POLYCHLORINATED DIBENZO-*p*-DIOXINS (PCDD) AND POLYCHLORINATED DIBENZOFURANS (PCDF) IN FOOD SAMPLES COLLECTED IN SOUTHERN MISSISSIPPI (USA)

K.R. Cooper[1], H. Fiedler[2], S. Bergek[3], R. Andersson[3], M. Hjelt[3], and C. Rappe[3]

[1]Rutgers University, E.O.H.S.I., P.O. Box 231, Piscataway, NJ 08855, USA
[2]University of Bayreuth, Chair of Ecological Chemistry and Geochemistry, D-95440 Bayreuth, Germany
[3]Umeå University, Institute of Environmental Chemistry, S-901 87 Umeå, Sweden

ABSTRACT

Each of the 38 food samples collected in southern Mississippi contained detectable levels of PCDD/PCDF; 2,3,7,8-TCDD was quantified in 31 of these samples. The levels (I-TEQ, lipid adjusted) in the meat (0.53-1.1 pg/g) and dairy products (0.42-0.97 pg/g) are slightly lower than those reported from other industrialized countries. The fish (1.19-28 pg/g) and shellfish (5.23-40.1 pg/g) have substantially higher levels compared to the meat and dairy products. A unique finding was that the farm raised catfish (10.2-27.8 pg/g) contained both 2,3,7,8-substituted and non-2,3,7,8-substituted congeners. This is unusual because vertebrate animals usually selectively eliminate and/or metabolize the non-2,3,7,8-substituted congeners. High PCDD/PCDF concentrations were found in the American oyster (21-32 pg/g) and the portions of the crustacea which contained the digestive gland (7.1-40.1 pg/g).

1. INTRODUCTION

Polychlorinated dibenzo-*p*-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF) have been detected in almost every matrix: soil, sediments, air, water, animals, and food (primarily meat, dairy products, fish and shellfish) (1-3). This widespread presence is due to the numerous sources that produce these compounds, as well as the compounds' fat solubility, long range transport and resistance to biotransformation (1). Only a few reported studies have examined PCDD/PCDF levels in food in the United States, limiting the ability to establish background exposure levels (4,5).

The food items in this study were selected based on their suspected high levels of PCDD/PCDF to the dietary intake (6-9); thus, food items such as vegetables, fruit, grain and cereal products were not sampled (3). This paper reports the levels of PCDD/PCDF found in locally consumed dairy products, meat, eggs, and seafood from southern Mississippi, USA.

2. MATERIALS AND METHODS

A total of 38 food items were purchased (Table 1) in April and May 1994 from local stores and seafood markets in George, Greene and Jackson Counties in southern Mississippi, USA.

Analytical Procedures: The samples (25-100 g) were carefully mixed with sodium sulfate (5-20 times by weight) and extracted, cleaned, and fractionated according to the method originally described by Smith et al. (1). Fat removal, however, was performed using a membrane technique described by Bergqvist et al. (11). Final separation and quantification of PCDD/PCDF were effected using a high resolution gas chromatograph equipped with either a 60 m DB-5 column (J&W, Folsom, CA) or a 60 m Restek Rt-2330 (non-bonded SP-2330/Silar 9CP) (Restek Corporation, Bellefonte, PA, USA) which was interfaced to a VG 70-250S double focusing mass spectrometer operating at a resolution of 8000-10000. In this study, we followed the most stringent QA/QC protocols, e.g., EPA Methods 1613 and 8290 and Meier et al. (12).

Table 1. Description of Food Items Collected from Southern Mississippi, USA

Food Item	Description	Number Analyzed
Whole Chicken	2 Whole Chickens (Meat with Skin)	3
Chicken Liver	1 lb. Container of Livers	3
Ground Beef	1 lb.	3
Sausage	Smoked Sausage (Mixed Pork and Beef)	3
Milk	½ Gallon Whole Milk (Plastic Container)	3
Eggs	1 Dozen Large Eggs	3
Cheese	1 lb. Cheddar	3
Butter	1 lb. Whole Butter	3
Fish	Farm Raised Catfish Nuggets[A]	3
	Gulf of Mexico Fish:	
	Mullet Fillet	2
	Spanish Mackerel Fillet	1
Oyster	Raw 1 lb. Shucked	3
Crawfish[B]	Head and Digestive Gland	2
	Tail Muscle	2
Blue crab[B]	Body (Soft-Shell)	3
	Claw Meat	3

[A]Nuggets are small pieces of fillet.
[B]These were composite samples and the tail muscle and claw meat were analyzed separately from the head and digestive gland of the crawfish and the body of the blue crab.

3. RESULTS

The levels for 2,3,7,8-TCDD and I-TEQ on both a wet weight basis and a lipid basis for the 38 food samples are reported in Table 2. In cases of non-detectable concentrations (ND), ½ the detection limit was used in

Figure 1. Mass Fragmentograms of 2,3,4,7,8-Pentachlorodibenzofurans in Spanish mackerel, farm raised catfish, blue crab body and American oyster.

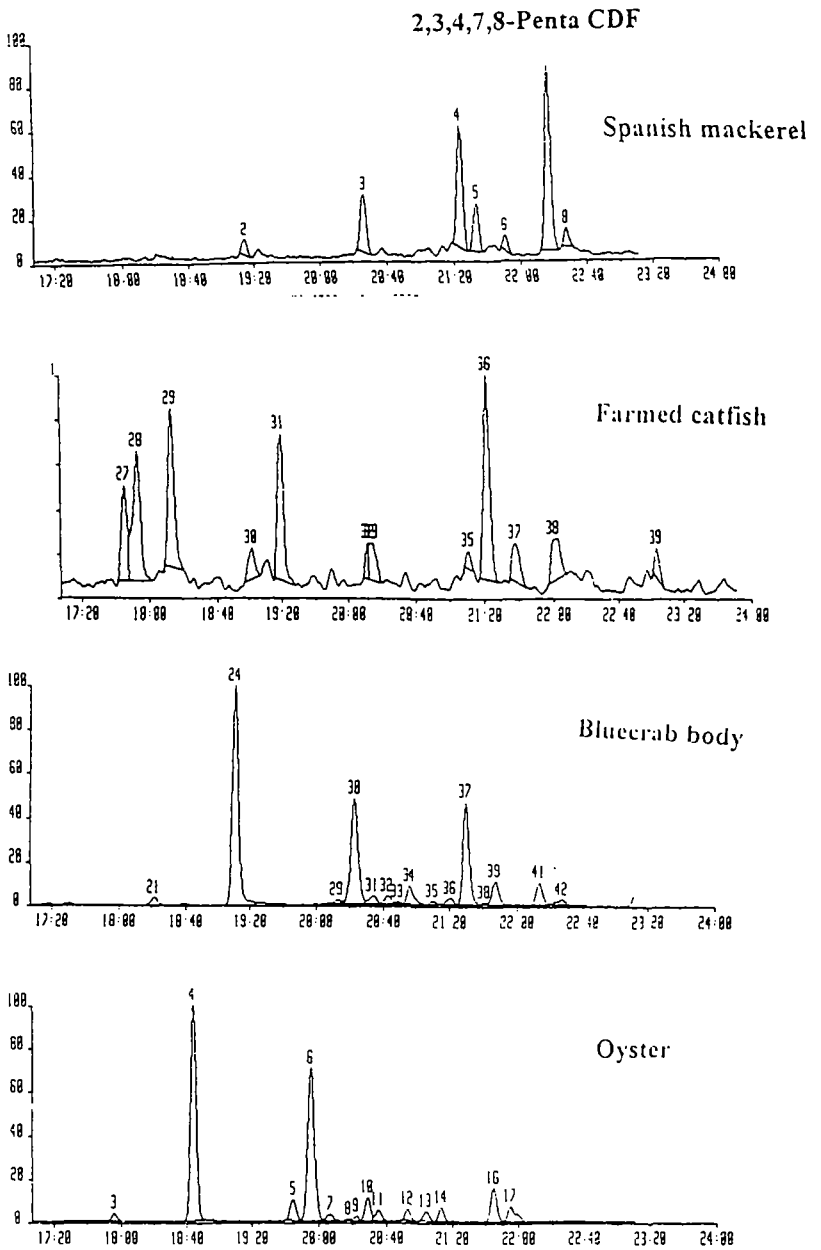

Table 2. Levels of 2,3,7,8-TCDD and I-TEQs in Food From Southern Mississippi[A]

Sample	N	2,3,7,8-TCDD pg/g sample	2,3,7,8-TCDD pg/g lipid	I-TEQ pg/g sample	I-TEQ pg/g lipid	I-TEQ Mean ± SD pg/g lipid
Catfish	3	0.30, 0.83, 0.69	2.5, 8.8, 6.3	1.19, 2.64, 2.57	10.2, 27.8, 23.5	20.5 ± 9.2
Mullet	2	0.051, ND(0.0092)	10, ND(0.41)	0.089, 0.027	17.9, 1.19	
Spanish Mackerel	1	0.18	7.15	0.72	28	
American Oyster	3	0.24, 0.19, 0.20	12, 7.2, 7.2	0.62, 0.53, 0.60	32, 21, 21	24.6 ± 6.4
Blue crab						
Claw	3	0.01, 0.02, 0.02	4.2, 3.2, 4.9	0.06, 0.10, 0.09	19.7, 17.0, 18.4	18.4 ± 1.4
Body	3	0.22, 0.18, 0.15	6.4, 5.8, 4.3	1.09, 1.14, 1.44	31.4, 35.9, 40.1	35.8 ± 4.4
Crawfish						
Tail	2	ND(0.014, 0.016)	ND(2.1, 2.0)	0.033, 0.087	5.23, 10.5	
Head	2	0.34, 0.31	1.3, 1.4	2.34, 1.55	8.8, 7.1	
Butter	3	0.069, 0.041, 0.042	0.088, 0.051, 0.053	0.683, 0.770, 0.552	0.87, 0.97, 0.70	0.85 ± 0.14
Milk	3	0.002, 0.002, 0.002	0.071, 0.060, 0.063	0.025,0.026,0.012	0.81, 0.81, 0.42	0.68 ± 0.23
Cheddar Cheese	3	0.023, 0.017, 0.018	0.067, 0.052, 0.051	0.300, 0.247, 0.254	0.86, 0.76, 0.74	0.79 ± 0.06
Eggs	3	0.008, ND(0.007, 0.005)	0.069, ND(0.067, 0.050)	0.038, 0.020, 0.019	0.20, 0.33, 0.18	0.24 ± 0.08
Ground Beef	3	0.015, 0.096, 0.011	0.048, 0.041, 0.037	0.196, 0.254, 0.152	0.63, 1.10, 0.53	0.75 ± 0.30
Chicken	3	0.008, 0.018, 0.016	0.15, 0.15, 0.19	0.043, 0.085, 0.053	0.78, 0.71, 0.61	0.70 ± 0.09
Chicken Liver	3	0.008, 0.018,0.024	0.18, 0.25, 0.45	0.031, 0.064, 0.070	0.75, 0.71, 0.61	0.69 ± 0.07
Sausage	3	ND(0.069, 0.024, 0.044)	ND(0.22, 0.088, 0.14)	0.178, 0.221, 0.282	0.56, 0.80, 0.87	0.74 ± 0.16

[A]ND stands for non detected and the values in the parentheses are the detection limits. ½ the detection limit was used in calculating the I-TEQ.

calculating the I-TEQ. Calculations using the full detection limits for ND resulted in insignificant and negligible differences in I-TEQ concentrations.

The I-TEQ pg/g lipid for the seafood was consistantly higher than both the dairy and meat products. The highest levels of 2,3,7,8-TCDD (wet weight) were found in the farm raised catfish (range 0.30-0.83 pg/g), crawfish head (0.31-0.34 pg/g) and American oyster (0.19-0.24 pg/g). The highest levels of 2,3,7,8-TCDD (lipid adjusted) were found in the American oyster (7.2-12 pg/g lipid), farm raised catfish (2.5-8.8 pg/g lipid) and blue crab body (4.3-6.4 pg/g lipid). The highest I-TEQ (wet weight) concentrations were found in the farm raised catfish (1.19-2.64 pg/g), crawfish head (1.55-2.34 pg/g) and blue crab body (1.09-1.44 pg/g). The highest I-TEQ (lipid adjusted) concentrations were found in the blue crab body (31.4-40.1 pg/g lipid), Spanish mackerel (28 pg/g lipid) and farm raised catfish (10.2-27.8 pg/g lipid). The mean and standard deviation (X $\pm$ SD) for the I-TEQ pg/g lipid are very similar for the dairy and meat products.

The farm raised catfish and the invertebrates contained 2,3,7,8-substituted and non-2,3,7,8-substituted congeners. This was expected for the invertebrates (1,6) but has not been previously reported for any fish. The congeners present in the shellfish had a complex pattern, most similar to that observed from analysis of sediment samples from the same area, sewage sludge and, to some extent, incineration (Figure 1) (1,14).

4. CONCLUSIONS

Based on the results presented in Table 2, the PCDD/PCDF concentrations reported in the dairy products, meat and eggs collected from southern Mississippi are slightly lower than those previously reported in Europe (2, 6, 8, 9, 15), United States (4) and Canada (7) but higher than those reported for New Zealand (16).

The Spanish mackerel had similar PCDD/PCDF levels to those reported for other salt water fish species (1, 6, 7).

The farm raised catfish had elevated PCDD/PCDF levels, the highest 2,3,7,8-TCDD pg/g levels for any food item sampled, and a congener pattern more similar to an invertebrate than a vertebrate (1, 17). Neither the source(s) of the PCDD/PCDF nor the explanation why farm raised catfish would contain such high concentrations or exhibit such a pattern is known at this time. Further investigation of this finding is planned.

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