ESTIMATION OF THE DAILY INTAKE OF POLYCHLORINATED BIPHENYLS FROM DIETARY EXPOSURE IN SOUTH KOREA

JONG-CHUL KIM, MIN-HUI SON, JUN-TAE KIM, EUN-JIN PARK, AND YOON-SEOK CHANG*

School of Environmental Science and Engineering, Pohang University of Science and Technology (POSTECH), San 31, Hyoja-dong, Nam-gu, Pohang, 790-784, Republic of Korea

Abstract
The dietary intake of polychlorinated biphenyls (PCBs) was estimated using the sum of 62 PCB congeners (∑62PCBs), including 7 indicator PCBs and 12 dioxin-like PCBs, in the South Korea. In this study, 200 individual food samples belonging to 40 different foodstuffs were investigated to estimate the distribution of PCB congeners in five sampling cities. PCB exposure was estimated using Korean dietary habits as established by the National Health and Nutrition Examination Survey (NHANES). The PCB concentrations in rice, the most frequently consumed food in Korea, was relatively low in whole food samples. The mean PCB levels measured in fish were the highest in this study, but each fish is consumed in relatively small amounts by the general population. Therefore, the daily dietary intake should also be considered with regard to human exposure to PCBs, especially with the consumption of contaminated foods. Dioxin-like PCB levels were also calculated using TEF values that were established in 2005. The average levels (pg TEQ/g) were 0.0002 for rice and 0.0098 for fish. The dioxin-like PCBs accounted for a relatively small percentage of the total PCBs, compared to previous studies. According to our research, the health risks associated with exposure to PCBs could be estimated using the tolerable daily intake (TDI) of the general population.

Introduction

Polychlorinated biphenyls (PCBs), a class of compounds consisting of 209 individual congeners, are considered to be highly representative of persistent organic pollutants (POPs). They have been extensively used in many industrialized countries as lubricants, plasticizers, flame retardants and insulating oils for capacitors and transformers because of their insulating and nonflammable properties [1]. In Korea, it is estimated that more than 5,000 tons of imported PCBs have been used over the past 2 decades, which may have leaked into the environment during the process of discarding the PCB containing products [2,3]. Accordingly, PCB residues, which have been attributed to potential adverse health effects, such as developmental toxicity, endocrine disruption and certain types of cancer, could still exist in various environmental media [4]. For these reasons, significant PCB monitoring in a wide diversity of environmental matrices is currently being considered[5, 6]. Nevertheless, the actual amounts of PCB-containing commercial products that had been imported have not been released so far, and thus the amounts can be estimated through environmental monitoring..

In the present study, we primarily investigated the distribution patterns of PCBs in various foodstuffs according to the eating habits of foodstuffs that are consumed by the general Korean population. Moreover, the comparative significances of specific food groups as they relate to the total intake of PCBs were estimated. In contrast to previous studies, we examined the degrees of contamination of various foodstuffs that were ingested by the general population and, not a specific, focused food groups, such as fish and meat, in light of the dietary habits of Koreans. This study also scrutinized the PCB levels of basic foodstuffs per se including 62 PCB congeners from mono- to deca-CBs. Furthermore, we assessed the amounts of PCBs exposure to the human body using the national dietary habits established by the National Health and Nutrition Examination Survey (NHANES) [6]. To our best knowledge, this is the first time that estimation of overall PCB exposure based on daily intake of staple foodstuffs has been reported in Korea.

Materials and Methods

Materials: All organic solvents used (i.e., acetone, methylene chloride, n-hexane and toluene) throughout this

* Corresponding author phone: (82)-54-279-2281; fax:(82)-54-279-8299; e-mail: yschang@postech.ac.kr

study were HPLC grade (Honeywell Burdick & Jackson, USA). Internal and recovery standards were purchased from Wellington Laboratories, Inc. (Guelph, Ontario, Canada). The 13C-labeled PCBs used as internal standard were PCB-3, 15, 31, 52, 118, 153, 180, 194, 206, and 209. The recovery standards that were used included PCB-19, 54, 104, 167, and 170. Silica was acquired from Supelco, Korea. and anhydrous sodium sulfate (Na2SO4, purity 98%) was purchased from Kando Chemical Co., Inc.

Sample collection: To evaluate the concentration of PCBs in foodstuffs, 200 individual food samples representing 40 different foodstuffs were collected from major supermarkets located in 5 different regions: Seoul, Daejeon, Gwangju, Gangneung and Busan.

Chemical analysis: Each sample aliquot was thoroughly mixed with anhydrous sodium sulfate (Na2SO4) to eliminate residual water and was then spiked with the internal standards. In the cases of the vegetables and fruits, these were extracted using acetone/hexane (1:4, v/v). The remaining samples were extracted with methylene chloride/hexane (1:1, v/v). To extract the 62 congeners of interest, we employed ultrasonication for half an hour two times. Next, the cleanup was performed with multi-layered silica columns

Instrument analysis: HRGC/HRMS analysis was performed using an Agilent 6890 gas chromatography that was connected to a JMS 800D mass spectrometer (JEOL, Japan) at a resolution of 10, 000 in selected ion monitoring (SIM) mode using the molecular ions of individual compounds. For all analyses, the HRMS was operated in the positive ionization mode at 38 eV and a 0.25 mm (i.d.) x 60 m DB-5MS capillary column (0.25 μm film thickness, J&W Scientific, USA) was used to separate the analytes.

Results and Discussion

Characteristics of PCB distribution patterns in each foodstuff

Compared with previous studies undertaken in Korea and in other countries, the number of congeners and food items in this study were increased to comprehend the overall exposure levels of PCBs [7, 8, 9, 10]. 62 PCB congeners (PCB-1, 3, 4, 8, 10, 15, 18, 19, 22, 33, 37, 44, 49, 54, 70, 74, 87, 95, 99, 104, 110, 112, 128, 149, 151, 155, 158, 168, 170, 171, 177, 178, 183, 187, 188, 191, 194, 199, 201, 202, 205, 206, 208, and 209 including 12 dioxin-like PCBs and 7 indicator PCBs) were measured in whole food samples. The total PCB concentrations and maximum PCB exposure concentrations caused by food intake, which were based on estimated daily intake obtained by the NHANES, are summarized in Table 1.

TEQ concentrations of dioxin-like PCBs

In the present study, the contributions of dioxin-like PCBs in each food group were not as great compared to non-dioxin-like PCB congeners. PCB-126 and PCB-169, which have the highest TEFs among the dioxin-like PCBs (i.e., 0.1 and 0.03, respectively) were not detected in any foodstuffs. This might be due to the amounts of samples used for analysis compared to previous studies [7, 8, 11]. Apart from fish, a few congeners, including PCB118, were detected in vegetables, fruits, milk, and even meat. Rice and eggs, however, only contained PCB-118 at low concentrations, and the sum of WHO-TEQ levels was also low.

As mentioned above, dioxin-like PCB contamination levels in fish and shellfish were quite high, and the amount of each congener that was detected varied as compared to other food groups. The contribution levels of fish in whole food groups was not great due to the low daily intakes of individual fish. To better understand the substance of this study, a comparison of the total TEQ concentrations in fish is summarized in Table 2. In one study [12], aqua-cultured and wild fish were investigated to estimate the levels of dioxin-like PCBs. The aqua-cultured fish had a higher lipid content, which might have been due to the commercial feed, and consequently had higher PCB levels than the wild fish. In general, lipid content has an effect on the accumulation of dioxin-like PCBs. It has previously been shown that PCB levels in fatty tissue were about 5 times higher than in lean tissue [13], which was also shown in this study (Table 1)

Table 1 Estimated dietary intake of indicator PCBs, dioxin-like PCBs and 62 total PCBs in selected Korean foodstuff

Food groups	Food items	Estimated daily intake (g/day)[a]	Lipid(%)	Concentration (ng/g wet weight)			Concentration (ng/person/day)[e]			Concentration (ng/kg body weight/day)[f]			Ratio (%)[g]
				$\sum_7$PCBs[b]	$\sum_{12}$PCBs[c]	$\sum_{62}$PCBs[d]	$\sum_7$PCBs	$\sum_{12}$PCBs	$\sum_{62}$PCBs	$\sum_7$PCBs	$\sum_{12}$PCBs	$\sum_{62}$PCBs	
	Potato	20.6	0.0	0.215	0.030	0.574	4.429	0.618	11.824	0.081	0.011	0.215	1.8
Vegetables	Kimchi	79.4	0.0	0.358	0.057	0.918	28.425	4.526	72.889	0.517	0.082	1.325	11.3
	Onion	23.4	0.0	0.289	0.038	0.701	6.763	0.889	16.403	0.123	0.016	0.298	2.5
Grains	Rice	183.2	0.0	0.324	0.023	0.903	59.357	4.214	165.430	1.079	0.077	3.008	25.5
Legumes	Tofu	21.7	0.0	0.305	0.040	0.730	6.619	0.868	15.841	0.120	0.016	0.288	2.4
Noodles	Noodle	12.8	0.0	0.305	0.019	0.884	3.904	0.243	11.315	0.071	0.004	0.206	1.7
Fruits	Apple	29.6	0.0	0.304	0.035	0.775	8.998	1.036	22.940	0.164	0.019	0.417	3.5
	Mandarin orange	21.7	0.0	0.313	0.021	0.867	6.792	0.456	18.814	0.123	0.008	0.342	2.9
Eggs	Egg	22.5	11.1	0.228	ND	0.740	5.130	ND	16.650	0.093	ND	0.303	2.6
Milk	Milk	73.3	2.0	0.090	ND	0.233	6.597	ND	17.079	0.120	ND	0.311	2.6
	Cheese	0.1	10.3	0.161	ND	0.401	0.016	ND	0.040	0.000	ND	0.001	0.0
Dairy products	Ice cream	6.9	3.1	0.189	ND	0.582	1.304	ND	4.016	0.024	ND	0.073	0.6
	Yogurt	7.3	0.2	0.133	ND	0.434	0.971	ND	3.168	0.018	ND	0.058	0.5
	Dried anchovy	2.5	5.6	1.666	0.301	3.877	4.165	0.753	9.693	0.076	0.014	0.176	1.5
	Cero	0.6	9.6	5.746	0.001	13.867	3.448	0.001	8.320	0.063	ND	0.151	1.3
	Clam	1.3	1.7	0.787	0.058	2.105	1.023	0.075	2.737	0.019	0.001	0.050	0.4
	Crab	1.6	3.1	1.449	0.229	3.090	2.318	0.366	4.944	0.042	0.007	0.090	0.8
	Eel	1.2	7.5	2.022	ND	4.102	2.426	ND	4.922	0.044	ND	0.089	0.8
	Fish paste	6.2	3.2	0.385	0.022	0.990	2.387	0.136	6.138	0.043	0.002	0.112	0.9
	Hair tail	2.4	6.7	1.943	ND	6.409	4.663	ND	15.382	0.085	ND	0.280	2.4
Fish and Shellfish	Mackerel	4.3	15.0	1.699	ND	4.958	7.306	ND	21.319	0.133	ND	0.388	3.3
	Oyster	0.8	2.5	0.608	0.048	1.576	0.486	0.038	1.261	0.009	0.001	0.023	0.2
	Polluck	2.6	0.2	0.547	0.044	1.356	1.422	0.114	3.526	0.026	0.002	0.064	0.5
	Salmon	0.4	13.1	7.670	0.001	15.865	3.068	ND	6.346	0.056	ND	0.115	1.0
	Pacific saury	1.1	23.2	1.343	ND	3.035	1.477	ND	3.339	0.027	ND	0.061	0.5
	Squid	4.1	0.4	0.516	0.033	1.299	2.116	0.135	5.326	0.038	0.002	0.097	0.8
	Canned tuna	1.6	0.9	0.243	0.020	0.483	0.389	0.032	0.773	0.007	0.001	0.014	0.1
	Yellow corvina	1.7	8.0	1.365	ND	3.684	2.321	ND	6.263	0.042	ND	0.114	1.0
	Beef	28.3	9.4	0.370	ND	0.855	10.471	ND	24.197	0.190	ND	0.440	3.7
	Bovine liver	0.0	1.6	0.237	ND	0.709	0.000	ND	0.000	0.000	ND	0.000	0.0
	Chicken	12.5	0.5	0.039	ND	0.103	0.488	ND	1.288	0.009	ND	0.023	0.2
Meat	Ham	2.9	18.0	0.164	ND	0.404	0.476	ND	1.172	0.009	ND	0.021	0.2
	Lean Pork	35.6	18.3	0.129	ND	0.361	4.592	ND	12.852	0.083	ND	0.234	2.0
	Pork belly	7.1	28.6	0.204	ND	0.514	1.448	ND	3.649	0.026	ND	0.066	0.6
	Mixed sausage	0.9	3.0	0.177	ND	0.548	0.159	ND	0.493	0.003	ND	0.009	0.1
Animal fat	Butter	0.1	74.9	0.449	ND	0.818	0.045	ND	0.082	0.001	ND	0.001	0.0
Plant fat	Soy oil	3.5	95.9	0.573	ND	1.964	2.006	ND	6.874	0.036	ND	0.125	1.1
Seaweeds	Seaweed	2.9	0.0	0.540	ND	1.299	1.566	ND	3.767	0.028	ND	0.068	0.6
Condiment	Soybean paste	6.3	0.0	0.559	ND	1.488	3.522	ND	9.374	0.064	ND	0.170	1.4
Sugars	Sugar	4.4	0.0	0.268	0.019	0.754	1.179	0.084	3.318	0.021	0.002	0.060	0.5
Sum		**639.4**		**34.912**	**1.039**	**85.255**	**204.272**	**14.584**	**543.764**	**3.713**	**0.265**	**9.886**	**100**

[a] NHANES (National Health and Nutrition Examination Survey) published in Korea Centers for Disease Control and Prevention(KCDC) in 2005 and 2008.

[b] Sum of Indicator PCBs, PCB-28, 52, 101, 118, 138, 153, and 180.

[c] Sum of Dioxin-like PCBs, PCB-77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189.

[d] Sum of 62 total PCBs, PCB-1, 3, 4, 8, 10, 15, 18, 19, 22, 28, 33, 37, 44, 49, 52, 54, 70, 74, 77, 81, 87, 95, 99, 101, 104, 105, 110, 112, 114, 118, 123, 126, 128, 138, 149, 151, 153, 155, 156, 157, 158, 167, 168, 169, 170, 171, 177, 178, 180, 183, 187, 188, 189, 191, 194, 199, 201, 202, 205, 206, 208, and 209.

[e] PCBs level based on daily dietary intake of general population estimated from NHANES

[f] PCBs level per kg body weight for a 55 kg average adult body per day (ng/kg body weight/day) based on NHANES

[g] The ratio was estimated for the total amount of the daily dietary intake of $\sum_{62}$PCBs in each foodstuff.

Table 2 Comparison of total TEQ concentration (dioxin-like PCBs) in seafood with other countries

	Food groups assessed	pg WHO-TEQ/g wet weight	References
Taiwan	Fish	0.49	[14]
Greece	Fish-aqua culture	1.19	[12]
	Fish wild	0.33	
Netherlands	Fish, fatty	3.16[a]	[13]
	Fish, lean	0.59[a]	
China	Fish	0.29	[15]
South Korea	Seafood	0.78	[8]
South Korea	Seafood	0.01[b]	This study

[a] PCDDs, PCDFs and dioxin-like PCBs (PCB-77, 81, 126, and 169)

[b] 2005 WHO-TEF was used for calculation of total TEQ concentration in this study

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