POLYCHLORINATED DIBENZO-*p*-DIOXINS/FURANS AND DIOXIN-LIKE POLYCHLORINATED BIPHENYLS IN FOODSTUFFS FROM KOREA

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Introduction

Recently, it has been reported that polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDDs/DFs) and dioxin-like polychlorinated biphenyls (non-*ortho*- and mono-*ortho*- substituted CBs) have been found in all variety of environmental media, such as air, soil, sediment and animal tissue. These hydrophobic chemicals are highly toxic persistent in the environment. Especially, these chemicals have a strong affinity for sediments and a high potential for accumulating in biological tissue lipid. Due to low excretion and biodegradation in humans, these toxic compounds concentrate in the body fat tissue, and their residue levels reflect external exposure.

The general population is mainly exposed to these toxic compounds through fatty foodstuffs consumption, especially meat, milk, fish and their by-products, leading to a background body burden of these compounds[1,2]. Therefore, in considering the human exposure assessment, it is an important issue to understand the contamination status for these food-groups. However, there is little information on the contamination status of these food-groups in Korea.

In this study, we examined the contamination levels of PCDDs/DFs and dioxin-like PCBs in foodstuffs, especially fish, pork, chicken and beef meat. Also, we quantify the contamination level and characterization of PCDDs/DFs and dioxin-like PCBs in foodstuffs collected from Korea.

Materials and Methods

Sample Collection

Raw foodstuffs were collected from different locations in Masan and Pusan from Korea during July 1994 to March 1995. Table 1 lists the details of food samples used in the present study collected from Korea. Fish samples representing different species, namely; Mackerel (*Scomber australasicus*), Croaker (*Argyrosomus argentatus*), Alaska Pollack (*Theragra chalcogramma*) and Hair tail (*Trichiurus iepturus*) were collected from fish markets in Masan and Pusan. Animal origin food samples such as chicken fat, pork fat and beef meat were obtained from butcher shop in markets, transported to laboratory with dry ice and preserved at –20℃ until analysis.

Determination of PCDDs/DFs and dioxin-like PCBs

Approximately 50 g from fish and meat samples was homogenized, freeze-dried and extracted using a Soxhlet apparatus with dichloromethane. Fractionation was carried out with an activated silica-gel and an alumina column. In the charcoal-impregnated silica-gel mixture column fractionation step, adsorbed PCDDs/DFs and dioxin-like PCBs were eluted into two fractions. The first fraction, eluted with 25% dichloromethane in hexane, consisted of mono-*ortho*-substituted

CBs. The second fraction, eluted with toluene, comprised PCDDs/DFs and non-*ortho*-substituted CBs.

Table 1. Details of market food samples collected from Korea.

Food item		Sample code no.	Daily intake (g/day)	Sampled year	Lipid content(%)
Fish	Mackerel (n=4)	FS1	10.9	1995	20
		FS2		1995	27
		FS3		1994	13
		FS4		1994	21
	Croaker (3)	FS5	10.2	1995	1.8
		FS6 [2]		1995	6
		FS7 [5]		1994	1.4
	Alaska pollack (3)	FS8	6.3	1995	1.1
		FS9		1995	0.7
		FS10		1994	0.6
	Hair tail (4)	FS11	9.3	1995	11
		FS12		1995	7.7
		FS13		1994	19
		FS14		1994	17
Meat	Pork (2)	PF1 [3]	21.6	1995	50
		PF2 [2]		1994	70
	Beef (2)	BM1 [4]	31.4	1995	74
		BM2 [4]		1994	73
	Chicken (2)	CF1 [2]	8.2	1995	61
		CF2 [3]		1994	46

Figures in brackets [ ] indicate the number of samples pooled.

Identification and quantification of PCDD/DF homologues and non-*ortho*- and mono-*ortho*- substituted CBs was performed by HRGC-HRMS (R>10000, 10% valley). The separation of PCDDs/DFs was achieved using a HP 6890 instrument equipped with DB-5 and DB-17 columns with splitless and solvent cut mode. Gas chromatographic separation of non-*ortho* and mono- *ortho*-substituted CBs was carried out on a DB-5 capillary column. The mass spectrometer was operated at an EI energy of 40 eV and the ion current was at 600 μA. PCDD/DF, non-*ortho* and mono-*ortho*-substituted CB congeners were monitored by SIM at the two most intensive ions at the molecular ion cluster.

Result and Discussion

Contamination level and TEQ of PCDDs/DFs and dioxin-like PCBs

The mean concentrations of individual 2,3,7,8-substituted PCDDs/DFs in the food samples collected from Korea are shown in Table 2. The total PCDDs/DFs were detected in all foodstuff samples. The mean concentrations on a wet weight basis of total PCDDs/DFs determined from the various foodstuffs were 0.43 pg/g (n=3) for alaska pollack, 0.85 pg/g (n=2) for chicken fat, 0.9 pg/g (n=2) for pork fat, 1.02 pg/g (n=3) for croaker, 1.69 pg/g (n=2) for beef meat, 1.86 pg/g (n=4) for hair tail and 2.84 pg/g (n=4) for mackerel.

The total dioxin-like PCB concentrations varied from a mean low value of 56 pg/g (on a wet weight basis) in pork fat samples to a mean high value of 175 pg/g in beef. For fish samples, dioxin-like PCB congeners were not analyzed.

On the whole, the contamination levels of total PCDDs/DFs in fish samples were higher than those in meat samples. Furthermore, the contributions of low-chlorinated PCDDs/DFs on total PCDD/DF concentrations in fish samples were relatively higher than those in meat samples. This phenomenon was also reflected in the percent composition of PCDDs /DFs-TEQ. The values of PCDDs/DFs and dioxin-like PCBs TEQ, based on WHO-TEFs for human [3)], for each sample are also shown in Table 2. The total PCDDs/DFs-TEQ concentrations ranged from 0.03 pg TEQ/g for pork fat to 0.89 pg TEQ/g for mackerel. Among the analyzed isomers of PCDDs/DFs in fish samples, percent contribution of TEQ for

Table 2. Mean concentrations (pg/g wet wt. basis) of foodstuffs collected from Korea.

compounds	congeners	WHO-TEF	Meat			Fish			
			beef (n=2)	pork(n=2)	chicken(n=2)	mackerel (n=4)	croaker(n=3)	alaska pollack(n=3)	hairtail(n=4)
PCDDs/DFs	2378-TeCDD	1	0.01	nd	0.00	0.12	0.02	0.04	0.16
	12378-PeCDD	1	0.06	0.01	0.01	0.21	0.10	0.04	0.21
	123478-HxCDD	0.1	0.02	0.01	0.01	0.04	nd	nd	nd
	123678-HxCDD	0.1	0.07	0.02	0.06	0.03	0.09	nd	0.08
	123789-HxCDD	0.1	0.02	nd	0.02	nd	0.15	nd	nd
	1234678-HpCDD	0.01	0.20	0.14	0.21	0.03	0.06	0.03	0.10
	OCDD	0.0001	0.51	0.59	0.32	nd	nd	nd	0.13
	2378-TeCDF	0.1	0.01	0.01	0.02	1.09	0.14	0.09	0.47
	12378-PeCDF	0.05	0.01	nd	0.01	0.32	0.20	0.05	0.20
	23478-PeCDF	0.5	0.10	0.02	0.02	0.82	0.03	nd	0.37
	123478-HxCDF	0.1	0.09	0.02	0.02	nd	0.06	0.04	0.08
	123678-HxCDF	0.1	0.08	nd	nd	0.14	0.03	0.01	nd
	123789-HxCDF	0.1	nd	nd	nd	nd	nd	nd	nd
	234678-HxCDF	0.1	0.09	0.01	0.02	0.02	0.07	0.01	0.01
	1234678-HpCDF	0.01	0.12	0.03	0.03	0.02	0.07	0.11	0.05
	1234789-HpCDF	0.01	0.01	nd	nd	nd	nd	0.01	nd
	OCDF	0.0001	0.28	0.05	0.10	nd	nd	nd	nd
	total PCDDs/DFs		1.69	0.90	0.85	2.84	1.02	0.43	1.86
	total PCDDs/DFs-TEQ		0.16	0.03	0.04	0.89	0.20	0.10	0.63
Non-ortho CBs	3,4,4',5-TeCB (81)	0.0001	0.20	0.01	0.31	na	na	na	na
	3,3',4,4'-TeCB(77)	0.0001	0.49	0.27	5.04	na	na	na	na
	3,3',4,4',5-PeCB(126)	0.1	1.89	0.23	0.44	na	na	na	na
	3,3',4,4',5,5'-HxCB(169)	0.01	0.34	0.26	0.09	na	na	na	na
	total Non-*ortho* CBs		2.92	0.77	5.88	-	-	-	-
Mono-ortho CBs	2',3,4,4',5-PeCB(123)	0.0001	3.20	0.44	4.55	na	na	na	na
	2,3',4,4',5-PeCB(118)	0.0001	120.5	29.6	56.4	na	na	na	na
	2,3,4,4',5-PeCB(114)	0.0005	3.30	0.93	1.64	na	na	na	na
	2,3,3',4,4'-PeCB(105)	0.0001	16.50	5.79	19.10	na	na	na	na
	2,3',4,4',5,5'-HxCB(167)	0.00001	6.18	1.26	2.47	na	na	na	na
	2,3,3',4,4',5-HxCB(156)	0.0005	15.40	13.60	6.71	na	na	na	na
	2,3,3',4,4',5'-HxCB(157)	0.0005	2.97	2.18	1.46	na	na	na	na
	2,3,3',4,4',5,5'-HpCB(189)	0.0001	3.91	1.56	1.12	na	na	na	na
	total Mono-*ortho* CBs		172.0	55.4	93.5	-	-	-	-
	total dioxin-like PCBs-TEQ		0.22	0.04	0.06	-	-	-	-

nd ; not detected, na ; not analyzed

2,3,4,7,8-PeCDF on total PCDDs/DFs was shown to be 42%. This suggests that fish consumption is a significant exposure source for low-chlorinated PCDDs/DFs on the general population in Korea.

Figure 1 shows the total TEQ (ΣPCDDs/DFs-TEQ +Σdioxin-like PCBs-TEQ) composition in meat samples. It was found that dioxin-like PCBs dominate the total TEQ in meat samples (up to 50% of the total TEQ). Many previous investigations have indicated that dioxin-like PCBs represent an important component of the total TEQ in various foodstuffs. Especially, dioxin like-PCBs in animal and fish dominate the total TEQ ingested by humans [4].

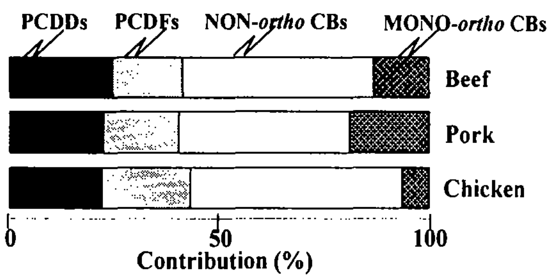

Figure 1. Contribution of individual compounds to the total TEQ in meat samples.

Daily intake of PCDDs/DFs via fish and meat consumption

The daily intake of PCDDs/DFs via fish and meat consumption was calculated to be 42 pg TEQ/person /day, using the 1994 Korean food consumption rate. The values are equivalent to 0.7pg/kg body weight/day for TEQ, by calculation under consideration of 60 kg for body weight.

In overall, the mean daily intake of PCDDs/DFs via fish consumption was about 10 times higher than those via meat consumption. The average daily intake of PCDDs/DFs-TEQ via fish consumption from various countries was published 0.13pg/kg/day for UK, 0.04 pg/kg/day for USA, 0.28 pg/kg/day for Canada and 0.98 pg/kg/day for Japan, respectively [5,6,7,8]. Although the daily dioxin TEQ intake via fish consumption in Korea (0.63 pg/kg/day) was lower than that in Japan, this value was higher than that for the other countries. It is important to note that fish consumption is one of the main sources of PCDDs/DFs exposure in the Korean population.

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