Lake trophic status and bioaccumulation of polychlorinated biphenyls in benthic organisms: a comparison between littoral and profundal invertebrates

Niklas Holmqvist, Patrik Stenroth, Olof Berglund, Per Nyström, Wilhelm Graneli, and Per Larsson

Abstract: We investigated the influence of lake trophic status on accumulation of polychlorinated biphenyls (PCBs) in littoral and profundal invertebrates from 19 lakes in southern Sweden. The PCB concentration in profundal chironomids (*Chironomus* sp.) was negatively correlated with lake trophy but the concentration in littoral crayfish (*Pacifasticus leniusculus*) was unrelated to shifting nutrient regimes. PCB concentrations in chironomids were negatively correlated, while crayfish did not correlate with lake total phosphorus. Two findings indicate that different processes determine bioaccumulation in littoral and profundal invertebrates: (*i*) a lack of correlation between PCB concentrations in crayfish and chironomids within lakes and (*ii*) some differences in congener distribution in the two species. We suggest a possible explanation in that chironomids are directly exposed to settling material from the pelagic zone, while crayfish are omnivorous and feed mainly in the littoral zone. A intensive investigation on one pond population could not reveal any individual factors, such as lipid content, size, or sex, influencing bioaccumulation of PCBs in crayfish.

Résumé : Nous avons étudié l'influence du statut trophique du lac sur l'accumulation des biphényles polychlorés (BPC) par les invertébrés des zones littorale et profonde de 19 lacs du sud de la Suède. La concentration de BPC dans les chironomidés (*Chironomus* sp.) de la zone profonde est fonction négative du statut trophique du lac, alors que celle dans les écrevisses littorales (*Pacifasticus leniusculus*) n'a pas de relation avec les divers régimes d'éléments nutritifs. Les concentrations de BPC chez les chironomidés sont en corrélation négative avec le phosphore total du lac, alors que celles de l'écrevisse ne montrent aucune corrélation. Deux résultats indiquent que des processus différents déterminent la bioaccumulation chez les invertébrés des zones littorale et profonde: (*i*) l'absence de corrélation entre les concentrations de BPC chez l'écrevisse et le chironomidé dans un même lac et (*ii*) les différences dans la répartition des congénères des BPC chez les deux espèces. Nous suggérons comme explication que les chironomidés sont directement exposés aux particules qui précipitent depuis la zone pélagique, alors que les écrevisses omnivores se nourrissent surtout dans la zone littorale. Une recherche détaillée sur une population d'étang n'a pu révéler aucun facteur particulier, tel que le contenu lipidique, la taille ou le sexe, qui influence la bioaccumulation des BPC chez l'écrevisse.

[Traduit par la Rédaction]

Introduction

Eutrophication and accumulation of persistent pollutants in biota are two major environmental problems in European marine and freshwater ecosystems. Lately, some investigations have focused on interactions between these two processes. A negative relationship between lake trophic status and levels of polychlorinated biphenyls (PCBs) has been shown for different components of the pelagic food web, from phytoplankton to piscivorous fish (e.g., Taylor et al. 1991; Larsson et al. 1992; Kidd et al. 1999). Few studies have focused on the interaction between eutrophication and accumulation of persistent organic pollutants (POPs) in benthic animals. Benthic animals potentially bioaccumulate sediment-borne contaminants as they spend their life in intimate contact with the sediment. Gunnarsson et al. (2000) found in laboratory experiments that POP concentrations in benthic animals could, contrary to pelagic biota, increase with increasing trophic status of water bodies. Owing to increasing settling of organic-rich particles, the benthic bioactivity increases with increasing disturbance of the sediment (bioturbidation) (Gilek et al. 1996; Björk and Gilek 1997;

N. Holmqvist,[1] **O. Berglund, and P. Larsson.** Department of Ecology, Institute of Ecotoxicology, Ecology Building, Lund University, SE-223 62 Lund, Sweden.
P. Stenroth, P. Nyström, and W. Graneli. Department of Ecology, Institute of Limnology, Ecology Building, Lund University, SE-223 62 Lund, Sweden.

[1]Corresponding author (e-mail: niklas.holmqvist@ekol.lu.se).

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Madsen et al. 1997). Bioturbation and increasing ventilation rates of sediments could enhance bioaccumulation of POPs in benthic invertebrates (Swackhamer and Skoglund 1993; Madsen et al. 1997; Gunnarsson et al. 2000).

The most commonly used explanation to the POP – lake trophy relationship in pelagic biota is that increased production or changes in the species composition of primary producers (phytoplankton), owing to excess of nutrients, cause a chain of events that results in reduced concentrations of POPs. The events include changes in the first link of the food chain where phytoplankton growth rate and lipid content affect bioconcentration of POP as well as increased sedimentation of organic matter (and POP) as a result of phytoplankton blooms. The growth dilution hypothesis is defined as the fast growth of phytoplankton, i.e., high cell division rates, that exceed uptake kinetics of POPs leading to decreasing concentrations in phytoplankton in nutrient-rich lakes (Taylor et al. 1991; Swackhamer and Skoglund 1993; Larsson et al. 2000). Alternatively, Berglund et al. (2001b) reported that decreasing concentrations of POPs with lake trophy is due to higher lipid content (and thus lipophilic POPs) in phytoplankton of oligotrophic systems owing to different species composition (mainly caused by diatom blooms) as a response to changing nutrient regimes.

Concentrations of various POPs in fish have been reported to follow the same pattern as in phytoplankton and decrease with increasing lake trophy. However, the explanation put forward was not a food chain related transport of pollutants dependent on growth rates or lipid content of phytoplankton. Instead, individual growth rate of fish was coupled to POP accumulation (Larsson et al. 1992): high nutrient status of the lake gives high growth rate of fish owing to high prey abundance leading to dilution of the pollutants following the same principle as for phytoplankton. Hence, by combining the different growth dilution events in plankton and at higher trophic levels, an additive effect of lake trophy in higher trophic levels within the pelagic food chain could be expected. However, Berglund et al. (2001b) could not find any such transport from phytoplankton to higher trophic levels (zooplankton and zooplanktivorous fish) within a lake trophy gradient.

The cycling of POPs in lakes is closely linked to the cycling of organic carbon. Organic carbon acts, owing to the lipophilic characters of POPs, as a major repository for POPs. Eutrophic lakes are characterized by large phytoplankton blooms that will increase the sedimentation of dead algae, organic carbon, when the blooms collapse (e.g., Larsson et al. 1998; Jeremiason et al. 1999). The total organic carbon mass in sediments will thus increase, by increased settling of dead algae, with increasing trophic status of lakes. The settling organic matter contains POPs that will accumulate in bottom sediments. This will lead to increased amounts of POPs in sediment but not necessarily increased concentrations. Lake morphometry affects the sedimentation process; in deep lakes, the major part of the organic carbon originating from phytoplankton is broken down, thereby concentrating POPs in sedimenting material (Larsson et al. 1998). The effect of lake trophic status on POP concentration in benthic biota is therefore not clear. Are high amounts of settling organic carbon and POPs in eutrophic lakes resulting in high levels of POPs in animals foraging in sedi-

ments or are processes in the pelagic compartment the major factor in determining exposure? Are the processes affecting uptake of POPs the same for animals living in the littoral and profundal zone?

We investigated the uptake of PCBs in signal crayfish (*Pacifastacus leniusculus*) and chironomid larvae (*Chironomus* sp.) as representatives for littoral and profundal benthic organisms in a trophic gradient of lakes. When studying between-lake differences, the variation between individuals could overshadow trends in the collected data. Therefore, an initial pilot study was made to investigate the intrapopulation variation within a crayfish population owing to differences in sex, size, and lipid content. However, we did not need to compensate for any of these variables, since none of them correlated with individual PCB concentrations in the population. By using both littoral and profundal animals, we wanted to investigate if processes affecting exposure, mechanisms of uptake, and concentrations of PCBs differed between the two compartments. Food source origin for benthic invertebrates may differ substantially between the littoral zone influence of terrestrial production and profundal areas where most energy input could be expected to originate from the pelagic photic zone.

Materials and methods

Study organisms

In this study, we used two different organisms, signal crayfish and chironomid larvae, as indicators of littoral and profundal food webs, respectively. The signal crayfish was introduced to Sweden in the 1960s and is today found in all types of freshwater ecosystems in southern Sweden and is common in both streams and lakes. It is generally found in the littoral zone but may inhabit hard bottoms in deeper areas of lakes. Diet examinations of crayfish have shown that individuals utilize a broad spectrum of food sources and shifts in diet may be induced by age and size, changing food resources, and changing environment (Nyström 1999). Chironomid larvae (Diptera: Chironomidae) are common in most lakes in southern Sweden and normally inhabit the sediment surface in the profundal zone. The larval stage may last for 1 to several years after which the insects metamorphose to the adult stage.

Intrapopulation variability in crayfish

During summer 2000, crayfish were collected from one population inhabiting a eutrophic pond in southern Sweden to examine if individual PCB levels were affected by intrapopulation variables such as sex, size, age, and fat content. Further, the variation of PCB concentration within the population was studied. From the crayfish population, a sample was drawn (n = 43) representing various sizes and both males (n = 24) and females (n = 19). The crayfish were collected using bated traps and were immediately frozen until analysis of lipid content and PCBs (see Sample preparation below).

Lakes

Lakes (n = 19) were investigated in the southern part of Sweden during August 2001. The lakes were chosen from different national lake databases to obtain high variation in

Table 1. Descriptive limnological data of lakes included in the study.

Lake	Latitude (°N)	Longitude (°E)	tot-N (mg·L^{-1})	tot-P (mg·L^{-1})	pH	Secchi depth (m)	Chl a (µg·L^{-1})	Crayfish (n)	Chironomid sample
Båtsjön	58°9′	16°9′	1.55	93	9.4	0.6	85.0	6	×
Bysjön	58°11′	15°58′	0.95	58	7.3	0.9	41.1	6	×
Växjösjön	56°51′	14°48′	0.83	54	7.9	1.3	20.0	5	×
Råbelövssjön	57°21′	15°23′	0.62	53	8.0	1.6	19.3	0	×
Lilla Nätaren	57°46′	14°35′	1.16	45	8.8	1.3	16.5	6	×
Getryggen	58°11′	16°4′	1.14	44	9.2	0.8	42.8	6	×
Skirösjön	57°10′	14°51′	0.86	42	7.1	1.0	3.2	6	×
Bergsjön	55°5′	14°44′	0.79	31	6.4	0.8	20.7	5	×
Stora Nätaren	56°5′	14°14′	0.85	30	8.5	1.5	7.7	6	×
Tolgsjön	57°4′	14°48′	0.42	25	6.7	1.8	8.9	6	×
Sommen	57°47′	14°33′	0.44	21	8.3	4.9	15.2	6	×
Bunn	57°56′	14°30′	0.56	19	7.6	2.9	4.9	6	
Örsjön	57°56′	14°34′	0.54	17	6.4	2.0	8.7	6	×
Hövern	58°20′	16°5′	0.54	16	7.3	2.4	4.0	6	×
Helgasjön	57°0′	14°41′	0.53	15	6.8	2.2	11.5	6	
Ören	56°27′	13°57′	0.43	12	8.0	4.9	2.9	6	×
Horsfjärden	58°9′	15°54′	0.33	8	6.5	6.5	3.0	6	
Vitavatten	56°15′	14°35′	0.29	6	6.3	6.8	1.8	6	×
Vättern	58°45′	14°50′	0.39	6	7.8	6.0	1.3	6	
Skärlen	58°7′	14°59′	0.33	5	6.6	7.7	4.1	4	×

Note: All parameters of lake trophic status were highly correlated, positively for total nitrogen (tot-N), total phosphorus (tot-P), and chlorophyll a (Chl a), (r = 0.64 to 0.89) and negatively for Secchi depth (r = –0.51 to –0.74). The last two columns show number of crayfish included in samples (n) and whether samples of chironomids were collected (×).

trophic status indicated by total phosphorus (tot-P), total nitrogen (tot-N), and chlorophyll a (Chl a). Water chemistry data from the sample event during this study are provided in Table 1. Other criteria considered were maximum depth (>5 m) and lake area (>50 ha). Contacts were taken with landowners to establish a rough picture of the crayfish population number and distribution. In this study, only lakes with an established crayfish population were included. The populations were considered established if (i) mean catch per unit effort was above five individuals per trap and (ii) the population was older than 5 years.

In all lakes included in the study, the crayfish populations were monitored using Swedish Standards Institute recording catch per unit effort, individual crayfish size, gender, etc. Crayfish traps (n = 100) were used in lines of five traps with 10 m distance reaching 40 m out from the beach. Each trap attracts and catches adult signal crayfish in a circle within a radius of approximately 2 m (Abrahamsson and Goldman 1970). Hence, the total area monitored by the 20 lines was approximately 1300 m^2 in each lake. From each population (i.e., lake), a subsample of six crayfish (three individuals with body length 80–100 mm of each sex except in two lakes where only five and in one lake where only four individuals within proper size range where caught; see Table 1) was taken for analysis of PCB and lipid content, resulting in a total of 110 animals. The crayfish were individually frozen at –20 °C within 24 h after capture.

Chironomid larvae were collected from the deepest part of each lake. Sediments were pumped to the lake surface and sediments mixed with lake water were flushed out in a sieve (mesh size 0.5 mm) and the chironomid larvae were collected. Chironomids were collected to obtain one composite sample (of at least >1 g wet weight, one sample for every

lake) for analysis of lipid content and PCBs. The samples were frozen and further treated similarly to crayfish samples.

Samples and measures of water chemistry (in the epilimnion) were taken at the same dates as the samples of benthic invertebrates and samples were determined according to Swedish standards (Swedish Standards Institute 1991). At the same time, Secchi depth was measured.

Sample preparation

Whole crayfish and pooled samples of chironomids were freeze-dried, homogenized, and kept dry (in closed containers at –20 °C) until time for extraction. The samples were Soxhlet extracted according to Bremle et al. (1995) using octachloronaphthalene as a surrogate standard. After extraction the solvent was reduced in a vacuum centrifuge. A subsample of the solvent was taken for lipid quantification and amounts were determined gravimetrically. The sample was redissolved in n-hexane and then transferred to an open column with two layers of silica gel for sample cleanup and separation of PCBs. The two layers had been treated with concentrated sulfuric acid and 1 mol $K_2CO_3 \cdot L^{-1}$, respectively. After column treatment, the solvent fraction containing PCBs was evaporated in a vacuum centrifuge, redissolved in isooctan, and stored in sealed capillary glass tubes containing copper grains (for elimination of sulfur). For every 12 samples extracted, a chemical blank was run. No PCBs were detected in the chemical blanks and thereby no blank correction was made.

PCB analyses

Samples were analyzed for PCBs by capillary gas chromatography – electron capture detection (Varian star 3400 cx electron capture detector, Varian, Inc., Palo Alto, Califor-

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nia) with a split–splitless injector, 25-m DB5 quartz capillary column (inside diameter 0.25 mm). PCB components were identified according to Schultz et al. (1989). The 18 predominant PCB congeners in the samples were included in ΣPCB in this study: International Union of Pure and Applied Chemistry (see Schultz et al. 1989) Nos. 52, 95, 101, 110, 149, 153, 138, 183, 128, 174, 177, 202, 180, 170, 201, 196, 194, and 206. Congeners were identified and quantified according to relative retention time and pentachlorbenzene was used as a standard for response factors for every run. External PCB standards of Clophen and Aroclor mixtures (Analytical Standards AB, Kungsbacka, Sweden) were included for every 10th sample for a quality check of both identification and quantification. Extraction efficiency for octachlornaphtalene was 94% ± 9% and no correction for recovery was made. The detection limits for ΣPCB were estimated to 1.1 ng·g dry weight^{-1} (sample size 1 g).

Statistical analysis

All concentration data on PCBs in crayfish or chironomids were log transformed owing to skewed distributions (Newton 1988) before statistical analysis. Possible relationships between variables were carried out by linear regression when the x variable could be expected to influence the y variable but not vice versa; otherwise, Pearson correlation was used. The crayfish intrapopulation variables were further tested using the data from lakes in an ANCOVA approach where log PCB concentration (dry weight) was the dependent variable in the analysis, lake was the independent grouping variable, and lipid content was the covariate. Comparisons of PCB concentrations between sexes were made using t tests in the intrapopulation study and paired t tests when comparing lake data.

The congener profile is presented as mean values normalized to the concentration of PCB 153 where mean PCB 153 concentration was set to 1. Mean values of individual congener concentrations were calculated on all samples of crayfish (n = 109) and chironomids (n = 15). The reason for normalizing for PCB 153 was that this congener had the most stable high concentration of the congeners present in both species. Normalization relative to PCB 153 has been used in earlier work (e.g., Derocher et al. 2003).

Results

Intrapopulation variability in crayfish

PCB concentrations in crayfish from the pond population (n = 43) varied between 3 and 198 ng·g dry weight^{-1}. The median concentration in the population was 12.5 ng·g dry weight^{-1}. As lipid content varied between 1% and 18% of the dry weight, individual PCB concentrations varied from 34 to 10 600 ng·g^{-1} on a lipid basis. Median concentration was 247 ng·g lipid weight^{-1}. The PCB concentration did not differ between male and female crayfish in the population (t test: df = 1, t = 0.133, p = 0.35), nor did lipid content (t test: df = 1, t = 4.0, p = 0.27). No correlation was found between size and lipid content (Pearson correlation: df = 42, r = 0.26, p = 0.09) of the crayfish or the concentration of PCB (Fig. 1) (Pearson correlation: df = 42, r = 0.25, p = 0.11). Lipid content was not correlated with carapace length (size

Fig. 1. Scattergram showing concentrations of PCBs (ΣPCB) in signal crayfish (*Pacifastacus leniusculus*) of increasing size in the single population data. Males are represented by solid squares and females by open squares. The linear regression shows no relationship between crayfish size and ΣPCB (r^2 = 0.15, p = 0.32, n = 43). This was also the case when data were separated into males (r^2 = 0.05, p = 0.29, n = 24) and females (r^2 = 0.06, p = 0.29, n = 19).
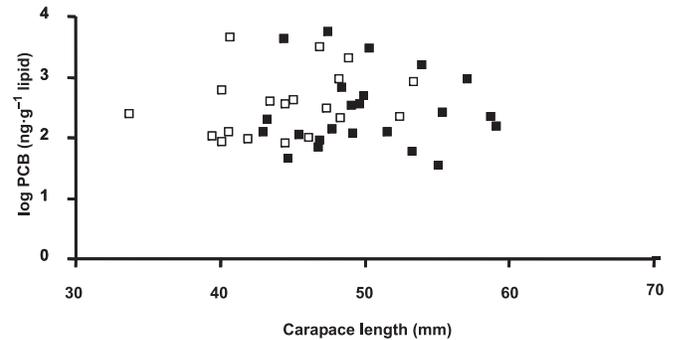
of crayfish) (Pearson correlation: df = 42, r = –0.05, p = 0.78).

We tested the general effect of lipid content on ΣPCB concentration in crayfish from all of the lakes in an ANCOVA. Log PCB concentration (dry weight) was the dependent variable in the analysis, lake was the independent grouping variable, and lipid content was the covariate. Since the interaction between lake and lipid was not significant ($F_{[1,71]}$ = 0.99, p > 0.47), this was omitted from the analyses. The ΣPCB concentration differed significantly between lakes (p < 0.001), but no significant effect of lipid content could be found on ΣPCB concentration ($F_{[1,89]}$ = 1.99, p > 0.16). The lipid content in lake crayfish varied between 0.5% and 13% (dry weight).
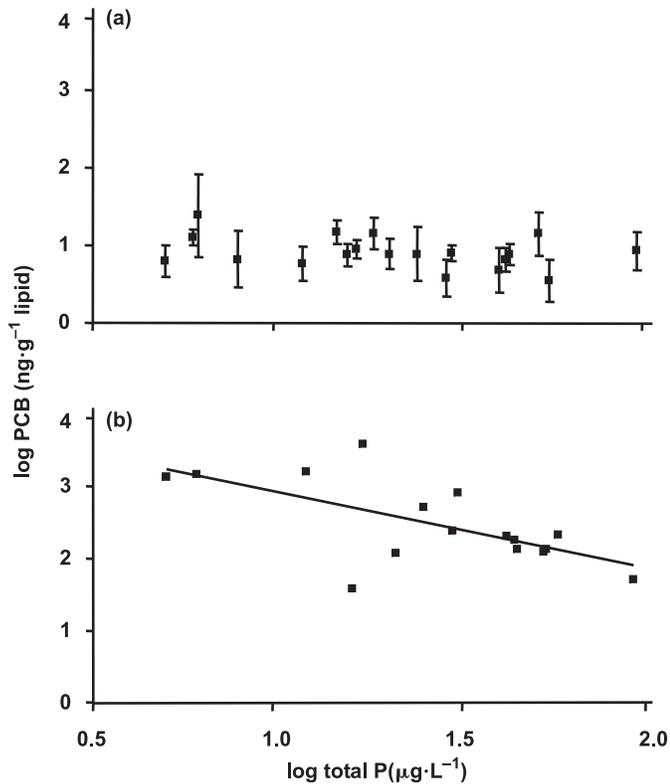
No difference could be found between crayfish males and females within lake data in ΣPCB (paired t test: df = 18, t = 0.94, p = 0.36), lipid content (paired t test: df = 18, t = 1.78, p = 0.09), and lipid-based ΣPCB (paired t test: df = 18, t = 0.16, p = 0.87).

Lake trophy related variability

The tot-P in the lakes varied from 5 to 93 μg·L^{-1}, tot-N from 0.3 to 1.6 mg·L^{-1}, and Chl a from 1.3 to 85 μg·L^{-1}. The Secchi depth varied from 0.6 to 7.7 m. All parameters of lake trophic status were highly correlated, positively for tot-N, tot-P, and Chl a (Pearson correlation: df = 18, r = 0.64–0.89, p < 0.01) and negatively for Secchi depth (Pearson correlation: r = –0.51–0.74, p < 0.01) (Table 1).

PCB concentrations varied between 1.5 and 260 ng·g dry weight^{-1} in crayfish (n = 110) in the studied lakes with a median of 8.4 ng·g dry weight^{-1} and between 4.0 and 780 ng·g dry weight^{-1} in chironomid larvae (n = 15) with a median of 14 ng·g dry weight^{-1}. The ΣPCB concentrations in chironomid larvae were negatively related to lake trophic status (tot-P, linear regression: df = 14, r^2 = 0.30, p = 0.03) and significance increased when values were lipid normalized (Fig. 2b) (linear regression: df = 14, r^2 = 0.38, p < 0.01). The lipid content in chironomid larvae partly explained PCB concentrations (linear regression: df = 14, r^2 = 0.41, p < 0.01). Lipid content in chironomids did not corre-

Fig. 2. (*a*) Mean concentration of PCBs (ΣPCB) in signal crayfish (*Pacifastacus leniusculus*) from lakes of increasing trophic status. Females (*n* = 3) and males (*n* = 3) were analyzed from each lake. The analysis is based on mean values, and error bars represent ±1 SD (linear regression: $r^2 = 0.08$, $p = 0.29$, $n = 19$). (*b*) ΣPCB in chironomid larvae from lakes of increasing trophic status (linear regression: $b = –0.65$, $r^2 = 0.42$, $p = 0.001$, $n = 15$).

Fig. 3. Scattergram showing mean concentration of PCBs (ΣPCB) for signal crayfish (*Pacifastacus leniusculus*) versus ΣPCB in pooled chironomid samples in lakes (*n* = 15). No correlation could be found between the two compartments (Pearson correlation $r = 0.18$, $p = 0.52$).

late with lake trophy in this study (Pearson correlation: df = 14, $r = 0.07$, $p = 0.80$). Lipid content in chironomid larvae samples ranged between 7% and 21% dry weight.

There was no relationship between lake trophic status (tot-P) and mean ΣPCB concentrations in crayfish on a dry weight basis (linear regression: df = 18, $r^2 = 0.08$, $p = 0.29$) or when values were lipid normalized (Fig. 2*a*) (linear regression: df = 18, $r^2 = 0.03$, $p = 0.56$). The lipid-based ΣPCB concentrations in crayfish and chironomid larvae within lakes were not correlated (Fig. 3) (Pearson correlation: df = 14, $r = 0.09$, $p = 0.76$). The PCB concentrations in chironomids were higher than in crayfish within lakes (paired *t* test: df = 14, $t = –10.8$, $p < 0.001$).
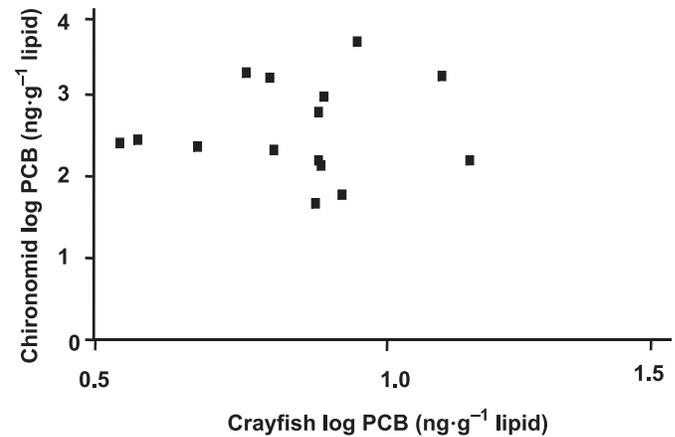
The PCB congener distribution showed slight differences in crayfish and chironomid samples (Fig. 4). The three dominating congeners in crayfish were the medium-chlorinated congeners PCB 149, PCB 153, and PCB 138. Dominating congeners in chironomids were the medium-chlorinated PCB 153 and the two highly chlorinated congeners PCB 180 and PCB 196.

Discussion

Lake trophy

Contrary to what could be expected, given the results from earlier experimental studies (Gilek et al. 1996; Björk and Gilek 1997; Gunnarsson et al. 2000), we found that PCB

concentrations in profundal benthos (chironomid larvae) were negatively related to lake trophic status. In earlier experiments, marine filter-feeding mussels were exposed to additions of algae at different concentrations. The concentrations of PCBs in mussels increased with increasing algae biomass, indicating a positive relationship between trophic status of the water and bioaccumulation of PCBs in benthic animals. Investigations of the relationship between trophic status of waters and POP accumulation in benthic animals have so far been restricted to a few experimental studies (summarized in Gunnarsson et al. 2000). One of the conclusions from these studies is a concern that increasing eutrophication may increase POP loads in benthic organisms because increasing settling of organic carbon rich particles increases the bioturbation of sediments. We found that different compartments of the benthic food web may differ considerably in bioaccumulation of PCBs within lakes. While the PCB concentrations of profundal organisms were negatively correlated with lake trophy, the PCB concentrations in littoral crayfish did not show any relationship with changing nutrient regimes. The lack of correlation within lakes between the two compartments indicates that different processes are governing the uptake of PCBs in littoral and profundal organisms.

A negative relationship between lake trophic status and ΣPCB concentrations in pelagic biota has been reported for freshwater phytoplankton (Berglund et al. 2001*a*, 2001*b*), zooplankton (Taylor et al. 1991), and fish (Larsson et al. 1992). The main explanation for decreasing levels of POPs in the piscivorious northern pike (*Esox lucius*) of eutrophic systems was increasing individual growth rates in the pike population (Larsson et al. 1992). Studies on pelagic plankton led to a hypothesis that a high growth rate of phytoplankton in nutrient-rich waters affects the concentration of POPs, i.e., that cell division rates of phytoplankton exceed the uptake kinetics of POPs (e.g., see Larsson et al. 2000) This restriction in uptake efficiency may lead to lower concentrations of POPs in the first link of the food chain in eutrophic environments (Larsson et al. 2000). However, Berglund et al. (2001*b*) reported that differences in phyto-

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Fig. 4. Congener pattern of the 18 most common PCBs in signal crayfish (*Pacifastacus leniusculus*; $n = 110$) (open bars) and chironomid larvae ($n = 15$) (solid bars). Mean values are calculated as individual congener concentration (ng·g lipid^{-1}) divided by congener 153 concentrations for each sample.
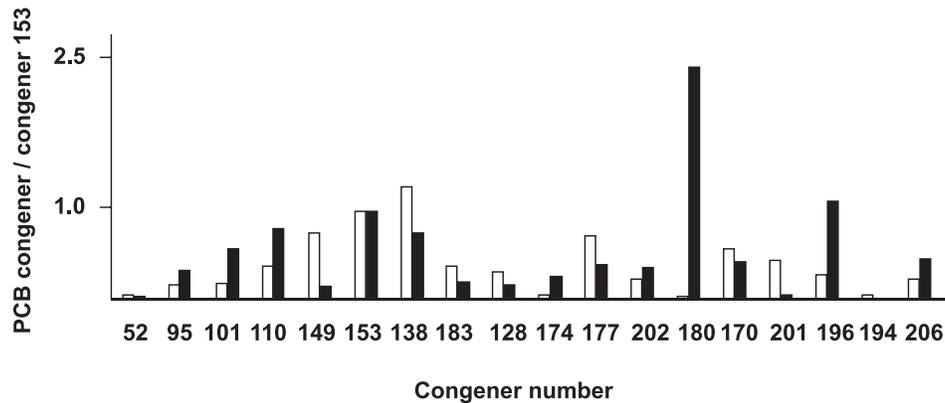
plankton lipids might explain most of the variance in ΣPCB concentrations. The higher levels of POPs in the phytoplankton of oligotrophic systems could thereby be explained by differences in species composition and lipid content as a response to nutrient stress. So far, investigations made on the relationship between bioaccumulation of POPs in biota and lake trophy have been focused on pelagic species.

After being incorporated in pelagic biota and (or) organic matter, parts of the particle-bound POPs will be transported to the bottom of lakes by sedimentation (see Fig. 5). The concentration of POPs in settling particles (i.e., sedimentating seston, see above) has been reported to be dependent on lake trophic status with higher concentrations in oligotrophic lakes. In Larsson et al. (1998), a comparison of a eutrophic and an oligotrophic lake showed that microbial breakdown of the organic matter concentrates POPs in the remaining organic matrix. The efficiency of this magnification process was calculated to be about four times more effective in the oligotrophic lake compared with the eutrophic lake. Hence, particles reaching the sediment have higher concentrations of POPs in oligotrophic systems, but owing to higher sedimentation rates in eutrophic lakes, the total amount reaching sediments is higher (Berglund et al. 2001*a*).

Internal lake processes redistribute PCBs between water, sediment, and biota (see below). There appears to be an intense (re-)cycling of organic carbon and PCBs at the sediment–water interface and the layer above, the nepheloid layer (Baker and Eisenrich 1985). The recycling of organic carbon and POPs has also been reported to be dependent on the trophic status of lakes; more organic carbon in the sediment will give a higher biological activity and thereby more bioturbation in eutrophic systems (e.g., Gunnarsson et al. 2000). However, bioaccumulation of sediment-derived POPs in chironomids and tubificides has been shown to be inversely related to organic carbon content in sediments (Bremle and Ewald 1995). Owing to their lipophilicity, POPs have high affinity for organic-rich material and thereby a low probability of being bioaccumulated in chironomids and tubificides in organic carbon rich sediments. Our data could in the light of these two findings give some support to the conclusion that high organic carbon content in

sediments binds the PCBs and makes them less available for bioaccumulation in sediments with high organic carbon content (i.e., eutrophic lakes).

Problems with finding a reliable equilibrium between benthic biota and POPs in sediments have been reported from several studies (e.g., Bremle and Ewald 1995; Gunnarsson et al. 1999). Further, when Bremle and Ewald (1995) compared the PCB congener patterns in chironomid samples from different parts of a lake, the variation could not be explained by congener variation in the sediments from the same locations alone. Instead, the variation was explained by varying quality of organic carbon, which influenced PCB sorption, and thereby bioaccumulation. Higher levels of POPs in benthic biota than expected from equilibrium calculations with sediments have also been explained with selective feeding on organic-rich and (or) highly contaminated particles (Gunnarsson et al. 1996, 1999; Morrison et al. 1996). In surface sediments, where most benthic invertebrates feed, particles with the highest nutritional value are newly settled particles (Larsson et al. 1998). After settling on the sediment surface, particles undergo rapid microbial transformations or breakdown. This transformation of particles rapidly decreases both organic carbon content and POP concentration in the surface sediment. Baker et al. (1991) showed that settling particles have 10–100 times higher levels of POPs compared with resuspended sediments. Thus, higher concentrations of PCBs in benthic invertebrates than to be expected from equilibrium calculations with sediment could be explained by selective feeding on fresh untransformed material from the pelagic compartment.

We suggest that the found negative relationship between PCB concentration and lake trophic status in the profundal organism is simply a mirror of the relationship found for pelagic biota. High concentrations of PCBs in the pelagic food webs, and especially in phytoplankton, in lakes will result in high concentrations in profundal benthic organisms regardless of varying organic carbon content in the sediment. The profundal organism used in this study, chironomid larvae, feeds in the upper layers of the sediment (Jonasson 1972) and is thereby directly exposed to settling material from the pelagic environment. Consequently, independent of organic carbon content or other biochemical variables in the sediment, PCB concentration and congener distribution in

Fig. 5. Conceptual view of fate of PCBs and internal pelagic–profundal processes in oligotrophic and eutrophic lakes. The flow of organic matter is represented by the size of boxes and arrows and the PCB concentration by the intensity of the grayscale. In the pelagic compartment of the eutrophic lake, there is a high amount of organic matter (biomass) but low PCB concentrations (e.g., Taylor et al. 1991). During sedimentation, degradation of organic matter magnifies PCB concentrations (Baker and Eisenrich 1985; Larsson et al. 1998) and this process is more efficient in the oligotrophic lake (Larsson et al. 1998), leading to higher PCB concentrations in freshly settled material. PCB concentrations in surface sediments are unrelated to lake trophy but amounts are positively related to lake trophy owing to the higher amount of settling of organic matter in eutrophic systems (Berglund et al. 2001a). PCBs can be buried into deep sediments and inactivated together with organic carbon (eutrophic lake: Jeremiason et al. 1999; Berglund et al. 2001a) or recycled back to the ecosystem owing to partitioning and bioturbation (oligotrophic lake: Gunnarsson et al. 2000). The results from this study show that the PCB concentration in benthic invertebrates feeding on the surface sediment is higher in the oligotrophic system, represented by the circle in the sediment box.
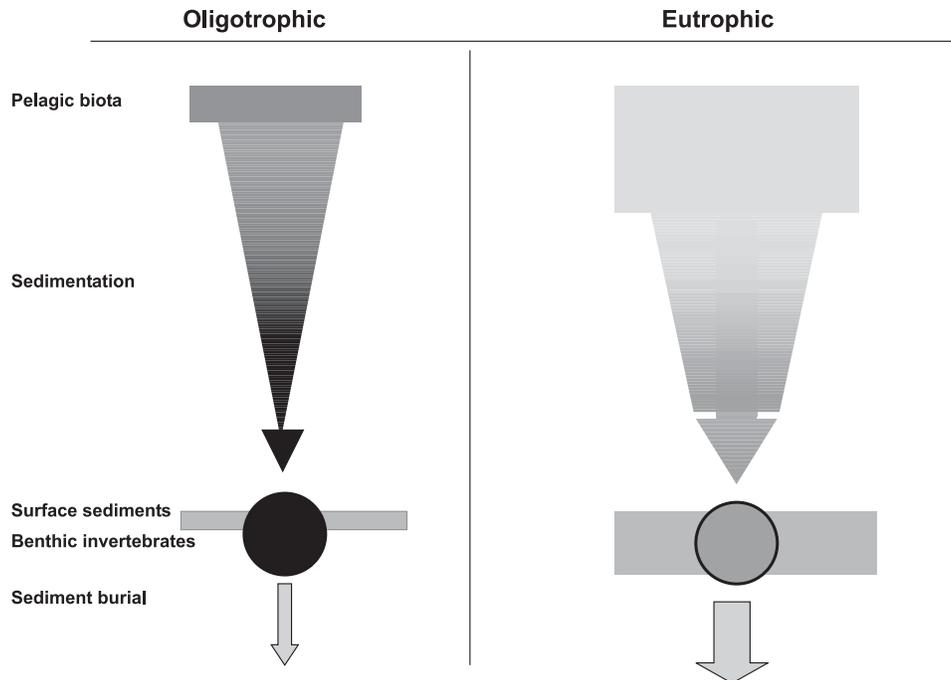
profundal sediment surface feeding biota will be determined by processes in the pelagic compartment and during sedimentation.

Profundal and littoral benthic food webs

Although we compared two different organisms, one in the littoral zone and one in the profundal zone, slight differences in congener pattern and the absence of a correlation between PCBs in chironomids and PCBs in crayfish indicate that different processes determine PCB accumulation in the two compartments. Owing to the hydrophobic nature of PCBs, they associate with particles and sorption is mainly governed by hydrophobicity of the compound and lipid and (or) organic content of the particle (Swackhamer and Skoglund 1993; Berglund et al. 2001a). During sedimentation, microorganisms process particles and less lipophilic congeners can be recycled back to the pelagic environment (Baker et al. 1991). The process leads to a higher proportion of highly lipophilic substances in particles entering the sediment surface (Baker et al. 1991). In Bremle and Ewald (1995), congener profiles in chironomid samples similar to those found in this study are reported. Proportionally high concentrations of highly chlorinated congeners compared with sediments were found. The differences in congener distribution could be a result of that profundal benthos mainly

feeds on settled pelagic phytoplankton. In littoral food webs, the major source of energy is primary production in macrophytes and periphytic algae with the addition of terrestrial production (Wetzel 1983). Both macrophytes and periphyton are important food sources for signal crayfish (Nyström 1999). Differences in species composition and thereby lipid quality and quantity in macrophytes and periphyton could change between lakes of different trophic status and species composition as found in phytoplankton. However, accumulation of PCBs in macrophytes and periphyton is a poorly investigated area. Vanier et al. (1999) reported PCB accumulation in macrophyte roots to be directly dependent on PCB concentration in the sediment, while the uptake in shoots is more likely dependent on other variables such as concentration in the surrounding water. Further, uptake in roots was proposed to be important in transporting sediment-buried PCBs back to biota in lake ecosystems. It can only be speculated that accumulation of PCBs in macrophytes and periphyton is independent of lake trophy and could thereby explain the lack of correlation between (i) crayfish uptake of PCBs and lake trophy and (ii) PCB accumulation in crayfish and that in chironomids within lakes.

Alternatively, the differences between the two compartments can be explained by terrestrial input of energy and PCB to the ecosystem. Especially nutrient-poor lake ecosys-

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tems have low production of autotrophic energy and may drive the system to be more dependent on terrestrial energy input (Wetzel 1983). The effect of terrestrial input of energy and PCBs is probably most obvious in the littoral food chain and could contribute to the differences between the two compartments. Changes to a more terrestrial-based diet especially in littoral organisms in oligotrophic lakes as well as changes in food web structure with changing lake trophy (Momot 1995; Nyström 1999) could thereby change the exposure of PCBs to crayfish.

Benthic–pelagic coupling

Berglund et al. (2001*b*) reported that the negative correlation between lipid content in primary producers and ΣPCB with lake trophy was not transferred to higher trophic levels, zooplankton and juvenile zooplanktivorous fish, in the pelagic food chain. This finding indicates that the decreasing concentrations of PCB with increasing lake trophy found in higher trophic levels, e.g., piscivorous fish (Larsson et al. 1992), are not due to a pelagic food chain mediated effect of bioaccumulation in phytoplankton. Instead, this relationship can, which has indeed been proposed by several authors (e.g., Larsson et al. 1992, 2000; Skei et al. 2000), be explained by excessive nutrient loads that increase individual growth rates and that high growth rate leads to a second level of growth dilution. These two processes could maybe work separately but with the same result giving decreasing levels of PCBs in the organisms with increasing eutrophication in the organisms. Alternatively, or complementary to, benthic invertebrates, such as chironomid larvae, act as transport vectors of the phytoplankton effect to higher trophic levels in the pelagic food web. As suggested earlier in this paper, pelagic processes could explain the negative relationship between levels of PCB in chironomid larvae and lake trophy. Consequently, consumption of chironomids by fish could act as a feedback loop of settled PCBs and complement the expected transport route phytoplankton–zooplankton–fish in the pelagic food chain. A feedback loop of PCB by phytoplankton – chironomids – benthivorous fish – pike could be an alternative or complementary explanation to the "growth dilution" hypotheses presented in Larsson et al. (1992). A possible transport of PCBs from sediments to fish via benthic invertebrates has been reported in other studies (e.g., Bremle and Ewald 1995; Gunnarsson et al. 2000).

Intrapopulation variability

In the part of the study where the variance in PCB concentration within one crayfish population was studied, we could not find any correlation with individual-based variables. The age of crayfish is hard to determine because growth varies considerably with food quality and quantity, water temperature, and individual size (Belchier et al. 1998). The lack of correlation between size (age) and PCB concentration in the crayfish population, which has been shown in freshwater fish (Boileau et al. 1979; Larsson et al. 1991; Madenjian et al. 1994), can be explained by the fact that, contrary to fish, the crayfish has continuously moulting stages during its life span. The process is energy demanding and, probably, means a total utilization of any energy stores, like those containing lipids. Lipophilic pollutants like PCBs may then be fully or partly eliminated with cycles of PCB

accumulation occurring from moult to moult, not over the whole life span as, e.g., in fish. Hence, no effect of age and size can be found in lipid content and (or) accumulation of PCBs. In fact, the different life history, compared with, e.g., fish, can explain the lack of differences between males and females as well. If lipids are not continuously stored during life, no differences can be established between sexes that are commonly found in other organisms owing to the loss of lipids during egg production and depuration (Harding 1986; Larsson et al. 1992; Johnston et al. 2002). This suggests that the positive relationship between fat content and accumulation of POPs in biota can be an effect of life history and a continuous storage of lipids during life that will result in increased accumulation of POPs with time. This process is not valid for crayfish and probably not for any aquatic invertebrate with continuously moulting stages.

In conclusion, bioaccumulation of PCBs in crayfish was unrelated to size, sex, and lipid content. We propose that internal lake processes in the pelagic environment directly influence PCB concentration of profundal chironomids. The negative relationship between lake trophy and pelagic species was also found in sediment surface living chironomids in the profundal zone. PCB concentrations in crayfish living in the littoral zone seemed unaffected by lake trophy and the concentrations were not correlated with chironomid concentration within lakes. Therefore, we speculate that different factors are involved in the exposure of PCBs in the two compartments.

Acknowledgements

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