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Calibration of the Gastrointestinal Magnification Model to Predict Maximum Biomagnification Potentials of Polychlorinated Biphenyls in a Bird and Fish

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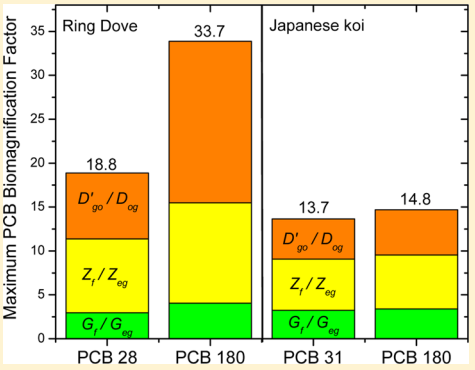
Supporting Information

ABSTRACT: The gastrointestinal magnification (GI-magnification) model was calibrated in ring doves and Japanese koi using matched data on dietary assimilation and fecal depuration of polychlorinated biphenyls (PCBs). Mass transport parameters describing PCB flux from gut contents to organism (D'_{go}; mol d^{-1} Pa^{-1}) and organism to gut contents (D_{og}; mol d^{-1} Pa^{-1}) were quantified to test the hypothesis that the ratio of these two terms approached unity. For birds, D'_{go}/D_{og} ranged from 2.9 to 6.3 and for fish the ratios ranged from 0.7 to 3.1. In both species, the ratio commonly exceeded 1. The GI-magnification model was used to predict maximum PCB biomagnification factors (BMF_{max}) for each species which ranged from 18.5 to 33.8 for ring doves and 7.9 to 14.8 for Japanese koi. Chemical losses via respiration reduced steady state biomagnification factor (BMF_{ss}) estimates by a negligible amount in birds, whereas for fish, predicted BMF_{ss} decreased to values from 0.5 to 7.2. This study demonstrated that chemical transfer efficiency during assimilation exceeds organism/feces transfer which contributes to elevated PCB biomagnification potentials in birds and fish. Combined with reduced losses of chemical across respiratory surfaces, higher D'_{go}/D_{og} ratios of birds contribute to elevated biomagnification in birds over fish.



INTRODUCTION

Food web biomagnification refers to the condition whereby chemicals increase in concentration with increasing trophic status of organisms in a food web.[1,2] Connolly and Pederson[3] further demonstrated that hydrophobic persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs), not only increase in chemical concentration, but also exhibit increases in chemical fugacity with animal trophic status; an observation that has been verified in numerous ecosystems.[4−7] In order to distinguish biomagnification from equilibrium partitioning bioaccumulation mechanisms (e.g., bioconcentration), Connolly and Pederson[3] redefined the term biomagnification for organic chemicals as the condition whereby the chemical fugacity in an organism exceeds the chemical fugacity of the respired media and food the animal consumes.

Gobas et al.[8] developed the gastrointestinal magnification model (GI-magnification model), which provides a mechanistic description of the biomagnification process applied to POPs. This model predicts biomagnification to be a consequence of reductions in the feces partitioning capacity and fecal egestion rates of organisms relative to the elevated fugacity capacity of ingested material and feeding rate of the animal. Combined, these processes increase the chemical fugacity of gastrointestinal (GI) contents as ingested materials proceed along the digestive tract where chemical fugacity becomes maximized in the lower intestine and feces.[8−10] As a result, the organism is hypothesized to approach equilibrium with the elevated chemical fugacity of the GI-contents rather than the food ingested or media respired. A number of predictions of the GI-magnification model have been tested and verified as exemplified below. Bioaccumulation studies on POPs in fish demonstrated that the chemical fugacity of feces and GI-contents are raised above food and animal during the initial uptake phase of the bioaccumulation curve.[10,11] Field studies demonstrated that chemical fugacities of POPs in fish exceeded chemical fugacities in collected food items.[10,12] Studies on humans demonstrated that chemical fugacity ratios of blood to food for POPs were consistently greater than one.[13] The GI-magnification model has subsequently been incorporated into several food web bioaccumulation models describing the trophodynamics of POPs in aquatic[14−16] and terrestrial[6,17] ecosystems.

To date the GI-magnification model has been calibrated for only a limited number of species.[10,11,18] Furthermore, some model assumptions commonly used to simplify calculations have not been tested. A critical assumption commonly used in the GI-magnification model is that the mass transport parameter describing chemical flux from the GI-contents to the animal (D_{go}) is equivalent to the mass transport parameter

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describing animal/feces transfer (D_{og}). This assumption is based on the hypothesis that both mass transport parameters are controlled by molecular diffusion of chemical in unstirred water layers (UWL) surrounding enterocytes of the small intestine.[8,10,11] Since uptake and elimination processes occur simultaneously along the GI-tract and are subject to the same UWL dimensions, the two terms are assumed to be equal. However, the assumption that $D_{go} = D_{og}$ has been questioned given understanding of physiological mechanisms governing long chain fatty acid, fat-soluble vitamin and hydrophobic drug assimilation in the GI-tract.[13,18−20] Micelle-mediated diffusion[20] and the "fat flush" effect[13] have been put forward as mechanisms to explain why diffusion from GI-contents to organism may be more efficient than animal/feces transfer.[18] Modification to the GI-magnification model incorporating the above changes have been proposed which involve defining an additional mass transport term (D_{mic}) between animal and GI-contents to account for chemical uptake from GI-contents via micelle-mediated diffusion in the uptake direction, but not allowing D_{mic} to participate in animal/feces depuration.[18,21,22] The effect of this modification is to allow ($D_{go} + D_{mic}$) to exceed D_{og} and therefore establish a maximum biomagnification potential for the animal.[7,18,21] In the present study, ($D_{go} + D_{mic}$) was grouped together into the single term D'_{go} to facilitate model solution and parameter calibration. This enabled testing the hypothesis that $D'_{go} = D_{og}$ or the ratio $D'_{go}/D_{og} = 1$ in two calibrated model organisms.

The objective of this study was to quantify the mass transport parameters D'_{go} and D_{og} for individual PCB congeners in a bird and fish species in order to test the hypothesis that $D'_{go}/D_{og} = 1$. A second objective was to complete the calibration of the GI-magnification model for the two species to estimate species specific maximum biomagnification factors (BMF_{max}) and steady state biomagnification factors (BMF_{ss}) in the two species to determine major causal factors related to differences in biomagnification potentials in a model bird and fish.

METHODS

Model. The GI-magnification model has been described elsewhere.[8,10,11] The model is further summarized in Supporting Information along with equations and assumptions associated with model solutions (Figure S1 and Tables S1, S2). For brevity, only the key equations tested and utilized are presented here. Assuming steady state is achieved between the animal and its environment and between the animal and its gut contents, the chemical fugacity in the animal is calculated by

$$f_{o(ss)} = [f_{w/a}\cdot D_{w/a}\cdot(D'_{go} + D_{eg}) + f_f\cdot D_f\cdot D'_{go}] / [D_{w/a}\cdot(D'_{go} + D_{eg}) + D_M\cdot(D'_{go} + D_{eg}) + D_{Gro}\cdot(D'_{go} + D_{eg}) + D_{og}\cdot D_{eg}] \quad (1)$$

where $f_{o(ss)}$, $f_{w/a}$, and f_f are the steady state chemical fugacities (Pa) in the organism, respired water (or air) and ingested food, $D_{w/a}$, D'_{go}, D_{eg}, D_f, D_M, D_{Gro}, and D_{og} are the mass transport parameters (mol Pa^{-1} d^{-1}) describing chemical flux between water (or air) and organism, gut contents to organism, loss of chemical to feces, intake of chemical via feeding, loss of chemical by biotransformation, growth dilution of chemical and organism to gut tract chemical exchange. The steady state biomagnification factor (BMF_{ss}) in the animal is subsequently calculated as

$$BMF_{ss} = \frac{f_{o(ss)}}{f_f} \quad (2)$$

The maximum biomagnification factor of an animal (BMF_{max}) is estimated under the condition where no chemical elimination occurs to respired media, biotransformation or growth dilution (i.e., $D_{w/a}$, D_M, and D_{Gro} approach 0). Under this condition, eq 1 is solved and rearranged to yield BMF_{max}

$$BMF_{max} = \frac{D_f\cdot D'_{go}}{D_{eg}\cdot D_{og}} = \frac{G_f\cdot Z_f\cdot D'_{go}}{G_{eg}\cdot Z_{eg}\cdot D_{og}} \quad (3)$$

The terms D_f and D_{eg} are readily measured experimentally and computed from the animal feeding (G_f; m^3 d), fecal egestion rates (G_{eg}; m^3 d), and fugacity capacities (mol m^{-3} Pa^{-1}) of ingested food (Z_f) and feces (Z_{eg}), respectively. The terms D'_{go} and D_{og} are more difficult to measure, but can be estimated with specific experimental measurements. During the initial uptake phase of a bioaccumulation study, when the organism chemical concentration approximates zero, the chemical assimilation efficiency from food is maximized (AE_{max}) and can be related to D'_{go} according to

$$D'_{go} = \frac{f_{f(up)}}{f_{g(up)}}\cdot AE_{max}\cdot D_f = \frac{C_f}{C_{eg}}\cdot AE_{max}\cdot G_f\cdot Z_{eg} \quad (4)$$

where $f_{f(up)}$ and $f_{g(up)}$ are chemical fugacities (Pa) in food and feces determined during the initial portion of the uptake study, G_f is the long-term feeding rate of the animal (m^3 d), Z_f and Z_{eg} are the fugacity capacities (mol m^3 Pa^{-1}) of ingested food and feces, C_f and C_{eg} represent the respective chemical concentrations (mol/m^3) in food and feces (See Supporting Information Table S1 and S2 for origin of eq 4). The AE_{max} is commonly measured by mass balance (Supporting Information Table S2), whereby an uncontaminated animal is provided a single contaminated meal and the assimilated fraction is measured either in the animal's body following fasting or subtracted from the chemical concentration determined in feces.

The term D_{og} can be examined under depuration conditions when a contaminated animal is provided the same type of food (containing no chemical) as provided in the dietary chemical assimilation study. During depuration, when the chemical concentration in food approaches zero and the only source of chemical to feces is from the animal, the term D_{og} is calculated by

$$D_{og} = \frac{f_{g(dep)}}{f_{o(dep)}}\cdot(D'_{go} + G_{eg}\cdot Z_{eg}) \quad (5)$$

where G_{eg} is the long-term fecal egestion rate (m^3 d^{-1}) of the animal and Z_{eg} is the fugacity capacity of feces (mol m^{-3} Pa^{-1}). In this case, the chemical fugacities in feces ($f_{g(dep)}$) and organism ($f_{o(dep)}$) refer to measurements taken under the experimental condition of depuration. The term D'_{go} is derived independently from the uptake study described above.

Henry's Law Constants (m^3 Pa^{-1} mol^{-1}; at 25 °C) and chemical K_{OW} values necessary to estimate fugacity capacities were obtained from Phillips et al.[23] and Hawker and Connell.[24] Nonlipid organic matter, calculated as the fraction of dry weight in the sample minus the fraction of lipid weight, was assumed to have a partition capacity equivalent to 5% of lipids.[25]

Experimental Data. Compatible uptake and depuration data for PCBs necessary to compute D'_{go} and D_{og} were available for ring doves (*Streptopelia risoria*) and Japanese koi (*Cyprinus carpio*).[20,26−28] Additional details regarding these studies are summarized in Supporting Information.

Dietary AE_{max} for PCBs were determined for ring doves[20] (176 g body weight) fed pellets spiked with Aroclor PCBs (1242:1254:1260) at 25 °C. The AE_{max} was calculated by mass balance by measuring the difference in chemical ingested relative to loss of chemical to feces. Congener specific D'_{go} values were calculated from eq 4. A 105-d depuration study,[26] using ring doves (dosed by interperitoneal injection with PCBs) of similar body weight, fed similar noncontaminated food and housed under identical conditions was used to estimate D_{og} according to eq 5. The lipid content of food differed slightly, although significantly ($p < 0.05$; ANOVA), between the two experiments; for the AE study it was 8.4 ± 1.8% and the depuration study it was 9.1 ± 0.5%.

Dietary AE_{max} for PCBs for Japanese koi (10 g body weight) fed koi pellets spiked with PCBs and held in aquaria at 16 °C are presented by Liu et al (2010).[27] AE_{max} was calculated by mass balance from the difference in chemical ingested relative to the assimilated mass of chemical in whole body following a 48 h fast after being fed a contaminated meal. To calculate D'_{go}, (C_{eg} was not measured during the uptake study) C_{eg} was estimated by assigning the unassimilated chemical mass to feces. The proximate composition of feces, used to measure Z_{eg}, was assumed to be the same as that measured in the depuration study (see below) where fish were provisioned with the same food. The difference in proximate composition (moisture, lipid and lean dry weight) of food and feces was used to establish a dry food digestion efficiency of 0.43. The mass of feces produced (E) was estimated as $(1 - 0.43)\cdot M_f$, where M_f is the mass of dry food ingested during the meal.

The fish fecal depuration study is described in Paterson et al.[28] Japanese koi (81.4 ± 3.0 g body weight) previously dosed with PCBs by intraperitoneal dose were placed in aquaria at 16 °C. Fish were switched between feeding and fecal collection aquaria daily in order to collect feces and establish a fecal mass balance for PCBs over the study duration. Owing to the small amount of feces collected, feces were pooled together from fecal collection tanks over 0−35, 35−70, 70−105, and 105−140 d time intervals. Pooled feces was used to measure $C_{eg(dep)}$, % moisture, and % lipid necessary to compute $f_{g(dep)}$. Five fish were sacrificed on days 0, 35, 70, 105, and 140 and homogenized to measure carcass $C_{o(dep)}$, % moisture, and % lipid necessary to calculated $f_{o(dep)}$. $C_{o(dep)}$ for PCBs was corrected for individual growth rates.[28] To match the pooled $f_{eg(dep)}$ and $f_{o(dep)}$, the average fish PCB concentration and Z_o across each time interval was determined using fish sacrificed at the beginning and end of the fecal collection intervals.

The fish data were less well matched across studies owing to different sizes of fish used in experiments. In addition, the low water temperature (16 °C) used in both studies likely contributed to underestimates of long-term feeding and fecal egestion rates of koi compared to higher temperatures. To make these data more comparable to ring dove studies, a bioenergetics model was used to estimate a size and temperature standardized feeding rate (G_f) and fecal egestion rates (G_{eg}) for koi which was used in conjunction with eqs 4 and 5 to estimate D'_{go} and D_{og}. The bioenergetics model was computed to estimate long-term feeding and fecal egestion rates for a 176 g fish at 25 °C to make D'_{go} and D_{og} consistent with the data for ring doves. The approach requires the following assumptions: (1) AE_{max} of 8−10 g fish used in the uptake study is similar to the AE_{max} of 81 g fish used in depuration studies; (2) AE_{max} and $f_{g(dep)}/f_{o(dep)}$ are assumed to be similar for the model 176 g koi acclimated to 25 °C.

The bioenergetic model used the relationship reported by Oikawa and Itazawa[29] to estimate oxygen consumption rates (μL O_2 min^{-1}) at 25 °C in common carp (*Cyprinus carpio*) based on respirometry trials

$$R_{O_2} = 4.53\cdot M_o^{0.827} \quad (6)$$

where R_{O_2} is the standard metabolic rate of oxygen consumption (μL $O_2\cdot fish^{-1}\cdot min^{-1}$) and M_o is the body mass of the organism (176 g). Oxygen consumption rates were converted to a standard metabolic rate (SMR) of 8.80 kJ $fish^{-1}$ d^{-1} using an oxygen gas density of 1.31 g L^{-1} and oxycalorific coefficient[30] of 14.3 kJ g^{-1} O_2^{-1}. The algorithms reported by Drouillard et al.[30] for yellow perch were used to estimate additional daily energy requirements due to specific dynamic action, energy excretion and activity (activity multiplier = 1) estimated from SMR to calculate a total daily energy demand of 11.14 kJ $fish^{-1}$ d^{-1}. The latter was divided by the food energy density of koi pellets (9.7 kJ g^{-1}) to establish a long-term daily feeding rate of 1.15 g food $fish^{-1}$ day^{-1} for a 176 g fish at 25 °C. Long-term fecal egestion rates were estimated to be 0.66 g feces $fish^{-1}$ d^{-1} based on the dry weight food digestion efficiency.

Calculation of BMF_{SS} in Birds and Fish. Steady state biomagnification factors were computed for nonbiotransformed PCBs to demonstrate how biomagnification is attenuated by loss of chemical to respired media. Gill ventilation rates (G_v; m^3 $fish^{-1}$ d^{-1}) were computed to estimate D_{wo} (see Supporting Information Table S2). The size (176 g) and temperature (25 °C) standardized total daily energy demand of Japanese koi (11.14 kJ $fish^{-1}$ d^{-1}) was reconverted into a daily oxygen consumption rate (0.78 g O_2 $fish^{-1}$ day^{-1}) using the oxycalorific coefficient. The daily oxygen consumption rate was divided by an average oxygen concentration of water (0.008 g L^{-1}) and oxygen extraction efficiency across the gills (0.6)[30] yielding a G_V of 0.162 m^3 water d^{-1}. Metabolic biotransformation of PCBs was assumed zero for fish.[28,31] Daily growth was assumed to be zero since concentration terms used to specify D_{og} were growth corrected.

For ring doves, a daily energy budget was estimated as the difference in caloric intake of food minus caloric value of feces calculated as 137.6 kJ $bird^{-1}$ d^{-1}. The daily energy intake was converted to an oxygen consumption rate using the same oxycalorific coefficient of fish, dividing by an average oxygen concentration of air (0.268 g L^{-1}) and oxygen extraction efficiency across the avian lung[32] of 0.55, yielding a G_V of 0.065 m^3 air d^{-1}. Metabolic biotransformation of PCBs was assumed to be zero for PCB congeners which have no open meta-, para-positions (i.e., PCB congeners having chlorine substitutions at the following positions: 4,4′; 3,4′,5; 3′,4, 5′ or 3,3′,5,5′).[33] Growth rates were assumed to be zero for adult ring doves.

BMF_{ss} simulations used f_f (fugacity of food) equivalent to the fugacity of PCBs measured in food used in the uptake studies for fish and birds. The fugacity of air (f_a) and water (f_w) were set at $0.2\cdot f_f$ to account for biomagnification in food items.

Uncertainty Propagation and Statistical Analysis. Standard deviations surrounding D'_{go} and D_{og} were computed using Monte Carlo techniques. Simulations used the mean and standard deviations as inputs for all measured parameters used

to compute D'_{go} (9 individual variables with error estimates) and D_{og} (7 variables plus error related to the D'_{go} estimate). Following initial evaluation of data, a log-normal distribution was selected and all simulations were performed for 1000 iterations. The standard deviation from Monte Carlo output across 1000 trials was used to estimate the variability around a given value of D'_{go} or D_{og}.

General linear models (GLM) were used to examine differences in D'_{go} and D_{og} across congeners and to build predictive models for these parameters based on chemical K_{OW}. Data were log-transformed prior to analysis after determining that log transformation was necessary to satisfy normality. Only congeners for which both D'_{go} and D_{og} were available in a given organism were included in the GLM. The GLM was first assessed to test for K_{OW}·Group interactions, that is, differences in the slope between D'_{go} and D_{og} with chemical K_{OW}. Where slopes were not different, the interaction term was removed from GLM to test for differences between D'_{go} and D_{og} (identified in the model as Group) and K_{OW} effects. A multiple regression model was generated to establish a predictive equation for both D'_{go} and D_{og}

$$\log(D'_{go} \text{ or } D_{og}) = A\cdot\log K_{OW} + B\cdot\text{Group} + \text{Constant} \quad (7)$$

where A, B, and Constant are fitted coefficients from the GLM and Group is a dummy variable given a value of 1 for D'_{go} estimates and 0 for D_{og}.

Given that PCBs are covariates of one another (i.e., the different chemicals were measured in the same set of samples) and that D'_{go} and D_{og} contained similar measurements in their computation (lack independence), testing for statistical differences between the two terms could not be directly performed. In lieu of statistical tests, sample size computations, based on power analysis, were performed to evaluate the number of replicates that would be required to detect a significant difference ($p < 0.05$; using a two sample t test) between D'_{go} and D_{og} for a given congener and organism. Sample size calculations assumed that computed D'_{go} and D_{og} values represent true population means and used the standard deviations from Monte Carlo simulations under a specified power of 0.5. As a general rule, if less than 5 independent replicate measures of D'_{go} and D_{og} were required for a given chemical to detect a statistical difference, a high likelihood of differences between computed values of D'_{go} and D_{og} was assigned.

RESULTS AND DISCUSSION

Dietary AE_{max} values were measured for 55 and 42 PCBs in ring doves and Japanese koi, respectively. The AE_{max} and D'_{go} estimates and their calculation are provided in Supporting Information (Tables S3 and S4). Depuration studies of fecal egestion of PCBs were available for 18 and 37 congeners for ring doves and Japanese koi and their calculations are outlined in Supporting Information (Tables S5 and S6). Figure 1 summarizes D'_{go} and D_{og} values measured in birds and fish along with standard deviations computed for each term from Monte Carlo simulations.

For ring doves, the initial GLM indicated no significant differences between slopes ($p > 0.3$; $n = 36$) of D'_{go} and D_{og} with log K_{OW}. After removal of the interaction term from the model, there was a highly significant ($p < 0.001$; t test on coefficient) K_{OW} dependence of parameters and a highly

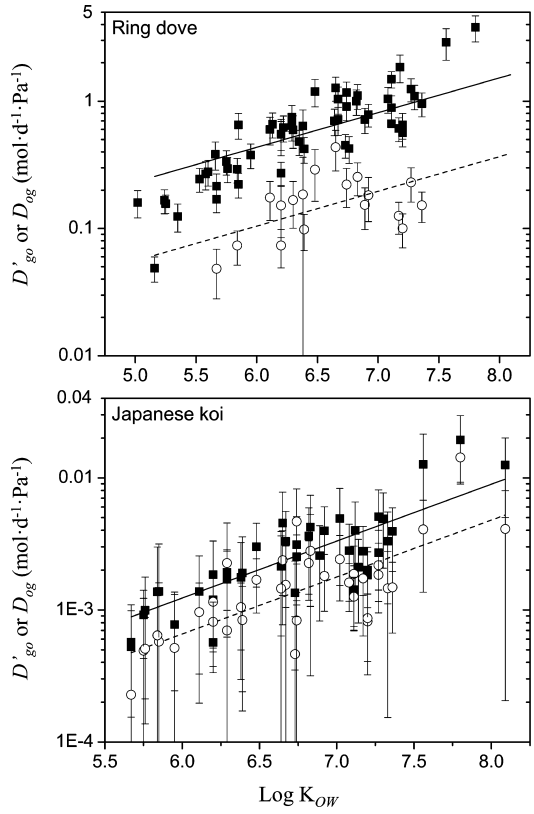

Figure 1. Mass transport parameters (D'_{go} and D_{og}) verses PCB hydrophobicity. D'_{go} represented by (■) and D_{og} represented by (○). Error bars represent standard deviations for D'_{go} or D_{og} estimated using Monte Carlo methods. Solid and dashed lines reflect linear regression fits to D'_{go} and D_{og} data, respectively. Top graphic presents data for ring doves, bottom graphic Japanese koi.

significant ($p < 0.001$; t test) effect of parameter type (D'_{go} and D_{og}). The fitted multiple regression equation was

$$\log(D'_{go} \text{ or } D_{og}) = 027 \pm 0.07\cdot\log K_{OW} + 0.62 \pm 0.07\cdot\text{Group} - 2.62 \pm 0.46;\ R^2 = 0.74 \quad (8)$$

Figure 1 presents the fit of eq 8 to all estimates of D'_{go} and D_{og} determined for ring doves. The fit was generally better for D'_{go} compared to D_{og}, although an over prediction for D'_{go} for PCB 16 + 32 and under predictions for PCBs 195 and 194 were noted. On the basis of the Group coefficient in eq 7, the model predicts D'_{go} to be 4.15 fold higher than D_{og}. This matches the mean ± standard deviation D'_{go}/D_{og} ratio across congeners calculated to be 4.23 ± 0.87 (range 2.92 to 6.28; Table 1). Sample size computations using power analysis indicated that <3 replicate samples would be needed to detect a statistical difference between D'_{go} and D_{og} for a given chemical (Supporting Information Table S5). These observations provide high confidence that the ratio of D'_{go}/D_{og} exceeds the theoretical value of one in ring doves.

For koi, the initial GLM revealed no significant differences ($p > 0.6$; $n = 74$) in the slope between D'_{go} and D_{og} and chemical K_{OW}. After removing the interaction term, there was a highly significant ($p < 0.001$; t test) dependence of parameters on K_{OW}, as well as highly significant ($p < 0.001$; t test) differences between parameter type. The multiple regression equation for the koi data was

Table 1. Maximum Biomagnification Factors (BMF_{max}) and Steady State Biomagnification Factors (BMF_{ss}) for Matched Polychlorinated Biphenyl (PCBs) Congeners in Ring Doves and Japanese Koi[a]

ring dove studies					
chemical	D'_{go}/D_{og}	Z_f/Z_{eg}	G_f/G_{eg}	BMF_{max}	BMF_{ss}
PCB 28	3.50	3.91	1.38	18.8	18.6
PCB 52	3.96	3.91	1.38	21.3	metabolized
PCB 66	3.61	3.91	1.38	19.4	19.4
PCB 56/60	3.43	3.91	1.38	18.5	18.4
PCB 99	4.29	3.91	1.38	23.1	23.0
PCB 101	3.44	3.91	1.38	18.5	metabolized
PCB 110	4.09	3.91	1.38	22.0	metabolized
PCB 118	4.10	3.91	1.38	22.1	22.0
PCB 105	2.92	3.91	1.38	15.7	15.7
PCB 153/132	4.30	3.91	1.38	23.1	23.1
PCB 138	4.36	3.91	1.38	23.5	23.5
PCB 187/182	4.88	3.91	1.38	26.3	26.2
PCB 183	5.63	3.91	1.38	30.3	30.2
PCB 170/190	5.41	3.91	1.38	29.1	29.1
PCB180	6.28	3.91	1.38	33.8	33.7
Japanese koi studies					
chemical	D'_{go}/D_{og}	Z_f/Z_{eg}	G_f/G_{eg}	BMF_{max}	BMF_{ss}
PCB 31	2.47	3.16	1.75	13.7	0.5
PCB 52	2.14	3.16	1.75	11.9	0.8
PCB 66	1.46	3.16	1.75	8.1	1.1
PCB 56/60	1.43	3.16	1.75	7.9	0.9
PCB99	2.27	3.16	1.75	12.6	1.9
PCB 101	1.68	3.16	1.75	9.3	1.7
PCB 110	1.78	3.16	1.75	9.9	1.8
PCB 118	3.03	3.16	1.75	16.8	3.0
PCB 105	1.91	3.16	1.75	10.6	2.5
PCB 153/132	2.20	3.16	1.75	12.2	4.3
PCB 138	1.51	3.16	1.75	8.4	3.1
PCB 187/182	1.61	3.16	1.75	8.9	4.8
PCB 183	2.24	3.16	1.75	12.4	5.3
PCB 170/190	2.33	3.16	1.75	12.9	6.5
PCB 180	2.66	3.16	1.75	14.8	7.2

[a]PCBs identified by IUPAC number. Chemicals with multiple congeners were co-eluting during analysis. PCB31 in fish was assumed to match with PCB28 on the basis of similar log K_{OW}. PCB 52, 101, and 110 are metabolized by birds and therefore BMF_{ss} would be overestimated for these compounds.

$$\log(D'_{go} \text{ or } D_{og}) = 043 \pm 0.04 \cdot \log K_{OW} + 0.27 \pm 0.05 \cdot \text{Group} - 5.77 \pm 0.28;\ R^2 = 0.66 \quad (9)$$

Based on the Group coefficient in eq 8, the regression model predicts D'_{go} to be 1.87 fold higher than D_{og} in fish. The mean ± standard deviation D'_{go}/D_{og} ratio across congeners in koi was 1.97 ± 0.59 (range 0.67 to 3.11; Table 1). Only two chemicals (PCBs 97 and 129) had D'_{go}/D_{og} values less than 1 (0.85 and 0.68, respectively) in koi. For koi, the D_{og} values plus one standard deviation frequently overlapped model predictions for D'_{go} and vice versa (Figure 1). Only PCB 153 + 132 showed no overlap of error bars for both D'_{go} and D_{og} and its counterpart GLM prediction. Sample size computations indicated variable results across congeners (Supporting Information Table S6). Twelve of the 37 congeners would require between 3 and 5 replicate samples to detect statistical differences between a given value of D'_{go} and D_{og}. This included PCBs 110, 118, 153 + 132, 179, 174, 183, 170 + 190, 180, 195, 199, and 206. Twelve congeners would require between 6 and 10 replicates per parameter to detect statistical differences and the remaining congeners required between 11 and 142 replicates (Supporting Information Table S6). After log transformation of sample sizes, there was a significant negative correlation ($p < 0.01$; Pearson's $r = -0.431$; $df = 35$) between the sample size needed to detect a significant difference between D'_{go} and D_{og} and log K_{OW}; indicating that the likelihood of true differences between D'_{go} and D_{og} increased with increasing chemical hydrophobicity.

Given the limited data on D'_{go} and D_{og}, it is difficult to make generalizations about these terms and how they vary across environments, changes in food composition and differences in digestive physiologies of organisms. Past studies have shown considerable variation in AE_{max} of fish when exposed to POPs via different diets.[27,34] Diet characteristics, such as energy density and undigested organic components, also contribute to differences in feeding rates and fecal fugacity capacities which impact the magnitude of D'_{go}. The D_{og} term is inversely proportional to the organism/feces fugacity gradient achieved during depuration. Several studies demonstrated addition of nondigestible organic constituents to the diet enhances fecal excretion of POPs.[26,35,36] Moser and McLachlan[36] demonstrated that the magnitude of enrichment of fecal elimination of POPs (defined as the excretion enhancement factor) in humans was not uniform across chemicals following amendment of the diet of volunteers with a nondigestible lipid surrogate, olestra. The greatest enhancement of fecal elimination (8- to 10-fold over controls) occurred for chemicals with log K_{OW} values between 5.5 and 6.5. Beyond log K_{OW} = 6.5, the excretion enhancement factor dropped with further increases in chemical hydrophobicity. Given that nondigestable organic matter is expected to alter Z_{eg} values for different POPs in a similar fashion, the observed changes in excretion enhancement factors must be explained as kinetic limitations of D_{og}.

Although the above studies indicate that D'_{go} and D_{og} can be manipulated by diet, it is not known if such manipulations can cause shifts in the ratio D'_{go}/D_{og}. Drouillard and Norstrom[26] provided evidence to suggest that the mass transport terms can be decoupled. Ring doves, previously dosed with Aroclor-PCBs, were given either high fat or low fat diets containing non-Aroclor PCBs permitting simultaneous chemical uptake and elimination rate measurements. The study demonstrated similar AE_{max} across diets, but higher feeding rates for birds fed the low fat diet resulted in 1.4- to 1.6-fold higher PCB uptake rates. Fecal egestion rates of PCBs was also lower in the low fat diet birds despite higher fecal egestion rates because of lower feces/carcass PCB distribution ratios. As such, estimated BMF_{ss} in birds fed the low fat treatment were approximately 2-fold higher compared to birds fed high fat food.

Table 1 summarizes ratios of Z_F/Z_{eg} and G_f/G_{eg} in birds and fish. For birds, the ratios of Z_F/Z_{eg} and G_f/G_{eg} were 3.91 and 1.38, respectively. For fish, the ratios of Z_F/Z_{eg} and G_f/G_{eg} were 3.16 and 1.75, respectively. Between the species, birds had slightly lower food digestion efficiencies and somewhat higher differences in food to feces partition capacities. These differences canceled one another out and produced similar D_f/D_{eg} (eq 3) ratios of 5.4 and 5.5 for birds and fish, respectively. Gobas et al.[9] reported a higher dry matter food assimilation efficiencies in small (1.3 g) goldfish (*Carassius auratus*) fed Tetramin fish flake treatments having different lipid contents (<0.2 to 13.5% lipid across three diet treatments) that ranged from 60 to 76% compared to the dry matter food

assimilation efficiency measured in the present study (43%). These differences are related to differences in the proximate composition of koi pellets compared to Tetramin fish flakes. The commercial koi food contains 8% clay and 4% non-digestible fiber while fish flakes are derived from ground up fish meal. For ring doves, dry matter food assimilation efficiency was 53% and somewhat lower than pigeons fed plant −based pellets with dry matter assimilation efficiencies ranging from 59% (hemp seed) to 82% (corn),[37] but well within the range of dry matter assimilation efficiencies reported for wild birds feeding on seeds and fibrous plant matter.[38] The difference in partition capacity of food relative to feces was consistent with what has been reported in other studies. Gobas et al.[10,11] provided measured Z_f/Z_{eg} ratios for fish using vial equilibration methods. For guppies, goldfish and rainbow trout, Z_f/Z_{eg} ratios for various POPs averaged 4.92, 4.97, and 3.9, respectively. Drouillard[21] reported a measured Z_f/Z_{eg} of 4 for tetrachlorobenze in ring doves fed a similar high fat pellet ration as described in the present study.

Calculated BMF_{max} values for ring doves ranged from 15.7 to 33.8 and were positively related to log K_{OW} (Table 1 and Supporting Information Table S7; linear regression statistics, $R^2 = 0.64$; $p < 0.001$; ANOVA). For Japanese koi, the calculated BMF_{max} ranged from 3.8 to 17.3. Unlike birds, no relationship was found between BMF_{max} with chemical K_{OW} for koi (Table 1 and Supporting Information Table S8; linear regression statistics, $R^2 = 0.02$; $p > 0.2$; ANOVA). The mean BMF_{max} computed for Japanese koi was 10.91 ± 0.53. When BMF_{max} was compared for matched congeners in birds versus fish, the bird BMF_{max} averaged 2.1 fold higher than for fish. These differences in BMF_{max} are attributed to the higher D'_{go}/D_{og} of birds.

Calculations of BMF_{ss} in birds were restricted to PCBs categorized as negligibly metabolized. The computation of BMF_{ss} for ring doves and Japanese koi are provided in Supporting Information Tables S7 and S8. Selected values of BMF_{ss} are presented in Table 1. For ring doves, the difference between BMF_{ss} and BMF_{max} was negligible. This is supported by past observations that indicate fecal elimination of PCB 28 (a negligibly metabolized PCB) in ring doves was the same as whole body losses of this compound.[26] For fish, there were larger differences between BMF_{ss} compared to BMF_{max}, and these differences were inversely related to chemical log K_{OW}. Differences between BMF_{max} relative to BMF_{ss} as it relates to chemical losses across respiratory surfaces in fish have been reported elsewhere.[28,39]

Steady state BMFs for PCBs as a function of chemical hydrophobicity are presented in Figure 2. For both ring doves and Japanese koi, the linear regression between BMF_{ss} and log K_{OW} yielded a slope that was significantly different from 0 ($R^2 = 0.65$; $p < 0.001$ for ring doves and $R^2 = 0.80$; $p < 0.001$ for Japanese koi). BMF_{ss} ranged from 15.7 to 33.7 in birds and 0.5 to 7.2 in fish. Braune and Norstrom[40] reported PCB concentrations in Lake Ontario herring gulls and their food (alewives) to allow calculation of lipid normalized field BMFs in birds for persistent PCBs. BMF_{lipid} for persistent categorized PCBs from the above study ranged from 18 to 59 and was K_{OW} dependent. Van Wezel et al.[41] reported lipid normalized BMFs ranging from 2 to 180 from field studies in a range of bird species. In fish, lipid normalized biomagnification factors for PCBs in the range of 3 to 5 are commonly reported.[5,6,12,42−44] Model predictions of BMF_{ss} in birds and fish were consistent with literature reports. In both cases, higher food digestion

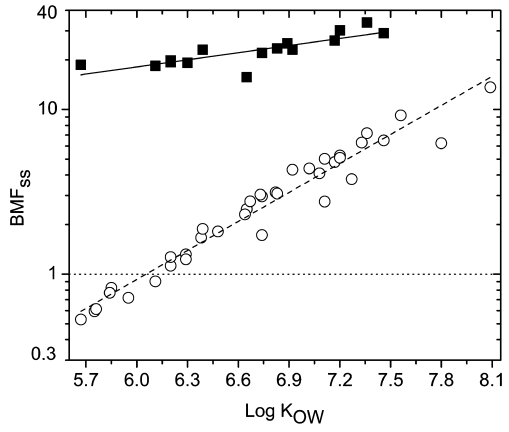

Figure 2. Steady state biomagnification factors (BMF_{ss}) versus PCB hydrophobicity. Data for ring doves represented by (■). Data for Japanese koi represented by (○). Solid and dashed lines reflect linear regression fits to ring dove and Japanese koi BMF_{ss} data, respectively.

efficiencies, as commonly observed in predatory birds and fish feeding on natural foods,[34,38] are likely to increase biomagnification potentials over those predicted for the calibrated organisms. Empirical studies demonstrating order of magnitude differences in BMF_{ss} for nonbiotransformed PCBs in birds compared to fish have been reported elsewhwere[43,45]

The GI-magnification model predicted BMF_{ss} for ring doves exceeded those of Japanese koi by factors ranging from 4.7 to 31.4-fold. Differences in steady state biomagnification potentials between the two species were K_{OW} dependent, such that the largest difference in biomagnification potentials occurred for lower K_{OW} PCBs. For the latter compounds, the difference in biomagnification potentials was mostly driven by species specific differences in chemical elimination across respiratory surfaces. For the highest K_{OW} PCBs, differences in biomagnification potentials between the two species were contributed to a similar extent by differences in respiratory losses of chemical and by differences the D'_{go}/D_{og} ratio. Kelly et al.[7] concluded that differences in the contribution of air compared to water as elimination vectors of POPs is a major reason for higher biomagnification potentials of terrestrial compared to aquatic organisms. This study reaffirms the above conclusion for low K_{OW}/negligibly biotransformed PCBs but also identifies elevated D'_{go}/D_{og} ratios in the birds to fish as a causal factor for high K_{OW} compounds.

The main hypothesis explored in this study was whether the ratio of D'_{go}/D_{og} approaches a value of 1 as commonly assumed in the simplified GI-magnification model. Unfortunately, the lack of independence of D'_{go} and D_{og} and covariation of PCBs place limitations on the ability to perform statistical tests on this ratio. Monte Carlo simulations were used to propagate uncertainty and estimate a standard deviation for each parameter. Sample size computations in conjunction with power analysis was then used in conjunction with D'_{go} and D_{og} values and their respective uncertainty as a as a proxy to ascertain whether the two parameters have a high degree of likelihood of being different or not. For ring doves, the average ratio of D'_{go}/D_{og} was 4.23 ± 0.87 across matched congeners and the small replicate size ($n < 3$) needed to detect statistical differences between the two terms for a given PCB imply that there is a high likelihood that this ratio is greater than 1. For Japanese koi, the average ratio of D'_{go}/D_{og} was 1.97 ± 0.59. In this case, the evidence was mixed. Approximately one-third of

the congeners were identified as having sufficient differences in computed D'_{go} and D_{og} values coupled with precise enough standard deviations to yield sample size calculations where a minimum replicate size ($n \leq 5$) would result in statistical differences between the two terms. A trend toward greater likelihood of differences between D'_{go} and D_{og} was also observed for more hydrophobic chemicals ($\log K_{OW} > 6.5$). This latter trend was a result of lower variation measured for these compounds (Monte Carlo estimated standard deviations) rather than differences in the magnitude of the D'_{go}/D_{og} ratio. Additional studies using consistent replicate sizes, more precise measurements of organism, food, and feces PCB concentrations, and proximate composition (lipids and nonlipid organic matter) of samples under matched uptake and depuration conditions would be useful to verify that $D'_{go}/D_{og} > 1$ for a larger number of chemicals in fish.

The observation of D'_{go}/D_{og} values exceeding 1 in birds and possibly in fish alters the conceptual premise of why POPs are able to biomagnify in animals and food webs. Under the simplifying assumption that $D'_{go}/D_{og} = 1$, the organism approaches equilibrium with the elevated fugacity generated in its GI-contents. However, in the more general model where $D'_{go}/D_{og} > 1$, there comes a point during bioaccumulation when the chemical must continue to transfer from GI-contents to organism even when the animal fugacity (f_o) exceeds the fugacity of chemical in the GI-contents (f_g). Schlummer et al.[13] observed this for humans and demonstrated that net positive dietary chemical assimilation continues to occur for several POPs even when the gut content fugacity exceeds the chemical fugacity in blood.

To explain their observations, Schlummer et al.[13] hypothesized a "fat flush" effect that interacts with GI-magnification. The "fat flush" effect considers fugacity capacity changes in gut tissues relative to animal and gut contents that result in temporary conditions of nonequilibrium between the organism and intestinal tissues. The effect is maximized during peak lipid assimilation post feeding, when the fugacity capacity of the gut contents are decreased raising f_g. At the same time, the enterocytes of the intestine are involved in the resynthesis of assimilated fatty acids into triglycerides, which temporarily increases Z and decreases chemical fugacity of enterocytes independent of the rest of the organism. This causes a favorable fugacity gradient to facilitate net chemical transfer into enterocytes even though the overall f_o/f_g exceeds 1. The "fat flush" effect is also thought to work in conjunction with the micelle-mediated diffusion, which considers the role of mixed micelles generated from bile salts and free fatty acids liberated from fats digested in the small intestine.[18−20] Mixed micelles are capable of dissolving hydrophobic chemicals into their interiors, and owing to their amphiphilic properties, traverse the UWL at faster rates than highly hydrophobic chemicals.[18−20] Furthermore, mixed micelles are believed to participate more strongly as uptake vectors than elimination vectors because the micelles interact with and are partially dissociated when they encounter a micro-pH gradient at the enterocyte surface.[18,46] Thus, micelle-mediated diffusion can explain the observation that $D'_{go} > D_{og}$, while the fat flush effect explains why it is possible for hydrophobic chemicals to continue to move against an apparent fugacity gradient between the organism and its gut contents.[18] Other physiological factors may complicate this process. For example, the contributions of microbial biomass associated with intestinal fauna and sloughing of intestinal epithelial cells to fugacity capacity changes of GI-contents are not explicitly considered by the GI-magnification model and would require additional compartments be added to consider their impacts.[47] These factors are not likely to alter the long-term magnitude of D'_{go}/D_{og} ratios, but may contribute to variation in measurements of these terms under experimental conditions described here. Additional research to characterize and quantify D'_{go}/D_{og} ratios in a greater diversity of species and to determine how environmental, physiological and ecological factors affect this ratio are necessary in to quantify the limits of POPs biomagnification in animals.

■ ASSOCIATED CONTENT

Supporting Information

Figure showing a conceptual overview of the GI-magnification model, tables summarizing the model equations associated with the GI-magnification model and the calculation of chemical fugacity, fugacity capacity, mass transport parameters, and the maximum assimilation efficiency, additional experimental details describing methods associated with the ring dove PCB assimilation and depuration studies and Japanese koi PCB assimilation and depuration studies, and tables presenting the calculation of D'_{go} and D_{og} for ring doves, the calculation of D'_{go} and D_{og} for Japanese koi, maximum (BMF_{max}) and steady state biomagnifications factor (BMF_{ss}) calculations for ring doves and Japanese koi, respectively. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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