LEVELS OF PCDDs, PCDFs AND CO-PCBs IN FRESH AND COOKED LEAFY VEGETABLES IN JAPAN

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Introduction

The most important route of exposure to PCDD/Fs and Co-PCBs in humans is through food consumption. Recently, a number of surveys have determined the levels of PCDD/Fs and Co-PCBs in food samples from various countries and regions. However, there are only a few reports concerning the levels of PCDD/Fs and Co-PCBs in leafy vegetables in Japan[1]. In this study, we report the latest levels of PCDD/Fs and Co-PCBs in popular leafy vegetables, spinach and komatsuna in Japan. Furthermore, little attention has been given to the effects of cooking on the levels of the PCDD/Fs and Co-PCBs. Only a few studies have centered on the effects of cooking on their levels in meat and fish samples[2, 3, 4]. Therefore, we also report the effects of the processing steps (washing and boiling) on the levels of PCDD/Fs and Co-PCBs in spinach samples.

Materials and methods

Samples. In order to survey the level of PCDD/Fs and Co-PCBs in leafy vegetables in Japan, spinach and komatsuna samples were obtained from seven different districts in Japan in 1998.
The Cooking effects on the levels of PCDD/Fs and Co-PCBs were investigated in two types of spinach samples. One was purchased in 1999 from a supermarket in Fukuoka, Japan, and the other was collected in 1999 from a private vegetable garden in Fukuoka, Japan. The sample from the garden had slightly adhered soil, while the sample from a supermarket had no observed soil adhesion.
Cooking. The spinach samples (150 g) had their roots cut off and then washed with tap water. The samples were then boiled in 1L of tap water for about 2 minutes. All uncooked (only removed roots) and cooked (washed and boiled) samples were chopped in a blender before extraction.
Extraction, cleanup and analysis. The extraction, cleanup and analysis of the PCDD/Fs and Co-PCBs generally followed a previously reported protocol [5]. Briefly, the samples were digested in 1 M-KOH/ethanol solution at room temperature. The alkaline hydrolysates were then extracted twice with *n*-hexane. After treatment with concentrated sulfuric acid, the extracts were purified on an Ag-silica gel column followed by an activated carbon column. On the activated carbon column, the PCDD/Fs and non-*ortho* PCBs were eluted with toluene. The elute was concentrated and dissolved in *n*-nonane. The Analyses of the PCDD/Fs and Co-PCBs were performed by HRGC/HRMS using a HP6890 plus gas chromatograph coupled to a Micromass Autospec mass spectrometer. The detection limits were 0.01 pg/g for TCDD/F and PeCDD/Fs, 0.02 pg/g for HxCDD/Fs and HpCDD/Fs, 0.05 pg/g for OCDD/F and 0.01 pg/g for non-*ortho* PCBs. The TEQ concentrations were calculated using the WHO-TEFs (1997).

Results and Discussion

Table 1 shows the measured concentrations of the PCDD/Fs and Co-PCBs in two kinds of leafy vegetables in Japan, 1998. The PCDDs, PCDFs and Co-PCBs were detected in all the spinach and komatsuna samples. The spinach samples had mean concentrations of the PCDDs, PCDFs and Co-PCBs of 12.48, 1.09 and 2.12 pg/g, respectively. The komatsuna samples had mean concentrations of the PCDDs, PCDFs and Co-PCBs of 3.85, 0.62 and 0.74 pg/g, respectively. Additionally, the mean TEQ concentrations were 0.196 pg TEQ/g for spinach and 0.094 pg TEQ/g for komatsuna. These TEQ value were similar to the values obtained from our previous study[1].

Table 2 shows the mean, minimum and maximum concentrations of the PCDD/Fs and Co-PCBs in uncooked and cooked spinach samples. Since the sample's weight changes during the processing, the concentrations were calculated on the basis of the original weights. The uncooked samples from the garden had higher PCDD/Fs and Co-PCBs levels than those from the supermarket due to the soil adhesion. The washing process reduced the mean total concentrations of the PCDDs, PCDFs and Co-PCBs to 41 %, 77% and 87% of the initial concentrations in the sample from the supermarket, and 35%, 68% and 89% of the initial concentrations in the sample from the garden, respectively. Washing followed by the boiling process indicated that the mean total concentrations of the PCDDs, PCDFs and Co-PCBs were reduced to 21%, 31% and 61% of the initial concentrations in the sample from the supermarket, and 21%, 38% and 60% of the initial concentrations in the garden sample, respectively. Furthermore, the mean total TEQ concentrations were reduced to about 30% of the initial TEQ concentrations by cooking in both samples. Our results suggest that washing followed by boiling effectively reduced the TEQ concentrations and the concentrations of the PCDD/Fs and Co-PCBs in the leafy vegetables. In this study, one of the most popular cooking methods used for spinach in Japan was examined to its effect on the levels of PCDD/Fs and Co-PCBs, however, different cooking processes may lead to different effects on their levels. Therefore, a further investigation of the effects on their levels by various cooking processes should be carried out to accurately estimate the intake of PCDD/Fs and Co-PCBs through food consumption.

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Table 1. Concentration of PCDD/Fs and Co-PCBs in leafy vegetable samples from seven different districts in Japan in 1998

Congeners		Spinach (pg/g)							Komatsuna (pg/g)						
		No. 1	No. 2	No. 3	No. 4	No. 5	No. 6	No. 7	No. 1	No. 2	No. 3	No. 4	No. 5	No. 6	No. 7
PCDDs	2,3,7,8-TCDD	0.02	0.03	0.02	0.04	0.03	0.01	ND	0.01	ND	0.01	0.03	0.01	0.01	ND
	1,2,3,7,8-PeCDD	0.06	0.05	0.05	0.07	0.04	0.06	ND	0.02	ND	0.03	0.04	0.02	ND	0.02
	1,2,3,4,7,8-HxCDD	0.04	ND	0.03	0.03	ND	0.06	ND	ND	ND	ND	ND	ND	ND	ND
	1,2,3,6,7,8-HxCDD	0.06	0.03	0.06	0.05	0.04	0.13	ND	ND	ND	0.03	ND	ND	ND	0.05
	1,2,3,7,8,9-HxCDD	0.05	0.03	0.04	0.04	0.02	0.12	ND	0.02	ND	0.03	ND	ND	ND	0.05
	1,2,3,4,6,7,8-HpCDD	0.49	0.17	0.61	0.28	0.13	4.70	0.08	0.17	0.05	0.22	0.10	0.11	0.20	0.90
	OCDD	4.20	0.94	3.80	2.20	0.54	67.00	0.89	0.93	0.61	1.10	0.79	1.40	3.00	17.00
PCDFs	2,3,7,8-TCDF	0.13	0.13	0.10	0.13	0.14	0.04	N.D.	0.04	0.01	0.04	0.05	0.03	0.02	0.02
	1,2,3,7,8-PeCDF	0.19	0.17	0.14	0.17	0.19	0.07	0.02	0.10	0.02	0.08	0.09	0.07	0.04	0.05
	2,3,4,7,8-PeCDF	0.11	0.08	0.09	0.11	0.10	0.04	ND	0.06	ND	0.06	0.06	0.04	0.02	0.04
	1,2,3,4,7,8-HxCDF	0.10	0.08	0.09	0.08	0.08	0.08	ND	0.06	ND	0.06	0.04	0.03	0.03	0.06
	1,2,3,6,7,8-HxCDF	0.11	0.08	0.11	0.11	0.08	0.05	ND	0.06	ND	0.07	0.05	0.03	ND	0.04
	1,2,3,7,8,9-HxCDF	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	2,3,4,6,7,8-HxCDF	0.11	0.06	0.13	0.09	0.07	0.05	ND	0.07	ND	0.09	0.05	0.02	0.02	0.05
	1,2,3,4,6,7,8-HpCDF	0.30	0.18	0.36	0.21	0.21	0.52	0.04	0.20	0.04	0.27	0.11	0.08	0.12	0.29
	1,2,3,4,7,8,9-HpCDF	0.05	0.02	0.05	0.02	ND	0.07	ND	ND	ND	0.03	ND	ND	ND	0.04
	OCDF	0.18	0.11	0.37	0.10	0.07	1.00	0.08	0.11	ND	0.19	0.08	0.09	0.13	0.80
Co-PCBs	3,3',4,4'-TCB	1.60	1.70	1.30	3.70	3.10	1.00	0.26	0.46	0.24	0.81	0.67	0.38	0.49	0.50
	3,3',4,4',5-PeCB	0.17	0.25	0.15	0.43	0.52	0.19	0.06	0.26	0.08	0.36	0.27	0.13	0.12	0.18
	3,3',4,4',5,5'-HxCB	0.06	0.06	0.06	0.07	0.10	0.03	ND	0.04	ND	0.05	0.05	0.02	0.02	0.03
Total PCDDs (mean)		4.92	1.25	4.61	2.71 (12.48)	0.80	72.08	0.97	1.15	0.66	1.42	0.96 (3.85)	1.54	3.21	18.02
Total PCDFs (mean)		1.28	0.91	1.44	1.02 (1.09)	0.94	1.92	0.14	0.70	0.07	0.89	0.53 (0.62)	0.39	0.38	1.39
Total Co-PCBs (mean)		1.83	2.01	1.51	4.20 (2.12)	3.72	1.22	0.32	0.76	0.32	1.22	0.99 (0.74)	0.53	0.63	0.71
Total TEQ (mean)		0.231	0.199	0.204	0.276 (0.196)	0.229	0.226	0.008	0.120	0.011	0.148	0.153 (0.094)	0.080	0.045	0.102

ND: not detected.

Table 2. Concentration of PCDD/Fs and Co-PCBs in uncooked and cooked spinach samples[a]

Congeners		Sample from a supermarket									Sample from a private vegetable garden								
		Uncooked, pg/g			Cooked, pg/g[b]						Uncooked, pg/g			Cooked, pg/g					
					Washed			Boiled						Washed			Boiled		
		Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max
PCDDs	2,3,7,8-TCDD	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND
	1,2,3,7,8-PeCDD	0.04	0.03	0.04	0.02	ND	0.03	0	ND	ND	0.09	0.08	0.10	0.07	0.05	0.08	0.03	ND	0.05
	1,2,3,4,7,8-HxCDD	0.01	ND	0.03	0.01	ND	0.02	0.01	ND	0.02	0.04	0.03	0.04	0.01	ND	0.02	0	ND	ND
	1,2,3,6,7,8-HxCDD	0.04	0.03	0.05	0.03	0.02	0.05	0.01	ND	0.02	0.11	0.08	0.13	0.05	0.04	0.08	0.02	ND	0.03
	1,2,3,7,8,9-HxCDD	0	ND	ND	0.01	ND	0.04	0	ND	ND	0.04	ND	0.08	0	ND	ND	0	ND	ND
	1,2,3,4,6,7,8-HpCDD	0.46	0.31	0.64	0.22	0.19	0.26	0.11	0.09	0.13	1.00	0.80	1.25	0.46	0.43	0.50	0.25	0.20	0.30
	OCDD	5.44	3.77	6.93	2.20	1.73	2.52	1.12	1.02	1.19	12.56	9.97	15.80	4.27	3.88	4.82	2.67	2.07	3.48
PCDFs	2,3,7,8-TCDF	0.07	0.06	0.08	0.07	0.06	0.08	0.04	0.03	0.05	0.07	0.07	0.08	0.07	0.06	0.08	0.04	0.03	0.06
	1,2,3,7,8-PeCDF	0.07	0.05	0.10	0.06	0.06	0.07	0.04	0.03	0.06	0.07	0.05	0.08	0.05	0.04	0.06	0.04	0.03	0.04
	2,3,4,7,8-PeCDF	0.04	0.03	0.05	0.03	0.03	0.03	0.01	ND	0.03	0.05	0.05	0.06	0.04	0.03	0.04	0.02	0.02	0.03
	1,2,3,4,7,8-HxCDF	0.05	0.04	0.06	0.04	0.03	0.04	0.01	ND	0.02	0.07	0.06	0.08	0.06	0.03	0.07	0.02	ND	0.04
	1,2,3,6,7,8-HxCDF	0.05	0.03	0.06	0.04	0.03	0.05	0.01	ND	0.02	0.05	0.04	0.06	0.05	0.04	0.06	0.02	ND	0.04
	1,2,3,7,8,9-HxCDF	0.01	ND	0.03	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND
	2,3,4,6,7,8-HxCDF	0.04	0.03	0.05	0.03	0.03	0.04	0.01	ND	0.02	0.14	0.12	0.16	0.08	0.07	0.08	0.04	0.03	0.05
	1,2,3,4,6,7,8-HpCDF	0.16	0.11	0.19	0.11	0.09	0.12	0.04	ND	0.06	0.27	0.22	0.32	0.14	0.13	0.15	0.08	0.07	0.10
	1,2,3,4,7,8,9-HpCDF	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND
	OCDF	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND	0	ND	ND
Co-PCBs	3,3',4,4'-TCB	0.73	0.65	0.78	0.63	0.55	0.73	0.46	0.37	0.62	1.77	1.60	1.91	1.57	1.51	1.60	1.06	0.83	1.31
	3,3',4,4',5-PeCB	0.12	0.11	0.13	0.11	0.09	0.15	0.07	0.05	0.09	0.23	0.19	0.25	0.20	0.19	0.22	0.13	0.11	0.16
	3,3',4,4',5,5'-HxCB	0.04	0.03	0.04	0.03	0.02	0.03	0.02	ND	0.03	0.04	0.03	0.05	0.05	0.04	0.05	0.03	0.02	0.04
Total PCDDs		5.98	4.14	7.58	2.48	2.01	2.83	1.23	1.13	1.36	13.84	11	17.3	4.86	4.42	5.47	2.96	2.30	3.86
(mean residue percent)						(41%)			(21%)						(35%)			(21%)	
Total PCDFs		0.48	0.35	0.59	0.37	0.34	0.40	0.15	0.12	0.24	0.71	0.61	0.83	0.48	0.43	0.51	0.26	0.20	0.35
(mean residue percent)						(77%)			(31%)						(68%)			(38%)	
Total Co-PCBs		0.89	0.79	0.94	0.77	0.67	0.91	0.54	0.42	0.73	2.04	1.82	2.21	1.82	1.75	1.88	1.23	0.96	1.51
(mean residue percent)						(87%)			(61%)						(89%)			(60%)	
Total TEQ		0.11	0.08	0.12	0.07	0.05	0.09	0.02	0.01	0.04	0.211	0.19	0.23	0.14	0.12	0.16	0.07	0.04	0.11
(mean residue percent)						(66%)			(21%)						(68%)			(34%)	

[a] Mean values (calculated at ND = 0) are the average of four individual samples.

[b] The concentrations were calculated on the basis of the original weights.