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DIETARY ACCUMULATION AND DEPURATION OF HYDROPHOBIC ORGANOCHLORINES: BIOACCUMULATION PARAMETERS AND THEIR RELATIONSHIP WITH THE OCTANOL/WATER PARTITION COEFFICIENT

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Abstract—Dietary accumulation of 23 hydrophobic organochlorines (OCs) by juvenile rainbow trout (*Oncorhynchus mykiss*) was studied with the objective of obtaining relationships between bioaccumulation parameters and the octanol/water partition coefficient (K_{ow}). A wide range of OCs were used including 16 polychlorinated biphenyls (PCBs 18, 28, 44, 52, 66, 101, 105, 118, 128, 138, 153, 187, 189, 195, 206, and 209), hexachlorobenzene, mirex, tris(4-chlorophenyl)methane (TCPMe), tris(4-chlorophenyl)methanol (TCPMeOH), and three toxaphene congeners (Cl_7-chlorobornane [CHB] [Hp-sed], Cl_8-CHB [T2], and Cl_9-CHB [T12]). Tris(4-chlorophenyl)methane (half-life [$t_{1/2}$] = 65 d) was more persistent than TCPMeOH ($t_{1/2}$ = 20 d), and TCPMe was not biotransformed to TCPMeOH by rainbow trout. Cl_7-chlorobornane ($t_{1/2}$ = 32 d) was more rapidly eliminated, and appears to be more readily metabolized, than Cl_8-CHB ($t_{1/2}$ = 43 d) and Cl_9-CHB ($t_{1/2}$ = 42 d). With the exception of TCPMeOH, Cl_7-CHB, and PCB 18, all of the OCs had biomagnification factors (BMFs) >1, implying a potential to biomagnify. Half-lives had a significant curvilinear relationship with K_{ow} (R^2 = 0.85, $p < 0.001$), with a maximum $t_{1/2}$ for OCs with log $K_{ow} \sim 7.0$. Decreasing $t_{1/2}$ for OCs of log $K_{ow} > 7.0$ may be related to slow kinetics of these super hydrophobic OCs and the short exposure phase, which results in insufficient time for the super hydrophobic OCs to reach slower clearing compartments of the rainbow trout. Assimilation efficiency was not as well described by K_{ow} as by $t_{1/2}$ and BMF, although a significant curvilinear relationship was observed (R^2 = 0.53, p = 0.004). The BMF had a significant curvilinear relationship with log K_{ow} (R^2 = 0.84, $p < 0.001$). Recalcitrant OCs with a log K_{ow} of ~7.0 would appear to have the greatest potential for food chain biomagnification in fish.

Keywords—Dietary accumulation Octanol/water partition coefficient Hydrophobic organochlorines Toxaphene congeners Tris(4-chlorophenyl)methane

INTRODUCTION

Fish, and many aquatic invertebrates, accumulate organochlorine compounds (OCs) from water (bioconcentration) and from food (biomagnification). The relative importance of these pathways varies with the water solubility of the OC and the trophic position of the organism. With decreasing water solubility [1–3] and increasing trophic position [1,4,5] there is greater accumulation from food. Greater than 99% of the polychlorinated biphenyls (PCBs) (log octanol/water partition coefficient [K_{ow}] > 5.0) in Lake Michigan lake trout have been estimated to be accumulated through the food chain [6]. Also, Thomann et al. [7] concluded that OCs of log $K_{ow} > 5$ in Lake Ontario sculpin are almost entirely derived from the food chain. Food has also been identified as the most important exposure route for PCBs in the aquatic food chain from the western basin of Lake Erie [8].

Bioaccumulation parameters, such as bioconcentration factor (BCF) or biomagnification factor (BMF), are important for the prediction of exposure to OCs in the aquatic environment. The relationship between the bioconcentration (BCF) of hydrophobic OCs and K_{ow} has been extensively studied [9–15]. However, application of BCF–K_{ow} relationships may not provide adequate data for fate and exposure assessment of recalcitrant hydrophobic OCs (log $K_{ow} > 5$) in the environment. Laboratory-derived BCF–K_{ow} comparisons were only useful in predicting field-derived BCFs for Lake Ontario rainbow trout (*Oncorhynchus mykiss*) for OCs of short half-lives ($t_{1/2}$ s), but not for OCs of longer half-life [2]. Swackhamer and Hites [16] concluded that laboratory-derived BCF–K_{ow} models gave a poor approximation of the bioaccumulation of hydrophobic OCs in lake trout and whitefish. Further, Thomann and Connolly [6] determined that PCB levels in Lake Michigan lake trout were not predicted by the BCF–K_{ow} relationship. Surprisingly, few attempts have been made to develop relationships between dietary accumulation of hydrophobic OCs and K_{ow}.

The lack of dietary accumulation–K_{ow} relationships is probably due to the scarcity and variability of kinetic data [17]. Dietary accumulation data obtained for hydrophobic OCs in laboratory-derived experiments are more variable than data from BCF experiments [18]. This variability is a function of the differences in food, the logistics of feeding fish, and differences in experimental methodology [19]. Dietary composition may also influence adsorption efficiency of organochlorines. Efficiency may be less in low-digestibility diets that are low in fat and protein and high in fiber [20]. Fish of different species, age, size, and sex will feed and digest food at different rates, which could influence assimilation efficiency. Although size and species differences also affect uptake of OCs from water, their influence on dietary accumulation parameters may

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be greater. Feeding rates (i.e., grams of food per day) established by the investigator can influence parameters. An increase in the rate of feeding was found to decrease the absorption efficiency of OCs [21]. Also, high concentrations in the food may cause fish to stop feeding through avoidance or because of a toxic response in the fish. In either case the fish are no longer being exposed, whereas fish exposed to high water concentrations cannot avoid the OCs.

Our objective was to develop physicochemical property–dietary bioaccumulation relationships using a wider range of hydrophobic OCs than had been studied previously. We exposed juvenile rainbow trout to dietary concentrations of 23 OCs, including 16 PCB congeners, hexachlorobenzene (HCBz), mirex, three toxaphene congeners (Cl_7-chlorobornane [CHB], Cl_8-CHB and Cl_9-CHB), tris(4-chlorophenyl)methane (TCPMe), and tris(4-chlorophenyl)methanol (TCPMeOH), to determine bioaccumulation parameters. We choose PCB congeners with 3 to 10 chlorines to provide a range of K_{ow}s that include the most hydrophobic of the PCBs. To reduce the influence of metabolic transformation, PCB congeners with meta and para chlorine substitution were chosen. Toxaphene, TCPMe, and TCPMeOH are global contaminants [22–24], but have not been studied in laboratory dietary bioaccumulation experiments. Toxaphene, and in particular Cl_8-CHB and Cl_9-CHB [25], have been found to bioaccumulate and biomagnify in aquatic food chains [26,27]. The PCB congeners, mirex, and HCBz have well-established K_{ow}s, and for many of these OCs data are available on dietary accumulation by fish. By exposing the rainbow trout to a mixture of all the OCs we have sufficient data to develop dietary accumulation parameter–K_{ow} relationships without many of the confounding problems of compiling data from different experiments and exposures.

METHODS AND MATERIALS

Chemicals and food preparation

Hexachlorobenzene, mirex, and the 16 PCB congeners (PCBs 18, 28, 44, 52, 66, 101, 105, 118, 128, 138, 153, 187, 189, 195, 206, and 209) were purchased from Ultra Scientific (North Kingstown, RI, USA). Tris(4-chlorophenyl)methane and TCPMeOH were synthesized at Carleton University (Ottawa, ON, Canada). The three toxaphene congeners (Cl_7-CHB [2-exo,3-endo,5-exo,6-endo,8c,9b (or 8b,9c),10a-heptachlorobornane], Cl_8-CHB [2-exo,3-endo,5-exo,6-endo,8,8,10,10-octachlorobornane], and Cl_9-CHB [2-exo,3-endo,5-exo,6-endo,8,8,9,10,10-nonachlorobornane]) were isolated and purified from Arctic mammal blubber using methods outlined previously [28,29].

Food was spiked by mixing a known quantity of each of the 23 organochlorines (listed above), dissolved in 150 ml of hexane, with 60 g of commercial fish food (Martin's Feed Mills, Elmira, ON, Canada) and slowly evaporating to dryness. Control food was treated in an identical manner but without the addition of the organochlorine compounds. Food was air-dried for 24 h and stored at 10°C. The fish food consisted of 41% protein, 14% lipid, and 3% fiber. Concentrations of each OC were determined in control and spiked food using the same analytical techniques used to determine concentrations in the rainbow trout tissue (Table 1).

Table 1. Concentrations (ng/g, wet weight) of organochlorines (mean ± 1 SE, n = 3) in the control, low, and high treatment food

Compound[a]	Control	Low	High
Miscellaneous organochlorines			
TCPMe	54.1 ± 27.6[b]	54.9 ± 22.0	185 ± 3.2
TCPMeOH	<0.01	33.7 ± 4.7	276.1 ± 4.9
Mirex	1.1 ± 0.3	20.6 ± 0.3	144.6 ± 4.4
HCBz	1.3 ± 0.1	13.7 ± 0.3	102.6 ± 5.6
Toxaphene congeners			
Cl_7-CHB	0.2 ± 0.0	20.8 ± 0.7	135.6 ± 2.0
Cl_8-CHB	2.6 ± 0.1	17.6 ± 0.1	120.6 ± 3.4
Cl_9-CHB	3.8 ± 0.3	17.2 ± 0.1	134.1 ± 2.4
Polychlorinated biphenyls			
PCB 18	<0.01	28.9 ± 0.9	182 ± 11.4
PCB 28	<0.01	15.9 ± 0.5	108.1 ± 8.8
PCB 44	0.9 ± 0.0	17.8 ± 0.4	129 ± 6.7
PCB 52	3.4 ± 0.2	21.6 ± 0.7	125 ± 6.9
PCB 66	1.0 ± 0.0	21.1 ± 0.2	158 ± 7.8
PCB 101	4.3 ± 0.3	20 ± 0.4	148 ± 14.0
PCB 105	<0.01	17 ± 0.6	133 ± 3.4
PCB 118	2.0 ± 0.1	20.3 ± 0.2	133 ± 9.1
PCB 128	<0.01	8.3 ± 1.1	99.4 ± 2.7
PCB 138	8.8 ± 0.3	31.3 ± 1.4	176 ± 7.0
PCB 153	7.1 ± 0.3	22.2 ± 0.3	124 ± 5.7
PCB 187	3.2 ± 0.1	19.4 ± 0.1	122 ± 5.1
PCB 189	0.1 ± 0.0	18.7 ± 0.2	138 ± 2.5
PCB 195	0.2 ± 0.0	24.0 ± 0.3	176 ± 3.1
PCB 206	0.4 ± 0.0	20.1 ± 0.3	141 ± 3.0
PCB 209	0.2 ± 0.0	61.5 ± 0.8	688 ± 56.4

[a] TCPMe = tris(4-chlorophenyl)methane; TCPMeOH = tris(4-chlorophenyl)methanol; HCBz = hexachlorobenzene; CHB = chlorobornane.

[b] TCPMe was not quantifiable in one control food subsample but had high levels in the other two subsamples. Control fish did not accumulate TCPMe to the extent expected from a concentration this high, which suggests that the high concentration reported for the control food is not accurate.

Experiments

Juvenile rainbow trout (initial weights 2–4 g) were exposed to the spiked food (containing all OCs) for 30 d followed by 160 d of depuration. Rainbow trout were maintained in fiberglass aquarium (40 L) with flow-through, ultraviolet and carbon dechlorinated, City of Winnipeg (MB, Canada) water (~10°C) with a 12 h light:12 h dark schedule. The daily rate of feeding was equal to 1.5% of the mean weight of the rainbow trout, corrected after each sampling period. All food was consumed in <1 min after being offered to the fish. Three fish were sampled from each treatment for organochlorine analysis on days 5, 10, 20, and 30 of the uptake period, and days 5, 10, 20, 40, 80, and 160 of the depuration period. One-liter water samples were also taken 1 h and 24 h after feeding on day 30 of the uptake phase. Sampled fish were separated into liver, gastrointestinal (GI) tract (includes stomach, pyloric caeca, spleen, intestines, and adipose fat associated with these organs, as well as gut contents), and carcass (whole fish minus liver and GI tract). Only results from the carcass samples were used for calculation of bioaccumulation parameters.

A second experiment exposed juvenile rainbow trout to trout food spiked with high concentrations of TCPMe to determine if TCPMe is biotransformed to TCPMeOH. After 21 d of feeding three rainbow trout were sacrificed and analyzed for TCPMe and TCPMeOH.

Organchlorine compound analysis

Tissue samples were weighed, frozen, freeze-dried, and homogenized in toluene. Polychlorinated biphenyl 30 was added to samples before the extraction step as a surrogate recovery

Table 2. Growth parameters (mean ± 1 standard error) of juvenile rainbow trout exposed to 25 organochlorines (significant differences [*t* test, $p < 0.05$] in body and liver growth rates between treatments are indicated by capital letters)

Treatment	Growth rate[a]		% Lipid		Liver somatic index		% Mortality
	Body (10^{-3}/d)	Liver (10^{-3}/d)	Day 30	Day 190[b]	Day 30	Day 190	
Control	12.9 ± 1.3 (0.75)[A]	9.8 ± 1.4 (0.61)[B]	3.4 ± 0.5	2.5 ± 0.4	1.6 ± 0.2	1.0 ± 0.1	0
Low	12.6 ± 1.4 (0.73)[A]	9.3 ± 1.3 (0.62)[B]	3.4 ± 0.3	5.9 ± 0.3	1.5 ± 0.1	0.9 ± 0.1	7.7
High	10.4 ± 1.5 (0.60)[A]	8.0 ± 1.5 (0.47)[B]	4.1 ± 0.6	6.3 ± 2.7	1.5 ± 0.1	1.1 ± 0.2	7.7

[a] The growth rates were calculated using the equation ln weight = $a + b$ time (d), where b is the growth rate (coefficient of determination for the model is shown in parentheses).
[b] Lipid percentages were similar between the three treatments throughout the experiment with the exception of day 190.

standard. Samples were centrifuged and the toluene supernatant was decanted and the toluene extraction was repeated and combined with the first extraction. The toluene was exchanged for 2 ml of hexane, and 250 μl was used to determine lipids gravimetrically. Lipids were removed from the sample by gel permeation chromatography (GPC). The GPC columns (inner diameter, 29.5 mm; length, 400 mm, reservoir, 500 ml) were packed with 60 g (dry weight) of 200- to 400-mesh Bio-Beeds® S-X3 beads (Bio-Rad Laboratories, Hercules, CA, USA). The column was eluted with 300 ml of dichloromethane (DCM): hexane; the first 125 ml contained lipids and was discarded. The lipid-free eluate, containing the OCs, was evaporated to 1 ml and applied to a Florisil (Fisher Scientific, Whitby, ON, Canada) column (8 g, 1.2% deactivated). The OCs were recovered by consecutive elution with 35 ml hexane (fraction 1 [F1]), 38 ml of 85% hexane:15% DCM (fraction 2 [F2]), and 52 ml of 50% hexane:50% DCM (fraction 3 [F3]). Fraction 1 contained 5% of the Cl_7-CHB; 90% of the HCBz, mirex, Cl_8-CHB, and Cl_9-CHB; and 100% of all the PCBs. Fraction 2 contained 10% of the HCBz, mirex, Cl_8-CHB, and Cl_9-CHB; 95% of the Cl_7-CHB; and 100% of the TCPMe. Fraction 3 contained 100% of the TCPMeOH. All fractions were evaporated, transferred to 2,2,4-trimethyl pentane, and evaporated to approximately 100 μl. Aldrin was added as a volume corrector.

Water samples (1 L) were extracted with 300 ml of DCM: hexane in a 2-L glass flask. The solvent was decanted, evaporated to 1 ml, and applied to a Florisil column using the same methods that were used for the fish tissue.

Samples were analyzed on a Varian 3600 gas chromatograph (GC; Varian Canada, Mississauga, ON, Canada) equipped with a 60-m DB-5 column (Chromatographic Specialties, Brockville, ON, Canada) and an ^{63}Ni-electron capture detector (ECD) [30]. The carrier gas was H_2 and N_2 was used as the make-up gas for the ECD.

Octanol/water partition coefficient

The K_{ow}s of PCB congeners were obtained from Hawker and Connell [31], of HCBz from Mackay et al. [32], and of mirex from Suntio et al. [33]. No published data are available on the K_{ow}s of TCPMe, TCPMeOH, or any toxaphene congeners; therefore, fragment constants were used to estimate their K_{ow}s [34]. The K_{ow}s of TCPMe and TCPMeOH were determined by adding and subtracting the appropriate fragments from dichlorodiphenyltrichloroethane (DDT). The K_{ow}s of the toxaphene congeners were determined by adding all the fragments and correcting by appropriate rules [34].

Data analysis

Growth rates were determined by fitting all fish and liver weight data to an exponential model (ln fish weight = $a + b \times$ time [d]; where a is a constant and b is the growth rate) [35]. All concentrations were corrected for growth dilution by multiplying the fish concentrations by a factor of ($1 + b \times$ time). Many of the compounds used (TCPMe, Cl_8-CHB, Cl_9-CHB, mirex, HCBz, and PCBs 52, 101, 118, 128, 138, 153, and 187) were found to have significant concentrations in the nonspiked food (Table 1) and control rainbow trout. For chemicals that were at steady state between food and control fish, and did not significantly increase in the control fish over the course of the experiment, a mean concentration was determined in the control fish and subtracted from the exposed fish concentration. For OCs that showed a significant increase in concentration in the control fish over the length of the experiment, concentrations in the exposed fish were corrected by subtracting the mean concentration of the control fish for the same collection day.

Depuration rate constants (k_ds) were determined by fitting the data to a first order decay curve (ln concn. = $a + b \cdot$time [d], were a is a constant and b is the k_d). The $t_{1/2}$ value is = ln $2/k_d$. Assimilation efficiency (α) was determined by fitting the concentration data to the integrated form of the kinetic rate equation for constant dietary exposure using iterative nonlinear regression [18]

$$C_{fish} = (\alpha F C_{food}/k_d) \times [1 - \exp(-k_d t)]$$

where F is the feeding rate ($F = 0.015$ g food/g of fish/d, lipid corrected), C_{fish} is the concentration in the fish (lipid corrected), C_{food} is the concentration in the food (lipid corrected), and t is the time (d). Equilibrium BMFs were predicted from the equation BMF = $\alpha F/k_d$.

Differences between growth rate constants among treatments, and depuration rates among treatments, were examined by testing the homogeneity of slopes in an analysis of covariance. Student's t test was used to compare pairs of elimination rate and growth rate constants at the $p < 0.05$ level of significance.

RESULTS AND DISCUSSION

Effects

No significant differences in whole fish or liver growth rates were found between control and exposure juvenile rainbow trout populations (Table 2). Liver somatic indices (LSI) were similar between treatment populations on days 30 and 190, although they decreased between days 30 and 190 (Table 2). Three out of 39 rainbow trout died in the low- and high-exposure treat-

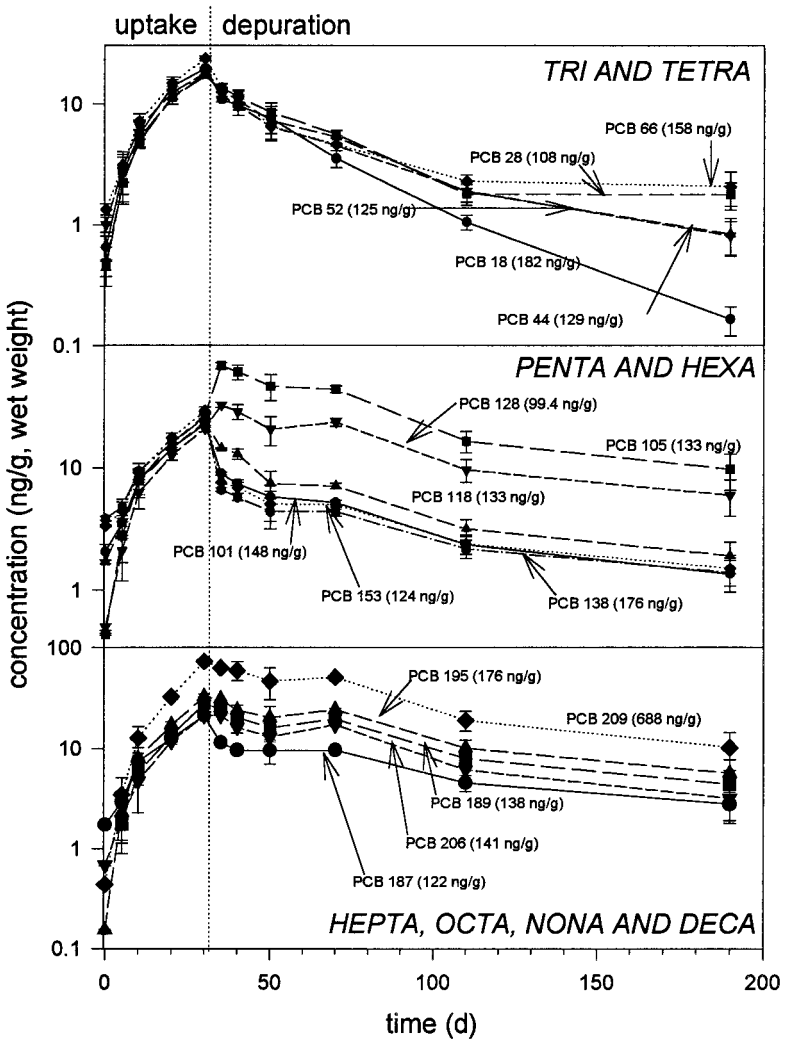

Fig. 1. Accumulation and depuration of 16 polychlorinated biphenyl congeners through dietary exposure to juvenile rainbow trout. Each point is the mean concentration ± 1 standard error of three fish carcasses (minus gastrointestinal tract and liver). Exposure concentrations (wet weight) are provided beside each organochlorine compound.

ments; no fish died in the control population. Lipid percentages in the fish increased with time and were similar between treatment populations on all sampling days except day 190, when lipid levels in the control were lower (Table 2).

Bioaccumulation parameters

All OCs were detected in the carcass after 5 d of exposure to spiked food (Figs. 1 and 2). None of the compounds reached steady state after 30 d (Figs. 1 and 2), and uptake curves and assimilation efficiencies were similar for most compounds (Figs. 1 and 2 and Table 3). Unfortunately, for OCs that had concentrations in the nonspiked food exceeding 1.0 ng/g (wet weight) (TCPMe, mirex, Cl_8-CHB, Cl_9-CHB, PCBs 52, 101, 118, 138, 153, and 187), the results for the lower concentration exposures could not be used. Because significant quantities of OCs were present in the nonspiked food used during the depuration phase, the rainbow trout continued to accumulate OCs during this phase. This resulted in an apparent, slower depuration rate and consequently higher assimilation efficiencies and BMFs that are incorrect. The source of the OCs in the food is probably from fish oils used by the manufacturer in the preparation of food. Concentrations of OCs in the nonspiked food were 17 to 300 times lower than those in the high-exposure food concentrations, and therefore had only a minor influence on the depuration rates from the high exposure.

For OCs not confounded by the presence of concentrations in the nonspiked food, bioaccumulation parameters were in good agreement between the two exposure treatments. Polychlorinated biphenyl 30 recoveries were 75.6 ± 5.9% (mean ± 1 SE), and no corrections were made for these recoveries. No OCs were detectable in the water (<1 pg/L), 1 and 24 h after feeding.

Polychlorinated biphenyls

All of the PCBs used in this study were selected based on the criteria of maximum, or near maximum, meta and para chlorine substitution. These PCBs represent a subset that should have the slowest elimination and greatest bioaccumulation potential [17,18,36]. With the exception of PCB 18, all PCBs in this experiment had BMFs >1. Polychlorinated biphenyls with similar meta and para chlorine substitution have been found to biomagnifiy in laboratory experiments using fish [17,18,21], and in aquatic food chains [4,37]. However, PCB BMFs from this study are as much as a factor of five lower than BMFs determined for adult Lake Ontario lake trout (4.1–5.2 kg) [38]. The difference in PCB BMFs could

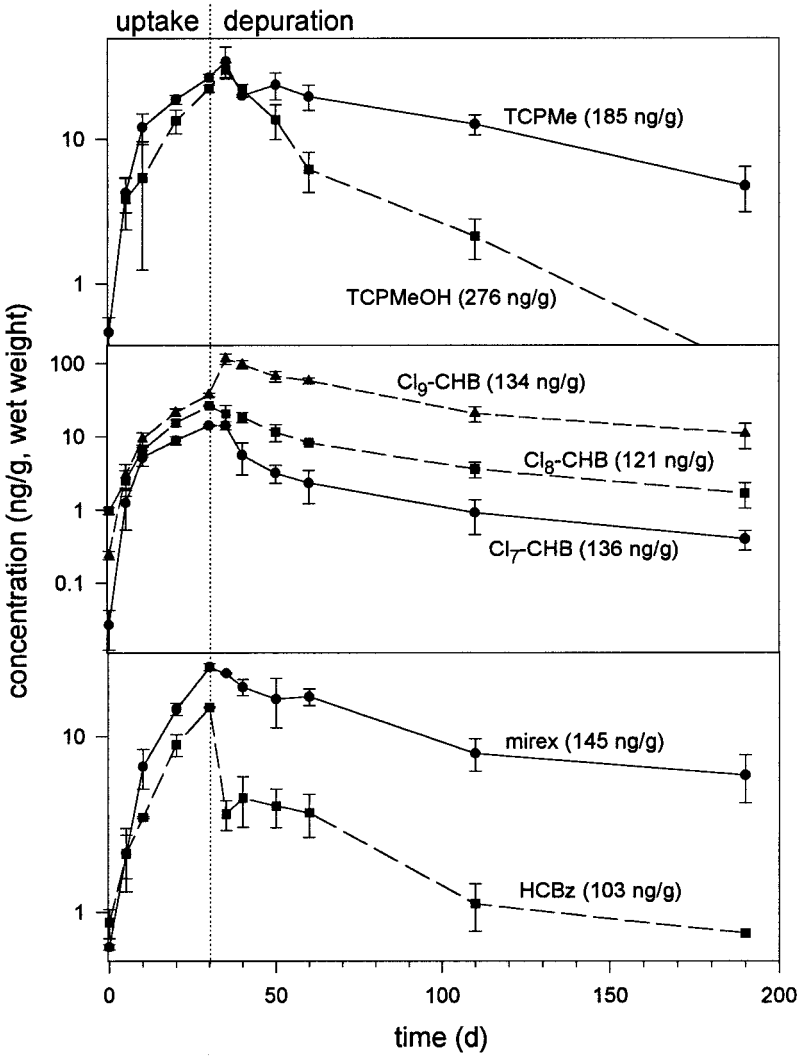

Fig. 2. Accumulation and depuration of tris(4-chlorophenyl)methane (TCPMe), tris(4-chlorophenyl)methanol (TCPMeOH), Cl_7-chlorobornane (CHB), Cl_8-CHB, Cl_9-CHB, hexachlorobenzene (HCBz), and mirex through dietary exposure to juvenile rainbow trout. Each point is the mean concentration ± 1 standard error of three fish carcasses (minus gastrointestinal tract tract and liver). Exposure concentrations (wet weight) are provided beside each organochloric compound.

be due to the small fish used in our study. As discussed below, PCB depuration rates are greater in smaller fish, which would result in lower BMF.

Assimilation efficiencies of PCBs (31–49%) and HCBz (34%) are similar to those reported by Gobas et al. [39] for goldfish and Sijm et al. [17] for guppies, but are much lower than those reported for rainbow trout (PCBs: 63–85% [36]; HCBz: 73–88% [40]). In both of Niimi and Oliver's [36,40] studies, trout were exposed to PCBs and HCBz in herring oil by gavage, which suggests that the high lipid content may have resulted in greater assimilation. In a gavage exposure, all lipid and most of the chemical is absorbed because there is no fecal elimination. Gobas et al. [39] found that increasing the lipid content of the food used for exposure resulted in a decrease in assimilation efficiencies of HCBz and PCBs, which they attributed to greater digestibility of the lower lipid food. Based on the results of Gobas et al. [39], the high lipid diet used in this experiment (14%) would underestimate the assimilation efficiency of these compounds in the environment. The digestibility of the food used to administer the contaminant to the fish must therefore be considered, particularly when comparisons are made to wild fish.

Generally, PCB $t_{1/2}$s increased with chlorine number up to a maximum $t_{1/2}$ for PCBs with 7 or 8 chlorines, but decreased for congeners with 9 and 10 chlorines. Niimi and Oliver [36] concluded that the $t_{1/2}$s of PCB congeners in whole rainbow trout was positively correlated with chlorine number, but also found that $t_{1/2}$s in muscle tissue were not as strongly correlated with chlorine number. In fact, Niimi and Oliver [36] found $t_{1/2}$s to decrease at high chlorine number (more than six chlorines), which they attributed to redistribution of the PCB congeners to other tissues and reduced lipid in the muscle. The K_{ow}, which is influenced by both chlorine number and substitution pattern, was also correlated with the $t_{1/2}$ of the PCB congeners in this study. Coristine et al. [41] concluded that the elimination of PCB congeners in rainbow trout is influenced by hydrophobicity and chlorine position.

Comparing the results reported by Oliver and Niimi [36] and Coristine et al. [41] with this work showed that PCB $t_{1/2}$s in rainbow trout are positively related to rainbow trout size (Fig. 3). Half-lives of similar PCB congeners were consistently highest in the study of Oliver and Niimi [36], who used 900-g rainbow trout, and lowest in the 10-g rainbow trout used

Table 3. Bioaccumulation parameters for 25 organochlorines for dietary exposures using juvenile rainbow trout data for carcass concentrations

Compound[a]	Food concn.[b] (ng/g)	Depuration rate[c] (10^{-2}/d)	$t_{1/2}$[d] (d)	BMF[e]	Assimilation efficiency[f] (%)
Miscellaneous organochlorines					
TCPMe	55	0.4 ± 0.1 (0.45)	178 ± 45	4.6	25 ± 2
	185	1.1 ± 0.1 (0.77)	65 ± 5.9	2.3	36 ± 2
TCPMeOH	34	3.6 ± 0.5 (0.81)	19 ± 2.6	0.6	32 ± 1
	276	3.3 ± 0.4 (0.85)	21 ± 2.5	0.5	23 ± 2
Mirex	21	1.7 ± 0.3 (0.77)	42 ± 7.4	1.8	41 ± 3
	145	0.9 ± 0.1 (0.70)	78 ± 8.7	2.9	37 ± 2
HCBz	14	1.6 ± 0.2 (0.77)	43 ± 5.4	2.3	50 ± 7
	103	1.7 ± 0.3 (0.60)	42 ± 7.4	1.4	34 ± 2
Toxaphene congeners					
Cl_7-CHB	21	1.6 ± 0.3 (0.61)	43 ± 8.1	0.8	18 ± 2
	136	2.2 ± 0.4 (0.63)	32 ± 5.8	0.9	28 ± 2
Cl_8-CHB	18	0.7 ± 0.1 (0.84)	95 ± 14	4.9	49 ± 5
	121	1.6 ± 0.2 (0.85)	43 ± 5.4	2.1	51 ± 3
Cl_9-CHB	17	0.8 ± 0.1 (0.86)	83 ± 10	4.6	53 ± 3
	134	1.7 ± 0.2 (0.87)	42 ± 4.9	2.6	63 ± 4
Polychlorinated biphenyls					
PCB 18	29	2.0 ± 0.2 (0.79)	36 ± 3.6	1.2	32 ± 2
	182	2.9 ± 0.1 (0.96)	24 ± 0.8	0.7	31 ± 2
PCB 28	16	1.6 ± 0.2 (0.85)	44 ± 5.5	2.1	45 ± 4
	108	1.5 ± 0.2 (0.77)	46 ± 6.1	1.8	39 ± 2
PCB 44	18	1.4 ± 0.1 (0.87)	49 ± 3.5	2.1	42 ± 7
	129	1.8 ± 0.1 (0.90)	38 ± 2.1	1.3	34 ± 2
PCB 52	22	1.1 ± 0.2 (0.74)	65 ± 12	2.9	43 ± 5
	125	1.8 ± 0.1 (0.91)	39 ± 2.2	1.5	38 ± 2
PCB 66	21	0.9 ± 0.1 (0.83)	82 ± 9.1	4.0	47 ± 6
	158	1.3 ± 0.2 (0.74)	55 ± 8.5	1.9	35 ± 2
PCB 101	20	0.5 ± 0.1 (0.62)	131 ± 26		
	148	1.2 ± 0.1 (0.86)	56 ± 4.7	2.0	37 ± 2
PCB 105	17	1.4 ± 0.2 (0.77)	48 ± 6.9	2.8	55 ± 5
	133	1.4 ± 0.1 (0.85)	50 ± 3.6	2.3	47 ± 3
PCB 118	20	0.7 ± 0.1 (0.65)	103 ± 15	6.0	55 ± 8
	133	1.3 ± 0.1 (0.84)	53 ± 4.1	2.2	42 ± 2
PCB 128	8	0.9 ± 0.1 (0.79)	75 ± 8.3	5.8	75 ± 8
	99	1.2 ± 0.1 (0.79)	59 ± 4.9	2.8	49 ± 3
PCB 138	31	0.5 ± 0.1 (0.59)	139 ± 28	7.1	49 ± 10
	176	1.1 ± 0.1 (0.81)	64 ± 5.8	2.4	38 ± 2
PCB 153	22	0.3 ± 0.1 (0.40)	224 ± 75	16	68 ± 15
	124	1.0 ± 0.1 (0.80)	69 ± 6.9	3.3	48 ± 3
PCB 187	19	0.5 ± 0.1 (0.68)	131 ± 26	6.1	45 ± 8
	122	1.0 ± 0.1 (0.78)	71 ± 7.1	2.8	40 ± 2
PCB 189	19	1.1 ± 0.1 (0.82)	64 ± 5.8	2.0	30 ± 2
	138	1.2 ± 0.2 (0.76)	58 ± 9.7	2.4	42 ± 3
PCB 195	24	1.0 ± 0.1 (0.81)	67 ± 6.7	2.4	34 ± 2
	176	1.1 ± 0.2 (0.76)	61 ± 11	2.4	40 ± 3
PCB 206	20	1.6 ± 0.2 (0.79)	45 ± 5.6	1.6	34 ± 4
	141	1.3 ± 0.2 (0.72)	53 ± 8.2	1.8	34 ± 2
PCB 209	62	1.3 ± 0.2 (0.81)	52 ± 8.0	1.1	21 ± 1
	688	1.3 ± 0.2 (0.74)	52 ± 8.0	1.1	22 ± 2

[a] TCPMe = tris(4-chlorophenyl)methane; TCPMeOH = tris(4-chlorophenyl)methane; HCBz = hexachlorobenzene; CHB = chlorobornane.
[b] Food concentration is wet weight.
[c] Depuration rate constants (k_ds) were calculated using the model ln concentration (lipid weight basis) = $a + b$ (time) for the elimination of toluene-extractable radioactivity for 120 d of depuration (coefficient of determination for the model is shown in parentheses).
[d] The half-life ($t_{1/2}$) is calculated from the equation $t_{1/2} = 0.693/k_d$.
[e] The biomagnification factor (BMF) is calculated from the equation BMF = $\alpha F/k_d$ where F is the feeding rate on a lipid basis.
[f] The assimilation efficiency (α) is determined by fitting the data to the integrated form of the kinetic rate equation for constant dietary exposure using iterative nonlinear regression: $C_{fish} = (\alpha F C_{food}/k_d)\cdot[1 - \exp(-k_d t)]$ where C_{fish} is the concentration in the fish (lipid basis and growth corrected), C_{food} is the concentration in the food (on a lipid basis), and t is the time of uptake (d).

in this work. Sijm and van der Linde [42] have also observed a positive correlation between PCB $t_{1/2}$ and fish size.

Mirex and HCBz

Mirex had a greater $t_{1/2}$ in juvenile rainbow trout than did HCBz, which agrees with similar studies on HCBz and mirex in rainbow trout [43]. Based on BMFs >1 (Table 3), both HCBz and mirex would biomagnify in aquatic food chains. Clark and Mackay [21] concluded that mirex would biomagnify but that significant biomagnification of HCBz is unlikely based on dietary uptake studies using guppies. Mirex has been found to biomagnify in aquatic food chains [44].

Tris(4-chlorophenyl)methane and TCPMeOH

The depuration rate of TCPMeOH was significantly greater than that of TCPMe; TCPMe was much more persistent in

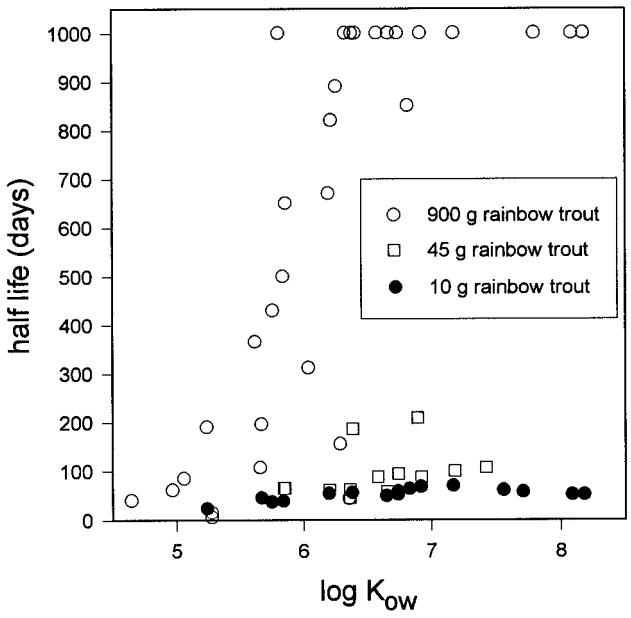

Fig. 3. Half-lives of polychlorinated biphenyl (PCB) congeners in rainbow trout versus log octanol/water partition coefficient (K_{ow}) for three sizes of rainbow trout ○, 900 g from [2] □, 100 g [41] ●, 10 g (this work). The PCB K_{ow}s are from [31].

juvenile rainbow trout (Table 3). Tris(4-chlorophenyl)methanol differs from TCPMe by a single hydroxyl group on the methane carbon, which may explain the significant differences in their bioaccumulation parameters. The assimilation efficiency of TCPMeOH (α = 23–32%) was slightly less than that of TCPMe (α = 36%), although this could be an artifact of the higher depuration rate of TCPMeOH. Tris(4-chlorophenyl)methane had a BMF >1, implying that it may biomagnify in aquatic food chains. The BMFs of TCPMeOH were 0.5 and 0.6, which suggests that this compound will not biomagnify but food chain transfer may still play an important role in the accumulation of TCPMeOH by aquatic organisms. This contrasts with the results of de Boer et al. [24], who concluded that TCPMeOH biomagnifies, based on concentrations in mussels, cod liver, and marine mammals.

In a separate experiment, TCPMeOH was not detectable in juvenile rainbow trout (liver and GI tract included) that had been exposed to TCPMe for 21 d and had accumulated TCPMe to high concentrations (~1,000 ng/g). Jarman et al. [22] hypothesized that TCPMeOH found in birds and mammals could be a derivative of either TCPMe or a chemically related compound not identified. Although our work does not rule out the biotransformation of TCPMe to TCPMeOH in birds and mammals, it suggests that salmonids cannot perform this biotransformation.

Toxaphene congeners

Toxaphene has been observed to biomagnify in aquatic food webs [26,27]; however, data is available on the bioaccumulation of individual toxaphene congeners. The data of Glassmeyer et al. [25] suggest that higher chlorinated toxaphene homologues (eight and nine chlorines) biomagnify in Great Lakes lake trout, but that lower chlorinated homologue groups do not. Bidleman et al. [23] reported that more highly chlorinated (octachloro- and nonachloro-) toxaphene congeners were selectively accumulated by Arctic biota. This is in agreement with the BMFs calculated for Cl_8-CHB (octachloro-) and Cl_9-CHB (nonachloro-) in rainbow trout (BMFs = 2.1 and 2.6, respectively) in this study. It is also in general agreement with the BMF of 0.8 to 0.9 for Cl_7-CHB, which suggests that toxaphene congeners with fewer than eight chlorines are not likely to biomagnify in aquatic food webs, although food will still play an important role in the accumulation of TCPMeOH by aquatic organisms. Other heptachloro-toxaphene congeners with different chlorine substitution patterns may have greater accumulation than this Cl_7-CHB. The congeners Cl_8-CHB and Cl_9-CHB have BMFs similar to PCBs with five chlorines, but have $t_{1/2}$s closer to PCBs with only four chlorines.

Bioaccumulation parameter–K_{ow} *relationships*

Bioaccumulation parameter–K_{ow} relationships were developed from the high-concentration treatment data only due to problems with the lower concentration treatment (discussed above). Bioaccumulation parameters ($t_{1/2}$, BMF, and assimilation efficiency) were log transformed for comparisons with log K_{ow}.

The OCs used to develop these relationships are slowly, if at all, metabolized in fish. Organochlorine compounds with $t_{1/2}$s and assimilation efficiencies that are lower than predicted are probably biotransformed by the fish [45,46]. For example, Fisk et al. [47], using an experimental protocol nearly identical to this work, reported a $t_{1/2}$ of approximately 45 d for a chlorinated *n*-alkane ($C_{16}H_{31}Cl_3$) in juvenile rainbow trout despite a log K_{ow} of 6.9. Based on our $t_{1/2}$–K_{ow} relationship, $C_{16}H_{31}Cl_3$ has a $t_{1/2}$ approximately one half of what is expected for an nonmetabolized OC with this K_{ow}. However, caution must exercised when using data from other studies. Fish species, fish size, and experimental protocol may all influence laboratory-derived bioaccumulation parameters [42,48].

Depuration rate constant–K_{ow} *relationships*

Depuration rate constants (k_ds) of the OCs were strongly related to the K_{ow}s. This is exemplified by the curvilinear relationship between log $t_{1/2}$ ($t_{1/2}$ = 0.693/k_d) and log K_{ow} (R^2 = 0.85, $p < 0.001$) (Fig. 4), with a maximum $t_{1/2}$ for OCs of log K_{ow} = 7.2. More rapid depuration above log K_{ow} > 7 is difficult to rationalize because greater K_{ow} implies less partitioning out of the fish lipids, and therefore greater retention in the tissue and longer $t_{1/2}$s. The tissue used to determine concentrations and bioaccumulation parameters did not include the GI tract; therefore, superhydrophobic OCs accumulated in lipid or sorbed to the epithelial cells of the GI tract and eliminated through subsequent cell turnover cannot explain this phenomena. Fox et al. [14] also observed a similar curvilinear relationship between K_{ow} and k_ds of PCBs by zebrafish (*Brachydanio rerio*), but made no attempt to explain this relationship. Gobas et al. [49] observed a linear relationship between log k_d and log K_{ow} for OCs with log K_{ow}s between 4 and 7, and a leveling off of elimination rates for OCs of log K_{ow} > 7.0. Although Gobas et al. [49] discuss the importance of fecal elimination as an additional elimination pathway of superhydrophobic OCs (log K_{ow} > 7.0) and its affect on BCF, no explanation was offered for the leveling off of depuration rates. A number of explanations are possible for the curvilinear relationship observed between k_d and K_{ow}.

Insufficient data. Only four compounds were used that had log K_{ow} > 7.2, and the curve could be interpreted as leveling off and not decreasing. However, PCBs 206 and 209 (log K_{ow} = 8.09 and 8.18, respectively) had significantly more rapid k_d than mirex and PCBs 153 and 187 (log K_{ow} = 6.9, 6.2, and

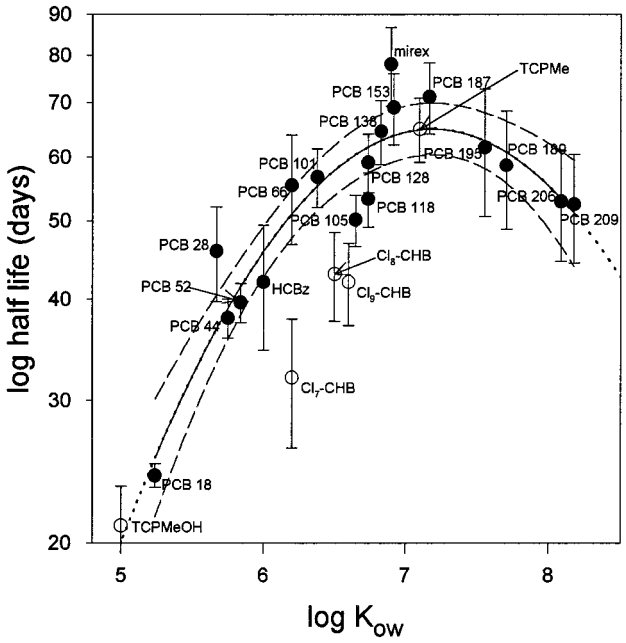

Fig. 4. Log half-life ($t_{1/2}$) of hydrophobic organochlorine compounds (OCs) in juvenile rainbow trout versus log octanol/water partition coefficient (K_{ow}). Solid circles represent OCs with published K_{ow} values. Open circles represent OCs that do not have published K_{ow} values but were estimated using the K_{ow} fragment-constant method [34]. The quadratic regression (solid line) was derived from the OCs that have published K_{ow} values (closed circles) ($\log t_{1/2} = -3.7 + (1.5 \cdot \log K_{ow}) - (0.1 \cdot \log K_{ow}^2)$, $r^2 = 0.85$, $p < 0.001$). The dashed line represents the 95% confidence intervals, and the dotted line carries the data past the last points to the axis. The K_{ow}s of polychlorinated biphenyls were taken from [31], of hexachlorobenzene (HCBz) were from [32], and of mirex were from [33].

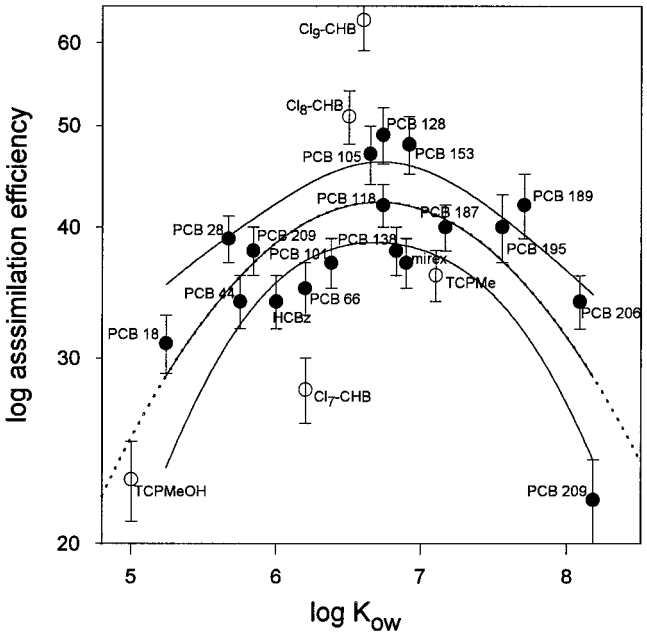

Fig. 5. Log assimilation efficiency of hydrophobic organochlorine compounds (OCs) by juvenile rainbow trout versus log octanol/water partition coefficient (K_{ow}). Solid circles represent OCs with published K_{ow} values. Open circles represent OCs that do not have published K_{ow} values but were estimated using the K_{ow} fragment-constant method [34]. The quadratic regression (solid line) was derived from the OCs that have published K_{ow} values (closed circles) (log assimilation efficiency $= -1.8 + (1.0 \cdot \log K_{ow}) - (0.08 \cdot \log K_{ow}^2)$, $r^2 = 0.53$, $p = 0.004$). The dashed line represents the 95% confidence intervals, and the dotted line carries the data past the last points to the axis. The K_{ow}s of polychlorinated biphenyls were taken from [31], of hexachlorobenzene (HCBz) were from [32], and of mirex were from [33].

7.2, respectively) (analysis of covariance [ANCOVA], $p < 0.05$). The k_d of octachlorodibenzo-*p*-dioxin (OCDD) and octachlorodibenzofuran (OCDF), compounds with very high K_{ow}s (8.2 and 8.0, respectively [32]), in juvenile rainbow trout were also much more rapid than the maximum rates observed in this study [50,51]. Therefore, it seems unlikely that insufficient data are the cause of the curvilinear relationship between k_d and K_{ow}, although it may influence the magnitude of equation coefficients.

Inaccurate K_{ow}. Two problems arise for the K_{ow} of superhydrophobic compounds. Measurement of an accurate K_{ow} of very hydrophobic compounds is difficult due to their extremely low water solubility [52], and estimates of high K_{ow}s are considered less accurate for compounds with higher K_{ow} [53]. Also, at high K_{ow}, a loss of linearity occurs between K_{ow} and lipidwater partition coefficients [45]. Chessells et al. [54] concluded that lower than expected BCFs of superhydrophobic OCs were partially due to relative lower lipid solubility that is not reflected in K_{ow}. Because our relationships were developed mainly from PCB data it is unlikely that inaccurate K_{ow}s could account completely for the curvilinear relationship between $t_{1/2}$ and K_{ow}, but they may have influenced the magnitude of equation coefficients.

Disequilibrium between fish compartments (experiment length). The time to reach steady state concentrations between compartments in the rainbow trout should be negatively correlated with water solubility. Therefore, by the end of the uptake phase, superhydrophobic OCs ($\log K_{ow} > 7$; e.g., PCBs 206 and 209) would be further from steady state between compartments of the fish than moderately hydrophobic OCs ($\log K_{ow} = 5$–7). These superhydrophobic OCs may not have reached slower clearing compartments during the uptake phase (30 d), and may therefore be in faster clearing compartments when compared with moderately hydrophobic OCs. Faster clearing compartments could include lipids and proteins of the blood system or even cell membranes. With a greater percentage of total burden in faster clearing compartments, superhydrophobic OCs are more available for elimination during the depuration phase compared to moderately hydrophobic OCs ($\log K_{ow} = 5$–7). Conversely, OCs with intermediate K_{ow}s may have a greater percentage of their total body burden in slower clearing compartments and therefore may be less available for elimination than superhydrophobic OCs. Sijm et al. [17] observed a negative linear relationship between PCB elimination rates, including those of PCB 209, and K_{ow} in guppies that had been exposed for 210 d. Also, de Boer et al. [24] observed no apparent elimination of higher chlorinated PCB congeners in wild populations of eels (*Anguilla anguilla*). Therefore, the length of exposure in bioaccumulation experiments using superhydrophobic OCs can be an experimental artifact, and may influence their k_ds and $t_{1/2}$s. Also, storage in various lipid compartments may vary depending on the method of exposure (feeding, gavage, or intraperitoneal injection) and should be considered when comparing results of different experiments.

Assimilation efficiency–K_{ow} *relationships*

Assimilation efficiency had a significant curvilinear relationship with K_{ow} ($R^2 = 0.56$, $p = 0.04$) (Fig. 5), but this

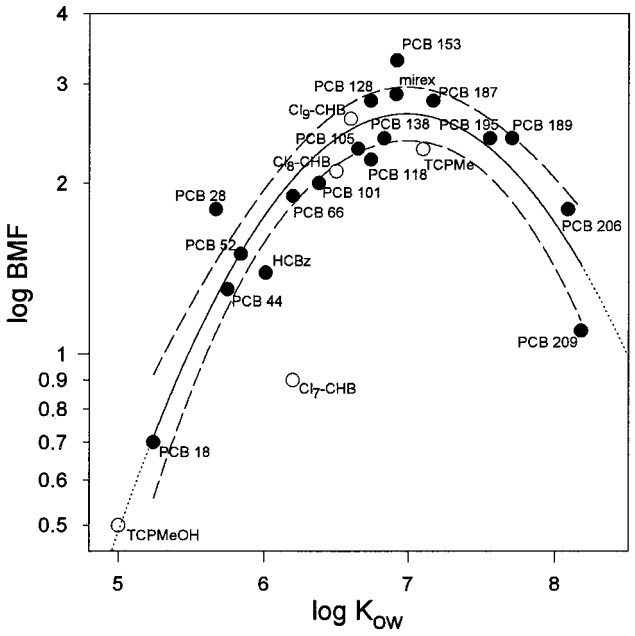

Fig. 6. Log biomagnification factor (BMF) of hydrophobic organochlorine compounds (OCs) in juvenile rainbow trout versus log octanol/water partition coefficient (K_{ow}). Solid circles represent OCs with published K_{ow} values. Open circles represent OCs that do not have published K_{ow} values but were estimated using the K_{ow} fragment-constant method [34]. The quadratic regression (solid line) was derived from the OCs that have published K_{ow} values (closed circles) ($\log BMF = -8.7 + (2.6 \cdot \log K_{ow}) - (0.2 \cdot \log K_{ow}^2)$, $r^2 = 0.84$, $p < 0.001$). The dashed line represents the 95% confidence intervals, and the dotted line carries the data past the last points to the axis. The K_{ow}s of polychlorinated biphenyls were taken from [31], of hexachlorobenzene (HCBz) were from [32], and of mirex were from [33].

relationship was not as strong as the $t_{1/2}$–K_{ow} and BMF–K_{ow} relationships. This result suggests that hydrophobicity does not have as strong an influence on uptake and assimilation as it does on $t_{1/2}$ and BMF. Bruggeman et al. [18] observed that the uptake rates of di-, tri-, and tetra-PCBs from food by goldfish were similar. Also, Gobas et al. [19] reported that absorption efficiencies for a range of hydrophobic OCs were constant up to a log $K_{ow} = 7$, but decreased for OCs of greater K_{ow}. Therefore, the bioaccumulation of OCs with log K_{ow}s < 7 are not as strongly influenced by the uptake rate or assimilation efficiency as they are by the k_d. However, at very high K_{ow}s (>7), a reduction in assimilation efficiency occurs, perhaps due to reduced bioavailability in the food or steric hindrance in crossing biological membranes.

Biomagnification factor–K_{ow} *relationships*

A significant curvilinear relationship was found between log BMF and log K_{ow} ($R^2 = 0.84$, $p < 0.001$) (Fig. 6), which is more easily rationalized than the $t_{1/2}$–K_{ow} relationship. At steady state the BMF is equal to the uptake rate divided by the k_d. Because the uptake is essentially constant, and the elimination rate decreases, for OCs with log K_{ow}s < 7, the BMF should increase with K_{ow} up to a log $K_{ow} = 7$. Oliver and Niimi [4] reported such a relationship for OCs, including PCBs, in Lake Ontario salmonids. For OCs with log K_{ow}s $>$ 7.0, lower assimilation efficiencies or lower uptake rates give rise to lower BMFs. Also, the k_ds increase above log $K_{ow} = 7$, further reducing BMFs.

A curvilinear relationship has been observed numerous times with BCF–K_{ow} relationships, where BCFs for compounds with log $K_{ow} > 7$ fall below the expected 1:1 relationship [1,14,49,55,56]. The curvilinear BCF–K_{ow} phenomenon has been attributed to a number of factors including overestimation of bioavailable water concentrations (or reduced bioavailability) [1,49], reduced membrane passage of the larger superhydrophobic chemicals [1,57], inaccurate K_{ow}s [54], and elimination into feces [49]. By exposing fish to dietary concentrations we have eliminated any problems associated with water uptake pathways, but the issues of disequilibrium, reduced membrane passage, inaccurate K_{ow}s, and elimination into feces remain.

Comparison of estimated K_{ow}*s: Fragment constant versus bioaccumulation parameter*–K_{ow} *relationships*

Half-life–K_{ow} relationships were used to predict the K_{ow} of TCPMe, TCPMEOH, Cl_7-CHB, Cl_8-CHB, and Cl_9-CHB. The K_{ow}s for TCPMe and TCPMeOH predicted by this relationship agreed with K_{ow}s determined by the fragment constant method (Table 4). This suggests that there is little, or no, metabolism of TCPMe or TCPMeOH by the juvenile rainbow trout. The K_{ow}s determined by the fragment-constant method for Cl_7-CHB, Cl_8-CHB, and Cl_9-CHB are lower than those predicted by the $t_{1/2}$–K_{ow} relationship. The fragment-constant method possibly overestimated the K_{ow}s of the toxaphene congeners. Greater uncertainty occurs with the fragment-constant method for highly hydrophobic compounds [34]. Alternatively, these toxaphene congeners are metabolized by the rainbow trout,

Table 4. Estimated log octanol/water partition coefficients (K_{ow}s) of tris(4-chlorophenyl)methane (TCPMe), tris(4-chlorophenyl)methanol (TCPMeOH), Cl_7-chlorobornane (CHB), Cl_8-CHB, and Cl_9-CHB using the fragment-constant method [54] and bioaccumulation parameter–K_{ow} relationships (Figs 4, 5, and 6). Because the bioaccumulation–K_{ow} relationships are curvilinear, two estimates have been given were appropriate[a]

Organochlorin compound	Fragment-constant method	$t_{1/2}$–K_{ow} relationship	α–K_{ow} relationship	BMF–K_{ow} relationship
TCPMe	7.1	6.8 or 7.6	6.4 or 7.7	6.4 or 7.5
TCPMeOH	5.0	5.0	4.9	5.0
Cl_7-CHB	6.2	5.4	5.4	5.2
Cl_8-CHB	6.5	5.8	6.7[b]	6.9 or 7.1
Cl_9-CHB	6.6	5.8	6.7[b]	6.3 or 7.7

[a] $t_{1/2}$ = half-life; α = assimilation efficiency; BMF = biomagnification factor.
[b] The assimilation efficiencies of Cl_8-CHB and Cl_9-CHB are greater than the α–K_{ow} equation and therefore the K_{ow} estimate represents the greatest α.

and therefore the $t_{1/2}$–K_{ow} relationship cannot accurately predict their K_{ow}. Based on a greater distance from the $t_{1/2}$–K_{ow} curve, Cl_7-CHB is more readily metabolized than Cl_8-CHB and Cl_9-CHB.

CONCLUSIONS

We have reported the first data on laboratory-derived bioaccumulation parameters of TCPMe, TCPMeOH, and three toxaphene congeners in juvenile rainbow trout. These compounds have high BMFs, confirming observations from field data. Dietary bioaccumulation parameter–K_{ow} relationships, derived from 18 recalcitrant OCs, revealed that assimilation efficiency, $t_{1/2}$, and BMF all have curvilinear relationships with K_{ow}. Organochlorine compounds that a have a log K_{ow} of approximately 7, and are not biotransformed, had the greatest persistence and biomagnification in fish. The curvilinear relationship of $t_{1/2}$ and K_{ow} suggests that internal kinetics of contaminants in fish play a significant role in their fate, and that the length of exposure of superhydrophobic OCs (log $K_{ow} > 7$) will affect their k_ds and $t_{1/2}$s. Variation in the $t_{1/2}$s of PCB congeners in various sizes of rainbow trout suggests that additional research is needed on the influence of fish size on bioaccumulation parameters. Also, differences in the composition of food effect the assimilation of PCB congeners in the digestive tract of fish but relationships are not clear and require further study.

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