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Periphyton density influences organochlorine accumulation in rivers

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Abstract

To investigate the influence of eutrophication on organochlorine dynamics in lotic systems, we related polychlorinated biphenyl (PCB) concentrations in brown trout (*Salmo trutta*) to the periphyton density and total phosphorus concentrations (Tot-P) in 10 Swedish rivers. Tot-P concentrations in the water ranged from 37 ± 4 to 156 ± 51 μg L^{-1}. Periphyton density in the rivers during the experiment ranged from 6 to 199 mg carbon (C) m^{-2} and was not related to the Tot-P concentration. The concentrations of PCB 52, PCB 66/95, PCB 90/101, PCB 132/153/105, PCB 160/138/158, PCB 180, and PCB 194 in brown trout were positively related to both periphyton density and Tot-P concentration, with periphyton density having the highest degree of correlation, 40–58%, compared to 6–44% for Tot-P. All seven PCB concentrations in brown trout were significantly, positively related to the areal mass (ng m^{-2}) of the PCB in periphyton ($r^2 = 0.69$–0.86). Contrary to what has been found for lakes, eutrophication and organochlorine accumulations are positively related in rivers. Thus, an increase in primary production should increase the exposure of the local biota but lead to a decrease of the downstream transport of these pollutants to coastal areas and oceans.

The interactions between two major environmental issues, eutrophication of aquatic environments and organochlorine pollution, have received increased attention during the past decade (Larsson et al. 1992; Jeremiason et al. 1999; Dachs et al. 2000). These two environmental problems are both concerns in their own right, but because they tend to co-occur, interactions are important for predicting organochlorine transport and accumulation in aquatic systems. Changes in the primary production, the primary effect of eutrophication, may alter the behavior of organochlorines in the systems. Earlier, it was suggested that increased primary production in lakes had the effect of lowering the concentrations of organochlorines in lake biota (i.e., there is a negative relationship between trophic status and organochlorine concentrations) (Taylor et al. 1991; Berglund et al. 2001*a*). This relationship may lead to conflicting environmental effects because trying to remedy eutrophication by reducing nutrient loads to lakes may further increase organochlorine concentrations in already contaminated ecosystems. One proposed theory to explain the established negative relationship between eutrophication and organochlorines in lakes is that the increased sedimentation rate causes the sediment burial of organochlorine compounds while the primary producer biomass increases (Berglund et al. 2001*b*). The lipophilic organochlorines associate mainly with organic particles in the water. When algae die and start to settle to the bottom, organochlorines are withdrawn from the water column. Hence, the pelagic food webs receive less exposure in eutrophic lakes, where the organochlorines instead will accumulate in the benthic sediment (Berglund et al. 2001*b*).

The interactions between eutrophication and organochlo-rines in lotic environments (i.e., streams and rivers) have received less attention, even though rivers are often the main recipients of anthropogenic effluents, including the organochlorines and nutrients causing eutrophication. There are several fundamental hydrological and ecological differences between lake and river systems, differences that should influence how eutrophication affects the fate of organochlorines. In an earlier study of streams and rivers, we observed a positive relationship between the organochlorine concentrations in brown trout and the trophic status of the river, with the latter being measured as the Tot-P concentrations in the river water (Berglund et al. 1997). Thus, there seemed to be a positive relationship between trophic status and organochlorines in rivers, contrary to the situation in lakes, where a negative relationship has been observed. One difference between rivers and lakes is the unidirectional flow of water in the former versus the turbulent mixing in the latter. This will result in a spiraling of compounds in rivers, which combines cycling with retention and accumulation in the ecosystem (Essington and Carpenter 2000). The "spiraling theory" has been developed and tested on nutrients and C in rivers and should be valid for organochlorines as well (Newbold et al. 1982, 1983; Stewart et al. 1993). Another difference is that benthic ecosystems have a higher relative importance in rivers than in lakes. In streams and rivers, benthic periphyton are the main primary producers, whereas in lakes, pelagic phytoplankton typically are of more importance. The influence of terrestrial, allochthonous C sources from the watershed may also be more important in the benthic river ecosystems than in the pelagic lake ecosystems (Essington and Carpenter 2000); additionally, the watershed influence on organochlorine input can be expected to be higher in rivers than in lakes. Increased primary production of benthic organisms in rivers may thus affect organochlorine dynamics in a way that is different from increased primary production of pelagic phytoplankton in lakes.

The goal of this study was to investigate the influence of eutrophication (increased benthic primary production) on organochlorine dynamics in rivers by relating the accumulation

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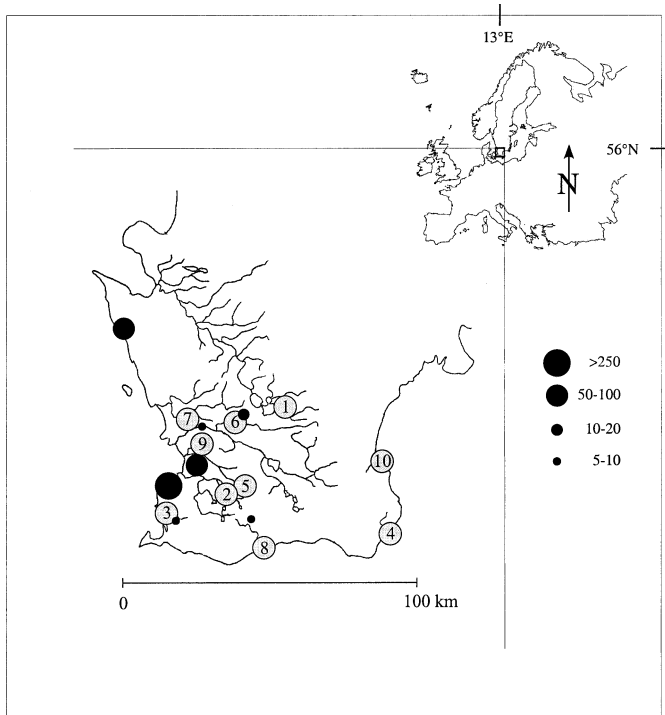
Fig. 1. Map of southern Sweden (13°E, 56°N) depicting the nine river systems and 10 sampling sites. Site 1: Rönne å, 2: Sege å, 3: Vällingebäcken, 4: Hobybäcken, 5: Höje å, 6: Bråån, 7: Saxån, 8: Dybäcksån, 9: Vallkärrabäcken, and 10: Mölleån. The major cities in the area and villages with >5,000 inhabitants in the watersheds of the sites are denoted by black circles (number of inhabitants × 10^3).

of organochlorines in brown trout to differences in the periphyton density.

Methods

Stream characteristics and sample collection—The experiment and sampling were conducted from July to October 1998. Ten different rivers in southern Sweden (13°E, 56°N) were selected for the experiment (Fig. 1). The rivers were chosen on a trophic gradient determined by the Tot-P concentrations in the water. The rivers are situated in a defined geographical area that receives similar amounts of organochlorines via atmospheric deposition with no known point sources (Berglund et al. 1997; Backe et al. 2000). Therefore, differences in organochlorine concentrations between the rivers are not caused by differences in input to the watersheds. The river stretches where the experiment was performed were similar in length (25 m), and riffle parts of the rivers were always included. At each site, stream width (wetted perimeter) was measured at four transects placed at evenly spaced locations along the stream bank. Stream velocity and depth were measured at five points (0, 1/4, 2/4, 3/4, and 4/4 of stream width) along each transect. Stream velocity was measured with a Nixon Streamflow 422 at 0.6 of total depth when the total depth was <0.8 m and at 0.2 and 0.8 of total depth in deeper sections. Light was measured at each of the transects as the percentage of light reaching the river

Table 1. Watershed area, land use in the watershed, morphological data, light penetration, total phosphorus (Tot-P) concentrations, and periphyton density for the studied rivers.

Site	Watershed area (km²)	Agriculture (%)	Rural (%)	Forest (%)	Mean width (m)	Mean depth (cm)	Mean current (cm s⁻¹)	Light penetration (%)	Tot-P (µg L⁻¹)	Periphyton density (mg C m⁻²)
Rönne å	15	5	5	90	2.5	10.3	6.3	19 ± 6	37 ± 4	6.2
Sege å	30	45	10	45	2.2	4.6	9.0	72 ± 10	50 ± 4	NM*
Vällingebäcken	17	60	40	0	1.4	7.4	15.8	98 ± 3	71 ± 26	199.4
Hobybäcken	26	95	5	0	1.3	10.9	26.5	26 ± 5	78 ± 19	127.9
Höje å	131	25	5	70	4.8	25.9	5.9	34 ± 17	86 ± 19	10.0
Bråån	150	80	10	10	5.2	7.6	27.2	38 ± 10	89 ± 6	32.2
Saxån	30	90	10	0	3.4	19.0	8.2	90 ± 2	100 ± 50	21.3
Dybäcksån	42	90	5	5	2.3	7.1	4.1	24 ± 31	101 ± 32	70.0
Vallkärrabäcken	10	60	40	0	2.3	11.4	5.4	37 ± 8	143 ± 10	16.8
Mölleån	25	40	10	50	4.4	13.3	10.4	16 ± 4	156 ± 51	30.4

* NM, not measured.

Table 2. Lipid content (% dry weight), organic carbon content (% of dry weight), and PCB concentrations (ng g organic carbon^{-1}) in periphyton from the studied rivers. Values are for one pooled sample from each river.

Site	Lipid content	Organic carbon content	PCB 52	PCB 66/95	PCB 90/101	PCB 132/153/105	PCB 160/138/158	PCB 180	PCB 194
Rönne å	1.21	11.7	0.72	0.90	0.97	1.22	1.39	1.24	0.18
Sege å	1.04	21.1	1.19	0.74	0.87	0.90	0.83	0.51	0.08
Vällingebäcken	0.70	17.6	1.00	1.43	1.41	1.42	1.51	0.74	0.11
Hobybäcken	1.08	9.3	0.27	0.22	0.27	0.77	0.88	0.42	0.08
Höje å	1.22	22.5	1.04	1.39	0.69	1.77	1.95	1.55	0.67
Bråån	1.29	12.2	1.91	3.80	3.50	7.09	8.42	6.28	1.32
Saxån	0.58	6.1	0.42	0.83	0.79	1.46	1.90	1.52	0.29
Dybäcksån	0.37	7.2	0.19	0.24	0.31	0.55	0.63	0.33	0.07
Vallkärrabäcken	1.04	10.6	0.95	1.74	1.42	2.94	3.31	2.11	0.71
Mölleån	0.77	13.2	0.70	1.41	1.41	3.37	3.98	3.03	0.38

surface to the total light measured adjacent to the river with no foliage cover. Means of stream width, depth, velocity, and light are presented in Table 1.

Periphyton was sampled on ceramic tiles. Twenty 20- by 20-cm tiles were placed in each of the 10 rivers. The tiles were left for colonization for 58–67 d from July to October. At the end of the experiment, the periphyton were scraped from the tiles with a brush into one precleaned container, giving one pooled sample for each river. The periphyton density was calculated as grams of organic C per square meter (g C m^{-2}). Organic C content was determined for periphyton with a LECO CR-12 carbon analyzer (Merry and Spounser 1988). One site was not included in the analysis of periphyton density because the tiles were destroyed during the experiment. At this site, periphyton for organochlorine analysis were sampled by scraping stones from the river. After periphyton sampling, two to three individuals of young-of-the-year (YOY) brown trout were sampled with electrofishing. All samples were frozen and then freeze-dried before analysis. Water samples for Tot-P analysis were taken three different times during the experiment (beginning, middle, and end). Tot-P concentrations were analyzed with standard methods (digestion with peroxodisulfate) at the Department of Ecology, Lund University.

Sample preparation and analysis—The methods for sample preparation and PCB analysis are described in detail elsewhere (Bremle et al. 1995). Briefly, PCBs in periphyton and whole brown trout samples were extracted in a modified Soxhlet apparatus with a mixture of acetone and *n*-hexane. The lipid content of periphyton and whole brown trout was determined gravimetrically. Octachloronaphthalene (OCN) and pentachlorobenzene were used as the internal standard and chromatographic standard, respectively. Concentrated extracts were cleaned and fractionated on acid/basic double-layer silicagel columns. Samples were analyzed for PCBs by capillary gas chromatography using an electron capture detector (Varian 3400 cx) with a split/splitless injector, a 25-m DB5 quartz capillary column (i.d. = 0.25 mm). PCB components were identified according to Mullin et al. (1984) and Schultz et al. (1989). A total of 65 peaks of PCB congeners were identified and quantified in the periphyton and brown trout samples. Domains 19 (IUPAC 52), 32 (66/95), 38 (90/101), D54 (132/153/105), D58 (160/138/158), D72 (180), and D84 (194) were used for further statistical analysis, as they were present in all of the samples in high concentrations and represent a wide range in lipophilicity (log K_{ow} = 5.8–7.8) (Hawker and Connell 1988).

Quality control—The analytical performance was regularly controlled with pesticide standards and PCB standards (Aroclor 1242 and Clophen A60). Extraction efficiency of the internal standard OCN was 117 ± 14%. Samples were not corrected for recovery. A procedural blank was run in parallel with every batch of eight tissue samples. Mean blank levels were 0–12% of the mean PCB levels in periphyton samples (PCB 52: 12%, PCB 66/95: 4%, PCB 90/101: 2%, PCB 132/153/105: 5%, PCB 160/138/158: 6%, PCB180: 3%, and PCB 194: 0%) and 0–3% of the PCB levels in mean brown trout (PCB 52: 3%, PCB 66/95: 0.8%, PCB 90/101: 0.2%, PCB 132/153/105: 0.8%, PCB 160/138/158: 1%, PCB180: 0.8%, and PCB 194: 0%). No blank corrections of the samples were made.

Statistical analysis—All concentrations and density data were log transformed before statistical analysis. Periphyton PCB concentrations were calculated as nanograms of PCB per gram of organic C (ng PCB g organic C^{-1}), and brown trout PCB concentrations were calculated as nanograms of PCB per gram of lipid (ng PCB g lipid^{-1}). For regression

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Fig. 2. Regressions between mean log Tot-P concentrations (μg L^{-1}) in the water and mean log PCB concentrations in brown trout, *S. trutta* (ng g lipid^{-1}) (left panels), and between log periphyton density (g C m^{-2}) and mean log PCB concentrations in brown trout (ng g lipid^{-1}) (right panels) in the investigated rivers. The lipophilicity (K_{ow}) of the PCBs increase from top to bottom in the figure (log K_{ow} for PCB 52: 5.8, PCB 66/95: 6.1, PCB 90/101: 6.4, PCB 132/153/105: 6.8, PCB 160/138/158: 6.8, PCB 180: 7.4, and PCB 194: 7.8 [from Hawker and Connell 1988]).

Olof Berglund Figure 2

log PCB concentration in brown trout (ng g lipid⁻¹)

Left column (vs. log total phosphorous):
- PCB #52: $y = 0.5x + 0.4$; $r^2 = 0.06$; $p = 0.51$
- PCB #66/95: $y = 0.8x + 0.2$; $r^2 = 0.11$; $p = 0.36$
- PCB #90/101: $y = 0.8x + 0.4$; $r^2 = 0.13$; $p = 0.30$
- PCB #132/153/105: $y = 0.9x + 0.5$; $r^2 = 0.26$; $p = 0.13$
- PCB #160/138/158: $y = 1.0x + 0.3$; $r^2 = 0.26$; $p = 0.13$
- PCB #180: $y = 1.1x - 0.1$; $r^2 = 0.29$; $p = 0.11$
- PCB #194: $y = 1.5x - 1.8$; $r^2 = 0.44$; $p = 0.036$

Right column (vs. log periphyton density):
- PCB #52: $y = 0.5x + 1.4$; $r^2 = 0.43$; $p = 0.057$
- PCB #66/95: $y = 0.5x + 1.6$; $r^2 = 0.40$; $p = 0.068$
- PCB #90/101: $y = 0.5x + 1.8$; $r^2 = 0.45$; $p = 0.047$
- PCB #132/153/105: $y = 0.5x + 2.2$; $r^2 = 0.54$; $p = 0.023$
- PCB #160/138/158: $y = 0.5x + 2.2$; $r^2 = 0.51$; $p = 0.031$
- PCB #180: $y = 0.5x + 1.9$; $r^2 = 0.46$; $p = 0.043$
- PCB #194: $y = 0.6x + 1.0$; $r^2 = 0.58$; $p = 0.018$

log total phosphorous (µg L⁻¹)   log periphyton density (g C m⁻²)

Table 3. Mean weight $\pm$ standard deviation (g dry weight), lipid content $\pm$ standard deviation (% dry weight), and PCB concentrations (range) (ng g lipid^{-1}) in brown trout (*Salmo trutta*) from the studied rivers. $n = 3$ except in Höje å and Bråån, where $n = 2$.

Site	Weight	Lipid content	PCB 52	PCB 66/95	PCB 90/101	PCB 132/153/105	PCB 160/138/158	PCB 180	PCB 194
Rönne å	0.66±0.12	8.7 ± 0.6	1.2 (1.0–1.5)	1.5 (1.3–1.7)	2.6 (2.3–2.8)	7.1 (6.6–7.5)	6.5 (5.8–6.9)	3.1 (2.8–3.3)	0.2 (0.2–0.3)
Sege å	1.85±0.08	14.4 ± 2.5	1.0 (1.0–1.1)	2.0 (1.4–2.6)	3.1 (3.0–3.2)	8.0 (7.9–8.2)	7.2 (6.4–7.7)	3.3 (3.1–3.4)	0.3 (0.3–0.3)
Vällingebäcken	2.00±0.07	10.5 ± 0.9	21.5 (17.6–24.3)	40.4 (32.7–45.2)	48.8 (40.0–55.4)	66.7 (59.8–70.6)	80.9 (70.7–87.4)	31.5 (29.4–33.0)	4.2 (3.9–4.4)
Hobybäcken	0.70±0.06	9.7±2.2	2.6 (2.2–3.4)	4.3 (3.4–5.8)	7.4 (5.2–12.8)	23.9 (20.4–29.0)	23.4 (20.2–28.9)	10.3 (9.3–12.3)	2.0 (1.7–2.3)
Höje å	2.40±0.50	14.5 ± 3.9	1.9 (1.8–1.90)	2.7 (2.4–3.0)	3.4 (3.2–3.7)	9.2 (8.4–10.1)	8.2 (7.5–8.8)	3.8 (3.5–4.2)	0.6 (0.5–0.7)
Bråån	3.67±0.14	15.5 ± 1.1	7.4 (6.8–8.0)	18.3 (17.9–18.7)	22.9 (22.8–23.1)	52.9 (52.0–53.7)	57.2 (56.8–57.5)	32.8 (31.5–34.1)	3.3 (3.0–3.5)
Saxån	0.83±0.09	8.5 ± 2.2	3.0 (2.3–4.7)	6.6 (5.2–8.5)	5.8 (4.7–6.9)	14.7 (11.6–20.3)	15.0 (11.8–20.4)	7.4 (5.8–9.6)	1.4 (1.2–1.7)
Dybäcksån	0.55±0.06	7.9 ± 1.2	2.1 (1.4–3.6)	2.7 (2.0–4.6)	3.7 (3.1–5.0)	12.7 (12.0–13.2)	12.6 (11.9–13.2)	6.2 (5.9–6.7)	1.1 (1.0–1.3)
Vallkärra-bäcken	1.60±0.28	12.9 ± 0.1	6.6 (6.2–6.9)	12.7 (9.8–19.6)	17.6 (16.1–18.8)	35.0 (32.5–37.8)	42.1 (38.8–45.2)	16.7 (14.7–17.9)	3.1 (2.8–3.3)
Mölleån	0.53±0.14	10.4 ± 2.9	1.3 (1.1–1.8)	3.2 (1.8–5.2)	7.7 (5.3–11.9)	32.5 (22.6–49.2)	33.4 (21.9–55.3)	18.0 (12.3–27.8)	2.1 (1.5-3.5)

analysis, means of the three Tot-P samples and means of the two or three brown trout PCB samples from each river were used. Simple regression analyses were performed using the StatView (SAS Institute) package.

Results

The mean Tot-P concentrations in the 10 streams ranged from 37 to 156 μg L^{-1}. The concentrations measured during the experiment were similar to the yearly means for six of the investigated rivers for which we could obtain data (simple regression: log mean Tot-P = 0.6 log yearly mean + 0.7, $r^2 = 0.78$, $P < 0.05$). The periphyton density in the nine streams ranged from 6 to 200 mg C m^{-2}. The periphyton density was not related to the Tot-P concentrations in the streams or to the light penetration or stream velocity (simple regressions, $P > 0.05$). The periphyton density was related to land use in the watershed, and the density decreased with the increasing proportion of forest in the watershed (simple regression, $r^2 = 0.47$, $P < 0.05$). The Tot-P concentrations were not related to land use in the watershed (simple regression, $P > 0.1$).

The organic C content in periphyton varied among the streams from 6.1% to 22.5% (Table 2). The organic C content was not related to the periphyton density or Tot-P concentration in the streams, nor was it related to the land use in the watershed.

Mean PCB concentrations in brown trout are shown in Table 3. Mean concentrations ranged from 0.2 to 80.9 ng g lipid^{-1}, with PCB 132/153/105 and PCB 160/138/158 having the highest concentrations.

PCB concentrations in brown trout were positively related to the Tot-P concentrations in the rivers but only significantly so for the most lipophilic PCB 194 (simple regressions, Fig. 2). The slope and predictability increased with increasing lipophilicity (increasing octanol–water partitioning coefficient, K_{ow}) of the PCBs (simple regression, $r^2 = 0.92$, $P < 0.01$, and $r^2 = 0.96$, $P < 0.01$, respectively). PCB concentrations in brown trout were positively related to the periphyton density in the rivers (simple regression, Fig. 2).

The PCB concentrations in periphyton are shown in Table 2. The concentrations ranged from 0.07 to 8.4 ng g organic C^{-1}; also, the PCBs of intermediate K_{ow} (PCB 132/153/105 and PCB160/138/158) were found in the highest concentrations. As for brown trout, the PCB concentrations in periphyton were positively related to the Tot-P concentrations in the streams but were not statistically significant (Fig. 3). The slope (k) and predictability (r^2) of the regressions increased with increasing K_{ow} of the PCBs (simple regression, $r^2 = 0.72$, $P < 0.05$, and $r^2 = 0.61$, $P = 0.05$, respectively). The PCB concentrations in periphyton were negatively related to periphyton density, but no statistical significance was found (Fig. 3).

The PCB concentrations in brown trout were positively

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Fig. 3. Regressions between mean log Tot-P concentrations (μg L^{-1}) in the water and log PCB concentrations in periphyton (ng g organic C^{-1}) (left panels) and between log periphyton density (g C m^{-2}) and log PCB concentrations in periphyton (ng g organic C^{-1}) (right panels) in the investigated rivers. The lipophilicity (K_{ow}) of the PCBs increase from top to bottom in the figure (log K_{ow} for PCB 52: 5.8, PCB 66/95: 6.1, PCB 90/101: 6.4, PCB 132/153/105: 6.8, PCB 160/138/158: 6.8, PCB 180: 7.4, and PCB 194: 7.8 [from Hawker and Connell 1988]).
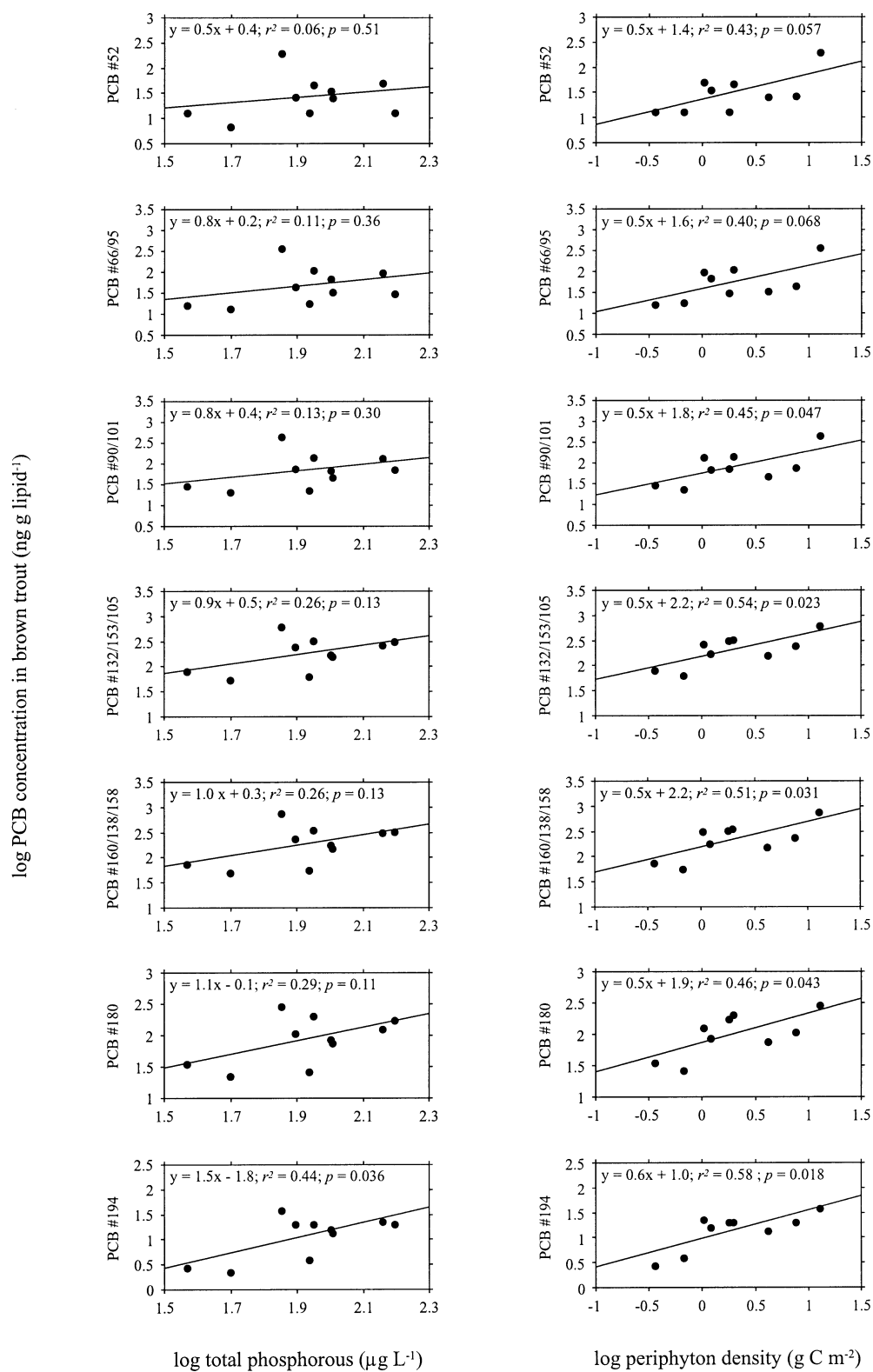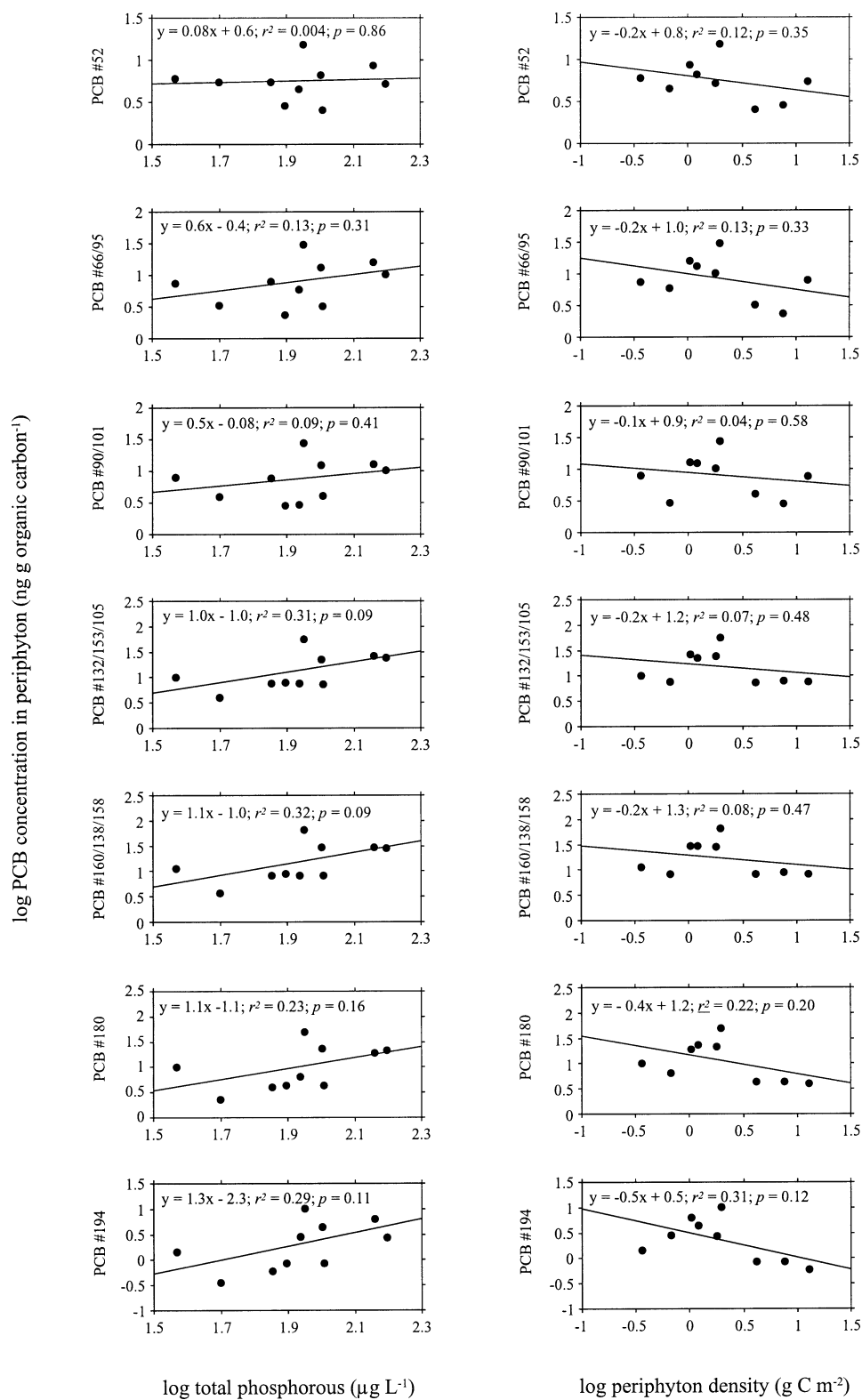
Olof Berglund Figure 3

log PCB concentration in periphyton (ng g organic carbon^{-1})

PCB #52 $y = 0.08x + 0.6$; $r^2 = 0.004$; $p = 0.86$

PCB #66/95 $y = 0.6x - 0.4$; $r^2 = 0.13$; $p = 0.31$

PCB #90/101 $y = 0.5x - 0.08$; $r^2 = 0.09$; $p = 0.41$

PCB #132/153/105 $y = 1.0x - 1.0$; $r^2 = 0.31$; $p = 0.09$

PCB #160/138/158 $y = 1.1x - 1.0$; $r^2 = 0.32$; $p = 0.09$

PCB #180 $y = 1.1x - 1.1$; $r^2 = 0.23$; $p = 0.16$

PCB #194 $y = 1.3x - 2.3$; $r^2 = 0.29$; $p = 0.11$

PCB #52 $y = -0.2x + 0.8$; $r^2 = 0.12$; $p = 0.35$

PCB #66/95 $y = -0.2x + 1.0$; $r^2 = 0.13$; $p = 0.33$

PCB #90/101 $y = -0.1x + 0.9$; $r^2 = 0.04$; $p = 0.58$

PCB #132/153/105 $y = -0.2x + 1.2$; $r^2 = 0.07$; $p = 0.48$

PCB #160/138/158 $y = -0.2x + 1.3$; $r^2 = 0.08$; $p = 0.47$

PCB #180 $y = -0.4x + 1.2$; $r^2 = 0.22$; $p = 0.20$

PCB #194 $y = -0.5x + 0.5$; $r^2 = 0.31$; $p = 0.12$

log total phosphorous (μg L^{-1})

log periphyton density (g C m^{-2})

Olof Berglund Figure 4

log PCB concentration in brown trout (ng g lipid^{-1})

Left column (PCB #52 through PCB #194):

PCB #52: $y = 0.4x + 1.1$; $r^2 = 0.06$; $p = 0.50$

PCB #66/95: $y = 0.6x + 1.1$; $r^2 = 0.23$; $p = 0.16$

PCB #90/101: $y = 0.7x + 1.3$; $r^2 = 0.27$; $p = 0.13$

PCB #132/153/105: $y = 0.5x + 1.7$; $r^2 = 0.26$; $p = 0.13$

PCB #160/138/158: $y = 0.5x + 1.7$; $r^2 = 0.27$; $p = 0.13$

PCB #180: $y = 0.4x + 1.6$; $r^2 = 0.19$; $p = 0.20$

PCB #194: $y = 0.3x + 1.0$; $r^2 = 0.09$; $p = 0.39$

Right column (PCB #52 through PCB #194):

PCB #52: $y = 0.7x + 0.8$; $r^2 = 0.69$; $p = 0.006$

PCB #66/95: $y = 0.8x + 0.8$; $r^2 = 0.83$; $p = 0.0007$

PCB #90/101: $y = 0.7x + 1.1$; $r^2 = 0.82$; $p = 0.0008$

PCB #132/153/105: $y = 0.6x + 1.5$; $r^2 = 0.85$; $p = 0.0004$

PCB #160/138/158: $y = 0.6x + 1.4$; $r^2 = 0.81$; $p = 0.001$

PCB #180: $y = 0.7x + 1.1$; $r^2 = 0.86$; $p = 0.0003$

PCB #194: $y = 0.7x + 0.7$; $r^2 = 0.74$; $p = 0.0028$

log PCB concentration in periphyton
(ng g organic carbon^{-1})

log areal mass of PCB in
periphyton (ng m^{-2})

related to the PCB concentrations in periphyton in the stream but were not statistically significant (Fig. 4). The PCB concentrations in brown trout were positively related to the areal mass of PCB in periphyton (ng PCB m^{-2}) (simple regression, $r^2 = 0.69$–0.86, $P < 0.01$, Fig. 4).

Discussion

Periphyton density—The periphyton density was not related to the Tot-P concentrations in the water. Primary production in rivers is not limited by nutrients to the same extent as in lakes because of the continuous flow and concomitant supply of nutrients (Kjeldsen 1996). The mean Tot-P concentrations in the investigated rivers were between 37 and 156 μg L^{-1}, and in this concentration range, the systems are probably not phosphorus limited (Chételat et al. 1999). Grazers were not excluded in the periphyton density measurements; therefore, these measurements reflected the realized density rather than the actual production. For the purpose of this study, a measure of the realized periphyton density was appropriate, as it is the realized periphyton density that should influence organochlorine dynamics. Grazer presence and density influence periphyton production and density, and grazers may obscure a relationship between Tot-P concentration and actual periphyton production (Rosemond et al. 1993). Light conditions and stream velocity are believed to limit periphyton production to a large extent in rivers (Rosemond et al. 2000). However, no relationships were found between light or stream velocity and periphyton density in this study. The mean Tot-P concentrations in the streams were not related to land use in the watershed, but periphyton density was weakly related to the proportion of forest/agricultural and urban land. Generally, phosphorus concentrations in rivers and land use in the watershed correlate well, and in an earlier study in this region of southern Sweden where we surveyed 53 streams and rivers, we found a positive relationship between Tot-P and proportion of agricultural land in the watershed (Berglund et al. 1997). However, as phosphorus may not exert a primary limitation on primary production in rivers and grazer presence may counteract a relationship between actual periphyton production and realized periphyton density, there was no coupling between phosphorus concentrations and periphyton density in the streams.

Trophic status and organochlorines—YOY brown trout were chosen, as they represent a time-integrated sample of the PCB exposure during the previous few months. YOY brown trout are stationary during their first year in the river and, consequently, reflect the PCB exposure at the specific sites where the other variables were measured (Elliott 1984). By choosing brown trout of the same age, differences in

PCB accumulation caused by differences in size, age, and food choice (trophic position) were also minimized (Olsson et al. 2000). The PCB concentrations in brown trout were similar to those found in previous studies of this region; the range of PCB concentrations was about one order of magnitude, indicating that there were no point sources of PCB to the streams (Berglund et al. 1997).

The PCB concentrations in brown trout were positively related to the Tot-P concentrations in the rivers (Fig. 2). This is supported by an earlier study, which demonstrated a positive correlation between organochlorines in brown trout and Tot-P concentrations, using data from 53 different streams and rivers in Sweden, that included the 10 rivers in the present study (Berglund et al. 1997). In the previous study, where periphyton density was not measured, we suggested three possible processes responsible for the correlation: (1) phosphorus and organochlorines have similar transport pathways to the rivers; (2) an increased retention of organochlorines and a decreased spiraling length caused by an increased periphyton biomass increase the probability of uptake according to the spiraling theory (Newbold et al. 1982, 1983; Stewart et al. 1993); and (3) rivers shift from heterotrophy to autotrophy with increasing trophic status, thereby shifting from a terrestrial C source with low organochlorine concentrations to an aquatic one with higher concentrations, which will then be biomagnified in the food chain (Peterson et al. 1985).

The PCB concentrations in brown trout were positively related to the periphyton density in the rivers. The predictability was higher than for Tot-P, and slopes were independent of K_{ow}. This supports the notion that the increased uptake of organochlorines was caused by decreased spiraling length. The spiraling length is defined as the distance (in meters) an atom or molecule travels downstream as it completes one cycle from dissolved phase, through one or more compartments in the river, back to dissolved phase (Newbold et al. 1983). The shorter the spiraling length, the higher the probability of uptake in the biota. Mainly, stream velocity and retentiveness will determine the spiraling length (Newbold et al. 1983). The retentiveness of the lotic system increases with increased periphyton density. Periphyton constitutes a large immobile surface area with which the lipophilic organochlorines can associate, decreasing their downward transport, and therefore increases the probability of uptake in biota. Hence, while an increased primary production in lakes appears to decrease the accumulation of organochlorines in pelagic biota by withdrawing the compounds from the water column, an increased primary production in rivers will increase the accumulation of organochlorines in biota by retarding the downstream transport (Essington and Carpenter 2000; Berglund et al. 2001*a,b*). No significant relationship was found between the other main

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Fig. 4. Regressions between log PCB concentrations in periphyton (ng g organic C^{-1}) and mean log PCB concentrations in brown trout, *S. trutta* (ng g lipid^{-1}) (left panels), and between log areal mass of PCB in periphyton (ng m^{-2}) and mean log PCB concentrations in brown trout (ng g lipid^{-1}) (right panels) in the investigated rivers. The lipophilicity (K_{ow}) of the PCBs increase from top to bottom in the figure (log K_{ow} for PCB 52: 5.8, PCB 66/95: 6.1, PCB 90/101: 6.4, PCB 132/153/105: 6.8, PCB 160/138/158: 6.8, PCB 180: 7.4, and PCB 194: 7.8 [from Hawker and Connell 1988]).
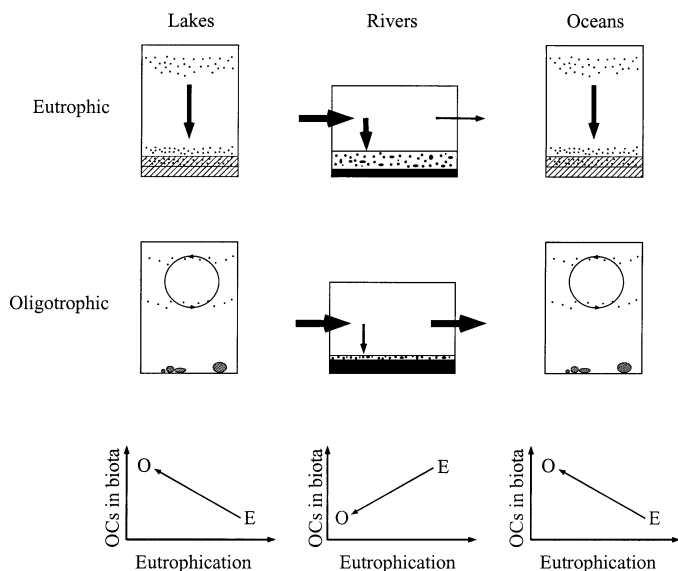
Fig. 5. Conceptual model of the fate of organochlorines in lakes, rivers, and oceans (coastal areas and estuaries) when the systems go from eutrophic to oligotrophic conditions and the effects on concentrations in biota. Under eutrophic conditions, the concentrations in pelagic lake biota will be low. The proposed explanatory mechanisms are sedimentation and burial and/or biodilution due to the large biomass of phytoplankton (Taylor et al. 1991; Dachs et al. 2000; Berglund et al. 2001*b*). In rivers, concentrations in biota will be high under eutrophic conditions because of the retention in the periphyton biomass analogous with the "spiraling theory" (Berglund et al. 1997; this study), and the transport to coastal areas and estuaries will be low. In coastal areas and estuaries, the concentrations in biota will be low because of factors similar to those in the lakes. When the systems become oligotrophic, the concentrations in pelagic lake biota will increase because of decreased sedimentation and burial and less biodilution in the smaller phytoplankton biomass. Also, changes in species composition of the phytoplankton community may increase concentrations (Berglund et al. 2001*a*). In rivers, concentrations will decrease in biota as the retentiveness of the system decreases with decreased periphyton biomass, and the transport to coastal areas and estuaries will increase. In oceans, concentrations will increase for reasons that are similar to those seen for the increases in lakes; also, the transport of organochlorines to the oceans, via lakes and rivers, will increase as the sinks of lake sediments and river periphyton and biota are reduced in magnitude.

determinant of spiraling length, stream velocity, and the PCB concentrations in brown trout. However, the range in stream velocity was low, as the rivers were chosen to differ in trophic status but to be similar in other characteristics.

Because the Tot-P concentrations and periphyton density varied independently of each other while the PCB concentrations in brown trout were related to both factors, the results suggest that two different processes influence the organochlorine accumulation in lotic environments.

First, transport pathways in the watershed determine the amount of organochlorines reaching the rivers after being deposited via the atmosphere. There should be similarities in the behavior of phosphorus and organochlorines, because both substances are associated with organic C in waters, are readily taken up by the biota in aquatic systems (albeit through different routes and mechanisms), and are highly

adsorptive (Sharpley et al. 1981; Eisenreich 1987). Hence, it is probable that transport processes and pathways from watershed areas to rivers are the same or similar for both types of compounds. Thus, their concentration should covary in time and space. In watersheds dominated by agricultural land, overland flow is a more common pathway than in watersheds, where the forest dominates (Allan 1995). Overland flow delivers higher amounts of phosphorus and organochlorines to the rivers than the subsurface flow dominating the forested watersheds (Sharpley and Syers 1979). High-chlorinated compounds with high K_{ow} have a higher affinity for adsorption to particles transported with overland flow relative to low-chlorinated compounds (Granier et al. 1990). As phosphorus also is associated with particles in the water, the relationship between phosphorus and organochlorines can be expected to be stronger for the highly chlorinated ones. This is supported by the fact that the slope and predictability of the regressions between Tot-P and PCBs in periphyton and brown trout increased with increasing K_{ow} of the PCBs. This may suggest an adsorption process during the transport of the PCBs rather than an absorption or uptake process, which typically is characterized by a "bell-shaped" relationship with the K_{ow} of the compounds (Bremle and Ewald 1995).

Second, once organochlorines have entered the rivers, the accumulation of these compounds in the river biota is influenced by the retentiveness of the system in accordance with the spiraling theory. A high periphyton biomass, with which the lipophilic organochlorines can associate, will increase the retention of these compounds and decrease their spiraling length. This will increase the probability that they will be taken up in river biota either directly from water or indirectly via food and, thus, decrease the downward transport of organochlorines. In aquatic environments, organochlorines are taken up by organisms either via water or food. The relative importance of the two uptake routes may vary depending on the organism and properties of the compounds. Generally, in larger organisms occupying higher trophic levels such as trout, the route via food is considered the most important, especially for compounds with low solubility with log K_{ow}s >6.5. An increased retention of PCBs in the river will increase the uptake of PCBs in trout both via partitioning with dissolved PCBs in the water and, perhaps more importantly, given the low solubility and exchange rate of the compounds, via trophic transfer from periphyton to invertebrates to trout. The net result of both processes—transport to the rivers and retention in the periphyton biomass—will determine the exposure and accumulation of organochlorines to biota in higher trophic levels. The total exposure of organochlorines resulting from the two processes may be quantified with the mass of organochlorines per unit area in the rivers. The concentrations of all seven investigated PCBs in brown trout were positively related to the areal mass of the PCBs in periphyton, explaining 69–86% of the total variation in brown trout PCB concentrations (Fig. 4).

An alternative explanation is that phosphorus, periphyton density, and PCB concentrations all covary in response to the same anthropogenic impacts. If this were the case, the variation in concentrations of PCB in brown trout among the streams would reflect different loadings to the streams of

both phosphorus and PCBs rather than in-stream processes such as spiraling and retention in the periphyton biomass. However, mean Tot-P concentrations and periphyton density were not related in the streams in this study, and the PCB concentrations in brown trout were only weakly related to the PCB concentrations in periphyton ($r^2 = 0.06$–0.27). The concentrations in periphyton should reflect the input and water concentration of PCB, as PCB will passively partition between water and periphyton according to the equilibrium partitioning theory. PCB concentrations in brown trout were significantly related to periphyton density in the streams ($r^2 = 0.40$–0.58), and when both the periphyton PCB concentrations and the density of periphyton were included and the brown trout concentrations were related to the areal mass of PCB in periphyton (ng m^{-2}), the relationship was improved ($r^2 = 0.69$–0.86). The above results, together with the fact that the land use in the watershed, mean Tot-P concentrations, and periphyton density did not all covary, suggest that although the inputs of PCB to the rivers from the watershed vary and affect PCB concentrations in brown trout to some degree in this study, the influence of in-stream processes such as spiraling and periphyton growth was of significant importance.

The source of PCBs for the region surrounding the rivers examined in this study is mainly the atmosphere; therefore, the watershed influence is high. In lotic systems that are direct recipients of organochlorines from point sources, primary production is likely even more important in determining the organochlorine exposure of river biota and in determining the downward transport of organochlorines. These findings may therefore have implications for risk assessments concerning sensitive species in recipient rivers and for modeling and predictions of downstream transport and loading of organochlorines to estuaries, coastal areas, and oceans. The results from this study demonstrate that the relationship between eutrophication and organochlorines is different between lakes and rivers. The effects of reducing nutrient loads to waters to remedy eutrophication would therefore have different effects on the organochlorine concentrations in lake and river biota. One would predict an increase in organochlorine concentrations in pelagic lake biota and a decrease in concentrations in river biota. However, the effect on biota in estuaries and coastal areas would also be an increase in organochlorine concentrations. The reasons may be twofold; first, the reduction of nutrients would affect organochlorine behavior in a manner similar to lakes, causing increased concentrations in pelagic biota. Second, the effects of reduced eutrophication of lakes and rivers will cause a reduced retention of organochlorines in these systems, mainly in the sediments in lakes and in the periphyton and biota in rivers. The elimination of these sinks would increase the loading of organochlorines from the watersheds via lakes and rivers into the estuaries and coastal areas, where biota would be exposed to the higher loads of organochlorines (Fig. 5).

References

ALLAN, J. D. 1995. Stream ecology. Chapman and Hall.

BACKE, C., P. LARSSON, AND L. OKLA. 2000. Polychlorinated biphenyls in the air of southern Sweden—spatial and temporal variation. Atmos. Environ. **34:** 1481–1486.

BERGLUND, O., P. LARSSON, C. BRÖNMARK, L. GREENBERG, A. EKLÖV, AND L. OKLA. 1997. Factors influencing organochlorine uptake in age-0 brown trout (*Salmo trutta*) in lotic environments. Can. J. Fish. Aquat. Sci. **54:** 2767–2774.

———, ———, G. EWALD, AND L. OKLA. 2001*a.* The effect of lake trophy on lipid content and PCB concentrations in planktonic food webs. Ecology **82:** 1078–1088.

———, ———, ———, AND ———. 2001*b.* Influence of trophic status on PCB distribution in lake sediments and biota. Environ. Poll. **113:** 199–210.

BREMLE, G., AND G. EWALD. 1995. Bioconcentration of polychlorinated biphenyls (PCBs) in chironomid larvae, oligochaete worms and fish from contaminated lake sediment. Mar. Freshwater Res. **46:** 267–273.

———, L. OKLA, AND P. LARSSON. 1995. Uptake of PCBs in fish in a contaminated river system: Bioconcentration factors measured in the field. Environ. Sci. Technol. **29:** 2010–2015.

CHÉTELAT, J., F. R. PICK, A. MORIN, AND P. B. HAMILTON. 1999. Periphyton biomass and community composition in rivers of different nutrient status. Can. J. Fish. Aquat. Sci. **56:** 560–569.

DACHS, J., S. J. EISENREICH, AND R. M. HOFF. 2000. Influence of eutrophication on air–water exchange, vertical fluxes, and phytoplankton concentrations of persistent organic pollutants. Environ. Sci. Technol. **34:** 1095–1102.

EISENREICH, S. J. 1987. The chemical limnology of nonpolar organic contaminants: Polychlorinated biphenyls in Lake Superior, p. 393–469. *In* R. A. Hites and S. J. Eisenreich [eds.], Sources and fates of organic contaminants. Advances in Chemistry Series 216. American Chemical Society.

ELLIOTT, J. M. 1984. Numerical changes and population regulation in young migratory trout *Salmo trutta* in a Lake District stream, 1966–83. J. Anim. Ecol. **53:** 327–350.

ESSINGTON, T. E., AND S. R. CARPENTER. 2000. Nutrient cycling in lakes and streams: Insights from comparative analysis. Ecosystems **3:** 131–143.

GRANIER, L., M. CHEVREUIL, A.-M. CARRU, AND R. LETOLLE. 1990. Urban runoff pollution by organochlorines (polychlorinated biphenyls and lindane) and heavy metals (lead, zinc and chromium). Chemosphere **21:** 1101–1107.

HAWKER, D. W., AND D. W. CONNELL. 1988. Octanol–water partitioning coefficients of polychlorinated biphenyl congeners. Environ. Sci. Technol. **22:** 382–387.

JEREMIASON, J. D., S. J. EISENREICH, M. J. PATERSON, K. G. BEATY, R. HECKY, AND J. J. ELSER. 1999. Biogeochemical cycling of PCBs in lakes of variable trophic status: A paired-lake experiment. Limnol. Oceanogr. **44:** 889–902.

KJELDSEN, K. 1996. Regulation of algal biomass in a small woodland stream: Field experiments on the role of invertebrate grazing, phosphorus and irradiance. Freshwater Biol. **36:** 535–546.

LARSSON, P., L. COLLVIN, L. OKLA, AND G. MEYER. 1992. Lake productivity and water chemistry as governors of the uptake of persistent pollutants in fish. Environ. Sci. Technol. **26:** 346–352.

MERRY, R. H., AND L. R. SPOUNSER. 1988. The measurements of carbon in soils using a microprocessor-controlled resistance furnace. Commun. Soil Sci. Plant. Anal. **19:** 707–720.

MULLIN, M. D., C. M. POCHINI, S. MCCRIDLE, M. ROMKES, S. H. SAFE, AND L. M. SAFE. 1984. High-resolution PCB analysis: Synthesis and chromatographic properties of all 209 congeners. Environ. Sci. Technol. **18:** 468–476.

NEWBOLD, J. D., J. W. ELWOOD, R. V. O'NEILL, AND A. L. SHELDON. 1983. Phosphorus dynamics in a woodland stream ecosystem: A study of nutrient spiralling. Ecology **64:** 1249–1265.

———, P. J. MULHOLLAND, J. W. ELWOOD, AND R. V. O'NEILL.

1982. Organic carbon spiralling in stream ecosystems. Oikos **38:** 266–272.

OLSSON, A., K. VALTERS, AND S. BURREAU. 2000. Concentrations of organochlorine substances in relation to fish size and trophic position: A study on perch (*Perca fluviatilis* L.). Environ. Sci. Technol. **34:** 4878–4886.

PETERSON, B. J., AND OTHERS. 1985. Transformation of a tundra river from heterotrophy to autotrophy by addition of phosphorus. Science **229:** 1383–1386.

ROSEMOND, A. D., P. J. MULHOLLAND, AND S. H. BRAWLEY. 2000. Seasonally shifting limitation of stream periphyton: Response of algal populations and assemblage biomass and productivity to variation in light, nutrients, and herbivores. Can. J. Fish. Aquat. Sci. **57:** 66–75.

———, ———, AND J. W. ELWOOD. 1993. Top-down and bottom-up control of stream periphyton: Effects of nutrients and herbivores. Ecology **74:** 1264–1280.

SCHULZ, D. E., G. PETRICK, AND J. C. DUINKER. 1989. Complete characterization of polychlorinated biphenyl congeners in commercial Aroclor and Clophen mixtures by multidimensional gas chromatography–electron capture detection. Environ. Sci. Technol. **23:** 852–859.

SHARPLEY, A. N., R. G. MENZEL, S. J. SMITH, E. D. RHOADES, AND A. E. OLNESS. 1981. The sorption of soluble phosphorus by soil material during transport in runoff from cropped and grassed watersheds. J. Environ. Qual. **10:** 211–215.

———, AND J. K. SYERS. 1979. Phosphorus inputs into a stream draining an agricultural watershed: II. Amounts and relative significance of runoff types. Water Air Soil Pollut. **11:** 417–428.

STEWART, A. J., W. R. HILL, AND H. L. BOSTON. 1993. Grazers, periphyton and toxicant movement in streams. Environ. Toxicol. Chem. **12:** 955–957.

TAYLOR, W. D., J. H. CAREY, D. R. S. LEAN, AND D. J. MCQUEEN. 1991. Organochlorine concentrations in the plankton of lakes in southern Ontario and their relationship to plankton biomass. 1991. Can. J. Fish. Aquat. Sci. **48:** 1960–1966.