Residue Levels of Polybrominated Diphenyl Ethers (PBDEs) in Food and Human Milk

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Introduction

Polybrominated diphenyl ethers (PBDEs) are chemicals widely used as flame retardants, because they dissolve in polymers , however many other kinds of flame retardants are available. There are 209 congeners of PBDEs exist all of which have specific chemical and physical properties, which lead to various biological and toxicological effects[1]. The clinical signs PBDE exposure include reduced the growth, diarrhoea, piloerection, tremors of forelimbs, red staining around eyes and nose, and continuous chewing[2].

PBDEs are existed in wide range of environments, and may enter the body in by many routes. The purpose of this study was to measure the PBDE contamination levels in food and human milk samples collected from Korea.

Materials and Methods

We used the standard analytical method of sample pretreatment with PBDEs to determine a recovery ratio. The target isomers in this research were selected between tri-BDE and hepta-BDE (#28, # 47, # 99, # 100, # 163, # 154, # 183) because they are used in electrical applications and their bioaccumulation properties were high. PBDE measurement was performed with an Agilent 6890 gas chromatograph and Jeol-700D high resolution mass spectrometer at resolution 10,000 using DB-5HT (30m x 0.25mm x 0.1 μm) capillary column.

Six species of fish (mackerel, hairtail, flatfish, anchovy, croaker and pollack) and two kinds of shellfishs (oyster and clam) were selected. Human milk was collected in the cities of Seoul and Jeju.

Pretreatment for food samples was applied that alkali digestion used for 1 N KOH/EtOH, multi-layer silica gel and activated carbon impregnated silica gel column were used for clean-up of sample. Elution solvent for each clean-up stage was 10 % and 25% DCM in n-Hexane, respectively. Digestion for breast milk samples was used small quantity of sodium oxalate, sodium chloride and ethanol of 20 mL. Clean-up method was the same for food samples.

Results and Discussion

The result of recovery test for clean-up stage was shown in table 1. Recovery rate of most target isomer was above 50%. In case of BDE-28 and 153, recovery ratio is lower than other isomer, however recovery ratio of clean-up stage include alkalii digestion and column clean-up, all target isomer was collected about 70%. This result shown that low recovery ratio of BDE-28 and 153 in activated carbon column was not due to elution condition. So suggested method in this study was to correct for pretreatment PBDEs samples.

The mean PBDE levels in food samples are shown in table 2. Classification PBDEs accumulation level, that were detected highest level in flatfish which it intakes without any cook processing. Detected level in pollack was lower than other species. Mackerel, which has the highest fat content, was shown t have lower levels than species with low fat content. Accumulation level of shellfishes (oyster and clam) was lower than fishes.

Both fish and shellfish samples, occupation ratio of BDE-47 were above 50% (not illustrated). And it had no difference between fish and shellfish samples about occupation ratio of BDE-28, fish and shellfish were 7.2 and 10.8%, respectively. However, in case of BDE-183 in high brominated compound occupation ratio in shellfish was higher than fish samples. The occupation levels in fish and shellfish were 0.5 and 15%, respectively.

Reference data from other countries were shown in table 3. PBDEs was detected from 190 to 410 ng/g (lipid base) at Baltic salmon muscle. And Lake trout in ontario was contaminated 945 ng/g. It show that PBDEs level in Korea

was lower than case in Europe or west country. Also in case of Japan, PBDEs level in mackerel was higher than this study. PBDE levels in cultured natural species were higher than free living organisms which may be due to feed and culture condition.

Table 1. Result of recovery test for treatment condition

(% sample added)

	BDE-28	BDE-47	BDE-100	BDE-99	BDE-154	BDE-153	BDE-183
Alkali digestion	108.15	103.74	82.01	97.69	104.12	100.37	103.68
Activated Carbon column	54.81	72.58	100.61	91.56	93.20	59.73	60.29
Clean-up stage	104.89	95.41	70.88	99.06	83.96	93.96	86.91

Table 2. PBDE concentration of fish and shellfish samples collected from Korea

(PBDE pg/g wet weight)

Samples	BDE-28	BDE-47	BDE-100	BDE-99	BDE-154	BDE-153	BDE-183
Flatfish(n=3)	42.41	507.90	154.18	95.57	64.16	27.17	ND
Pollack(n=3)	3.00	22.32	4.33	7.08	1.89	1.63	1.48
Hairtail(n=5)	12.51	87.65	29.83	18.73	33.65	10.57	0.54
Croaker(n=4)	62.23	341.57	59.51	54.84	83.28	22.51	0.73
Mackerel(n=4)	3.78	24.74	9.50	9.50	9.14	3.91	ND
Anchovy (pooled)	18.03	215.67	9.93	27.44	14.90	7.33	4.84
Oyster(pooled)	7.63	60.30	6.91	10.46	7.43	2.16	0.98
Clam(pooled)	9.17	34.38	4.84	14.06	5.97	4.13	1.17

ND : Not Detected

PBDE levels in breast milk were shown in table 4. Levels of BDE-47 which bioaccumulation property is high, was not difference between Seoul and Jeju sample. The interested fact in breast milk, BDE-154 and 183 was not detected and occupation ratio of BDE-153 was higher than case of fish and shellfish samples.

Reference data of Japan[6] were illustrated in Fig 1. The figure shown that PBDE level was increased, progressively in same area. This result was same with the result at sweden[7]. The PBDEs level which is detected Japan Kanagawa on 1999 had many difference in spite of it is and identical area. The occupation ratios of each isomer were same with the result of our research, and total PBDEs level was almost same.

Table 3. Level of PBDEs in fish and shellfish from various countries.

Class of samples	Concentration (ng/g lipid)	Target congener
Baltic salmon muscle[3]	190 ~ 410	47, 99, 100, 153, 154
Lake trout(1998, L. Ontario)[4]	945	47, 99, 100
Yellow tail(cultured natural)[5]	1.58 ~ 1.72 (ww)	28, 47, 99, 100, 153, 154
Mackerel(natural)[5]	1.28 ~ 1.55 (ww)	28, 47, 99, 100, 153, 154
Yellow tail(natural)[5]	0.99 ~ 1.32 (ww)	28, 47, 99, 100, 153, 154

ww : wet weight

Table 4. PBDEs concentration in human milk samples collected from Seoul and Jeju

(PBDE Congener pg/g lipid weight)

Sampling Area	Congeners						
	BDE-28	BDE-37	BDE-100	BDE-99	BDE-153	BDE-154	BDE-183
Seoul (n=20)	113.81	1,082.32	438.01	226.61	ND	720.63	ND
Jeju (n=20)	157.88	1,167.95	404.23	349.92	ND	1,209.85	ND

ND : Not Detected

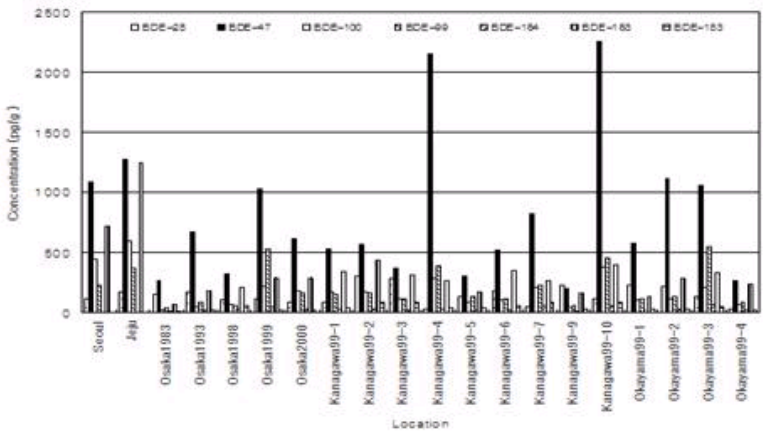

Fig. 1. Comparison of each congener between Seoul, Jeju and Japan(Akutsu, 2003)

Conclusion

This research was initial investigation of PBDEs level in food and human breast milk at Korea. This result will proffer important information about exposure route of PBDEs to human body and method which pretreatment samples. After this study, we have to estimate PBDEs level in food and human samples continually, in order to find increasing tendency properties at Korea.

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