

LEVELS OF POLYBROMINATED DIPHENYL ETHERS IN BREAST MILK ASSOCIATED WITH THE CONSUMPTION OF FISH AND SHELLFISH IN CENTRAL TAIWAN

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Introduction

Polybrominated diphenyl ethers are a group of brominated compounds with the wide use as brominated flame retardants¹. Currently, little is known about the sources of human exposure to PBDEs and the adverse health effects for public health². Breast milk and house dust had been indicated the possibly main exposure pathways for PBDEs intake of breastfed infants after delivery³. In the Great Boston's study, there was a statistically significant and positive correlation between PBDE concentrations in house dust and breast milk⁴. Infants with prenatal and postnatal exposure to PBDEs might be associated with PBDEs coated on house dust in their parent's home. Up to date, the data are limited to suggest that breast milk is a significant source of infant exposure, but inhalation and dust may be important.

Breast milk is the only food for the breastfed infants. Biomonitoring of PBDEs in breast milk provides a good marker of exposure from the environment and the food chain especially for mothers and their nursing infants. From our previous study⁵, PBDE levels in breast milk and the daily intake of a breastfed infant in Taiwan were no higher than the reports from other countries. In a recent biological study in Taiwan⁶, the contaminants of PBDEs in fish samples caught from Taiwan were reported to have higher magnitudes compared to those from European countries. The goal of the present study was examine the association between PBDEs in breast milk and the consumption of animal fat in Taiwanese general population. The indicators of exposure from the dietary habits and socioeconomic statuses were also determined.

Materials and Methods

Participants were selected from the cohort of the dioxin survey described previously⁵. The study protocol was approved by the Institutional Review Boards (IRB) of the Human Ethical Committee at the National Health Research Institutes (NHRI), Taiwan. Ethical standards formulated from the Helsinki Declarations of 1964 and revised in 2000 were followed. All participants gave informed consent after receiving detailed explanations prior

to enrollment. Participants, who were healthy pregnant women, were recruited from a medical center in central Taiwan between December 2000 and November 2001. Our participants answered a detailed questionnaire including maternal age, maternal weight before pregnancy, maternal height, socioeconomic status (i.e. home annual income), smoking and dietary habits (i.e. the frequent and amount consumption of fish), drinking alcohol, and medical and pregnancy history. 176 individual breast milk samples were collected with the provision of breast milk. 20 subjects were further randomly selected.

Breast milk was collected approximately two weeks after the delivery and milk samples were stored at -20°C refrigerator. The milk samples of 25 ml were shipped frozen to ERGO laboratory in Germany for chemical analysis. Chemical analysis for PBDEs in breast milk was described in detail previously⁵. The extract was identified and quantified by high-resolution gas chromatograph equipped with high-resolution mass spectrometry (HRGC/HRMS: HP GC5890 II / VG-AutoSpec) using a DB-5 column (J&W Scientific) for gas chromatographic separation. Typical HRGC and HRMS conditions had been published previously⁵. The congeners of 12 PBDEs (IUPAC 17, 28, 47, 66, 85, 99, 100, 138, 153, 154, 183, 209) were measured to determine the levels in human milk and the correlated factors.

We defined PBDEs as the sum of 9 individual congeners without BDE-17, 66, and 138 based on all the measurements of these congeners lower than LODs. Log-normality of PBDEs was used for further statistical analyses by the Kolmogorov-Smirnov method. Spearman's rank correlation coefficients were initially used to find the correlations between individual PBDEs and subjects' demographic characteristics (systolic blood pressure) or the associated factors (i.e. fish consumption). We compared PBDE levels in breast milk with dietary habits, socioeconomic status, and demographic characteristics. When two groups (i.e. parity) were compared, Student *t*-tests were used. Analysis of variance (ANOVA) and covariance (ANCOVA) and Bonferroni post-hoc test were used for comparisons among the socioeconomic groups (e.g., education level). Multiple linear regressions were used to evaluate differences in the groups (i.e. eating fish) after maternal age, maternal pre-pregnant BMI, and parity were adjusted. Analyses were carried out using the Statistical Package for Social Science (SPSS) 13.0 version.

Results and Discussion

The participants of the present study were described previously in detail elsewhere⁵. Maternal age was not correlated with PBDE levels in breast milk in Table 1. The significant differences were found in the congeners of BDE-28 and BDE-183 between the lower ($<22\text{ kg/m}^2$) and higher ($\geq 22\text{ kg/m}^2$) body mass index (BMI) groups. Multipara mothers had a significantly higher BDE-28 level than Primopara mothers did ($p=0.029$). There was a significant difference in BDE-28 concentrations between higher and lower systolic blood pressure groups

($p=0.045$), but no significant difference was presented after maternal age, pre-pregnant BMI, and parity were adjusted ($p=0.113$). When maternal age, pre-pregnant BMI, and parity were adjusted, we still found that women with higher diastolic blood pressure (≥ 76 mmHg) had a higher PBDE magnitude than those with lower pressure (<76 mmHg) ($p=0.038$).

Table 2 was shown that levels of PBDEs were in the different socioeconomic groups. There were no significant differences in PBDE concentrations between lower and higher annual income groups before and after the adjustment of maternal age, pre-pregnant BMI, and parity. We found that PBDE levels in breast milk were positively associated with women with higher education levels particularly for the statistical significance of BDE-47, BDE-100, and BDE-153. There were no significant differences in individual PBDEs and total PBDEs among these three education level groups. Women reported Taoist had higher BDE-28 levels than those reported other religion after maternal age, pre-pregnant BMI, and parity ($p=0.010$).

Women who consumed more fish (≥ 4 times/week) had a higher PBDE level than those who consumed less ones (<4 times/week), but there were no statistical differences in these two groups after maternal age, pre-pregnant BMI, and parity were adjusted. Body burden of BDE-85 in the higher meat consumption group (≥ 11 times/week) was higher compared to those in the lower consumption group after the adjustment of maternal age, pre-pregnant BMI, and parity ($p=0.025$). The finding of the present study was indicated that PBDEs in breast milk was significantly correlated with the lower consumption of shellfish group ($p=0.002$) after maternal age, pre-pregnant BMI, and parity were adjusted.

Table 1. PBDE levels in Taiwanese breast milk (ng/g lipid)

	BDE28	BDE47	BDE85	BDE99	BDE100	BDE153	BDE154	BDE183	BDE209	PBDEs
Maternal age										
<29 yr (n=9)	0.078 ^a	1.21	0.023	0.351	0.339	0.741	0.145	0.067	0.170	3.40
≥ 29 yr (n=11)	0.067	1.48	0.024	0.451	0.377	0.834	0.097	0.059	0.152	3.78
<i>p</i> value	0.575	0.397	0.945	0.481	0.383	0.583	0.310	0.617	0.826	0.584
pre-pregnant BMI										
< 22 kg/m ² (n=13)	0.069	1.59	0.018	0.439	0.378	0.884	0.148	0.077	0.192	4.07
≥ 22 kg/m ² (n=7)	0.077	0.995	0.010	0.351	0.327	0.643	0.075	0.044	0.113	2.89
<i>p</i> value	0.732	0.041 [*]	0.159	0.521	0.281	0.148	0.089	0.038 [*]	0.308	0.081
Parity										
Primipara (n=17)	0.0635	1.28	0.026	0.427	0.357	0.803	0.109	0.064	0.181	3.56
Multipara (n=3)	0.143	1.83	0.014	0.303	0.375	0.726	0.169	0.058	0.078	3.88
<i>p</i> value	0.029 [*]	0.270	0.254	0.462	0.777	0.740	0.424	0.789	0.227	0.756
Systolic pressure										
<121 mmHg (n=8)	0.049	1.22	0.024	0.408	0.351	0.845	0.083	0.072	0.168	3.48
≥ 121 mmHg (n=12)	0.093	1.37	0.026	0.423	0.343	0.683	0.117	0.052	0.128	3.46
<i>p</i> value ^b	0.045 [*]	0.674	0.880	0.928	0.886	0.361	0.421	0.269	0.633	0.986
<i>p</i> value ^c	0.113	0.998	0.551	0.714	0.728	0.383	0.655	0.310	0.957	0.887
Diastolic pressure										
<76 mmHg (n=7)	0.062	1.38	0.045	0.546	0.376	0.894	0.075	0.094	0.239	3.97
≥ 76 mmHg (n=13)	0.076	1.28	0.019	0.373	0.335	0.691	0.116	0.049	0.115	3.28
<i>p</i> value ^b	0.584	0.789	0.019 [*]	0.365	0.468	0.306	0.533	0.028 [*]	0.230	0.432
<i>p</i> value ^c	0.779	0.705	0.145	0.524	0.410	0.452	0.418	0.038 [*]	0.480	0.484

^a Geometric mean

^b Without any adjustment

^c Maternal age, prepregnant BMI, and parity were adjusted by multiple linear regression.

* $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$

Table 2. PBDE levels in breast milk from the different socioeconomic groups (ng/ g lipid)

	BDE28	BDE47	BDE85	BDE99	BDE100	BDE153	BDE154	BDE183	BDE209	PBDEs
Annual income										
< \$18000 (n=8)	0.094 ^a	1.344	0.025	0.364	0.328	0.757	0.124	0.070	0.149	3.54
≥ \$ 18000 (n=12)	0.060	1.356	0.023	0.436	0.382	0.814	0.112	0.058	0.167	3.66
<i>p</i> value ^b	0.098	0.969	0.892	0.598	0.183	0.738	0.802	0.503	0.826	0.854
<i>p</i> value ^c	0.271	0.635	0.260	0.758	0.604	0.678	0.521	0.473	0.911	0.660
Education level^d										
≤ High school (n=8)	0.080	1.41	0.023	0.356	0.362	0.752	0.120	0.080	0.158	3.65
College (n=8)	0.069	1.02	0.020	0.344	0.308	0.633	0.095	0.046	0.112	2.83
≥ University (n=4)	0.063	2.18	0.037	0.733	0.482	1.36	0.164	0.071	0.330	5.74
<i>p</i> value ^b	0.817	0.033*	0.050	0.184	0.022*	0.015*	0.596	0.124	0.283	0.013*
<i>p</i> value ^c	0.450	0.219	0.848	0.311	0.254	0.250	0.246	0.307	0.784	0.228
Maternal Religion										
Taoist (n=5)	0.111	1.28	0.022	0.364	0.338	0.634	0.120	0.029	0.099	3.17
Buddhist (n=6)	0.096	2.02	0.023	0.425	0.468	1.04	0.203	0.073	0.247	4.96
Other (n=9)	0.046	1.07	0.025	0.417	0.312	0.745	0.079	0.053	0.155	3.14
<i>p</i> value ^b	0.007**	0.041*	0.054	0.963	0.934	0.192	0.105	0.521	0.398	0.079
<i>p</i> value ^c	0.010*	0.260	0.971	0.978	0.285	0.895	0.415	0.319	0.847	0.554

^a Geometric mean^b Without any adjustment^c Maternal age, prepregnant BMI, and parity were adjusted by multiple linear regression^d Maternal education level**p*<0.05, ***p*<0.01, ****p*<0.001

Table 3. The deference in PBDE levels between the different food consumption groups

	BDE28	BDE47	BDE85	BDE99	BDE100	BDE153	BDE154	BDE183	BDE209	PBDEs
Eating fish										
<4 times/week (n=4)	0.071 ^a	0.887	0.017	0.270	0.294	0.682	0.114	0.041	0.113	2.73
≥4 times/week (n=16)	0.072	1.50	0.026	0.449	0.378	0.820	0.117	0.070	0.174	3.87
<i>p</i> value ^b	0.984	0.057	0.390	0.211	0.007**	0.492	0.955	0.005**	0.498	0.142
<i>p</i> value ^c	0.657	0.161	0.370	0.330	0.216	0.666	0.852	0.060	0.789	0.482
Eating meat										
<11 times/week (n=10)	0.072	1.61	0.015	0.353	0.379	0.916	0.172	0.063	0.128	3.96
≥11 times/week (n=10)	0.072	1.14	0.038	0.465	0.346	0.682	0.079	0.062	0.198	3.29
<i>p</i> value ^b	0.992	0.126	0.012*	0.404	0.403	0.162	0.038*	0.981	0.388	0.335
<i>p</i> value ^c	0.305	0.393	0.025*	0.506	0.580	0.173	0.054	0.897	0.627	0.541
Eating shellfish										
<9 times/month (n=15)	0.082	1.48	0.025	0.402	0.380	0.892	0.151	0.072	0.193	3.98
≥9 times/month (n=5)	0.056	0.811	0.017	0.316	0.275	0.453	0.039	0.039	0.059	2.15
<i>p</i> value ^b	0.249	0.023*	0.412	0.549	0.001**	0.006**	0.118	0.046*	0.049*	0.004**
<i>p</i> value ^c	0.604	0.019*	0.281	0.497	0.026*	<0.001***	0.015*	0.029*	0.029*	0.002**

^a Geometric mean^b Without any adjustment^c Maternal age, prepregnant BMI, and parity were adjusted by multiple linear regression**p*<0.05, ***p*<0.01, ****p*<0.001

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