日本産コメ、コムギ、ダイズおよびアズキ中のダイオキシン類濃度とその推定摂取量

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Report

Levels of Dioxins in Rice, Wheat, Soybean, and Adzuki Bean Cultivated in 1999 to 2002 in Japan and Estimation of Their Intake

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A total of 369 samples of rice ($n=311$), wheat ($n=10$), soybean ($n=44$), and adzuki bean ($n=4$) collected from various locations in Japan between 1999 and 2002 were analyzed for PCDDs, PCDFs (PCDD/Fs) and coplanar PCBs. Sampling points within about 1 km of operational municipal waste incinerators that were considered sources of dioxins were defined as "near-source" areas, and all other sampling points were defined as "general" areas. The toxic equivalent quantity (TEQ) values of soybean samples collected from near-source areas were significantly higher ($p<0.05$) than those from general areas. A significant difference of TEQs among sampling years in rice in general areas was also found. However, the differences could not be explained by the presence or absence of incineration plants in the area surrounding the sampling point or by a temporal decrease of air pollution. The TEQs of the crops varied widely, but the median value of each crop was quite low, at 0.000021, 0.00013, 0.0000095, and 0.00016 pg-TEQ/g wet wt. in rice, wheat, soybean and adzuki bean, respectively. On the basis of these survey results, the daily intake of PCDD/Fs and coplanar PCBs from rice, wheat, soybean, and adzuki bean was calculated. The daily intakes from these crops were estimated to be 0.0056 pg-TEQ/kg B.W./day on the assumption that "not detected" (ND) could be taken as zero, ND=0, and 0.18 pg-TEQ/kg B.W./day if ND is put equal to 1/2 LOD (half the limit of detection). In comparison with the tolerable daily intake set in Japan for PCDD/Fs and coplanar PCBs (4 pg-TEQ/kg B.W./day), it was considered that the levels of contamination by PCDD/Fs and coplanar PCBs in these crops cultivated in the environment of Japan do not present a problem.

Key words: polychlorinated dibenzo-*p*-dioxin; polychlorinated dibenzofuran; coplanar polychlorinated biphenyl; rice; wheat; soybean; adzuki bean; daily intake

Introduction

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and coplanar polychlorinated biphenyls (coplanar PCBs) show strong toxicities, including carcinogenicity, teratogenicity, impairment of the immune system and other adverse effects. Processes of combustion and incineration in municipal waste incinerators[1] and Kanechlors[2] were cited as sources of PCDD/Fs and coplanar PCBs. In addition, these chemicals were contained as impurities in organochlorine pesticides such as pentachlorophenol (PCP) and 1,3,5-trichloro-2-(4-nitrophenoxy) benzene (CNP)[3-6], which were previously used as herbicides. Therefore, considerable amounts of PCDD/Fs and coplanar PCBs have contaminated agricultural fields.

The Law Concerning Special Measures against Dioxins (The Dioxins Law) was enacted in 1999, and came into force in 2000. This law prescribes effluent control, the tolerable daily intake (TDI), and the values of environmental standards for PCDD/Fs and coplanar PCBs in the atmosphere, water, sediments and soils. In this context, the Ministry of Agriculture, Forestry and Fisheries and the Ministry of the Environment conducted a national survey to review PCDD/Fs and coplanar PCBs contamination of agricultural soils and crops from 1999 to 2002. In our previous study, levels of PCDD/Fs and coplanar PCBs in fruits were reported[7]. Rice, wheat and soybean are the major foods in Japan. The sum intake of these crops accounts for approximately 40% of total agricultural products intake on a weight basis[8-10]. Accordingly, it is important that we grasp the current status of PCDD/Fs and coplanar PCBs contamination of these grains and bean crops. In this paper, we analyze the results of the survey of PCDD/Fs and coplanar PCBs in rice, wheat, soybean, adzuki bean, and the soils in which these crops were cultivated in order to 1) evaluate the influence of municipal waste incinerators on the contamination of the crops, 2) compare the contamination levels in the crops, and 3) estimate the daily human intake of these contaminants from the crops.

Materials and Methods

Samples

Samples of the edible parts of rice, wheat, soybean,

and adzuki bean were collected from all over Japan between 1999 and 2002. In addition, samples of the soils in which these crops were grown were collected. Sampling points within about 1 km of operational municipal waste incinerators that were considered sources of dioxins were defined as "near-source" areas, and all other sampling points were defined as "general" areas. The distance between the sampling point and the nearest operational incinerator was calculated based on Global Positioning System (GPS) data. The numbers of samples collected from near-source and general areas are shown by sampling year in Table 1.

Sample preparation procedures were described in detail elsewhere[11]. In brief, for crop samples, brown rice without chaff, threshed wheat, and seeds without pods (in the case of soybean and adzuki bean) were milled uniformly. Soil samples sifted through a mesh screen (<2 mm) after air-drying were used for analysis. These samples were preserved in a freezer at −20C° until extraction.

Analysis

Analytical methods (extraction, purification and measurement with HRGC/HRMS) of crops and soils were described in our previous report[7], according to the guideline for PCDD/Fs and coplanar PCBs analysis in foods and soils[12), 13)]. Limits of detection (LODs) for each congener of PCDD/Fs and coplanar PCBs in crops were calculated based on the *Provisional guidelines for analysis of polychlorinated dibenzo-p-dioxins, dibenzofurans and coplanar PCBs in foods*[12] and the limits of quantitation (LOQs) for congeners of PCDD/Fs and coplanar PCBs in soils were calculated based on the *Manual for the Survey and Determination of Dioxins in Soils*[13]. These values are shown in Table 2. Recovery rates of $^{13}C_{12}$-labeled PCDD/Fs and coplanar PCBs added before extraction ranged from 40% to 120% [average in rice: 80 % (range 40–120%), in wheat: 77% (range 54–116%), in soybean: 69% (range 40–114%), in adzuki bean: 83% (range 44–113%), in soil: 84% (range 50–120 %)]. If the values obtained were beyond this range, the sample was reanalyzed by repeating the processes used after extraction. WHO-toxicity equivalent factors (WHO-TEFs)[14] were used to calculate the values of toxic equivalent quantity (TEQs).

Statistical analysis

Statistical analysis of differences in levels was conducted by nonparametric testing, because the measurement values obtained did not show a normal distribution in each near-source or general area group by sampling year. The Mann–Whitney *U* test and Kruskal Wallis test were performed by using SPSS 12.0J for Windows® (SPSS Inc., US), and Spearman's correlation coefficient by rank test was performed by using Statcel 2 (OMS Publishing, Tokorozawa).

Results and Discussion

Comparison of PCDD/Fs and coplanar PCBs concentrations in crops collected from near-source and general area

Concentrations (TEQs) of PCDD/Fs and coplanar PCBs in rice, wheat, soybean, and adzuki bean are shown by sampling year and point (near-source or gen-

Table 2. Limits of Detection (LODs) in Crop Samples and Limits of Quantitation (LOQs) in Soil Samples Collected between 1999 and 2002

	Crops LOD (pg/g wet wt.)	Soils LOQ (pg/g dry wt.)
PCDD/Fs		
TeCDDs	0.01	1
PeCDDs	0.01	1
HxCDDs	0.02	2
HpCDDs	0.02	2
OCDD	0.05	5
TeCDFs	0.01	1
PeCDFs	0.01	1
HxCDFs	0.02	2
HpCDFs	0.02	2
OCDF	0.05	5
Coplanar PCBs		
Non-*ortho*-PCBs		
3,3′,4,4′-TeCB (#77)	0.1	2
3,4,4′,5-TeCB (#81)	0.1	2
3,3′,4,4′,5-PeCB (#126)	0.1	2
3,3′,4,4′,5,5′-HxCB (#169)	0.1	2
Mono-*ortho*-PCBs		
2,3,3′,4,4′-PeCB (#105)	1	2
2,3,4,4′,5-PeCB (#114)	1	2
2,3′,4,4′,5-PeCB (#118)	1	2
2′,3,4,4′,5-PeCB (#123)	1	2
2,3,3′,4,4′,5-HxCB (#156)	1	2
2,3,3′,4,4′,5′-HxCB (#157)	1	2
2,3′,4,4′,5,5′-HxCB (#167)	1	2
2,3,3′,4,4′,5,5′-HpCB (#189)	1	2

Table 1. Numbers of Crop Samples Collected between 1999 and 2002

Fiscal year	Rice			Wheat			Soybean			Adzuki bean		
	Source[a]	General[b]	Total	Source	General	Total	Source	General	Total	Source	General	Total
1999	28	18	46		2	2	6	8	14			
2000	57	77	134	4	2	6	4	4	8	2	2	4
2001	36	30	66				4	6	10			
2002	39	26	65		2	2	5	7	12			
Total	160	151	311	4	6	10	19	25	44	2	2	4

[a] Sampling points located within about 1 km of operational municipal waste incinerators
[b] Sampling points more than about 1 km from operational municipal waste incinerators

Table 3. TEQ Levels of PCDD/Fs and Coplanar PCBs (pg-TEQ/g wet wt.) in Rice, Wheat, Soybean, and Adzuki Bean Collected between 1999 and 2002

Crops	Fiscal year	Source[a]						General[b]						Mann–Whitney *U* test[d]
		Mean	SD	Median	Minimum	Maximum	Kruskal Wallis test[c]	Mean	SD	Median	Minimum	Maximum	Kruskal Wallis test	
Rice							ns[e]						*[f]	
	1999	0.0020	0.0061	0.000057	0.000006	0.027		0.00089	0.0018	0.00038	0.000006	0.0079		ns
	2000	0.00096	0.0023	0.000037	0	0.010		0.00033	0.00059	0.000020	0	0.0030		ns
	2001	0.00024	0.00042	0.000021	0	0.0017		0.00046	0.0015	0.000009	0	0.0079		ns
	2002	0.00019	0.00031	0.000022	0	0.0014		0.0032	0.015	0.000010	0	0.077		ns
Wheat							nt[g]						nt	
	1999							0.00013	0.00014	0.00013	0.000037	0.00023		nt
	2000	0.00059	0.00060	0.00046	0.000037	0.0014		0.000010	0	0.000010	0.000010	0.000010		nt
	2002							0.00013	0.000021	0.00013	0.00011	0.00014		nt
Soybean							ns						ns	
	1999	0.00017	0.00016	0.00013	0.000035	0.00044		0.0075	0.021	0.000021	0	0.060		*
	2000	0.0026	0.0034	0.0015	0.000005	0.0072		0	0	0	0	0		*
	2001	0.000034	0.000058	0.000009	0	0.00012		0.000004	0.000011	0	0	0.000026		ns
	2002	0.00024	0.00027	0.00016	0.000006	0.00070		0.000036	0.000057	0.000008	0	0.00014		*
Adzuki bean							nt						nt	
	2000	0.00016	0.00021	0.00016	0.000016	0.00031		0.00017	0.00022	0.00017	0.000015	0.00032		nt

[a] Sampling points located within about 1 km of operational municipal waste incinerators
[b] Sampling points more than about 1 km from operational municipal waste incinerators
[c] TEQ values differences among 4 sampling years were tested with the Kruskal Wallis test at a level of significance of 0.05.
[d] TEQ values in each year were compared between "Source" and "General" with the Mann–Whitney *U* test at a level of significance of 0.05.
[e] ns: not significant
[f] *: significant difference
[g] nt: not tested

eral) in Table 3. These values were calculated by taking the concentration of the PCDD/F and coplanar PCB congener as equal to zero when the congener concentration was below its LOD (Table 2). When TEQs in rice and soybean were compared between near-source and general areas with the Mann–Whitney U test at a level of significance of 0.05, no significant differences were observed between sampling areas for rice. However, for soybean, the TEQs for samples collected from near-source areas were significantly higher than those in samples collected from general areas in 1999, 2000, and 2002.

Next, comparisons of the TEQs among sampling years were performed in rice and soybean, which had been collected in all four years. Temporal changes in the TEQs in each crop in near-source and general areas are shown separately (Table 3). When the differences among 4 sampling years were tested with the Kruskal Wallis test at a level of significance of 0.05, there were no significant differences among years in the TEQs in soybean in near-source or general areas, or in rice in near-source areas. A significant difference among years was observed only for rice in general areas. Comparison between near-source and general areas, and among the 4 sampling years were not performed or were impossible for wheat and adzuki bean, because the sample numbers of these crops were too small for statistical analysis.

Sakai *et al.*[15] reported the congener composition of PCDD/Fs and coplanar PCBs in the gases emitted from a typical municipal waste incinerator in Japan. Contributions of PCDD/Fs and coplanar PCBs congeners in these gases to the TEQ values were calculated using the data. The contributions of six congeners were as follows: 1,2,3,7,8-PeCDD (16.5%), 2,3,4,7,8-PeCDF (33.8%), 1,2,3,4,7,8-HxCDF (7.3%), 1,2,3,6,7,8-HxCDF (7.3%), 2,3,4,6,7,8-HxCDF (9.1%), and 3,3′,4,4′,5-PeCB (#126) (6.2%). These six congeners greatly contributed to the TEQ, accounting for 80% of the total PCDD/Fs and coplanar PCBs (0.065 ng-TEQ/Nm^3) in the gases emitted. With regard to the composition of PCDD/Fs and coplanar PCBs in the atmosphere in Japan, the contributions of the congeners to the TEQ values were calculated using data from the national survey ($n=100$) conducted in 1998[16]. The results showed a trend very similar to that for gases from municipal waste incinerators. The contributions of the six congeners that contributed highly to the TEQ were as follows: 1,2,3,7,8-PeCDD (10.8%), 2,3,4,7,8-PeCDF (31.3%), 1,2,3,4,7,8-HxCDF (7.7%), 1,2,3,6,7,8-HxCDF (6.5%), 2,3,4,6,7,8-HxCDF (10.9%), and 3,3′,4,4′,5-PeCB (#126) (8.2%). The six congeners accounted for 75% of the mean concentration of PCDD/Fs and coplanar PCBs (0.23 pg-TEQ/m^3) in the atmosphere. Therefore, it was expected that the six congeners contained mainly in the atmosphere in Japan or in the gases emitted from municipal waste incinerators would reflect differences in the total concentrations (TEQs) of PCDD/Fs and coplanar PCBs between near-source and general areas in the case of soybean or among sampling years in the case of rice in general areas.

However, the six dominant congeners [1,2,3,7,8-PeCDD, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 2,3,4,6,7,8-HxCDF, and 3,3′,4,4′,5-PeCB (#126)] in the atmosphere and the gases emitted from municipal waste incinerators were detected in only two soybean samples in 2001 (one sample each collected from a near-source and a general area) and in one sample in each year among rice samples collected in 1999–2002 from general areas. On the other hand, in most crop samples (321 of total 369 samples), only four congeners—1,2,3,4,6,7,8-HpCDD, OCDD, 1,2,3,4,6,7,8-HpCDF, and 3,3′,4,4′-TeCB (#77)—were detected, and the other congeners were below their LODs. The contribution of these four congeners to the TEQ in emission gases was only 5.0%, and their contribution to the atmosphere in Japan was only 8.4%. Accordingly, the congeners that contributed mainly to the TEQs in the crops were clearly different from the dominant congeners in the atmosphere and in the gases emitted from municipal waste incinerators. Therefore, it is difficult to judge the influence of incineration plants or temporal trends of air pollution on PCDD/Fs and coplanar PCBs concentrations in rice and soybean samples, though significant differences were observed by statistical analysis.

Comparison of PCDD/Fs and coplanar PCBs concentrations in crops

The TEQs in each crop were compared by compiling all the data into crop groups, because the differences were not clear by sampling area and year. The TEQs in each crop were shown by means of a boxplot (Fig. 1). As

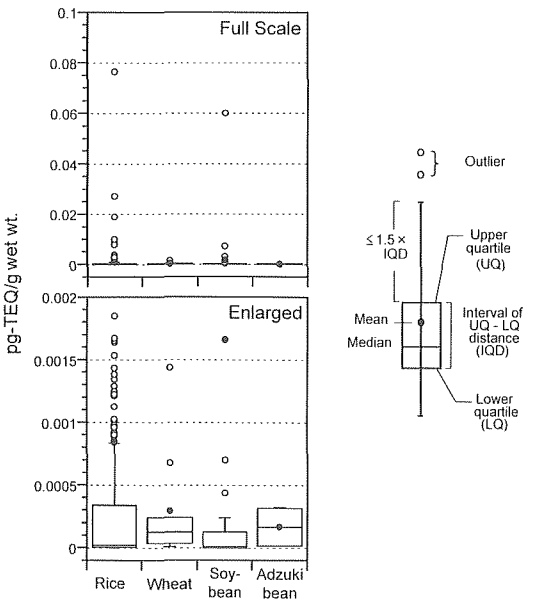

Fig. 1. TEQs in crops collected from 1999 to 2002 in Japan

In the box plots, the line extending up from the UQ (or down from the LQ) was stopped when there was no point in the range.

shown in this graph, the TEQs in each crop varied widely, containing a substantial number of upper "outliers" [points equivalent to >UQ+1.5×IQD, where UQ =upper quartile and IQD=the difference between upper quartile and lower quartile (interval of UQ–LQ distance)], except in adzuki bean. In particular, one outlier each in rice and soybean was obviously higher than the other values; thus, the mean TEQs in rice and soybean became four to eight times higher than those in wheat and adzuki bean. To determine the sources of contamination of these samples, the contributions of each congener to the TEQ in each crop and soil were calculated (Fig. 2). Among rice samples with outliers, in a sample collected in general areas in 2002 (No. 02G56) that showed a notably higher TEQ value of 0.077 pg-TEQ/g wet wt., the congener composition was almost identical with that of the soil. On the other hand, the congener compositions of the other rice samples and soybean samples were completely different from the composition of the soil. Accordingly, we can assume that sample No. 02G56 of rice, whose congener composition was identical to that of the soil, was contaminated with PCDD/Fs and coplanar PCBs owing to intrusion of soil in the sampling or preparation process. In fact, the TEQ in the soil at the point of sampling of No. 02G56 was 2000 times higher (170 pg-TEQ/g dry wt.) than that in the rice. Thus, the TEQ of the rice would be greatly influenced even if only a very slight amount of soil was present as a contaminant. The relationship between the TEQs in the crops and the corresponding soils (n=221) was examined in groups for which we had both crop and soil samples (Fig. 3). Based on Spearman's correlation coefficient, rank correlation coefficients were 0.21 in rice (n=183), 0.89 in wheat (n=6), 0.40 in soybean (n=30), and 0.25 in all crops (n=221). In these cases, rank correlations were detected at a level of significance of 0.05. However, the TEQs in most crop samples were below 0.001 pg-TEQ/g wet wt. when the TEQs of the soils ranged from 0 to 300 pg-TEQ/g dry wt. Therefore, the TEQs in the crops might not, for practical purposes, have been influenced by those in the soils. In this study, only one (rice) sample among all the crop samples clearly showed the influence of the soil, and this soil contamination was considered to be an exceptional case. Among the soybean samples with "outliers", one sample collected in general areas in 1999 (No. 99G8) had particularly high (0.060 pg-TEQ/g wet wt.) TEQ value. In this soybean sample, only 2,3,7,8 -TeCDD (Other PCDD/Fs in Figure 3) was detected at the same level as its LOD among congeners with WHO-TEF values. The congener 2,3,7,8-TeCDD was not detected at all in other soybean samples, including those with "outliers", other than No. 99G8. In addition, it was impossible to explain its source in terms of the composition of PCDD/Fs and coplanar PCBs in the soil. Further consideration was impossible, because air samples were not collected.

It is difficult to compare the mean values among crops because the TEQs in rice and soybean did not

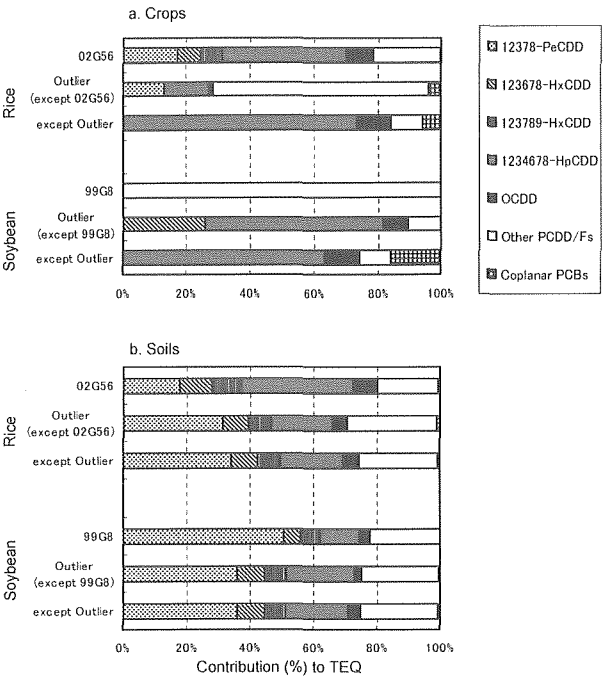

Fig. 2. TEQ composition in crops and cultivated soils

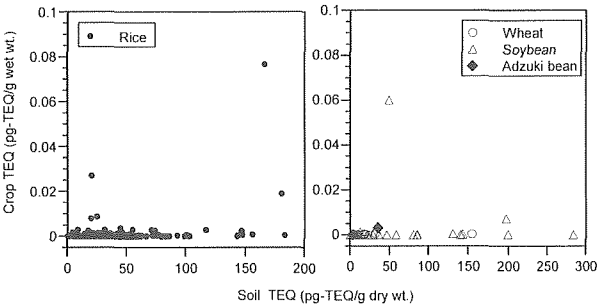

Fig. 3. Relationship between TEQs in crops and the cultivated soils

show a normal distribution in addition to having marked "outliers". Accordingly, when the TEQs in each crop were compared using the median values, the level of each value was extremely low, at 0.000021, 0.00013, 0.0000095, and 0.00016 pg-TEQ/g wet wt. in rice, wheat, soybean, and adzuki bean, respectively. When a rank comparison for the TEQs was performed with the Kruskal Wallis test at a level of significance of 0.05, it was determined that there were significant differences among the crops. However, considering the levels of the LODs, it was considered that there were no meaningful differences in the TEQs among the four crops. In our previous report, mean TEQ values of PCDD/Fs and coplanar PCBs in various fruit samples collected between 1999 and 2002 ranged from 0 to 0.033 pg-TEQ/g wet wt.[7] The TEQ levels of rice, wheat, soybean and adzuki bean were nearly the same or lower than that of fruits. The reason for the extremely low levels of TEQs in each crop was considered to be the fact that rice, wheat, soybean, and adzuki bean—the targets of this study—are not susceptible to air pollution because the analyzed parts of these crops were covered by their

chaff, bran, or pod.

Daily intakes of PCDD/Fs and coplanar PCBs from crops

Daily intakes of PCDD/Fs and coplanar PCBs from rice, wheat, soybean, and adzuki bean were estimated (Table 4). For all the crop samples collected from near-source and general areas between 1999 and 2002, TEQs were calculated by assuming that the congener concentration was equal to either zero (ND=0) or half the LOD (1/2 LOD) when each congener concentration was below the LOD. These TEQs were multiplied by the daily intakes of individual crops[8)-10)], and the values were divided by an average body weight of 50 kg. In the case of adzuki bean, the daily intake value including cow pea, kidney bean, and lentil pea was used. The sums of daily intakes of PCDD/Fs and coplanar PCBs at ND=0 from these grains and beans were 0.00039 pg-TEQ/kg B.W./day using the median values and 0.0057 pg-TEQ/kg B.W./day using the mean values; at ND=1/2 LOD they were 0.18 pg-TEQ/kg B.W./day using both the median and mean vvalues. In our previous report, the daily intake of PCDD/Fs and coplanar PCBs from fruits (apple, chestnut, grape, Japanese apricot, mandarin orange, peach, pear, persimmon and ponkan orange) in the same government survey was estimated to be 0.0082 pg-TEQ/kg B.W./day (ND=0) or 0.072 pg-TEQ/kg B.W./day (ND=1/2 LOD)[7)]. The Japanese daily intake of rice, wheat, soybean and adzuki bean (359.4 g/day) is three times greater than that of these fruits (121.9 g/day)[8)-10)]. However, the mean value of the daily intake of PCDD/Fs and coplanar PCBs from the former crops was lower (ND=0), or only 2.5 times much (ND=1/2 LOD) than that from the latter. Tsutsumi et al.[17)] calculated daily intakes of PCDD/Fs and coplanar PCBs from 14 food groups in 1999 and 2000 by the Total Diet Study (TDS), and reported that they were 2.25 pg-TEQ/kg B.W./day (ND=0) and 3.22 pg-TEQ/kg B.W./day (ND=1/2 LOD). The daily intakes of PCDD/Fs and coplanar PCBs contained in rice, wheat, and beans, which were calculated using the concentrations of PCDD/Fs and coplanar PCBs obtained from the results in this study, was only 5.6% of 3.22 pg-TEQ/kg B.W./day (ND=1/2 LOD) based on the TDS, or 4.5% of 4 pg-TEQ/kg B.W./day, which is the tolerable daily intake of PCDD/Fs and coplanar PCBs set in Japan, even if the mean value at ND=1/2 LOD was used. Therefore, PCDD/Fs and coplanar PCBs of rice, wheat, soybean and adzuki bean cultivated in the environment of Japan seem not to be present at problematic levels, under the usual conditions used for food, in the light of the consumed amounts of crop products.

Table 4. Estimated Daily Intakes of PCDD/Fs and Coplanar PCBs (pg-TEQ/kg B.W./day) from Rice, Wheat, Soybean, and Adzuki Bean in Japan

	Rice	Wheat	Soybean	Adzuki bean	Sum[a)]
Intake of crop (g/day)[b)]	185.1[c)]	116.8	56.1[c)]	1.4[d)]	
	ND=0[e)]				
Mean	0.0031	0.00069	0.0019	0.000005	0.0057
	(0.00085)[f)]	(0.00029)	(0.0017)	(0.00017)	
SD	0.018	0.0010	0.010	0.000005	0.021
	(0.0049)	(0.00045)	(0.0091)	(0.00017)	
Median	0.000078	0.00030	0.000011	0.000005	0.00039
	(0.000021)	(0.00013)	(0.0000095)	(0.00016)	
Minimum	0	0.000023	0	0.0000004	0.000023
	(0)	(0.000010)	(0)	(0.000010)	
Maximum	0.28	0.0034	0.067	0.000009	0.35
	(0.077)	(0.0014)	(0.060)	(0.00032)	
	ND=1/2LOD[g)]				
Mean	0.10	0.064	0.032	0.00076	0.18
	(0.028)	(0.027)	(0.029)	(0.027)	
SD	0.014	0.00060	0.0093	0.000003	0.016
	(0.0038)	(0.00026)	(0.0083)	(0.00012)	
Median	0.10	0.063	0.030	0.00076	0.18
	(0.027)	(0.027)	(0.027)	(0.027)	
Minimum	0.10	0.063	0.030	0.00076	0.18
	(0.027)	(0.027)	(0.027)	(0.027)	
Maximum	0.34	0.065	0.092	0.00076	0.45
	(0.091)	(0.028)	(0.082)	(0.027)	

a) The sum of the four crops
b) Intake of crops was obtained from National Surveys in 1998, 1999 and 2000.
c) Including the products
d) Including kidney bean, cowpea and lentil
e) Each congener concentration which was below the LOD was assumed to be equal to zero.
f) TEQ values (pg-TEQ/g wet wt.) used for the calculation of the daily intakes were shown in parentheses.
g) Each congener concentration which was below the LOD was assumed to be equal to half of the LOD.

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References

1) Hiraoka, M., Okajima, S., Source control technologies in MSW incineration plants. Organohalogen Compounds, **19**, 275–291 (1994).
2) Wakimoto, T., Kannan, N., Ono, M., Tatsukawa, R., Masuda,Y., Isomer-specific determination of polychlorinated dibenzofurans in Japanese and American polychlorinated biphenyls. Chemosphere, **17**, 743–750 (1988).
3) Yamagishi, T., Miyazaki, T., Akiyama, K., Morita, M., Nakagawa, J., Hori, S., Kaneko, S., Polychlorinated dibenzo-*p*-dioxins and dibenzofurans in commercial diphenyl ester herbicides, and in freshwater fish collected from the application area. Chemosphere, **10**, 1,137–1,144 (1981).
4) Hagenmaier, H., Brunner, H., Isomer specific analysis of pentachlorophenol and sodium pentachlorophenate for 2,3,7,8-substituted PCDD and PCDF at sub-ppb levels. Chemosphere, **16**, 1,759–1,764 (1987).
5) Masunaga, S., Takasuga, T., Nakanishi, J., Dioxin and dioxin-like PCB impurities in some Japanese agrochemical formulations. Chemosphere, **44**, 873–885 (2001).
6) Seike, N., Otani, T., Ueji, M., Takasuga, T., Tsuzuki, T., Temporal change of polychlorinated dibenzo-*p*-dioxins, dibenzofurans and dioxin like polychlorinated biphenyls source in paddy soils. Kankyou Kagaku (J. Environ. Chem.), **13**, 117–131 (2003).
7) Seike, N., Miwa, T., Otani, T., Ueji, M., Levels of PCDDs, PCDFs and dioxin-like PCBs in fruits in Japan. Shokuhin Eiseigaku Zasshi (J. Food Hyg. Soc. Japan), **46**, 256–262 (2005).
8) Kenkou Eiyou Johou Kenkyukai ed. "Kokumin Eiyou No Joukyou, Heisei 10 Nen Kouseiroudou Shou Kokumin Eiyou Chousa Kekka (National Nutrition Survey in 1998, Japan)", Tokyo, Daiichi Shuppan, 2000. (ISBN 4-8041-0920-X)
9) Kenkou Eiyou Johou Kenkyukai ed. "Kokumin Eiyou No Genjou, Heisei 11 Nen Kouseiroudou Shou Kokumin Eiyou Chousa Kekka (National Nutrition Survey in 1999, Japan)", Tokyo, Daiichi Shuppan, 2001. (ISBN 4-8041-0961-7)
10) Kenkou Eiyou Johou Kenkyukai ed. "Kokumin Eiyou No Genjou, Heisei 12 Nen Kouseiroudou Shou Kokumin Eiyou Chousa Kekka (National Nutrition Survey in 2000, Japan)", Tokyo, Daiichi Shuppan, 2002. (ISBN 4-8041-0994-3)
11) Nouyaku No Sakumotsu Zanryu Shiken Zissi Youkou (Execution Points for Pesticides Residue in Crops) (Apr., 1973), Ministry of Agriculture, Forestry and Fisheries of Japan.
12) Shokuhinchu No Daiokishinrui Oyobi Kopurana PCB No Sokuteihouhou Zantei Gaidorain (Provisional Guidelines for Analysis of Polychlorinated Dibenzo-*p*-dioxins, Dibenzofurans and Coplanar PCBs in Foods) (Sept., 1999), Environmental Health Bureau, Ministry of Health and Welfare of Japan.
13) Daiokishinrui Ni Kakaru Dojo Chousa Sokutei Manual (Manual for investigation and determination of polychlorinated dibenzo-*p*-dioxins, dibenzofurans and coplanar PCBs in soils) (Jan., 2000), Water Quality Conservation Bureau, Ministry of Environment of Japan.
14) Van den Berg, M., Birnbaum, L., Bosveld, A.T.C., Brunstrom, B., Cook, P., Feeley, M., Giesy, J.P., Hanberg, A., Hasegawa, R., Kennedy, S.W., Kubiak, T., Larsen, J.C., Rolaf van Leeuwen, F.X., Djien Liem, A.K., Nolt, C., Peterson, R.E., Poellinger, L., Safe, S., Schrenk, D., Tillitt, D., Tysklind, M., Younes, M., Wærn, F., Zacharewski, T., Toxic equivalency factors (TEFs) for PCBs, PCDDs, and PCDFs for humans and wildlife. Environ. Health Perspect., **106**, 775–792 (1998).
15) Sakai, S., Hayakawa, K., Takatsuki, H., Kawakami, I., Dioxin-like PCBs released from waste incineration and their deposition flux. Environ. Sci. Technol., **35**, 3,601–3,607 (2001).
16) Regarding the Results of the Urgent Simultaneous Nationwide Survey of Dioxins (Conducted in 1998) (Sept., 1999), Environmental Agency of Japan.
17) Tsutsumi, T., Yanagi, T., Nakamura, M., Kono, Y., Uchibe, H., Iida, T., Hori, T., Nakagawa, R., Tobiishi, K., Matsuda, R., Sasaki, K., Toyoda, M., Update of daily intake of PCDDs, PCDFs, and dioxin-like PCBs from food in Japan. Chemosphere, **45**, 1,129–1,137 (2001).