DECREASE OF PCDDs, PCDFs AND Co-PCBs LEVELS IN HUMAN MILK FROM OSAKA (1973-1996)

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Introduction

Recently, environmental pollution by dioxins has been attracting social attention and it has been pointed out that dioxins might be related to so called endocrine disrupters. An animal experiment revealed that endometriosis was induced by dioxins in Rhesus monkeys[1)] and another report showed that the amount of dioxins ingested by human infants via human milk was negatively correlated with their thyroid hormone level in blood[2)]. Furthermore, there are reports showing that the number of female infants became transiently larger than that of male ones in the area exposed to high-level dioxins, which were leaked because of a plant accident in Seveso, Italy[3)]. Therefore, it is thought that the effects of various endocrine disrupters including dioxins on human bodies are especially severe for the fetus and the infant.

Here, we determined the dioxin-levels in human milk samples which had been stored in a freezer after taking from mother in Osaka and demonstrated that there were significant changes in the levels during a 24-year period from 1973 to 1996.

Material and Methods

Samples: Human milk fats stored in a freezer after taking from mothers living in Osaka (within 30 days from parturition) in a period of 1973-1996 were used as the samples. Each 0.1 g of human milk-fat samples from 19-33 primipara ranging in 25-29 years of age was taken to mix each other. Thus obtained homogeneous mixture was used as test sample for each year.

Analytical method for dioxins: Analysis of the PCDDs, PCDFs and Co-PCBs in human milk was made according to the procedures shown in Fig. 1.

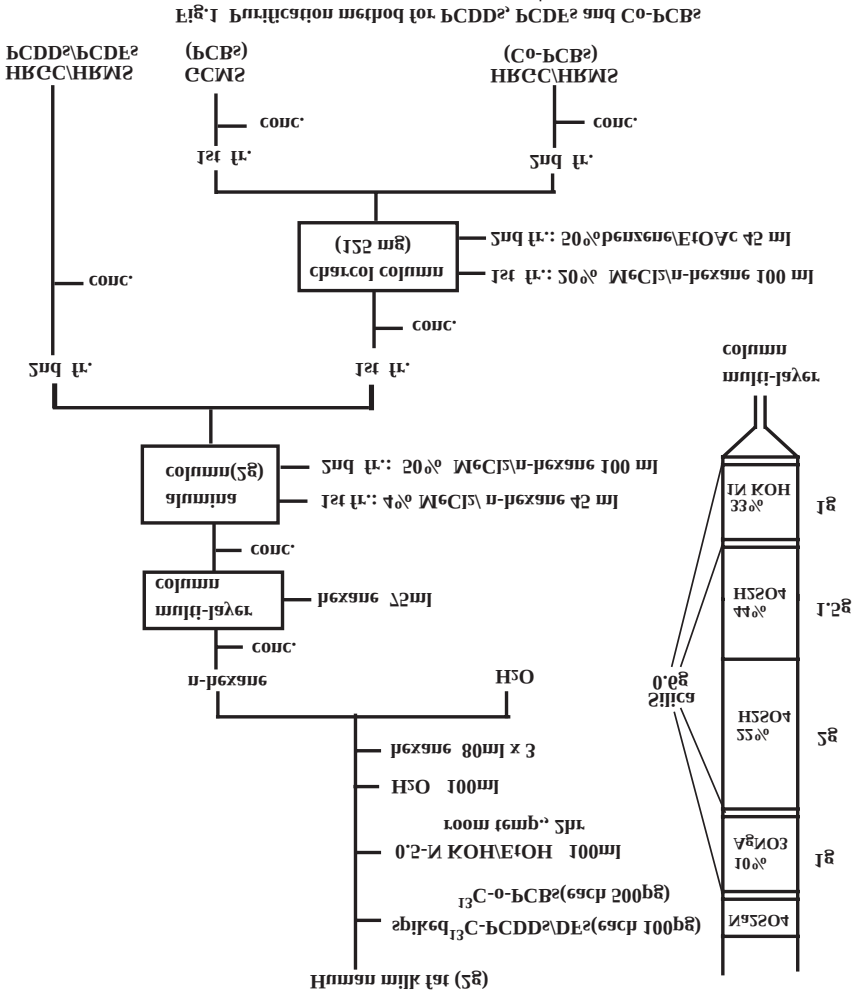
Results and Discussion

PCDDs, PCDFs and Co-PCBs Ievels in human milk :The annual reports, 1994 and 1995 of the Ministry of Health and Welfare showed that the mean level of PCDDs+PCDFs in human milk fat from Japanese women was 26.6 pg-TEQ/g fat. Regarding the levels for other countries, it is known to be 29-37 pg-TEQlg fat for England, 28-37 pg-TEQlg for Germany and 16-23 pg-TEQlg for Canada[4)]. Thus, it was thought that PCDDs+PCDFs levels in human milk were almost equal among these advanced countries including Japan.

Here, we determined the levels of PCDDs, PCDFs, Co-PCBs and PCBs in human milk from mothers living in Osaka and demonstrated significant changes in the levels during the period, 1973-1996 as shown in Fig. 2and Table 1.

The human milk sample for 1996 included 124.5 pg/g fat of PCDDs, 26.4 pg/g fat of PCDFs, 150.9pg/g fat of PCDDs +PCDFs and 129.0 pg/g fat of Co-PCBs and the levels for 1973 were 1292.6 pg/g fat for PCDDs, 58.5 pg/g fat for PCDFs, 1351.1 pg/g fat for PCDDs + PCDFs and 568.9 pg/g fat for Co-PCBs. Thus, it was demonstrated that all the four levels were markedly decreased to 9.6% for PCDDs, 45.1% for PCDFs, 11.2% for PCDDs + PCDFs and 22.7% for Co-PCBs. Meanwhile, TEQ levels of PCDDs, PCDFs and Co-PCBs in human milk fat were as follows; TEQ levels (pg/g fat) for 1996 was 9.5 pg for PCDD, 6.8 pg for PCDF, 16.3 pg for PCDDs + PCDFs and 7.8 pg for Co-PCBs. Since TEQ Ievels for 1973 were 14.3 pg for PCDDs,

11.4 pg for PCDFs, 25.7 pg for PCDDs + PCDFs and 31.4 pg for Co-PCBs, the four levels were found to be reduced to be 66.4, 59.6, 63.4 and 24.8%, respectively during 23 years.
Changes in the isomer levels in PCDDs, PCDFs and Co.-PCBs :With regard to the isomers of PCDDs, marked decreases in 2,3,7,8-TCDD 1,2,3,4,7,8-HxCDD, 1,3,3,4,6,7,8-HpCDD and OCDD in the period of 1973-1996 was found, but the proportion of 1,2,3,6,7,8-HxCDD was increased. With regard to PCDFs, there were significant decreases in its isomers except OCDF and the proportions of 3,3',4,4'-TCB and 3,3',4,4',5-PeCB in Co-PCBs were also decreased.



Comparison of isomer compositions for PCDDs, PCDFs and Co-PCBs between 1975 and 1996. :There were marked changes in the isomer composition of PCDDs between 1975 and 1996, i.e., OCDD occupied 87% in 1975, whereas its proportion decreased to 53.9% in 1996 and HxCDD increased from 2.2% (1975) to 30.4% (1996). Whereas the composition of PCDFs showed no marked change although only the proportion of HpCDF was slightly decreased (from 13.5% to 7.6%). However, there were significant changes in the isomer composition of Co-PCBs

as well as PCDDs during the period. Namely, a decrease in 3,3',4,4'-TCB and an increase in 3,3',4,4',5,5'-HxCB were found in 1996. Thus, it was demonstrated that PCDDs+PCDFs level in human milk fat decreased to nearly a half level during about 20 years. This decrease was thought to have been caused by the following reasons: 1) Lowering of food pollution level has resulted from the decrease of environmental pollution, 2) The quality of diets has been changed associating with diversification of food materials and increases in the kinds of imported foods.

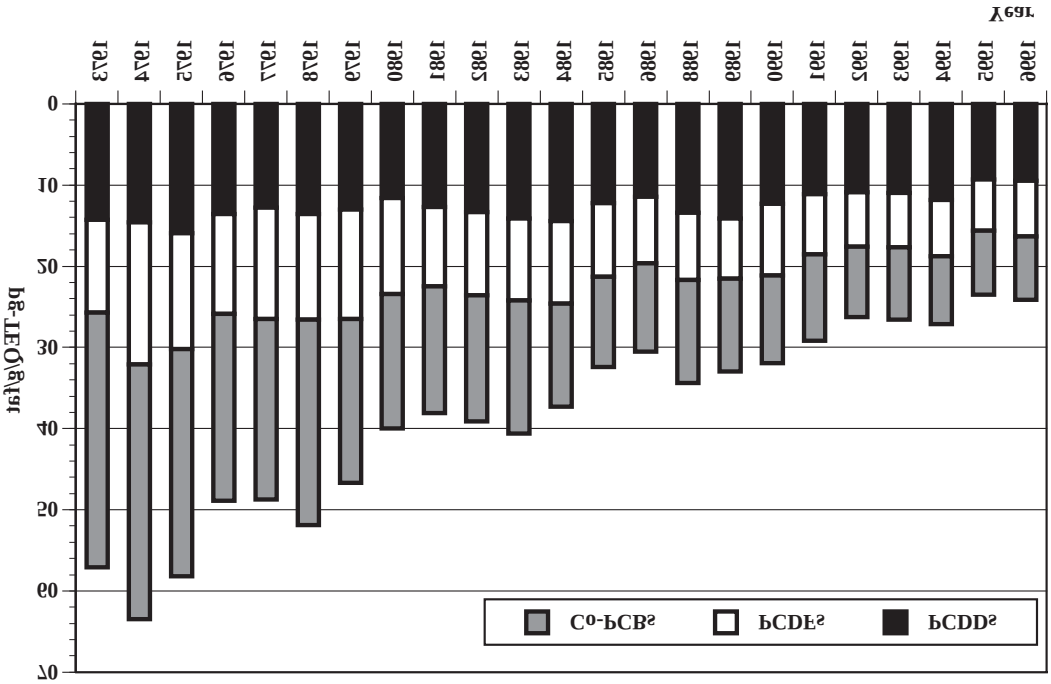


Fig.5 Time-course of PCDDs, PCDFs and Co-PCBs levels in human milk of mothers living in Osaka

However, the real reason is not unclear at present.

The amount of PCDDs+PCDFs ingested by infants via mother's milk was estimated to be 115.2 pg-TEQ/kg/day for 1973 and 73.4 pg-TEQ/kg/day for 1996. The both amounts were much higher than either of the tolerable daily intake (TDI) levels of 10 pg and 1- 4 pg-TEQ/kg/day defined by Japan authority concerned and WHO, respectively. When it was supposed that the infants had artificial breeding milk containing dioxins at 0.3 pg-TEQ/ fat instead of mother's milk, the amount of PCDDs+PCDFs ingested to infants was estimated as about 1.4 pg-TEQ/kg/day.The pollution of foods and human milk by organochlorine compounds, which are regarded as endocrine disrupters were much severer in 1970's than now. Therefore, there is a possibility that the environmental pollution in 1970's might have affected the fertilities of young men and women in the twenties at present because they were exposed to the pollution at the age of infant. The endocrine disturbing effects of dioxins and organochlorine compounds on human health should be investigated from long-term viewpoints in consideration of not only the exposure at present but also in the periods of fetus and infant. Therefore, it is essential to make a long-lasting monitoring of dioxins from such viewpoints.

Table 1. Average levels(fat basis) of PCDDs, PCDFs, Co-PCBs and PCBs in human milk(Osaka)

year	PCDDs pg-TEQ/g	PCDFs pg-TEQ/g	PCDDs/DFs pg-TEQ/g	Co-PCBs pg-TEQ/g	PCBs ug/g
1973	14.3	11.4	25.6	31.4	1.43
1974	14.6	17.5	32.1	31.4	1.58
1975	15.9	14.3	30.2	28.0	1.24
1976	13.6	12.3	25.9	23.0	1.13
1977	12.8	13.7	26.5	22.2	1.15
1978	13.6	13.0	26.6	25.3	1.34
1979	13.0	13.5	26.4	20.1	1.11
1980	11.6	11.8	23.4	16.6	1.00
1981	12.7	9.80	22.5	15.6	1.04
1982	13.3	10.3	23.7	15.0	1.04
1983	14.1	10.1	24.2	16.4	1.17
1984	14.4	10.2	24.6	12.7	0.89
1985	12.2	9.1	21.3	11.1	0.74
1986	11.4	8.2	19.6	10.9	0.67
1988	13.4	8.3	21.7	12.7	0.52
1989	14.1	7.4	21.5	11.5	0.53
1990	12.3	8.8	21.1	10.8	0.48
1991	11.1	7.4	18.5	10.7	0.36
1992	10.9	6.7	17.5	8.6	0.31
1993	11.0	6.7	17.7	8.8	0.28
1994	11.8	7.0	18.8	8.3	0.32
1995	9.3	6.3	15.6	7.9	0.28
1996	9.5	6.8	16.3	7.8	0.33

Acknowledgments

We gratefully acknowlege and thank all study participants. This work was supported in part by grants from Health Science Research Grants (Research on Environmental Health)

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