Environ. Sci. Technol. **1996**, *30,* 3377−3384

Development and Verification of a Bioaccumulation Model for Organic Contaminants in Benthic Invertebrates

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A novel nonequilibrium, steady-state model is presented to predict the bioaccumulation of organic chemicals by filter feeding and detritivorous benthic invertebrates. This model accounts for chemical disequilibria between overlying water, diet and sediment, biomagnification, and benthic invertebrate feeding preferences and strategies. The results of a field study of PCB congener bioaccumulation in various benthic invertebrate species in western Lake Erie are reported to verify the model. A comparison of model-predicted and field data demonstrate that the predictability of this model is better than that of the widely used equilibrium partitioning model for assessing bioaccumulation in benthic organisms and for developing sediment quality criteria.

Introduction

In aquatic systems, the organic carbon fraction of sediment acts as a major repository for organic contaminants. Benthic invertebrates bioaccumulate sediment-derived contaminants because they spend much of their lifecycle in intimate contact with sediment (e.g., refs 1−3). As important prey items for many fish species, these organisms can transfer contaminants to higher trophic levels in aquatic food webs. Hence, the ability of contaminants to bioaccumulate in higher trophic levels is often dependent on the extent of chemical accumulation in benthic invertebrates (*4*). The most widely used model for predicting the extent of organic chemical accumulation in benthic invertebrates is the equilibrium partitioning (EP) model. This model assumes that chemical concentrations in benthic invertebrates, sediment, and pore water are in thermodynamic equilibrium (*5*, *6*). The equilibrium assumption enables chemical concentrations in benthic invertebrates to be predicted from chemical concentrations in the sediment or porewater, using appropriate partition coefficients. The applicability of this model has been reviewed extensively by DiToro et al. (*7*). It has been considered for adoption by regulatory agencies in Canada (*8*) and has been adopted in the United States to establish sediment quality guidelines (*9*).

The EP model is simple and easy to use. It assumes that the ratio of lipid-normalized chemical concentration in the organism to the organic carbon-normalized concentration in sediment, a ratio sometimes referred to as the biota sediment accumulation factor (BSAF), should be constant and independent of the chemical, organism, and sediment properties (*7*). Parkerton (*10*) did an extensive literature review of measured BSAFs in benthic invertebrates. Contrary to the EP predictions, he found that ratios varied by 5 orders of magnitude among PCB congeners of differing log K_{OW} and with similar feeding type (e.g., filter feeders, deposit feeders, and omnivores). BSAFs for PCBs with the same log K_{OW} and within feeding type also varied up to 4 orders of magnitude. Furthermore, BSAFs exhibited a parabolic dependence on log K_{OW}.

Other studies of less generality described a smaller but still considerable degree of variation in BSAFs. For example, Markwell et al. (*1*) measured BSAFs in oligochaetes that ranged from 5.5 to 22.7 with an average of 11.0. Van der Oost et al. (*11*) found that BSAFs in zebra mussels and crustaceans ranged from 2.0 to 21.3, and those measured in Lake St. Clair mayflies by Gobas et al. (*6*) varied from 0.2 to 1.0. Furthermore, Landrum et al. (*12*) reported considerable seasonal changes in the accumulation of PAHs in *Diporeia* spp., and Lake et al. (*13*) found that BSAFs varied in response to the level of contamination and the organic carbon content of the sediment. In addition to the differences in the BSAFs among chemicals, species, and location, it is important to note that BSAFs can achieve values that are much greater than the values of 1−2 predicted by the EP model and used in the development of sediment quality criteria.

There are several factors that may cause the EP model to inaccurately predict BSAFs and concentrations in benthic invertebrates. First, the EP model does not include biomagnification, which can result in concentrations in benthic invertebrates that are in excess of their chemical equilibrium concentrations in the diet due to digestion (*14*). Biomagnification can explain why many observed BSAFs are greater than expected by the EP model. Second, the EP model does not adequately distinguish between different feeding strategies among benthic invertebrates such as filter feeding and detritus consumption, which may be responsible for considerable differences in the BSAF among species. Third, the EP model assumes chemical equilibrium between pore water, sediment, and the organism. However, for chemical uptake the overlying water may be more relevant than the porewater because it is in contact with the respiratory surface of the organism. Chemical concentrations in sediment and overlying water can be in considerable disequilibrium in aquatic ecosystems as a result of temporal changes in chemical inputs (*15*), relatively slow chemical kinetics between sediment and water

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S0013-936X(96)00280-5 CCC: $12.00 © 1996 American Chemical Society

compared to organisms and water (*16*), and changes in the organic carbon pool (*17*). In addition, there are several other factors relating to contaminant kinetics, rates of metabolism, mode of feeding, and seasonal changes in feeding, reproductive status, and lipid contents (*12*) that cause significant errors in the estimates of EP models. Consequently, this paper reports (i) the development of an alternative model for the bioaccumulation of organic chemicals in filter feeders and detritivores under nonequilibrium conditions and (ii) the results of field studies in western Lake Erie to verify the applicability of the model for predicting chemical concentrations in various benthic invertebrate species. The main objective of the model is to provide a more realistic description of organic chemical bioaccumulation in benthic invertebrate species. In order to achieve this, biomagnification, disequilibria at the sediment−water interface, and two different feeding strategies (filter feeding and detrital feeding) have been incorporated into the model. However, at the same time, we have attempted to keep the model simple and generic to facilitate its use on a broad scale for regulatory purposes, such as the development of sediment quality criteria.

Theory

Although chemicals exhibit a natural tendency toward achieving a thermodynamic equilibrium in the environment, biological, environmental, and in some cases chemical (e.g., reaction and metabolic transformation) processes can prevent chemical equilibrium from being achieved. The resulting disequilibrium may reflect a temporary situation in response to changing environmental conditions, or it can be maintained over extended periods of time reflecting a kinetically controlled equilibrium, here referred to as steady-state. We hypothesize that, under most field conditions, BSAFs of hydrophobic organic chemicals, in benthic detritivores and filter feeders, reflect this steady-state condition with chemical concentrations in overlying water, sediment, suspended solids, and porewater that may be in a prolonged or continuous chemical disequilibrium. This is believed to be due to the small size and corresponding short chemical half-life of many sediment-dwelling organisms and filter feeders in relation to the large size and long half-life of the sediments and water in which these organisms reside. In our one-compartment, steady-state model, we assume that the predominant routes of chemical uptake are via the respiratory surface from the overlying water (U_W, mol d^{-1}) and from the diet (U_D, mol d^{-1}). The primary routes for chemical elimination are to the water via the respiratory surface (D_W, mol d^{-1}) and into fecal matter (D_F, mol d^{-1}). If the chemical can be metabolized, there is a loss of the chemical through metabolic transformation (D_M, mol d^{-1}). These pathways are illustrated in Figure 1.

At steady-state, chemical intake from water (U_W, mol d^{-1}) and food (U_D, mol d^{-1}) equals the sum of chemical elimination from gills, feces, and metabolism. Hence

$$U_W + U_D = D_W + D_F + D_M \quad (1)$$

In filter feeders and benthic detritivores, the uptake of chemical from water via the respiratory surface can be described as the product of the concentration of chemical in the overlying water (C_W, mol m^{-3}), the water ventilation rate across the respiratory surface (G_W, m^3 d^{-1}), and the efficiency of chemical transfer across the respiratory surface (E_W):

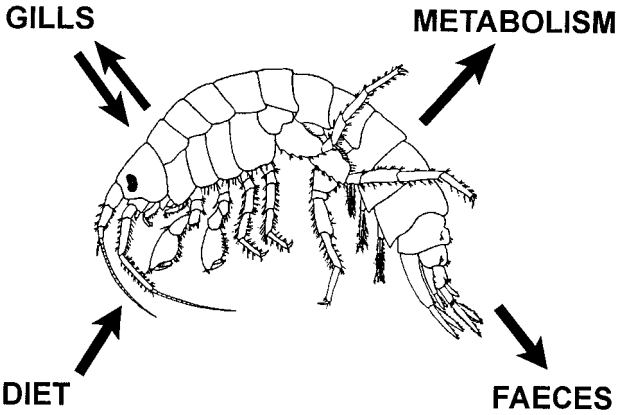

FIGURE 1. Conceptual diagram of routes of chemical intake and elimination considered for benthic filter feeders and detritivores (diagram of amphipod from ref 30).

$$U_W = C_W G_W E_W \quad (2)$$

Chemical uptake in detritivores and filter feeders through food intake can be described as the product of the concentration of chemical in food (C_D, mol m^{-3}), the ingestion rate of food (G_D, m^3 d^{-1}), and the efficiency of chemical transfer between gut contents and the organism (E_D):

$$U_D = C_D G_D E_D \quad (3)$$

For filter feeders, the ingestion rate can be described as the product of the gill ventilation rate, the concentrations of plankton and other suspended solids in the water column (V_{pl} , m^3 m^{-3}), and the particle scavenging efficiency (σ) describing the fraction of particles that are removed from the water column by the organism:

$$G_D = G_W V_{pl} \sigma \quad (4)$$

Consequently, chemical uptake in filter feeders through food intake can be described as follows:

$$U_D = C_{pl} G_W V_{pl} \sigma E_D \quad (5)$$

where C_{pl} is the concentration of chemical in plankton and other particulate matter in the water column.

Elimination of chemical via the respiratory surface of detritivores and filter feeders can be described as

$$D_W = \frac{C_B G_W E_W}{K_{BW}} \quad (6)$$

where C_B is the chemical concentration in the detritivore or filter feeder (mol m^{-3}) and K_{BW} is the organism−water partition coefficient.

Elimination of chemical via feces can be described as

$$D_F = \frac{C_B G_F E_D}{K_{BF}} \quad (7)$$

where G_F is the egestion rate (m^3 d^{-1}) and K_{BF} is the organism−feces partition coefficient. Elimination of chemical via metabolic transformation can be described as

$$D_M = C_B V_B k_M \quad (8)$$

where k_M (d^{-1}) is the rate of metabolic transformation and V_B (m^3) is the volume of the organism.

By combining eqs 1−8 and assuming that steady-state conditions apply, the following expressions for the bioaccumulation of organic chemicals in benthic filter feeders (ff) and detritivores (det) result:

$$C_{B(ff)} = \frac{C_W G_W E_W + C_{pl} G_W V_{pl} E_D \sigma}{\frac{G_W E_W}{K_{BW}} + \frac{G_F E_D}{K_{BF}} + V_B k_M} \quad (9)$$

and

$$C_{B(det)} = \frac{C_W G_W E_W + C_D G_D E_D}{\frac{G_W E_W}{K_{BW}} + \frac{G_F E_D}{K_{BF}} + V_B k_M} \quad (10)$$

To better understand the factors controlling the BSAF in benthic organisms, it is beneficial to express eqs 8 and 9 in fugacity format as described by Mackay et al. (*18*). This can be done by replacing concentrations and partition coefficients with their corresponding fugacity expressions, i.e., the concentration (C) (in mol m^{-3}) is replaced by the product of the chemical fugacity (f) (in units of Pa) and the fugacity capacity (Z) (in units of mol m^{-3} Pa^{-1}), hence, C is fZ. The partition coefficient of a chemical between two media, such as the organism and water (K_{BW}) is the ratio of the chemical's fugacity capacities in the two media, i.e., K_{BW} equals Z_B/Z_W. Likewise, K_{BF} equals Z_B/Z_F.

The fugacities represent the equilibrium status of the chemical. Equal fugacities of a chemical in different media (e.g., $f_S = f_{B(ff)}$) represent a chemical equilibrium. The fugacity capacity (Z) is a measure of the media's potential for "storing" a chemical and is hence related to the chemical's solubility in that medium. The fugacity capacity of a phase such as diet (Z_D) can be expressed as $Z_D = Z_W \delta_D \phi_D K_{OW}$ if the diet contains lipids (e.g., prey items of crayfish such as zebra mussels) or $Z_D = Z_W \delta_D \phi_D K_{OC}$ if the diet contains organic carbon (e.g., prey items of zebra mussels such as phytoplankton) (*18*). In these expressions, Z_W is the fugacity capacity of the water, δ_D is the density of the diet (kg L^{-1}), ϕ_D is either the fraction of lipid or organic carbon in the diet, and K_{OW} and K_{OC} are the octanol−water and organic carbon−water partition coefficients, respectively. In filter feeders and detritivores, the fugacity capacity of the faecal matter (Z_F) is only a fraction of the fugacity capacity of the diet (Z_D) due to the removal of lipids and organic carbon upon digestion. Hence $Z_F = (1 - \alpha)Z_D$, where α is the fraction of organic carbon or lipid in the diet that is removed upon digestion. Digestion also causes the fecal egestion rate G_F to be less than the dietary ingestion rate G_D, i.e., $G_F = (1 - \beta)G_D$, where β is the fraction of ingested diet absorbed by the organism. If these substitutions are made into eqs 9 and 10, chemical accumulation by filter feeders relative to sediment can be expressed in fugacity terms as

$$\frac{f_{B(ff)}}{f_S} = 0.62\text{BSAF} = \frac{E_W \frac{f_W}{f_S} + E_D V_{pl} \sigma \frac{f_D}{f_S} \delta_D \phi_D (K_{OW} \text{ or } K_{OC})}{E_W + E_D (1 - \alpha)(1 - \beta) V_{pl} \sigma \delta_D \phi_D (K_{OW} \text{ or } K_{OC}) + \left(\frac{V_{ff} k_M K_{BW}}{G_W}\right)} \quad (11)$$

and chemical accumulation by detritivores relative to sediment is described as

$$\frac{f_{B(det)}}{f_S} = 0.62\text{BSAF} = \frac{E_W G_W \frac{f_W}{f_S} + E_D G_D \frac{f_D}{f_S} \delta_D \phi_D (K_{OW} \text{ or } K_{OC})}{E_W G_W + E_D (1 - \alpha)(1 - \beta) G_D \delta_D \phi_D (K_{OW} \text{ or } K_{OC}) + V_{det} k_M K_{BW}} \quad (12)$$

In equation 11 and 12, K_{OW} is used when lipids are the main source of energy in the diet, and K_{OC} is used when organic carbon is the primary source of energy in the benthic invertebrate's diet. If the diet contains a mixture of organic carbon and lipid rich items, then K_{OC} is used to describe those items for which organic carbon is the major energy storage form, and K_{OW} is used to describe those items in which lipid is the major energy storage form.

The fugacity ratio of filter feeders and detritivores to sediment ($f_{B(ff)}/f_S$ and $f_{B(det)}/f_S$) can be converted to a BSAF by dividing the ratio by 0.62. The rationale for this conversion factor is as follows: The biota to sediment fugacity ratio (f_B/f_S) equals the product of chemical concentration in biota (C_B) and the capacity of sediment (Z_S) divided by the product of the chemical concentration in sediment (C_S) and the fugacity capacity of biota (Z_B); i.e., f_B/f_S equals $C_B Z_S / C_S Z_B$. Substituting $Z_W \delta_S \phi_S K_{OC}$ for Z_S and $Z_W \delta_B \phi_B K_{OW}$ for Z_B and assuming K_{OC} equals $0.41 K_{OW}$ (*19*), it follows that the ratio of the lipid based concentration C_B/ϕ_B and the organic carbon based concentration C_S/ϕ_S (i.e., the BSAF) equals $f_B \delta_B / f_S \delta_S$ 0.41. Assuming that the density of the organism (δ_B) equals 1.0 kg L^{-1} (*18*) and the density of sediment (δ_S) is 1.5 kg L^{-1} (*6*), it follows that the BSAF equals $1.6 f_B/f_S$ or f_B/f_S equals 0.62BSAF. Given the natural variability in the densities of the sediments as well as the variability in the sorption tendency among sediments, it follows that BSAFs ranging between approximately 1 and 2 can reflect a thermodynamic equilibrium between the organism and the sediments.

This steady-state model (eqs 11 and 12) illustrates the factors controlling the bioaccumulation of organic substances in benthic filter feeders and detritivores. Specifically, it highlights the role of chemical hydrophobicity (as indicated by K_{OW}) and environmental disequilibrium (as indicated by f_W/f_S and f_D/f_S) in determining the BSAF of a benthic invertebrate. For instance, the equation shows that the second term in the numerator (i.e., $E_D G_D (f_D/f_S) \cdot \delta_D \phi_D K_{OW \text{ or } OC}$), which describes the contribution of diet to the invertebrates' contaminant burden, becomes more important relative to the first expression (i.e., $E_W G_W (f_W/f_S)$), which describes the intake of chemical from water via the respiratory surface, when K_{OW} or K_{OC} increases. The second term in the denominator (i.e., $E_D(1 - \alpha)(1 - \beta)G_D \cdot \delta_D \phi_D K_{OW \text{ or } OC}$), which describes elimination via feces, also becomes more important relative to the first term (i.e., $E_W G_W$), which describes elimination of chemical via the gills, when K_{OW} or K_{OC} increases. Hence, for low K_{OW} substances (i.e., log K_{OW} less than 6 as is illustrated in the Results) that are not metabolized, the fugacity ratio $f_{B(ff)}/f_S$ and $f_{B(det)}/f_S$ or 0.62BSAF approaches f_W/f_S. If the chemical concentrations in water and sediments are in a chemical equilibrium (i.e., $f_W = f_S$), then $f_{B(ff)}/f_S$ and $f_{B(det)}/f_S$ approach 1.0 and the BSAF is approximately 1.6. If the chemical fugacity in the overlying water is less than that in the

TABLE 1

Observed Concentrations of PCB Congeners in Western Lake Erie Benthic Invertebrates (μg kg^{-1} Wet Weight), Sediments (μg kg^{-1} Dry Weight), and Water (ng L^{-1})

congener	log K_{OW}[b]	sediment (n = 9)		water (n = 3)[a]		plankton (n = 5)		zebra mussels (n = 20)		caddisfly larvae (n = 1)	gammarus (n = 4)		crayfish (n = 5)	
		mean	SD	mean	SD	mean	SD	mean	SD	mean	mean	SD	mean	SD
	% lipid					1.2	0.24	1.3	0.34	1.7	2.1	1.04	1.7	0.11
	% OC	7.4	1.78											
28/31	5.6	*1.949	0.7309	0.008	0.0031	0.350	0.2353	0.431	0.4642	0.369	0.666	0.2768	0.392	0.2407
52	6.1	4.024	1.4404	0.028	0.0156	0.834	0.2399	*1.473	0.5168	1.704	2.060	1.1426	0.660	0.0624
49	6.1	2.042	0.7850	0.013	0.0060	0.394	0.1462	*0.696	0.2691	0.872	0.967	0.4325	0.244	0.0230
44	6.0	2.295	0.8216	0.013	0.0053	0.494	0.1115	*0.795	0.2311	1.275	1.146	0.4454	0.142	0.0164
42	5.6	*1.479	0.5501	0.005	0.0021	0.252	0.0581	*0.352	0.2313	0.000	0.419	0.2471		
64	6.1	0.440	0.1939	0.001	0.0005	0.068	0.0421	*0.136	0.0570	0.065	0.183	0.0875		
74	6.1	1.882	0.6916	0.005	0.0026	0.272	0.1914	0.554	0.2128	0.994	0.840	0.4339	0.390	0.0400
70	5.9	3.398	1.2745	0.017	0.0110	0.542	0.2844	0.830	0.2632	0.821	0.949	0.4539	0.144	0.0230
66/95	5.8	6.587	2.1076	0.026	0.0141	1.790	0.7137	2.242	0.6836	2.063	2.822	1.1957	1.074	0.1036
60	5.9	2.856	1.2365	0.007	0.0034	0.478	0.2125	0.644	0.4736	0.000	1.037	0.5871	0.566	0.0981
101	6.4	5.045	1.9068	0.018	0.0067	1.694	0.4525	2.868	0.8139	3.925	2.983	1.2448	1.912	0.2302
99	6.6	2.464	0.9362	0.008	0.0047	0.762	0.2896	*1.611	0.6134	2.567	2.243	1.1347	1.692	0.2129
97	6.6	1.580	0.6074	0.004	0.0020	0.400	0.1338	0.629	0.2388	0.698	0.743	0.4850	0.378	0.1195
87	6.5	2.206	0.8281	0.009	0.0043	0.588	0.1794	*1.013	0.3226	1.361	1.094	0.4077	0.550	0.0930
110	6.5	6.202	2.4848	0.011	0.0056	1.476	0.4845	2.253	0.6983	1.767	3.203	1.8491	0.562	0.1137
151	6.9	1.762	0.6666	0.003	0.0022	0.992	0.3298	1.588	0.4800	1.922	2.154	1.0334	0.956	0.1322
149	6.8	5.848	2.3524	0.008	0.0052	2.890	0.8772	4.674	1.0940	4.874	5.396	2.6169	2.026	0.2805
118	6.4	4.514	1.8449	0.007	0.0044	0.750	0.4919	*2.156	0.8847	4.780	3.113	1.7881	2.242	0.3628
146	6.9	1.211	0.5074	0.001	0.0001	0.466	0.1326	1.264	0.4853	3.061	1.681	0.8925	1.218	0.1878
153	6.9	5.841	2.2982	0.006	0.0037	2.344	0.6357	7.066	2.2491	10.423	8.529	4.1301	6.230	0.9667
105	6.4	2.703	1.0659	0.003	0.0020	0.666	0.1881	1.627	1.6470	1.109	1.611	0.7505	0.606	0.1101
141		1.667	0.6238	0.001	<0.0001	0.514	0.1812	1.118	0.3237	1.424	1.146	0.5498	0.880	0.1492
138	7.0	9.737	4.9230	0.008	0.0058	2.576	0.7204	7.093	2.3238	10.446	9.064	5.0972	6.446	1.1187
129	7.3	0.858	0.3575	<0.001	<0.0001	0.246	0.0802	0.558	0.2143	0.770	0.929	0.5870	0.542	0.1078
182/187	7.2	3.494	1.5063	0.002	0.0006	1.188	0.3638	3.873	1.1775	6.196	5.001	2.7107	3.744	0.6297
183	7.0	1.517	0.6822	0.001	<0.0001	0.512	0.1491	1.667	0.4905	2.957	1.991	1.0698	1.056	0.1936
185	7.0	0.227	0.0718	nd	<0.0001	0.090	0.0316	0.145	0.0800	0.065	0.189	0.0865	0.094	0.0167
174	7.0	2.549	1.2685	0.001	0.0001	0.716	0.2185	1.914	0.5222	2.014	2.244	1.2816	1.098	0.1974
171	6.7	0.755	0.2188	<0.001	0.0001	0.210	0.0678	0.745	0.2464	0.285	0.956	0.5550	0.522	0.1094
200		0.549	0.2379	nd		0.108	0.1741	0.326	0.0934	0.216	0.423	0.2684	0.230	0.0324
172		0.422	0.1831	<0.001	<0.0001	0.094	0.0321	0.300	0.1080	0.321	0.461	0.2769	0.300	0.0534
180	7.4	6.041	2.5446	0.003	0.0027	1.336	0.4032	5.127	1.6875	8.266	6.789	4.2289	5.002	1.0381
170/190	7.3	3.770	1.8822	0.001	<0.0001	0.604	0.1845	2.257	0.7543	3.378	3.027	1.7870	1.774	0.3895
201	7.5	2.320	1.1580	0.002	0.0003	0.392	0.1819	1.374	0.4204	1.784	1.637	0.9936	1.168	0.2544
203	7.1	1.442	0.6904	<0.001	<0.0001	0.206	0.0615	0.869	0.2636	1.368	1.043	0.6253	0.466	0.1024
195	7.1	*0.954	0.4614	<0.001	<0.0001	0.330	0.4179	0.365	0.0996	0.318	0.387	0.2496	0.278	0.0832
194	7.1	*5.296	5.2338	nd		0.134	0.0619	0.631	0.2023	0.889	1.209	0.7835	0.560	0.1308
206	7.2	0.729	0.3682	<0.001	<0.0001	0.040	0.0894	0.211	0.0970	0.065	0.549	0.6770	0.104	0.0207

[a] From ref 31. [b] Values from Shiu and Mackay (*32*) except for congeners 110, 182/187, 180, and 170/190, which are from Hawker and Connell (*33*). An asterisk (*) indicate that lipid- or organic carbon-normalized data statistically differed (Kruskal−Wallis, $p < 0.05$) between years.

sediments, then the BSAF will be less than 1.6. Also, if metabolic transformation occurs, the BSAF will be less than 1.6. With increasing K_{OW}, the second term in the numerator and denominator will become more important, causing the fugacity ratio $f_{B(ff)}/f_S$ and $f_{B(det)}/f_S$ and 0.62BSAF to approach $f_D/(f_S(1 - \alpha)(1 - \beta))$ for non-metabolizing substances, indicating that the BSAF is dependent (i) on the fugacities in the diet of the benthic invertebrates relative to the fugacity in the sediments, (ii) the extent of reduction in the fugacity capacity of the diet upon digestion, and (iii) the rate of food absorption, which is reflected in the extent to which the fecal egestion is lower than the dietary absorption rate. For example, if the chemical in the prey items and sediments are in equilibrium (i.e., $f_D = f_S$), α is 0.75, and β is 0.1, then the BSAF is 7.1. The model further illustrates that differences in chemical fugacities in the overlying water, bottom sediments, and diet of the organisms can have an important effect on the BSAF. Also, the rates of chemical uptake across the respiratory surface in relation to dietary absorption rates affect the BSAF. The model requires information about ventilation rates and dietary ingestion rates, which have been compiled in Morrison (*20*) for 56 species of benthic invertebrates. However, despite the fact that the model requires physiological data regarding respiration and ingestion, the model appears to be fairly insensitive to the precise values of these rates because they appear both in the numerator and denominator and, hence, have a tendency to "cancel out", which makes the model fairly robust.

Methods

Field Study. During July and August 1993, four (30 g wet weight) sediment, one composite caddisfly larvae (*Hydropsyche alterans*), and 16 composite zebra mussel (*Dreissena polymorpha*, shell lengths approximately 1.5 cm) samples were collected offshore of Middle Sister Island (41°51′ N 83°00′ W) in western Lake Erie. In July and August 1994, eight (30 g wet weight) sediment, five (3−5 g wet weight) plankton, four composite zebra mussel, four composite amphipod (*Gammarus fasciatus*), and five crayfish (*Orconectes propinquus*) samples were collected offshore of Middle Sister Island and nearby East Sister Island

TABLE 2

Definition of Symbols

parameter	units	definition
U_W, U_D	mol d^{-1}	rate of chemical uptake via the respiratory surface and diet, respectively
D_W, D_F, D_M	mol d^{-1}	rate of chemical elimination via the respiratory surface, feces, and metabolic transformation, respectively
C_W, C_D, C_B, C_{pl}	mol m^{-3}	chemical concentration in overlying water, diet, biota, and plankton and particulates, respectively
G_W, G_D, G_F	m^3 d^{-1}	rate of water ventilation across the respiratory surface, ingestion of food, and egestion, respectively
V_{pl}	m^3 m^{-3}	concentration of suspended solids in water column
E_W, E_D	%	efficiency of chemical transfer across the respiratory surface and between gut contents and the organism, respectively
σ	%	particle scavenging efficiency
K_{OW}, K_{OC}, K_{BW}, K_{BF}		chemical partition coefficient between, respectively, octanol and water, organic carbon and water, organism and water, and organism and feces
k_M	d^{-1}	rate of metabolic transformation
V_B	m^3	volume of organism
ff		filter feeder
det		detritivore
BSAF		biota−sediment accumulation factor
f_B, f_S, f_W, f_D	Pa	fugacity of biota, sediment, water, and diet, respectively
Z_B, Z_S, Z_W, Z_D, Z_F	mol m^{-3} Pa^{-1}	fugacity capacity of biota, sediment, water, diet, and feces, respectively
α	%	fraction of organic carbon or lipid in diet that is removed upon digestion
β	%	fraction of ingested diet absorbed by the organism
δ	kg L^{-1}	density of a phase
ϕ	%	fraction of lipid or organic carbon in a phase

(41°49′ N 82°51′ W). These sampling sites were chosen because they are distant from shoreline point sources of contamination and are located in one of the more contaminated sites in the Great Lakes. All of the benthic invertebrates were rinsed or hand-picked from on or beneath rocks. Plankton was collected by vertical towing throughout the water column with a 125-μm mesh net, and sediment was collected with a petite ponar grab. Environment Canada extracted PCB congeners from water at a collection site in close proximity to Middle Sister Island and East Sister Island in 1994. Three 50.5-L samples were extracted with a Goulden large volume extractor according to the methods of L'Italien and Fay (*21*). Immediately after collection, the samples were placed in either hexane-rinsed glass jars or hexane-rinsed aluminum foil and frozen at −20 °C until analysis for PCBs.

Chemical Analysis. Preparation and cleanup of tissue, sediment, and water samples were done by the Great Lakes Institute for Environmental Research (GLIER) according to the methods of Lazar et al. (*22*). Tissue samples (approximately 2−5 g wet weight) were randomly sorted before being taken to the laboratory. Each sample was homogenized with 20 g of Na_2SO_4 using 300 mL of DCM/hexane (1:1). Cleanup of tissue and water extracts was performed on 1 × 35 cm glass columns containing 6 g of Florisil (60−100 μm mesh) topped with 2 cm of anhydrous Na_2SO_4. The column was then eluted with 50 mL of hexane and roto-evaporated to 2 mL. Sediment samples (approximately 20 g wet weight) were mixed with 70 g of anhydrous Na_2SO_4. Chemicals were extracted from this mixture using acetone/hexane (1:1) in a Soxhlet extractor for 16 h. The extract was roto-evaporated to 50 mL, sent through a 50-g Na_2SO_4 column, and eluted with 250 mL of hexane. The sample was roto-evaporated to 2 mL and transferred to a 24-g Florisil column and eluted with 200 mL of hexane. This fraction was roto-evaporated to 2 mL and transferred to a 10-mL volumetric flask containing activated copper powder.

PCB congeners listed in Table 1 were quantified using gas chromatography-electron capture detection (GC-ECD). The detection level was 0.05 μg kg^{-1}, and recoveries were greater than 90%. Canadian Wildlife Service (CWS) herring gull (*Larus argentatus*) egg homogenate standards and reference material were analyzed every tenth sample, and blanks were run every six samples. The lipid content was determined on subsamples of the extracts and measured gravimetrically using one-tenth of the extract. The lipid was reported as a percent of organism wet weight. The organic carbon content of plankton and sediment was estimated by loss on ignition according to the methods of Hakanson and Jansson (*23*) and Standard Methods for the Examination of Water and Wastewater (*24*), respectively. Characterization of the sediment was as follows (%, ± SE, $n = 6$): moisture 67.4 ± 0.30; solids 32.6 ± 0.30; solvent-extractable organics 0.206 ± 0.0298; solvent-extractable total organic carbon 0.055 ± 0.0048. Solvent-extractable organics were determined gravimetrically, and solvent-extractable organic carbon was determined using inductively coupled plasma-atomic emission spectroscopy (ICP-AES) analysis. Due to the nature of the sediment, it was not possible to accurately quantify the inorganic carbon content, but Thomas et al. (*25*) analyzed the mineralogical composition of sediments from the same region of western Lake Erie and found that inorganic carbon (mean ± standard deviation) was 1.45 ± 0.99%.

Model Parametrization. Table 2 defines all of the variables used in the derivation of this model. The input parameters used in the model and their literature source are listed in Table 3. Because there is extensive resuspension of sediment in the Great Lakes (*26*−*28*), the diet of filter feeders was considered to contain 25% resuspended sediment. Metabolic transformation was ignored ($k_M = 0$) because it is not considered to be a significant route of chemical elimination for the high K_{OW} PCBs and the benthic invertebrates used in this study (*29*). Finally, the value for β was estimated to be 0.05, representing a fecal egestion rate that is close to the dietary ingestion rate, which reflects the small fraction of the diet of these organisms that is actually "absorbable".

TABLE 3

Model Parametrization[a]

environmental properties
- 1994 particulate organic carbon (POC): 5.4×10^{-7} ($\pm 3.27 \times 10^{-7}$) $m^3\ m^{-3}$ [b]
- concentration of plankton and suspended solids (V_{pl}): $4.0 \times 10^{-5}\ m^3\ m^{-3}$ [c]
- density of aquatic organisms (δ_B): 1.0 kg L^{-1} [d]
- density of plankton (δ_{pl}): 1.0 kg L^{-1} [d]
- density of sediment (δ_S): 1.5 kg L^{-1} [d]

benthic invertebrate properties
- organic carbon assimilation efficiency (α): 46 ($\pm$ 15.7)% [e]
- fraction of ingested diet absorbed (β) 5% [f]
- PCB assimilation efficiency from food (E_D): 72 ($\pm$ 28.1)% [g]
- PCB assimilation efficiency from water (E_W): 100% [f]
- metabolic rate constant (k_M): 0.0 d^{-1} [h]

Gammarus fasciatus
- mean size: 0.0013 g dry weight
- ingestion rate (G_D): $1.9 \times 10^{-8}\ m^3\ d^{-1}$ wet weight [i]
- gill ventilation rate (G_V): $6.0 \times 10^{-6}\ m^3\ d^{-1}$ [j]
- diet: 10% sediment/90% plankton [k]

crayfish (*Orconectes propinquus*)
- mean size: 1.8 g dry weight
- ingestion rate (G_D): $8.8 \times 10^{-7}\ m^3\ d^{-1}$ wet weight [l]
- gill ventilation rate (G_V): $1.6 \times 10^{-2}\ m^3\ d^{-1}$ [i]
- diet [m]: 15% sediment/25% plankton/45% zebra mussels/6% caddisfly larvae/9% *Gammarus*

zebra mussel (*Dreissena polymorpha*) and caddisfly larvae (*Hydropychidae alterans*)
- scavenging efficiency (σ): 100% [n]
- diet: 25% sediment [o]/75% plankton

[a] Standard deviations are given in parentheses. [b] From ref 34. [c] POC converted to volume of plankton according to Strathmann (*35*). [d] From ref 18. [e] Average of estimates from refs 36–42. [f] Estimated. [g] Average of estimates from refs 43–48. [h] From ref 29. [i] From ref 20. [j] Estimated from *Gammarus pulex* (*49*). [k] Used as a surrogate for zebra mussel pseudofaeces. [l] From ref 50. [m] Estimates based on work by refs 50–*52*. [n] From refs 53 and 54. [o] Used as a surrogate for resuspended sediment.

Results and Discussion

Table 1 lists the observed concentrations of PCB congeners in caddisfly larvae, zebra mussels, *Gammarus*, and crayfish. Because only 8 of 38 PCB congener concentrations differed significantly (Kruskal–Wallis test, $p < 0.05$) between those samples (sediment and zebra mussel) collected in 1993 and 1994 (Table 1), these data were combined. Observed organism-to-sediment fugacity ratios ranged between 0.29–7.7, with corresponding BSAFs of 0.5–12.4, for the two species of filter feeders and between 0.13 and 4.9, with corresponding BSAFs of 0.2 and 7.9, for the detritivores, whereas a fugacity ratio of 1.0 reflects a thermodynamic equilibrium (Figure 2). With the exception of crayfish, the logarithms of observed fugacity ratios showed statistically significant correlations with log K_{OW} (ANOVA, $p < 0.05$) illustrating a trend toward higher fugacity ratios with increasing K_{OW}. However, rather than a linear relationship, logarithms of the organism-to-sediment fugacity ratios showed a tendency to follow a parabolic relationship with log K_{OW} (Figure 2). In general, observed organism-to-sediment fugacity ratios of PCBs with a log K_{OW} less than 6.0 were either approximately 1.0 or somewhat below 1.0 (Figure 2). Fugacity ratios less than 1.0 suggest that in this system PCB concentrations in benthic invertebrates have not reached their equilibrium concentrations with sediments. Observed organism-to-sediment fugacity ratios of PCBs with a log K_{OW} greater than 6.0 were generally above 1.0–2.0, which indicate that PCB concentrations in the benthic invertebrates are greater than their equilibrium concentrations with sediment (Figure 2).

TABLE 4

Two Goodness-of-Fit Tests Comparing Predictions of This Steady-State Model (SS) and Equilibrium Partitioning Model (EP) with Observed Organism-to-Sediment Fugacity Ratios (FR)[a]

		test 1: SRSE		test 2: 95% CL	
species	*n*	EP model	SS model	EP model	SS model
CL	31	143.1	33.4	4.7	3.2
ZM	31	117.5	48.0	2.6	1.9
G	33	113.2	3.3	2.5	2.1
C	27	56.2	16.4	9	7

[a] Test 1: sum of relative squared errors (SRSE) between observed and predicted data ($SRSE = \sum((FR_{obs,i} - FR_{pred,i})/FR_{obs,i})$). Test 2: 95% confidence limits of the model predictions. CL = caddisfly larvae; ZM = zebra mussels; G = *Gammarus*; C = crayfish. The 95% confidence limit reflects the factor that should be applied to the model predictions in order to account for 95% of the observed data. This calculation is based on a log normal distribution of the deviations between observed and predicted fugacity ratios.

Observed ratios of PCB fugacities between water and sediment (f_W/f_S) were in all cases less than 1.0 (mean = 0.3 and standard error = 0.04) and did not show a statistically significant ($p < 0.05$) relationship with log K_{OW}. This indicates that PCB concentrations in the bed sediments exceeded their equilibrium concentration with the overlying water at the experimental sites and that the extent of disequilibrium was approximately the same for all PCB congeners. Observed ratios of PCB fugacities between the diet of the benthic invertebrates and sediment (f_D/f_S) were close to 1.0 (mean, standard errors, sample size, respectively: *Gammarus* 1.0, 0.05, 33; zebra mussels and caddisfly larvae 1.0, 0.18, 33; crayfish 1.6, 0.57, 33) and did not show a statistically significant ($p < 0.05$) relationship with log K_{OW}. These fugacity ratios indicate that PCB concentrations in the sediments and the diet of the benthic invertebrates are approximate in chemical equilibrium for all PCB congeners.

While the EP model predicts organism-to-sediment fugacity ratios of 1.0 for all PCBs, the steady-state model presented in this study predicts a nonlinear relationship between the fugacity ratio and K_{OW}, similar to the relationship observed (Figure 2). A goodness-of-fit test comparing the sum of relative squared errors (Table 4) illustrates that, for both filter feeders and detritivores, our steady-state model is a much better predictor of bioaccumulation in benthic invertebrates than the EP model. In addition, the quality of model predictions, as expressed by the 95% confidence limit of the model predictions of the organism-to-sediment fugacity ratios (Table 4), are better using this steady-state model as compared to the EP model. The 95% confidence limits express the range of model predictions (f_B/f_S or BSAF) that are able to account for 95% of the observed BSAFs in this study. Table 4 illustrates that with the exception of crayfish, 95% confidence limits of model predictions are less than a factor of 3.2.

A sensitivity analysis was performed on the model to identify the most important model inputs affecting the fugacity ratio between benthic invertebrates and sediment. In separate simulations, the value of each parameter was lowered 10%, and the effect of this change on model output was evaluated. Model parameters describing the digestibility and absorption of food, i.e., α and β, and the fugacity ratio between diet and sediment were the most sensitive

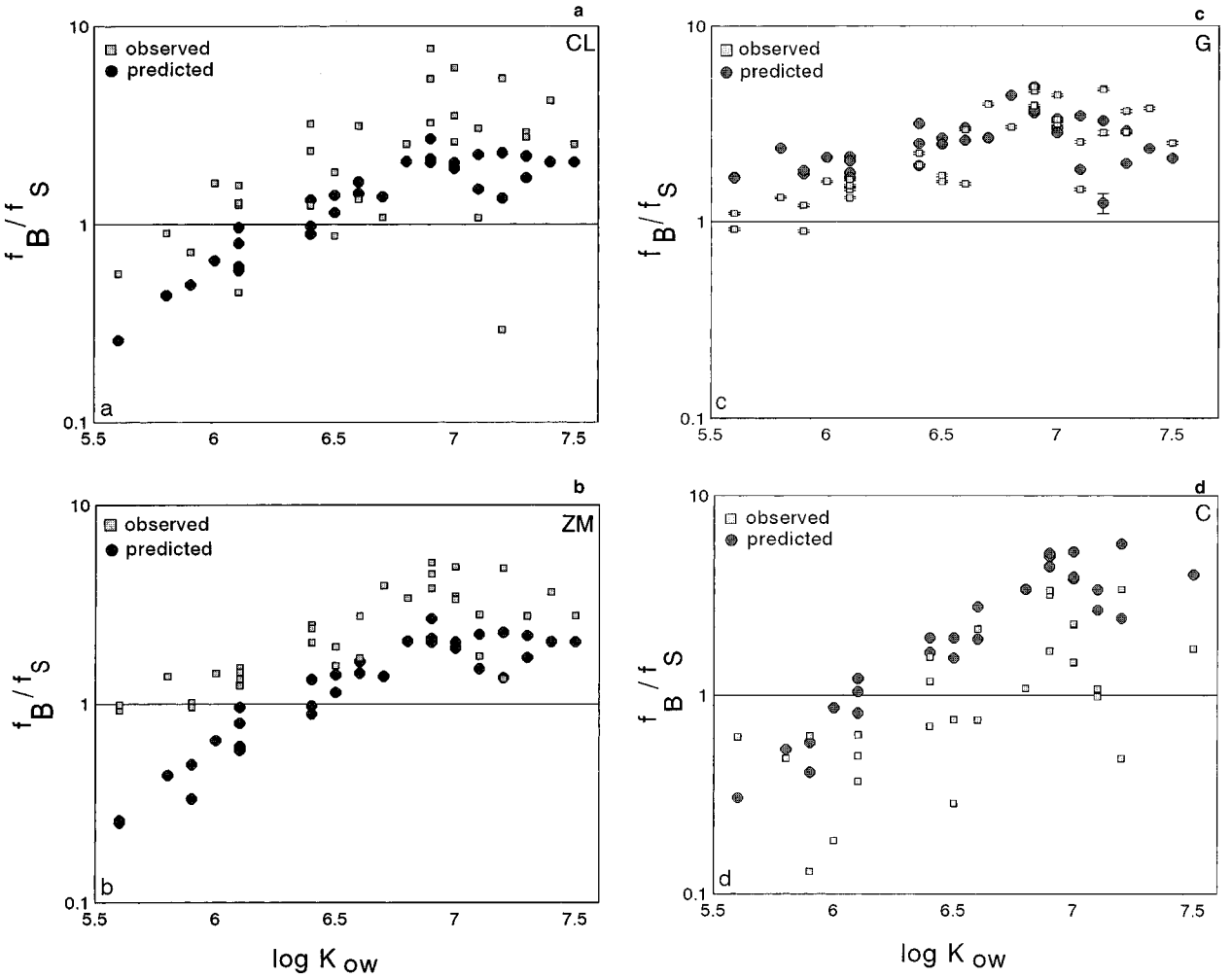

FIGURE 2. Mean observed and predicted fugacity ratios for individual PCB congeners in (a) caddisfly larvae (CL), (b) zebra mussels (ZM), (c) *Gammarus* (G), and (d) crayfish (C) from the western basin of Lake Erie. The line represents the predictions of the EP model.

TABLE 5

Change in Model Output Due to a 10% Decrease in Input Parameter Values[a]

	% change (±SE)			
parameter	caddisfly larvae	zebra mussels	*Gammarus*	crayfish
f_W/f_S	−0.4 (± 0.73)	−0.4 (± 0.73)	0	−0.2 (± 0.94)
f_D/f_S	−7.8 (± 0.70)	−7.8 (± 0.70)	−10.0 (± 0.01)	−8.2 (± 0.69)
E_W	3.6 (± 0.35)	3.6 (± 0.36)	0.2 (± 0.05)	3.8 (± 0.38)
E_D	−3.6 (±0.32)	−3.6 (± 0.32)	−0.3 (± 0.05)	−3.8 (± 0.34)
α	5.7 (± 0.49)	5.7 (± 0.49)	10.0 (± 0.06)	5.9 (± 0.51)
G_W			0.2 (± 0.05)	4.6 (± 0.71)
G_D			−0.3 (± 0.05)	−2.9 (± 0.63)
δ_D	−2.6 (± 0.65)	−2.6 (± 0.65)	−0.2 (± 0.05)	−2.9 (± 0.63)
ϕ_D	−4.0 (± 0.72)	−4.0 (± 0.72)	−0.3 (± 0.06)	−3.9 (± 0.69)
V_{pl}	−2.6 (± 0.65)	−2.6 (± 0.65)		

[a] n = 29, 30, 31, and 25 for caddisfly larvae, zebra mussels, *Gammarus*, and crayfish, respectively. A sensitivity value of magnitude less than 0.05 was reported as zero.

for every benthic invertebrate (Table 5). Conversely, the fugacity ratio between water and sediment was the least sensitive for all organisms. The sensitivity of model output to diet-related parameters indicates that it is important to account for different feeding preferences because different diets are likely to differ in their digestibility and extent of equilibria (disequilibria) with sediment.

Because model output is most sensitive to diet-related parameters, discrepancies between predicted and observed organism-to-sediment fugacity ratios may be due to unrealistic diet assignments. For instance, the fugacity ratios for crayfish to sediment were systematically overpredicted by the model. The observed fugacity ratios of the crayfishes' assigned diet to sediment were, on average, the highest of the four benthic invertebrates. It is possible that the composition of the diet and/or the chemical concentrations measured in the diet at the time of collection do not reflect the actual diet of these crayfishes. Systematic underpredictions of the model for caddisfly larvae and zebra mussels may also be due to the same phenomena.

In conclusion, this steady-state model, which incorporates disequilibria, biomagnification, and species' specific feeding strategies, better describes bioaccumulation of hydrophobic contaminants by benthic invertebrates as compared to the EP model. Furthermore, this model highlights three important aspects of chemical accumulation in benthic invertebrates. First, chemical disequilibria between diet, water, and sediment can have an important effect on the extent of chemical bioaccumulation in benthic invertebrates. Second, biomagnification or dietary accumulation can raise the BSAF several fold over its thermodynamically controlled partition coefficient of approximately 1−2. Third, feeding strategy and prey digestibility are important factors controlling the BSAF. Finally, the model illustrates how the various factors interrelate and control the BSAF. Because the model is generic and adjustable to reflect site-specific conditions, it may be useful for developing site-specific water quality criteria and related regulatory purposes.

Acknowledgments

We thank Katy Haralampides, Geoff Riddle, and the staff of the analytical laboratory of the Great Lakes Institute for Environmental Research for their help. We also thank D. M. Whittle for his support. We are grateful for the financial support provided by an NSERC-Environment Canada (GLURF) grant to G.D.H. and F.A.P.C.G. and by the Department of Fisheries and Oceans, Greenplan Toxic Chemical Program to D.M.W.

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Received for review March 28, 1996. Revised manuscript received June 18, 1996. Accepted June 25, 1996.[⊗]

ES960280B

[⊗] Abstract published in *Advance ACS Abstracts,* September 15, 1996.