

### Persistent Organochlorine Concentrations in Sediment and Mussel Tissues from the Lowermost Tennessee River and Kentucky Lake, U.S.A.

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#### Introduction

Environmental monitoring and assessment for the lowermost Tennessee River (Kentucky Dam Tailwater) and Kentucky Lake are becoming increasingly important due to increased incidence of mussel die-offs (extirpations that typically involve hundreds to thousands of mussels dying during a short period (days to weeks)) and the poor quality and quantity of shells harvested for the Japanese pearl industry from these regional water bodies during recent years [1,2]. These episodic events have raised considerable concern, both politically and environmentally. Levels of metal contamination in sediments and biota of this region have been studied in detail. However, very little is known on the levels of synthetic organic contaminants. The present study is our most recent effort in understanding the levels of contamination by selected chlorinated hydrocarbons and trace metals (data not presented here) in sediments and bivalve mollusks taken from the lowermost Tennessee River and Kentucky Lake (Fig 1). Bivalve mollusks have been demonstrated as excellent biomonitors of pollutants in the aquatic ecosystems, since they are sedentary, relatively hardy, of reasonable size, relatively insensitive to pollution and high bio-concentrative capacity for many organic and inorganic contaminants. The organic pollutants of concern are industrial chemicals, polychlorinated biphenyls (PCBs) and agricultural insecticides including, chlorinated hydrocarbon pesticides (DDT and its metabolites, hexachlorobenzene (HCB) and chlordane compounds). The organic pollutants are known for their widespread occurrence, biomagnification in the food chain, and long-term health effects in wildlife and humans [3,4]. This paper presents the concentrations of persistent, highly toxic organochlorine contaminants including PCB congeners, HCB, DDT and its metabolites, chlordane compounds in surface sediments and several species of mussel tissues collected from the lowermost Tennessee River and Kentucky Lake.

#### Material and Methods

Surface sediment (0-5 cm) samples were collected using Ponar Grab sampler. Pre-cleaned 1-Chem bottles were used to store (under -20 °C) the samples. Mussel samples were collected

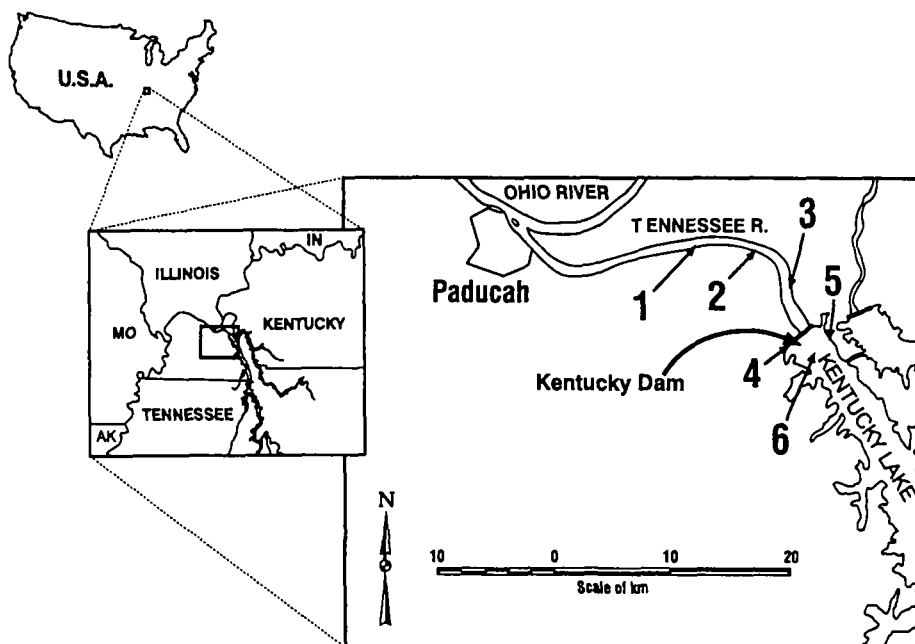


Fig. 1. Map showing sediment and mussels sampling locations.

Sampling was performed on May 21, 1997.

by SCUBA diving. The mussels were identified, measured, wet weight and age were determined (Table 1). The mussels were separated from the shells. Individuals of same species and age were pooled and transferred to pre-cleaned glass jars and stored under  $-20^{\circ}\text{C}$  until analysis. PCBs and chlorinated pesticides were analyzed following the published procedures [5,6] with some modifications. Sediment and mussel samples were freeze dried for over 60 hrs using Labconco FreeZone Freeze Dry System Model 77535. About 20 gm of dry sediment was Soxhlet extracted for 16 hrs using 3:1 mixture of methylene chloride and acetone was used as extraction solvent. Sample extract volume reduction was done using K-D apparatus. Silica gel column chromatography was used to separate PCBs and pesticides. Activated copper treatment and sulfuric acid clean-up procedures were followed to remove interfering materials in the instrumental analysis. PCB congeners were analyzed using Shimadzu model GC-17A gas chromatograph (GC) with Shimadzu model AOC-17 autoinjector. The GC was equipped with DB-5 (30m; 0.25mm i.d.; 0.25 $\mu$  film thickness) capillary column and a  $^{63}\text{Ni}$  electron capture detector. The column oven temperature program was  $90^{\circ}\text{C}$  (1.0 min) ...@  $5^{\circ}\text{C}/\text{min}^{-1}$  ... $150^{\circ}\text{C}$  (0 min.) ...@  $2^{\circ}\text{C}/\text{min}^{-1}$  ... $260^{\circ}\text{C}$  (15 min.). Injector and detector temperatures were set at  $270^{\circ}\text{C}$  and  $330^{\circ}\text{C}$  respectively. Helium (2 mL/ $\text{min}^{-1}$ ) and nitrogen (28 mL/ $\text{min}^{-1}$ ) were used as carrier and makeup gases respectively. PCB calibration standard SRM-2262 obtained from National Institute of Standards and Technology (NIST) was used for quantitations of PCB congeners 8, 18, 29, 50, 28, 52, 104, 44, 66+95, 101+90, 87, 77, 154, 118, 188, 153, 105+132, 138+163+164, 126, 187+182+159, 128, 200, 180, 170+190, 195, 206 and 209 in the samples. Pesticide calibration standard NIST-SRM-2261 was used for quantitations of pesticides. Analyte confirmation was done using GC (HP 5890 Series II) -MS (HP 5989A) for representative samples. Mussel tissues

**Table 1.** Details of mussel samples collected from the lowermost Tennessee R. & KY Lake.

Site No.	Common name	Genus and species	Length (mm)	Height (mm)	Width (mm)	Wet wt. (g)	Age (yrs.)
1	Mapleleaf	<i>Quadrula quadrula</i> (5)	59.7-77.8	53.0-62.9	31.6-42.2	70.7-129	8-10
	Threeridge	<i>Amblema plicata</i> (6)	66.8-81.7	57.0-65.2	36.7-45.6	101-176	8-10
2	Threeridge	<i>Amblema plicata</i> (5)	72.7-82.4	60.3-70.5	36.3-42.9	113-163	8-11
	Pink heelsplitter	<i>Potamilus alatus</i> (1)	115.1	87.6	38.3	159.1	9
3	Mapleleaf	<i>Quadrula quadrula</i> (4)	75.8-92.0	61.5-70.9	40.1-44.3	135-212	11-12
4	NC	NC	NC	NC	NC	NC	NC
5	Mapleleaf	<i>Quadrula quadrula</i> (1)	58.7	55.1	33.5	86.7	6
	Threeridge	<i>Amblema plicata</i> (1)	75.5	59.8	42.3	134	7
	Ebonysheal	<i>Fusconaia ebena</i> (1)	85.1	75.8	52.0	259	13
6	Washboard	<i>Megaloniais nervosa</i> (3)	136-143	97.0-100	55.0-56.2	495-561	23-27

( ): Values in parenthesis indicate number of specimens collected and used for analysis.

NC - mussels were not collected at this site for safety reasons

also analyzed using the above procedure except that about 2 to 5 g dry wt. (20 to 60 g wet wt.) of pooled (2 to 6 specimens) mussel tissue were used for extraction and lipid removal was done using Florisil dry column chromatography technique.

## Results and Discussion

Total PCB concentrations (ng g<sup>-1</sup> dry wt.) (sum of the congener quantitated) in the sediment and mussel tissues are presented in Table 2.

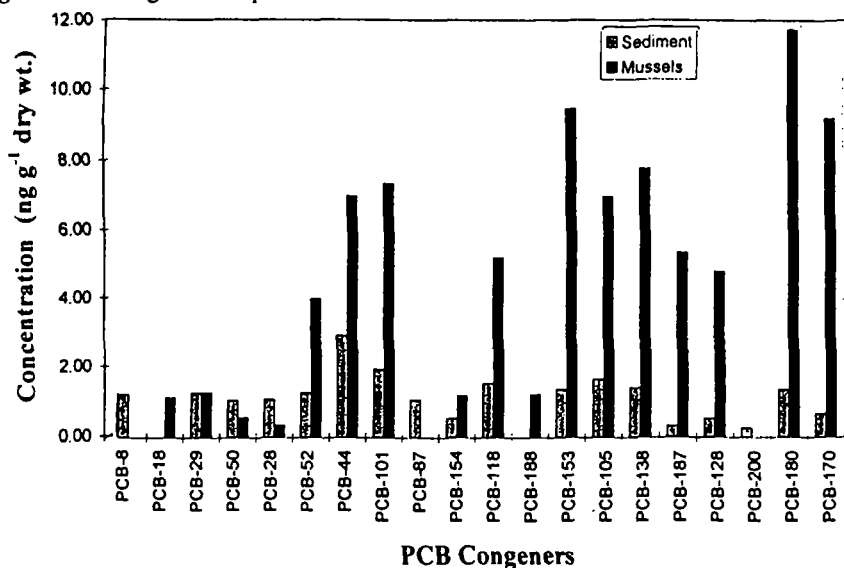
Site No.	Location (L= left bank , R= Right bank )	Total PCBs (ng g <sup>-1</sup> d wt.)	
		Sediments	Mussels
1	KY Dam Tailwater- TRM 15.2/ L	6.88	50.32
2	Air Products outfall- TRM 17.7/ L	26.32	82.72
3	I-24 Bridge 20m down- TRM 20.9/ R	1.00	40.04
4	KY Dam Marina- TRM 22.9/ L	6.30	NA
5	KY Lake Near Barges- TRM 23.1/ R	1.34	35.68
6	KY Lake- TRM 23.2/ L	0.90	18.94

TRM= Tennessee River Mile.

NA= Mussel samples were not collected at this site for safety reasons.

Total PCB concentrations ranged from detection limit (1.0 ng g<sup>-1</sup> d wt.) to 26.36 ng g<sup>-1</sup> d wt. in sediments and 18.94 ng g<sup>-1</sup> d wt. to 82.72 ng g<sup>-1</sup> d wt. in mussel tissue homogenates. In general, total PCB concentrations were relatively higher in the lowermost TN River sites (site #s 1-3) than in the sample from KY lake sites. The greatest concentration of total PCBs was recorded in the samples (both sediment and mussels) collected from site #2. Considering the PCB congener composition (Fig. 2), presence of PCB-28 (2,4,4'-trichloro), PCB-52 (2,2',5,5'-tetrachloro), PCB-101 (2,2',4,5,5'-pentachloro), PCB-153 (2,2',4,4',5,5'-hexachloro) and PCB-138 (2,2',3,4,4',5'-hexachlorobiphenyl) in most of the mussel tissue and also in sediment samples indicate that the samples were exposed to both lower chlorinated (Aroclor 1016 and Aroclor 1242) and higher chlorinated (Aroclor 1254 and Aroclor 1260) PCBs. Among the PCB congener measured, PCB-101, PCB-153 and PCB-138 (indicative of Aroclor 1254 and Aroclor 1260) were relatively higher in concentration and detected in most of the sediment and mussel tissue samples analyzed suggesting more exposure to the

Fig. 2. PCB congener composition of sediment and mussel tissues collected from site #2.



persistent Aroclors. Among the chlorinated pesticides, hexachlorobenzene (HCB) was consistently detected in almost all of the sediment and mussel tissues with highest concentrations (2.32 ng g<sup>-1</sup> d wt. sediment and over 500 ng g<sup>-1</sup> d wt. in tissues) at site 2. Other pesticides detected were DDT and chlordane compounds, and the concentrations were detected at a few ng/g. Presence of detectable concentrations of 4,4'-DDD and 4,4'-DDT in the samples collected from a few sites may indicate recent input of DDT.

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#### References

- Owen DA, Babbs SE, Sickel JB. 1990, *Manganese in Kentucky Reservoir Sediments, benthic water and mussels*. CRR, Murray State University, Murray, KY. 40 pp.
- Doubinis-Gray L, Maddox WE, Owen DA and Sickel JB. 1993, *Manganese accumulation and toxicity in mussels of Kentucky Lake*. Report to TVA, Knoxville, TN. 34 pp.
- Loganathan BG and Kannan K. 1994, *Ambio* 23,187-191.
- Kannan K, Tanabe S, Giesy JP and Tatsukawa R. 1997, *Rev. Environ. Contam. Toxicol.* 152, 1-55.
- Loganathan BG, Tanabe S, Goto M and Tatsukawa R; *Environ. Pollut.* 1989, 62, 237-251.
- Loganathan BG, Irvine KN, Kannan K, Pragatheeswaran V and Sajwan KS; *Arch. Environ. Contam. Toxicol.* 1997. 33. 130-140.