

Levels of PCDDs/PCDFs and DL-PCBs in Foodstuffs Imported to Korea in 2004-2006.

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Abstract

In this study, the total daily exposure(TDE) and the average Korean(body weight=55kg) intakes(AKI) of dioxin-like compounds(excluding dioxin-like PCBs) were determined. TDE in food was estimated as 6.940 pgTEQ/day in 2005 and 3.902 pgTEQ/day in 2006, respectively. In 2005, AKI from food was calculated to be 0.126 pgTEQ/kg body weight/day, 3.15% of the WHO TDI (4 pgTEQ/kg bw/day). In 2006, AKI were calculated to be 0.071 pgTEQ/kg body weight/day, 1.77% of the WHO TDI (4 pgTEQ/kg bw/day). In conclusion, the levels of PCDDs/PCDFs and DL-PCBs currently detected in foodstuffs imported to Korea are at a safe level, provided there is not a high consumption of oily fish such as mackerel.

Introduction

Food could be the major route of human intake of toxic dioxin-like compounds, which include WHO Polychlorinated Dibenzo-p-Dioxins(PCDDs) and Polychlorinated Dibenzo Furans(PCDFs). Also the inhalation route contributes only a negligible extent, because these chemicals have strong tendencies to bioaccumulate in lipid-rich compartments of organisms.^{1,2} The contamination levels of dioxin-like compounds depend on food types. Foods with a higher fat content may have higher contamination levels so that detectable levels are found in fatty food such as meat, dairy products and fishes. Therefore, it is important to measure the levels and to do a risk assessment for dioxin-like compounds in food (especially fatty food). The aim of this study was to measure the levels of PCDDs/PCDFs and DL-PCBs of imported foodstuffs in Korea and then to assess the health risks potentially associated with the dietary intake of PCDDs/PCDFs and DL-PCBs.

Materials and Methods

Sample: Imported and domestic samples were randomly collected in local markets, discount stores, and department stores in Busan, Korea. The samples included rice, meat (beef, pork, chicken), fish (hair tail, mackerel, crab, shrimp), green tea, salt, and milk products (cheese).

Sample preparation: All food samples were homogenized by mixer and samples were kept at -20°C before analysis. The aliquots of each transferred to a pre-extracted cellulose thimble(43x123mm). It was mixed with 40g of anhydrous sodium sulfate. The samples were then fortified with the ¹³C-labeled standards and extracted with Soxhlet extractor using a mixture of hexane/methylene chloride(1:1) for 24 hours. The extracts were cleaned up using sulfuric acid impregnated silica gel and purified on a series of silica gel, alumina and carbon column prior to analysis by HRGC/HRMS.

In the case of salt, 20g of salt was dissolved in water and adjusted to pH 2.0. The samples were then fortified with the ¹³C-labeled standards and extracted with methylene chloride. After extraction, The next step for the salt was same as the other food samples. For each run, samples were prepared including a method blank and a QC sample.

Instrumental conditions: The analysis was performed by HRGC/HRMS using a HP6890 plus gas chromatography coupled to a Micromass Autospec mass spectrometer. The determination of PCDDs/PCDFs and DL-PCBs was performed in DB-5MS capillary column(60m, 0.25mm id, 0.25µm film thickness). The oven temperature for PCDDs/PCDFs analysis was programmed as follows: 160°C for 4min then to 220°C at 5°C /min, hold for 15min then to 290°C at 5°C /min, hold for 10min then to 300°C at 5°C /min and held for 7min. In the case of DL-PCBs, the oven temperature program was set 150°C for 1min then to 185°C at 20°C /min, hold for 3min then to 245°C at 2°C /min, hold for 3min then to 300°C at 6°C /min and held for 4min. The carrier gas was Helium and flow rate was at 1.0 ml/min. Injector and transfer line temperature were 290°C. HRMS analysis was performed with Micro-Mass AutoSpec-Ultima in SIR mode operating positive EI ionization at a resolving power of >10,000 at m/z 331 of PFK. The MS source was set at 35eV, the trap current was 400 µA and The ion source temperature was 260 °C^{3,4}. Under these conditions, all 17 congeners of PCDDs/PCDFs and all 12 congeners of

DL-PCBs were separated.

Results and Discussion

The levels of contamination were determined as the TEQ values by multiplying with the corresponding WHO-TEFs for each congener.⁵ The results for each food type are shown in table 1, 2, and 3. The levels of PCDDs/PCDFs (pg WHO-TEQ/g ww) in the food were <0.001 for rice, 0.024~0.043 for beef, 0.017~0.048 for domestic beef, 0.004~0.012 for pork, 0.011~0.019 for domestic pork, 0.002 for chicken, 0.006~0.057 for hair tail, 0.032~0.140 for mackerel, 0.010~0.529 for crab, 0.003~0.031 for shrimp, 0.027~0.213 for green tea, 0.009~0.112 for domestic green tea, <0.001 for salt, and 0.034~0.072 for cheese, respectively.

The levels of DL-PCBs (pg WHO-TEQ/g ww) in the food were <0.001 for rice, 0.003~0.014 for beef, 0.085~0.086 for domestic beef, 0.001~0.016 for pork, 0.012~0.015 for domestic pork, 0.009 for chicken, 0.066~0.155 for hair tail, 0.365 for mackerel, 0.002~0.086 for crab, 0.002~0.003 for shrimp, 0.041~0.080 for green tea, 0.011~0.026 for domestic green tea, <0.001 for salt, and 0.016~0.062 for cheese, respectively.

The highest level of PCDDs/PCDFS and DL-PCBs was detected in the mackerel (0.505 pgTEQ/g ww in 2005, 0.216 pgTEQ/g ww in 2006), whereas the lowest level was detected in the rice sample (<0.001 pgTEQ/g) as expected, since the rice was dehusked (thus removing surface contaminants due to environmental pollution).

TDE and AKI of dioxin-like compounds (excluding dioxin-like PCBs) were determined. TDE in food was estimated as 6.940 pgTEQ/day in 2005 and 3.902 pgTEQ/day in 2006, respectively. In 2005, AKI were calculated to be 0.126 pgTEQ/kg body weight/day, 3.15% of the WHO TDI (4 pgTEQ/kg bw/day). In 2006, AKI were calculated to be 0.071 pgTEQ/kg body weight/day, 1.77% of the WHO TDI (4 pgTEQ/kg bw/day). In conclusion, the levels of PCDDs/PCDFS and DL-PCBs currently detected in imported foodstuffs imported to Korea are at a safe level, provided there is not a high consumption of oily fish such as mackerel.

Table 1. Contaminated PCDDs/PCDFS level in different foodstuffs imported to Korea in 2004-2006

Source of exposure	PCDDs/PCDFS (pgTEQ/g)		
	2004	2005	2006
Hair tail	0.057	0.023	0.006
Mackerel	0.099	0.140	0.032
Mackerel(Domestic)	-	-	0.032
Crab	0.118	0.529	0.010
Chicken	0.002	0.002	-
Chicken(Domestic)	-	-	0.003
Cheese	0.034	0.072	0.046
Pork	0.008	0.004	0.012
Pork(Domestic)	0.011	0.040	0.019
Beef	0.043	0.024	0.027
Beef(Domestic)	-	0.017	0.048
Shrimp	0.031	0.003	0.007
Rice	<0.001	<0.001	<0.001
Green tea	0.213	0.050	0.027
Green tea(Domestic)	0.009	0.112	0.015
Salt	<0.001	<0.001	<0.001

Table 2. Contaminated DL-PCBs level in different foodstuffs imported to Korea in 2005-2006

Source of exposure	DL-PCBs (pgTEQ/g)	
	2005	2006
Hair tail	0.155	0.066
Mackerel	0.365	0.365
Mackerel(Domestic)	-	0.003
Crab	0.086	0.002
Chicken	0.009	-
Chicken(Domestic)	-	0.002
Cheese	0.062	0.016
Pork	0.016	0.001
Pork(Domestic)	0.012	0.015
Beef	0.014	0.003
Beef(Domestic)	0.085	0.086
Shrimp	0.003	0.002
Rice	<0.001	<0.001
Green tea	0.080	0.041
Green tea(Domestic)	0.011	0.026
Salt	<0.001	<0.001

Table 3. Estimated daily Exposure to PCDDs/PCDFS and DL-PCBs in 2005-2006

Source of exposure	Contaminated level (pgTEQ/g)		Food intake (g/day)	Estimated daily Exposure (pgTEQ/day)	
	2005	2006		2005	2006
Source of exposure			Food intake (g/day)		
Hair tail	0.178	0.072	2.2	0.392	0.158
Mackerel	0.505	0.216*	5.8	2.929	1.253*
Crab	0.615	0.012	2.1	1.292	0.025
Chicken	0.011	0.005	15.2	0.167	0.076
Cheese	0.134	0.062	0.6	0.080	0.037
Pork	0.036*	0.039*	25.9	0.932*	1.010*
Beef	0.071*	0.083*	16.1	1.143*	1.336*
Shrimp	0.006	0.009	0.8	0.005	0.007
Rice	<0.001	<0.001	205.7	-	-
Green tea	0.127*	0.055*	0.0	-	-
Salt	<0.001	<0.001	3.2	-	-
Sum			277.6	6.940	3.902

*: average of imported and domestic

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