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Comparison of the Concentrations of Polychlorinated Dibenzo-*p*-dioxins, Polychlorinated Dibenzofurans, and Polychlorinated Biphenyls in the Blood of Yusho Patients Measured in 2004 with those Measured in 2014

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Abstract

We have been measuring the concentrations of PCDDs (polychlorinated dibenzo-*p*-dioxins), PCDFs (polychlorinated dibenzofurans), and dioxin-like PCBs (polychlorinated biphenyls) in blood collected from Yusho patients in medical health examinations since 2002. The present study extends our previous studies by reporting changes in the concentrations of individual congeners of PCDDs, PCDFs, and dioxin-like PCBs in the blood of Yusho patients from 2004 to 2014. Out of 243 and 246 Yusho patients who received medical health examinations in 2004 and 2014, respectively, there were 118 patients in whom the blood concentrations of these dioxin-like compounds were measured in both years. The concentrations of individual congeners of PCDDs, PCDFs, and PCBs in the blood of these 118 Yusho patients measured in 2004 were compared with those measured in 2014. The total toxicity equivalence (TEQ) concentrations of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs in the blood of 118 Yusho patients in 2004 and 2014 were 5.2–533 (mean : 80, median : 52) and 11–545 (mean : 78, median : 73) pg TEQ g⁻¹ lipid, respectively. Among the individual congeners of PCDDs, PCDFs, and dioxin-like PCBs, most congeners did not significantly decrease from 2004 to 2014. Though the concentrations of 1,2,3,6,7,8-hexaCDD, 2,3,4,7,8-pentaCDF, 1,2,3,4,7,8-hexaCDF, and 1,2,3,6,7,8-hexaCDF—all considered characteristic congeners in the blood of Yusho patients—decreased significantly from 2004 to 2014, the decreasing ratios of these congeners tended to decrease slightly. These findings suggest that the half-lives of individual congener concentrations of PCDDs, PCDFs and dioxin-like PCBs in the blood are proving to be long to near infinity in the majority of Yusho patients.

Keywords : Polychlorinated dibenzo-*p*-dioxins, Polychlorinated dibenzofurans, Polychlorinated biphenyls, Human blood, Yusho

Introduction

The 1968 Yusho poisoning accident affected

over 1,800 people in western Japan, and was caused by the accidental ingestion of rice bran oil containing PCBs, PCDFs, PCDDs, polychlorinated

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quarterphenyls (PCQs), and polychlorinated terphenyls (PCTs)¹⁾. Since the Yusho outbreak, the National Study Group for the Therapy of Yusho has carried out medical care and health examinations of the affected population. In 2001, technological advances made it possible to measure PCDDs, PCDFs, and non-*ortho* PCBs in small amounts of blood^{2)~4)}. We have measured the concentrations of PCDDs, PCDFs, and dioxin-like PCBs in the blood collected from Yusho patients in medical health examinations since 2002^{5)~7)}. Moreover, we have conducted a congener-specific analysis of non-dioxin-like PCBs in the blood of these patients since 2004^{8)~10)}. Based on these results, 2,3,4,7,8-pentaCDF has been recognized as the most important causative agent for the subjective symptoms of Yusho¹¹⁾. We also reported that Yusho patients continue to have higher concentrations of PCDFs in their blood than unaffected people, and that the concentration of PCDFs in the blood is significantly correlated with the intensity of Yusho symptoms¹²⁾¹³⁾. To provide useful information related to the health risks of PCDDs, PCDFs, and dioxin-like PCBs in Yusho patients, we previously reported on changes in the individual congener concentrations of PCDDs, PCDFs, and dioxin-like PCBs in the blood of Yusho patients¹⁴⁾. The objective of this study was to extend our previous studies.

Out of 243 and 246 Yusho patients who received medical health examinations in 2004 and 2014, respectively, there were 118 patients in whom the blood concentrations of PCDDs, PCDFs, and PCBs were measured both years. In these 118 Yusho patients, we compared the individual congener concentrations of PCDDs, PCDFs, and dioxin-like PCBs in the blood measured in 2004 with those measured in 2014.

Materials and Methods

1. Sampling

Medical health examinations have been performed annually on Yusho patients to determine their health status since the Yusho incident. The

medical health examination is open not only to those persons officially registered as Yusho patients but also to Yusho-suspected persons who regard themselves as potential victims. Both officially registered Yusho patients and Yusho-suspected persons are examined based on the "Diagnostic Criteria for Yusho"¹¹⁾. The blood samples examined in this study were collected from 243 and 246 participants who received medical health examinations in 2004 and 2014, respectively, each of whom gave informed consent to participate in this study. Blood samples of 10 ml were collected using a vacuum blood-collecting tube containing heparin and were stored at 4°C until analyses for the concentrations of PCDDs, PCDFs, and PCBs.

2. Materials

Native congeners of PCDDs, PCDFs, dioxin-like PCBs, and non-dioxin-like PCBs were purchased from Wellington Laboratories (Guelph, Canada). [¹³C₁₂] – congeners of PCDDs, PCDFs, dioxin-like PCBs, and non-dioxin-like PCBs as internal standards were also purchased from Wellington Laboratories. Active carbon columns were prepared as follows : active carbon was purchased from Nacalai Tesque (Kyoto, Japan), refluxed with toluene for 1 hour 3 times, and dried in vacuum, after which 500 mg of the active carbon was mixed with 500 g of anhydrous sodium sulfate (Wako Pure Chemical Industries, Ltd., Tokyo, Japan). A silver nitrate/silica gel was purchased from Wako Pure Chemical Industries, Ltd. All reagents and solvents used in this experiment were of the analytic grade of dioxin that is commercially available.

3. Analysis of PCDDs, PCDFs, dioxin-like PCBs, and non-dioxin-like PCBs

The extraction and purification of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs from blood samples were performed using a previously reported method²⁾⁸⁾. Concentrations of PCDDs, PCDFs, and dioxin-like PCBs and concen-

trations of 58 non-dioxin-like PCB congeners were determined by a previously reported method²⁾⁸⁾.

4. Quality control

To evaluate the accuracy and reliability of the analysis of PCDDs, PCDFs, dioxin-like PCBs, and non-dioxin-like PCBs, our laboratory participated quality control studies of the analysis of PCDDs, PCDFs, and dioxin-like PCBs in 2013, 2015, 2017, and 2019 and non-dioxin-like PCBs in 2014, 2016, and 2018. Each quality control study involved the participation of various laboratories that perform measurements for these compounds in human blood in Japan. In each quality control study, our results were compared with those of participating laboratories, and tests confirmed that the average variation among values obtained by each organization performing the analysis was within 10%. These results indicated that our laboratory's analytical methods regarding PCDDs, PCDFs, dioxin-like PCBs, and non-dioxin-like PCBs in human blood provided accurate results.

5. Data analysis

To estimate the toxicity equivalence (TEQ) concentrations, we introduced ND (less than the detection limit) values to half values of the detection limit and calculated based on the toxic equivalency factor (TEF) values proposed by the WHO¹⁵⁾. Statistical analysis was conducted using the Wilcoxon signed-rank test in the software programs from IBM SPSS Statistics 24 (Advanced Analytics, Inc). Significant probabilities (p values) were calculated for the respective number of samples analyzed.

Results and Discussion

The objective of the present study was to extend our previous studies by reporting the changes in the individual congener concentrations of PCDDs, PCDFs, and dioxin-like PCBs in the blood of Yusho patients from 2004 to 2014¹⁴⁾. Out of 243 and 246 Yusho patients who received

medical health examinations in 2004 and 2014, respectively, there were 118 patients in whom the blood concentrations of PCDDs, PCDFs, and dioxin-like PCBs were measured both years. In these 118 Yusho patients, the individual congener concentrations of PCDDs, PCDFs, and dioxin-like PCBs in the blood samples measured in 2004 were compared with those measured in 2014 (Table 1). The total TEQ concentrations of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs in the blood of 118 Yusho patients in 2004 and 2014 were 5.2–533 (mean : 80, median : 52) and 11–545 (mean : 78, median : 73) pg TEQ g⁻¹ lipid, respectively. The TEQ concentrations of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs in the blood of Yusho patients were 16, 51, 12, and 1.7 pg TEQ g⁻¹ lipid in 2004, respectively, and 16, 45, 14, and 2.0 pg TEQ g⁻¹ lipid in 2014, respectively, indicating that the concentrations of PCDFs in the blood of Yusho patients significantly decreased from 2004 to 2014 ($p < 0.001$).

We previously reported that the concentrations of 1,2,3,6,7,8-hexaCDD, 2,3,4,7,8-pentaCDF, 1,2,3,4,7,8-hexaCDF, and 1,2,3,6,7,8-hexaCDF in the blood of Yusho patients were higher than those of normal controls⁸⁾. These can be considered the characteristic congeners in the blood of Yusho patients. Of these four congeners, the concentrations of 1,2,3,6,7,8-hexaCDD, 2,3,4,7,8-pentaCDF, 1,2,3,4,7,8-hexaCDF, and 1,2,3,6,7,8-hexaCDF were 46, 151, 37, and 15 pg g⁻¹ lipid in 2004, respectively, and 41, 137, 25, and 12 pg g⁻¹ lipid in 2014, respectively, indicating that these congeners decreased significantly from 2004 to 2014 ($p < 0.001$). In addition, the concentrations of 1,2,3,7,8,9-hexaCDD, 1,2,3,4,6,7,8-heptaCDD, octaCDD, and 1,2,3,4,6,7,8-heptaCDF showed a trend of decrease from 2004 to 2014 ($p < 0.001$). However, among individual congeners of dioxin-like PCBs, most congeners did not significantly decrease from 2004 to 2014.

Among the 118 patients, the blood concentrations of 2,3,4,7,8-pentaCDF were under

Table 1 Concentrations of PCDDs, PCDFs, and dioxin-like PCBs in the blood of 118 Yusho patients

Congeners	Concentration (pg/g lipid)										Ratio	
	2004					2014						
	Mean	Median	SD	Maximum	Minimum	Mean	Median	SD	Maximum	Minimum	2014/2004	<i>p</i> Values
2,3,7,8-TetraCDD	1.3	1.3	0.7	4.4	0.5	1.6	0.9	1.5	4.2	0.5	1.2	< 0.001
1,2,3,7,8-PentaCDD	8.7	8.1	4.3	33	1.0	9.6	5.4	8.9	41	1.9	1.1	0.001
1,2,3,4,7,8-HexaCDD	2.5	2.4	1.5	8.3	1.0	2.2	1.6	2.1	12	1.0	0.9	0.011
1,2,3,6,7,8-HexaCDD	46	37	35	247	4.8	41	35	29	248	3.0	0.9	< 0.001
1,2,3,7,8,9-HexaCDD	4.3	3.3	3.4	23	1.0	3.2	3.0	2.6	18	1.0	0.7	< 0.001
1,2,3,4,6,7,8-HeptaCDD	49	44	25	196	16	37	24	33	208	11	0.8	< 0.001
OctaCDD	714	624	359	2,305	181	511	292	424	1,779	95	0.7	< 0.001
Total PCDDs	825	741	393	2,459	205	605	318	517	1,930	138	0.7	< 0.001
2,3,7,8-TetraCDF	1.6	1.3	1.3	7.2	0.5	2.6	5.5	1.4	43	0.5	1.6	0.329
1,2,3,7,8-PentaCDF	0.8	0.5	0.7	4.1	0.5	1.1	1.1	0.5	5.6	0.5	1.4	0.002
2,3,4,7,8-PentaCDF	151	79	192	1,240	4.1	137	180	67	1,261	5.8	0.9	< 0.001
1,2,3,4,7,8-HexaCDF	37	14	63	514	1.0	25	48	8.6	402	1.0	0.7	< 0.001
1,2,3,6,7,8-HexaCDF	15	8.2	20	176	1.0	12	19	6.9	174	1.0	0.8	< 0.001
2,3,4,6,7,8-HexaCDF	ND					ND						
1,2,3,7,8,9-HexaCDF	ND					ND						
1,2,3,4,6,7,8-HeptaCDF	2.5	1.5	2.6	24	1.0	1.8	2.3	1.0	21	1.0	0.7	< 0.001
1,2,3,4,7,8,9-HeptaCDF	ND					ND						
OctaCDF	ND					ND						
Total PCDFs	213	109	273	1,946	13	186	244	101	1,875	16	0.9	< 0.001
TriCB-77	ND					ND						
TriCB-81	ND					ND						
PentaCB-126	82	66	62	441	5.0	91	65	77	428	17	1.1	< 0.001
PentaCB-169	121	105	79	361	11	180	124	149	677	23	1.5	< 0.001
Total Non- <i>ortho</i> PCBs	219	193	120	696	26	283	161	254	856	50	1.3	< 0.001
PentaCB-105	3,111	2,239	2,425	15,888	563	3,197	2,503	2,654	19,926	606	1.0	0.134
PentaCB-114	1,721	1,339	1,363	8,660	208	2,033	1,712	1,622	10,177	234	1.2	< 0.001
PentaCB-118	14,872	11,534	11,468	80,220	2,355	16,238	14,513	12,544	133,844	2,853	1.1	0.005
PentaCB-123	265	195	220	1,385	5.0	249	223	196	1,502	16	0.9	0.150
HexaCB-156	23,217	17,570	19,537	90,316	5.0	28,518	27,967	19,752	175,909	1,961	1.2	< 0.001
HexaCB-157	6,639	4,980	5,510	25,277	351	7,332	7,635	5,311	47,932	318	1.1	0.050
HexaCB-167	3,057	2,360	2,108	13,497	483	3,957	2,838	3,318	20,467	541	1.3	< 0.001
HeptaCB-189	3,402	2,629	2,720	11,402	5.0	4,276	3,843	3,261	23,833	137	1.3	< 0.001
Total Mono- <i>ortho</i> PCBs	56,283	45,958	34,608	159,176	5,971	65,802	47,855	56,430	268,679	6,767	1.2	< 0.001
TEQ from PCDDs	16	14	8.3	63	2.4	16	9.2	15	72	3.7	1.0	0.288
TEQ from PCDFs	51	26	66	442	1.7	45	60	23	437	2.7	0.9	< 0.001
TEQ from PCDDs/PCDFs	67	40	72	505	4.1	62	68	38	509	6.4	0.9	< 0.001
TEQ from non- <i>ortho</i> PCBs	12	10	7.4	50	0.8	14	8.6	13	52	2.4	1.2	< 0.001
TEQ from mono- <i>ortho</i> PCBs	1.7	1.4	1.0	4.8	0.2	2.0	1.4	1.7	8.1	0.2	1.2	< 0.001
TEQ from dioxin-like PCBs	14	12	8.2	54	1.0	16	9.7	15	57	2.6	1.2	< 0.001
Total TEQ	80	52	75	533	5.2	78	73	54	545	11	1.0	0.288

ND (less than the determination limit) values introduced to half values of the detection limit.

SD : standard deviation.

CDD : chlorinated dibenzo-*p*-dioxin.

CDF : chlorinated dibenzofuran.

CB : chlorinated biphenyl.

100 pg g⁻¹ lipid in 67 patients and over 100 pg g⁻¹ lipid in 51 patients. In the 51 over 100 pg g⁻¹ lipid group, the arithmetic mean TEQ concentrations of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs in the blood were 21, 101, 13, and 2.2 pg TEQ g⁻¹ lipid in 2004, respectively, and 22, 90, 17, and 2.7 pg TEQ g⁻¹ lipid in 2014, respectively, with the total TEQ concentrations of these dioxin-like compounds ranging from 50 to 533 (mean : 137, median : 84) and 47 to 545 (mean : 132, median : 83) pg TEQ g⁻¹ lipid in 2004 and 2014, respectively (Table 2). With respect to the characteristic congeners in the blood of Yusho patients, the concentrations of 1,2,3,6,7,8-hexaCDD, 2,3,4,7,8-pentaCDF, 1,2,3,4,7,8-hexaCDF, and 1,2,3,6,7,8-hexaCDF were found to decrease slightly from year to year. These results in the high-2,3,4,7,8-pentaCDF patients were almost the same as those in the total 118 Yusho patients.

Of the 118 Yusho patients, 61 were men and 57 were women. The arithmetic mean TEQ concentrations of PCDDs, PCDFs, non-*ortho* PCBs, and mono-*ortho* PCBs in the blood of the 61 men were 5.2–252 (mean : 60, median : 45) in 2004 and 11–249 (mean : 58, median : 43) pg TEQ g⁻¹ lipid in 2014 (Table 3). The concentrations in the 57 women were 12–533 (mean : 102, median : 93) in 2004 and 15–545 (mean : 100, median : 91) pg TEQ g⁻¹ lipid in 2014, indicating that the total TEQ concentrations in women in 2004 and 2014 were significantly higher than those in men (Table 4). Regarding the characteristic congeners in the blood of Yusho patients, the concentrations of 1,2,3,6,7,8-hexaCDD, 2,3,4,7,8-pentaCDF, 1,2,3,4,7,8-hexaCDF, and 1,2,3,6,7,8-hexaCDF showed a trend of decrease from 2004 to 2014 in both men and women. The decreasing ratios of individual congener concentrations of PCDDs, PCDFs, and dioxin-like PCBs from 2004 to 2014 in women were almost the same as those in men, suggesting that there is probably no sex difference regarding the enzyme that catalyzes the metabolism of dioxin-like compounds in

humans.

According to the results of the present study, among the individual congeners of PCDDs, PCDFs, and PCBs, most congeners of these compounds did not significantly decrease from 2004 to 2014. However, the concentrations of 1,2,3,6,7,8-hexaCDD, 2,3,4,7,8-pentaCDF, 1,2,3,4,7,8-hexaCDF, and 1,2,3,6,7,8-hexaCDF in the blood of Yusho patients significantly decreased from 2004 to 2014. In addition, the concentrations of 1,2,3,7,8,9-hexaCDD, 1,2,3,4,6,7,8-heptaCDD, octaCDD, and 1,2,3,4,6,7,8-heptaCDF were also showed a trend of decrease from 2004 to 2014.

Although over 50 years have passed since the outbreak of Yusho, many patients still suffer various symptoms such as chloracne, general fatigue and neuropathy. There are patients who continue to have much higher concentrations of PCDDs, PCDFs, and dioxin-like PCBs in their blood than unaffected persons. The investigations conducted in the present study suggest that the PCDDs, PCDFs, and dioxin-like PCBs that have remained in the bodies of Yusho patients are very difficult to excrete from the body, and that the half-lives of individual congener concentrations of these dioxin like-compounds in the blood are proving to be long to near infinity in the majority of Yusho patients.

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Table 2 Concentrations of PCDDs, PCDFs, and dioxin-like PCBs in the blood of 51 Yusho patients

Congeners	Concentration (pg/g lipid)										Ratio	
	2004					2014						
	Mean	Median	SD	Maximum	Minimum	Mean	Median	SD	Maximum	Minimum	2014/2004	<i>p</i> Values
2,3,7,8-TetraCDD	1.4	0.8	1.3	4.4	0.5	1.8	0.8	1.7	4.0	0.5	1.2	0.003
1,2,3,7,8-PentaCDD	11	5	11	33	4.1	13	6.1	11	41	5.0	1.1	0.010
1,2,3,4,7,8-HexaCDD	2.67	1.33	2.61	6.74	1.00	2.4	1.4	2.3	6.3	1.0	0.9	0.042
1,2,3,6,7,8-HexaCDD	69	39	61	247	23	64	40	49	248	17	0.9	0.001
1,2,3,7,8,9-HexaCDD	5.0	4.1	3.6	23	1.0	3.9	3.5	3.0	18	1.0	0.8	< 0.001
1,2,3,4,6,7,8-HeptaCDD	48	19	44	98	16	35	14	33	84	13	0.7	< 0.001
OctaCDD	687	297	612	1,760	265	542	254	493	1,317	107	0.8	< 0.001
Total PCDDs	825	330	738	1,973	324	662	269	653	1,463	161	0.8	< 0.001
2,3,7,8-TetraCDF	2.22	1.64	1.92	7.20	0.50	2.3	2.5	1.8	16	0.5	1.1	0.687
1,2,3,7,8-PentaCDF	0.95	0.69	0.50	3.10	0.50	1.1	1.0	0.5	4.8	0.5	1.2	0.256
2,3,4,7,8-PentaCDF	301	211	231	1,240	106	273	203	211	1,261	87	0.9	< 0.001
1,2,3,4,7,8-HexaCDF	76	81	49	514	14	52	64	29	402	3.7	0.7	< 0.001
1,2,3,6,7,8-HexaCDF	27	26	20	176	6.6	23	26	15	174	2.8	0.8	< 0.001
2,3,4,6,7,8-HexaCDF	ND					ND						
1,2,3,7,8,9-HexaCDF	ND					ND						
1,2,3,4,6,7,8-HeptaCDF	2.5	1.8	2.0	8.4	1.0	2.0	2.9	1.0	21	1.0	0.8	0.002
1,2,3,4,7,8,9-HeptaCDF	ND					ND						
OctaCDF	ND					ND						
Total PCDFs	415	314	333	1,946	135	359	290	262	1,875	112	0.9	< 0.001
TriCB-77	ND					ND						
TriCB-81	ND					ND						
PentaCB-126	78	55	64	354	27	92	56	79	323	24	1.2	< 0.001
PentaCB-169	174	78	156	361	49	264	128	241	677	79	1.5	< 0.001
Total Non- <i>ortho</i> PCBs	268	119	256	696	91	367	161	333	856	132	1.4	< 0.001
PentaCB-105	2,761	1,988	2,180	12,894	941	2,977	1,665	2,607	9,721	909	1.1	0.694
PentaCB-114	2,516	1,493	1,964	8,660	848	3,065	1,769	2,474	10,177	837	1.2	0.001
PentaCB-118	13,441	9,333	10,335	59,893	4,613	15,409	8,394	12,698	47,970	4,863	1.1	< 0.001
PentaCB-123	223	198	180	1,268	5.0	219	149	168	827	64	1.0	0.023
HexaCB-156	36,739	20,764	31,628	90,316	7,320	46,431	32,682	38,547	175,909	9,250	1.3	< 0.001
HexaCB-157	10,130	5,947	8,477	25,277	1,996	12,335	8,943	10,501	47,932	2,437	1.2	< 0.001
HexaCB-167	3,444	2,031	2,626	10,040	1,002	4,467	2,038	3,956	10,483	1,655	1.3	0.001
HeptaCB-189	5,035	2,718	4,422	11,402	960	6,539	4,243	5,543	23,833	1,348	1.3	< 0.001
Total Mono- <i>ortho</i> PCBs	74,289	35,228	68,154	159,176	22,680	91,442	49,916	79,465	268,679	28,912	1.2	< 0.001
TEQ from PCDDs	21	9.1	20	63	7.5	22	10	20	72	8.0	1.0	0.238
TEQ from PCDFs	101	73	79	442	34	90	69	67	437	28	0.9	< 0.001
TEQ from PCDDs/PCDFs	122	81	107	505	44	112	78	87	509	38	0.9	< 0.001
TEQ from non- <i>ortho</i> PCBs	13	6.8	12	43	4.7	17	8.0	16	41	6.2	1.3	< 0.001
TEQ from mono- <i>ortho</i> PCBs	2.2	1.1	2.0	4.8	0.7	2.7	1.5	2.4	8.1	0.9	1.2	< 0.001
TEQ from dioxin-like PCBs	15	7.7	14	48	5.5	20	9.2	18	45	7.5	1.3	< 0.001
Total TEQ	137	84	119	533	50	132	83	113	545	47	1.0	0.023

ND (less than the determination limit) values introduced to half values of the detection limit.

SD : standard deviation.

CDD : chlorinated dibenzo-*p*-dioxin.

CDF : chlorinated dibenzofuran.

CB : chlorinated biphenyl.

Table 3 Concentrations of PCDDs, PCDFs, and dioxin-like PCBs in the blood of 61 men

Congeners	Concentration (pg/g lipid)										Ratio	
	2004					2014						
	Mean	Median	SD	Maximum	Minimum	Mean	Median	SD	Maximum	Minimum	2014/2004	<i>p</i> Values
2,3,7,8-TetraCDD	1.3	0.8	1.3	4.4	0.5	1.6	0.8	1.5	3.8	0.5	1.2	0.004
1,2,3,7,8-PentaCDD	7.8	3.4	7.7	18	1.0	8.4	4.0	7.9	21	1.9	1.1	0.018
1,2,3,4,7,8-HexaCDD	2.5	1.6	2.4	8.3	1.0	2.3	1.8	2.1	12	1.0	0.9	0.267
1,2,3,6,7,8-HexaCDD	40	28	34	132	4.8	33	26	25	115	3.0	0.8	< 0.001
1,2,3,7,8,9-HexaCDD	4.0	3.1	3.1	19	1.0	2.7	2.6	2.1	17	1.0	0.7	< 0.001
1,2,3,4,6,7,8-HeptaCDD	48	28	41	196	16	40	31	34	208	11	0.8	0.001
OctaCDD	705	376	594	1,811	181	531	335	422	1,779	107	0.8	< 0.001
Total PCDDs	809	418	712	2,135	205	619	369	504	1,930	161	0.8	< 0.001
2,3,7,8-TetraCDF	1.6	1.2	1.3	7.2	0.5	3.0	5.5	1.3	28	0.5	1.8	0.178
1,2,3,7,8-PentaCDF	0.8	0.7	0.5	4.1	0.5	1.3	1.3	0.5	5.0	0.5	1.6	0.003
2,3,4,7,8-PentaCDF	92	107	55	541	4.1	81	93	50	479	5.8	0.9	< 0.001
1,2,3,4,7,8-HexaCDF	20	33	10	234	1.0	12	22	6	164	1.0	0.6	< 0.001
1,2,3,6,7,8-HexaCDF	10	12	7.2	85	1.0	8.1	9.5	5.6	70	1.0	0.8	< 0.001
2,3,4,6,7,8-HexaCDF	ND					ND						
1,2,3,7,8,9-HexaCDF	ND					ND						
1,2,3,4,6,7,8-HeptaCDF	2.8	3.4	2.0	24	1.0	1.8	1.7	1.0	12	1.0	0.7	0.001
1,2,3,4,7,8,9-HeptaCDF	ND					ND						
OctaCDF	ND					ND						
Total PCDFs	133	150	78	871	13	113	122	75	725	16	0.8	< 0.001
TriCB-77	ND					ND						
TriCB-81	ND					ND						
PentaCB-126	91	78	69	441	5.0	96	77	80	428	17	1.1	0.180
PentaCB-169	118	73	107	361	11	169	116	146	677	23	1.4	< 0.001
Total Non- <i>ortho</i> PCBs	225	137	182	696	26	278	171	237	856	50	1.2	< 0.001
PentaCB-105	3,424	2,733	2,890	15,888	563	3,290	2,355	2,709	12,923	606	1.0	0.028
PentaCB-114	1,378	997	1,101	5,098	208	1,505	1,181	1,269	6,021	244	1.1	0.717
PentaCB-118	15,612	11,372	13,364	62,223	2,355	15,995	11,383	12,318	62,596	2,853	1.0	0.053
PentaCB-123	289	244	231	1,385	5.0	255	227	202	1,484	17	0.9	0.558
HexaCB-156	20,945	17,096	16,866	90,316	393	24,609	24,675	18,604	161,802	1,961	1.2	< 0.001
HexaCB-157	5,927	4,867	4,691	23,883	351	6,062	6,562	4,545	43,561	318	1.0	0.006
HexaCB-167	2,988	2,269	2,323	13,497	483	3,765	2,768	3,327	18,126	541	1.3	0.957
HeptaCB-189	3,352	2,654	2,654	11,402	311	4,080	3,898	2,940	23,833	137	1.2	0.001
Total Mono- <i>ortho</i> PCBs	53,915	34,349	44,754	159,176	5,971	59,560	43,870	48,702	268,340	6,767	1.1	0.026
TEQ from PCDDs	14	6.8	13	33	2.4	14	7.0	13	37	3.7	1.0	0.838
TEQ from PCDFs	31	36	18	195	1.7	27	31	16	168	2.7	0.9	< 0.001
TEQ from PCDDs/PCDFs	46	41	33	225	4.1	41	36	33	204	6.4	0.9	< 0.001
TEQ from non- <i>ortho</i> PCBs	13	9.1	10	50	0.8	15	10	13	52	2.4	1.2	< 0.001
TEQ from mono- <i>ortho</i> PCBs	1.6	1.0	1.3	4.8	0.2	1.8	1.3	1.5	8.1	0.2	1.1	0.026
TEQ from dioxin-like PCBs	14	10	12	54	1.0	16	11	14	57	2.6	1.2	< 0.001
Total TEQ	60	45	49	252	5.2	58	43	48	249	11	1.0	0.200

ND (less than the determination limit) values introduced to half values of the detection limit.

SD : standard deviation.

CDD : chlorinated dibenzo-*p*-dioxin.

CDF : chlorinated dibenzofuran.

CB : chlorinated biphenyl.

Table 4 Concentrations of PCDDs, PCDFs, and dioxin-like PCBs in the blood of 57 women

Congeners	Concentration (pg/g lipid)										Ratio	
	2004					2014						
	Mean	Median	SD	Maximum	Minimum	Mean	Median	SD	Maximum	Minimum	2014/2004	<i>p</i> Values
2,3,7,8-TetraCDD	1.3	0.6	1.3	3.1	0.5	1.5	0.9	1.5	4.2	0.5	1.2	0.006
1,2,3,7,8-PentaCDD	9.7	5.0	8.3	33	3.3	11	6.3	9.7	41	2.8	1.1	0.014
1,2,3,4,7,8-HexaCDD	2.5	1.3	2.4	6.8	1.0	2.1	1.3	2.2	6.3	1.0	0.8	0.013
1,2,3,6,7,8-HexaCDD	52	40	43	247	6.4	49	41	40	248	8.1	0.9	0.010
1,2,3,7,8,9-HexaCDD	4.6	3.8	3.8	23	1.0	3.7	3.4	2.9	18	1.0	0.8	< 0.001
1,2,3,4,6,7,8-HeptaCDD	50	20	47	104	16	34	14	30	80	12	0.7	< 0.001
OctaCDD	722	341	630	2,305	242	489	234	424	1,251	95	0.7	< 0.001
Total PCDDs	843	364	762	2,459	306	590	253	568	1,366	138	0.7	< 0.001
2,3,7,8-TetraCDF	1.7	1.4	1.3	5.8	0.5	2.3	5.6	1.5	43	0.5	1.4	0.925
1,2,3,7,8-PentaCDF	0.8	0.6	0.5	2.9	0.5	0.9	0.8	0.5	5.6	0.5	1.2	0.400
2,3,4,7,8-PentaCDF	213	238	136	1,240	5.3	197	225	136	1261	8.5	0.9	0.034
1,2,3,4,7,8-HexaCDF	55	80	29	514	1.0	39	62	17	402	2.2	0.7	< 0.001
1,2,3,6,7,8-HexaCDF	20	25	11	176	1.0	17	25	9.3	174	1.0	0.9	< 0.001
2,3,4,6,7,8-HexaCDF	ND					ND						
1,2,3,7,8,9-HexaCDF	ND					ND						
1,2,3,4,6,7,8-HeptaCDF	2.2	1.4	1.0	5.8	1.0	1.8	2.8	1.0	21	1.0	0.8	0.003
1,2,3,4,7,8,9-HeptaCDF	ND					ND						
OctaCDF	ND					ND						
Total PCDFs	298	340	178	1,946	14	264	309	166	1,875	20	0.9	< 0.001
TriCB-77	ND					ND						
TriCB-81	ND					ND						
PentaCB-126	72	37	64	157	25	85	47	76	211	22	1.2	< 0.001
PentaCB-169	124	84	104	361	23	191	130	164	588	36	1.5	< 0.001
Total Non- <i>ortho</i> PCBs	211	98	198	493	61	287	150	268	712	69	1.4	< 0.001
PentaCB-105	2,776	1,989	2,180	13,355	843	3,098	2,649	2,607	19,926	773	1.1	0.896
PentaCB-114	2,087	1,588	1,736	8,660	272	2,599	1,989	2,034	10,177	234	1.2	< 0.001
PentaCB-118	14,081	11,517	10,573	80,220	4,180	16,499	17,241	12,698	133,844	4521	1.2	< 0.001
PentaCB-123	239	187	187	1,178	5.0	243	219	189	1,502	16	1.0	0.008
HexaCB-156	25,647	21,586	18,613	83,648	5.0	32,702	30,557	26,711	175,909	3,292	1.3	< 0.001
HexaCB-157	7,400	6,031	5,596	25,277	770	8,691	8,427	6,797	47,932	694	1.2	< 0.001
HexaCB-167	3,132	1,917	2,376	9,186	736	4,162	2,897	3,251	20,467	982	1.3	0.005
HeptaCB-189	3,455	2,788	2,615	11,211	5.0	4,487	3,773	3,736	19,231	460	1.3	< 0.001
Total Mono- <i>ortho</i> PCBs	58,818	34,705	51,520	158,563	13,428	72,481	50,941	62,139	268,679	14,193	1.2	< 0.001
TEQ from PCDDs	18	9.4	16	63	6.2	18	11	16	72	4.7	1.0	0.214
TEQ from PCDFs	72	82	45	442	2.1	65	76	43	437	3.3	0.9	0.001
TEQ from PCDDs/PCDFs	89	90	60	505	8.3	84	85	61	509	9.4	0.9	0.030
TEQ from non- <i>ortho</i> PCBs	11	4.8	11	23	3.5	14	6.9	13	30	3.3	1.3	< 0.001
TEQ from mono- <i>ortho</i> PCBs	1.8	1.0	1.5	4.8	0.4	2.2	1.5	1.9	8.1	0.4	1.2	< 0.001
TEQ from dioxin-like PCBs	13	6	12	28	3.9	16	8.1	15	36	3.8	1.3	< 0.001
Total TEQ	102	93	70	533	12	100	91	73	545	15	1.0	0.790

ND (less than the determination limit) values introduced to half values of the detection limit.

SD : standard deviation.

CDD : chlorinated dibenzo-*p*-dioxin.

CDF : chlorinated dibenzofuran.

CB : chlorinated biphenyl.

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平成 16 および平成 26 年度に測定した油症患者血液中 ダイオキシン類濃度の比較

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平成 16 および平成 26 年度, その両年度に油症検診を受診した 118 名の血液中ダイオキシン類濃度の比較を行い, 各異性体の濃度推移を調べた. 今回調査した異性体の中で, 最も濃度が減少していたのは 1,2,3,4,7,8-hexaCDF で, 約 30%の濃度低下が認められた. 油症の主要原因物質である 2,3,4,7,8-pentaCDF は約 10%濃度が減少していた. 男女間での比較では, 女性の方が高い値を示したが, 10 年間の濃度推移に関しては, 男女間での相違は認められなかった.

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