

ner pattern from a presumed original pattern.

The composition and quantity of PCB discharged to the Hudson River from two General Electric (GE) capacitor factories for approximately 30 years was essentially unmonitored (2). Cooling drainage, steam condensation, detergent washing processes, and a network of interconnected diffuse sources supplied PCB from one of the plants to the river (3). A significant portion of the PCB derived from the other plant was discharged to the river from a municipal sewage treatment plant (3). The discrepancy between the composition of PCB in river sediment and the proportions of Aroclors in GE's incomplete purchase records has been known since the early 1970s (2).

Simple quantitative analyses invalidate the presumption of sediment with a PCB composition like Aroclor 1242. A summary of our recent analyses of PCB composition in sediment within 12 km of GE discharges is shown in Table 1. Mono-, di-, hexa-, and heptachlorobiphenyls are enriched relative to Aroclor 1242, whereas tetra- and pentachlorobiphenyls are in proportions similar to that of Aroclor 1242. If Aroclor 1242 is used as a yardstick, the logic of Brown *et al.* would indicate chlorination as well as dechlorination.

When the authors' pattern A is used as a reference, there are substantial differences between patterns A and B that cannot be explained by the proposed dechlorination scheme. J. F. Brown provided detailed data for 18 consecutive sections of the sediment core 18-6. These data have been summarized elsewhere (6) and have been used to calculate numbers of halftimes for the dechlorination process (7). According to the authors, 2,4,2',4'-tetrachlorobiphenyl appears to be stable, with a net reduction of approximately 5% occurring over 15 years (7).

Using their data we calculated the mole ratio of 2,4,2',4'-tetrachlorobiphenyl to total PCB. The four samples with pattern A have mole ratios of 2.2 to 2.9%. Mole ratios of the six samples with pattern B ranged from 1.4 to 1.7%. The averages of these ratios imply a 68% enrichment of PCB molecules relative to 2,4,2',4'-tetrachlorobiphenyl in pattern B. Moreover, the sum of the comparatively stable 2,4,2',4'-, 2,4,2',5'-, and 2,5,2',5'-tetrachlorobiphenyls (7) indicates enrichment of 36% in pattern B relative to pattern A. The mole proportion calculations indicate pattern modification with enrichment by selective deposition. The average total PCB concentration in pattern B samples in this core is 1100 µg/g, which is ten times the average of pattern A samples. In the data provided to

Table 1. Composition of PCB in Hudson River sediment the Fort Edward Area (4) and in Aroclors (5).

PCB isomer group	Sediment (%)	PCB Aroclors			
		1221 (%)	1242 (%)	1254 (%)	1260 (%)
Monochlorobiphenyl	6.5	51	1	0.1	
Dichlorobiphenyl	21.7	32	16	0.5	
Trichlorobiphenyl	23.5	4	49	1	
Tetrachlorobiphenyl	29.1	2	25	21	
Pentachlorobiphenyl	10.0	0.5	8	48	12
Hexachlorobiphenyl	4.2		1	23	38
Heptachlorobiphenyl	3.5		0.1	6	41
Octachlorobiphenyl					8

us, the concentrations of all individual PCB congeners are higher in pattern B samples than in pattern A samples. A sedimentological and PCB transport framework for the proposed dechlorination scheme appears necessary for the interpretation of real and relative differences between PCB congener concentrations in recent and in earlier sediment deposits in the Hudson River. Such a framework is lacking in the three publications by Brown and his colleagues (1, 6, 7) regarding anaerobic dechlorination of PCB in the environment.

The hypothesis of anaerobic microbial dechlorination of PCB in the Hudson River is perhaps not confirmable by the use of logic that requires an assumption of a prior composition of PCB in river sediment. An alternative hypothesis, that pattern variations could have been caused primarily by a number of physical and chemical processes in the factories, in wastewater treatment, and during the process of river transport and sedimentation, cannot be rejected. We remain unconvinced that microbial PCB dechlorination has occurred in the Hudson River.

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REFERENCES

1. J. F. Brown *et al.*, *Science* **236**, 709 (1987).
2. A. D. Sofaer, *Interim Opinion and Order, in the Matter of Alleged Violations of Sections 17-0501, 17-0511, and 11-0503 of the Environmental Conservation Law of the State of New York by General Electric Company, Respondent* (1976).
3. Clark Dietz and Associates, "Engineering Report, wastewater monitoring program—phase III" (General Electric Co., Hudson Falls, NY, 1975).
4. B. Bush, L. A. Shane, M. Wahlen, M. Brown, *Chemosphere* **16**, 733 (1987).

5. W. Y. Shiu and D. Mackay, *J. Phys. Chem. Ref. Data* **15** (1986).

6. J. F. Brown *et al.*, *Northeast. Environ. Sci.* **3**, 167 (1984).

7. J. F. Brown, *Environ. Toxicol. Chem.* **6**, 579 (1987).

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Response: We disagree with the representation by M. P. Brown and his coauthors (1) of (i) the extent of the available information on the original composition of the PCBs deposited in the sediments of the Hudson River immediately below Fort Edward, New York; (ii) the nature of the sediment sample subpopulation that was selected for reporting in table 1 of their comment; (iii) our reported data on the stability of 2,4,2',4'-tetrachlorobiphenyl (24-24 CB) levels in upper Hudson River sediments; and (iv) the compositional alterations that would be expected if deposition occurred by means of their proposed (2) sedimentation hypothesis.

Although the middle and lower portions of the Hudson River have received PCBs from many sources, it is generally agreed that most of those in upper Hudson Reaches 8 and 9 (river miles 188.5 to 194.8) came from General Electric capacitor manufacturing plants located just upstream in Hudson Falls and Fort Edward, New York. Releases occurred mainly between 1955 and 1971. In 1971, major control measures were taken that sharply reduced the discharges and changed their pattern to that reported in reference 3 of M. P. Brown's comment (1). Monsanto sales records made publicly available in 1982 show that during the period 1955 through 1971 these plants' PCB purchases were 97.4% Aroclor 1242 (50.6×10^6 kg) and 2.6% Aroclor 1254 (1.4×10^6 kg). The composition of the General Electric releases was described by M. P. Brown *et al.* as predominantly Aroclor 1242 as recently as early 1987 (2). The overall composition of the minor, non-General Electric contributions to the Reach 8–9 deposits is not known; however, we have seen lightly contaminated sediment and fish specimens, collected both

Table 1. Estimated original and observed 1984 PCB compositions in upper Hudson River hot spots and cold spots.

Biphenyl parameters	PCBs (%)		
	Estimated original	Cold spot*	Hot spot†
Monochloro	1.4	6.5	34.6
Dichloro	10.7	21.7	36.5
Trichloro	46.1	23.5	17.0
Tetrachloro	32.0	29.1	7.7
Pentachloro	6.3	10.0	2.9
Hexachloro	2.6	4.2	0.9
Heptachloro	0.9	3.5	0.2
Octachloro	0.2	—	0.0
Total PCB's	—	2.8‡	345.7‡
Summary (per biphenyl)			
Ortho Cl's	1.46	—	1.44
Meta + para Cl's	1.92	—	0.50

*Unweighted average PCB composition and median PCB level as reported in (1) and (2). †Weighted average PCB composition and mean PCB level for all 60 core sections collected in 1984 for studies reported in (3–5), including those showing alteration patterns A, B, B', C, and E. ‡Parts per million.

above and below the subject area, that showed unambiguous patterns for the more heavily chlorinated products, Aroclors 1254, 1260, 1268, and 1270. Our analyses of minimally altered Reach 9 remnant deposits suggest the original composition of the PCBs in the "hot spots" (sediments with more than 50 ppm PCB) to have averaged approximately 94% Aroclor 1242, 5% Aroclor 1254, and 1% Aroclor 1260/1268. The homolog distribution calculated for such a composition from our own Aroclor composition data is shown in the first column of our Table 1.

M. P. Brown's testimony before the New York State Industrial Hazardous Wastes Facility Siting Board in May and June of 1987 indicated that his group had collected and analyzed more than 500 Reach 8 sediment samples during 1984. His testimony also indicated that the sediment PCB compositions restated in table 1 of his comment (1) did not come from these 500 samples as a whole, but instead represented the simple, unweighted average of the compositions of the ten sediment cores of the series described in table 3 of (2). None of these cores averaged more than 50 ppm PCB, and only two (both showing heavily altered patterns) averaged above 20 ppm. The median PCB level for the reported cores was only 3.8 ppm, and most of that occurred within a few centimeters of the surface. Clearly, the sample subpopulation selected for reporting consisted mainly of lightly contaminated surface deposits, sampled many years after the time of release. Such deposits would be expected to have lost much of their lower homolog content by means of aerobic biodegradation and elution into the water column, with consequent increases (which we

have repeatedly observed) in the proportion of more heavily chlorinated homologs in the residue. The contrast between this "cold spot" composition (restated in column 2 of our Table 1) and that of the Reach 8 hot spots (column 3) is striking. In the hot spots, where we have reported anaerobic microbial dechlorination to be occurring (3–5), there is a sharp increase in the proportions of mono- and dichlorobiphenyls, corresponding decreases in those of tri-, tetra-, and pentachlorobiphenyls, and sharp decreases also in the average numbers of chlorine atoms attached to meta and para positions on the biphenyl nuclei, while the number of ortho chlorines per biphenyl nucleus remains constant.

We did not state in our report (5) that the proportion of 24-24 CB in the PCBs of Pattern A/B sediments is stable. What table 1 of (5) actually reports is that, with increasing depth in such sediments the relative sizes of the 24-24, 24-25, and 25-25 CB peaks all first increase and then fall. Even the uppermost layers of Core 18-6, which exhibited only a modest (Pattern A+B) level of dechlorination (3), showed more 24-24 CB than did Aroclor 1242. Table 3 of (5) indicates that for one specific Pattern B sediment section, taken low in a core, the *net* changes (relative to Aroclor 1242) in the levels of 24-24, 24-25, and 25-25 CB, as a group were low. This resulted from the net effect of their formation by means of the dechlorination of the corresponding penta- and hexachlorobiphenyls (estimated rate data given) and their concurrent removal by dechlorination to trichlorobiphenyls. In short, the variations in 24-24, 24-25, and 25-25 CB levels were reported; they are consistent with the proposed stepwise dechlorination scheme; and hence they require no special ad hoc explanation in terms of selective adsorption by the sediments.

The original basis for the sedimentation hypothesis (2) was the observation that the unusual PCB composition seen in the Reach 8 hot spots was similar to that often seen in water samples collected from the same area. From this it was postulated that the PCBs were entering the sediments by means of adsorption and deposition from the water column. This conclusion might have been plausible if based upon observations made during the period 1955 through 1971, when significant PCB discharge and deposition was occurring. However, the observations were made during 1983 and 1984, long after the discharge had ceased, and when the PCB flux was entirely from the sediments to the water. Clearly, the reason why the PCB in the water then resembled that of the sediments was because that is where it was coming from.

We have even more serious objections to the argument that a sequence of two-phase transfer steps (partial elution of PCBs from bulk Aroclor 1242 into the water column followed by partial adsorption from the water onto sediment particles) could account for the huge changes in composition represented by upper Hudson hot spot patterns B, B', C, and E, and those of other aquatic sediments as well (4, 5). First, the distribution coefficients describing the behavior of many individual PCB congeners in both of the phase transfer steps have been determined and found to be closely correlated with each other (and those of other simple phase transfer processes) by means of linear free energy relations (6–8). Because of this, one must anticipate that the effects of the two-phase transfer processes on PCB composition would very nearly cancel each other out, so that no sizable net change in composition would be expected.

In addition, the sedimentation hypothesis advocated by M. P. Brown *et al.* does not explain the extensive but selective loss of nonortho chlorines, the formation of novel PCB congeners (4, 5), the net increases in the masses of lower congeners present (5), the variability in the alteration patterns seen (3–5), or the tendency of the alterations to be most extensive in the deeper and more heavily contaminated sediment layers. Carefully controlled laboratory experiments by Quensen, Tiedje, and Boyd have recently demonstrated the anaerobic dechlorination of PCBs in Reach 8 Hudson River sediments by the indigenous microbes (9). These experiments showed depletion of selected congeners having meta and para chlorines and corresponding increases in the predicted dechlorination products.

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REFERENCES

1. M. P. Brown, B. Bush, G. Y. Rhee, L. Shane, *Science* **240**, 1675 (1988).
2. B. Bush *et al.*, *Chemosphere* **16**, 833 (1987).
3. J. F. Brown, Jr., *et al.*, *Northeast. Environ. Sci.* **3**, 167 (1984).
4. J. F. Brown, Jr., *et al.*, *Science* **236**, 709 (1987).
5. J. F. Brown, Jr., *et al.*, *Environ. Toxicol. Chem.* **6**, 579 (1987).
6. S. W. Karickhoff, *Chemosphere* **10**, 833 (1981).
7. D. MacKay *et al.*, *Physical Behavior of PCBs in the Great Lakes*, D. MacKay, S. Paterson, S. J. Eisenreich, M. S. Simmons, Eds. (Ann Arbor Science, Ann Arbor, MI, 1983), chap. 4.
8. G. D. Vieth and P. Kosianin, *ibid.*, chap. 15.
9. J. F. Quensen III, J. M. Tiedje, S. A. Boyd, abstract (No. 416) of paper presented at the Eighth Annual Meeting of the Society of Environmental Toxicology and Chemistry, Pensacola, FL, 9 to 12 November 1987.

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