EFFECT OF SUBLETHAL CHLORINATED DISCHARGES ON PCB ACCUMULATION IN TRANSPLANTED ASIATIC CLAMS (*CORBICULA FLUMINEA*)

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Abstract. From 1987–1990, uncontaminated Asiatic clams (*Corbicula fluminea*) were placed in cages and transplanted into two streams receiving industrial discharges to help identify and quantify polychlorinated biphenyl (PCB) contamination to the streams. Clams accumulated substantial PCB residues at most sites monitored, with the exception of the sites closest to chlorinated discharges. Clams placed nearest to the chlorinated stream reaches consistently underestimated PCB contamination, based on the amount of PCBs found in fish and sediment at those sites. In a separate experiment, clams exposed in stream-side tanks to untreated (total residual chlorine ranged from 0.02–0.07 mg^{-1} L daily) and dechlorinated stream water exhibited differing degrees of valve movement, growth, and PCB accumulation after a four-week exposure to the two treatments. Clams exposed to untreated (chlorinated) stream water closed their shells more often, exhibited less growth, and accumulated substantially lower PCB concentrations than clams exposed to dechlorinated stream water. Clams apparently close their shells to avoid chlorine exposure, thus isolating clam tissues from PCBs found in the stream water and in the clams' food. Because chlorine and PCBs occur together in many industrial discharges, this finding is a significant consideration for monitoring programs that utilize clams to assess PCB bioavailability.

1. Introduction

Bivalves have been widely used as bioindicators of environmental contamination in aquatic systems (Phillips 1977; Leard *et al.*, 1980; Johnston and Hartley 1981; Farrington *et al.*, 1983; Doherty 1990). A common monitoring method using bivalves has been to transplant uncontaminated, caged bivalves to contaminated sites and analyze the soft tissue of the bivalves for contaminant accumulation after predetermined exposure periods (Foster and Bates 1978; Adams *et al.*, 1981; Kauss and Handy 1985; Czarnezki 1987; Muncaster *et al.*, 1990). In freshwater the Asiatic clam (*Corbicula fluminea*) has been often used for this purpose, in part because of the species widespread abundance and general tolerance of polluted conditions (Joy *et al.*, 1983; Hartley and Johnston 1983; Foe and Knight, 1986; Belanger *et al.*, 1990). *Corbicula* has been shown to readily detect polychlorinated biphenyl (PCB) contamination (Claeys *et al.*, 1975; Elder and Mattraw 1984; Tatem 1986; Southworth 1990). Uncontaminated *Corbicula* can be placed in cages and transplanted into small streams or discharges near industry where suitable resident organisms are often not present, providing an opportunity to detect localized sources of PCBs.

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Caged clams placed near or within some industrial discharges, however, may be susceptible to the effects of toxic constituents of the discharges that may attenuate the bioaccumulation capability of the clams. Chlorine is a common component of industrial cooling water discharges and effluents from wastewater treatment plants. Such facilities may also be potential sources of PCB contamination. If the presence of residual chlorine reduces the ability of clams to accumulate PCBs, the use of transplanted clams to locate discrete sources of PCB contamination could lead to very erroneous conclusions.

The objective of this study was to evaluate the effects of sublethal chlorinated discharges on PCB accumulation in caged *Corbicula fluminea*. Clams were temporarily transplanted into two PCB contaminated streams and monitored for PCB accumulation at sites near chlorinated effluents and at various distances downstream from chlorinated effluents. In a separate experiment we compared measurements of *Corbucula's* growth, valve movement, and PCB accumulation, with and without chlorine removed from the effluent.

2. Materials and Methods

PCB contamination was monitored in two East Tennessee streams [East Fork Poplar Creek (EFPC) and White Oak Creek (WOC)] using transplanted *Corbicula* on an approximately yearly basis between 1987 and 1990. The clam monitoring effort was part of broader-based biological monitoring programs mandated by National Pollution Discharge Elimination System (NPDES) permits issued to the U.S. Department of Energy's (DOE) facilities in Oak Ridge, Tennessee. PCB contamination in the two streams has been well documented since 1984 (Southworth 1990; Kornegay *et al.*, 1991). Locating and evaluating the relative importance to biota of the various PCB sources in the streams (i.e., from contaminated sediment, disposal sites, or effluent releases) is a major objective of the on-going monitoring programs.

Clams used in this study were initially obtained from one of three reference streams (Beaver and Bull Run Creeks, Knox County, Tennessee, and Little Sewee Creek, Meigs County, Tennessee) where clams were shown in preliminary investigations to contain PCB concentrations typical of uncontaminated streams. After collection, clams of similar size (approximately 16–20 mm in total length) were transported by water-filled buckets to the laboratory, and held in clean flowing water overnight. The following day, approximately 20 to 30 clams were placed in each of several 15×15×15 cm polypropylene cages with 1 cm mesh. The caged clams were then placed in water-filled buckets and transported to the monitoring sites. A subsample of the clams was frozen for PCB analysis as a baseline control for each monitoring period. One cage with associated clams was placed at each site monitored on EFPC and WOC (Figure 1).

For each monitoring period caged clams were exposed to the stream for four weeks, after which they were removed, wrapped in aluminum foil, placed on ice for

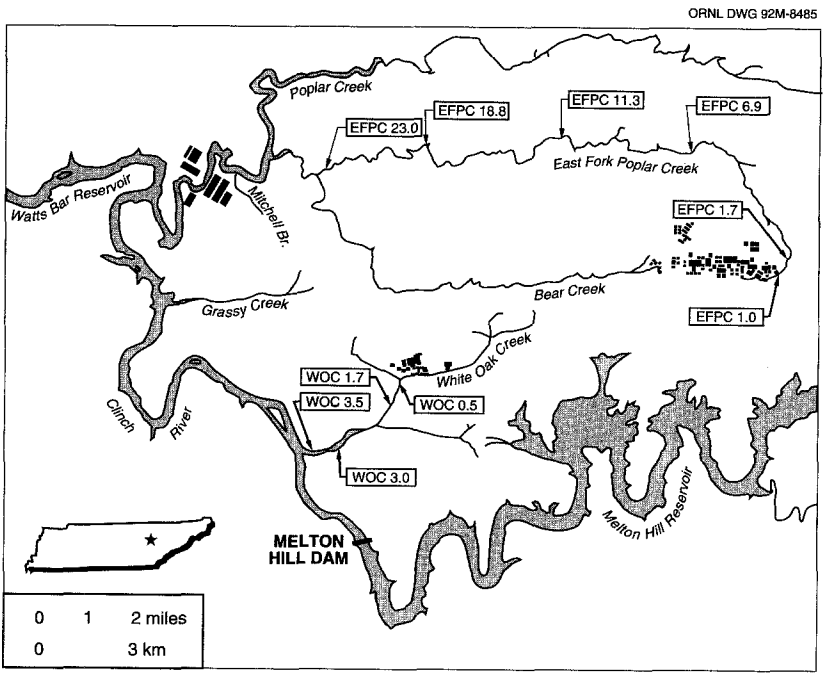

Fig. 1. Map showing locations of monitoring sites on East Fork Poplar Creek and White Oak Creek. Site designations refer to the approximate kilometer distance downstream of the facilities' chlorinated discharge(s).

transport, and frozen at -20 °C at the laboratory. Later the soft tissue was removed (without thawing) from the clam shells, placed in prewashed 20 ml glass vials, and stored at -20 °C prior to PCB analysis. A composite of approximately 10 clams was used for each PCB measurement sample, with two or three composite samples prepared for each site monitored.

Water containing elevated total residual chlorine (TRC) concentrations was discharged into EFPC and WOC primarily from sewage treatment facilities and the use of potable water in once-through cooling systems associated with operations at the DOE facilities (Kornegay *et al.*, 1990, Stewart 1992). Although the maximum concentration of chlorine in the effluents during this study (probably near the potable water concentration of 1 mg^{-1} L, total residual) was acutely toxic to most biota, the level of chlorine in the receiving streams decreased rapidly with distance downstream (Kornegay *et al.*, 1990, Ryon *et al.*, 1990; Kornegay *et al.*, 1991). At the monitoring sites nearest the chlorinated effluents (i.e., WOC 0.5 and EFPC 1.0; Figure 1), no mortality was observed in caged clams at the end of the four-week exposure periods.

In order to directly measure the possible effect of chlorine (measured as TRC) on PCB accumulation, clams were experimentally exposed to water from EFPC 1.0, a site known to have elevated concentrations of chlorine. This was accomplished using two 76-L, flow-through tanks positioned alongside EFPC 1.0 in March/April

1991. One tank received untreated stream water from EFPC, and the other tank received EFPC water treated with sodium thiosulfate to chemically reduce the chlorine in the water. Two cages were placed in each tank with ten similarly-sized uncontaminated clams in each cage. The length and weight of the ten unmarked clams in each cage were measured individually at 0 and 4 weeks exposure to obtain estimates of mean growth for the clams in each cage. The ten clams in each cage comprised one sample for PCB analysis, such that two duplicate PCB samples were prepared for each treatment. Valve movement (percent time open or closed) was monitored in two uncaged clams placed in each tank over a 36 day period coinciding with the caged-clam phase of the experiment. An automated computer system monitored the valve movements in the individual clams (Ham and Peterson, in press).

Fish tissue samples were extracted with methylene chloride followed by adsorption column cleanup, solvent exchange, and evaporative concentration prior to analysis by packed column gas chromatography using electron capture detection (EPA, 1980; EPA, 1984). PCBs were quantified against standard commercial mixtures (Aroclor 1254 and 1260). Concentrations of PCBs in clam composite samples from the uncontaminated reference streams averaged 0.06 ± 0.04 μg^{-1} g (mean ± sd, n=14). The analytical detection limit was 0.01 μg^{-1} g, wet weight. The mean recovery of known PCB quantities spiked into clam composite samples was 93 ± 9% (± sd, n=6).

3. Results and Discussion

3.1. PCB MONITORING

The mean PCB concentrations in *Corbicula fluminea* placed for four weeks at the most upstream sites on WOC and EFPC (WOC 0.5 and EFPC 1.0) were consistently lower than the concentrations in *Corbicula* placed at downstream sites throughout the entire study (Tables I and II). Mean PCB concentrations in clams placed at WOC 0.5 and EFPC 1.0 were similar to reference stream values (Tables I and II). In contrast, clams placed approximately 1 km downstream of the most upstream sites (WOC 1.7 and EFPC 1.7) accumulated substantial PCB residues (average of 0.76 and 0.45 μg/g at WOC 1.7 and EFPC 1.7, respectively). PCB concentrations in clams placed at sites greater than 1 km downstream from the facilities' discharges averaged an order of magnitude higher than PCB concentrations in reference stream clams. The results of clam monitoring strongly suggests that during the period of this study much of WOC and EFPC were PCB contaminated, with the exception of the stream reaches nearest to the industrial facilities.

The very low level of PCBs found in clams placed at WOC 0.5 and EFPC 1.0 suggests that the source(s) of PCBs to the two streams was downstream of those sites. However, the results of concurrent monitoring of fish and sediment during this time period revealed that substantial PCB contamination was evident at the most upstream sites (Rogers *et al.*, 1989, Kornegay *et al.*, 1990, Southworth 1990,

TABLE I

Concentrations of PCBs (μg g^{-1}, wet wt) in caged clams (*Corbicula fluminea*) held for four week exposure periods in White Oak Creek, 1987–1990. Values are the mean ± S.E. of duplicated composite samples, with the number of samples in parentheses

	Sites				
Month/Year	WOC 0.5	WOC 1.7	WOC 3.0	WOC 3.5	Reference stream[a]
July 1987	0.17±0.02 (2)	1.50 (2)	1.35±0.05 (2)	–	0.05±0.01 (2)
March 1988	0.06±0.00 (2)	–	0.82±0.05 (2)	0.37±0.01 (2)	0.05±0.01 (3)
November 1988	0.05±0.01 (2)	0.23±0.03 (2)	0.52±0.01 (2)	0.32±0.03 (2)	0.05±0.02 (2)
April 1990	0.06±0.04 (2)	0.54±0.03 (2)	0.90±0.11 (2)	0.74±0.46 (2)	0.01±0.00 (2)
Mean	0.09±0.03	0.76±0.38	0.90±0.17	0.48±0.13	0.04±0.01

[a] Clams were obtained each year from the following reference streams: Bull Run Creek in 1987 and March 1988, Beaver Creek in November 1988, and Little Sewee Creek in 1990.

TABLE II

Concentrations of PCBs (μg g^{-1}, wet wt) in caged clams (*Corbicula fluminea*) held for four week exposure periods in East Fork Poplar Creek in April of each year from 1987–1990. Values are the mean ± S.E. of duplicated composite samples, with the number of samples in parentheses

	Sites						
Year	EFPC 1.0	EFPC 1.7	EFPC 6.9	EFPC 11.3	EFPC 18.8	EFPC 23.0	Reference stream[a]
1987	0.14±0.06 (3)	0.57±0.11 (3)	0.50±0.03 (3)	0.49±0.03 (3)	–	–	0.08±0.01 (3)
1988	0.04±0.01 (3)	0.32±0.02 (3)	–	0.50±0.04 (3)	0.33±0.01 (3)	–	0.05±0.01 (3)
1989	0.04±0.03 (2)	0.19±0.01 (2)	–	–	–	–	0.12±0.01 (2)
1990	–	0.73±0.25 (2)	–	0.31 (1)	–	0.21±0.08 (2)	0.01 (2)
Mean	0.07±0.03	0.45±0.12	0.50	0.43±0.06	0.33	0.21	0.07±0.02

[a] Clams were obtained each year from the following reference streams: Beaver Creek in 1987 and 1989, Bull Run Creek in 1988, and Little Sewee Creek in 1990.

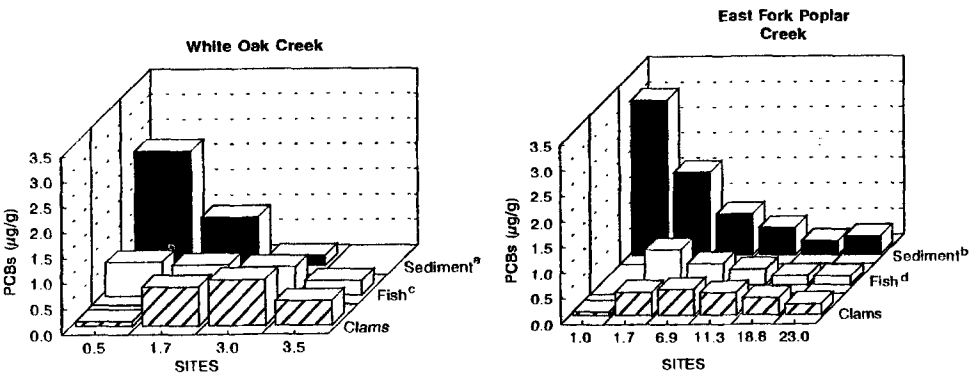

Fig. 2. Comparison of mean PCB concentrations (µg/g) found in surface sediment, fish, and caged clams (*Corbicula fluminea*) at sites on White Oak Creek and East Fork Poplar Creek; all available sampling data combined over the 1987–1990 time period. (a) Sediment not sampled in 1987. Concentrations represent estimated values (some samples were below detection limits). Data from Rogers *et al.*, 1989, Kornegay *et al.*, 1990, and Kornegay *et al.*, 1991). (b) Sediment sampled in 1986 and 1989. (c) N = 8 fish per site per year, 1987–1990. Data from Southworth 1990 and Kornegay *et al.*, 1991. (d) N = 16 fish per site per year, 1987–1990. Data from Southworth and Kornegay *et al.*, 1991.

and Kornegay *et al.*, 1991; Figure 2). Mean concentrations of PCBs in surface sediment (all sampling data combined over the 1986–1990 time period) at WOC 0.5 and EFPC 1.0 were 2.27 and 2.96 μg^{-1} g, respectively (Rogers *et al.*, 1989, Kornegay *et al.*, 1990, and Kornegay *et al.*, 1991). The substantial PCB concentrations in sediment at the most upstream sites, coupled with the steady decrease in sediment PCBs with distance away from the facilities, suggests ongoing PCB sources upstream of WOC 0.5 and EFPC 1.0. Although no fish were obtained from EFPC 1.0, fish collected from WOC 0.5 were highly contaminated with PCBs, averaging 0.67 μg^{-1} g from 1987–1990 (Figure 2). Thus, all of the relevant data suggest that the clams substantially underestimated the PCB exposure at the sites closest to facility discharges. At monitoring sites further downstream in each creek, clams appeared to be effective indicators of PCB contamination; the pattern of PCB accumulation in clams was similar to the pattern in sediment and fish (Figure 2).

3.2. Assessment of chlorine effects

The two most likely causes of lower PCB accumulation in clams placed nearest the facilities were hypothesized to be either (1) the absence of suitable or available food, which would decrease PCB uptake through the food chain, or (2) the presence of a toxicant(s) that caused physiological or behavioral stress in clams, resulting in decreased filtering/feeding activity. As mentioned previously, chlorine was implicated because it is a known toxicant discharged from the facilities on White Oak Creek and East Fork Poplar Creek.

Approximate concentrations of chlorine that the caged clams were exposed to can be estimated from TRC concentrations measured in water samples collected for toxicity testing near some of the clam placement sites during 1986–1991 (Stewart

A. J., ORNL Toxicology Laboratory, personal communication). Concentrations of TRC measured in the vicinity (within about 200 m) of the most upstream sites on each stream averaged approximately 0.04 mg^{-1} L from 1986-1991, except for samples taken prior to 1989 in EFPC, when chlorine concentrations were substantially higher (average of 0.17 mg^{-1} L, TRC; A. J. Stewart, ORNL Toxicology Laboratory, personal communication). A pronounced chlorine gradient has been observed on both creeks; mean TRC concentrations in the stream were found to be highest near some discharge points upstream of WOC 0.5 and EFPC 1.0, while TRC concentrations in stream water from further downstream in the vicinity of WOC 1.7 and EFPC 1.7 were on average below the limit of reliable detection (0.01 mg^{-1} L). Chlorine concentrations in the upper reach of WOC and EFPC have exhibited a diel cycle, at least partially due to photolysis of chlorine by sunlight (Stewart *et al.*, 1992). Thus, caged clams at EFPC 1.0 and WOC 0.5 were probably exposed to variable, and on average relatively low-level ($<$0.05 mg^{-1} L), TRC concentrations that were not lethal to clams over the various four week exposure periods.

Corbicula were exposed in stream-side tanks to untreated and dechlorinated water from EFPC 1.0 in order to directly measure the possible effects of exposure to sublethal concentrations of chlorine on PCB accumulation in clams. Clam stress at EFPC 1.0 was also assessed, in part, by measuring clam growth (i.e. length and weight increase) and behavior (i.e. valve movement) in these clams. Clams placed in untreated EFPC 1.0 water were exposed to a daily range of average hourly TRC concentrations of 0.02–0.07 mg^{-1} L (based on 18 days of TRC measurements taken during the second half of the monitoring period). The concentrations of chlorine in this experiment were similar to the concentrations observed near EFPC 1.0 from 1989–1991. The valve movement study showed that clams exposed to untreated water opened their shells only 30% of the time they were monitored (Figure 3D). In contrast, clams had their shells open almost 80% of the time when exposed to dechlorinated water from EFPC 1.0.

Clams exposed to the untreated water from EFPC 1.0 probably did not increase their filtering rate to compensate for the decreased time the shell was open, because clams in untreated water grew very little (mean length and weight increase were 0.12 mm and 0.01 g, respectively) in comparison to clams exposed to dechlorinated water from EFPC 1.0 (mean length and weight increase of 0.54 mm and 0.15 g, respectively; Figure 3A and 3B). The length increase of clams exposed to dechlorinated water from EFPC 1.0 was similar to the length increase in clams placed for 4 weeks in cages in an uncontaminated reference stream (Little Sewee Creek) in the summer of 1990 (0.49 mm increase). The substantial amount of growth in clams placed in dechlorinated water from EFPC 1.0 suggests that if chlorine had not been present at EFPC 1.0, food availability would not have been a problem for caged *Corbicula* placed at this site.

Clams placed in untreated water from EFPC 1.0 had a mean PCB concentration ($\pm$ S.E.) of 0.11 $\pm$ 0.02 μg^{-1} g, which was approximately six times lower than that observed in clams placed in dechlorinated water from EFPC 1.0 (0.63 $\pm$ 0.02 μg^{-1} g;

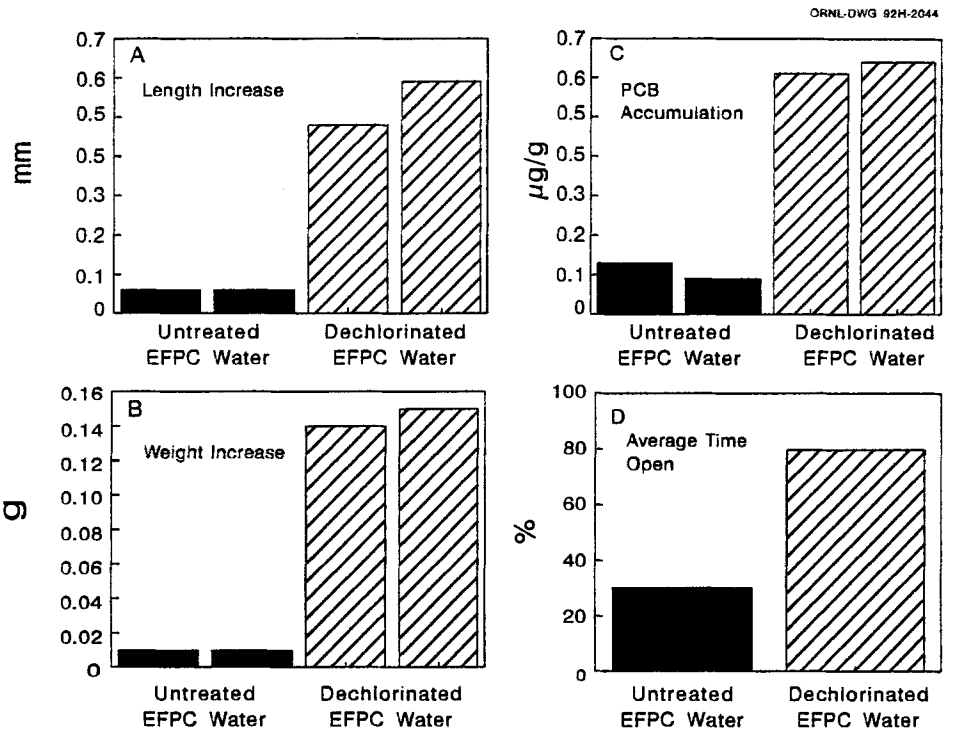

Fig. 3. Mean length increase (A), weight increase (B; each bar equals the mean of the ten clams in each cage), and PCB accumulation (C; each bar represents the concentration of a ten-clam composite sample) in clams (*Corbicula fluminea*) after four weeks exposure to untreated and dechlorinated water from EFPC 1.0. The percent time clam shells were open over the length of the experiment (n = 2 clams, each treatment) is shown in (D).

Figure 3C). The concentration of PCBs in clams exposed to untreated water from EFPC 1.0 was similar to the concentrations of PCBs found in caged clams placed at EFPC 1.0 in prior monitoring studies (Table II). Clams placed in dechlorinated water accumulated substantial PCB residues typical of those found in caged clams placed at monitoring sites further downstream. The results of the stream-side tank experiment suggest that clams used to monitor PCB contamination in chlorine impacted waters close their shells to avoid chlorine exposure, thus reducing filtering/feeding time and, consequently, PCB uptake.

4. Conclusion

This study showed that the presence of even very low and variable chlorine concentrations can cause transplanted *Corbicula* to substantially underestimate the potential PCB exposure at a site. This finding is a significant consideration for bioaccumulation monitoring programs using clams, because chlorine or other toxicants may exist near point sources of PCBs. Heavy users of electricity and electric power plants often are potential sources of PCB contamination, due to

their historic use of PCB-containing transformers. Municipal wastewater treatment plants, which receive wastes from a wide variety of customers, are also potential PCB sources. The effectiveness of using clams to locate PCB sources in close proximity to such facilities may be severely impaired by chlorine that is commonly discharged by these facilities as a concequence of wastewater disinfection, cooling water systems, or control of biofouling.

Before using clams to monitor bioaccumulation, the possible presence of chlorine (or other toxicants) at the site should be evaluated. To the extent possible, caged clams should be placed away from any toxicant exposure. If measurable levels of a stressor such as chlorine exist at a monitoring site, factors such as water quality, clam condition, and the amount of contaminant accumulation in other components of the stream ecosystem must be evaluated when interpreting the results of biological monitoring programs that utilize caged clams to assess contaminant bioavailability.

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References

Adams, T. G., Atchison, G. J. and Vetter, R. J.: 1981, *Hydrobiologia.* **83**, 67.

Belanger, S. E., Farris, J. L., Cherry, D. S. and Cairns J., Jr.: 1990, *Can. J. Fish Aquat Sci.* **47**, 904.

Claeys, R. R., Caldwell, R. S., Cutshall, N. H. and Holton, R.: 1975, *Pest Monitor J.* **9**, (1).

Czarnezki, J. M.: 1987, *Bull. Environ. Contam. Toxicol.* **38**, 641.

Doherty, F. G.: 1990, *Environmental Monitoring and Assessment* **15**, 143.

Elder, J. F. and Mattraw, H. C. Jr.: 1984, *Arch. Environ. Contam. Toxicol.* **13**, 453.

Environmental Protection Agency (EPA): 1980, 'Interim Methods for the Sampling and Analysis of Priority Pollutants in Sediments and Fish Tissues,' EPA 600/4-81-055. Environmental Monitoring and Support Laboratory, U.S. Environmental Protection Agency, Cincinnati, Ohio. 60 pp.

Environmental Protection Agency (EPA): 1984, 'Extraction and Analysis of Priority Pollutants in Biological Tissue, Method PPB 12/83' Environmental Services Division, Region IV, Analytical Support Branch, U.S. Environmental Protection Agency, Athens, Georgia. Mimeo, 10 pp.

Farrington, J. W., Goldberg, E. D., Risebrough, R. W., Martin, J. H. and Bowen, V. T.: 1983, *Environ. Sci. Technol.* **17**, 490.

Foe, C. and Knight, A.: 1986, 'A Method for Evaluating the Sublethal Impact of Stress Employing *Corbicula fluminea*, in Prezant RS (ed.), *Proceedings of the Second International Corbicula Symposium*, American Malacological Bulletin, Special Edition No. **2**, pp. 133–142.

Foster, R. B. and Bates, J. M.: 1978, *Environ. Sci. Technol.* **12**, 958.

Ham, K. D. and Peterson, M. J.: in press, *Environ. Toxicol. Chem.*

Hartley, D. M. and Johnston, J. B.: 1983, *Bull. Environ. Contam. Toxicol.* **31**, 33.

Johnston, J. B. and Hartley, D. M.: 1981, 'Bivalves as Monitors for Persistent Pollutants in Marine and Freshwater Environments', in S. M. Somani and F. L. Cavender (eds.), *Environmental Toxicology: Principles and Policies*, Charles C. Thomas, Springfield, pp. 184–198.

Joy, J. E., Pritchard, A. J. and Danford, D.: 1983, *West Virginia Acad. Sci.* **55**, 113.

Kauss, P. B. and Hamdy, Y. S.: 1985, *J. Great Lakes Res.* **11**(3), 247.

Kornegay, F. C., West, D. C., Goodpasture, S. T., Kimbrough, C. W., Tardiff, M. F., Jacobs, V. A. and Wilson, A. R.: 1990, 'Oak Ridge Reservation Environmental Report for 1989', ES/ESH-13/V1 & 2. Martin Marietta Energy Systems, Inc., Oak Ridge, Tennessee. Vol. **1**, 315 pp; Vol. **2**, 251 pp.

Kornegay, F. C., West, D. C., Goodpasture, S. T., Evans, R. A., Tardiff, M. F. and Wilson, A. R.: 1991, 'Oak Ridge Reservation Environmental Report for 1990', ES/ESH-13/V1 & 2. Martin Marietta Energy Systems, Inc., Oak Ridge, Tennessee. Vol. **1**, 283 pp; Vol **2**, 279 pp.

Leard, R. L., Grantham, B. J. and Pessoney, G. F.: 1980, *Pest. Monitor. J.* **14**, (2), 47.

Muncaster, B. W., Hebert, P. D. N. and Lazar, R.: 1990, *Arch. Environ. Contam. Toxicol.* **19**, 25.

Phillips, D. J. H.: 1977, *Environ. Pollut.* **13**, 281.

Rogers, J. G., Daniels, K. L., Goodpasture, S. T., Kimbrough, C. W. and Prince, N. L.: 1989, 'Oak Ridge Reservation Environmental Report for 1988', ES/ESH-8/V1 & V2. Environmental and Safety Activities, Martin Marietta Energy Systems, Inc., Oak Ridge, Tennessee. Vol. **1**, 304 pp.; Vol. **2**, 250 pp.

Ryon, M. G., Loar, J. M., Southworth, G. R., Stewart, A. J., Adams, S. M. and Kszos, L. A.: 1990, 'Evaluation of Fish Kills During November 1986 and July 1987 in Upper East Fork Poplar Creek Near the Oak Ridge Y-12 Plant', ORNL-3514. Oak Ridge National Laboratory, Oak Ridge, Tennessee, 48 pp.

Southworth, G. R.: 1990, *Water, Air, and Soil Poll.* **51**, 287.

Stewart, A. J., Hill, W. R. and Ham, K. D.: 1992, 'Sunlight and Periphyton Drive Chlorine Dynamics in Area Streams', pp. 68–69, *In Environmental Sciences Division Annual Progress Report for Period Ending September 30, 1991.* ORNL-3826. Oak Ridge National Laboratory, Oak Ridge, Tennessee. 279 pp.

Tatem, H. E.: 1986, *Arch. Environ. Contam. Toxicol*, **15**, 171.