- Supporting information -

Including Ecotoxic Impacts on Warm-blooded Predators in Life Cycle Impact Assessment

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Contents

1. The Model OMEGA

The model OMEGA estimates accumulation of organic compounds and metals in aquatic and terrestrial food chains. Here, a brief explanation of main processes and equations on accumulation of organic substances is given. More detailed information can be found in Hendriks and others (2001) and Hendriks and Heikens (2001). Standard food chains in OMEGA consist of four trophic levels. The mass of organisms in such food chains results from four basic flows (see Figure S1):

1) absorption and excretion of water
2) ingestion and egestion of food
3) (re)production of mass
4) mortality of tissues

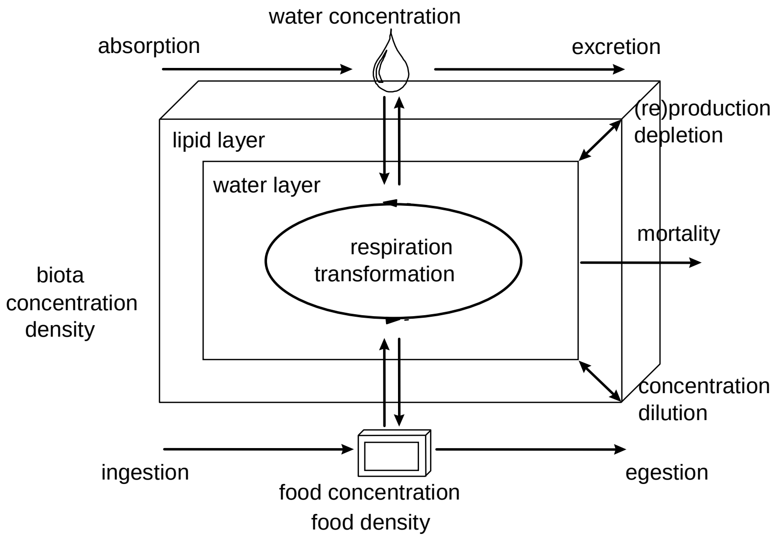


Figure S1. The densities of organisms and of their food are determined by metabolic flows at rate constants for absorption and excretion of water, ingestion and egestion of food, (re)production, respiration and mortality of mass. The concentrations in organisms and their food are determined by the lipid and water resistance as well as by the metabolic flows that carry substances into and out of organisms.

OMEGA calculates steady-state substance residues in biota as the sum of influx via water and uptake of food divided by the total elimination rate (Equation S1). Symbols are explained in Table S1. Different routes of elimination exist: efflux via water, food and biomass (growth dilution). Mostly, metabolic transformation is not explicitly accounted for, but in this study rate constants for metabolic transformation were based on the data of Arnot et others (2008). Furthermore, uptake and elimination via air were taken into account based on the calculations of Veltman and others (2009).

The concentration in an organism ($C_{x,i}$) is determined by a trophic-level-specific combination of all routes of uptake and elimination described in Equation S1.

$$C_{x,i} = \frac{k_{x,w,in,i} \cdot C_{x,w} + k_{x,f,in,i} \cdot C_{x,i-1} + k_{x,a,in,i} \cdot C_{x,a}}{\sum k_{x,w,out,i} + k_{x,f,out,i} + k_{x,p,out,i} + k_{x,m,out,i} + k_{x,a,out,i}} \qquad \textbf{(}S1)$$

Values for the rate constants in Equation S1 are derived by Hendriks and others (2001) and Veltman and others (2009). Rate constants for influx and efflux are predicted based on species-weight following allometric relationships. For organic substances, these constants are inversely proportional to resistances substances encounter in water and lipid layers and specific flows.

Defining the uptake and elimination rate constants requires taking into account the partitioning between the tissue or blood of an organism and the exchange compartments air or water. This partitioning depends on the fractions of water, proteins, polar and neutral lipids in the tissue or blood (Hendriks et al. 2005; Veltman et al. 2009). The relationships between these tissue or blood components and the K_{ow} of a substance differ for polar and nonpolar substances (Hendriks et al. 2005). For nonpolar substances (i.e. substances not containing H-bonding groups (Briggs 1981)), the tissue-water partition coefficient was defined as:

$$K_{tw} = p_{nl,t} \cdot K_{ow} + p_{pl,t} \cdot K_{ow}{}^{0.94} + p_{p,t} \cdot K_{ow}{}^{0.63} + p_{g,t} \cdot K_{ow}{}^{0.63} + p_{H2O,t} \cdot K_{ow}{}^{0} \qquad \text{(S2)}$$

and for polar substances (i.e. substances containing H-bonding groups (Briggs 1981)) as:

$$K_{tw} = 0.04 p_{nl,t} \cdot K_{ow} + 2 p_{pl,t} \cdot K_{ow}{}^{0.94} + 2.9 p_{p,t} \cdot K_{ow}{}^{0.63} + 2.9 p_{g,t} \cdot K_{ow}{}^{0.63} + p_{H2O,t} \cdot K_{ow}{}^{0} \qquad \text{(S3)}$$

Abbreviations are explained in Table S1. Using the fractions of water, proteins, polar and neutral lipids in blood instead of tissue, the blood-water partition coefficient can be calculated in a similar way.

The outflux rate constants $k_{x,w,out}$ and $k_{x,f,out}$ were calculated by multiplying the influx, as described by Hendriks and others (2001), with the inverse of tissue-water partition coefficient.

The affinity of substances for water, proteins and different lipids in the food was defined similar to the substances' affinity for these components in the tissue. Instead of the fractions of different components found in the trophic level of interest, the fractions of the trophic level below, i.e. i-1 were used. The affinity for these food components was used in the calculation of the influx rate constant $k_{x,f,in}$ and the outflux rate constant $k_{x,f,out}$ according to the work of Hendriks et al (2001) adjusted as described above.

Uptake and excretion via water were described by:

$$k_{x,w,in,i} = \frac{1}{\rho_{H_2O,w} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa} + \frac{\rho_{CH_2,i}}{K_{ow}} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa} + \frac{1}{\gamma_w} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa}} \qquad \text{(S4)}$$

$$k_{x,w,out,i} = \frac{1}{K_{tw}} \cdot \frac{1}{\rho_{H_2O,w} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa} + \frac{\rho_{CH_2,i}}{K_{ow}} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa} + \frac{1}{\gamma_w} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa}} \qquad \text{(S5)}$$

Uptake from food and egestion with faeces were described by: (S6)

$$k_{x,f,in,i} = \frac{p_f}{1 - p_f} \cdot \frac{1}{K_{tw,i-1}} \cdot \frac{1}{\rho_{H_2O,f} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa} + \frac{\rho_{CH_2,i}}{K_{ow} \cdot q_T} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa} + \frac{1}{K_{tw,i-1} \cdot (1 - p_f) \cdot \gamma_f \cdot q_T} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa}}$$

$$k_{x,f,out,i} = \frac{1}{K_{tw}} \cdot \frac{1}{\rho_{H_2O,f} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa} + \frac{\rho_{CH_2,i}}{K_{ow} \cdot q_T} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa} + \frac{1}{K_{tw,i-1} \cdot (1 - p_f) \cdot \gamma_f \cdot q_T} \cdot \left(\frac{w_i}{w^*}\right)^{\kappa}} \qquad \text{(S7)}$$

The outflux rate constant for growth dilution equaled:

$$k_{x,p,out,i} = \gamma_p \cdot q_T \cdot \left(\frac{w_i}{w^*}\right)^{-\kappa} \qquad \text{(S8)}$$

We used the outflux rate constant for biotransformation ($k_{x,m,out}$) in fish of the third trophic level presented by Arnot et al (2008) in EPI Suite ™ 4.0. The model calculates $k_{x,m,out}$ as a whole body value, namely the fraction of the mass in the whole body biotransformed per unit of time. The biological half-life, on which $k_{x,m,out}$ is based, is normalized to a 10 g fish at 15 °C. The study-specific $k_{x,m,out}$ can be calculated from the normalized one:

$$k_{x,m,out} = k_{x,m,out,n} \cdot \left(\frac{w_i}{0.01 \cdot w^*}\right)^{-\kappa} \cdot \left(\frac{\exp(T)}{\exp(T_n)}\right)^{0.01} \qquad \text{(S9)}$$

For inhalation and exhalation via air, $k_{x,a,in}$ and $k_{x,a,out}$ the concentration in the organism was calculated depending on the blood-air partition coefficient (K_{ba}) and the tissue-air partition coefficient (K_{ta}) as described by Veltman et al (2009).

$$k_{x,a,in} = \frac{1}{\frac{1}{G_A} + \frac{\beta_A}{d_w \cdot A_A \cdot K_{ba}} + \frac{1}{G_B \cdot K_{ba}}} \tag{S10}$$

$$k_{x,a,out} = \frac{1}{K_{ta}} \cdot \frac{1}{\frac{1}{G_A} + \frac{\beta_A}{d_w \cdot A_A \cdot K_{ba}} + \frac{1}{G_B \cdot K_{ba}}} \tag{S11}$$

$$G_A = 0.67 \cdot 667.008 \cdot w_i^{0.745} \tag{S12}$$

$$\beta_A = 269.8 \cdot (1000 \cdot w_i)^{0.0618} \cdot 10^{-8} \tag{S13}$$

$$d_w = \frac{2.7 \cdot 10^{-8}}{molweight_x^{0.71} \cdot 100 \cdot 60 \cdot 60 \cdot 24} \tag{S14}$$

$$A_A = 293 \cdot w_i^{0.97} \tag{S15}$$

$$K_{ba} = \frac{K_{bw}}{K_{aw}} \tag{S16}$$

$$G_B = 321.408 \cdot w_i^{0.75} \tag{S17}$$

$$K_{ta} = \frac{K_{tw}}{K_{aw}} \tag{S18}$$

The bioaccumulation factor due to direct fresh water uptake and indirect (i.e. via food) was calculated for each trophic level separately in order to determine the higher predators' intake via food. For trophic level 1 the BF was calculated as:

$$BF_{x,1} = \frac{k_{x,w,in,1}}{\sum k_{x,out,1}} \qquad (S19)$$

Species from higher trophic levels also take up chemicals from the water via food. For trophic level 2 the BF was defined as:

$$BF_{x,2} = \frac{k_{x,w,in,2} + k_{x,f,in,2} \cdot BF_{x,1}}{\sum k_{x,out,2}} \qquad (S20)$$

and for trophic level 3 as:

$$BF_{x,3} = \frac{k_{x,w,in,3} + k_{x,f,in,3} \cdot BF_{x,2}}{\sum k_{x,out,3}} \qquad (S21)$$

In these equations BF_x is the bioaccumulation factor of substance x in organism for trophic levels 1, 2 and 3, due to uptake from the fresh water compartment, i.e. water or food ($m^3 \cdot kg_{wwt}^{-1}$), $k_{x,w,in}$ is the influx rate constant for water absorption of substance x per trophic level ($L \cdot kg_{wwt}^{-1} \cdot d^{-1}$), $k_{x,f,in}$ is the influx rate constant for assimilation of substance x from food per trophic level ($L \cdot kg_{wwt}^{-1} \cdot d^{-1}$), and $\sum k_{x,out}$ is the sum of the rate constants for the different elimination routes per trophic level (all in d^{-1}). These elimination routes are excretion and growth dilution for each trophic level, egestion for trophic levels 2 and 3, and biotransformation for trophic level 3.

Table S1. Factors used in equations with typical values for parameters

Symbol	Description	Unit	Typical value	Reference
A_A	Alveolar surface area per kg body weight	$dm^2 \cdot kg^{-1}$	equation S15	(Veltman et al. 2009)
β_A	Diffusion distance across alveolar cells	dm	equation S13	(Veltman et al. 2009)
BF_x	Bioaccumulation Factor of substance x in organisms per trophic level in the water compartment	$L \cdot kg_{wwt}^{-1}$	equations S19 – S21	
$C_{x,a}$	Concentration of substance x in the air	$kg \cdot L^{-1}$	USES-LCA 2.0	(van Zelm et al. 2009)
$C_{x,i}$	Concentration of substance x in organisms of trophic level i	$kg \cdot kg_{wwt}^{-1}$	equation S1	(Hendriks et al. 2001; Veltman et al. 2009)
$C_{x,w}$	Concentration of substance x in the water	$kg \cdot L^{-1}$	USES-LCA 2.0	(van Zelm et al. 2009)
d_w	Aqueous diffusion in cytosol	$dm^2 \cdot d^{-1}$	equation S14	(Veltman et al. 2009)
γ_w	Water absorption – excretion coefficient Tropic level 1-3 Trophic level 4	$kg^{\kappa} \cdot d^{-1}$	200 0.2	(Hendriks et al. 2001)
γ_p	Biomass (re)production coefficient	$kg^{\kappa} \cdot d^{-1}$	0.0006	(Hendriks et al. 2001)

γ_f	Food ingestion coefficient	$kg^{\kappa}\cdot d^{-1}$	0.005 ($i \geq 2$)	(Hendriks et al. 2001)
G_A	Alveolar ventilation rate	$dm^3\cdot kg^{-1}\cdot d^{-1}$	equation S12	(Veltman et al. 2009)
G_B	Cardiac output	$dm^3\cdot kg^{-1}\cdot d^{-1}$	equation S17	(Veltman et al. 2009)
$k_{x,a,in}$	Rate constant for inhalation	$L\cdot kg^{-1}\cdot d^{-1}$	equation S12	(Veltman et al. 2009)
$k_{x,a,out}$	Rate constant for exhalation	$L\cdot kg^{-1}\cdot d^{-1}$	equation S10	(Veltman et al. 2009)
$k_{x,f,in}$	Rate constant for assimilation	$L\cdot kg^{-1}\cdot d^{-1}$	equation S6	
$k_{x,f,out}$	Rate constant for egestion with faeces	$kg\cdot kg^{-1}\cdot d^{-1} = d^{-1}$	equation S7	
$k_{x,m,out}$	Rate constant for metabolic transformation	$kg\cdot kg^{-1}\cdot d^{-1} = d^{-1}$	equation S9	(Arnot et al. 2008)
$k_{x,m,out,n}$	Normalized rate constant for metabolic transformation	$kg\cdot kg^{-1}\cdot d^{-1} = d^{-1}$		(Arnot et al. 2008)
$k_{x,p,out}$	Rate constant for dilution of biomass by reproduction or growth	$kg\cdot kg^{-1}\cdot d^{-1} = d^{-1}$	equation S8	(Hendriks et al. 2001)
$k_{x,w,\ in}$	Rate constant for absorption	$L\cdot kg^{-1}\cdot d^{-1}$	equation S4	(Hendriks et al. 2001)
$k_{x,w,out}$	Rate constant for excretion	$kg\cdot kg^{-1}\cdot d^{-1} = d^{-1}$	equation S5	
K_{aw}	Air – water partition coefficient at 310 °K	[-]	USES-LCA 2.0	(van Zelm et al. 2009)
K_{ba}	Blood – air partition coefficient	[-]	Equation S16	(Veltman et al.

				2009)
K_{bw}	Blood – water partition coefficient			(Hendriks et al. 2005; Veltman et al. 2009)
	Nonpolar substances	[-]	Equation S2	
	Polar substances	[-]	Equation S3	
K_{ow}	Octanol – water partition coefficient at 298 °K	[-]	USES-LCA 2.0	(van Zelm et al. 2009)
K_{ta}	Tissue – air partition coefficient	[-]	Equation S18	(Veltman et al. 2009)
K_{tw}	Tissue – water partition coefficient			(Hendriks et al. 2005; Veltman et al. 2009)
	Nonpolar substances	[-]	Equation S2	
	Polar substances	[-]	Equation S3	
κ	Rate exponent	[-]	0.25	(Hendriks 1999; West et al. 1999)
p_f	Fraction of ingested food assimilated			(Hendriks 1999)
	Herbivores	$kg \cdot kg^{-1}$	0.4	
	Carnivores	$kg \cdot kg^{-1}$	0.8	
p_g	Fraction of lignin in organism	$kg \cdot kg^{-1}$	Table S2	
p_{H2O}	Fraction of water in tissues (t) or blood (b)	$kg \cdot kg^{-1}$	Table S2	
p_{nl}	Fraction of neutral lipids in tissues (t) or blood (b)	$kg \cdot kg^{-1}$	Table S2	
p_p	Fraction of proteins in tissues (t) or blood (b)	$kg \cdot kg^{-1}$	Table S2	

p_{pl}	Fraction of polar lipids in tissues (t) or blood (b)	$kg \cdot kg^{-1}$	Table S2	
q_T	Temperature correction factor	$kg \cdot kg^{-1}$		(Hendriks 1999)
	Cold-blooded species		1	
	Warm-blooded species		10	
$\rho_{CH2,i}$	Lipid layer permeation resistance	$d \cdot kg^{-\kappa}$		(Hendriks et al. 2001)
	plants		$4.6 \cdot 10^{3}$	
	animals		68	
$\rho_{H2O,w}$	Water layer resistance from / to water	$d \cdot kg^{-\kappa}$	$2.8 \cdot 10^{-3}$	(Hendriks et al. 2001)
$\rho_{H2O,f}$	Water layer resistance from / to food	$d \cdot kg^{-\kappa}$	$1.1 \cdot 10^{-5}$	(Hendriks et al. 2001)
T	System temperature in model	°K	285	(van Zelm et al. 2009)
T_n	Normalized water temperature	°K	288	(Arnot et al. 2008)
x	Substance	[-]		
w*	Reference mass	kg	1	
w_i	Species wet weight			
	Trophic level 1 – 3	kg	$1.0 \cdot 10^{-12}$ – $1.0 \cdot 10^{-2}$	
	Trophic level 4	kg	3.0	

Table S2. Fractions of different components in tissue and blood per trophic level

Trophic level	**Body part**	p_{H2O} (a)	p_{nl}	p_{pl}	p_p	p_g
1	Tissue	0.93	0	0.01 (1)	0.05 (1)	0.01 (1)
2	Tissue	0.88	0.01 (1,2)	0.01 (1)	0.10 (1)	0
3	Tissue	0.77	0.04 (1)	0.01 (1)	0.18 (1)	0
4	Tissue	0.70 (1)	0.09 (1)	0.01 (1)	0.20 (1)	0
4 (b)	Blood	0.80 (3)	0.002 (3)	0.002 (3)	0.20 (3)	0

(a) The water fractions are considered to be the remaining fractions after subtracting the other components.

(b) based on data for mammals

(1) (Hendriks et al. 2005)

(2) (Hendriks et al. 2001)

(3) (Veltman et al. 2009)

2. Chemical Properties

Table S3. Chemical properties of the example chemicals highlighted

Property name	Symbol	Unit	Acephate	Aldicarb	Lindane	DDT
Type of chemical	-	-	polar	polar	nonpolar	nonpolar
Octanol-water partition coefficient	K_{ow}	-	$1.00 \bullet 10^{-1}$	$1.26 \bullet 10^{1}$	$5.01 \bullet 10^{3}$	$1.55 \bullet 10^{6}$
Air-water partition coefficient	K_{aw}	-	$2.04 \bullet 10^{-11}$	$5.12 \bullet 10^{-8}$	$6.01 \bullet 10^{-5}$	$5.20 \bullet 10^{-4}$
Organic carbon-water partition coefficient	K_{oc}	$L \cdot kg^{-1}$	$2.00 \bullet 10^{0}$	$1.66 \bullet 10^{1}$	$9.55 \bullet 10^{2}$	$4.27 \bullet 10^{5}$
Rate constant for degradation in air	$k_{deg(air)}$	s^{-1}	$2.55 \bullet 10^{-5}$	$3.38 \bullet 10^{-5}$	$3.47 \bullet 10^{-6}$	$1.81 \bullet 10^{-6}$
Rate constant for degradation in water	$k_{deg(water)}$	s^{-1}	$1.53 \bullet 10^{-7}$	$1.07 \bullet 10^{-7}$	$1.10 \bullet 10^{-7}$	$1.66 \bullet 10^{-7}$
Rate constant for degradation in soil	$k_{deg(soil)}$	s^{-1}	$3.65 \bullet 10^{-6}$	$9.44 \bullet 10^{-8}$	$7.09 \bullet 10^{-8}$	$3.93 \bullet 10^{-9}$
Rate constant for degradation in sediment	$k_{deg(sed)}$	s^{-1}	$1.19 \bullet 10^{-6}$	$4.58 \bullet 10^{-8}$	$5.50 \bullet 10^{-7}$	$9.06 \bullet 10^{-8}$
Normalized rate constant for biotransformation in fish of the third trophic level	$k_{x,m,out}$	s^{-1}	$1.08 \bullet 10^{-3}$	$1.23 \bullet 10^{-4}$	$1.60 \bullet 10^{-7}$	$9.49 \bullet 10^{-8}$
Orally hazardous dose that is lethal to 50 percent of the individuals in 50 percent of the predator species	$HD50_{predator}$	$mg \cdot kg_{BW}^{-1}$	$2.34 \bullet 10^{2}$	$2.03 \bullet 10^{0}$	$9.81 \bullet 10^{1}$	$8.11 \bullet 10^{2}$

3. Additional Graphs

Figure S2: Correlation plots of K_{aw} and the contribution of uptake via air to CB

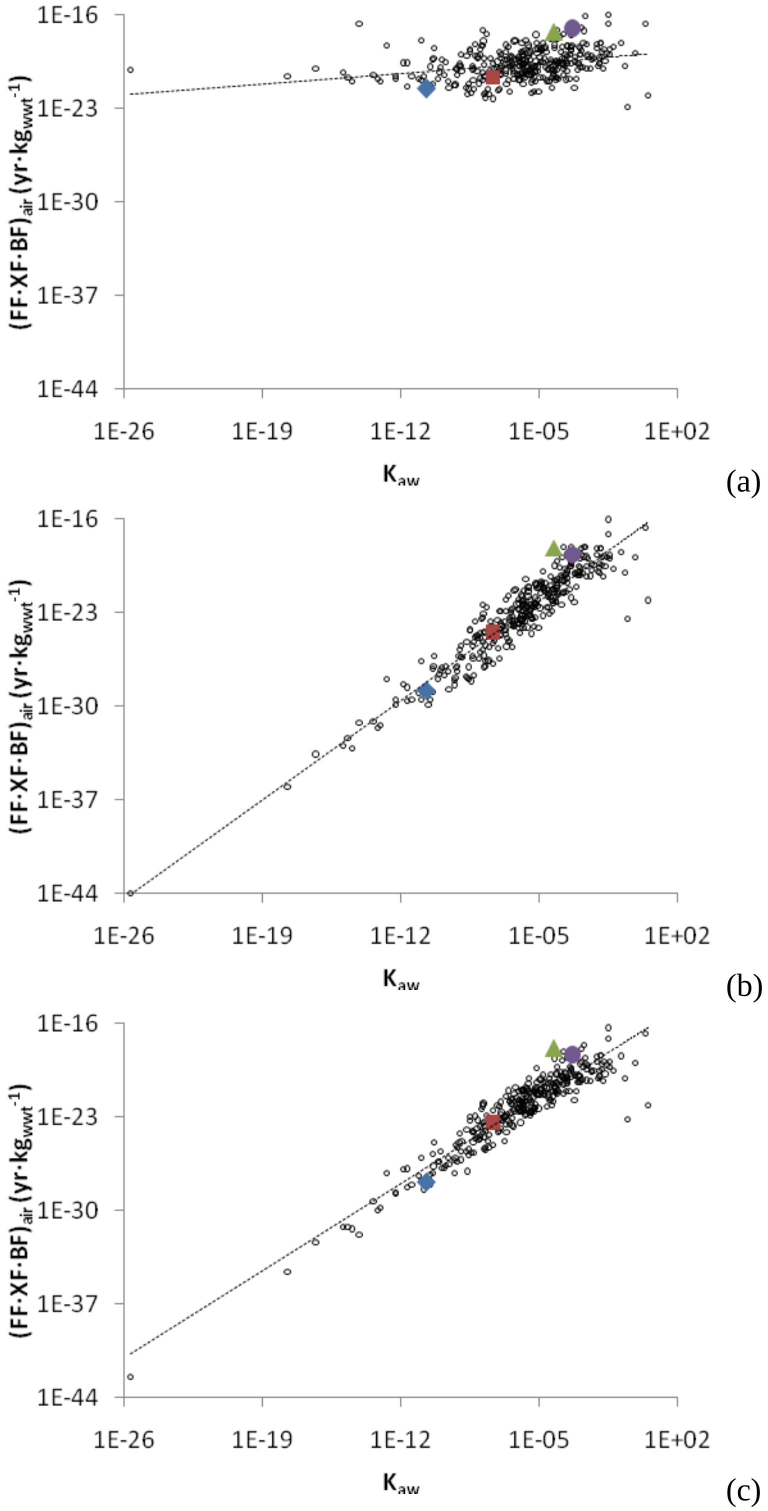
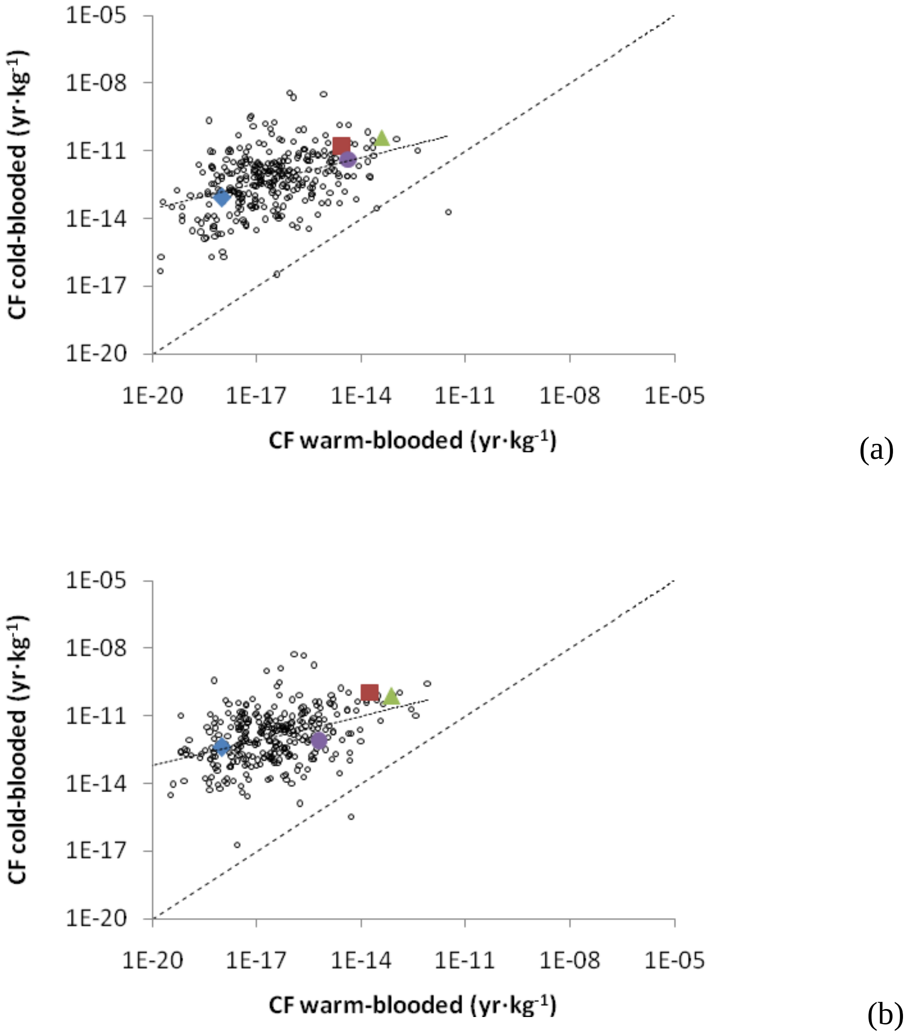


Figure S2: The contribution of uptake via air to the chemicals' CB was positively correlated with the chemicals' K_{aw} for an emission to air (a, R^2=0.09), fresh water (b, R^2=0.89), and agricultural soil (c, R^2=0.89), respectively. Acephate (◊), Aldicarb (□), Lindane (Δ), and DDT (O) are highlighted. The dotted line shows the linear fit for the data.

Figure S3: Correlation plots of CFs for warm-blooded predators and CFs for cold-blooded species



(a)

(b)

Figure S3: Correlation between our new CFs for warm-blooded predators and CFs for cold-blooded species calculated according to existing methodologies, for an emission to air (a, R^2=0.16), and agricultural soil (b, R^2=0.18), respectively. Acephate (◊), Aldicarb (□), Lindane (Δ), and DDT (O) are highlighted. The dashed line indicates the 1:1 relation, whereas the dotted line shows the linear fit for the data.

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