Arch. Environ. Contam. Toxicol. 17, 721–729 (1988)

Archives of Environmental Contamination and Toxicology

Polychlorinated Biphenyl Congeners in Sediments, Plankton, Molluscs, Crustaceans, and Eel in a Freshwater Lake: Implications of Using Reference Chemicals and Indicator Organisms in Bioaccumulation Studies

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Abstract. The concentrations of six polychlorobiphenyl (PCB) congeners in sediments and four classes of biotic species of the aquatic food chain were investigated in a freshwater lake near Amsterdam. Despite the low concentrations of the contaminants in the sediment, significant amounts of PCBs were found in plankton, macro-invertebrates and fish. The composition of the PCB mixtures found in the various organisms cannot be explained in terms of simple partitioning of the PCB congeners between sediment, water, and organisms. In addition to bioconcentration, it is likely that biomagnification via consumption of contaminated food also contributed significantly to the total PCB concentrations. This is most pronounced for the higher trophic food-chain organisms. Studying bioaccumulation processes by monitoring just one type of organism is probably not very suitable, due to the pronounced differences in accumulation patterns demonstrated for the different organisms. In addition, significant differences were found in the accumulation of the six PCBs in the various biotic samples. This indicates that the fate of mixtures of PCBs is determined by the fate of the individual congeners and therefore cannot be monitored in terms of total PCBs concentrations.

Polychlorinated biphenyls (PCBs) are well known environmental micropollutants. Due to their hydrophobicity and their low degradability, these compounds can accumulate to high concentrations in organisms relative to the concentrations found in the environment (Thomann and Connolly 1984). In particular, high concentrations can be found in natural aquatic organisms, while at the same time concentrations found in water are very low, even below detection limits.

Accumulation of PCBs and other classes of poorly degradable hydrophobic chemicals has been shown in fish and other types of aquatic species (Bruggeman *et al.* 1984; Opperhuizen *et al.* 1985; Biddinger and Gloss 1984). It has been argued that a compound's bioaccumulation factor (the ratio between the concentration in the organism and the concentration in the surrounding environment during a steady state) for various species may be comparable if it is expressed in terms of the organism lipid weight (Geyer *et al.* 1985; Chiou 1985). If so, the bioaccumulation factor determined for one indicator organism may be used to calculate the bioaccumulation factor in other organisms, if the lipid content of the species involved is known.

For the natural environment, little is known about how hydrophobic chemicals, such as the PCBs, enter organisms. In general, it is assumed that chemicals are bioaccumulated after being taken up directly from the water (bioconcentration) or from contaminated food (biomagnification) (Thomann and Connolly 1984; Landrum *et al.* 1987; Eaton *et al.* 1983; McCarthy 1983). Uptake directly from contaminated particles or sediments ingested by the organism, is usually considered to be less important. On the other hand, contaminated particles or sediments in the aquatic ecosystem can act as a source of these contaminants which then can be bioconcentrated in the organism (Rubinstein *et*

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al. 1985). It has been shown that even if direct uptake of the chemicals from water doesn't contribute significantly to the total concentration in organisms, the ratio between these concentrations in the organisms and those in sediments tends to be constant (Connor 1984; Breck 1985; Rubinstein *et al.* 1983). This ratio may be dependent on the organic carbon content of the sediment. Hence, organic carbon standardized ratios may represent the capacities of PCBs and other hydrophobic chemicals to accumulate in biota of natural aquatic ecosystems.

Polychlorinated biphenyls are released into the environment as mixtures, containing many congeners. It has been shown that the bioaccumulation capabilities of the individual congeners are determined by their physico-chemical properties and their molecular configuration. A linear relationship between the logarithms of the octanol/water partition coefficients and the bioconcentration factor has accordingly been found for various PCB congeners (Spacie and Hamelink 1982; Bruggeman *et al.* 1984). On the other hand, significant differences have also been demonstrated between the bioaccumulation potential of several tetrachloro congeners, which cannot be explained by the small differences between the respective hydrophobicities (W. A. Bruggeman, Bioaccumulation of polychlorinated biphenyls and related hydrophobic chemicals in fish, Ph.D. Thesis University of Amsterdam, 1983).

Furthermore, it has been shown that the bioaccumulation of individual highly chlorinated biphenyls is not significantly altered if they are administered as a mixture (Opperhuizen and Jongeneel 1986). Therefore, after exposure of aquatic organisms to mixtures of hydrophobic chemicals, the composition found in the organism will not necessarily resemble that of the original mixture, which is due to the different bioconcentration and biomagnification factors of the congeners (Opperhuizen and Jongeneel 1986; Calambokidis *et al.* 1979).

In a series of studies, the authors' laboratories are investigating the fate and ecotoxicological effects of organic micropollutants on freshwater ecosystems. In the present study, the distribution of six polychlorinated biphenyls in sediments, water, and biota of a freshwater lake was investigated, using four different classes of aquatic organisms, *i.e.*, plankton, molluscs, crustaceans and fish. The aim was to find whether or not the lipid-standardized bioaccumulation factors would be comparable for different PCB congeners in various aquatic organisms. The ratio between the lipid-standardized concentrations in the organisms and the concentrations in the organic fraction of the sediment was taken as a measure of the biotic fate of these hydrophobic chemicals in aquatic ecosystems. Whether these ratios are a manifestation of steady states between the concentrations in water, sediment and organisms, or are also influenced by the uptake of contaminated food, will be investigated.

In addition, it will be discussed whether the biotic fate of the six different PCB congeners may be representative for the fate of mixtures of extremely hydrophobic chemicals.

Experimental

Sample Collection

In Lake Nieuwe Meer (a fresh-water lake located in the southwestern part of Amsterdam) contaminated dredged materials from the Amsterdam city canals have been discharged for over thirty years. All samples were taken in or close to the discharge area of the lake. Samples were collected of water, top sediment, plankton (mainly *Microcystis aeruginosa* and *Daphnia longispina*), molluscs (*Dreisena polymorpha*), crustaceans (*Gammarus tigrinus, Asellus aquaticus* and *Orchestra carimana*) and fish (eel = *Anguilla anguilla*).

Ten L samples of surface water (0–25 cm depth) were collected in clean brown-glass bottles. Samples of top sediment (0–20 cm) were collected with a bottom sediment grab sampler. At each site, four separate grabs were taken and homogenized. The homogenized sample was placed in a clean glass jar, sealed with aluminium foil and stored at 4°C. Plankton was collected by towing two plankton nets (55 and 335 μ mesh) at a depth of 0 to 1 m through the water. The collected plankton was transferred to clean glass jars sealed with aluminium foil. In the laboratory, the samples were centrifuged for 10 min at 4,000 rpm (Christ UJ1S). The condensed plankton pellets were stored in a freezer at −25°C. Molluscs and crustaceans were hand-picked and identified. Both macro-invertebrates and eel were stored in a freezer (−25°C) until analysis.

Polychlorobiphenyl Analyses

Analysis of polychlorinated biphenyls was performed by means of hexane extraction, followed by column chromatographic clean-up and analysis on a gas chromatograph with electron capture detector (GC-ECD). Hexane and acetone were obtained from J. T. Baker Chemical Co., Phillipsburg, NJ 08865 (Resi-analyzed) and ethanol from E. Merck, Darmstadt (Reinst 95%). Sodium sulphate from E. Merck, Darmstadt (anhydrous, pro analysis) was dried for at least 12 hr at 300°C before use. Deionized water was extracted with hexane before use. Aluminium oxide (Woelm B super I, ICN Pharmaceuticals Inc.) was activated at 150°C for at least 16 hr and de-activated with 11% w/w water for at least 16 hr. Silicon dioxide—Kieselgel 60 (70–230 mesh ASTB, E. Merck) was activated at 150°C for at least 16 hr and de-activated with 5% w/w water for at least 16 hr. Individual PCBs for use as standards were obtained from Dr. S. Ehrenstorfer, D8900 Augsburg 1, Germany.

Sediment: The organic matter content of the sediments was determined by the modified method reported by Mebius (1960). For PCB analysis, samples of 50 g were shaken with 50 ml of

acetone for 20 min and equilibrated for at least 4 hr. The acetone layer was filtered over an acetone-washed paper filter and collected in a separating funnel. Fifty ml of fresh acetone were added to the remaining sediment and the procedure was repeated. After combining the aliquots, the acetone extract was desulphurated with 5 ml of sodium sulphite, extracted twice with 50 ml of hexane and washed twice with 250 ml of water together with 15 ml of saturated sodium chloride solution. The combined hexane extract (100 ml) was dried by adding 3 g of anhydrous Na_2SO_4 and shaking the suspension for 1 min. This extract was transferred quantitatively over a hexane-washed glass microfibre filter into a 3-ball Snyder apparatus, in which it was concentrated to 5–10 ml. The extract was concentrated to 1 ml under nitrogen.

Organisms: The lipid weight of the organisms was determined by the method reported by Bligh and Dyer (1959). For PCB analysis organisms (3–30 g depending on their lipid weight) were weighed, and, when necessary, cut into small pieces. The tissues were heated under reflux with 25 ml of ethanol and 25 ml of a 40% (w/v) KOH solution for 4 hr. The ethanol extract was transferred quantitatively into a 1L separatory funnel, and extracted once with 50, and twice with 25 ml of hexane. The combined hexane extracts were washed with 25 ml of water and shaken 4 times with 25 ml of a 2% (w/v) Na_2SO_4 solution. The extract was dried and concentrated in the same way as the sediment extracts.

Water: For PCB analysis, water samples (10 L) were extracted consecutively by shaking amounts of 1 L twice with 50 ml hexane after adding 50 ml of a saturated NaCl solution to the water. After extracting the sample, the two hexane extracts were combined and treated similar to the sediments extracts.

Clean-up: The concentrated PCB-extracts (1 ml) were eluted with 10 ml of *n*-hexane through a 1.50 g alumina (11% H_2O) column, which was previously washed with 2 ml of hexane. The eluate was concentrated in a calibrated test tube to 1 ml under nitrogen. The concentrated eluate was eluted with 10 ml of *n*-hexane through a 0.60 g silica (5% w/w H_2O) column, prewashed with 2 ml hexane. The eluate was concentrated to 2 ml under nitrogen.

Gas Chromatographic Analysis

For analysis, 1 μl of the extracts was automatically injected into a gas chromatograph equipped with a splitless Grob-injector. A Packard-Becker 438-S gas chromatograph was used, equipped with a Packard LS607 automatic sampler and a 25m × 0.22mm i.d. fused silica capillary column (CP-S8 CB, Chrompack). The system was connected to a Nelson Analytical 760 series interface and a Hewlett-Packard 9000-216 computer for data analysis.

For analysis the following conditions were used:

Purge valve switch	0.5 min
Initial temperature	60°C
Hold time	1 min
Program rate 1	10°/min to 180°C, then
Program rate 2	4°C/min to 250°C
End temperature time	27 minutes
Injector temperature	305°C
Detector temperature	310°C
Carrier gas	nitrogen
Nitrogen pressure	60 kPa

The individual PCB-congeners were selected to span the whole range of chlorination. Usually, these six congeners (Table 1) are the most prominent in sediments of Dutch waters. In the investigated sediments, these selected PCBs represented an average of over 20% of the total PCB concentration. The PCB-congeners were identified and quantified by comparing retention times and areas with those of PCB standards. With the electron capture detector, 5 pg amounts of the individual congeners could be detected, so that the detection limit was approximately 5 ng/ml extract. If the concentration of a congener was below the detection limit, a value of 50% of the detection limit was arbitrarily used for the calculation of the mean PCB contents. The detection limits for the six separate PCB congeners in the various compartments are listed in Table 1. These values are based upon the mean weights of the samples, the organic matter fractions or the lipid contents, final extraction volumes and the response factors of each congener.

Extraction and analysis were reproducible with a standard deviation less than 10%. Analysis of samples spiked with standard solutions of the PCBs in hexane, demonstrated that the recovery of the analytical procedures was between 90 and 100% for each of the test compounds. Every series of PCB analyses was accompanied by blanks, in which no detectable PCBs were found.

Statistical Analysis

The data were analyzed by the Mann-Whitney-Wilcoxon rank sum nonparametric test, using the SPSS/PC+ statistical software on a Tulip PC. Significant differences were determined at a 0.05 level of probability using a 2-tailed test. Linear regression was calculated by a least-squares approximation.

Results

During the period from June to September 1986, a large number of samples (Table 2) were taken from Lake Nieuwe Meer. The concentrations of six PCB congeners in these samples were determined. In general, no detectable concentrations of PCBs were found in the water samples, the detection limit being approximately 0.3 ng/L. On the contrary, in most of the sediment and organism samples PCBs were found, although not all the investigated PCB congeners were detected in all samples. The mean PCB concentrations expressed on a dry weight (sediment) or fresh weight basis (organisms) are listed in Table 2.

Since it is usually assumed that hydrophobic chemicals sorb on the organic phase of particles, all concentrations are expressed on the basis of the organic carbon content (OC) of the sediment (Table 3). The mean organic carbon content of the sediments was 9.7%. Bioaccumulation of hydrophobic chemicals may be dependent on the lipid content of the various organisms; thus, PCB concentrations are expressed on a lipid weight basis (Table 3). The mean lipid weights of the organisms studied were: plankton 0.65%, molluscs 1.74%, crustaceans

Table 1. Octanol/water partition coefficients and detection limits of the PCB congeners

PCB-no Name	log $K_{d,oct}$[a]	Detection limit[b] wat.	sed.	pla.	mol.	cru.	eel
28:2,4,4'-tri	5.46	0.24	1.2	17.2	37.3	49.0	5.2
52:2,2',5,5'-tetra	5.64	0.60	3.0	42.9	93.3	122.4	12.9
101:2,2',4,5,5'-penta	6.83	0.46	2.3	32.9	71.5	93.8	9.9
138:2,2',3,4,4',5'-hexa	6.92	0.28	1.4	20.0	43.5	57.1	6.0
153:2,2',4,4',5,5'-hexa	7.23	0.20	1.0	14.3	31.1	40.8	4.3
180:2,2',3,4,4',5,5'-hepta	7.40	0.10	0.5	7.2	15.6	20.4	2.2

[a] source: Opperhuizen, unpublished results
[b] wat. = water (ng PCB/l); sed. = sediment (ng PCB/g OC); pla. = plankton (ng PCB/g lipid); mol. = molluscs (ng PCB/g lipid); cru. = crustaceans (ng PCB/g lipid); eel = eel (ng PCB/g lipid)

Table 2. PCB concentrations in sediment (ng PCB/g dry weight) and organisms (ng/g fresh weight), and their standard deviations (in parentheses)

PCB-no Compartment[a]	28	52	101	138	153	180
Sediment	1.35	2.34	4.09	13.74	7.21	3.37
(n = 11)	(0.90)	(1.94)	(3.63)	(11.68)	(7.13)	(3.77)
Plankton	0.23	0.30	0.53	0.69	0.54	0.22
(n = 14)	(0.22)	(0.28)	(0.50)	(0.64)	(0.47)	(0.23)
Molluscs	0.52	0.89	3.85	4.52	5.77	1.25
(n = 5)	(0.36)	(0.88)	(0.64)	(1.26)	(2.20)	(0.88)
Crustaceans	2.93	3.15	4.31	4.18	3.97	0.80
(n = 7)	(1.41)	(1.61)	(1.96)	(1.66)	(3.24)	(0.62)
Eel	3.98	14.75	28.42	97.35	89.02	40.92
(n = 6)	(3.42)	(12.15)	(23.67)	(68.93)	(60.07)	(33.94)

[a] n = number of samples analyzed

0.86% and eel 14.9%. Fresh weight PCB concentrations in the sampled organisms, when arranged in order of ascending trophic level (Table 2), show that the PCB concentrations are higher in higher trophic level organisms. The PCB concentrations on a lipid weight basis do not show a similar pattern for the various PCB congeners. These data are consistent with those reported for Lake Geneva (Mowrer *et al.* 1982).

Although the standard deviations of the PCB concentrations in the different types of samples are rather high, the statistical significance of the differences between the concentrations in the different types of samples can be investigated by applying the Mann-Whitney-Wilcoxon Rank Sum test (Table 4).

In Figure 1, the logarithms of the ratios between PCB concentrations in the organisms and the average concentration in the sediments are plotted against the cumulative probability distribution of the samples for each separate congener and each class of organisms involved. The median ratios were determined by interpolation, *i.e.*, the 50% cumulative probability ratios of the plots after linear regression. The results are shown in Table 5.

In Figure 2, the median C_{org}/C_{sed} ratios are shown for each PCB congener. The lower chlorinated compounds (28,52,101) accumulate in the crustaceans, while no clear distinction between the other organisms can be observed. For the higher chlorinated compounds (138,153,180) bioaccumulation increases with higher trophic levels.

Figure 3 shows the ratio between the concentrations in the organisms and sediments of the four classes of organisms investigated. For plankton and molluscs, no distinction can be observed between these ratios of the six PCB congeners studied. For crustaceans, the ratio decreases as the chlorine content of the biphenyls increase. For eel, however, the reverse is found.

Discussion

In the aquatic environment, hydrophobic chemicals such as PCBs partition between water and various non-aqueous compartments such as biotic lipids, organic phases of sediments or suspended particles. Generally, it is assumed that these partitioning processes can be described by first order kinetics.

Table 3. PCB-concentrations in sediment (ng PCB/g organic carbon) and organisms (ng PCB/g lipid weight) and their standard deviations (in parentheses)

PCB-no Compartment	28	52	101	138	153	180
Sediment	17	25	42	129	65	33
	(10)	(13)	(18)	(72)	(47)	(28)
Plankton	68	102	206	259	209	60
	(56)	(78)	(173)	(212)	(157)	(56)
Molluscs	36	57	220	258	322	75
	(15)	(44)	(20)	(61)	(97)	(46)
Crustaceans	362	400	532	529	505	107
	(192)	(251)	(284)	(280)	(524)	(106)
Eel	21	83	186	986	932	436
	(12)	(76)	(131)	(1162)	(1148)	(600)

Table 4. Statistical differences between PCB concentrations with the Mann-Whitney-Wilcoxon Rank Sum Test

PCB-no Compartments[a]	28	52	101	138	153	180
sed-pla	<*	<*	<*	<	<	<
sed-mol	<*	<	<*	<*	<*	<
sed-cru	<*	<*	<*	<*	<*	<
sed-eel	<	<	<*	<*	<*	<*
pla-mol	>	>	<	<	<	<
pla-cru	<*	<*	<*	<	<	<
pla-eel	>*	>	>	<	<	<*
mol-cru	<*	<*	<*	<	<	=
mol-eel	>	<	>	<	<	<
cru-eel	>*	>*	>*	<	<	<

* = significant difference (2-tailed p-value < 0.05)

[a] sed = sediment; pla = plankton; mol = molluscs; cru = crustaceans

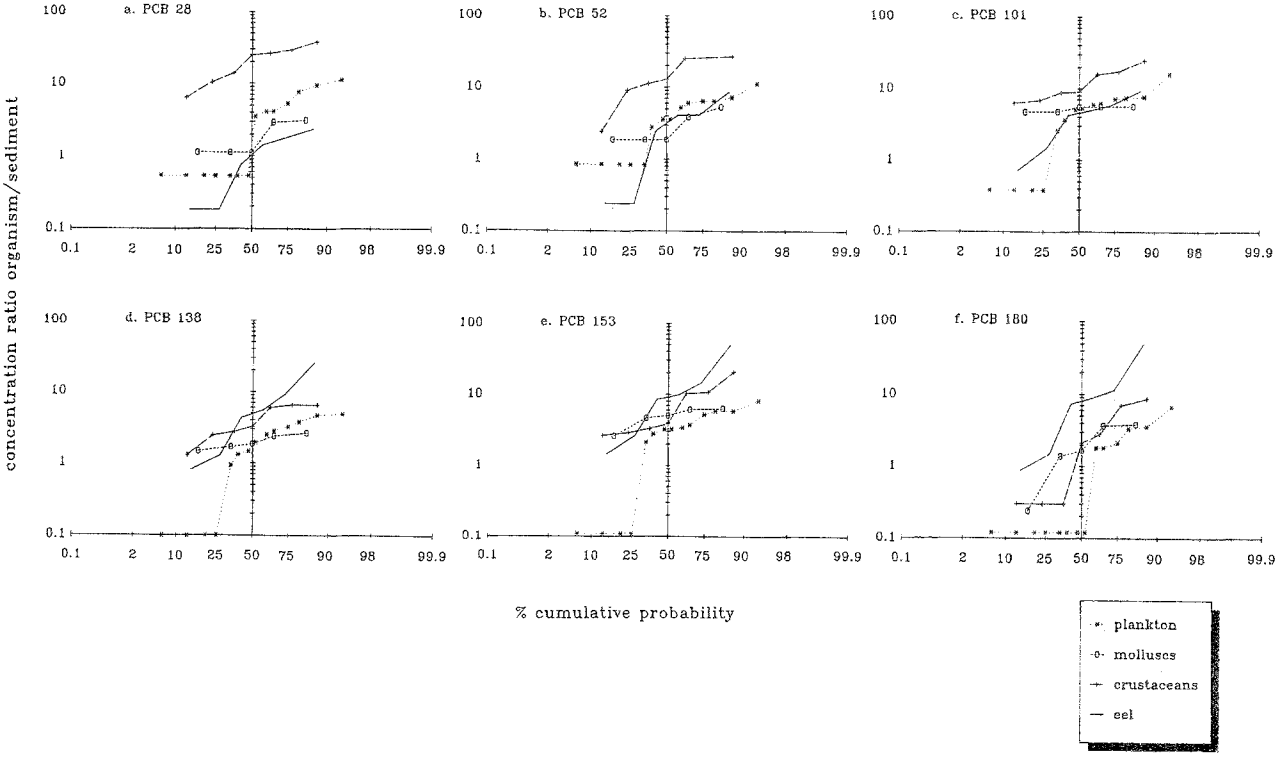

Fig. 1. Logarithms of C_{org}/C_{sed} ratio versus the cumulative probability distribution of the samples for each PCB congener. (non-detectable concentrations are represented by their 50% detection limit levels)

For the partitioning of chemicals between water and lipid phases of organisms, the bioconcentration factor (K_c) is defined as:

$$K_c = \frac{C_{org}}{C_w} \quad (1)$$

If the sorption of hydrophobic chemicals is also considered as a partitioning between water and the organic fraction of the sediment then, during equilibrium, the sorption coefficient (K_a) can be expressed as:

$$K_a = \frac{C_{sed}}{C_w} \quad (2)$$

If the processes of bioconcentration and sorption on sediments have both reached an equilibrium, then equations 1 and 2 can be combined:

$$C_{org} = K_c/K_a\, C_{sed} \quad (3)$$

Table 5. Median PCB C_{org}/C_{sed} ratios in different organisms

PCB-no / Organism	28	52	101	138	153	180
Plankton	3.3	3.8	4.5	1.7	3.1	1.3
Molluscs	2.8	3.2	5.2	2.0	4.8	1.9
Crustaceans	18.2	12.8	11.2	3.4	5.8	2.1
Eel	1.5	3.0	3.3	4.3	7.8	6.8

From Figures 1 and 2, it is clear that for the three lower chlorinated congeners the median C_{org}/C_{sed} ratios found for crustaceans are significantly higher than those found for the other organisms, despite the variation in concentrations. This is confirmed by Mann-Whitney-Wilcoxon statistics (Table 4). For the higher chlorinated compounds, however, no such significantly higher ratio is found for the crustaceans. Higher C_{org}/C_{sed} ratios of PCB's in crustaceans than in fish are consistent with the data reported by Connor (1984), but the lower ratio for molluscs found in the present study contradicts the literature data.

The present data indicate that the C_{org}/C_{sed} ratio of the PCB congeners in natural organisms is not just a reflection of the partitioning of the chemicals between water, organic carbon of the sediments and organisms' lipids. Otherwise, for instance, the ratio of 2,4,4′-trichlorobiphenyl should be equal for the various organisms. In Figure 2, the median C_{org}/C_{sed} ratios of the various PCB congeners are shown for the different organisms. Experimental data on the ratios between concentrations in biotic species and sediments are very scarce. It has been argued that such ratios may be comparable for a large series of chemicals, because the process of sorption and the process of bioaccumulation are both determined by the hydrophobicities of the chemicals involved. The hydrophobicity of a chemical is often expressed by its octanol/water partition coefficient. For both the bioconcentration factor and the sorption coefficient of polychlorinated biphenyls and structurally related compounds, relationships have been obtained with the octanol/water partition coefficient (Connor 1984; Shaw and Connell 1984; Karickhoff 1984; Kenaga and Goring 1980; Spacie and Hamelink 1982). After combining these results, a relationship between the C_{org}/C_{sed} and the octanol/water partition coefficient should result (Connor 1984; Breck 1985). As is shown in Figures 3a and 3b for plankton and molluscs, the ratios between the median concentrations in organisms and in sediment are almost independent of the physico-chemical properties of the individual congeners. This is consistent with the assumption that there will be equilibria between the concentrations in the sediment, water and biota (Brown *et al.* 1982; Clayton *et al.* 1977). This can be explained by the relatively high rate constants of the exchange processes between the various environmental compartments. It has been previously shown that both the uptake rates of hydrophobic chemicals in organisms and the sorption rates are high (Bruggeman *et al.* 1984; Opperhuizen *et al.* 1985; Opperhuizen and Jongeneel 1986; Karickhoff 1984). Since, for the lower chlorinated compounds, both the elimination rate in organisms and the rate of desorption are relatively high, equilibrium will be achieved relatively fast (Wang *et al.* 1982; Vreeland 1974; Calambokidis *et al.* 1979; Hiraizumi *et al.* 1979; Shaw and Connell 1987). In addition, the aqueous solubilities of the lower chlorinated congeners are sufficient to enable water to act as an important medium for the transfer of chemicals between sediments and organisms.

On the other hand, for the higher chlorinated congeners it can be seen (Figure 3), that the partitioning of the compounds between the sediments and the various organisms' lipids is not comparable, because the C_{org}/C_{sed} ratios for the various organisms are significantly different. As is shown in the Figures 2a and 2b, only in plankton and molluscs is the distribution of the higher chlorinated congeners between organisms and sediment comparable to that of the lower chlorinated congeners. In eel, however, the accumulation of the higher chlorinated compounds is significantly higher than that of the lower chlorinated congeners. Simple partitioning of these PCB congeners between sediment, water and eel, is not a satisfactory explanation for this phenomenon. Therefore, it is likely that uptake by consumption of contaminated food contributes significantly to the total accumulation of PCBs in eel. This is consistent with data obtained in other fish species (Thomann and Connolly 1984). The increase of the C_{org}/C_{sed} ratio with increasing hydrophobicity may be explained by the difference between the biomagnification factors of the various congeners. Generally, biomagnification can be described by:

$$dC_{org}/dt = E\, f\, C_{food} - k_2\, C_{org} \qquad (4)$$

in which E denotes the uptake efficiency of the chemical by fish from food, f the feeding rate, and k_2 the elimination rate constant. During a steady state in fish the biomagnification factor can be defined as:

$$K_m = C_{org}/C_{food} = E\, f/k_2 \qquad (5)$$

This equation shows that the biomagnification factor increases with decreasing elimination rate constant. Since elimination rate constants decrease

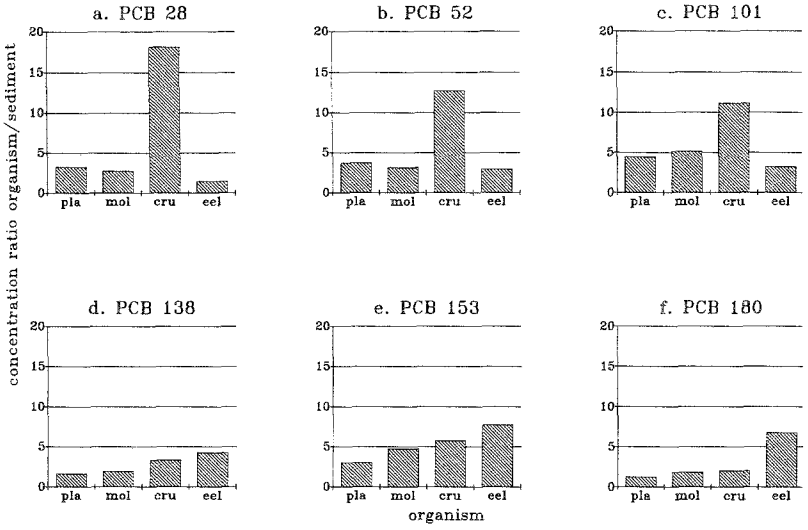

Fig. 2. C_{org}/C_{sed} ratio for different PCB congeners. Organisms: pla = plankton; mol = molluscs; cru = crustaceans; eel = eel

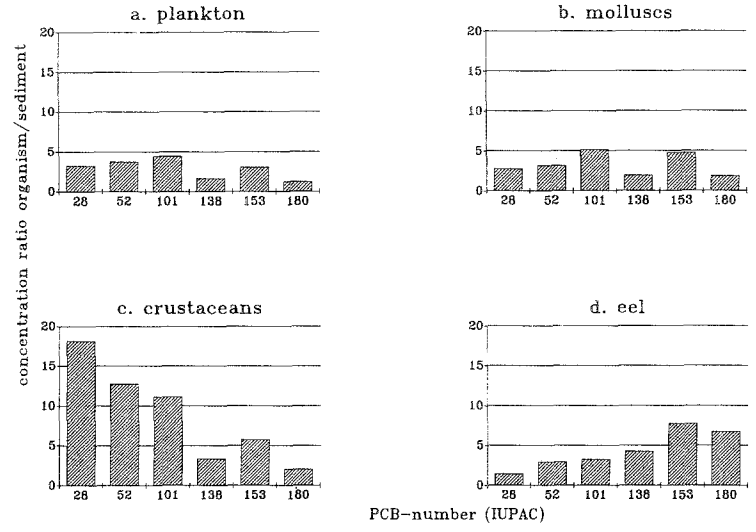

Fig. 3. C_{org}/C_{sed} ratio of PCBs for different organisms of Lake Nieuwe Meer

with increasing hydrophobicity of the chemical (Opperhuizen *et al.* 1985; Bruggeman *et al.* 1984) and uptake efficiencies of PCBs in fish are not dependent on their hydrophobicity (Opperhuizen and Schrap, 1988) it is clear that biomagnification factors increase with increasing hydrophobicity. It should be noted that biomagnification factors, in contrast to bioconcentration factors, are not partition coefficients. They do not merely reflect the partitioning of the chemical between food and organism, but represent the ratio between the uptake rate from food and the elimination rate from the fish (Brown *et al.* 1982).

The concentration pattern in the crustaceans is the reverse of that in eel. The significantly higher concentrations of PCBs found in crustaceans, relative to the concentrations found in plankton and molluscs, is in agreement with previous observations (Pinkney *et al.* 1985). These results can be explained by the contribution of uptake via the food, and is probably not due to the low mixed-function oxidase enzyme activity as was suggested by Connor (1984), since all six PCB congeners are biotransformed very poorly, if at all (Bruggeman *et al.* 1984).

The decreasing C_{org}/C_{sed} ratios for crustaceans with increasing hydrophobicity is probably due to the kinetics of the accumulation process. As is shown in equation 4, the time required to achieve steady state concentrations in organisms after dietary exposure is dependent upon the uptake efficiency and the elimination rate constant. A possible reason for the higher concentrations of the lower chlorinated PCBs in crustaceans could be a more efficient uptake of these congeners. In several studies, however, it was demonstrated that the dif-

ference between clearance rates for the PCB congeners rather than between uptake rates account for different biomagnification factors (Bruggeman *et al.* 1981; Opperhuizen and Schrap, 1988). As was demonstrated earlier for the higher chlorinated congeners in aquatic organisms, the elimination rate constants are extremely low. For example, the elimination rate constant of 2,2′,4,4′,5,5′-hexachlorobiphenyl in fish is 0.007 d^{-1} (Bruggeman *et al.* 1984). For other aquatic organisms the elimination rate constants are often comparable with those determined in fish (Yamato *et al.* 1983; Gooch and Hamdy 1982). After integration of equation 4 and substituting the value of 0.007 for k_2, it can be calculated that it will take nearly two years of continuous exposure before 90% of the steady state concentration will be reached. For organisms such as crustaceans with a short lifespan relative to the biological half lives of the chemicals, this means that steady state concentrations are probably never achieved. So, we suggest that the lower concentrations of the higher chlorinated PCBs in crustaceans are a manifestation of bioaccumulation kinetics rather than of equilibrium. The concentrations of the higher chlorinated biphenyls in fish may also be kinetically controlled (Bruggeman *et al.* 1984; Schrap and Opperhuizen, 1988). However, since the mean age of the eel significantly exceeds the biological half-lives of the PCBs it is clear that the concentrations of the higher chlorinated PCB congeners in eel are closer to their steady state values than those in crustaceans. It should be noted that the greater mobility of fish can also be a factor influencing the different concentration ratio's. Since biomagnification probably contributes to the PCB-accumulation in eel and crustaceans, differences in feeding habits of both groups may be important for the composition of the bioaccumulation patterns.

Conclusions

After analysis of the concentrations of six polychlorinated biphenyl congeners in four types of biota and in the sediments of a fresh water lake, it can be concluded that:

A. Despite the relatively low levels of contamination of the lake sediments studied, significant amounts of polychlorinated biphenyls tend to accumulate in various classes of organisms (plankton, molluscs, crustaceans and fish) of Lake Nieuwe Meer. This accumulation cannot be explained by equilibrium partitioning of the chemicals between the organic matter of the sediments and the organisms' lipids. Uptake of contaminated food contributes significantly to the organisms' concentrations of higher chlorinated PCB-congeners.
B. Studying one class of organisms as a monitoring species for bioaccumulation of hydrophobic chemicals is inadequate, since pronounced species differences between the accumulation of PCBs in different organisms exist. These differences are probably caused by differences in the contribution of uptake via the food and by differences in the period of exposure.
C. Investigation of the environmental fate of six PCB congeners support the idea that the biotic fate of mixtures of PCB's can be understood as the sum of the fates of the individual components of the mixture. Hence, analysis should be carried out in terms of individual components rather than as technical formulations (Duinker and Hillebrand 1983). If only a few mixture components are used to monitor the aquatic fate of mixtures of PCBs, it is of paramount importance that these reference congeners cover at least a wide range of physico-chemical properties in order to be representative for the whole mixture.

In future research, bioaccumulation of PCB-congeners, organochlorine pesticides and polycyclic aromatic hydrocarbons (PAH) will be investigated in relation to the possible induction of sublethal toxic effects.

Acknowledgments. The authors thank Dr. John R. Parsons and Prof. Dr. N. P. E. Vermeulen for their useful comments and suggestions and Ed van der Wal for his technical assistance.

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Manuscript received November 18, 1987 and in revised form March 7, 1988.