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Effects of Cooking on Concentrations of Polychlorinated Dibenzo-*p*-dioxins and Related Compounds in Fish and Meat

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We investigated the cooking-induced changes in concentrations of polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyls (PCBs) (dioxins) using mackerel and beef. The concentrations of dioxins (29 congeners) were determined by isomer specific analyses and were compared between uncooked and cooked samples. The cooking procedures examined in this study included grilling as a fillet, boiling as a fillet, and boiling as tsumire (small, hand-rolled balls) for mackerel and boiling as a slice, broiling as a slice, and broiling as a hamburger for beef. Three trials were carried out for each cooking method. Generally, concentrations of dioxins were reduced in every cooking trial. When nondetected congener concentrations were assumed to be half the limit of detection for mackerel, the maximum percentage reductions of total concentrations given as 2,3,7,8-tetraCDD equivalents (TEQ) were 31% in grilling as a slice, 14% in boiling as a slice, and 21% in boiling as tsumire under the conditions of this study. In contrast, for beef, the reductions were 42% in boiling as a slice, 42% in broiling as a slice, and 44% in broiling as a hamburger. These results suggest that ordinary cooking processes with heating undoubtedly reduce the dioxin content in animal products, and the reductions estimated should be considered when dioxin intake is evaluated using contamination data for individual food items.

KEYWORDS: PCDDs; PCDFs; dioxin-like PCBs; food; animal products

INTRODUCTION

In 1999, the Environmental Agency and the Ministry of Health and Welfare of Japan reevaluated the tolerable daily intake (TDI) of dioxins and determined it to be 4 pg/kg body weight/day when indicated as 2,3,7,8-tetraCDD equivalents (TEQ). It is generally accepted that food consumption is a major source of exposure to dioxins. Therefore, the estimation of dietary intake of dioxins based on typical Japanese dietary habits is likely to represent most of the average intake of dioxins in the Japanese consumer. The estimation-based total diet study (TDS) demonstrated that the recent Japanese dietary intake of dioxins is below the TDI and that the mean daily intake has declined during the past 20 years in the Kansai district of western Japan (*1*−*3*), which may indicate that the levels of exposure to dioxins fall under the TDI in the rest of Japan. However, the possibility of adverse effects on human health caused by long-term exposure to dioxins and their accumulation in the body cannot be neglected. Ongoing data collection and updates regarding human dietary exposure to dioxins should continue to be examined, with the long-term goal of a continued reduction of dioxin levels in the human population.

Generally, the daily dietary intake of food contaminants is evaluated via a TDS (*1*−*3*) or a duplicate diet study (*4*, *5*). The estimation values derived from these methods include effects of cooking and/or processing, since the food items are cooked and homogenized prior to analysis. On the other hand, the intake can be quickly calculated from data on individual foods by combining their mean contamination (concentration) with their average consumption (fresh weight) in a specified area (*6*, *7*). These methods do not take into account any effects of cooking, which means that studies of the changes in dioxin content induced by ordinary cooking are indispensable for estimating the actual intake from an individual food. To our knowledge, typical cooking procedures and dietary habits have not been considered in studies of dioxin content. In particular, evaluating the cooking-induced changes in animal products is most important, as these kinds of food are considered to be the most prominent source of the human dietary intake of dioxins in Japan (*1*, *2*) as well as other nations (*8*−*10*). We have previously reported that dioxin levels in leafy vegetables decrease during ordinary Japanese cooking procedures (*11*, *12*); however, we

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have found no surveys of cooking-induced changes in dioxin concentrations, or those of 29 toxic congeners, for animal products.

In the present study, we employed mackerel and beef from the market as food models and examined concentrations of dioxins before and after the cooking of these foods to investigate the effects of cooking on the dioxin content of food. We believe that the data gathered are useful for estimating the actual Japanese dietary intake of dioxins.

MATERIALS AND METHODS

Preparation of Fish Sample (Mackerel). We purchased 18 Japanese inshore mackerel (about 25 cm long) in January 2000. Cooked and control samples were prepared according to the method reported by Trotter et al. (*13*). Namely, one fillet from each fish was homogenized raw and used as a control, while the other side was cooked. We examined three different cooking protocols, as follows: (i) Grill: grilling for about 8 min. The released juice was discarded. (ii) Boil: boiling in 800 mL of tap water for 10 min and discarding the stock. (iii) Tsumire: chopping in a food processor and hand-rolling the chopped fish into balls the size of a mouthful (the Japanese often name these preparations and/or balls tsumire). The tsumire were then boiled in 800 mL of tap water for 10 min, and the stock was discarded.

The sample weights were recorded before and after cooking. After cooking, each sample was chopped in a food processor. Overall, three trials were carried out for each cooking method using individual sets of mackerel.

Preparation of Meat Sample (Beef). We purchased three chunks of domestic beef (about 1 kg apiece) in March 2000. Each chunk was cut into 3 mm thick slices from one end. The slices were then arranged into four piles (about 250 g in a pile), with each having almost the same dioxin content that would be obtained from a chunk. Of the four piles, one was chopped in a food processor without being cooked and served as a control. Using the remaining three piles, we carried out three different cooking procedures. The protocols were as follows: (i) Boil: boiling in 700 mL of tap water for 5 min and discarding the stock. (ii) Broil: broiling both sides of each slice on a hot plate until the surface was slightly burned (for approximately 5 min). The released juice was discarded. (iii) Hamburger: chopping in a food processor and hand-forming a hamburger patty and then broiling both sides of each patty for approximately 10 min on a hot plate. The juice was discarded.

The sample weights were recorded before and after cooking. After cooking, each sample was chopped in a food processor. Overall, three trials were carried out for each cooking method using the individual chunks of meat.

Sample Extraction and Cleanup. The chemicals and standards used in this study were described in our previous paper (*14*). We determined 29 kinds of dioxins congeners, all of which have toxic equivalent factors (TEFs) of the World Health Organization (*15*). Each sample (50 g) was spiked with 17 kinds of $^{13}C_{12}$-labeled polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) and 12 kinds of $^{13}C_{12}$-labeled polychlorinated biphenyls (PCBs) as an internal quantification standard. The samples were treated with 300 mL of 1 N potassium hydroxide/ethanol for 2 h with stirring at room temperature. In view of the percent recovery of internal quantification standards and analysis of certified reference material, the alkaline treatment used in this study did not induce degradation of dioxins during the process (data not shown). The alkaline hydrolyzate was extracted twice with 150 mL of *n*-hexane, and the extract was then treated with 15 mL of concentrated sulfuric acid and subsequently transferred to a silver nitrate-impregnated silica gel column. The eluate with 100 mL of *n*-hexane was evaporated and then loaded on an active carbon column on which mono-*ortho*-PCBs were eluted with 50 mL of 10% (v/v) dichloromethane/*n*-hexane. Non-*ortho*-PCBs and PCDD/Fs were then eluted with 150 mL of toluene. In the first fraction, coelution of some matrices (maybe a kind of aliphatic hydrocarbon) was observed and disturbed the gas chromatography/mass spectrometry (GC/MS) analysis. Accordingly, the first eluate was further purified using the acetonitrile/*n*-hexane partition technique. Both fractions were concentrated to a final volume of 20−25 μL, respectively. We used $^{13}C_{12}$-labeled 2,3,3′,5,5′-pentaCB and $^{13}C_{12}$-labeled 1,2,3,4-tetraCDD as internal recovery standards.

Table 1. Selected Ions on HRGC/HRMS Analysis

	monitor ions (*m*/*z*)		
	native compound		$^{13}C_{12}$-labeled
compound	for quantification	for identification	compound
tetraCDD	321.8936	319.8965	333.9339
pentaCDD	355.8546	353.8576	367.8949
hexaCDD	389.8159	391.8127	401.8559
heptaCDD	423.7766	425.7737	435.8169
octaCDD	459.7348	457.7377	471.7750
tetraCDF	305.8987	303.9016	317.9389
pentaCDF	339.8597	337.8208	351.9000
hexaCDF	373.8208	375.8178	385.8610
heptaCDF	407.7818	409.7789	419.8220
octaCDF	443.7399	441.7428	455.7801
tetraCB	291.9194	289.9224	303.9597
pentaCB	325.8804	323.8834	337.9207
hexaCB	359.8415	361.8385	371.8817
heptaCB	393.8025	395.7986	405.8428

Analysis of PCDD/Fs and Dioxin-like PCBs. High-resolution (HR) GC/HRMS analysis was performed using an HP 6890 gas chromatograph (Hewlett-Packard, CA) coupled with an AutoSpec ULTIMA mass spectrometer (Micromass, United Kingdom). An SP-2331 (60 m length, 0.32 mm i.d., 0.2 μm film thickness, Supelco, PA) capillary column was used to determine tetra-, penta-, and hexa-CDD/Fs. The GC conditions were as follows: column temperature program, 130 (held for 1 min) to 180 °C at 20 °C/min, 240 °C (held for 15 min) at 3 °C/min; injector temperature, 260 °C; and injection volume, 2 μL. A BPX-5 capillary column (60 m length, 0.25 mm i.d., 0.25 μm film thickness, SGE, Australia) was used to determine hepta- and octa-CDD/Fs as well as non-*ortho*-PCBs. The GC conditions were as follows: column temperature program, 150 (held for 1 min) to 220 °C at 20 °C/min, 320 °C (held for 3.2 min) at 3 °C/min; injector temperature, 280 °C; and injection volume, 2 μL. An HT-8 capillary column (50 m length, 0.22 mm i.d., 0.25 μm film thickness, SGE, Australia) was used to determine mono-*ortho*-PCBs. The GC conditions were as follows: column temperature program, 130 (held for 1 min) to 220 °C at 20 °C/min, 280 °C at 3 °C/min, 320 °C at 20 °C/min (held for 1 min); injector temperature, 280 °C; and injection volume, 1 μL. The monitoring ions used in the GC/MS analysis are given in **Table 1**, and other MS conditions were described in our previous paper (*16*). A limit of detection (LOD) for each congener was determined according to the guidelines for food analysis of dioxins issued by the Japanese government ("Guideline") (*17*). The LODs in this study were 0.005 pg/g for tetraCDD/Fs and pentaCDD/Fs, 0.01 pg/g for hexaCDD/Fs and heptaCDD/Fs, 0.025 pg/g for octaCDD/F, 0.1 pg/g for non-*ortho*-PCBs, and 1 pg/g for mono-*ortho*-PCBs. The TEQs were calculated using the TEFs. The concentrations of nondetected congeners were principally replaced by half of the LOD (ND = LOD/2), although they were also assumed to be zero (ND = 0).

Measurement of Fat Content. Extraction of fat from fish and meat samples was performed according to "Guideline". Approximately 5 g of homogenized mackerel or beef was weighed in a glass centrifuge bottle and stirred continually with 20 g of sodium sulfate anhydrate. The obtained dehydrated powder was extracted three times with diethyl ether/*n*-hexane (1:2, v/v). The extract was then washed twice with 10 mL of distilled water and dried over sodium sulfate anhydrate. The solution was evaporated until dry, and the resolved residue was transferred to a 50 mL beaker. The solvent was dried in the atmosphere, and the residue was gravimetrically measured as fat content.

Measurement of Water Content. Approximately 5 g of homogenized mackerel or beef was weighed in a 50 mL beaker and subsequently heated in an aging oven at 130 °C for 3 h. The weight decrease was gravimetrically measured as the water content.

Table 2. Description of the Appearance of Food Samples before and after Cooking

A: Fish

	grill-1	grill-2	grill-3	boil-1	boil-2	boil-3	tsumire-1	tsumire-2	tsumire-3
weight									
before cooking (g)	235.0	224.1	232.2	224.8	191.6	235.7	176.2	164.9	220.2
after cooking (g)	179.5	173.6	179.6	194.1	160.0	202.2	168.6	162.2	202.2
% of weight change	–24	–23	–23	–14	–16	–14	–4.3	–1.6	–8.2
fat content									
before cooking (%)	20.7	13.3	10.7	9.4	8.0	13.9	12.5	11.8	13.0
after cooking (%)	21.7	14.5	12.1	10.7	10.5	12.5	12.3	10.9	11.2
change (%)	+1.0	+1.2	+1.4	+1.3	+2.5	–1.4	–0.2	–0.9	–1.8
water content									
before cooking (%)	53.6	63.2	65.4	66.4	68.2	65.0	57.9	63.2	50.6
after cooking (%)	50.4	56.9	59.8	63.0	64.0	61.3	65.7	67.1	64.6
change (%)	–3.2	–6.3	–5.6	–3.4	–4.3	–3.7	7.8	4.0	14

B: Meat

	boil-1	boil-2	boil-3	broil-1	broil-2	broil-3	hamburger-1	hamburger-2	hamburger-3
weight									
before cooking (g)	257.7	250.2	252.5	252.6	250.8	248.1	226.8	238.3	230.9
after cooking (g)	173.2	175.1	159.3	184.2	174.5	172.7	187.4	189.4	174.3
% of weight change	–33	–30	–37	–27	–30	–30	–17	–21	–25
fat content									
before cooking (%)	11.0	15.9	20.2	11.0	15.9	20.2	11.0	15.9	20.2
after cooking (%)	13.7	17.6	21.8	12.7	19.6	24.3	13.2	16.8	18.8
change (%)	+2.7	+1.7	+1.6	+1.7	+3.7	+4.1	+2.2	+0.9	–1.4
water content									
before cooking (%)	66.2	62.1	58.4	66.2	62.1	58.4	66.2	62.1	58.4
after cooking (%)	57.0	54.1	50.5	57.6	51.4	46.2	59.0	55.0	53.6
change (%)	–9.2	–8.0	–7.9	–8.6	–10.7	–12.1	–7.1	–7.2	–4.8

RESULTS

Effects of Cooking on Mackerel. The average dioxin concentrations of uncooked mackerel (control) were 2.4 pg TEQ/g (ranging from 0.84 to 5.6 pg TEQ/g) when ND = 0 was adopted. This was similar to the results of a recent national survey for the edible portion of fish and shellfish: 1.49 pg TEQ/g on average (rangeing from 0.003 to 23.093 pg TEQ/g). It can therefore be said that the fish samples used here are suitable for simulating typical household cooking in Japan.

Cooking-induced changes in the appearance of fish samples are described in **Table 2A**. Decreases in sample weights were found in all procedures: Grilling reduced the weight by 51−66 g, boiling reduced the weight by 31−34 g, and tsumire reduced the weight by 2.7−18 g; these values correspond to 23−24, 14−16, and 1.6−8.2% decreases, respectively. Although cooking must have induced the release of fat from the food, an increase in fat content was observed in all grill preparations and two boil preparations. We believe the increased fat content was caused by a loss of moisture, which made the overall proportion of fat greater. In contrast with grill and boil preparations, an increase in water content and a decrease in fat content were observed in all of the tsumire preparations, which means that the water was absorbed by the fish balls during the boiling process. These changes in weight made the cooking effects unclear: Most of the concentrations calculated based on the weight after cooking decreased, although some increases were also found.

To correct for these weight changes, we recalculated the concentrations in cooked samples on the basis of the original weights. Consequently, reductions in the concentrations of detected isomers were obtained in all trials, as shown in **Tables 3−5**. The mean total TEQ calculated at ND = LOD/2 was reduced to 62−73% of the initial concentrations by grilling, 84−87% of the initial concentrations by boiling, and 76−85% of the initial concentrations by tsumire (**Table 6**). Generally, the largest effects were obtained in grilled preparations, while boiling resulted in the smallest effects. However, significant differences among the three procedures could not be verified, as dioxin concentrations were originally different among the control samples.

Effects of Cooking on Beef. The average dioxin concentrations of uncooked beef (control) were 0.13 pg TEQ/g (ranging from 0.0361 to 0.315 pg TEQ/g) when ND = 0 was adopted. These findings are almost the same as those of a recent national survey for meat: 0.191 pg TEQ/g on average (ranging from <0.001 to 1.434 pg TEQ/g). The meat samples used here are believed to be typical of those consumed in Japan.

Similar to the case with mackerel, we found that with some cooking methods dioxin concentrations were increased somewhat in beef. The procedures reduced sample weights as follows: boiling, 75−93 g; broiling, 68−76 g; and hamburger broiling, 47−57 g. Moreover, an increased fat content was observed in all preparations, except for a preparation of broiled hamburger (**Table 2B**). We conclude that these fat increases were induced by excessive weight loss, as with the mackerel.

Tables 7−9 show the calculations for concentrations of dioxins in the cooked beef on the basis of the original weights, which show decreases in most isomers. The mean total TEQ calculated at ND = LOD/2 was reduced to 48−68% of initial concentrations by boiling, 50−69% of initial concentrations by broiling, and 48−63% of initial concentrations by hamburger broiling (**Table 10**). We found no obvious differences among the three cooking procedures.

DISCUSSION

We examined three cooking procedures each for mackerel and beef to study their effects on dioxin concentrations. Although cooking seemed to increase the concentrations of some dioxins in the samples, this finding was most likely a consequence of excess weight changes. Accordingly, the original

Table 3. Concentrations (pg/g)[a] of PCDD/Fs and Dioxin-like PCBs in Uncooked and Cooked Mackerel Samples (Grill)

	grill-1		grill-2		grill-3	
congener	uncooked	cooked	uncooked	cooked	uncooked	cooked
2,3,7,8-tetraCDD	ND	ND	ND	ND	0.068	0.036
1,2,3,7,8-pentaCDD	0.18	0.081	0.17	0.11	0.15	0.14
1,2,3,4,7,8-hexaCDD	ND	ND	ND	ND	0.0290	ND
1,2,3,6,7,8-hexaCDD	ND	ND	ND	ND	0.0250	ND
1,2,3,7,8,9-hexaCDD	ND	ND	ND	ND	ND	ND
1,2,3,4,6,7,8-heptaCDD	0.052	0.046	0.042	0.029	0.045	0.039
octaCDD	0.046	0.040	0.047	0.043	0.075	0.066
2,3,7,8-tetraCDF	0.74	0.44	1.3	0.97	0.76	0.58
1,2,3,7,8-pentaCDF	0.18	0.13	0.26	0.14	0.23	0.15
2,3,4,7,8-pentaCDF	0.48	0.34	0.81	0.66	0.66	0.58
1,2,3,4,7,8-hexaCDF	0.065	0.040	0.058	ND	0.060	0.043
1,2,3,6,7,8hexaCDF	ND	ND	0.069	0.033	0.058	0.037
1,2,3,7,8,9-hexaCDF	ND	ND	ND	ND	ND	ND
2,3,4,6,7,8-hexaCDF	0.084	0.060	ND	ND	0.068	0.036
1,2,3,4,6,7,8-heptaCDF	0.039	0.027	0.042	0.022	0.048	0.025
1,2,3,4,7,8,9-heptaCDF	ND	ND	ND	ND	ND	ND
octaCDF	ND	ND	ND	ND	ND	ND
3,3′,4,4′-tetraCB (#77)	16	14	59	41	21	17
3,4,4′,5-tetraCB (#81)	1.8	1.2	4.6	3.8	3.1	2.7
3,3′,4,4′,5-pentaCB (#126)	8.5	5.0	20	14	14	9.4
3,3′,4,4′,5,5′-hexaCB (#169)	2.2	1.4	5.5	4.5	3.4	2.5
2′,3,4,4′,5-pentaCB (#123)	4.3	3.8	10	4.9	5.2	4.1
2,3′,4,4′,5-pentaCB (#118)	379	318	963	660	700	524
2,3,4,4′,5-pentaCB (#114)	16	14	50	35	37	28
2,3,3′,4,4′-pentaCB (#105)	147	114	341	252	255	172
2,3′,4,4′,5,5′-hexaCB (#167)	38	30	89	64	72	50
2,3,3′,4,4′,5-hexaCB (#156)	45	42	139	99	105	77
2,3,3′,4,4′,5′-hexaCB (#157)	14	12	44	34	34	25
2,3,3′,4,4′,5,5′heptaCB (#189)	9.4	6.2	20	14	17	12

[a] Concentrations in cooked samples were calculated on the basis of the original weight.

Table 4. Concentrations (pg/g)[a] of PCDD/Fs and Dioxin-like PCBs in Uncooked and Cooked Mackerel Samples (Boil)

	boil-1		boil-2		boil-3	
congener	uncooked	cooked	uncooked	cooked	uncooked	cooked
2,3,7,8-tetraCDD	0.11	0.11	0.031	0.031	0.25	0.20
1,2,3,7,8-pentaCDD	0.25	0.23	0.11	0.087	0.40	0.36
1,2,3,4,7,8-hexaCDD	0.022	0.020	0.023	0.014	ND	ND
1,2,3,6,7,8-hexaCDD	0.060	0.043	0.029	0.024	0.043	0.037
1,2,3,7,8,9-hexaCDD	0.011	0.013	0.011	ND	0.014	0.011
1,2,3,4,6,7,8-heptaCDD	0.043	0.039	0.025	0.018	0.022	0.018
octaCDD	0.047	0.030	0.041	0.038	0.037	0.037
2,3,7,8-tetraCDF	1.4	1.3	0.48	0.47	1.3	1.1
1,2,3,7,8-pentaCDF	0.30	0.28	0.17	0.15	0.25	0.21
2,3,4,7,8-pentaCDF	1.2	1.1	0.54	0.52	2.2	1.8
1,2,3,4,7,8-hexaCDF	0.073	0.056	0.028	0.028	0.051	0.036
1,2,3,6,7,8-hexaCDF	0.11	0.079	0.052	0.049	0.074	0.065
1,2,3,7,8,9-hexaCDF	0.015	0.010	ND	ND	0.044	0.034
2,3,4,6,7,8-hexaCDF	0.085	0.076	0.055	0.053	0.060	0.056
1,2,3,4,6,7,8-heptaCDF	0.028	0.016	0.024	0.023	0.035	0.021
1,2,3,4,7,8,9-heptaCDF	ND	ND	0.022	ND	0.061	ND
octaCDF	ND	ND	ND	ND	ND	ND
3,3′,4,4′-tetraCB (#77)	67	55	16	14	37	36
3,4,4′,5-tetraCB (#81)	5.4	4.2	2.2	0.9	1.9	1.2
3,3′,4,4′,5-pentaCB (#126)	22	18	13	11	30	25
3,3′,4,4′,5,5′-hexaCB (#169)	7.2	5.7	3.2	2.8	10	8.4
2′,3,4,4′,5-pentaCB (#123)	57	45	18	15	67	21
2,3′,4,4′,5-pentaCB (#118)	961	879	592	538	1658	1427
2,3,4,4′,5-pentaCB (#114)	59	54	30	28	116	105
2,3,3′,4,4′-pentaCB (#105)	380	354	219	197	821	695
2,3′,4,4′,5,5′-hexaCB (#167)	110	105	72	64	209	189
2,3,3′,4,4′,5-hexaCB (#156)	171	158	103	96	379	333
2,3,3′,4,4′,5′-hexaCB (#157)	52	52	33	31	112	104
2,3,3′,4,4′,5,5′-heptaCB (#189)	26	23	14	13	60	51

[a] Concentrations in cooked samples were calculated on the basis of the original weight.

sample weights were adopted to evaluate the concentrations more legitimately, with the result that most of the isomers showed obvious downward trends.

On the other hand, some dioxin concentrations increased or hardly changed, even after the original sample weights are applied for analyzing cooked samples. Three sets of data could

Table 5. Concentrations (pg/g)[a] of PCDD/Fs and Dioxin-like PCBs in Uncooked and Cooked Mackerel Samples (Boil as "Tsumire")

	tsumire-1		tsumire-2		tsumire-3	
congener	uncooked	cooked	uncooked	cooked	uncooked	cooked
2,3,7,8-tetraCDD	0.077	0.061	0.031	0.013	0.10	0.058
1,2,3,7,8-pentaCDD	0.18	0.16	0.15	0.045	0.14	0.097
1,2,3,4,7,8-hexaCDD	0.037	ND	ND	ND	ND	ND
1,2,3,6,7,8-hexaCDD	0.046	0.034	ND	ND	0.057	0.030
1,2,3,7,8,9-hexaCDD	0.029	0.012	0.022	ND	ND	ND
1,2,3,4,6,7,8-heptaCDD	0.022	0.021	0.021	0.020	0.030	0.028
octaCDD	0.035	0.029	ND	ND	0.032	0.032
2,3,7,8-tetraCDF	0.71	0.68	0.34	0.27	0.56	0.45
1,2,3,7,8-pentaCDF	0.22	0.22	0.14	0.11	0.24	0.16
2,3,4,7,8-pentaCDF	0.56	0.50	0.39	0.32	0.66	0.63
1,2,3,4,7,8-hexaCDF	0.041	0.025	0.024	0.020	0.039	ND
1,2,3,6,7,8-hexaCDF	0.064	0.053	0.038	0.037	0.093	0.062
1,2,3,7,8,9-hexaCDF	ND	ND	ND	ND	ND	ND
2,3,4,6,7,8-hexaCDF	0.061	0.039	0.054	0.042	0.039	0.018
1,2,3,4,6,7,8-heptaCDF	0.020	0.016	0.014	0.013	0.030	0.021
1,2,3,4,7,8,9-heptaCDF	ND	ND	ND	ND	ND	ND
octaCDF	ND	ND	ND	ND	ND	ND
3,3′,4,4′-tetraCB (#77)	26	22	7.7	7.1	20	17
3,4,4′,5-tetraCB (#81)	2.0	1.8	1.0	0.63	1.4	0.93
3,3′,4,4′,5-pentaCB (#126)	10	8.8	5.5	5.0	18	13
3,3′,4,4′,5,5′-hexaCB (#169)	2.9	2.8	1.8	1.5	4.8	3.8
2′,3,4,4′,5-pentaCB (#123)	6.4	4.8	4.1	3.6	25	19
2,3′,4,4′,5-pentaCB (#118)	777	449	445	258	1036	840
2,3,4,4′,5-pentaCB (#114)	26	22	19	14	41	34
2,3,3′,4,4′-pentaCB (#105)	232	167	118	92	406	315
2,3′,4,4′,5,5′-hexaCB (#167)	38	39	30	27	93	86
2,3,3′,4,4′,5-hexaCB (#156)	70	64	59	40	154	135
2,3,3′,4,4′,5′-hexaCB (#157)	17	17	20	13	38	32
2,3,3′,4,4′,5,5′-heptaCB (#189)	13	9.5	6.4	6.1	20	18

[a] Concentrations in cooked samples were calculated on the basis of the original weight.

Table 6. TEQ Concentrations (pg TEQ/g)[a] of PCDD/Fs and Dioxin-like PCBs in Uncooked and Cooked Mackerel Samples

	grill-1		grill-2		grill-3		
	uncooked	cooked	uncooked	cooked	uncooked	cooked	mean % decrease
PCDDs	0.18 (0.18)	0.084 (0.081)	0.17 (0.17)	0.12 (0.11)	0.22 (0.22)	0.14 (0.14)	41 (41)
PCDFs	0.34 (0.34)	0.23 (0.23)	0.56 (0.56)	0.43 (0.43)	0.44 (0.44)	0.36 (0.36)	24 (24)
dioxin-like PCBs	0.96 (0.96)	0.60 (0.60)	2.3 (2.3)	1.7 (1.7)	1.6 (1.6)	1.1 (1.1)	32 (32)
total dioxins	1.5 (1.5)	0.91 (0.91)	3.0 (3.0)	2.2 (2.2)	2.2 (2.2)	1.6 (1.6)	31 (31)
	boil-1		boil-2		boil-3		
	uncooked	cooked	uncooked	cooked	uncooked	cooked	mean % decrease
PCDDs	0.37 (0.37)	0.34 (0.34)	0.15 (0.15)	0.12 (0.12)	0.66 (0.66)	0.57 (0.57)	13 (13)
PCDFs	0.76 (0.76)	0.70 (0.70)	0.34 (0.34)	0.33 (0.33)	1.3 (1.25)	1.0 (1.03)	10 (10)
dioxin-like PCBs	2.6 (2.6)	2.1 (2.1)	1.5 (1.5)	1.3 (1.3)	3.6 (3.6)	3.1 (3.1)	16 (16)
total dioxins	3.7 (3.7)	3.2 (3.2)	2.0 (2.0)	1.7 (1.7)	5.6 (5.6)	4.7 (4.7)	14 (14)
	tsumire-1		tsumire-2		tsumire-3		
	uncooked	cooked	uncooked	cooked	uncooked	cooked	mean % decrease
PCDDs	0.27 (0.27)	0.22 (0.22)	0.19 (0.19)	0.060 (0.058)	0.25 (0.25)	0.16 (0.16)	40 (41)
PCDFs	0.38 (0.38)	0.34 (0.34)	0.25 (0.25)	0.20 (0.20)	0.4 (0.41)	0.4 (0.38)	13 (13)
dioxin-like PCBs	1.23 (1.23)	1.02 (1.02)	0.68 (0.68)	0.58 (0.58)	2.1 (2.1)	1.6 (1.6)	19 (19)
total dioxins	1.9 (1.9)	1.6 (1.6)	1.1 (1.1)	0.84 (0.84)	2.8 (2.8)	2.1 (2.1)	21 (21)

[a] Concentrations in cooked samples were calculated on the basis of the original weight. TEQ concentrations were calculated and summed at ND=LOD/2, while calculations at ND = 0 are shown in parentheses.

be found as follows: the concentration of 2,3′,4,4′,5,5′-pentaCB (PCB #167) in trial tsumire-1, the concentration of OCDD in trial tsumire-3, and 3,3′,4,4′-tetraCB (PCB #77) in trial hamburger-2. As far as we know, there is no information on new formation of dioxin elements induced by ordinary cooking temperatures. Therefore, it is reasonable to consider that the concentrations of these isomers are overestimated by coelution of small matrices at the same retention time, although evident matrices were not found on the chromatograms simultaneously monitored at the lock mass *m*/*z* from perfluorokerosene.

To statistically examine the relative significance of the different cooking protocols, identical control homogenates that cover all cooking trials should be prepared. In this study, however, we prepared different pairs of uncooked and cooked samples using fish as fillets or meat as slices, so that the universal effects of household cooking forms could be demon-

Table 7. Concentrations (pg/g)[a] of PCDD/Fs and Dioxin-like PCBs in Uncooked and Cooked Beef Samples (Boil)

	boil-1		boil-2		boil-3	
congener	uncooked	cooked	uncooked	cooked	uncooked	cooked
2,3,7,8-tetraCDD	0.0080	ND	0.013	ND	0.0090	0.0065
1,2,3,7,8-pentaCDD	ND	ND	ND	ND	0.090	0.067
1,2,3,4,7,8-hexaCDD	ND	ND	ND	ND	0.059	0.025
1,2,3,6,7,8-hexaCDD	0.015	ND	0.034	ND	0.16	0.11
1,2,3,7,8,9-hexaCDD	0.021	0.0081	0.013	ND	0.066	0.050
1,2,3,4,6,7,8-heptaCDD	0.078	0.048	0.12	0.078	0.56	0.43
octaCDD	0.29	0.19	0.34	0.25	2.4	1.6
2,3,7,8-tetraCDF	0.015	ND	0.010	ND	ND	ND
1,2,3,7,8-pentaCDF	0.018	0.0067	0.017	ND	ND	ND
2,3,4,7,8-pentaCDF	0.033	0.022	0.037	0.031	0.14	0.093
1,2,3,4,7,8-hexaCDF	0.028	0.012	0.035	0.028	0.11	0.082
1,2,3,6,7,8-hexaCDF	0.020	0.019	0.039	0.014	0.095	0.062
1,2,3,7,8,9-hexaCDF	ND	ND	ND	ND	ND	ND
2,3,4,6,7,8-hexaCDF	0.015	0.009	0.025	0.017	0.089	0.070
1,2,3,4,6,7,8-heptaCDF	0.028	0.015	0.039	0.029	0.13	0.086
1,2,3,4,7,8,9-heptaCDF	ND	ND	ND	ND	0.012	0.011
octaCDF	ND	ND	ND	ND	0.035	0.025
3,3′,4,4′-tetraCB (#77)	0.14	ND	0.12	0.078	0.14	0.086
3,4,4′,5-tetraCB (#81)	ND	ND	ND	ND	0.12	0.076
3,3′,4,4′,5-pentaCB (#126)	0.25	0.17	0.27	0.22	0.65	0.43
3,3′,4,4′,5,5′-hexaCB (#169)	ND	ND	ND	ND	0.26	0.19
2′,3,4,4′,5-pentaCB (#123)	ND	ND	ND	ND	ND	ND
2,3′,4,4′,5-pentaCB (#118)	22	18	47	29	49	34
2,3,4,4′,5-pentaCB (#114)	ND	ND	1.7	1.1	1.6	1.1
2,3,3′,4,4′-pentaCB (#105)	5.9	4.6	13	8.6	11	7.2
2,3′,4,4′,5,5′-hexaCB (#167)	1.3	1.0	3.4	2.0	3.5	2.3
2,3,3′,4,4′,5-hexaCB (#156)	2.3	1.9	5.5	3.1	5.6	4.0
2,3,3′,4,4′,5′-hexaCB (#157)	ND	ND	1.4	1.0	1.8	1.2
2,3,3′,4,4′,5,5′-heptaCB (#189)	ND	ND	ND	ND	ND	ND

[a] Concentrations in cooked samples were calculated on the basis of the original weight.

Table 8. Concentrations (pg/g)[a] of PCDD/Fs and Dioxin-like PCBs in Uncooked and Cooked Beef Samples (Broil)

	broil-1		broil-2		broil-3	
congener	uncooked	cooked	uncooked	cooked	uncooked	cooked
2,3,7,8-tetraCDD	0.0080	ND	0.013	0.0042	0.0090	0.0077
1,2,3,7,8-pentaCDD	ND	ND	ND	ND	0.090	0.076
1,2,3,4,7,8-hexaCDD	ND	ND	ND	ND	0.059	0.030
1,2,3,6,7,8-hexaCDD	0.015	0.012	0.034	0.029	0.16	0.13
1,2,3,7,8,9-hexaCDD	0.021	0.009	0.013	0.009	0.066	0.061
1,2,3,4,6,7,8-heptaCDD	0.078	0.057	0.12	0.075	0.56	0.47
octaCDD	0.29	0.206	0.34	0.275	2.4	2.0
2,3,7,8-tetraCDF	0.015	ND	0.010	0.008	ND	ND
1,2,3,7,8-pentaCDF	0.018	ND	0.017	0.005	ND	ND
2,3,4,7,8-pentaCDF	0.033	0.025	0.037	0.033	0.14	0.12
1,2,3,4,7,8-hexaCDF	0.028	ND	0.035	0.026	0.11	0.097
1,2,3,6,7,8-hexaCDF	0.020	ND	0.039	0.021	0.095	0.085
1,2,3,7,8,9-hexaCDF	ND	ND	ND	ND	ND	ND
2,3,4,6,7,8-hexaCDF	0.015	0.012	0.025	0.016	0.089	0.074
1,2,3,4,6,7,8-heptaCDF	0.028	0.023	0.039	0.029	0.13	0.097
1,2,3,4,7,8,9-heptaCDF	ND	ND	ND	ND	0.012	0.010
octaCDF	ND	ND	ND	ND	0.035	0.031
3,3′,4,4′-tetraCB (#77)	0.14	0.091	0.16	0.13	0.14	0.11
3,4,4′,5-tetraCB (#81)	ND	ND	ND	ND	0.12	0.086
3,3′,4,4′,5-pentaCB (#126)	0.25	0.16	0.27	0.21	0.65	0.53
3,3′,4,4′,5,5′-hexaCB (#169)	ND	ND	ND	ND	0.26	0.23
2′,3,4,4′,5-pentaCB (#123)	ND	ND	ND	ND	ND	ND
2,3′,4,4′,5-pentaCB (#118)	22	21	47	28	49	40
2,3,4,4′,5-pentaCB (#114)	ND	ND	1.7	1.1	1.6	1.4
2,3,3′,4,4′-pentaCB (#105)	5.9	5.7	13	8.4	11	9.0
2,3′,4,4′,5,5′-hexaCB (#167)	1.3	1.2	3.4	2.3	2.8	7.7
2,3,3′,4,4′,5-hexaCB (#156)	2.3	2.0	5.5	3.5	5.6	4.6
2,3,3′,4,4′,5′-hexaCB (#157)	ND	ND	1.4	1.0	1.8	1.4
2,3,3′,4,4′,5,5′-heptaCB (#189)	ND	ND	ND	ND	ND	ND

[a] Concentrations in cooked samples were calculated on the basis of the original weight.

strated. As a result, relatively large variations were observed in the cooking protocols due to application of the same cooking conditions to samples having different dioxin contents.

Our interest in this study was the toxicity changes brought about by household cooking on fish and meat samples, the toxicity of which makes up more than 90% of the average

Table 9. Concentrations (pg/g)[a] of PCDD/Fs and Dioxin-like PCBs in Uncooked and Cooked Beef Samples (Broil as Hamburger)

	hamburger-1		hamburger-2		hamburger-3	
congener	uncooked	cooked	uncooked	cooked	uncooked	cooked
2,3,7,8-tetraCDD	0.0080	ND	0.013	ND	0.0090	0.0082
1,2,3,7,8-pentaCDD	ND	ND	ND	ND	0.090	0.062
1,2,3,4,7,8-hexaCDD	ND	ND	ND	ND	0.059	0.021
1,2,3,6,7,8-hexaCDD	0.015	ND	0.034	ND	0.16	0.096
1,2,3,7,8,9-hexaCDD	0.021	ND	0.013	ND	0.066	0.063
1,2,3,4,6,7,8-heptaCDD	0.078	0.071	0.12	0.079	0.56	0.47
octaCDD	0.29	0.27	0.34	0.33	2.4	1.6
2,3,7,8-tetraCDF	0.015	0.015	0.010	ND	ND	ND
1,2,3,7,8-pentaCDF	0.018	ND	0.017	ND	ND	ND
2,3,4,7,8-pentaCDF	0.033	0.024	0.037	0.033	0.14	0.094
1,2,3,4,7,8-hexaCDF	0.028	0.019	0.035	ND	0.11	0.082
1,2,3,6,7,8-hexaCDF	0.020	0.018	0.039	ND	0.095	0.055
1,2,3,7,8,9-hexaCDF	ND	ND	ND	ND	ND	ND
2,3,4,6,7,8-hexaCDF	0.015	0.013	0.025	0.009	0.089	0.068
1,2,3,4,6,7,8-heptaCDF	0.028	0.026	0.039	0.021	0.13	0.078
1,2,3,4,7,8,9-heptaCDF	ND	ND	ND	ND	0.012	0.0091
octaCDF	ND	0.023	ND	ND	0.035	ND
3,3′,4,4′-tetraCB (#77)	0.14	0.11	0.12	0.12	0.14	0.097
3,4,4′,5-tetraCB (#81)	ND	ND	ND	ND	0.12	0.11
3,3′,4,4′,5-pentaCB (#126)	0.25	0.18	0.27	0.26	0.65	0.42
3,3′,4,4′,5,5′-hexaCB (#169)	ND	ND	ND	ND	0.26	0.16
2′,3,4,4′,5-pentaCB (#123)	ND	ND	ND	ND	ND	ND
2,3′,4,4′,5-pentaCB (#118)	22	20	47	27	49	35
2,3,4,4′,5-pentaCB (#114)	ND	ND	1.7	0.96	1.6	1.1
2,3,3′,4,4′-pentaCB (#105)	5.9	5.7	13	7.2	11	7.3
2,3′,4,4′,5,5′-hexaCB (#167)	1.3	1.4	3.4	1.7	3.5	2.6
2,3,3′,4,4′,5-hexaCB (#156)	2.3	2.2	5.5	3.0	5.6	3.9
2,3,3′,4,4′,5′-hexaCB (#157)	ND	ND	1.4	ND	1.8	1.2
2,3,3′,4,4′,5,5′-heptaCB (#189)	ND	ND	ND	ND	ND	ND

[a] Concentrations in cooked samples were calculated on the basis of the original weight.

Table 10. TEQ Concentrations (pg TEQ/g)[a] of PCDD/Fs and Dioxin-like PCBs in Uncooked and Cooked Beef Samples

	boil-1		boil-2		boil-3		
	uncooked	cooked	uncooked	cooked	uncooked	cooked	mean % decrease
PCDDs	0.015 (0.012)	0.013 (0.0013)	0.022 (0.019)	0.011 (0.00081)	0.13 (0.13)	0.10 (0.10)	31 (71)
PCDFs	0.039 (0.023)	0.015 (0.015)	0.031 (0.031)	0.022 (0.022)	0.24 (0.10)	0.069 (0.069)	54 (33)
dioxin-like PCBs	0.031 (0.029)	0.021 (0.020)	0.038 (0.037)	0.029 (0.028)	0.078 (0.078)	0.052 (0.052)	29 (29)
total dioxins	0.085 (0.065)	0.049 (0.036)	0.091 (0.087)	0.062 (0.051)	0.45 (0.31)	0.22 (0.22)	42 (39)
	broil-1		broil-2		broil-3		
	uncooked	cooked	uncooked	cooked	uncooked	cooked	mean % decrease
PCDDs	0.015 (0.012)	0.0067 (0.0027)	0.022 (0.019)	0.011 (0.0087)	0.13 (0.13)	0.11 (0.11)	41 (52)
PCDFs	0.039 (0.023)	0.015 (0.014)	0.031 (0.031)	0.024 (0.024)	0.24 (0.10)	0.088 (0.087)	49 (27)
dioxin-like PCBs	0.031 (0.029)	0.020 (0.020)	0.038 (0.037)	0.028 (0.027)	0.078 (0.078)	0.052 (0.064)	31 (27)
total dioxins	0.085 (0.065)	0.042 (0.036)	0.091 (0.087)	0.063 (0.060)	0.45 (0.32)	0.25 (0.26)	42 (32)
	hamburger-1		hamburger-2		hamburger-3		
	uncooked	cooked	uncooked	cooked	uncooked	cooked	mean % decrease
PCDDs	0.015 (0.012)	0.0061 (0.00074)	0.022 (0.019)	0.0060 (0.00082)	0.13 (0.13)	0.093 (0.093)	54 (73)
PCDFs	0.039 (0.023)	0.019 (0.019)	0.031 (0.031)	0.019 (0.018)	0.24 (0.10)	0.07 (0.069)	54 (32)
dioxin-like PCBs	0.031 (0.029)	0.023 (0.022)	0.038 (0.037)	0.032 (0.032)	0.078 (0.078)	0.052 (0.051)	25 (25)
total dioxins	0.085 (0.065)	0.048 (0.041)	0.091 (0.087)	0.057 (0.050)	0.45 (0.32)	0.21 (0.21)	44 (37)

[a] Concentrations in cooked samples were calculated on the basis of the original weight. TEQ concentrations were calculated and summed at ND = LOD/2, while calculations at ND = 0 are shown in parentheses.

dietary intake of dioxins in Japan (*1*, *2*). Generally, in dietary exposure study of hazardous chemicals including dioxins, the concentrations of nondetected congeners are replaced by ND = LOD/2 as well as ND = 0 and are subsequently converted to their TEQ concentrations (*1*−*5*). The validity of such procedures has also been demonstrated from a statistical viewpoint (*18*). Therefore, with the intention of maintaining a correspondence with these estimates, a conversion of concentrations into TEQ values was carried out.

As shown in **Table 10**, the estimated TEQ reductions differed from those obtained using the method of handling nondetects where large numbers of nondetects are found. With a view to enhancing the comparability of the pairs of control and cooked data, it might beneficial to use artificially contaminated meat, as described by Rose et al. (*19*). In conclusion, our results also suggest that ordinary cooking and heating can have significant effects, regardless of the form they take, as each cooking procedure examined in this study had the effect of reducing

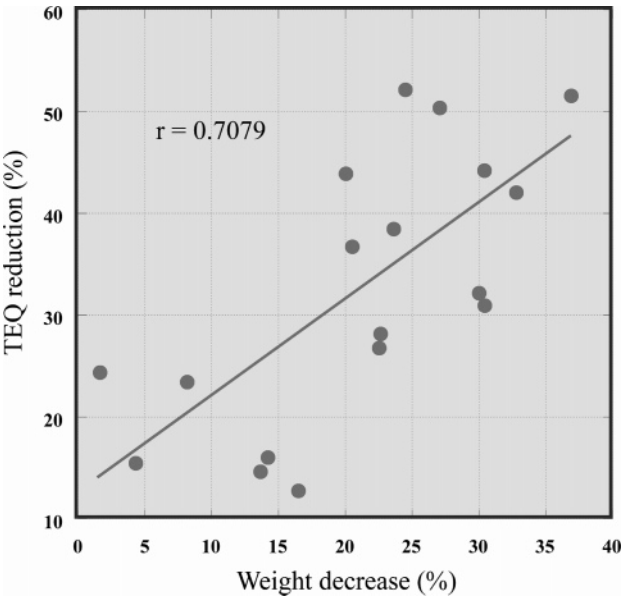

Figure 1. Relationship between weight decrease and TEQ reduction on fish and meat samples ($P = 0.001$).

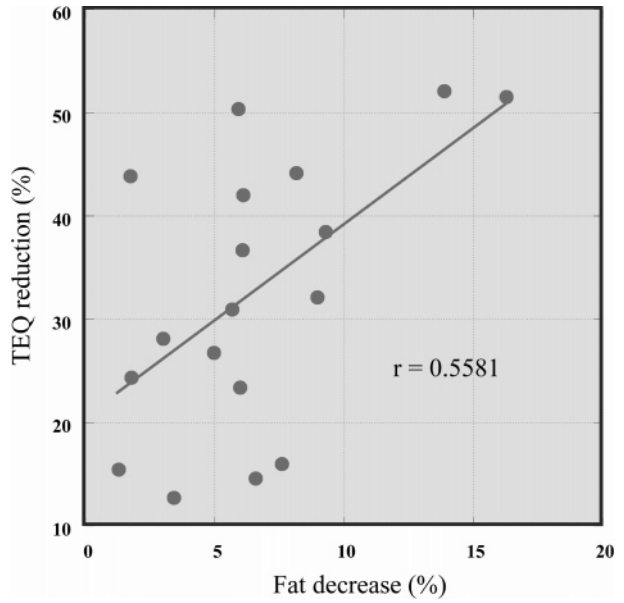

Figure 2. Relationship between fat decrease and TEQ reduction on fish and meat samples ($P = 0.044$).

levels of dioxin congeners in mackerel and beef. As shown in **Figure 1**, we observed relationships between the degree of weight change and that of TEQ reduction, similar to the findings of Schecter et al. as a result of cooking ground beef as hamburger (*20*). Furthermore, a slight relationship between the estimated fat release and the degree of TEQ reduction was also observed, indicating that fat and dioxins generally behave in the same way when they are released from fish and/or meat during heating (**Figure 2**). This observed behavior could be explained by the high lipophilicity of dioxin and related compound molecules: log K_{ow} for 2,3,7,8-tetraCDD is estimated to be 6.64 or 6.80 at 25 °C (*21*).

We can now successfully refer to the extensive body of data on the dioxin levels of Japanese farm products, which has been acquired in national investigations conducted by the Ministry of Health, Labor, and Welfare of Japan since 1996. One can estimate dietary intake from an item of fish or meat by simply combining the mean contamination (concentration) with average consumption data (fresh weight). However, such estimating without regard to the cooking-induced reductions will lead to overestimation to some extent: Our results suggest that an approximately 40% cooking-induced reduction might be considered in estimations regarding meat, which occurs exclusively due to heating. On the other hand, additional assumptions are needed with regard to cooking style, specifically as to whether it involves heating, since the Japanese often eat fish raw without heating, for example, eating slices as "sashimi". Moreover, in some cases in which dioxins are released with fat and/or moisture during heating processes and then reintroduced into the diet in sauces, soups, and broths; as such, the cooking-induced reduction as presented here would not be fully demonstrated in terms of the total diet. Recent total diet surveys suggest that the Japanese dietary intake of dioxins originates primarily in fish and shellfish (*1*, *2*), likely due to both fish containing relatively higher levels of dioxins than other animal products and the Japanese more often consuming fish than other animal products. It is also the case that dietary exposure to dioxins from fish is considered to not be serious for average dietary consumers. However, the cooking-induced reductions recognized in the present study would be worthwhile to consider, especially for heavy fish consumers wishing to decrease their risk of exposure to dioxins.

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