EFFECTS OF ORGANOCHLORINE PESTICIDES, PCDDs, PCDFs, PCBs, TEQ and PBDEs IN "FEBRA" EXPOSED JAPANESE DURING TWO YEAR INVESTIGATION

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Introduction

Organochlorine pollutants (OCPs) are known for their contamination of the global environment, bio-magnification in the food chain, and long-term health effects in wildlife and humans. Among the OCPs used, DDTs, HCHs, chlordane compounds (CHLs) and HCB are of significant importance[1]. Besides, contamination and health impacts of polychlorinated dibenzo-p-dioxins (PCDDs), dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) are considered to me major importance in humans. These chlorinated compounds are recognized as environmental pollutants that do not decompose readily in the environment after agricultural, domestic and industrial use and disposal[2]. Among PCDD/DFs and dioxin-like PCBs the most potent of these chemicals is 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD). 2,3,7,8-TCDD and its congeners produce a wide spectrum of toxic effects including immunotoxicity, teratogenicity, carcinogenicity and lethality to humans and wild animals[3]. In addition, recent toxicological studies have demonstrated that PBDEs can elicit serious health effects such as thyrodiogenic, estrogenic and dioxin-like activity.

With frequent occurrence and considerable toxic effects of OCPs, PCDDs, PCDFs, PCBs and PBDEs in human tissue, biological degradation of these chemicals should be introduced. In rats, dietary fiber and chlorophyll shown to activate fecal excretion of PCDD/DFs probably due to inhibition of their absorption in the digestive tract and consequent decrease in liver. For instance, FEBRA the [fermented brown rice with *Aspergillus oryzae*] and rich dietary fiber seems to a play a major role for efficient removal of organic contaminants from human body[4]. Based on these observations, we studied the dynamics OCPs, PCDDs, PCDFs, PCBs, TEQ [toxic equivalent quantity by PCDD/DFs and PCBs] and PBDEs in blood of male and female of nine families as they have classified as FEBRA exposed and non-exposed categories over two-year study period from Japan.

Materials and Methods

Study Design and Sample Collection: FEBRA has been manufactured for 30-years by Genmaikouso Corporation located at Sapporo, Japan and about 100, 000 peoples consuming this staple food[4,5]. Eleven adult married couples of 37 to 48 year old were voluntarily participated for this study and they were further grouped in to two categories that match age and sex. First group

consumed about 7.5 to 10.5 g FEBRA immediate after their meal for two-year period and second group didn't. Besides, the exposure gender also categorized/varied in each family. Whole blood (approximately 50 mL) of each subject from nine family couples were collected in chemically clean heparin tubes for 6 different time periods [0 and 1-week of study period "namely 0 year study period", ½ an year study, 1-year study, 1½ year study and 2-year study]. The collected blood was transported to the laboratory and stored at –20^{o}C until chemical analysis.

Analysis and Quantification: The known weight of blood samples were ground with sodium sulfate (Na_2SO_4) until dry powder, extracted with dichloromethane for 20 h using Soxhlet extractor. Extracted samples were then concentrated to 20 mL using rotary evaporator and from aliquot of the extract, fat content was determined. The remaining extracts were sub-divided into 3 portions for OCPs, PCDD/DFs, PCBs and PBDEs analysis, respectively. The internal standards of all OCPs, 2378-PCDD/DFs, PCBs and PBDEs were spiked in according extracts. For OCPs analysis, it was subjected to a column chromatographic clean-up procedure[6]. For PCDD/DF and PCB analysis, extracts were subjected into multilayer silicagel column clean-up[6]. Immediately after the multilayer silicagel column, trace/known amount of sample was stored for total PCBs determinations. For PCDD/DF and dioxin-like PCBs clean-up, the multilayer cleaned samples was concentrated to 2 mL and sequentially subjected to, alumina and silica gel impregnated activated carbon column chromatography. Eventually, for BFRs analysis, it was subjected to a multi layer column chromatographic clean-up procedure. Analysis of PBDEs was slightly modified and was reported elsewhere[6]. Identification and quantification of OCPs, PCDD/DFs, PCBs and PBDE was performed using Hewlett Packard 6890 Series high-resolution gas chromatography interfaced with a Micromass Autospec - Ultima high-resolution mass spectrometer[6].

Results and Discussion

Organochlorine Pesticides: Among OCPs analyzed, DDTs was predominant accumulant in human blood followed by HCHs, CHLs and HCB (Table 1). DDTs, HCHs, CHLs and HCB, respectively contributed 40 - 91%, 3.2 - 60%, 1.3 - 24% and 1 - 8.6% to the total OCPs. In general, FEBRA exposed individual males contained greater DDTs levels than the non-exposed individuals. While FEBRA exposed females had greater HCHs than DDTs, CHLs and HCB. Perhaps, we appraised a greater co-incidence of most of FEBRA exposure subjects chosen in this study were from OCPs elevated individuals. The slight variation in concentrations of OCPs in inter family couples suggests slightly different feeding habits among husband (e.g., husband eats at working environment) and wife. However, food volume intake between individuals, metabolism, excretion and other habits also considerable. The contamination pattern was different in between family and these trends found to be a reflective of geographical variation of DDT contamination in Japan. In order to investigate the impact of OCPs in FEBRA exposed and non-exposed groups, we computer normalized 0.5 year, 1 year, 1.5 year and 2 year concentrations from the average concentration of 0 year and 1-week which normalized as 1.0 or 100[5]. The overall normalized data exhibited slightly promoted elimination of DDTs in FEBRA exposed groups [1.18 or 118] and non-exposed individual [1.22 or 122]. For HCHs, the estimations suggested slightly elevated exposure in FEBRA exposed groups [1.25 or 125] than the non-exposed groups [1.17 or 117]. In case of CHLs the results explicit intake of FEBRA has no effect in reducing overall accumulation of CHLs due to slightly higher levels in exposed groups [1.09 or 109] than non-exposed groups [0.99 or 99]. The HCB showed similar accumulation levels in FEBRA exposed group [0.96 or 96] and non-exposed group [0.97 or 97].

PCDD/DFs: The maximum concentration of PCDDs was noticed in female (Sample ID. 59514-

11) while lowest end was found in female (Sample ID.59514-079) as shown in Table 1. PCDFs accumulated lowest level among PCBs and PCDDs. Considerably, FEBRA-exposed individuals had lower levels of PCDD/DFs than non-FEBRA exposed individuals. In contrast to organochlorine pesticides and PCBs, PCDDs and PCDFs in most of males had lower levels than females (Table 1). The computer normalized data showed significantly decreased PCDD/DFs in FEBRA-exposed groups than non-FEBRA exposed groups. The overall normalized data exhibited that considerable reductions of PCDDs in between exposed [0.75 or 75] and non-exposed [0.82 or 82] groups. Particularly, reductions of PCDFs in between exposed [0.68 or 68] and non-exposed [0.83 or 83] groups were so prominent. Altogether, 6-times more decrease was observed for PCDDs in exposed subjects when compare to non-exposed individuals. Whereas, 15-times greater decrease was noticed for PCDFs in exposed subjects when compare to non-exposed individuals.

PCBs: Total PCBs were major accumulants in human blood than followed by dioxin-like PCBs and PCDD/DFs (Table 1). The maximum and minimum concentration of sum PCBs was noticed in 1-year FEBRA exposed male and 0-year non-FEBRA exposed female, respectively (Table 1). In general, there is no significant difference between the concentrations of PCBs in FEBRA-exposed and non-FEBRA exposed groups. Husbands from 8 among 9 family had greater PCB levels than wives. Reduction of organic pollutants in females can be explained as gestation and lactation transfer to the offspring. On the other hand, few reports documented greater intake of meat food, smoking habit, and vice-versa played a vital role in greater accumulation in males. The greater contamination in some female individuals suggestive of the greater in take of seafood and dairy products than males (who eats lots of farm raised animal meat). The computer normalized concentrations of PCBs exhibited indictable variation of PCBs in between exposed [1.14 or 114] and non-exposed groups [2.5 or 250]. The isomer/congener-specific trend of DL-PCBs implies a domination of CB-126 followed by CB-169 (in male), CB-77 (in female) and CB-81 among non-ortho PCBs (Data not shown). CB-118 was predominant congeners followed by CB-156, CB-105, CB-167, CB-157, CB-114, CB-189 and CB-113 among mono-ortho PCBs in either gender (Data not shown).

Toxic Equivalent Quantity (TEQ): The maximum TEQ (0.110 ng/g fat) was noticed in 0.5-year FEBRA exposed male (Sample ID. 59514-39). The minimum TEQ (0.007 ng/g fat) was observed in 1-year non-FEBRA exposed female (Sample ID. 59514-077&095). In most of the cases, PCDD/DF TEQ was abundant than PCBs TEQ. In order to investigate the changes in the TEQ levels in blood more in detail, their relative concentrations were computed based upon their respective original ones (0 and 1-week sample). In the FEBRA-intake group, in general, the PCDD and PCDF TEQ has been declined with average relative level of [0.62 or 62]. In the non-intake group, however, the relative concentrations showed a bit increase or no change with the mean relative level was [0.66 or 66]. The 41% of TEQ concentrations were reduced after intake of FEBRA for 1 year in exposed group. While non-exposed group showed only a 34% reduction. Although, PCDD/DF TEQ showed clearly reductive concentration and TEQ in FEBRA-exposed group, the DL-PCBs showed no reductions and therefore, the total TEQ was slightly un-changed in these groups. Collectively, PCDDs was prevalent TEQ contributors in all the samples followed by non-ortho PCBs, PCDFs and mono-ortho PCBs. In males however, contribution of PCDFs and mono-ortho PCBs were found equal. Altogether, FEBRA-exposed males reduced TEQ congeners from PCDDs but increased non-ortho PCB TEQ. In case of non-FEBRA exposed males and both groups females showed almost similar trend of TEQ contribution by 4 homologue groups.

PBDEs: In this study, DiBDE #15, TrBDE #28, TeBDEs #49, 47, 66, 77, PeBDEs #100, 119, 99, HxBDEs #154, 153, HpBDE #183 and DeBDE #209 were detected in blood samples and their sum were shown in Table 1. There was no notable difference of concentrations in between exposed and non-exposed as well as male and females (Table 1). The average DeBDE concentrations were 8.6 to 8.8 (in non-exposed male), 7.9-17 (in exposed female) on ng/g fat weight. Likewise, the DiBDE analyzed in two families had lower contamination levels (Table 1). On the whole, mean and ranges of tri- through hepta-BDEs were in males and females were 1.9-4.4 and 1.4 to 2.1 ng/g fat wt., respectively. The DeBDE (#209) was predominant congener followed by TeBDE (#47), HxBDE (#153), HpBDE (#183), PeBDE (#99), PeBDE (#100), TrBDE (#28), DiBDE (#15), HxBDE (#154) and TeBDE (#49). The isomers such as PeBDE (#126), HxBDEs (#139), (#138) and TeBDE (#77) were rarely detected in some samples. This is the first report of mono-through hepta-BDEs in human blood. It should be indicated that blank samples had noticeable levels of DeBDE and thus the results of DeBDE needed to be re-confirmed with further analysis. There was not apparent correlation in between total TEQ Vs TeBDE, TEQ Vs PeBDE, TEQ Vs HxBDE and PCDD/DF TEQ Vs PCDD/DF concentrations (results not shown). The exposure route of PBDEs probably suggests from working conditions of males and females. The TBBPA were also analyzed in this study, however due to lack of well defined extraction procedure and possibility of non-reproducibility of the data, we ignore to discuss more in detail.

In order to investigate the concentration flux or elimination rates of PBDEs more in detail, we computer normalized 0.5-year, 1-year, 1.5-year and 2-year concentrations from the average concentrations of 0-year and 1-week (data not shown) which normalized as 1.0 to 100. The overall The computer normalized concentrations of BFRs reduced in FEBRA non-exposed (0.79 or 79) and increased in exposed (1.15 or 115) individuals. In general the levels of PBDEs were lower in 1 and 1.5 year but elevated levels were noticed in 2-year samples. These results suggested that all family uniformly exposed elevated concentrations of PBDEs during second year of investigation. Altogether, the FEBRA not likely had any impact on exposed individuals due to the slight elevation with those of non-exposed individuals. These findings are similar to those to the organochlorine pesticides but contrast to PCDDs and PCDFs. The 7 family males among 9 family had comparatively greater concentrations than females although average concentrations were greater in females due to one individual had elevated concentrations. Greater food intake and working exposure by males probably a possible explanation of higher levels. As indicated earlier, the temporal variation seems very prominent in all the family. Particularly, the sample collected in 0.5-year and 2-year had higher concentrations while 1 and 1.5-year samples showed minimum levels.

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Table 1. The sample details, concentrations (ng/g fat wt.) of organochlorine pesticides, PCDDs, PCDFs, Total PCBs, WHO-TEQs and total PBDEs in human blood of FEBRA exposed and non-exposed groups from Japan.

Sample I.D.	Sample Code	Gender[a] (FEBRA)	Sample Period[b]	Sample number	Fat %	DDTs[c]	HCHs[d]	CHLs[e]	HCB[f]	2378-PCDDs	2378-PCDFs	Total PCBs	Total TEQ	Total PBDEs
59514-01&2	G01A	M (Y)	0	(n=2)	0.21	280	55	56	23	0.47	0.16	330	0.044	3.3
59514-37	G01A	M (Y)	0.5	(n=1)	0.40	170	39	33	9.9	0.28	0.25	250	0.033	8.5
59514-0648&082	G01A	M (Y)	1	(n=2)	0.27	210	39	35	17	0.18	0.00	330	0.029	3.8
59514-101	G01A	M (Y)	1.5	(n=1)	0.33	200	33	39	22	0.17	0.04	290	0.025	3.6
73140-001&002	G01A	M (Y)	2	(n=2)	0.46	210	36	33	18	0.13	0.02	240	0.020	58
59514-03&04	G01B	F (N)	0	(n=2)	0.32	330	160	34	17	0.74	0.05	190	0.022	1.8
59514-48	G01B	F (N)	0.5	(n=1)	0.41	250	180	24	7.8	0.71	0.04	210	0.018	6.1
59514-065&083	G01B	F (N)	1	(n=2)	0.33	290	170	23	11	0.52	0.02	200	0.016	2.4
59514-102	G01B	F (N)	1.5	(n=1)	0.38	240	120	22	12	0.54	0.03	200	0.016	2.4
73140-003&004	G01B	F (N)	2	(n=2)	0.44	230	140	18	8.1	0.56	0.02	190	0.015	13
59514-05&06	G02A	M (N)	0	(n=2)	0.30	160	19	43	12	0.20	0.07	180	0.014	12[g]
59514-38	G02A	M (N)	0.5	(n=1)	0.36	100	27	44	5.2	0.14	0.06	170	0.015	14[g]
59514-066&084	G02A	M (N)	1	(n=2)	0.30	160	29	48	8.7	0.10	0.01	210	0.013	13[g]
59514-103	G02A	M (N)	1.5	(n=1)	0.38	130	23	35	8.8	0.11	0.02	170	0.011	4.0[g]
73140-005&006	G02A	M (N)	2	(n=2)	0.32	130	22	32	7.2	0.09	0.01	190	0.010	37[g]
59514-07&08	G02B	F (Y)	0	(n=2)	0.27	410	200	53	17	1.1	0.20	660	0.067	12[g]
59514-49	G02B	F (Y)	0.5	(n=1)	0.46	350	430	60	8.2	0.62	0.04	370	0.034	20[g]
59514-067&085	G02B	F (Y)	1	(n=2)	0.34	550	560	50	19	0.47	0.03	600	0.041	8.4[g]
59514-104	G02B	F (Y)	1.5	(n=1)	0.39	500	470	73	21	0.70	0.04	540	0.042	6.9[g]
73140-007&008	G02B	F (Y)	2	(n=2)	0.36	490	480	63	16	0.57	0.03	560	0.038	19[g]
59514-09&10	G03A	M (Y)	0	(n=2)	0.76	710	480	140	33	0.54	0.06	800	0.071	10
59514-39	G03A	M (Y)	0.5	(n=1)	0.73	520	710	150	29	0.79	0.12	130	0.110	6.9
59514-068&086	G03A	M (Y)	1	(n=2)	0.72	710	620	83	45	0.40	0.04	980	0.071	3.6
59514-105	G03A	M (Y)	1.5	(n=1)	0.69	730	670	150	49	0.62	0.06	1000	0.079	4.0
73140-009&010	G03A	M (Y)	2	(n=2)	0.87	720	550	130	29	0.45	0.05	980	0.067	86
59514-11&12	G03B	F (N)	0	(n=2)	0.40	210	42	29	14	1.9	0.05	270	0.031	4.4
59514-50	G03B	F (N)	0.5	(n=1)	0.53	140	46	22	7.4	1.3	0.03	270	0.024	4.0
59514-069&087	G03B	F (N)	1	(n=2)	0.30	230	56	22	16	1.3	0.03	360	0.029	3.3
59514-106	G03B	F (N)	1.5	(n=1)	0.40	260	70	28	22	1.6	0.04	310	0.029	2.5
73140-011&012	G03B	F (N)	2	(n=2)	0.42	260	61	31	16	1.5	0.03	340	0.027	13
59514-17&18	G05A	M (N)	0	(n=2)	0.76	180	41	51	13	0.14	0.03	280	0.025	4.9
59514-41	G05A	M (N)	0.5	(n=1)	0.46	190	66	49	14	0.17	0.11	290	0.032	85
59514-070&088	G05A	M (N)	1	(n=2)	0.51	300	91	71	22	0.15	0.03	460	0.037	4.4
59514-107	G05A	M (N)	1.5	(n=1)	0.69	250	73	49	16	0.17	0.03	380	0.031	4.8
73140-013&014	G05A	M (N)	2	(n=2)	0.65	210	57	47	10	0.13	0.02	340	0.024	8.9
59514-19&20	G05B	F (Y)	0	(n=2)	0.36	270	220	9.2	16	0.91	0.06	120	0.025	2.6
59514-52	G05B	F (Y)	0.5	(n=1)	0.46	150	190	10	7.6	0.60	0.04	95	0.019	3.7
59514-071&089	G05B	F (Y)	1	(n=2)	0.30	340	420	13	22	0.65	0.04	170	0.023	2.7
59514-108	G05B	F (Y)	1.5	(n=1)	0.47	280	310	11	15	0.70	0.05	150	0.019	3.6
73140-015&016	G05B	F (Y)	2	(n=2)	0.36	310	300	13	11	0.64	0.03	140	0.016	470
73140-035&038	G05B	F (Y)	2.5	(n=2)	0.59	230	230	12	9.3	0.55	0.03	98	0.015	7.7
59514-21&22	G07A	M (Y)	0	(n=2)	0.40	1100	97	110	75	0.27	0.08	1000	0.067	15
59514-42	G07A	M (Y)	0.5	(n=1)	0.28	1000	120	120	31	0.30	0.13	1000	0.098	9.5
59514-072&090	G07A	M (Y)	1	(n=2)	0.57	1100	120	130	34	0.13	0.04	1100	0.062	6.5
59514-109	G07A	M (Y)	1.5	(n=1)	0.38	1600	72	75	72	0.16	0.04	900	0.058	5.4
73140-017&018	G07A	M (Y)	2	(n=2)	0.32	860	80	84	21	0.14	0.05	870	0.050	32
59514-23&24	G07B	F (N)	0	(n=2)	0.42	220	380	45	26	1.5	0.05	430	0.045	6.0
59514-53	G07B	F (N)	0.5	(n=1)	0.46	160	240	30	17	0.95	0.04	310	0.031	4.5

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Table 1 Continues.,

Sample I.D.	Sample Code	Gender[a] (FEBRA)	Sample yr[b]	Sample number	Fat %	DDTs[c]	HCHs[d]	CHLs[e]	HCB[f]	2378-PCDDs	2378-PCDFs	Total PCBs	Total TEQ	Total PBDEs
59514-073&091	G07B	F (N)	1	(n=2)	0.30	230	460	53	33	0.87	0.04	400	0.038	4.0
59514-110	G07B	F (N)	1.5	(n=1)	0.39	440	340	45	66	1.2	0.05	380	0.043	3.0
73140-019&020	G07B	F (N)	2	(n=2)	0.28	240	450	46	23	1.1	0.05	470	0.041	22
59514-25&26	G08A	M (N)	0	(n=2)	0.60	240	30	39	12	0.14	0.02	310	0.023	18[g]
59514-43	G08A	M (N)	0.5	(n=1)	0.57	310	37	42	13	0.16	0.09	290	0.027	23[g]
59514-074&092	G08A	M (N)	1	(n=2)	0.40	480	50	84	26	0.13	0.03	430	0.030	8.9[g]
59514-111	G08A	M (N)	1.5	(n=1)	0.40	920	47	55	77	0.16	0.03	430	0.032	6.9[g]
73140-021&022	G08A	M (N)	2	(n=2)	0.37	600	61	88	23	0.17	0.04	540	0.033	26[g]
59514-27&28	G08B	F (Y)	0	(n=2)	0.36	91	48	20	11	0.30	0.04	180	0.019	54[g]
59514-54	G08B	F (Y)	0.5	(n=1)	0.44	110	57	24	12	0.20	0.02	140	0.016	34[g]
59514-075&093	G08B	F (Y)	1	(n=2)	0.31	160	92	37	16	0.21	0.03	230	0.022	7.3[g]
59514-112	G08B	F (Y)	1.5	(n=1)	0.38	300	71	21	34	0.22	0.02	210	0.019	5.0[g]
73140-023&024	G08B	F (Y)	2	(n=2)	0.32	160	79	29	11	0.20	0.02	270	0.018	10[g]
59514-29&30	G09A	M (Y)	0	(n=2)	0.35	310	51	28	12	0.43	0.03	210	0.020	7.0
59514-44	G09A	M (Y)	0.5	(n=1)	0.23	450	78	32	16	0.72	0.08	440	0.034	7.4
59514-076&094	G09A	M (Y)	1	(n=2)	0.32	390	86	37	16	0.35	0.02	240	0.015	3.2
59514-113	G09A	M (Y)	1.5	(n=1)	0.41	1000	75	26	27	0.78	0.04	330	0.025	3.0
73140-025&026	G09A	M (Y)	2	(n=2)	0.28	510	79	32	19	0.49	0.04	330	0.020	210
59514-31&32	G09B	F (N)	0	(n=2)	0.35	650	17	10	5.4	0.51	0.09	90	0.014	9.4
59514-55	G09B	F (N)	0.5	(n=1)	0.15	110	23	9.1	10	1.3	0.16	220	0.029	4.1
59514-077&095	G09B	F (N)	1	(n=2)	0.35	93	22	13	9.9	0.30	0.01	87	0.007	2.0
59514-114	G09B	F (N)	1.5	(n=1)	0.46	180	16	11	23	0.41	0.05	89	0.011	1.4
73140-027&028	G09B	F (N)	2	(n=2)	0.32	99	22	9.5	7.4	0.38	0.02	98	0.010	23
59514-33&34	G10A	M (Y)	0	(n=2)	0.29	180	21	25	6.2	0.35	0.19	230	0.030	23
59514-45	G10A	M (Y)	0.5	(n=1)	0.28	200	20	23	8.2	0.27	0.12	250	0.030	4.5
59514-078&096	G10A	M (Y)	1	(n=2)	0.35	210	20	22	12	0.13	0.02	200	0.011	2.4
59514-115	G10A	M (Y)	1.5	(n=1)	0.33	550	20	35	22	0.19	0.02	260	0.015	5.0
73140-029&030	G10A	M (Y)	2	(n=2)	0.31	160	20	31	9.7	0.20	0.03	290	0.015	25
59514-35&36	G10B	F (N)	0	(n=2)	0.41	100	43	28	7.5	0.22	0.17	160	0.019	10
59514-56	G10B	F (N)	0.5	(n=1)	0.23	210	67	38	15	0.27	0.05	210	0.025	NA
59514-079&097	G10B	F (N)	1	(n=2)	0.49	130	43	24	11	0.13	0.02	140	0.013	1.7
59514-116	G10B	F (N)	1.5	(n=1)	0.55	270	36	23	24	0.12	0.01	120	0.009	2.4
73140-031&032	G10B	F (N)	2	(n=2)	0.36	130	30	29	7.8	0.12	0.02	140	0.011	38
59514-46&47	G11A	M (N)	0	(n=2)	0.21	540	88	63	19	0.24	0.08	420	0.040	32
59514-080	G11A	M (N)	0.5	(n=1)	0.34	520	65	51	19	0.12	0.02	290	0.020	3.4
59514-117&119	G11A	M (N)	1	(n=2)	0.37	1000	70	38	34	0.18	0.02	290	0.020	4.0
73140-033	G11A	M (N)	1.5	(n=1)	0.27	530	56	42	15	0.16	0.04	320	0.019	47
73140-036&039	G11A	M (N)	2	(n=2)	0.51	350	42	28	11	0.13	0.02	190	0.013	8.7
59514-57&58	G11B	F (Y)	0	(n=2)	0.35	86	59	14	9.4	0.62	0.04	120	0.014	6.4
59514-081	G11B	F (Y)	0.5	(n=1)	0.4	120	43	17	13	0.32	0.02	150	0.011	1.8
59514-118&120	G11B	F (Y)	1	(n=2)	0.53	250	76	12	24	0.49	0.02	120	0.012	1.8
73140-034	G11B	F (Y)	1.5	(n=1)	0.39	98	52	15	9.7	0.47	0.03	130	0.011	17
73140-037&040	G11B	F (Y)	2	(n=2)	0.51	100	50	16	9.3	0.44	0.02	120	0.012	6.2

[a]M=male and F=female; [b]Time of sampling by year;(FEBRA) exposed (Y) and not exposed (N); [c]sum of o,p'-/p,p'-DDE, DDD and DDTs

[d]sum of -a,-b,-g,-d-HCH isomers; [e]sum of cis-/trans-chlordane, cis-/trans-nonachlor and oxychlordane; [f]Hexachlorobenzene

Two family G04 and G06 discontinued with in one week of the study period, while, G05B female participate for 2.5 years.

[g]Inclusion of DiBDE and DeBDE