Bioaccumulation and biotransformation of polychlorinated *n*-alkanes (PCAs) by juvenile rainbow trout (*Oncorhynchus mykiss*): Relationships with carbon chain length, chlorine content and K_{ow}

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Introduction

Polychlorinated alkanes (PCAs) are a class of high molecular weight organochlorines used for a variety of industrial processes. Commercial PCA formulations, also known as chlorinated paraffins (CPs), are complex mixtures of chlorinated *n*-alkanes with carbon chain lengths between 10 and 30 and chlorine contents between 30 and 70% by weight. PCAs are one of the last high molecular weight organochlorines and one of the largest groups of chlorinated hydrocarbons produced and used in North America and western Europe [1]. PCAs have physical chemical properties, e.g., low water solubilities and vapor pressures [2,3], which are similar to other high molecular weight organochlorine pollutants (e.g., PCBs and DDT). PCAs have been found in biotic and abiotic samples from North America and western Europe, including the arctic, and in many cases exhibit the highest concentration of any organochlorine measured [1].

The objective of this research was to develop relationships between PCA bioaccumulation and biotransformation with carbon chain length, chlorine content and K_{ow} using data generated with juvenile rainbow trout (*Oncorhynchus mykiss*) and a series of PCA standards with varying carbon chain length (C_{10}, C_{11}, C_{12}, C_{14}, C_{16} and C_{18}) and chlorine content (4 to 13 Cl atoms)[4,5,6].

Methods and Material

PCAs synthesis, extraction and analysis methods, data analysis and experimental protocols have been described previously [4,5,6]. In brief: Juvenile rainbow trout (*Oncorhynchus mykiss*)(initial weights ~ 5 g) were exposed to PCA-spiked food for 40 days followed by 160 days of clean food. Three fish were sampled from each treatment for PCA determination on days 5, 10, 20, 30 and 40 of the uptake period, and days 5, 10, 20, 40, 80 and 160.

Results and Discussion

PCA half lives ranged from 7 days for a $C_{10}H_{17}Cl_5$ to 91 day for $C_{18}H_{31}Cl_7$, and are in the range of half lives reported for other hydrophobic organochlorines in juvenile rainbow trout [7,8,9]. Biomagnification factors (BMFs) ranged from 0.4 for a $C_{10}H_{17}Cl_5$ to 3.0 for $C_{14}H_{25}Cl_5$. PCA half lives and BMFs were positively correlated with # carbons, # chlorines, # (carbons + chlorines) and log K_{ow} (Figure 1).

Comparing PCA half lives with the half life-log K_{ow} relationship reported in Fisk et al. [10] provides insight into biotransformation and provides an estimate of the biotransformation rate (Figure 2). The half life-log K_{ow} relationship was generated with a series of recalcitrant chemicals and should represent the maximal half life, or minimal depuration rate, for chemicals with log K_{ow}s between 4.5 and 7.5. Subtracting the minimal depuration rate of the PCAs, calculated form their log K_{ow} and the relationships of Fisk et al. [10], from the measured depuration rates provides an estimate of

biotransformation rate. Biotransformation rates approaching zero, or that are negative, suggest minimal or no biotransformation.

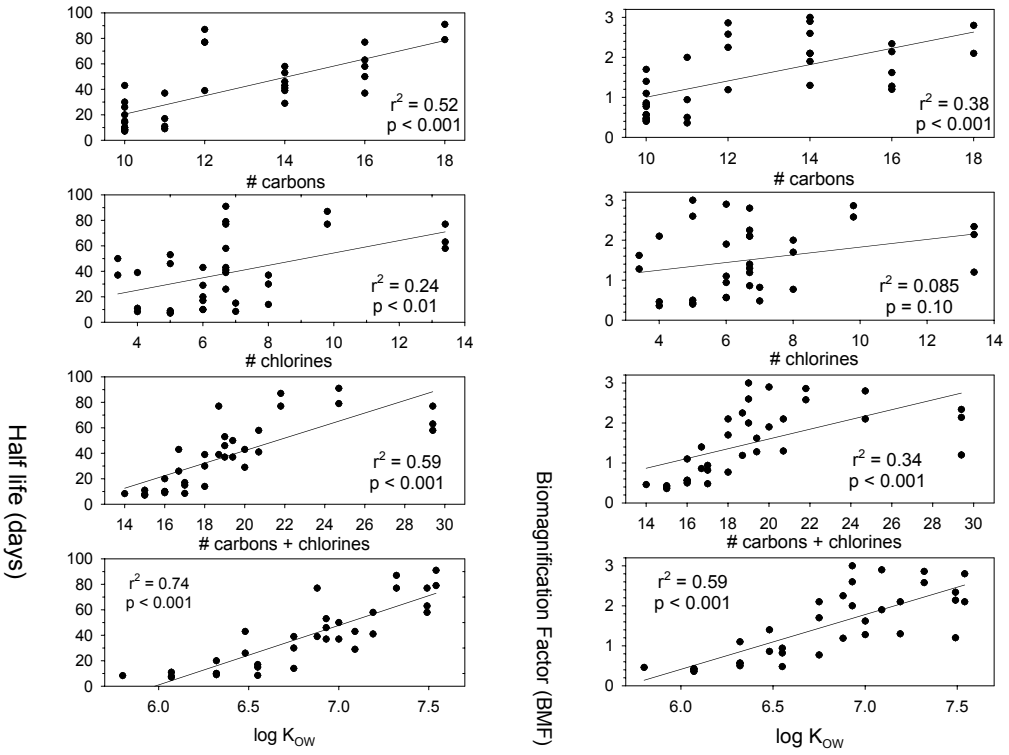

Figure 1: Relationships between PCA $t_{1/2}$ with C # ($t_{1/2}$ = -52 + 7.2 * #C, n = 34, r^2 = 0.52, p < 0.001), Cl # ($t_{1/2}$ = 5.6 + 4.9 * #Cl, n =34, r^2 = 0.24, p = 0.004), total C and Cl # ($t_{1/2}$ = -56 + 4.9 * #(C+Cl), n = 34, r^2 = 0.59, p < 0.001), and log K_{ow} ($t_{1/2}$ = -280 + 47 * log K_{ow}, n = 34, r^2 = 0.74, p < 0.001) and PCA BMFs (assuming 50% assimilation efficiency) with C # (BMF = -1.0 + 0.20 * #C, n = 34, r^2 = 0.38, p < 0.001), Cl # (BMF = 0.86 + 0.096 * #Cl, n =34, r^2 = 0.084, p = 0.095), total C and Cl # (BMF = -0.84 + 0.12 * #(C+Cl), n = 34, r^2 = 0.34, p < 0.001), and log K_{ow} (BMF = -7.8 + 1.4 * log K_{ow}, n = 34, r^2 = 0.59, p < 0.001).

Most PCAs were biotransformed to some extent by the rainbow trout, with biotransformation decreasing with increasing carbon chain length and chlorine content (Figure 3). A curvilinear relationship using total number of carbon and chlorine atoms explained the greatest percentage of variation in the biotransformation rate. Log K_{ow} also explained a large percentage of the variation in biotransformation rate. The relationship between biotransformation rate and total carbon and chlorine atoms suggest that PCAs with a total number of carbons and chlorine atoms of approximately 21 to 30 are not biotransformed (BTR ≈ 0) by juvenile rainbow trout, and likely fish in general. Decreases in biotransformation of PCAs with carbon chain length and chlorine content have been observed with birds and mammals [11,12].

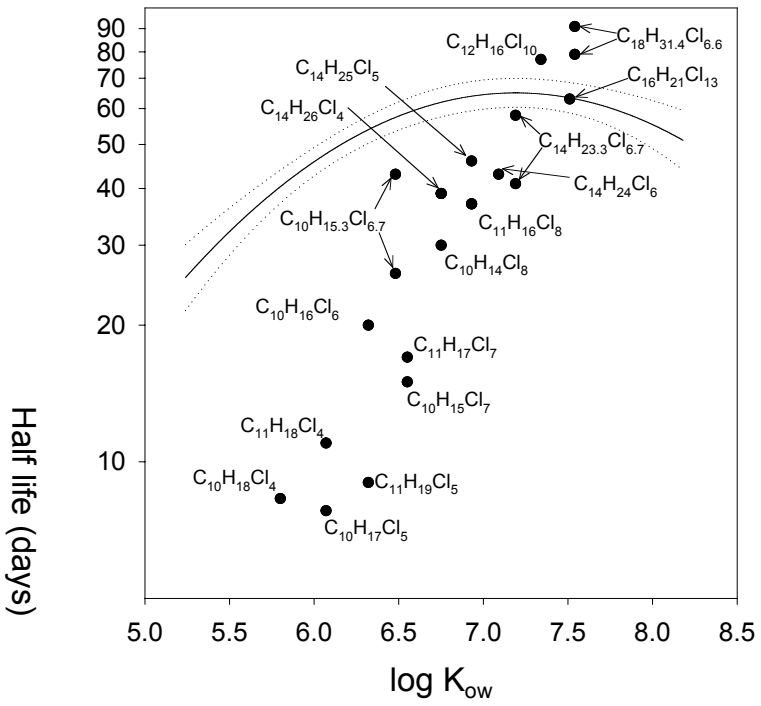

Figure 2: Log half life of PCAs in juvenile rainbow trout vs log K_{ow}. Log K_{ow} values for the PCAs are from Sijm and Sinnige (1996). The quadratic regression (solid line) is from Fisk et al. (1998b) and represents a series of recalcitrant OCs that are not metabolized by juvenile rainbow trout (no symbols; $\log t_{1/2} = -3.7 + (1.5 \cdot \log K_{ow}) - (0.1 \cdot \log K_{ow}^2)$, $r^2 = 0.85$, $p < 0.001$). The dashed line represents the 95% confidence intervals.

PCAs are readily accumulated from food by fish and PCA bioaccumulation is strongly influenced by carbon chain length and to a lesser extent chlorine content. The number of carbons and chlorine influence PCA bioaccumulation through control of physical chemical properties and biotransformation. Increasing carbon chain length and chlorine content result in greater bioaccumulation of PCAs by reducing partition-based (i.e., greater K_{ow} results in lower elimination by diffusion) and metabolic (i.e., biotransformation) elimination processes. Due to the high bioaccumulation potential and low biotransformation rates of medium (C_{14-17}) and long (C_{18-30}) carbon chain PCAs and highly chlorinated PCAs, additional data on environmental levels of these PCAs in aquatic food chains is warranted.

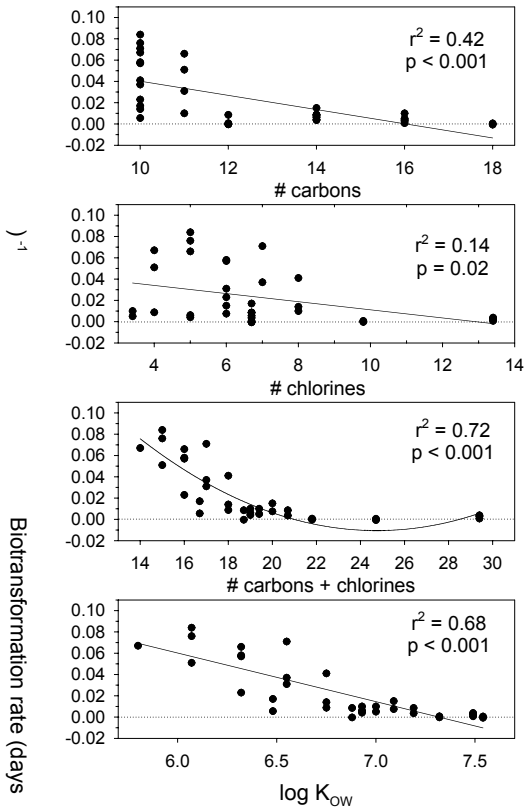

Figure 3: Relationships (solid lines are regressions) between PCA BTR with C # (biot rate = 0.12 - 0.007 * #C, n = 34, r^2 = 0.44, p < 0.001), Cl # (biot rate = 0.062 - 0.004 * #Cl, n =34, r^2 = 0.15, p = 0.024), total C and Cl # (biot rate = 0.49 - (0.039 * #(C+Cl)) + (0.001*(#(C+Cl)^2), n = 34, r^2 = 0.75, p < 0.001), and log K_{ow} (biot rate = 0.37 - 0.049 * log K_{ow}, n = 34, r^2 = 0.71, p < 0.001).

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