Time trends of PCDDs/DFs in human adipose tissues in Japan

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INTRODUCTION

The levels of PCDDs/DFs in human adipose tissues from general population in various countries have been reported. However, only few information is available on contamination trends in human adipose tissues. Earlier, in our laboratory, Ono[1] reported the trends of PCDDs/DFs levels in human adipose tissues from Ehime district, Japan, since 1959 to 1985. In order to further understand the recent levels and patterns, we analyzed human adipose tissues for PCDDs/DFs collected in Ehime district during 1990-1993. Three high peaks were found in 1970, 1980 and 1991, and the causes for such elevated levels are discussed in this report. Apart from Ehime district, we analyzed human adipose tissues in two more districts to know the PCDDs/DFs contamination levels in other districts.

MATERIALS AND METHODS

As the major objective of this study was to elucidate temporal trends of PCDDs/DFs in human adipose tissues rather than describing spatial distribution, we feel that undermentioned samples are representative and suitable for such studies.

Materials: Human adipose tissues were obtained from three regions

(1) Ehime district : Totally 11 adipose samples were collected during 1990 to 1993. They were all males and the average age was 54.4 years.

(2) Kinki district : Totally 7 adipose samples were collected in 1993. Four samples were males with the average age of 65.5 years and three samples were females with the average age of 55 years.

(3) Tyukyou district : Totally 11 adipose samples were collected during 1990 to 1992. Seven samples were males with the average age of 60.6 years and four samples were females with the average age of 48.8 years

Analytical method: Fat samples were extracted from adipose tissues using Soxhlet apparatus in 450 ml dichloromethane for 24 hrs. About 10 g of the fat samples were used for analysis. The samples were treated with H_2SO_4 and they were cleaned up using silica gel, alumina, activated carbon column chromatography and HPLC. The determination of PCDDs/DFs was performed by HRGC/HRMS (HP5890 II /JMS-SX102A), using CP-SIL 88 fused silica capillary column. (50m × 0.25mm I.D. × 0.2 μm)

RESULTS AND DISCUSSION

PCDDs/DFs trends in human adipose tissues

The levels of PCDDs/DFs in human adipose tissues are shown in Table 1. All detected compounds were 2,3,7,8-substituted isomers. Based on the results, we could figure out the time trends of PCDDs/DFs levels in human adipose tissues from 1959 to 1993 in Ehime district. We observed three peaks through out the period, in 1970, 1980 and 1991. The first peak in 1970 might be caused by the use of chlorinated organic compounds such as organochlorine pesticides, PCBs and so on. Since the production and use of these compounds have been banned, it seemed that the occurrence of PCDDs/DFs was also decreased later. But, the trend of PCDDs/DFs was upward again in 1980. The increased contamination during this period is assumed to be caused by the consumption of chickens and eggs as shown by the results of food analysis in our earlier study.

In this study, the highest peak was observed in 1991. The trend seemed to increase in 1990,1991 and then decline in 1992, 1993. The number of samples analyzed is very few, therefore the average value is much skewed towards the elevated value. At present, we have no explanation for increasing of PCDDs/DFs level in recent years. Because there are multiple contamination sources in the environment. The PCDDs trends is shown in Figure 1, where PCDDs congeners are represented by TEQ (2,3,7,8-TeCDD Equivalent Quantity) based on I-TEF. Figure 1 showed that the composition of PCDDs congeners varied during the period 1959-1993. The congeners with highest level in all the periods were as follows; OCDD in 1970, TeCDD in 1980 and HxCDD in 1991. The HxCDD isomers detected were 1,2,3,4,7,8-, 1,2,3,6,7,8- and 1,2,3,7,8,9-HxCDD. 1,2,3,6,7,8-HxCDD was the highest among them. 1,2,3,7,8-PeCDD was the second predominant congener in all the periods. The time trends of human contamination by PCDFs was not complex as that of PCDDs. We observed that PeCDFs was the predominant congener in all the periods and the main isomer was 2,3,4,7,8-PeCDF.

To find out the regional difference, samples from Kinki and Tyukyou districts were analyzed and compared. The results are shown in Figure 2. In Tyukyou, the PCDDs levels were highest in 1991 as in Ehime prefecture and declined since then. For PCDFs, the levels exhibited almost same trend. In Kinki, same trend is also expected. Analysis of the sample of Kinki from 1990 to 1992 will throw light on this aspect. The time trends of PCDDs/DFs levels had slight difference between these regions. We observed the congener contribution to the total dioxin toxic equivalents in three regions. We found that HxCDD, PeCDF, PeCDD and TeCDD mainly contributed for the TEQ in three regions. The composition of PCDDs/DFs was similar in three regions. In Germany[2], it was reported that PeCDF, HxCDD, PeCDD and TeCDD were mainly contributed to the TEQ. In the USA[2], TeCDD, HxCDD PeCDD and PeCDF were mainly contributed to the TEQ. The present result implies that the composition pattern of major toxic congeners in Japanese adipose tissue is different from those of other countries.

Table 1 : Levels (pg/g, fat basis) of PCDDs/DFs in human adipose tissues in Japan

District	Ehime											
Year	1990			1991			1992			1993		
n	3		mean	3		mean	2		mean	3		mean
2,3,7,8-TeCDD	4.1	6.1	5.4	7.3	21	12	3.6	3.7	3.7	ND	(<2.4)	N D
1,2,3,7,8-PeCDD	21	26	24	24	86	45	14	18	16	6.6	9.6	8.1
1,2,3,4,7,8-HxCDD	9.1	12	11	6.3	17	13	6.2	8.9	7.6	ND	(<2.9)	N D
1,2,3,6,7,8-HxCDD	120	240	180	110	340	210	110	140	130	34	52	42
1,2,3,7,8,9-HxCDD	24	51	34	13	57	31	21	25	23	<5.3	10.2	10.2
1,2,3,4,6,7,8-HpCDD	100	180	140	32	73	54	50	62	56	28	41	34
OCDD	2000	13000	6200	2100	3100	2500	1900	4500	3200	200	730	390
2,3,7,8-TeCDF	7.0	34	19	8.9	47	31	5.2	6.7	6.0	<1.8	4.9	4.9
1,2,3,7,8-PeCDF	5.0	13	9.1	7.1	24	14	2.6	4.1	3.4	ND	(<1.3)	N D
2,3,4,7,8-PeCDF	40	46	43	52	170	93	30	31	31	9.6	15	12
1,2,3,4,7,8-HxCDF	12	14	13	13	28	18	6.5	10	8.4	4.3	4.4	4.4
1,2,3,6,7,8-HxCDF	12	15	14	15	37	23	6.8	8.2	7.5	3.5	5.4	4.5
1,2,3,7,8,9-HxCDF	ND	(<1.6)	N D	ND	(<1.5)	N D	ND	(<0.9)	N D	ND	(<1.1)	N D
2,3,4,6,7,8-HxCDF	<1.6	6.3	6.3	3.5	4.1	3.7	1.4	4.5	3.0	ND	(<1.1)	N D
1,2,3,4,6,7,8-HpCDF	15	16	16	8.9	16	12	3.7	5.6	4.7	ND	(<3.1)	N D
1,2,3,4,7,8,9-HpCDF	ND	(<2.4)	N D	ND	(<2.9)	N D	ND	(<0.9)	N D	ND	(<3.1)	N D
OCDF	ND		N D	ND		N D	ND		N D	ND		N D
PCDDs/DFs	2369.2	13659.4	6714.8	2401	4020.1	3059.7	2161	4827.7	3500.3	286	872.5	510.1
TEQ	57	95	75.4	65.2	207.2	118.8	43.8	54.5	50.4	12.8	21	17.4
Recovery rate (%)	37			41			47			64		

District	Tyukyou						Kinki		
Year	1990	1991			1992		1993		
n	1	7		mean	3		mean	7		mean
2,3,7,8-TeCDD	2.7	5.4	12	7.7	1.8	4.4	3.0	2.0	3.9	2.8
1,2,3,7,8-PeCDD	15	9.1	77	33	12	30	20	6.2	14	9.5
1,2,3,4,7,8-HxCDD	8.8	17	100	31	7.8	29	20	3.1	7.5	5.3
1,2,3,6,7,8-HxCDD	73	63	180	97	90	180	120	46	82	63
1,2,3,7,8,9-HxCDD	ND (<7.0)	9.3	53	23	8.5	44	21	6.7	15	9.7
1,2,3,4,6,7,8-HpCDD	22	43	260	120	16	430	210	16	81	34
OCDD	77	170	630	390	190	11000	4600	240	1400	650
2,3,7,8-TeCDF	2.2	2.7	6.9	4.7	1.4	4.6	3.2	1.3	3.3	2.1
1,2,3,7,8-PeCDF	ND (<0.3)	1.2	3.0	2.2	1.1	1.4	1.3	1.1	2.4	1.9
2,3,4,7,8-PeCDF	10.1	9.2	21	15	6.9	18	14	7.7	23	14
1,2,3,4,7,8-HxCDF	3.5	2.5	11	4.9	1.3	5.6	3.1	3.6	9.3	6.3
1,2,3,6,7,8-HxCDF	ND (<1.3)	2.6	4.4	3.2	1.1	6.8	3.2	4.7	9.8	7.3
1,2,3,7,8,9-HxCDF	ND (<1.3)	ND	(<1.2)	N D	ND	(<1.7)	N D	1.1	1.2	1.2
2,3,4,6,7,8-HxCDF	ND (<1.3)	ND	(<1.2)	N D	ND	(<1.7)	N D	2.2	5.5	3.6
1,2,3,4,6,7,8-HpCDF	ND (<1.3)	NA		N A	NA		N A	7.0	16	9.6
1,2,3,4,7,8,9-HpCDF	ND (<1.3)	NA		N A	NA		N A	<1.1	2.2	2.2
OCDF	ND	ND		N D	ND		N D	ND		N D
PCDDs/DFs	214.3	335	1358.3	731.7	337.9	11753.8	5018.8	348.7	1676.1	822.5
TEQ	24.4	25.1	100.9	50.3	22.8	70.9	43.8	16.5	38.8	25.6
Recovery rate (%)	39	41			28			82		

ND=Not Detected
NA=Not Analyzed

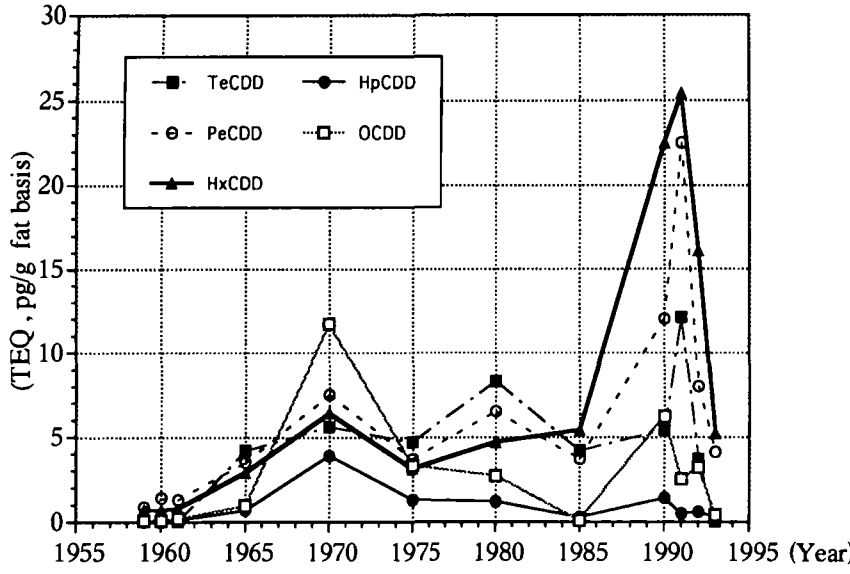

Figure 1 : Time trends of PCDDs (TEQ pg/g) in human adipose tissues in Ehime district

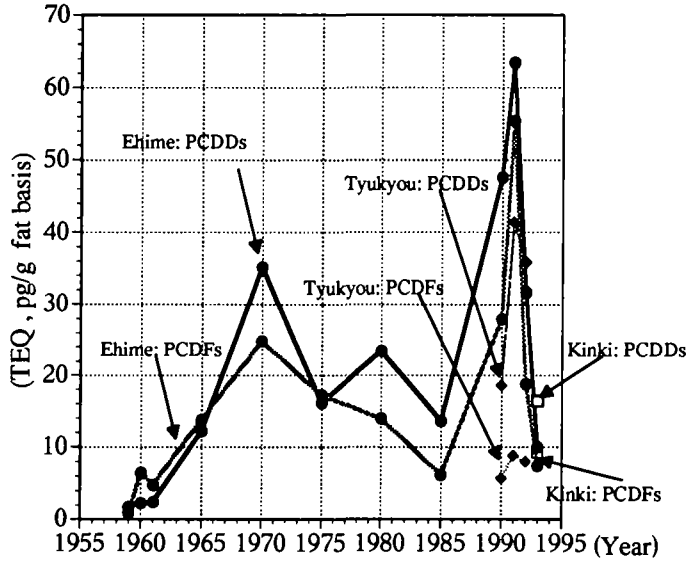

Figure 2 : Time trends of 2,3,7,8-TeCDD Equivalent (TEQ pg/g) in human adipose tissues in three regions

CONCLUSIONS

The results of the present study can be summarized as following;

1: Three peaks were observed in time trends of PCDDs/DFs level in human adipose tissues in Japan from 1959 to 1993.

2: Since 1990, A similar trends was found for Ehime and Tyukyou.

3: Since 1985, the main composition of PCDDs was 1,2,3,6,7,8-HxCDD whereas PCDFs was 2,3,4,7,8-PeCDF. The second most predominant isomer was 1,2,3,7,8-PeCDD.

As the major objective of this study was to elucidate temporal trends of PCDDs/DFs in human adipose tissues rather than describing spatial distribution, we feel that analyzed samples are representative and suitable for such studies. As for this results, it may be concluded that PCDDs/DFs trends in human adipose tissues the composition of congeners are similar in three regions. It is necessary to continue the research to know the sources of present PCDDs/DFs pollution in human body.

REFERENCES

1) Ono Mitsuhiro : Study of PCDDs and PCDFs levels and these sources in human adipose tissues in Japan : *A thesis for doctorate (1988)*

2) A. Schecter, P. Fürst, C. Fürst, O. Päpke, M. Ball, J. J.Ryan, Hoang Dinh Cau, Le Cao Dai, Hoang Trong Quynh, H.Q.Cuong, Nguyen Thi Ngoc Phuong, Pham Hoang Phiet, A. Beim, J. Constable, J. Startin, M. Samedy, and Yit Kim Seng : Chlorinated Dioxins and Dibenzofurans in Human Tissue from General Populations: A Selective Review : *Environmental Health Perspectives Supplements Vol. 102 Suppl 1:159-171 (1994)*