, shark, swordfish, and marlin, accumulate high levels of environmental pollutants by way of the food web. Of these species, tuna are particular important as a marine resource, and knowledge regarding the contamination level of mercury (Hg) and related health risks is of great interest to consumers. The permitted levels of total mercury (T-Hg) and methylmercury (M-Hg) in fish and shellfish set by the Japanese Ministry of Health and Welfare are 0.4 and 0.3 μg/wet g, respectively. However, this legislation does not cover the Hg contamination in some of the large predatory fish mentioned previously. Due to concerns over the impact of M-Hg on developing fetuses, the Food and Agriculture Organization/World Health Organization Joint Expert Committee on Food Additives (JECFA 2003) lowered its guideline value for provisional tolerable weekly intake of M-Hg from 3.3 to 1.6 μg/kg body weight (JECFA 2003).

In addition to Hg, tuna accumulate anthropogenic lipophilic compounds, such as polychlorinated biphenyls (PCBs), dichloro-diphenyl-trichloroethane (DDT) and its metabolites (DDTs: *p,p'*-DDT, *p,p'*-DDD, and *p,p'*-DDE), chlordane-related compounds (CHLs: trans-chlordane, cis-chlordane, trans-nonachlor, cis-nonachlor, and oxychlordane), and hexachlorobenzene (HCB) (Ueno et al. 2002, 2003; Hisamichi et al. 2010) as well as naturally produced compounds of tribromoanisol (TBA) (Penta-Abaurrea et al. 2009) and 2,3,3′,4,4′,5,5′-heptachloro-1-2′-bipyrrole (referred to as Q1) (Hisamichi et al. 2010). In contrast to the great emphasis placed on Hg contamination, a little attention has been paid to the potential human health problems associated with the contamination of PCBs and other lipophilic pollutants in tuna. We previously reported that contamination levels of Hg in three tuna species caught off the southern region of Japan were greater than those in tuna caught in the central and northern regions, whereas

contamination levels of PCBs and *p,p*-DDE (a major metabolite of DDTs) caught off the southern region were lower, probably reflecting the contamination levels in the respective marine environment (Hisamichi et al. 2010).

Japan is the world's largest consumer of tuna, and the majority of consumption is in the form of slices of raw fish (sashimi and sushi). Because people living in countries other than Japan have also recently come to eat much more tuna, tuna numbers have begun to dwindle. Recently, the demand for muscle containing a lot of fat (*toro*) has increased as a result of changing preferences among the Japanese population. As a result, the price of *toro* is greater than that of *akami* (lean meat), although there is no fixed standard for distinguishing *akami* from *toro*. To supplement the lack of wild tuna, fatty meat in particular, the business of farming tuna has expanded in Japan.

The technology for farming (culturing) tuna, in which wild tuna are caught and used to stock farms, has been well developed and applied commercially for the Atlantic bluefin tuna (*Thunnus thynnus*) in Mediterranean countries, for the southern bluefin tuna (*T. maccoyii*) in Australia, and for the Pacific bluefin tuna (*T. orientalis*) in Japan and Mexico. The aim of tuna farming in countries other than Japan, which operates through the confinement of captured tuna for short periods of time (usually 2–6 months), is mostly to increase the fat content in flesh (Tudela and Garcia 2004). In contrast, tuna culturing in Japan is usually aimed at cultivating fish captured in the larval stage for long periods (approximately 2.5–3 years) and to up to a fish weight of approximately 30–50 kg. Tuna culturing in Japan has been developed in temperate regions, such as the Okinawa, Kagoshima, Kochi, Nagasaki, Wakayama, and Mie Prefectures (Fig. 1). Recently, due to the development

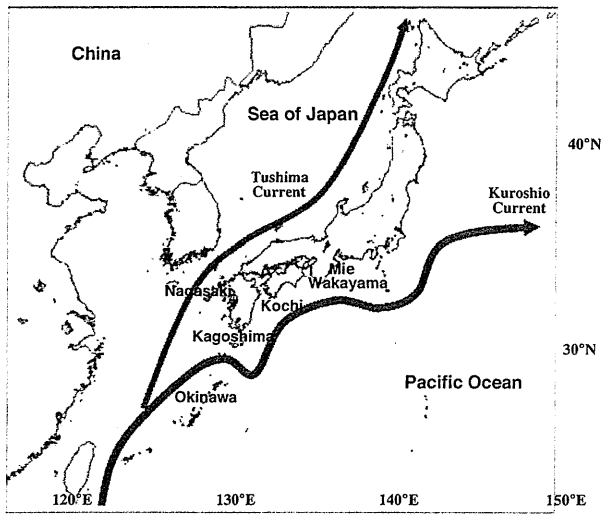

Fig. 1 Map of Japan showing six Prefectures in Japan where bluefin tuna is cultured

of hatchery technology, complete aquacultivation of tuna from gametes (full-cycle cultured Pacific bluefin tuna [FC tuna]), not from wild or larval tuna, has been achieved at Kinki University, Wakayama Prefecture (Nakao et al. 2007; Ando et al. 2008), and the meat of FC tuna cultured for approximately 3 years is now on the market.

Generally in wild tuna, Hg accumulation increases as the size of prey fish increases. However, the size and species of prey fish in farmed tuna can be controlled. Trials for the control of Hg concentration in the muscle (edible portion) have been undertaken in FC tuna (Nakao et al. 2007; Ando et al. 2008) and the farmed southern bluefin tuna (Balshaw et al. 2008a, b), because the increase in lipid content as well as rapid growth of the tuna could result in a decreased Hg concentration in muscle. In contrast, Padula et al. (2008) analyzed the contamination levels of lipophilic pollutants, dioxins, and PCBs in farmed and wild southern bluefin tuna and reported the greater levels of lipophilic contaminants in the farmed tuna than in the wild tuna. However, little is known about the contamination levels of those compounds in the tuna cultured in Japan.

Stable isotope ratios of $\delta^{13}C$ and $\delta^{15}N$ have been used to investigate feeding ecology. The $\delta^{15}N$ value is used to determine the trophic position of the studied species, and the $\delta^{13}C$ value is used to determine the source of carbon by the primary producer in a trophic web, providing information on the foraging habits of the species studied (Kelly 2000). Furthermore, the $\delta^{13}C$ and $\delta^{15}N$ values are known to vary by habitat. For instance, both $\delta^{15}N$ and $\delta^{13}C$ values in wild Pacific bluefin tuna caught off the northern region of Japan were lower than those in tuna caught off the southern region, probably reflecting their wide-ranging migration (Hisamichi et al. 2010). Furthermore, the latitudinal effects on $\delta^{13}C$ and $\delta^{15}N$ values in wild yellowfin and albacore tuna caught off the coast of central and southern Japan have been reported (Hisamichi et al. 2010); $\delta^{13}C$ values in those fish tended to decrease with an increase in latitude, whereas $\delta^{15}N$ values tended to increase. Enrichment of $\delta^{15}N$ by farming was reported in bluefin tuna farmed in the Mediterranean Sea (Vizzini et al. 2010). However, $\delta^{13}C$ and $\delta^{15}N$ values in tuna cultured in Japan have not yet been analyzed and compared with those of wild tuna.

The aim of the present study was to analyze the contamination levels of T-Hg, M-Hg, 13 PCB congeners, *p,p'*-DDE, trans-nonachlor (a major chemical among CHLs), Q1, HCB, and TBA in *akami* and *toro* samples of bluefin tuna cultured at different locations in Japan. Furthermore, we analyzed $\delta^{13}C$ and $\delta^{15}N$ levels in *akami* samples of tuna cultured at different locations. These results were compared among locations and with those for wild bluefin tuna reported previously (Hisamichi et al. 2010).