POLYCHLORINATED DIBENZO-P-DIOXIN/FURAN AND DIOXIN-LIKE PCB CONCENTRATIONS IN SEDIMENTS AND MUSSEL TISSUES FROM KENTUCKY LAKE, USA

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Introduction

Polychlorinated dibenzo-p-dioxins (PCDDs), dibenzofurans (PCDFs) and dioxin-like PCB congeners are among the most toxic chemicals to a variety of animals species including humans^{1,2}. PCDDs, PCDFs are found as byproducts in the production of PCBs, polychlorinated naphthalenes and chlorinated phenols. These pollutants are widely dispersed in the environment^{2,3} and the residues of these chemicals have been reported in air, water, soil, sediment, aquatic and terrestrial organisms including humans⁴⁻⁷. Due to persistent and bioaccumulative properties of these compounds, higher trophic level animals receive the largest amount of these contaminants and are implicated in a variety of health effects including, body weight loss, thymic atrophy, dermal disorder, hepatic damage, terratogenicity, reproductive toxicity and immunotoxicity in some animals⁸⁻¹⁰. The westernmost Kentucky is endowed with the highest densities of major rivers and reservoirs in the world, variety of industries and state-of-the-art agricultural operations. However, little is known on the levels of highly toxic dioxins, furans and dioxin-like polychlorinated biphenyls in sediments and biota of this watershed. The objective of the study was to determine the concentrations of 2,3,7,8-chlorine substituted PCDDs, PCDFs and non- and mono-ortho-chlorine substituted PCBs in surface sediment and mussels tissues collected from selected locations of Kentucky Lake and Kentucky Dam Tailwater.

Materials and Methods

Sampling locations and Samples

Kentucky lake is one the major man-made lakes in the U.S. Figure 1 shows the map of westernmost Kentucky, the Kentucky lake watershed and the sediment and mussel sampling locations. Ledbetter embayment of Kentucky Lake is considered relatively unpolluted, whereas Kentucky Dam Tailwater receives industrial wastewater from several industries (chemical, metallurgical etc) located in the Calvert City Industrial Complex. Selected locations in the Kentucky Lake including Ledbetter embayment and Kentucky Dam Tailwater were sampled for sediments and freshwater mussels during 1999 and 2000. Surface sediments (0-5 cm) samples were collected using PONAR grab sampler. Freshwater mussel samples were collected by SCUBA diving. The mussels were identified, measured for length, height and width, wet weight and age were determined. Mussel species collected and analyzed included, mapleleaf (*Quadrula*)

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quadrula), threeridge (Amblema plicata), ebonyshell (Fusconaia ebena) and washborard (Megalonaias nervosa).

Chemical Analysis

Sediment and mussel samples were freeze-dried and Soxhlet extracted using methylene chloride for 16h. Details of the analytical procedures were described elsewhere¹¹. Identification and quantitation of 2,3,7,8-substituted congeners of PCDDs/DFs and dioxin-like PCBs were performed using a Hewlett Packard 6890 Series High Resolution Gas Chromatography interfaced with a Micromass Autospec - Ultima High Resolution Mass Spectrometer.

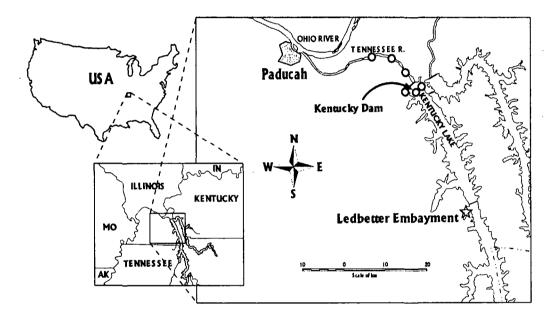


Figure 1. Map showing sediment and mussel sampling locations in the Ledbetter embayment (\bigstar) and the Kentucky Dam Tailwater (**o**).

Results and Discussion

Sediment and mussel tissues were analyzed for non-*ortho* and mono-*ortho*-chlorine substituted PCBs, polychlorinated dibenzo-*p*-dioxins and furans (PCDDs/DFs) and the mean concentrations of the analytes are presented in Tables 1. Average total dioxin-like PCBs in sediment from Ledbetter embayment were 120 pg g⁻¹ dry wt. Mussel tissues from the same general location recorded 35,000 pg g⁻¹ fat wt. Elevated concentrations of the dioxin-like PCBs were found in sediments (570 pg g⁻¹ dry wt.) and mussel tissues (390,000 pg g⁻¹ fat wt.) collected from Kentucky Dam Tailwater indicating relatively higher exposure to these compounds in this region. Among 2,3,7,8- substituted PCDD and PCDF concentrations in sediments and mussel tissues from Ledbetter embayment and Kentucky Dam Tailwater, OCDD/DF were most prominent detects in all of the samples analyzed (Table 1).

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Table 1. Mean concentrations of dioxin-like PCBs, polychlorinated dibenzo-*p*-dioxins (PCDDs) dibenzofurans (PCDFs) and TEQs in sediment (pg g^{-1} dry wt) and mussel tissues (pg g^{-1} fat wt) from Ledbetter embayment of Kentucky Lake and Kentucky Dam Tailwater, USA.

Compound	Sediment		Mussel Fissue	
_	Ledbetter	KY Dam	Ledbetter	KY Dam
	Embayment(n=5)	Tailwater(n=5)	Embayment(n=5)	Tailwater(n=5)
Non-ortho PCBs				
81	0.04	15	9.2	9.4
77	5.1	56	1300	1200
126	0.6	41	3100	110
169	0.1	5.3	22	12
Mono-ortho PCBs				
105	26	81	7900	8200
114	3.2	8.3	170	370
118	75	270	19000	24000
123	3.6	16	< 0.01	<0.01
156	6.1	44	1400	2600
157	2.0	11	520	610
167	2.4	17	1200	1500
189	0.5	3.0	67	560
TEQ (Mean)	0.1	0.3	19	0.7
PCDDs				
2,3,7,8-D	0.04	0.1	11	4.5
1,2,3,7,8-D	0.1	0.2	73	8.9
1,2,3,4,7,8-D	0.3	0.5	98	6.2
1,2,3,6,7,8-D	0.6	0.1	88	13
1,2,3,7,8,9-D	0.1	0.2	11	1.6
1,2,3,4,6,7,8-D	24	490	310	140
OCDD	830	1500	5400	2800
TEQ (Mean)	0.58	0.60	110	17.34
PCDFs				
2,3,7,8-F	0.2	0.6	66	88
1,2,3,7,8-F	0.1	0.9	470	25
2,3,4,7,8-F	0.1	0.3	83	13
1,2,3,4,7,8-F	0.3	3.8	63	27
1,2,3,6,7,8-F	0.2	1.2	57	15
2,3,4,6,7,8-F	0.1	0.3	93	11
1,2,3,7,8,9-F	0.03	0.2	64	4.5
1,2,3,4,6,7,8-F	2.1	22	68	48
1,2,3,4,7,7,8,9-F	0.1	2.8	71	29
OCDF	3.2	170	140	320
TEQ (Mean)	0.2	0.4	62.4	14
Total TEQ	0.9	1.4	192	32

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In general, PCDD homologues were abundant contaminants in sediments and mussel tissues indicating multiple local sources in the Kentucky Lake and Kentucky Dam Tailwater. Greater concentrations of OCDD and OCDF in the samples suggested agricultural and/or recreational (golf courses) sources that used pentachlorophenol-containing herbicides.

Toxic equivalents (TEQs) were calculated using WHO-TEF values proposed in 1998. Mammal TEFs and fish TEFs were used for sediment and mussel tissues respectively. TEQ data indicated that sediments from KY Dam Tailwater (1.3 pg g⁻¹ dry wt) and mussel tissues (190 pg g⁻¹ fat wt) from Ledbetter embayment contain higher toxic potential than sediment from Ledbetter embayment and mussel tissues from KY Dam Tailwater (Table 1). PCDD homologues greatly contributed to the toxicity followed by PCDFs, non-*ortho*-PCBs and mono-*ortho*-PCBs in all samples analyzed. In particular, 2,3,7,8-TCDD, 1,2,3,7,8-PnCDD, 2,3,7,8-TCDF, 1,2,3,7,8-PnCDF and 2,3,4,7,8-PnCDF comprised greater composition in toxic equivalents. Elevated concentrations of PCDD/DFs in mussel tissues indicate bioaccumulation of these compounds. Very little is known on the concentrations of dioxins and dioxin-like compounds in Kentucky Lake watershed. Further studies with greater number of samples from this watershed is needed to elucidate distribution, source, bioaccumulation and toxic effects.

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