Dioxin Concentrations in Commercial Health Tea Materials in Japan

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This study determined the concentrations of polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyls (dioxin-like PCBs) in five selected plant materials [dokudami (from houttuynia herb), rose hip (from rosa fruit), ebisugusa (from cassia seed), rooibos, and tochu (from eucommia leaf)] used as health teas in Japan. The toxic equivalent (TEQ) levels for dioxins in the samples ranged from < 0.001 to 0.27 pg-TEQ/g weight, when undetectable and trace amounts were taken as zero. The mean of total TEQ level in commercial tea materials was estimated as 0.08 pg-TEQ/g (n = 5). The total TEQ in these samples was mainly dominated by the levels of PCDD/Fs (representing *ca.* 80% of the total TEQ).

Key words —— dioxin, polychlorinated dibenzo-*p*-dioxin, polychlorinated dibenzofuran, dioxin-like polychlorinated biphenyl, health tea, food

INTRODUCTION

In Japan, foods have been generally recognized as the main route of human intake of polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyls (PCBs), which are referred to collectively as dioxins, contributing to more than 95% of the daily intake of these compounds.[1)] Thus, it is very important to survey the levels of dioxins in various kinds of foodstuffs in order to evaluate the risk to humans. For food safety and security, we have been regularly performing daily intake surveys of dioxins by total diet studys and dioxin-pollution surveys of individual foods in Japan. From previous results, fishery products have been regarded as the main source of dioxins in the Japanese diet.[2,3)] In addition, the levels of dioxins in retail fish and shellfish including dioxin contamination in the edible parts of Japanese common squid and saury were elucidated.[4,5)]

In recent years, the demand for functional foods, including health teas, has expanded significantly.[6)] Functional foods tend to be concentrates of selected foods; therefore, they may have a risk of excessive intake of harmful chemical contaminants in the foods even if those chemicals in individual food are at such a small level that they have no influence on health routinely. Previously, the levels of dioxin contaminants in fish oil supplements on the Japanese market were reported.[7)] As a part of these ongoing studies on the actual situation of dioxin contamination in functional foods in Japan, it is reported herein on the dioxin levels in health teas, which have not been described previously with the exception of studies on green tea leaf.[8,9)]

MATERIALS AND METHODS

Samples and Reagents —— Five tea materials [sample 1: dokudami (from houttuynia herb; aerial part of *Houttuynia cordata*), sample 2: ebisugusa (from cassia seed; seed of *Cassia obtusifolia* or *Cassia tora*), sample 3: rooibos (from the leaves of *Aspalatus linearis*), sample 4: rose hip (from the fruit of *Rosa* spp.) and sample 5: tochu (from the leaves of *Eucommia ulmoides*)] were obtained from stores in Japan (in 2006). Reagents used in this study were the same as described in the previous paper.[10)]

Extraction and Cleanup Procedure —— The method of extraction and cleanup followed that of the tentative guideline for the analysis of dioxins in foods in Japan.[11)] Briefly, 50 g of homogenized tea material was spiked with a mixture of ^{13}C-labeled internal quantitative standards [seventeen PCDD/Fs and twelve dioxin-like PCBs that have toxic equivalency factor (TEF) values proposed by the World Health Organization (WHO)], then extracted twice

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by shaking with acetone-*n*-hexane (1 : 1, total 200–360 ml) for 1 hr at room temperature, and filtered (0.8 μm filter, GPF filter paper of Kiriyama, Tokyo, Japan). After extraction, sulfuric acid treatment and column chromatography (CC) procedures (silver nitrate-silica gel CC, alumina CC, and activated carbon mixed silica gel CC) were carried out for cleanup, and each fraction obtained was spiked with ^{13}C-labeled recovery standards before analysis by high-resolution gas chromatography (HRGC)/high-resolution mass spectrometry (HRMS). The details were described in the previous paper.[10)]

Dioxin Analysis —— Dioxin analysis was performed under HRGC/HRMS conditions using an Agilent 6890 plus gas chromatograph (Agilent Technologies, Santa Clara, CA, U.S.A.) coupled to a Micromass Autospec mass spectorometer (Micromass, Manchester, U.K.). The approximate limits of quantification in this study were; 0.01 pg/g for Tetra (Te)CDDs, Penta (Pe)CDDs, Tetra (Te)CDFs, and Penta (Pe)CDFs; 0.02 pg/g for Hexa (Hx)CDDs, Hepta (Hp)CDDs, Hexa (Hx)CDFs, and Hepta (Hp)CDFs; 0.05 pg/g for Octa (O)CDD and Octa (O)CDF; 0.1 pg/g for non-*ortho* PCBs; and 1 pg/g for mono-*ortho* PCBs. The toxic equivalent (TEQ) values for congeners were calculated using the WHO-TEFs.[12)] The total TEQ in a sample was calculated based on the assumption that all isomer concentrations lower than the limits of quantification were equal to zero.

RESULTS AND DISCUSSION

Table 1 shows the concentrations of the sums for PCDDs, PCDFs, and dioxin-like PCBs, as well as the total TEQ, for five plant materials used as health teas. The recoveries calculated for the spiked compounds were always in the range defined as sufficient recovery in the tentative guideline.[11)] On the whole, the samples had low dioxin levels around the limit of quantification or below. The highest total concentration in pg/g product weight was 37.15 pg/g in sample 5 (tochu). Subsequently samples 1 (dokudami) and 4 (rose hip) were found to have contamination levels of 17.46 and 11.42 pg/g, respectively. The dioxin concentrations of samples 2 (ebisugusa) and 3 (rooibos) were low, 2.46 and 0.66 pg/g, respectively. The mean of dioxin concentrations of commercial tea materials was 13.83 pg/g (n = 5, 3.18 pg/g for PCDDs, 0.79 pg/g for PCDFs, and 9.86 pg/g for dioxin-like PCBs, respectively). The maximum TEQ values corresponded to sample 5 (0.27 pg-TEQ/g) followed by sample 1 (0.13 pg-TEQ/g), and the mean of total TEQ level in commercial tea materials was 0.08 pg-TEQ/g (n = 5).

Figure 1 graphically illustrates the TEQ con-

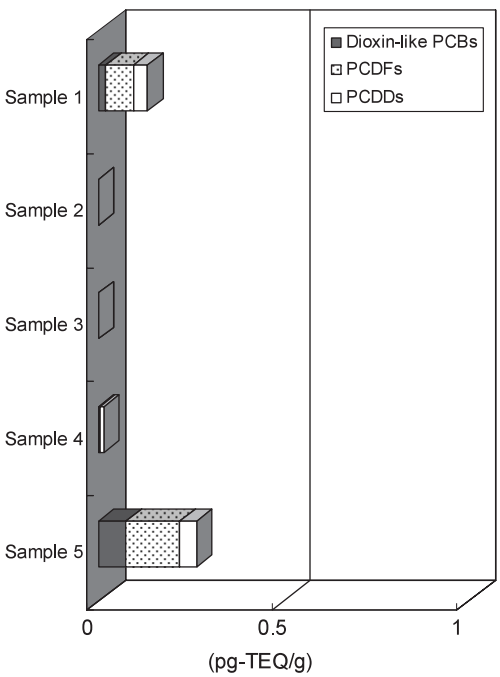

Fig. 1. TEQ Concentrations in Five Commercial Health Tea Materials
Name of samples: see Materials and Methods.

Table 1. Concentrations of PCDD/Fs and Dioxin-like PCBs in Commercial Health Tea Materials in Japan

Tea Materials	Concentration (pg/g prepared weight)				Total TEQ concentration (pg-TEQ/g prepared weight)
	PCDDs	PCDFs	Dioxin-like PCBs	Total	
Sample 1	3.35	1.01	13.1	17.46	0.13
Sample 2	0.16	nd	2.3	2.46	< 0.001
Sample 3	0.06	nd	0.6	0.66	< 0.001
Sample 4	8.50	0.42	2.5	11.42	0.014
Sample 5	3.83	2.52	30.8	37.15	0.2
Average (n = 5)	3.18	0.79	9.86	13.83	0.08

nd: not detected. TEQ was calculated as WHO-TEFs. Values below the limit of detection were takes as zero.

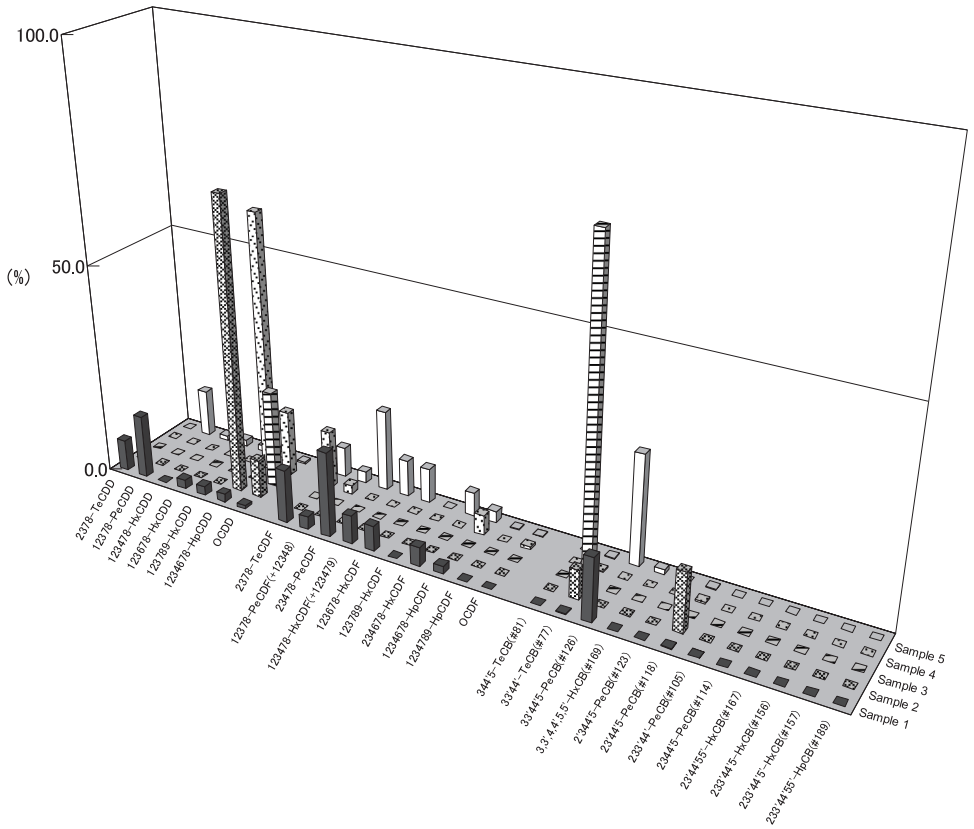

Fig. 2. Percentage Contributions of Congeners Using WHO-TEFs to Total Contamination in Five Commercial Health Tea Materials
Name of samples: see Materials and Methods.

centrations and their components in each health-tea sample. As can be seen, PCDD/Fs were the predominant congeners detected in the samples.

Figure 2 shows the percentage contribution of isomers to the total TEQ levels of tea materials. Total TEQ levels were dominated by 1,2,3,7,8-PeCDD, 2,3,7,8-TeCDF, 2,3,4,7,8-PeCDF, and 3,3′,4,4′,5-PeCB (#126) in samples 1 and 5; 1,2,3,4,6,7,8-HpCDD, OCDD and 2,3,7,8-TeCDF in sample 4; 1,2,3,4,6,7,8-HpCDD and 2,3′,4,4′,5-PeCB (#118) in sample 2; and OCDD and 3,3′,4,4′-TeCB (#77) in sample 3. It was reported previously that the dioxin concentrations in vegetables reflect in the contamination source.[13] For example, dioxin analysis of leafy vegetables indicated that 1,2,3,7,8-PeCDD, 2,3,4,7,8-PeCDF, and 3,3′,4,4′-5-PeCB (#126), which are indicator isomers of total TEQ in environmental samples (exhaust gas, ash, and ambient air), are the dominant isomers. In the present study, the dominant isomers contaminating samples 1 and 5 were similar to those found in leafy vegetables. Thus, it is suggested that the dioxin contamination in the teas was caused by absorption or adhesion from the atmosphere to the surface of the plants.

The previous report indicated that the dioxin levels of green tea leaf were also in the range of 0.053–0.856 pg-TEQ/g.[8,9] The present TEQ levels for dioxins in health tea materials were < 0.001–0.27 pg-TEQ/g with the roughly same values as those of green tea. Generally, the plant materials for health teas are decocted in boiling water for drinking. As dioxins are insoluble in water, they are considered not to be ingested from health teas.

In conclusion, the levels of dioxins in health tea materials were surveyed for the first time in Japan. The samples analyzed had low dioxin levels similar to those of leafy vegetables.

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