Bull. Environ. Contam. Toxicol. 31, 377–385 (1983)

Temporal Trends Toward Stability of Hudson River PCB Contamination

Ronald J. Sloan,[1] Karl W. Simpson,[2] Roy A. Schroeder,[3] and Charles R. Barnes[4]

[1]New York State Department of Environmental Conservation, 50 Wolf Road Albany, NY 12233; [2]New York State Department of Health, Empire State Plaza, Albany, NY 12201; [3]U.S. Geological Survey, 24000 Avila Road, Laguna Niguel, CA 92677; and [4]U.S. Geological Survey, P.O.B. 744, Albany, NY 12201

Polychlorinated biphenyls (PCB), widely employed for 50 years by industry in a variety of applications, are regarded as ubiquitous environmental contaminants, although their production has stopped and use is restricted to "closed" containers (NATIONAL RESEARCH COUNCIL 1979).

PCB was used in the manufacture of electrical equipment at two General Electric Company (GE) facilities located on the upper Hudson River about 1 km apart in Fort Edward and Hudson Falls (Washington Co.), New York (Fig. 1). Table 1 synopsizes PCB contamination chronology and outlines the amounts of PCB used since 1966. Discharges of PCB from these plants resulted in concentrations in bottom sediments of the Hudson River which now exceed those of other major rivers by about two orders of magnitude (BOPP et al. 1981) and those of small remote streams by more than three orders of magnitude (CRUMP-WIESNER et al. 1973).

Intensive monitoring was initiated in 1977 by the New York State Department of Environmental Conservation (DEC) to ascertain the magnitude of and trends in contaminant conditions of biotic and physical strata. This paper summarizes PCB trends from 1977 to 1981 in three major monitoring components - water, multiplate residues and fish.

MATERIALS AND METHODS

Water is possibly the major route of PCB accumulation in organisms (BROWN et al. 1982; ARMSTRONG & SLOAN 1981) and is utilized by several municipalities along the river (SCHROEDER & BARNES 1983). Multiplate residues, composed of silt, algae and invertebrates collected on hardboard plates over a five-week exposure period, reflect PCB availability in the water column at a specific time and place. Concentrations are much higher than in water samples, minimizing problems with analytical detection limits. Fish accumulate exceptionally high levels of PCB (ARMSTRONG & SLOAN 1981) and therefore pose the principal environmental health hazard associated with PCB pollution.

Sample sites for water, multiplate samplers and fish are shown in Fig. 1. Control sites were located upstream from the former PCB discharges. Results from the upper and lower river sections are presented separately due to the hydrological differences between these two reaches. The upper river from Troy to Ft. Edward is part of the Champlain Canal and consists of a series of step-type navigation pools. Of the 270,000 kg of PCB thought to have been discharged to the Hudson River, 2/3 of this amount probably

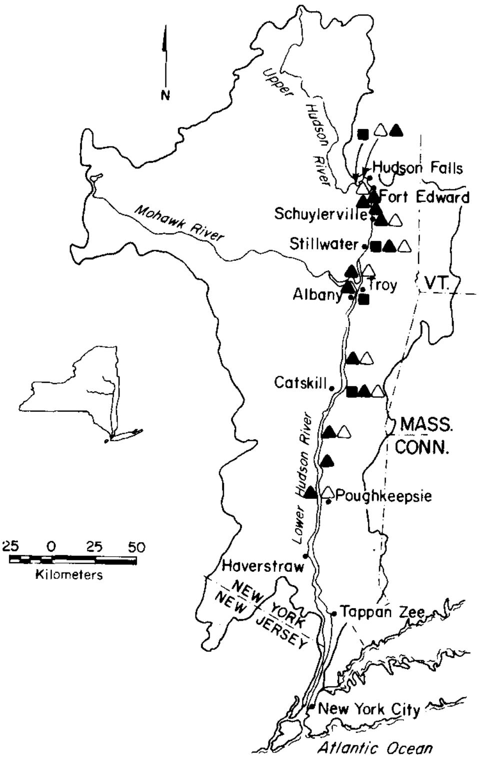


FIGURE 1. Hudson River locations sampled for PCB in water (△), multiplate samples (▲) and resident fish (■). Control areas denoted by arrows were upstream of the primary sites of PCB discharge. The upper and lower river sections are separated by a dam at Troy.

is still contained in sediments north of Troy (TOFFLEMIRE et al. 1979). The most highly contaminated sediments occur in the northernmost pool closest to the previous discharge sites (U.S. ENVIRONMENTAL PROTECTION AGENCY 1981). The lower river, extending from the mouth in New York City to Troy, is unimpounded and experiences tidal fluctuations throughout. All sampling sites in the lower river were located in the freshwater zone, which extends northward from Poughkeepsie (HOWELLS 1972). Water samples were collected throughout the year but only those during summer low-flow periods are included here to correspond with collection times of biota and to reduce the variable influences of sporadic high flow conditions. Low flow is defined here as <500 m^3/s as measured at the Waterford U.S. Geological Survey station just upstream from the confluence with the Mohawk River. At greater flows (>600 m^3/s), resuspension of bottom sediment becomes the dominant mechanism injecting PCB into the water (TURK 1980). Summer flow is usually between 100 and 300 m^3/s. Samples collected by passing a depth integrating sampler through the water column were placed in 1-liter glass bottles and extracted with hexane. Unfiltered samples were analyzed for total hexane-

TABLE 1. Timing of events related to the Hudson River PCB problem.

Year(s)	Event
1950* - 1976	PCB used by General Electric (GE) at Ft. Edward and Hudson Falls; discharges to Hudson River
1966 - 1975	PCB obtained by GE from Monsanto Corporation totals 37,284,807 kg (SOFAER 1976); quantity bought prior to 1966 unknown Purchases: 1966-70 Aroclor 1254 = 225,850 kg 1966-73 Aroclor 1242 = 22,819,048 kg 1971-75 Aroclor 1016 = 14,187,755 kg 1971-75 Aroclor 1221 = 52,154 kg
1973	Dam at Ft. Edward, retaining major PCB deposits, removed
1975	PCB-contaminated biota linked to discharge
1976	Restrictions placed on commercial and recreational fishing in Hudson River
1976	GE/DEC Settlement Agreement of $6 million to assess extent of contamination and evaluate possible remedial actions; PCB discharge at GE controlled
1976;1977	Last major spring flood stages in Hudson River
1977	PCB monitoring projects for fish, multiplate residues and water fully implemented; PCB discharge at GE terminated

* Earliest year established for PCB usage by GE (U.S. ENVIRONMENTAL PROTECTION AGENCY 1981; C. MCFARLAND of GE, personal communication, 1982). Aroclor is the trade name for various PCB mixtures produced by Monsanto Corporation, Sauget, Illinois.

extractable PCB and results therefore include dissolved and suspended fractions, though PCB is predominantly in the dissolved phase at low flow (TURK 1980).

Each multiplate sampler (HESTER & DENDY 1962) had a total surface area of 4620 cm^2 consisting of 10 hardboard plates stacked with 3 mm to 6 mm spaces between each 15.2 cm square board and was suspended 1 m beneath the water surface from a navigation buoy. Each year, multiplate samplers were initially installed during the first week in June, collected after a 5-week exposure period (July) and replaced with fresh samplers. Subsequent collections, after 5-week exposures, were made in August and September. Accumulated organisms and silt were scraped off the plates and placed in glass jars. Samples were lyophilized and hexane extracted prior to standard

Florisil clean-up. Resident fish were collected during the spring of each year to reduce the potential for seasonal variation in contaminant loads (WILSON & FORESTER 1978; EDGREN et al. 1981). Fillets were prepared from one side of the fish with the skin intact. Weighed portions of the ground, homogenized fish tissue were dried with anhydrous sodium sulfate, extracted with petroleum ether and cleaned up on a Florisil column (ARMSTRONG & SLOAN 1981).

Fish were individually analyzed for PCB but the data presented here represent composite values of up to 7 species for each river reach and year. This procedure increased the sample sizes providing a more robust temporal trend assessment. Patterns of PCB contamination in individual fish species were evaluated and presented elsewhere (ARMSTRONG & SLOAN 1981; SLOAN & ARMSTRONG 1981). Fish species composited in the upper Hudson River included brown bullhead (Ictalurus nebulosus), largemouth bass (Micropterus salmoides), yellow perch (Perca flavescens) and goldfish (Carassius auratus). In the lower river, the species mix was more varied between years with the composite samples utilizing brown bullhead, largemouth bass, smallmouth bass (Micropterus dolomieui), yellow perch, white perch (Morone americana), goldfish, common carp (Cyprinus carpio) and redbreast sunfish (Lepomis auritus). Anadromous species were not included with these analyses because the contaminant patterns differ from those of resident fish (SLOAN & ARMSTRONG 1981).

Samples were analyzed using gas chromatographs (GC) with electron capture detectors (U.S. ENVIRONMENTAL PROTECTION AGENCY 1979). Analyses of water samples were conducted by the U.S. Geological Survey laboratory in Doraville, Georgia; multiplate samples were analyzed by laboratories within the New York State Departments of Health and Environmental Conservation and fish by Hazleton Raltech, Inc., Madison, Wisconsin. Laboratory procedures varied somewhat for the three monitoring strata but results were consistent for each as reflected through quality control checks (SLOAN & ARMSTRONG 1981; SCHROEDER & BARNES 1983). PCB concentrations were determined by comparing chromatogram peak height or area with measurements of selected peaks corresponding to various Aroclor standards. Sample chromatograms rarely exactly matched those of the Aroclor standards. Hence, use of the specific Aroclor terms is meant to provide a common basis for comparison and discussion, and does not imply the occurrence of precise Aroclor mixtures in the samples. "Aroclor 1221", "Aroclor 1016" and "Aroclor 1254" refer to PCB mixtures with average percent chlorine by weight saturations in the biphenyl ring of approximately 21%, 40-42% and 54%, respectively. Aroclor 1016 was not distinguishable from Aroclor 1242 by the GC techniques used. GE used both Aroclors and the results reflect various combinations of these two mixtures. Concentrations in water are expressed as total PCB based on comparisons of chromatograms to an Aroclor or mixtures of two Aroclors to which a sample most closely corresponded (SCHROEDER & BARNES 1983).

The weight of material extracted by hexane or petroleum ether for the multiplate and fish samples was used as an estimate of lipid content. PCB concentration was expressed per unit lipid which has been shown to reduce the inter- and intraspecific variability for biological samples (ARMSTRONG & SLOAN 1981; CLAYTON et al. 1977; MONOD & KECK 1982).

RESULTS AND DISCUSSION

Background concentrations of PCB in river water measured above Hudson Falls (Fig. 1) were always below the detection limit of 0.05 µg/l (ERDMAN et al. 1982). Control site annual mean concentrations for multiplate residues and fish ranged from 17-88 and 12-90 µg/g lipid, respectively.

Each year, all three sample types from the most highly contaminated portion of the upper Hudson River had higher PCB concentrations than those from the lower river (Fig. 2 and Table 2) by factors ranging from 1.4 to 4.8. Both river sections exhibited declining PCB concentrations over the course of the study, but the upper river response was more pronounced. For water, the greatest measured declines occurred in 1980 and 1981 but limited unpublished data in the upper section immediately above the confluence with the Mohawk River reflected the possibility that the highest water concentrations occurred in 1976. That year the average summer low flow concentration was 0.70 µg/l (N=6) compared to 0.40 µg/l (N=20) in 1977 at the same location. The multiplate residues and fish experienced their initial significant declines in 1979 ($P < 0.05$) (Fig. 2). However, the multiplate sample concentrations rose sharply in 1981.

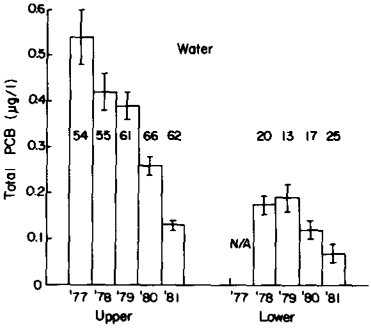
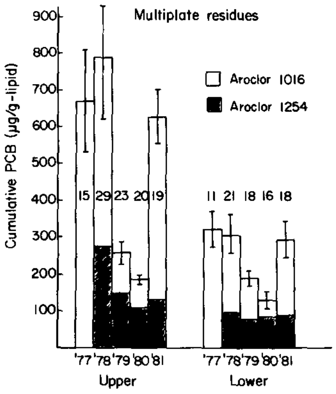


FIGURE 2. Mean annual concentrations of PCB in water, multiplate samples and fish from upper and lower Hudson River sections, 1977-81. Sample sizes appear in or above the histogram bars. Standard errors of the means for total PCB are denoted at the topof each bar. Water concentrations were not measured in 1977 in the lower river. Aroclor types were not differentiated for the 1977 multiplate sample data since the contracting laboratory that year did not use quantitation methods distinguishing PCB mixtures. The 1981 fish samples from the upper river were lost due to a freezer malfunction.



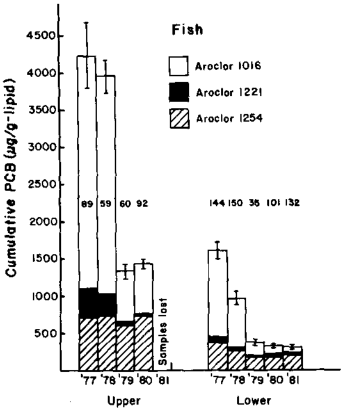


Table 2. Mean annual PCB concentrations (± S.E.s) in water (μg/1), multiplate samples (μg/g of lipid) and fish (μg/g of lipid) collected from the Hudson River. Sample sizes are noted in Fig. 2

			Years*				
Stratum	Location	PCB type	1977	1978	1979	1980	1981
Water	Upper	Total	.54±.06	.42±.04	.39±.03	.26±.02	.13±.01
	Lower	Total	N/A	.18±.02	.19±.03	.12±.02	.07±.02
Multiplate samples	Upper	Aro 1016	N/A	518±3	110±15	75±6	493±58
		Aro 1254	N/A	274±36	148±17	111±8	133±14
		Total	671±140	792±168	257±31	186±12	626±71
	Lower	Aro 1016	N/A	210±51	110±13	45±7	207±29
		Aro 1254	N/A	98±10	79±9	86±17	90±21
		Total	323±49	307±58	189±21	131±23	297±49
Fish	Upper	Aro 1221	383±42	295±18	49±5	33±5	N/A
		Aro 1016	3108±368	2913±161	674±48	664±32	N/A
		Aro 1254	721±64	743±53	607±39	734±39	N/A
		Total	4217±61	3951±214	1332±103	1431±66	N/A
	Lower	Aro 1221	79±8	40±2	13±6	36±5	19±3
		Aro 1016	1154±82	667±62	176±18	106±7	87±6
		Aro 1254	371±34	262±25	182±23	185±20	213±16
		Total	1604±110	969±84	371±38	327±25	319±22

* N/A denoted information that was not available or not analyzed.

Changes in total PCB for both multiplate samples and fish were due almost entirely to changes in concentration of Aroclor 1016. From 1978-1981 there was no appreciable change in the concentration of Aroclor 1254.

The 1981 fish samples from the upper river were lost but lower river results reflected the continuing trend toward declining levels observed in 1979-1980. Aroclor 1221 consistently declined in the fish from both river sections and, for the most part, is now less than the detection limit of 0.1 µg/g of wet tissue. This mixture was never quantified in the multiplate samples, but levels were substantially lower compared to those in the fish.

The consistently higher PCB concentrations in the samples from the upper river supports the contention that the majority of PCB now contaminating the river originates from sediments in the upper section. Further downstream, PCB concentrations are lowered by the admixture of less contaminated tributary waters. Hydrological events may have played a key role in the recent declines in PCB concentrations. The last major flood occurred in the spring of 1977 and resulted in substantial resuspension of contaminated sediments. Approximately 40% (± 20%) of all PCB transported in 1977 occurred during that flood event (TURK 1980). Recent flows in the river have been comparatively low and the contaminated sediments have been relatively undisturbed. Another major flood might increase the contaminant levels.

Other factors involved in the reduction of PCB since 1978 include the elimination of the point sources in 1977 and the maintenance dredging of some sections for the Champlain Canal. Although performed to deepen the navigation channel, the dredging removed some contaminated sediments from the river (TOFFLEMIRE et al. 1979). Also, removal in 1973 of the Ft. Edward hydroelectric dam below the previous discharge sites released major deposits of contaminated sediments allowing their transport downstream. The highly contaminated shoreline and redeposited sediments at the village of Ft. Edward received some remedial attention (dredging and streambank stabilization) in 1974-75 and again in 1977-78 following a spring flood event in 1976. This artificial stabilization was intended to inhibit further PCB release to the river through erosion (HETLING et al. 1979, U.S. ENVIRONMENTAL PROTECTION AGENCY 1981).

The surge of PCB in 1981 multiplate samples is not consistent with the water and fish data, and the reason for the increase is not known. There were no major dredging activities in 1981 and the hydrologic profile is similar to that for 1980 when low concentrations were found. The 1982 water samples, not reported here, do not show an increase over 1981 levels. Also, yearling pumpkinseed (*Lepomis gibbosus*) collected in the fall of 1981, as part of an additional study but not included here, and the spring 1982 fish samples from the lower river have Aroclor 1016 values comparable to those in 1980-81.

Accumulation kinetics of biphenyl molecules do vary with chlorine substitution (BRUGGEMANN et al. 1981). Some congeners may have selectively accumulated over the short-term exposures in the multiplate samples, as opposed to water or fish samples. However, such detail is not available from the chromatograms produced by the GC techniques used. Differential accumulation of silt and algae was not determined but such effects could not explain the 1981 increase since concentrations in caddisfly larvae (trichoptera: Hydropsychidae) collected as part of another study (unpublished) also showed a similar increase in Aroclor 1016 concentrations.

Initial responses to eliminating a contaminant source may be rapid for PCB (ARMSTRONG & SLOAN 1981; WILSON & FORESTER 1978) and DDT (GRIER 1982) but at least for PCB, within a few years relatively stable conditions ensue (WILSON & FORESTER 1978), or might be expected (THOMANN & ST. JOHN 1979). The temporal constancy of Aroclor 1254 concentrations in both multiplate and fish samples indicates the persistence of the more highly chlorinated compounds in biological tissues and is consistent with laboratory studies on fish, bivalves, soils and microbial degradation (HALTER & JOHNSON 1977; LANGSTON 1978; TUCKER et al. 1975; FURUKAWA et al. 1979) related to differing chemical and physical properties of PCB congeners such as volatility, solubility and stability (BALLSCHMITER et al. 1978; ROBERTS & MARSHALL 1980; U.S. ENVIRONMENTAL PROTECTION AGENCY 1976). Lack of significant change in the higher chlorinated PCB component for the period 1977-81 suggests that a dynamic equilibrium has been reached and that these compounds may continue to exist at present levels. This has serious economic implications, since current PCB levels in many Hudson River fish species are above the U.S. Food and Drug Administration temporary tolerance level of 5 parts per million on a wet weight basis (HILE 1977). The once-productive commercial fishery in the Hudson River has been severely restricted since 1975 due to PCB contamination. Continuing high levels of PCB will also preclude full development of the economic potential of the recreational fishery (U.S. ENVIRONMENTAL PROTECTION AGENCY 1982).

The history of Hudson River contamination is unique because of the discharge of massive quantities of closely related compounds from a point source over an extended period. This comparative simplicity has provided an opportunity to study the natural elimination response of the river. Our observations should be broadly transferable to other large rivers where contamination is severe.

Acknowledgments. This work was supported by GE-DEC settlement funds, step one grant monies from the U.S. Environmental Protection Agency, and supplemental services of the U.S. Geological Survey and the New York Departments of Health and Environmental Conservation. We thank Marian O'Sullivan for typing the manuscript.

ARMSTRONG, R.W. and R.J. SLOAN. Proc. Hudson R. Environ. Soc. Norrie Point, N.Y., 1981 (in press).

BALLSCHMITER, K., M. ZELL, H.J. NEU. Chemosphere 2, 173 (1978).

BOPP, R.F., H.J. SIMPSON, C.R. OLSEN, N. KOSTYK. Environ. Sci. Technol. 15, 210 (1981).

BROWN, M.P., J.J.A. MCLAUGHLIN, J.M. O'CONNOR, K. WYMAN. Ecol. Model. 15, 29 (1982).

BRUGGEMAN, W.A., L.B.J.M. MARTRON, D. KOOIMAN, O. HUTZINGER. Chemosphere 10, 811 (1981).

CLAYTON, J.R., Jr., S.P. PAVLOU, N.F. BREITNER. Environ. Sci. Technol. 11, 676 (1977).

CRUMP-WIESNER, H.J., H.R. FELTZ, M.L. YATES. J. Res. U.S. Geol. Surv. 1, 603 (1973).

EDGREN, M., M. OLSSON, and L. REUTERGARD. Chemosphere 10, 447 (1981).

ERDMANN, D.E., E.R. ANTHONY, G.R. PERRYMAN. U.S. Geol. Surv. Open-file Rep. 82-766 (1982).

FURUKAWA, K., N. TOMIZUKA, A. KAMIBAYASHI. Appl. Environ. Microbiol. 38, 301 (1979).

GRIER, J.W. Science 218, 1232 (1982).

HALTER, M.T. and H.E. JOHNSON. In: Aquatic Toxicology and Hazard Evaluation ASTM STP 634. American Society for Testing and Materials (1977).

HESTER, F.E. and J.S. DENDY. Trans. Am. Fish. Soc. 91. 420 (1962).

HETLING, L.J., T.J. TOFFLEMIRE, E.G. HORN, R. THOMAS, R. MT. PLEASANT, Ann. N.Y. Acad. Sci. 302, 630 (1979).

HILE, J.P. Fed. Reg. 42, 17487 (1977).

HOWELLS, G.P. Proc. R. Soc. B. 180, 521 (1972).

LANGSTON, W.J. Mar. Biol. 46, 35 (1978).

MONOD, G. and G. KECK. Bull. Environ. Contam. Toxicol. 29, 570 (1982).

NATIONAL RESEARCH COUNCIL. Polychlorinated Biphenyls. National Academy Sciences, Washington, D.C. (1979).

ROBERTS, J.R. and W.K. MARSHALL. Ecotoxicol. Environ. Safety 4, 158 (1980).

SCHROEDER, R.A. and C.R. BARNES. U.S. Geol. Surv. Water-Resources Invest. (submitted for public. 1983).

SLOAN, R.J. and R.W. ARMSTRONG. Proc. Hudson R. Environ. Soc. Norrie Point, N.Y. 1981 (in press).

SOFAER, A. Interim Order and Opinion, File #2833. New York Department of Environmental Conservation, Albany (1976).

THOMANN, R.V. and J.P. ST. JOHN. Ann. N.Y. Acad. Sci. 320, 610 (1979).

TOFFLEMIRE, T.J., L.J. HETLING, S.O. QUINN. N.Y. Dept. Environ. Conserv. Tech. Paper #55. Albany (1979).

TUCKER, E.S., W.J. LITSCHGI, W.M. MEES. Bull. Environ. Contam. Toxicol. 13, 86 (1975).

TURK, J.T. In: Contaminants and Sediments. Ann Arbor Science, Ann Arbor, Mich. (1980).

U.S. ENVIRONMENTAL PROTECTION AGENCY. Criteria Document PCBs. Office Toxic Substances, Washington, D.C. (1976).

U.S. ENVIRONMENTAL PROTECTION AGENCY. Fed. Reg. 44, 69501 (1979).

U.S. ENVIRONMENTAL PROTECTION AGENCY. Draft Environmental Impact Statement on the Hudson River PCB Reclamation Project. EPA, New York (1981).

U.S. ENVIRONMENTAL PROTECTION AGENCY. Final Environmental Impact Statement on the Hudson River PCB Reclamation Demonstration Project. New York (1982).

WILSON, A.J. and J. FORESTER. Bull. Environ. Contam. Toxicol. 19, 637 (1978).

Accepted June 6, 1983.