

## CURRENT STATUS OF MATERNAL AND FETAL EXPOSURE TO BROMINATED FLAME RETARDANTS, PCBS AND DIOXINS IN JAPAN

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### Introduction

We are investigating on fetal contamination by persistent organic pollutants (POPs) and in our previous papers, we insisted that the best sample to assess the fetal exposure to persistent chemicals would be umbilical cord, not cord serum because the assessment using cord serum may result in an underestimation of the level of chemicals<sup>1,2</sup>. The purpose of this current study was to detect and calculate the human fetal exposure to brominated flame retardants, polybrominated diphenyl ethers (PBDEs), using umbilical cord. We also compared the data of concentration level of polychlorinated biphenyls (PCBs), chlorinated dioxins {polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and coplanar-PCBs (co-PCBs)}, in umbilical cord, cord blood and maternal blood.

### Materials and Methods

#### (1) Samples

Fifty sets of samples (umbilical cord, cord blood and maternal blood) were collected at the delivery in Chiba University Hospital (Japan) from 2002 to 2004. This study was approved by the "Congress of Medical Bioethics" of Chiba University and University of Yamanashi, and all the samples were obtained after receipt of written informed consent.

#### (2) Chemicals measured

Twenty seven congeners of PBDEs (#3, 7, 15, 17, 28, 47, 49, 66, 71, 85, 77, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, 191, 196, 197, 207, 206 and 209), twenty nine congeners of dioxins (7 PCDDs, 10 PCDFs, 4 non-ortho co-PCBs, 8 mono-ortho co-PCBs) and PCBs (10 groups of PCB congeners summed up by their number of chlorines from 1 to 10) were measured by GC/MS.

#### (3) Analysis

Fifty sets of samples were used individually for dioxin and PCB measurement. For measurement of PBDEs, dioxins and PCBs, two mixed sample sets (sample set A and set B) were used. Sample set A was consisted of the tissues from 5 volunteers, and sample set B was consisted of the tissues from 3 volunteers. However, the maternal serum of sample set A was not measured because the amount was not enough to measure. The umbilical cord samples were homogenized with ethanol/hexane (1:3) and sodium sulfuric anhydride by a Polytron PT3100. After filtration, the filtrate and an additional filtrate of the re-homogenate of the residue were washed with water twice. The resulting hexane extract was dehydrated using sodium sulfuric anhydride and concentrated by evaporation (crude extract). The maternal serum and cord serum samples were extracted twice using an ether/hexane (3:1) mixture. The resulting ether/hexane extract was dehydrated using sodium sulfuric anhydride and concentrated by evaporation (crude extract). The crude extract was purified through a silica gel column for PCB measurement, through a silica gel column

and a charcoal column for dioxins and PBDEs measurement.

## Results and Discussion

### (1) Dioxins and PCBs

Dioxins were detected in all three tissues (Table 1). On measured basis, almost all dioxins (>97 %) were mono-ortho PCBs such as 2,3',4,4',5-PentaCB (#118), 2,3,3',4,4',5-HexaCB (#156) and 2,3,3',4,4'-PentaCB (#105). On TEQ basis, major congeners were 1,2,3,7,8-PentaCDD (about 30 %), 2,3,4,7,8- PentaCDF (about 20 %), 3,3',4,4',5-PentaCB(#126) (about 15 %), 1,2,3,6,7,8- HexaCDD (about 10 %), 2,3,7,8-TetraCDD and 2,3,3',4,4',5-HexaCB (#156).

**Table 1.** Total dioxin concentrations in the three types of tissues. Upper: on wet basis (pg/g-wet), lower: on fat basis (ng/g-fat)

	n	Mean ± SD	Median	Min. - Max.
Umbilical Cord	42	8.7 ± 3.6 9.9 ± 4.6	9.6 7.1	2.6 - 23 2.2 - 21
Cord Serum	10	12 ± 6.0 4.8 ± 2.7	11 5.3	3.5 - 21 1.3 - 9.2
Maternal Serum	21	61 ± 34 11 ± 5.1	57 10	22 - 160 4.2 - 24

PCBs were also detected in all three tissues (Table 2). Comparing the congeners according to the number of chlorines, the level of HexaCBs were the highest in all tissues, followed by PentaCBs and HeptaCBs. Concentrations on fat basis, HexaCBs, PentaCBs and OctaCBs were higher in umbilical cord than maternal serum.

**Table 2.** Total PCB concentrations in the three types of tissues. Upper: on wet basis (pg/g-wet), lower: on fat basis (ng/g-fat)

	n	Mean ± SD	Median	Min. - Max.
Umbilical Cord	42	88 ± 65 83 ± 57	71 73	29 - 390 32 - 350
Cord Serum	42	170 ± 120 82 ± 66	140 65	56 - 780 30 - 390
Maternal Serum	42	880 ± 940 110 ± 99	590 78	220 - 5700 29 - 570

### (2) Brominated flame retardants

Mixed samples were used for PBDE concentration measurement. Dioxin and PCB concentrations in the same mixed samples were also measured, and the value was in the range of the value measured individually. PBDEs were detected in all types of samples (Table 3). Total concentrations of PBDEs measured were 110 pg/g-wet in maternal serum and 6.3 pg/g-wet in umbilical cord A, 1.9 pg/g-wet in umbilical cord B. DecaBDE (#209) was 56pg/g-wet in maternal serum, 55 pg/g-wet in cord serum A, 23 pg/g-wet in cord serum B, but it was very low (N.D.) in umbilical cord. Top 6 PBDE congeners in cord serum A depending on the number of bromine atom were, from the highest, DecaBDE (#209), TetraBDE (#47) (9.8 pg/g-wet), PentaBDE (#99)(9.5 pg/g-wet), HexaBDE (#153)(2.2 pg/g-wet), HexaBDE (#154) (1 pg/g-wet) and PentaBDE (#100)(1.8 pg/g-wet).

Hites et al.<sup>3</sup> and Ryan and Oostdam<sup>4</sup> measured PBDEs in maternal and cord serum each at Indianapolis and at northern Canada. Both groups did not measure DecaBDE, however, both studies showed that TetraBDE (#47) and PentaBDE (#99) were higher than other congeners. In comparison with the results of major PBDE congeners (e.g., #47, #99) analyzed between the studies, our results indicated that the concentration levels of PBDEs were lower in Japan than in North America.

**Table 3.** Total and individual congener concentrations of PBDEs in the three types of tissues. Upper: on wet basis (pg/g wet), lower: on fat basis (ng/g fat)

	Umbilical Cord A	Umbilical Cord B	Cord Serum A	Cord Serum B	Maternal Serum A
total PBDEs	6.3	1.9	83	43	110
	5	1.7	35	19	20
TetraBDE (#47)	2.3	0.58	9.8	6.2	14
	1.8	0.52	4.2	2.7	2.6
TetraBDE (#49)	0.19	N.D.	0.35	0.22	0.53
	0.15	N.D.	0.15	0.097	0.096
PentaBDE (#99)	0.94	0.28	9.5	8.3	8.1
	0.75	0.25	4.1	3.6	1.5
PentaBDE (#100)	0.46	0.17	1.8	1.3	3.1
	0.37	0.15	0.76	0.56	0.56
HexaBDE (#153)	0.8	0.29	2.2	1.3	9.1
	0.64	0.26	0.95	0.55	1.7
HexaBDE (#154)	0.21	N.D.	1	0.8	2.1
	0.17	N.D.	0.45	0.35	0.38
DecaBDE (#209)	N.D.	N.D.	55	23	56
	N.D.	N.D.	23	10	10

In this study, we measured PBDE concentrations including DecaBDE in three types of human tissues (umbilical cord, cord serum, maternal serum). Higher concentration ratios of cord to maternal serum observed for PBDEs (>1) than for chlorinated dioxins and PCBs (<1) suggest that PBDEs transfer from mother to fetus through placenta more easily than those chlorinated compounds. On the other hand, transfer from blood to tissues (cord) seems to occur more easily in the case of chlorinated dioxins and PCBs than PBDEs. The fact that DecaBDE existed large amount in cord serum, but is very low in umbilical cord, is an important finding about fetal exposure and transfer of PBDEs among tissues.

## References

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