

DIETARY EXPOSURE TO PBDEs IN KOREA

Seo JJ¹, Shin JH¹, Jung KK², and Kang IH²

¹Korea Basic Science Institute, Seoul, 136-701 Korea; ²National Institute of Toxicological Research, Seoul, 122-704 Korea

Introduction

PBDEs have come under increased scrutiny because of their potential to impact upon the environment and human health. Food is major exposure source of persistent organic pollutants (POPs). Over 90% of human exposure of dioxin and PCBs is estimated to occur through the diet¹. Given the lipophilic nature of PBDEs and their presence in consumer products and house dust, suspected routes of human exposure include both diet and the indoor environment². Unlike other POPs, however, the key routes of human exposure are not thought to be food, but rather are from their use in household consumer products³. The indoor environment and diet both play prominent roles in human exposure to PBDEs.

A market basket study is a useful method for estimating the average intake level of PBDEs in regions, based on a model of the average total diet. It is possible to provide information for the daily intake of food groups. Preliminary estimates of mean intake of PBDEs, based on a limited number of samples from Canada, some European countries, Japan, and the USA, as reported in published studies and reports, range from 13 to 113 ng/day⁴. These monitoring provide current estimates of the exposure of human to PBDEs and are a valuable tool to improve risk assessments and to develop the appropriate strategies to manage risks that may be associated with these contaminant.

In the present study, we analyzed brominated dioxins and PBDEs in food mixtures from each of 13 food group from 3 regions (Seoul, Ulsan, and Ansan) in Korea and estimated daily intake levels of PBDEs.

Materials and Methods

The food and food group consumption data used in the composition of the market baskets and in the intake calculations consist of the average consumption figures taken from the 2007 National Health and Nutrition Examination Survey.⁵ Selected products were purchased at supermarkets and local markets. Fish and meat products were also purchased at a local fish store and a local butcher's. The samples were homogenized immediately after collection and stored at $-20\text{ }^{\circ}\text{C}$ until further treatment. In case of food items where wastage could be supposed, inedible parts such as bone, skin, etc. were removed prior to homogenization.

A 300-g aliquot of sample was homogenized, and a 10-g aliquot was spiked with the ¹³C-labeled compounds. The sample was mixed with anhydrous sodium sulfate, dried for a minimum of 30 minutes, and extracted for 18-24 hours in a Soxhlet extractor. The extract was evaporated to dryness, and the lipid content was determined. After extraction, a labeled cleanup standard was spiked into the extract and the extract was concentrated. Tissue extracts are first cleaned

up using an anthropogenic isolation column, and all extracts are cleaned up using back-extraction with sulfuric acid, and gel permeation, silica gel, as required. PBDE values are based on analyses of the 27 PBDE congeners BDE-3, 7, 15, 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, 191, 196, 197, 206, 207 and 209 in 44 food samples by gas chromatography-isotope dilution high resolution mass spectrometry.

Table 1 Description of food items (and their matching food groups) sampled in Korean market baskets (n = 9), purchased in three different cities in Korea.

<i>No.</i>	<i>Food group</i>	<i>Main items (%)</i>	<i>Daily Intake g/day</i>
1	Cereal products	Rice, noodle, ramen noodles, bread, mixed grain	283.0
2	Potatoes and products	Potato	30.3
3	Sweeteners	Sugar	7.6
4	Seasonings	Soybean paste, soy sauce	29.9
5	Pulses and bean products	Bean curd	37.7
6	Vegetables	Chinese cabbage, radish, onion, bean sprouts, tomato, leek, spinach, zucchini, cucumber	287.5
7	Fruit	Orange, apple, strawberry	175.7
8	Beverages & Alcohol	Beer, beverages, Soju, coffee, Korean rice wine	153.1
9	Oil & fats	Beans oil, butter	7.2
10	Meat & products	Pork, beef, chicken	93.9
11	Egg	Egg	21.2
12	Fish and shellfish	Boiled fish paste, mackerel, croaker, Alaska pollack, clam, anchovy, flatfish	52.0
13	Dairy products	Milk, ice cream	88.6
	Total		1283.2

Results and Discussion

A food market-basket, representative for the general Korean population, containing various meat, fish and dairy food products, was assembled and analyzed by gas chromatography-high resolution mass spectrometer for its polybrominated diphenyl ether (PBDE) content. The concentrations, as pg/g fresh weight (fw), of PBDEs in 13 market baskets and in the total diet basket are presented in Table 2. We could not observe any differences in PBDE concentrations between the foods from the different supermarkets and cities, therefore averages were calculated using all analyses regardless of the origin. However, we have found a wide variation in sum of PBDE congeners concentrations across the food groups sampled.

Fish and shellfish had the highest average PBDEs levels (2.83 - 539.66 pg/g ww), followed by meat & products (20.36 - 153.57 pg/g ww), egg (16.49 pg/g ww), and oil and fat (16.89-127.18 pg/g ww). PBDE intake calculations were based on the average daily food consumption in Korea and were estimated. In the sum of PBDEs the major contributors to daily intake were fish and shellfish (30%), meat & products (36%), cereal products (16%), and dairy products (9%).

Table 2 Lower bound concentration of PBDEs in food as pg/g wet weight in Korea.

Food Group	Food	Average	SD	Median	Min	Max
Cereal products	rice	12.99	3.55	11.71	6.86	29.72
	noodle	18.71	7.64	15.71	10.26	31.69
	ramen noodles	12.70	2.28	12.32	4.92	23.16
	bread	16.98	2.81	19.20	2.40	34.93
	mixed grain	7.61	1.37	8.08	1.41	15.24
Vegetables	Chinese cabbage	4.08	1.27	3.33	2.01	10.20
	radish	5.09	1.27	4.82	3.23	7.20
	onion	5.85	0.10	5.88	4.72	6.65
	bean sprouts	3.08	0.24	3.73	ND	5.06
	tomato	5.32	0.40	4.89	4.33	7.07
	leek	6.59	0.35	6.57	5.61	8.67
	spinach	5.04	0.68	4.94	4.13	6.05
	zucchini	4.43	0.84	4.57	3.02	6.09
	cucumber	4.82	0.16	4.58	3.65	6.35
Potatoes	potato	6.44	1.64	6.31	4.29	9.05
Sweeteners	sugar	3.72	1.31	3.42	1.73	7.20
Beverages & Alcohol	beer	1.07	0.80	0.17	0.07	4.36
	beverages	1.42	0.16	1.57	0.98	1.77
	Soju	1.45	0.41	1.42	ND	3.30
	coffee	1.03	0.70	0.68	0.19	3.68
	Korean rice wine	1.35	0.51	1.19	0.18	2.66
Bean products	bean curd	4.40	2.62	2.77	0.98	9.84
Egg	egg	16.49	7.22	13.56	6.62	30.21
Fruit	orange	6.77	1.12	7.01	5.18	9.82
	apple	3.27	1.13	4.31	0.42	6.19
	strawberry	8.87	1.67	8.08	5.51	13.66
Oil & fats	beans oil	42.72	8.37	43.68	28.04	54.67
	butter	52.32	4.79	25.33	16.89	127.18
Meat	pork	117.90	20.76	121.77	57.97	153.57
	beef	46.62	2.73	44.58	37.22	55.57
	chicken	39.81	6.68	40.71	20.36	54.63
Fish and shellfish	boiled fish paste	66.50	0.49	73.51	22.18	99.26
	mackerel	155.57	80.36	138.27	54.97	288.92
	croaker	105.67	46.18	116.93	15.41	170.18
	Alaska pollack	84.78	2.89	86.23	2.83	157.28
	clam	35.70	4.39	36.09	28.42	48.23
	anchovy	250.21	62.98	262.99	3.59	539.66
	flatfish	113.54	33.55	101.03	57.25	201.43
Dairy products	milk	25.91	0.76	27.14	19.08	30.62
	powdered milk	10.58	2.78	12.34	3.22	14.77
	ice cream	35.75	8.24	36.40	17.41	57.36
Seasonings	soybean paste	7.23	2.89	6.57	3.69	11.56
	soy sauce	3.38	5.71	1.19	ND	18.30

Multivariation analysis was carried to fine the distribution pattern of PBDE congeners in food. Most of food was influenced by BDE209 source. Fish and shellfish showed similar pattern with pentabromodiphenyl oxide (TBDE71).

The intake calculations were based on a theoretical estimate of the average daily food consumption. The sum of PBDEs intake was 25.8pg/day in the total diet basket.

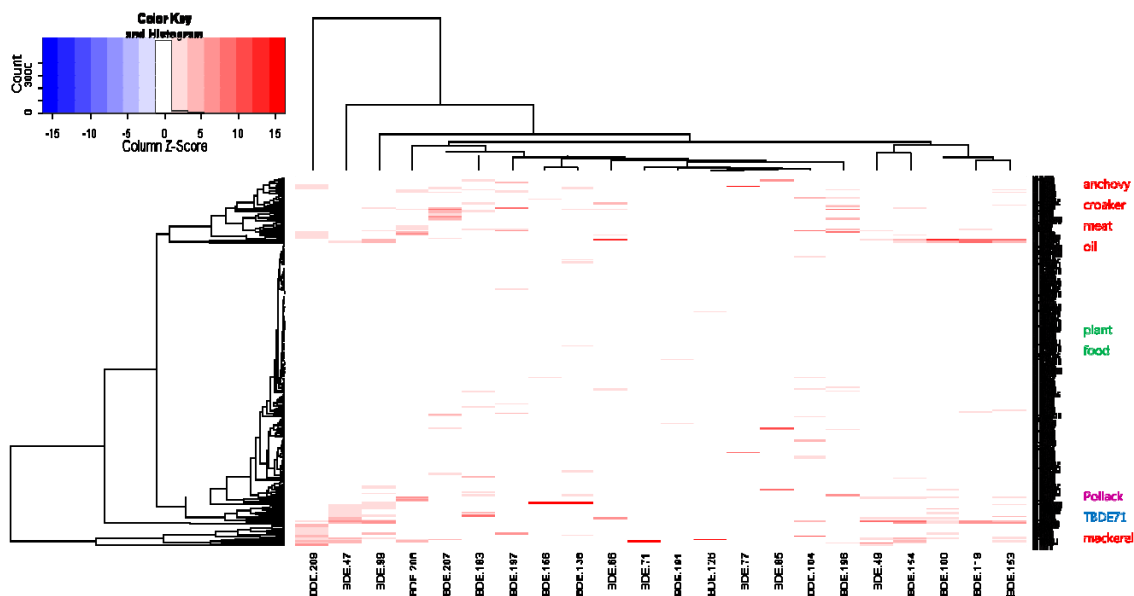


Fig. 1 HCA clusters and heat map of food for PBDEs

Acknowledgements

This research has been funded Korean Food and Drug Administration.

References

1. Liem, A.K., Furst, P., Rappe, C. (2000); *Food Addit. Contamin.* 17: 241-259
2. Wu, N., Herrmann, T., Joeltickn r, O., Hale, R., Harey, E., Guardia, M. L., Mcclean , M. D., Webster, T. F (2007); *Environ. Sci. Technol.* 41: 1584-1589
3. Lorber, M. (2008) *J. of Exposure Science and Environmental Epidemiology* 18: 2-19
4. WHO (World Health Organization). WHO Technical Report Series 930: Evaluation of certain food contaminants. Geneva: Joint FAO/WHO Expert Committee on Food Additives; 2006.
5. Ministry for Health, Welfare and Family Affairs, The Third Korea National Health & Nutrition Examination Survey-Nutrition Survey 2007