# NATIONAL SURVEY ON CONTAMINATION LEVEL OF PCDD/DFS AND RELATED COMPOUNDS IN HUMAN BREAST MILK COLLECTED FROM KOREA

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#### Introduction

Chlorinated hydrocarbons, such as polychlorinated biphenyls(PCBs) and polychlorinated dibenzo-p-dioxins and dibenzofurans(PCDD/DFs) are detected in high concentrations in human tissues from some developed and industrialized countries. These toxic compounds have attracted considerable attention regarding public health. However, the contamination statuses of these toxic compounds are not clear in the general population in Korea. Therefore, Korea Food & Droug Administration has been coordinating monitoring program on PCDD/DFs and Dioxin-like PCBs since the last years. The 4th round of the KFDA-coordinated exposure study has been initiated in March 2002. The aim of this study was to measure the contamination levels and to investigate the residue profiles of PCDD/DFs and dioxin-like PCBs in human breast milk collected from residents of Seoul and Busan area in Korea. Furthermore, we investigated the variation of PCDD/DFs and dioxin-like PCB levels in human milk during lactation.

### **Materials and Methods**

### Sample Collection

The human breast milk samples analyzed in this study were collected at maternal clinics in Seoul city (a metropolitan area) and Busan city (a port area). As donors of human breast milk, 55 mothers in total were chosen ; 36 mothers from were Seoul (mean age :  $31 \pm 3.7$ years) and 19 mothers from Busan (mean age :  $30 \pm 5.1$ years). The human milk samples were collected in about 50mL each time on 0, 10, 30 and 150 day after delivery. The samples were immediately frozen in clean glass bottles and kept frozen at -20 until analysis.

Extraction and clean-up of milk samples were made according to the slightly modified method of US EPA 1613 and CDC (Centers for Disease Controls & Prevention). Identification and quantification of dioxin like PCBs was performed by HRGC-HRMS (Jeol-700D, Japan).

### **Result and Discussion**

# Contamination level of PCDD/DFs and dioxin like PCBs

The mean concentrations of individual PCDD/DF and dioxin-like PCB congeners in the human breast milk samples collected from Seoul and Busan are shown in Table 1. The total concentrations of PCDD/DFs and dioxin-like PCB were determined in 26 colostrums samples,

Sampling Area		Seoul city				Busan city			
Days after delivery (day)		colostrum	10th	30th	150th	colostrum	10th	30th	150th
Number of donors		26	24	22	22	20	13	6	6
Lipid content(%)		2.02	2.36	2.6	2.79	2.56	2.61	2.5	2.48
PCDDs	2378-TeCDD	0.44	0.28	0.09	0.07	0.46	0.17	0.36	ND
	12378-PeCDD	2.28	0.36	0.59	0.84	2.65	2.44	0.48	0.69
	123478-HxCDD	2.13	1.06	0.78	1.58	2.09	1.07	1.01	0.78
	123678-HxCDD	5.39	3.66	2.39	3.75	4.7	3.98	3.81	1.04
	123789-HxCDD	1	0.23	0.08	0.21	0.75	0.45	0.39	0.05
	1234678-HpCDD	9.59	3.76	1.57	3.17	5.8	3.98	3.32	0.33
	OCDD	51.16	41.53	34.81	26.86	59.25	49.7	46.63	27.83
PCDFs	2378-TeCDF	1.19	1.03	0.43	0.21	3.19	0.86	0.67	0.21
	12378-PeCDF	1.79	0.14	0.04	0.43	1.56	0.38	ND	ND
	23478-PeCDF	5.48	4.39	3.32	2.94	5.98	4.89	5.09	1.95
	123478-HxCDF	4.81	3.02	2.86	2.04	3.47	3.37	2.5	1.24
	123678-HxCDF	3.86	2.2	2.2	1.72	2.76	2.92	2.29	1.12
	123789-HxCDF	1.69	0.24	ND	0.15	0.95	0.4	ND	ND
	234678-HxCDF	4.97	0.85	0.55	0.75	2.48	1.28	0.78	0.05
	1234678-HpCDF	9.11	1.88	0.73	0.76	3.67	2.7	2.97	2.33
	1234789-HpCDF	1.75	0.3	ND	0.14	1.09	0.58	ND	ND
	OCDF	3.77	1.95	ND	ND	4.16	ND	ND	ND
	Total PCDD/DFs	110.42	66.85	50.43	45.6	105	79.19	70.31	37.6
	PCDD/DFs-TEQ	8.27	4.12	3.3	3.48	8.33	6.58	4.6	2.14
Non-	344`5(81)	3.7	3	1.2	1.1	3.7	3	1.2	1.1
ortho CBs	33`44`(77)	21.5	16.6	13.7	12.2	21.5	16.6	13.7	12.2
	33`44`5(126)	30	19.3	12.9	12.8	30	19.3	12.9	12.8
	33`44`55`(169)	15.6	10.7	9.1	6.3	15.6	10.7	9.1	6.3
Mono-	2`344`5(123)	307.4	230.7	213.5	117.2	307.4	230.7	213.5	117.2
ortho CBs	23`44`5(118)	3521.4	2549.6	2156.7	2122.6	3521.4	2549.6	2156.7	2122.6
	2344`5(114)	255.1	195.2	150.8	130.4	255.1	195.2	150.8	130.4
	233`44`(105)	1657.4	849.6	629.1	571.3	1657.4	849.6	629.1	571.3
	23`44`55`(167)	418.6	381.1	191.5	193.4	418.6	381.1	191.5	193.4
	233`44`5(156)	1134.5	839.9	690.1	556	1134.5	839.9	690.1	556
	233`44`5`(157)	358.6	289.6	266.9	174.3	358.6	289.6	266.9	174.3
	233`44`55`(189)	111.7	108.8	19.5	16.2	111.7	108.8	19.5	16.2
	Total.Dioxin-like PCBs	7835.4	5494.1	4355.1	3913.8	7835.4	5494.1	4355.1	3913.8
	Dioxin-like PCBs-TEQ	. 4.6	. <u>3.08</u>	2.24	2.06	4.6 .	3.08	WHO TEE	2.06
	2,3,7,0-10000	ionic equ	Invalents(TEQ)	were	calculate	u using	uie	WHU-IEF	•

Table 1. Average concentrations of PCDD/DFs and dioxin-like PCBs (pg/g lipid weight basis) in human breast milk sample	es
collected from Korea.	

24 samples of human breast milk sampled at the 10th day after delivery, 24 samples at 30th day and 21 samples at 150th day, all collected from Seoul area. The mean Total-TEQ concentrations of PCDD/DFs and dioxin-like PCBs ( $\sum$ PCDD/DFs-TEQ +  $\sum$ Dioxin-like PCBs-TEQ) in colostrums and samples collected at, 10th day, 30th day and 150th day after delivery were 12.9, 7.2, 5.5 and 5.5 pg WHO-TEQ/g, respectively. Furthermore, the total concentrations of PCDD/DFs and dioxin-like PCB congeners were determined in 20 colostrums samples, 13 samples of human milk sampled at the 10th day after delivery, 6 samples at 30th day and 6 samples at 150th day, all collected from Busan area. The mean Total-TEQ concentrations in colostrums, and samples collected at 10th day, 30th day and 150th day after delivery human milk samples were 11.5, 9.6, 6.8 and 3.8 pg WHO-TEQ/g, respectively. In general, we found that the residue levels of PCDD/DFs and dioxin-like PCB congeners in human milk samples collected from Seoul and Busan area were similar.

### The temporal trend of Toxic Equivalents in the Korean population

In recent years, several studies have reported declining tendency of PCDD/DFs and related compounds in human breast milk collected from some developed country <sup>1,2,3</sup>. However, the temporal changes of PCDD/DF and related compound levels in the Korean population were not clear, until now. In table 2, the monitored levels in Korean human milk are compared with those from human breast milk surveys performed in 1999 and 2002. When expressed on I-TEQ basis, the 1999 survey showed an average total TEQ level of 32 pg I-TEQ/g(lipid weight basis) which is 65 % higher than the average of 11 pg I-TEQ/g in 2002. However, this declining tendency was not shown in the case of PCDD/DFs and coplanar PCB-TEQ.

Year	1999	2000	2001	2002	
Sampling Area	Seoul	Seoul, Chungbuk	Seoul, Chungbuk	Seoul, Busan	
Sampling Time	Colostrum	5th day after birth	Colostrum	Colostrum	
Number of donors	59	66	43	46	
PCDD/DFs-TEQ	31.8	10.2	3.63	7.12	
Coplanar-PCBs-TEQ**	0.19	2.6	6.44	3.82	
Total TEQ ***	31.99	12.8	10.1	10.9	

Table 2. Temporal trend of PCDD/DF and Coplanar-PCB-TEQ in human milk samples collected from Korea (from 1999 to 2002).

\* 2,3,7,8-TeCDD toxic equivalents(TEQ) were calculated using the I-TEF.

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** Coplanar-PCBs -TEQ : # 77 + # 126 + #169
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\*\*\*Total TEQ :  $\Sigma$  PCDDs -TEQ +  $\Sigma$  PCDFs-TEQ +  $\Sigma$  Coplanar PCBs-TEQ

# The decline trend of residue levels during the lactation periods.

Both PCDD/DFs and dioxin-like PCBs TEQ concentrations in human breast milk collected from Seoul were significantly reduced during the period of observation in this study. This declining tendency of residues was also observed samples collected from Busan area (Fig. 1).

Donors from Seoul had collected milk samples from their first lactation to 150 days after delivery. The total concentrations of PCDD/DFs and dioxin-like PCBs were reduced during that period by about 59% and 50%, respectively. On the other hand, the decline tendency was not observed with the fat content in human milk over the lactation period. There have been several investigations on the changes in organochlorine residue levels in human milk during lactation, which demonstrated a gradual decrease of organochlorine residue levels in milk during lactation period <sup>4,5)</sup>. Our results were in general agreement with these references.



Fig. 1. The decline in residue concentrations of Total –TEQ ( $\sum$  PCDD/DFs -TEQ +  $\sum$  Dioxin-like PCBs-TEQ ) during the lactation periods of donors from Seoul and Busan.

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