CONGENER SPECIFIC DETERMINATION OF PCBs IN HUMAN BREAST MILK COLLECTED FROM HOKKAIDO, JAPAN

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Abstract
We determined concentrations of 73 PCB isomers in breast milk collected from 60 mothers about one month after parturition from Hokkaido prefecture during 2004. PCBs were detected in all the samples collected. Among PCB isomers, mean concentrations of PCB#153, #138, #180, #118 and #182 were higher than those of other isomers. In this study, we also determined PCB isomers concentrations in the blood collected from the same mothers to compare with those in the breast milk. Similar to breast milk, mean concentrations of PCB#153, #138, #180, #118 and #182 in the blood were higher than those of other isomers. Concentrations of total PCBs and dominant PCB isomers in the breast milk showed significant positive correlations with those in the blood. These results suggest long-term accumulation of persistent PCBs in humans despite the ban on usage of PCBs for more than three decades. The health risks to infants by lactational exposure to PCBs are of concern.

Introduction
Polychlorinated biphenyls (PCBs) have been of great concern because of their worldwide distribution, high persistency, potential for magnification in the food web, and toxic effects on human health and wildlife. In recent years, there has been a growing concern over the potential effects of PCBs on infants via breast milk. Thus, many investigations on levels of PCBs in human breast milk have been conducted to assess risks for infants all over the world [1,2]. In the present investigation, we analyzed 73 PCB isomers and determined their concentrations in human breast milk collected from Hokkaido prefecture in the north of Japan during 2004.

Materials and Methods
The breast milk samples were collected from primipara (n = 30, 21–40 years: mean; 30.1 years) and pluripara (n = 30, 21–47 years: mean; 32.2 years) about one month after parturition from Hokkaido prefecture during 2004. No statistically significant difference was observed between age of primipara and pluripara. The samples were stored at -20^{o}C until analysis. The details of the analysis for breast milk (2 g) including lipid extraction, purification and mass-spectrometric measurements have been described elsewhere[3]. PCB concentrations are expressed on a lipid weight basis (ng/g lipid wt.) in the present study.

Results and Discussion
PCBs were detected in all the breast milk samples analyzed with total PCB concentrations ranging from 31.1–387 ng/g lipid wt. and a mean of 106 ng/g lipid wt (Table 1). Concentration of Total PCB in breast milk from Hokkaido was lower than that from other areas of Japan and developed countries (Fig. 1) [2]. Among the PCB isomers, average concentrations of PCB#153, #138, #180, #118 and #182 were higher

Table 1. Concentrations of PCBs (ng/g lipid wt.) in breast milk collected from Hokkaido, Japan (2004).

	Compounds	No.	Mean	S.D.	Maximum	Minimum
TriCBs	244'-TrCB	28	0.93	0.41	2.75	0.25
	245-TrCB	29	<0.01	<0.01	<0.01	<0.01
	344'-TrCB	37	<0.01	<0.01	<0.01	<0.01
TetraCBs	22'35'-TeCB	44	0.10	0.05	0.27	<0.01
	22'44'-TeCB	47	0.22	0.11	0.61	0.06
	22'45'-TeCB	49	0.08	0.04	0.26	0.02
	22'55'-TeCB	52	0.39	0.36	2.07	<0.01
	233'4'-/2344'-TeCBs	56/60	0.31	0.17	1.12	0.06
	234'5-TeCB	63	0.06	0.03	0.23	<0.01
	23'44'-TeCB	66	0.84	0.48	3.15	0.17
	23'4'5-TeCB	70	0.06	0.05	0.30	<0.01
	23'4'6-TeCB	71	0.04	0.04	0.20	<0.01
	244'5-TeCB	74	4.05	2.74	20.0	0.85
	33'4'4'-TeCB	77	0.01	0.00	0.03	<0.01
	344'5-TeCB	81	0.01	0.00	0.01	<0.01
PentaCBs	22'344'-PeCB	85	0.10	0.07	0.32	<0.01
	22'345'-PeCB	87	0.23	0.13	0.80	0.01
	22'355'-PeCB	92	0.34	0.21	1.10	0.07
	22'35'6-PeCB	95	0.25	0.15	0.75	0.03
	22'44'5-PeCB	99	4.69	2.51	16.6	0.95
	22'455'-PeCB	101	0.76	0.45	2.52	0.16
	233'44'-PeCB	105	1.82	0.02	6.95	0.30
	233'4'5-PeCB	107	0.39	0.25	1.67	0.04
	233'4'6-PeCB	110	0.18	0.12	0.77	0.03
	2344'5-PeCB	114	0.44	0.01	1.71	0.09
	234'56-PeCB	117	0.29	0.18	1.10	0.06
	23'44'5-PeCB	118	7.58	0.08	29.1	1.44
	2'344'5-PeCB	123	0.13	4.22	0.53	0.02
	33'44'5-PeCB	126	0.04	0.26	0.16	<0.01
HexaCBs	22'33'44'-HxCB	128	0.40	0.24	1.29	0.07
	22'33'45'-HxCB	130	0.85	0.50	3.28	0.15
	22'33'46'-HxCB	132	0.04	0.07	0.34	<0.01
	22'33'56-HxCB	134	0.01	0.01	0.06	<0.01
	22'33'56'-HxCB	135	0.18	0.10	0.54	0.03
	22'344'5-HxCB	137	0.84	0.49	3.19	0.22
	22'344'5'-HxCB	138	14.0	7.63	51.3	3.95
	22'344'6-/22'34'5'6-HxCBs	139/149	0.28	0.07	0.90	<0.01
	22'3455'-HxCB	141	0.12	0.17	0.41	0.01
	22'34'55'-HxCB	146	3.69	2.12	13.3	0.75
	22'34'56-HxCB	147	0.14	0.08	0.42	<0.01
	22'355'6-HxCB	151	0.43	0.24	1.17	0.05
	22'44'55'-HxCB	153	25.2	14.4	92.3	6.92
	233'44'5-HxCB	156	2.08	1.02	7.84	0.42
	233'44'5'-HxCB	157	0.48	0.45	1.86	0.10
	233'4'5'6-HxCB	164	4.18	2.26	14.3	1.05
	233'55'6-HxCB	165	<0.01	<0.01	<0.01	<0.01
	23'44'55'-HxCB	167	0.76	1.22	3.18	0.19
	33'44'55'-HxCB	169	0.02	0.28	0.06	<0.01
HeptaCBs	22'33'44'5-HpCB	170	3.80	2.23	13.7	0.99
	22'33'455'-HpCB	172	0.57	0.34	1.94	0.15
	22'33'4'56-HpCB	177	1.28	0.78	4.89	0.23
	22'33'55'6-HpCB	178	1.32	0.80	4.77	0.31
	22'33'566'-HpCB	179	0.07	0.05	0.22	<0.01
	22'344'55'-HpCB	180	10.2	5.84	34.5	2.81
	22'344'56-HpCB	181	0.01	0.01	0.05	<0.01
	22'344'56-HpCB	182	5.13	3.08	18.4	1.36
	22'344'5'6-HpCB	183	1.28	0.69	4.18	0.42
	233'44'55'-HpCB	189	0.19	0.11	0.67	0.05
	233'44'5'6-HpCB	191	0.12	0.07	0.44	0.03
OctaCBs	22'33'44'55'-OcCB	194	0.94	0.56	3.09	0.24
	22'33'44'56-OcCB	195	0.25	0.14	0.85	0.08
	22'33'45'66'-OcCB	200	0.05	0.03	0.19	0.01
	22'33'455'6-/22'33'4'55'6-OcCBs	201/198	1.12	0.22	4.46	0.27
	22'33'55'66'-OcCB	202	0.33	0.74	1.28	0.09
	22'344'55'6-OcCB	203	0.92	0.55	3.19	0.26
	233'44'55'6-OcCB	205	0.04	0.02	0.15	0.01
NonaCBs	22'33'44'55'6-NoCB	206	0.20	0.13	0.75	0.04
	22'33'44'566'-NoCB	207	0.04	0.02	0.12	<0.01
	22'33'455'66'-NoCB	208	0.08	0.06	0.32	<0.01
DecaCB	22'33'44'55'66'-DeCB	209	0.08	0.06	0.38	0.01
	Total TrCBs		0.94	0.53	2.75	<0.01
	Total TeCBs		6.15	1.14	20.0	<0.01
	Total PeCBs		17.3	2.20	29.1	<0.01
	Total HxCBs		53.8	6.32	92.3	<0.01
	Total HpCBs		24.0	3.13	34.5	<0.01
	Total OcCBs		3.64	0.46	4.46	<0.01
	Total NoCBs		0.32	0.08	0.75	<0.01
	Tota PCBs		106	57.8	387	31.1
	Lipid (%)		3.97	1.10	7.05	1.29
	Age (year)		31.2	5.45	21	47

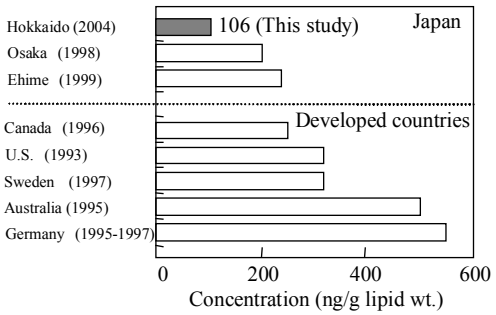

Fig. 1. Comparison of total PCBs concentration in human breast milk from Japan and developed countries[2].

than those of other isomers in the breast milk. The sum total of these five isomers concentrations contributed about 60% to the total PCB concentrations (Fig. 2).

In the present investigation, no significant difference in levels of total PCBs in the breast milk were observed between primipara (mean; 110 ng/g lipid wt.: range; 47.2–387 ng/g lipid wt.) and pluripara (mean; 102 ng/g lipid wt.: range; 31.1–284 ng/g lipid wt.). Generally, in the case of primipara, it is shown that levels of PCBs in human breast milk were higher than pluripara [4]. On the other hand, there was a significant positive correlation between concentrations of total PCBs in breast milk and mother's age (r=0.65, p<0.001), in agreement with previous studies[2]. These results indicate long-term accumulation of persistent PCBs in mothers and that it might pose health risks to infants.

We also determined PCB isomers concentrations in the blood collected from the same mothers to compare with those in the breast milk, which was determined by the method reported by Hori et al[3]. The mean total PCB concentration in the blood was 134 ng/g lipid wt. (range: 38.2–353 ng/g lipid wt.) and was significantly higher than that in the breast milk (p=0.008) (Fig.3).

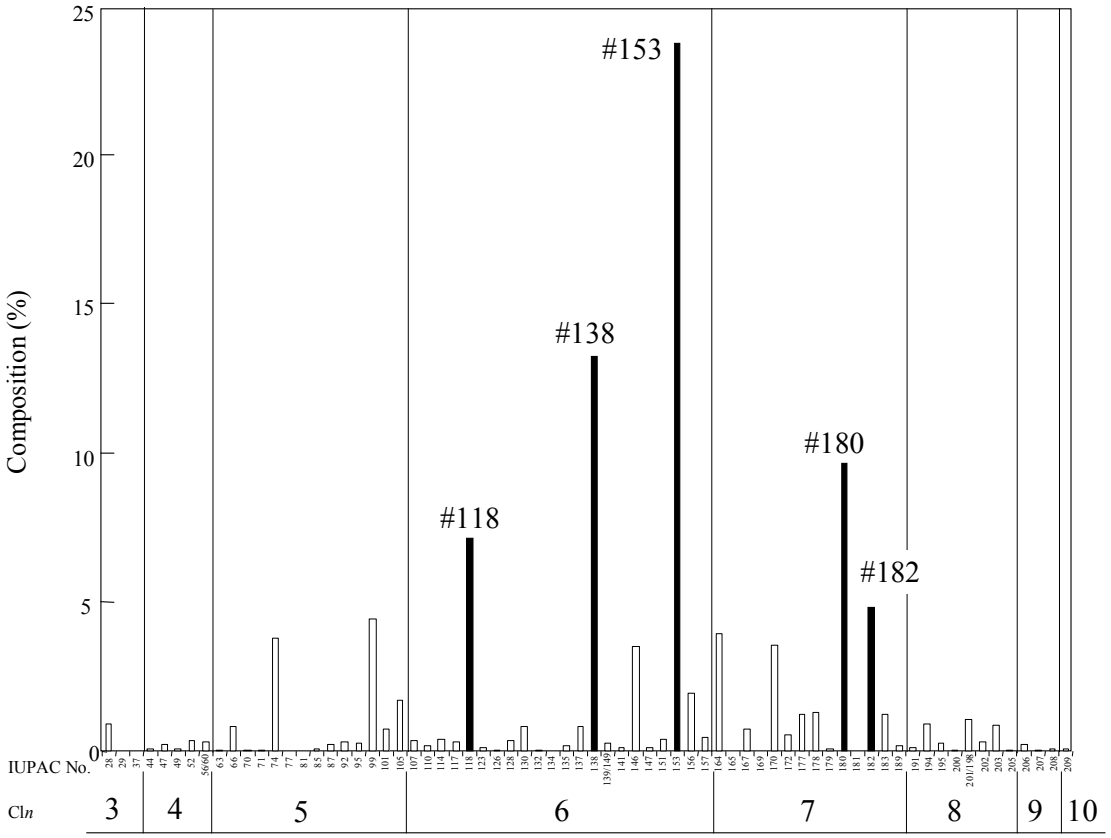

Fig. 2. PCB isomers and congener composition of breast milk from Hokkaido in 2004. Vertical bars represent concentrations of individual congeners relative to total PCBs.

In addition, the dominant isomers in blood samples were similar to those in breast milk. Concentrations of total PCBs and dominant PCB isomers in the breast milk showed significant positive correlations with those in the blood (Fig. 4). These results suggest that PCBs accumulated in the blood may be transferred to the breast milk. On the other hand, breast milk to blood concentration ratios of each PCB isomer tended to significantly decrease with increase of octanol/water partition coefficient (K_{ow}) (Fig. 5, r=-0.72, p<0.001). This result may suggest that higher lipophilic PCBs (higher chlorinated PCBs) in blood are not easily transferred to breast milk.

Our results recommend continuous investigations on concentrations of PCBs in breast milk to assess health risk of infants in Japan.

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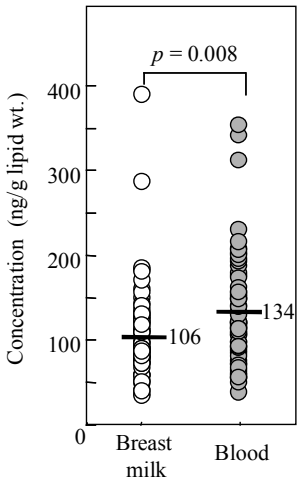

Fig. 3. Comparison of total PCBs concentrations in breast milk and blood. The cross bars indicate mean.

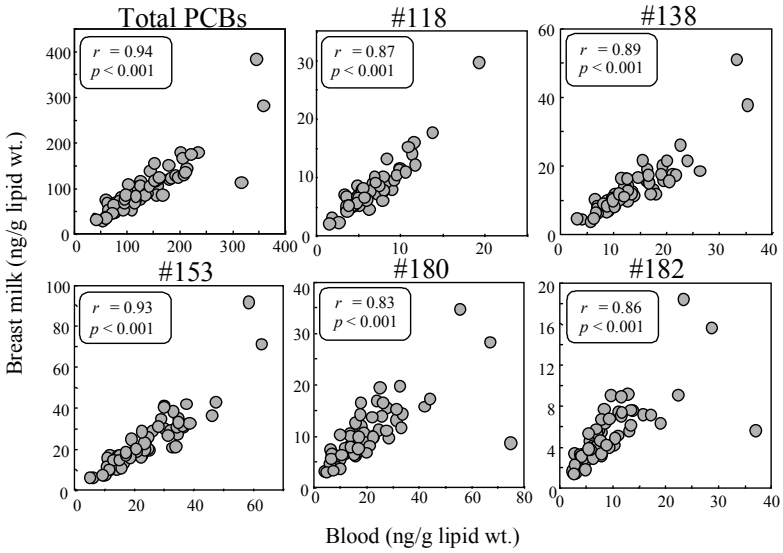

Fig. 4. Relationships between PCBs concentrations in breast milk and blood.

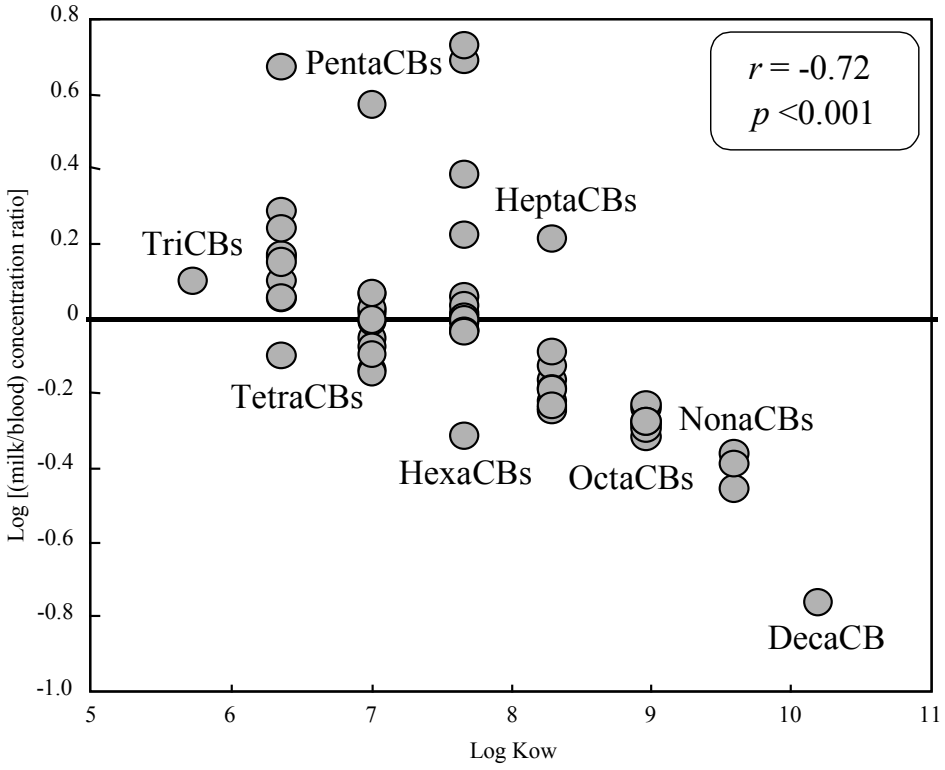

Fig. 5. Relationship between Kow and milk/blood concentration ratios of PCBs congeners.