Levels of Dioxins, Dibenzofurans, DDE and PCB Congeners in Pooled Food Samples Collected at Supermarkets across the United States

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Abstract. Food, particularly milk, meat and fish, has been identified as the primary immediate source of polychlorinated dibenzo-p-dioxin (PCDDs) and polychlorinated dibenzofuran (PCDFs) and PCBs in the general population. Estimated intake of these PCDD/Fs has been previously reported from limited U.S. food data. The present study extends our previous work by increasing the sample size and geographical base and by analyzing for selected PCB, as well as PCDD and PCDF congeners. Food samples were obtained from five cities across the U.S.; Binghamton, NY, Atlanta, GA, Chicago, IL, San Diego CA, and Louisville, KY. This study consisted of 10 PCDD/F and PCB congener specific analyses of pooled food samples.

Keywords. Food, dioxins, dibenzofurans, polychlorinated biphenyls, PCDD, PCDF, PCB, DDE

Methods.

A. Collection Food was collected and frozen from supermarkets in Binghamton, New York, Chicago, Illinois, Louisville, Kentucky, Atlanta, Georgia and San Diego, California. They were kept separate and frozen until they reached the dioxin laboratory, where they were thawed, weighed and mixed by weight into pooled samples for analysis. All geographical regions were included in each analysis.

B. Chemistry Samples were prepared by grinding samples of the same types (eg., beef, chicken) together using an auger-fed Hobart® grinder. Each sample composite was ground and homogenized three times. Equal amounts of each type of individual food were taken for a composite sample. One hundred grams of each sample type was taken and mixed with approximately 300-g of pre-cleaned sodium sulfate, then the sample was split and placed into two soxhlet extractors. The sample was fortified with ^{13}C-labeled analogs consistent with Method 8290 and extracted for 16 hours with 1:1 dichloromethane:hexane. After extraction the solvent was removed and the percent extractable lipid was gravimetrically determined. The lipid was re-dissolved in 200 mL of hexane and slurried for 2 hours in 100-g of acidified

silica gel. The extract was then eluted through an acidified silica gel column, a neutral alumina column, and an AX-21/Celite 545 carbon column. The final toluene extract was reduced in volume to 10 μL with ^{13}C-labeled recovery standards added. Ten percent of the extract was taken after the acid silica gel step and analyzed for mono- and di-ortho PCBs.

The extract was analyzed on a VG70-250S at a resolution of 10,000. A 60-m DB-5 GC-MS column was used to provide chromatographic resolution between 2,3,7,8-TCDD and all other TCDD isomers. Method 8290 analysis criteria were met. The ions monitored were changed to accommodate the monitoring of coplanar PCBs in the same analytical run. Results were calculated on a wet weight and on a extractable lipid weight.

Results. Table 1 presents a summary of our previous food dioxin intake study, which found 0.3 to 3.0 pg TEQ kg/day of intake from PCDD/Fs in an average US adult. The estimates of intake for nursing infants were found to be far higher, from 35-53 pg TEQ kg/day[1].

Table 2 presents levels of DDE and PCBs 118, 114, 153, 105, 138, 128, 156 and 180 for pooled samples of beef, chicken, ocean fish, fresh water fish and pork, half of the samples planned for analysis in this study. Results are presented on a lipid basis.

Table 1. Calculated PCDD and PCDF Intake from Various Food Groups for the U.S. Adult General Population

Food Group	Consumption Rate (g/day)*	Range of PCDD/F TEq in Food (wet weight; pg/g)		Daily Human Intake			
				Range Total TEq (pg)		Range TEq/kg B/W (pg)**	
		LOW	HIGH	LOW	HIGH	LOW	HIGH
Beef	88	0.04	1.50	3.52	132.00	0.054	2.031
Pork	28	0.03	0.30	0.84	8.40	0.013	0.129
Poultry	31	0.03	0.03	0.93	0.93	0.014	0.014
Fish	18	0.02	0 13	0.36	2.34	0.006	0.036
Milk	254	0.04	0.04	10.16	10.16	0.156	0.156
Other Dairy Products	55	0.04	0.70	2.20	38.50	0.034	0.592
Fruits & Vegetables	283	---	—	—	—	—	—
		Total Range		18.0	192.3	0.3	3.0

* Consumption rates from Yang and Nelson(ref 2)
** Assuming a 65 kg adult weight

TABLE 2. FOOD SAMPLE RESULTS MONO- AND DI-ORTHO PCB CONGENERS AND DDE (EXTRACTABLE LIPID BASIS)

Sample Description	% Extractable Lipid	DDE Final conc. (ng/g)	118 Final conc. (ng/g)	114 Final conc. (ng/g)	153 Final conc. (ng/g)	105 Final conc. (ng/g)	138 Final conc. (ng/g)	128 Final conc. (ng/g)	156 Final conc. (ng/g)	180 Final conc. (ng/g)
Method Blk	-	-	-	-	-	-	-	-	-	-
Beef	13.1	3.30	0.717	-	0.629	-	-	-	-	-
Chicken	5.33	2.81	3.70	-	2.08	1.47	0.747	-	-	4.32
Ocean Fish	1.43	68.6	22.3	-	27.4	8.34	30.2	-	-	11.8
Fresh Fish	4.83	206	36.3	-	39.3	12.4	37.4	5.59	7.27	12.6
Pork	9.18	2.25	-	-	0.785	-	1.07	-	-	-

Table 3 presents PCDD/F levels for the same pooled food samples on a lipid basis for the 2,3,7,8- substituted congeners as well as total homologue levels.

TABLE 3. POLYCHLORINATED DIOXIN, DIBENZOFURAN LEVELS IN FOOD (pg/g, lipid basis)

Congener	Method Blank b1	BEEF (N=9)	CHICKEN (N=7)	OCEAN FISH (N=13)	FRESH FISH (N=10)	PORK (N=7)
2378TCDF	U(.0323 EMPC)(a)	0.488	U(1.88 EMPC)	U(11.6 EMPC)	14.4	1.97
2378TCDD	U(.038)(b)	U(.19)	U(.467)	2.3	3.09	U(.349 EMPC)
12378PECDF	U(.125)	U(.95)	U(2.34)	U(8.74)	U(7.59 EMPC)	U(1.36)
23478PECDF	U(.125)	U(.95)	2.6	U(8.74)	7.56	U(1.36)
12378PECDD	U(.125)	U(.95)	U(2.34)	U(8.74)	5.2	U(1.36)
123478HXCDF	U(.125)	U(1.22 EMPC)	U(2.34)	U(10.8)	U(3.36 EMPC)	U(1.47 EMPC)
123678HXCDF	U(.125)	U(2.15 EMPC)	U(2.84 EMPC)	U(16.6 EMPC)	U(19.9 EMPC)	U(6.46 EMPC)
234678HXCDF	U(.125)	U(.95)	U(2.34)	U(9.51)	U(2.58)	U(1.36)
123789HXCDF	U(.125)	U(.95)	U(2.91)	U(14.5)	U(2.58)	U(1.36)
123478HXCDD	U(.125)	U(1.39 EMPC)	U(2.34)	U(8.74)	3.01	U(1.36)
123678HXCDD	U(.125)	4.92	U(2.34)	U(8.74)	5.31	1.81
123789HXCDD	U(.125)	1.04	U(2.34)	U(8.74)	4.11	U(1.36)
1234678HPCDF	U(.125)	U(10.8 EMPC)	U(5.49 EMPC)	U(48.7 EMPC)	U(31 EMPC)	U(20.2 EMPC)
1234789HPCDF	U(.125)	U(.95)	U(2.34)	U(8.74)	U(2.72)	U(1.36)
1234678HPCDD	U(.125)	20.9	8.09	11.7	23.5	17.1
12346789OCDF	U(.25)	U(3 EMPC)	U(4.67)	U(17.5)	U(5.17)	U(3.26 EMPC)
12346789OCDD	U(.375 EMPC)	32.7	20.2	31.6	122	87.1
TCDF	U(.025)	0.949	42.2	17.8	40.4	4.7
TCDD	U(.038)	U(.19)	U(.467)	4.06	4.44	U(.272)
PECDF	U(.125)	U(.95)	30.8	8.75	21.7	U(1.36)
PECDD	U(.125)	U(.95)	U(2.34)	U(8.74)	5.2	U(1.36)
HXCDF	U(.125)	1.13	U(2.34)	U(11)	U(2.58)	U(1.36)
HXCDD	U(.125)	6.23	2.39	U(8.74)	12.5	1.93
HPCDF	U(.125)	U(.95)	U(2.34)	U(8.74)	U(2.58)	U(1.36)
HPCDD	U(.125)	20.9	8.09	11.7	23.5	17.1
% Lipid	-	13.13	5.33	1.43	4.83	9.18
I-TE-min(c)	0.00	0.89	0.10	2.45	12.51	0.64
I-TE-max(d)	0.27	2.86	5.17	21.14	16.07	3.97

(a)-Undetected due to an interference. An estimated maximum possible concentration (EMPC) is given as the detection limit.
(b)-Undetected, with the curve based detection limit in pg/g.
(c)-Calculated using a value of zero for undetected isomers.
(d)-Calculated using the detection limit value for undetected isomers.

Discussion. This paper confirms and extends our previous US food studies which were based on PCDD/F analyses of individual food samples collected at one New York supermarket.

- The findings again suggest that the general US food supply is contaminated with similar levels of dioxins and dibenzofurans as is the case for a number of industrial countries including Canada, England, Germany and the Netherlands[3].

- For the first time, measurement of the levels of certain PCB congeners in US food is reported. More PCB congeners will be analyzed and presented. Also, larger number

of pooled samples will be reported.

- Insufficient US food data exits for temporal trends to be evident. There is some indication that human tissue levels may be decreasing over the past decade for PCBs and possibly also for some dioxin congeners. Further work will be needed to establish the existence of a decrease in PCDD/F body burden in the United States over the past decade.

- Comparisons with food dioxin levels from other countries may also contribute to understanding fate and transport as well as levels in food worldwide at the present time.

- Direct congener specific measurement of dioxins, dibenzofurans and PCB congeners will help provide direct evidence of human intake of dioxins and help decrease reliance on indirect modelling which does not take into account measured food data. Such collection, analytic determinations and estimates of intake based on consumption patterns can be established for a relatively modest cost.

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