Bull. Environ. Contam. Toxicol. (1991) 46:276–283

Bulletin of Environmental Contamination and Toxicology

Accumulation Factors for Eleven Polychlorinated Biphenyl Congeners

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According to the fugacity approach (Mackay 1979), pollutant uptake by an organism is determined by the chemical fugacity differential between the organism and its environment. The Accumulation Factor [AF = (concentration of pollutant in animal tissue, C_t (ng/g dry wt)/animal lipid (%/100))/(concentration of pollutant in sediment, C_s (ng/g dry wt)/sediment total organic carbon, TOC (%/100))] is a simple, fugacity-based model which has been shown to be useful for predicting the bioaccumulation potential of hydrophobic neutral organic compounds in sediment-dwelling animals (Rubinstein et al. 1987; McElroy and Means 1988; Clarke et al. 1988; Ferraro et al. 1990). The theoretical basis for the AF model is discussed in Mackay (1979), Mackay and Paterson (1981, 1982), McFarland (1984), McFarland and Clarke (1986), and Lake et al. (1987). The model assumes chemical equilibrium or steady-state in the animals and the sediments to which they are exposed, no chemical transformation or phase transfer resistance, and chemical partitioning primarily between the organic pool in the sediment and the lipid pool in the animal. If AFs are constant for a given chemical (or group of chemicals), AFs can be used to predict C_t (or an upper bound for C_t) given C_s, TOC and % animal lipids by ratio estimation (Snedecor and Cochran 1967). The applied value of AFs is as a cost-effective, environmentally protective screening tool for evaluating sediment quality (McFarland 1984; Lake et al. 1987; Rubinstein et al. 1987; Ferraro et al. 1990).

Ferraro et al. (1990) tested the constancy of AFs for ten hydrophobic neutral organic compounds by exposing clams (Macoma nasuta) in the laboratory for 28 days to six field-collected sediments varying widely in C_s, TOC, and other chemical and physical characteristics. Sediment and tissue samples from that study were archived and latter analyzed for 11 polychlorinated biphenyl (PCB) congeners. In this paper we report mean and maximum AFs (AF_{max}) for 11 PCB congeners and test the constancy of the AFs across 5 sediments (treatments) by congener and across the 11 congeners by treatment.

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MATERIALS AND METHODS

Details of the experimental methods and design are presented in Ferraro et al. (1990).

Field sediments were collected from two depth strata (0-2 cm, and either 4-8 cm or 8-12 cm, depending upon the depth of penetration of the grab) at each of three stations representing low (station R), intermediate (station S), and high (station D) pollution at 60 m depth in the Southern California Bight. We measured C_s, TOC, water content, and grain size in subsamples from each of the 6 sediment treatments (3 stations x 2 depths). The station S, 8-12 cm deep treatment, however, has been omitted from this study due to analytical problems measuring C_s in the sample from that treatment.

Bioaccumulation tests were conducted with individual clams as replicates with each clam placed in separate 100-mL beakers containing 100 g (wet wt) of test sediment and filtered (1 μm), 25 °/oo salinity, gently aerated overlying seawater. Water temperature was 12±1°C and the light cycle was 12L:12D. The exposures were static with replacement of seawater 3 times per week. A 2 g, freshly frozen (<2 d) wafer of test sediment was added to each replicate beaker after each water change to replenish nutrients and maintain C_s as constant as possible in the surface sediments upon which Macoma feeds. After 28 days of exposure, clams from each treatment were placed in separate aquaria with clean sieved (1 mm) sediment and overlying, aerated, filtered seawater (25 °/oo salinity) for 24 h to permit gut purging. The following day, clams were removed from the aquaria, rinsed with filtered deionized water, measured, weighed, wrapped in baked-out aluminum foil, and frozen for subsequent analysis of C_t and lipids.

Sediment and tissue samples were extracted, enriched and analyzed for 10 organic compounds as described in Ferraro et al. (1990). These extracts were stored approximately 1.5 yr at 4°C. The extracts, amended with internal standards (4-bromobiphenyl and 2,2',3,4,4',5,6,6' octachlorobiphenyl), were treated specifically for the enrichment of PCB congeners using open column chromatography (1.0 cm id) with 5 g silica gel preceded by 1 cm depth silica gel impregnated with 40% sulfuric acid (w/w) and elution with 45 mL hexane. These procedures were necessary to remove interferences from biogenic materials and other electron-capture active contaminants. Determinations of 11 PCB congeners [chemical structure (IUPAC No.): 2,2',5,5' tetrachlorobiphenyl (52), 2,2',4,5,5' pentachlorobiphenyl (101), 2,3,3',4,4' pentachlorobiphenyl (105), 2,3,3',4',6 pentachlorobiphenyl (110), 2,3',4,4',5 pentachlorobiphenyl (118), 2,2',3,3',4,4' hexachlorobiphenyl (128), 2,2',3,4,4',5' hexachlorobiphenyl (138), 2,2',3,5,5',6 hexachlorobiphenyl (151), 2,2',4,4',5,5' hexachlorobiphenyl (153), 2,3,3',4,4',5 hexachlorobiphenyl (156), and 2,2',3,4,4',5,5' heptaclorobiphenyl (180)] were made by capillary gas chromatography using electron capture detection and

hydrogen carrier gas. Quantifications were carried out using the internal standards and calibrations with individual PCB congeners (Ultra Scientific, North Kingston, RI), except for PCBs 105, 110 and 156 which were estimated from their % in Aroclor 1254 as described in Ferraro et al. (1990). No correction was needed for methods blanks since all blanks were below detection limits. The average method recovery from the silica gel procedure was approximately 90%. Detection limits were 0.2-0.6 ng/g, dry wt, for both sediments and tissues.

Mean AFs were calculated using the appropriate treatment means for C_t, C_s, and TOC, and the mean percentage lipid of clams in all experimental treatments since % lipids were not significantly different across treatments (Ferraro et al. 1990). The variances of treatment AFs were estimated using the rule for combining errors of products and quotients in Beers (1962). Accumulation factors were tested for homogeneity of variance by F_{max} (Sokal and Rohlf 1981). Differences in AFs among treatments by congener and among congeners by treatments were determined by multiple comparisons tests (experimentwise $\alpha = 0.05$) using the Tukey-Kramer method, or, the Games and Howell method when variances were heterogeneous (Sokal and Rohlf 1981).

RESULTS AND DISCUSSION

Summary statistics for C_s and C_t are provided in Tables 1 and 2, respectively. The C_s of each PCB congener was consistently lowest at station R, intermediate at station S and highest at station D, and C_s was higher in the 8-12 cm than the 0-2 cm sediments at station D.

Table 1. Mean ± SE[a] sediment concentrations (ng/g, dry wt) of 11 PCB congeners in five treatments

IUPAC No.	Treatment 1 R, 0-2[b]	2 R, 4-8	3 S, 0-2	4 D, 0-2	5 D, 8-12
52	0.5 ±0.00	0.4	6.0±1.08	43 ± 6.0	121 ± 6.5
101	1.93±0.067	1.60	15.2±1.27	44 ± 8.8	156 ±11.6
105	1.51±0.032	1.26	8.6±0.37	20 ± 3.7	70 ± 7.6
110	3.80±0.055	3.14	26.0±2.92	76 ±17.7	250 ±44
118	2.93±0.067	2.50	16.5±1.42	45 ± 9.2	162 ±16.5
128	0.4 ±0.00	0.3	2.4±0.48	3.8 ± 0.32	20.1± 0.87
138	4.23±0.120	3.40	21.7±1.19	46 ± 9.6	155 ±14.9
151	0.33±0.033	0.30	2.4±0.46	3.47± 0.240	20.5± 1.51
153	4.00±0.153	4.00	19 ±3.1	37 ± 6.5	112 ± 9.0
156	0.60±0.019	0.48	3.4±1.73	11.6 ± 2.29	34 ± 5.3
180	0.83±0.033	0.70	11 ±4.5	18 ± 3.4	61 ± 5.5

[a] Sample size = 3 except for treatment 2 where n = 1.
[b] Station designation and depth in cm.

Table 2. Mean ± SE[a] clam tissue concentrations (ng/g, dry wt) of 11 PCB congeners in five treatments

IUPAC No.	Treatment 1 R, 0-2[b]	2 R, 4-8	3 S, 0-2	4 D, 0-2	5 D, 8-12
52	6.7±1.67	4.9 ±1.06	19.1 ±1.81	44 ±4.1	50 ± 8.1
101	18.1±2.89	11.7 ±1.23	31.8 ±1.65	48 ±3.2	64 ±10.2
105	6.6±0.83	1.8 ±0.67	8.2 ±0.75	11.9 ±0.84	20.3± 2.83
110	20 ±2.8	13.5 ±1.69	39 ±3.3	53 ±3.7	75 ±11.6
118	20 ±3.0	12.0 ±1.89	28.9 ±2.60	40.3 ±2.64	66 ± 8.9
128	3.8±0.90	1.18±0.149	2.38±0.417	2.69±0.226	5.5± 0.71
138	21.1±2.51	14.6 ±1.39	22.6 ±1.54	26.7 ±1.78	40 ± 6.3
151	3.5±0.95	2.5 ±0.71	3.6 ±0.32	4.9 ±0.34	8.2± 1.19
153	19.6±2.26	18.7 ±2.90	20.7 ±1.12	25.0 ±1.63	34 ± 5.9
156	2.6±0.59	1.93±0.284	2.61±0.192	2.89±0.215	4.1± 0.77
180	6.1±1.36	2.3 ±0.36	3.8 ±0.62	4.1 ±0.37	7.3± 1.03

[a] Sample size = 9 except for treatment 2 where n = 4.
[b] Station designation and depth in cm.

Sediment TOC and grain size in each treatment are reported in Ferraro et al. (1990). Mean TOC was significantly different across treatments (ANOVA, $p < 0.001$) varying from 0.84% (station R, 4-8 cm) to 7.37% (station D, 8-12 cm). There was no significant difference in the mean % lipids (5.52 ± 0.203%, dry wt, n = 25; $F_{(4,20)} = 1.65$, $p > 0.05$) among treatments.

Statistically significant differences between some treatment AF means were found for 6 congeners (IUPAC No. 101, 105, 110, 118, 138, 151) (Table 3). The magnitude of the differences were small, however, the mean AFs differing by a factor of about 2-3 for a given congener. Mean AFs exceeded 2 for PCB-52 in 2 treatments (R, 0-2 and S, 0-2); all other mean AFs were < 2. Individual clam AFs exceeded 2 for PCB-52 (5 in treatment S, 0-2, $AF_{max} = 3.11$; 4 in treatment R, 0-2, $AF_{max} = 5.42$; 1 in treatment R, 0-4, $AF_{max} = 3.04$), PCB-101 (2 in treatment R, 0-2, $AF_{max} = 2.81$), PCB-128 (2 in treatment R, 0-2, $AF_{max} = 3.74$), PCB-151 (2 in treatment R, 0-2, $AF_{max} = 4.58$; 1 in treatment R, 4-8, $AF_{max} = 2.23$), and PCB-180 (1 in treatment R, 0-2, $AF_{max} = 2.62$). More than half (10 of 18) of the individual clam AFs which exceeded 2 were for PCB-52, and most (13 of 18) individual clam AFs > 2 occurred in clams exposed to station R (low C_s and low TOC) sediment. Of the total number of AFs determined (40 clams in 5 treatments x 11 PCB congeners = 440 AFs) only 18, or about 4% exceeded 2, the integer value of the maximum AF predicted by McFarland (1984) and McFarland and Clarke (1986).

Mean AFs were not constant across the 11 PCB congeners in 3 of the 5 treatments (Table 3). Accumulation factors appear to be related to the hydrophobicity (or lipophilicity) of the congeners since there were highly significant negative rank correlations between individual clam AFs and log K_{ow} in each of the treatments

Table 3. Mean ± SE[a] accumulation factor (AF) of clams, results of tests for homogeneity of variance (F_{max}), differences between mean AFs by multiple comparisons (MC)[b], and maximum AFs (AF_{max}) by PCB congener

IUPAC No.	log K_{ow}[c]	Treatment 1 R, 0-2[d]	2 R, 4-8	3 S, 0-2	4 D, 0-2	5 D, 8-12	F_{max}	MC	AF_{max}
A. 52	5.84	2.1 ±0.54	1.9 ±0.44	2.1 ±0.98	0.94±0.131	0.56±0.098	**		5.42
B. 101	6.38	1.46±0.253	1.11±0.164	1.40±0.131	1.00±0.147	0.55±0.096	ns	5≠1,3	2.81
C. 105	6.65	0.68±0.096	0.22±0.084	0.64±0.072	0.56±0.081	0.39±0.064	ns	2≠1,3	1.02
D. 110	6.48	0.82±0.126	0.66±0.104	0.99±0.122	0.63±0.103	0.41±0.080	ns	3≠5	1.54
E. 118	6.74	1.08±0.177	0.73±0.136	1.17±0.140	0.82±0.120	0.54±0.086	ns	3≠5	1.99
F. 128	6.74	1.5 ±0.36	0.60±0.095	0.65±0.143	0.65±0.075	0.37±0.053	**		3.74
G. 138	6.83	0.78±0.106	0.66±0.091	0.70±0.068	0.54±0.082	0.35±0.062	ns	5≠1,3	1.17
H. 151	6.64	1.7 ±0.46	1.2 ±0.40	0.99±0.152	1.30±0.131	0.53±0.087	**	4≠5	4.58
I. 153	6.92	0.76±0.102	0.71±0.132	0.72±0.089	0.62±0.085	0.40±0.078	ns		1.20
J. 156	7.18	0.67±0.158	0.61±0.110	0.51±0.156	0.23±0.034	0.16±0.035	**		1.69
K. 180	7.36	1.15±0.265	0.51±0.095	0.23±0.068	0.21±0.032	0.16±0.026	**		2.62
F_{max}		**	ns	**	**	*			
MC			A≠C-G,I-K	K≠B,E	H≠J,K				

[a] Sample size = 9 except for treatment 2 where n = 4.
[b] Multiple comparisons by the Tukey-Kramer method, or, when variances were heterogeneous by the Games and Howell method. Experimentwise α = 0.05.
[c] Hawker and Connell (1988).
[d] Station designation and depth in cm.
ns = $p > 0.05$; * = $p < 0.05$; ** = $p < 0.01$.

(Table 4). The structural explanation for these correlations is unclear. The relationship could reflect a direct effect of physicochemical properties on bioavailability, an indirect effect of other factors (e.g., the time to reach equilibrium or the rates of chemical degradation or metabolism- see Ferraro et al. (1990) and references therein) on AFs, or a more complex underlying structure.

Table 4. Spearman rank correlations (r_s) between accumulation factors and log K_{ow}[a] by treatment

Treatment	n	r_s	p
R, 0-2	99	-0.31	<0.01
R, 4-8	44	-0.50	<0.01
S, 0-2	99	-0.77	<0.01
D, 0-2	99	-0.71	<0.01
D, 8-12	99	-0.53	<0.01

[a] See Table 3.
n = sample size; p = significance probability of correlation.

The overall results for the 11 PCB congeners were similar to those reported for 10 other compounds in the same experiment (Ferraro et al. 1990). Although AFs for individual PCB congeners were not always homogeneous across treatments, AF variability was low; AFs were usually < 2; AFs > 2 were exclusive to clams exposed to sediments with low C_s (≤ 6 ng/g, dry wt) and, with one exception, low TOC (≤ 0.86%); and AFs were not constant across chemicals in all treatments.

The predicted value of C_t using an AF of 3.11 (the maximum AF for all 11 PCB congeners in an individual clam exposed to sediments from stations S or D, i.e., excluding sediments with low C_s and low TOC) overestimates the mean C_t of the 11 PCB congeners in this study by a factor of 1.8-19.7, while an AF of 2 overestimates the mean C_t by a factor of 1.2-12.7 for all but PCB-52 at stations R and S. Less liberal estimates of C_t may be obtained using maximum AFs by congener. High or maximum AFs for single chemicals or groups of chemicals can be used to predict upper bounds for C_t in animals exposed to polluted sediments, thereby providing a reliable way to screen out sediments in which animals have a low probability of concentrating pollutants to unacceptable levels (Rubinstein et al. 1987; Ferraro et al. 1990).

The low variability of AFs for animals exposed to polluted sediments in this and other recent studies (Rubinstein et al. 1987; McElroy and Means 1988; Clarke et al. 1988; Ferraro et al. 1990) suggests the AF model is a "justifiable simplification" (sensu Karickhoff 1981) of chemical partitioning, and that AFs may be used to predict C_t.

Acknowledgments. We thank N. Rubinstein, J. Lake and V. McFarland for reviewing the manuscript. The contents of this paper do not necessarily reflect the views and policies of the EPA, nor does mention of trade names or commercial products constitute endorsement or recommendation for use. Contribution No. N-107 of the U.S. Environmental Protection Agency, Environmental Research Laboratory, Narragansett, RI.

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Received March 1, 1990; accepted June 4, 1990.