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

Running Title – Ecotoxic Impacts on Warm-blooded Predators

Laura Golsteijn *†, Rosalie van Zelm †, Karin Veltman ‡, Gijs Musters †, A Jan Hendriks †, and Mark AJ Huijbregts †

†   Radboud University Nijmegen, Institute for Water and Wetland Research, Department of Environmental Science, PO Box 9010, 6500 GL, Nijmegen, The Netherlands
‡  Norwegian University of Science and Technology, Industrial Ecology Programme, NO-7491 Trondheim, Norway
*   Email: L.Golsteijn@science.ru.nl

ABSTRACT

In current Life Cycle Impact Assessment (LCIA), the focus of ecotoxicity is on cold-blooded species. We developed a method to calculate Characterization Factors (CFs) for the impact assessment of chemical emissions on warm-blooded predators in freshwater food chains. The method was applied to 329 organic chemicals. The CF for these predators was defined as a multiplication of the Fate Factor (FF), Exposure Factor (XF), Bioaccumulation Factor (BF), and Effect Factor (EF). FFs and XFs were calculated with the model USES-LCA 2.0. BFs were calculated with the model OMEGA, for chemical uptake via fresh water, food and air. EFs were calculated based on experimental, median lethal doses (LD50). The chemicals’ Concentration Buildup (CB, i.e. FF, XF, and BF over the 3 routes of exposure) showed a range of 7 to 9 orders of magnitude, depending on the emission compartment. EFs displayed a range of 7 orders of magnitude. CFs ranged 9 orders of magnitude. After emissions to fresh water, the relative contribution of the uptake routes to CB were 1% (90%-CI: 0-2%) for uptake from air, 43% (11-50%) for uptake from water, and 56% (50-87%) for uptake from food. After an emission to agricultural soil, the contribution was 11% (0-80%) for uptake from air, 39% (5-50%) for uptake from water, and 50% (11-83%) for uptake from food. Uptake from air was mainly relevant for emissions to air (on average 42%, 90%-CI: 5-98%). CFs for cold-blooded species were typically 4 orders of magnitude higher than CFs for warm-blooded predators. The correlation between both types of CFs was low, which means that a high relative impact on cold-blooded species does not necessarily indicate a high relative impact on warm-blooded predators. Depending on the weighing method to be considered, the inclusion of impacts on warm-blooded predators can change the relative ranking of toxic chemicals in a life cycle assessment. Keywords – organic chemicals, pesticides, bioaccumulation, aquatic food chain, warm-blooded predators
INTRODUCTION

A Life Cycle Assessment (LCA) quantifies the resource use and emissions of a product or service for its complete life cycle. The impact categories of interest, e.g. fossil fuel use, global warming, and ecotoxicity, are determined in a Life Cycle Impact Assessment (LCIA) (Pennington et al. 2004b). In current LCIA of freshwater ecotoxicity, the focus is on cold-blooded species (e.g. algae, invertebrates, and fish), excluding the impact of chemicals on warm-blooded predators (e.g. mammals and birds). The impact of chemicals on cold-blooded species is estimated from direct exposure to concentrations in fresh water, whereas uptake of chemicals via food is not accounted for. Although both cold-blooded and warm-blooded predators in aquatic food chains can be exposed to chemical pollutants via water and food, the inclusion of uptake from food is of much greater importance for warm-blooded predators than for carnivorous fish (Hendriks 1995a; Kelly et al. 2007). Furthermore, the effects per unit of exposure may differ between cold-blooded and warm-blooded species. Therefore, we developed a method to assess impacts of chemicals on warm-blooded predators in freshwater ecosystems.

The impact of a product or service for the different impact categories is quantified with Characterization Factors (CFs). CFs for ecotoxicity depend on the fate, exposure and effects of each chemical emission in the environment (Pennington et al. 2004b). The fate and exposure factors of chemicals are generally modeled with multimedia fate and exposure models (McKone 1993; Pennington et al. 2005; Rosenbaum et al. 2008; van Zelm et al. 2009b). Effect factors are modeled from experimental toxicity data, applying species sensitivity distributions (Hauschild and Pennington 2002).

In order to develop characterization factors for the ecotoxicological impacts of organic chemicals on warm-blooded predators at the end of freshwater food chains, we calculated fate and exposure factors for water and air. Subsequently, we introduced bioaccumulation factors in the CF-calculations. This way, we accounted for bioaccumulation in three uptake routes of the warm-blooded predators, i.e. absorption from freshwater, assimilation from food, and inhalation of air. Internal effect factors were calculated based on LD50-values for mammals and birds. To conclude, we made a comparison between our new characterization factors for warm-blooded predators and characterization factors for cold-blooded species currently applied in LCIA.
METHODOLOGY

Ecotoxicity Characterization Factors

In this study, the CF for warm-blooded predators in freshwater food chains was defined as the change in ecotoxic effects of a chemical x on warm-blooded predators, resulting from a change in emission of chemical x. It consists of a multiplication of the Fate Factor (FF_{x,i,j}), Exposure Factor (XF_{x,j}), Bioaccumulation Factor (BF_{x,j}), and Effect Factor (EF_x) of a chemical:

\[ CF_{x,i} = \sum_{j} \left( FF_{x,i,j} \cdot XF_{x,j} \cdot BF_{x,j} \right) \cdot EF_x \]  \hspace{1cm} (1)

where CF_{x,i} is the ecotoxicological characterization factor of a chemical x emitted into an environmental compartment of emission (i) (yr·kg^{-1}). The fate factor describes the fraction of the chemical x transferred from the emission compartment i to a compartment of reception (j), and its subsequent residence time in compartment j (yr·m^{-3}). The dimensionless exposure factor is the fraction of the chemical x in the receiving compartment j that is bioavailable for uptake by organisms. The bioaccumulation factor for substance x represents the predators’ uptake potential of the bioavailable concentration in fresh water, food and air (quantified as unit of environmental volume per unit of wet weight, i.e. m^3·kg_{wwt}^{-1}). For the remainder of this paper, we will refer to the product of FF_{x,i,j}, XF_{x,j}, and BF_{x,j}, summed for uptake from fresh water, food, and air, as the chemical’s Concentration Buildup (CB_{x,i} in yr·kg_{wwt}^{-1}). CB_{x,i} is the change in the internal concentration of chemical x in warm-blooded predators, resulting from a change in emission of chemical x of 1 kilogram per year. EF_x is the effect factor of chemical x describing the effects of chemical x on warm-blooded predators per unit of internal concentration (kg_{wwt}·kg^{-1}). It is based on the assimilated dose that has lethal effects on 50 percent of the species (kg chemical per kg wet weight, i.e. kg·kg_{wwt}^{-1})

The freshwater food chain modeled in this study consists of four trophic levels, i.e. algae, invertebrates, fish, and warm-blooded predators such as mammals or birds (see Figure 1). In order to quantify the predators’ internal concentration for each chemical, the exposure and bioaccumulation in trophic level 1 up to and including trophic level 3 were taken into account.
Fate and Exposure

The fate factor is the change in total steady state concentration of substance x in receiving compartment j (dC_{j,x,total} in kg·m^{-3}) due to a change in emission of substance x to compartment i (dM_{i,x} in kg·yr^{-1}):

\[ FF_{x,i,j} = \frac{dC_{j,x,total}}{dM_{i,x}} \] (2)

The exposure factor for fresh water (XF_{x,w}) is the fraction of chemical x dissolved:

\[ XF_{x,w} = \frac{dC_{x,w,diss}}{dC_{x,w,total}} \] (3)

where dC_{x,w,diss} represents the change in concentration of chemical x dissolved in the freshwater compartment due to a change in the total concentration of chemical x in the freshwater compartment (dC_{x,w,total} both in kg·L^{-1}). For air, the exposure factor was set to 1, since both chemical attached to aerosols and chemical in the gaseous phase contribute to the exposure.

USES-LCA 2.0 was used to calculate fate and exposure factors (van Zelm et al. 2009b).

Bioaccumulation

Bioaccumulation is defined as the net process by which the chemical concentration in an organism achieves a level exceeding the concentration in air, water or organic solids. We distinguished three bioaccumulation factors (BFs) for warm-blooded predators, namely for uptake from water, uptake from food, and uptake from air.

The bioaccumulation factor for uptake from water was defined as:

\[ BF_{x,w} = \frac{dC_{x,predator}}{dC_{x,w,diss}} = \frac{k_{x,w,in}}{\sum k_{x,out}} \] (4)

where BF_{x,w} is the bioaccumulation factor of chemical x in warm-blooded predators due to uptake from fresh water (m^{3}·kg_{wwt}^{-1}), dC_{x,predator} is the change in predators' internal concentration of chemical x (kg·kg_{wwt}^{-1}), k_{x,w,in} is the influx rate constant for chemical x via water absorption for warm-blooded predators (L·kg_{wwt}^{-1}·yr^{-1}), and \( \sum k_{x,out} \) is the sum of the rate constants for the
different elimination routes in warm-blooded predators, i.e. excretion, egestion, biotransformation, growth dilution, and exhalation (yr⁻¹).

For the bioaccumulation factor of uptake from food (BFₓ,f), the concentration change in predators results from a change in the dissolved chemical concentration in water, via a concentration change in the predators’ food:

\[
BFₓ,f = \frac{dCₓ,predator}{dCₓ,w,diss} = \frac{kₓ,f,in \cdot BFₓ,3}{\sum kₓ, out}
\]  

(5)

where BFₓ,f is the bioaccumulation factor of chemical x in warm-blooded predators attributable to uptake from food (m³·kgwwt⁻¹), kₓ,f,in is the predators’ influx rate constant for assimilation of chemical x from food (L·kgwwt⁻¹·yr⁻¹), and BFₓ,3 (m³·kgwwt⁻¹) is the bioaccumulation factor of trophic level 3 attributable to freshwater uptake which is both direct and indirect (i.e. via food). The equation describing BFₓ,3 can be found in the SI.

The concentration of a chemical in predators of trophic level 4 can further increase due to uptake from the air via inhalation. The resulting bioaccumulation was described by BFₓ,a (m³·kgwwt⁻¹):

\[
BFₓ,a = \frac{dCₓ,predator}{dCₓ,a} = \frac{kₓ,a,in}{\sum kₓ,out}
\]  

(6)

where dCₓ,a is the change in concentration of chemical x in the air (kg·L⁻¹), and kₓ,a,in is the predators’ uptake rate constant for inhalation (L·kgwwt⁻¹·yr⁻¹).

The bioaccumulation factors were calculated with the bioaccumulation model OMEGA (Optimal Modeling for Ecotoxicological Applications) of Hendriks et al (2005; 2001) supplemented by the calculations of Veltman et al (2009) to predict accumulation of air pollutants in various mammals. OMEGA is based on classical fugacity theory for accumulation of organic substances and defines rate constants for uptake and elimination as a function of the partitioning and biotransformation of a chemical, the fractions of water, proteins, polar and neutral lipids in the tissue or blood of the species, and the trophic level of the species. The partitioning between the blood or tissue of organisms and the exchange compartments water or air was implemented separately in the model calculations for polar and nonpolar chemicals (Hendriks et al. 2005). More information about the calculations in OMEGA can be found in the Electronic Supporting Information. It also includes a description of how typical species characteristics were implemented per trophic level (Tables S1 and S2).
Effect

The effect factor expresses the effect of a chemical on warm-blooded predators in freshwater food chains per unit of internal exposure. We applied the linear approach of Pennington et al. (2004a) to calculate the effect factor of chemical x ($EF_x$ in kg$_{wwt} \cdot$ kg$^{-1}$):

$$EF_x = \frac{dPAF}{dC_{x,\text{predator}}} = \frac{0.5}{BB50_x}$$  \hspace{1cm} (7)

where $dPAF$ is the dimensionless change in the potentially affected fraction (PAF) of species, and $BB50_x$ is the median hazardous body burden of chemical x lethal to 50 percent of the individuals in 50 percent of the species (kg·kg$_{wwt}^{-1}$). We calculated the predators’ hazardous body burden for each chemical as the fraction of the orally hazardous dose that is assimilated:

$$BB50_x = p_{x,\text{ass}} \cdot HD50_x$$  \hspace{1cm} (8)

The fraction of the ingested dose that is assimilated by predators ($p_{x,\text{ass}}$) was obtained by dividing the rate constant for assimilation by the rate constant for food ingestion as calculated in OMEGA (Hendriks et al. 2001). The orally hazardous dose of chemical x (HD50$_x$ in kg·kg$_{wwt}^{-1}$) represents the oral dose that is lethal to 50 percent of the individuals in 50 percent of the species:

$$\log HD50_x = \frac{1}{n} \cdot \sum_n \log LD50_x$$  \hspace{1cm} (9)

where $n$ is the number of species tested and LD50$_x$ is the dose of chemical x lethal to 50 percent of the individuals of a certain species (kg·kg$_{wwt}^{-1}$).

Data Collection

The majority of the 329 organic chemicals modeled in this study were pesticides. The Electronic Supporting Information gives the complete list of chemicals, and details on how they were classified as nonpolar or polar. For fate and exposure modeling, the physicochemical properties of the chemicals were taken from USES-LCA 2.0 (van Zelm et al. 2009b).

For bioaccumulation modeling, the biotransformation rate constants ($k_{x,m,\text{out}}$) in fish of the third trophic level were taken from EPI Suite™ 4.0 (Arnot et al. 2008). Arnot and colleagues defined biotransformation as the change of a chemical to another molecule or a conjugated form.
of that chemical. Experimental biotransformation rates were available for 69 out of the 329 chemicals modeled in this study (Arnot et al. 2008). We used model estimates for the biotransformation rates of the remaining chemicals (see Electronic Supporting Information). Biotransformation rates in warm-blooded predators were assumed to be five times faster than biotransformation rates in fish of the third trophic level on a per body weight basis, based on the work of Arnot and others (2010). We did not take elimination via biotransformation in algae and invertebrates into account due to lack of data. For bioaccumulation modeling, the chemicals’ $K_{ow}$-values and $K_{aw}$-values were taken from USES-LCA 2.0 (van Zelm et al. 2009b).

For all 329 organic chemicals, experimental LD50-values for mammals and birds were obtained from literature (ATSDR 2006; Gaines 1960; 1969; Hudson et al. 1979; Luttik and Aldenberg 1997; Mineau et al. 2001; Schafer and Bowles 1985; Schafer et al. 1983; Vernot et al. 1977). We grouped the effect data available for mammals and birds in order to calculate effect factors for warm-blooded predators.

Model Comparison

We compared our characterization factors for warm-blooded predators with characterization factors for cold-blooded species calculated by USES-LCA 2.0 (van Zelm et al. 2009b).

RESULTS

Figure 2 shows that the chemicals’ concentration buildup (the product of $FF_{x,ij}$, $XF_{x,j}$, and $BF_{x,j}$, summed for uptake from fresh water, food, and air) ranged 7 orders of magnitude for an emission to air, and 9 orders of magnitude for an emission to fresh water or agricultural soil. For illustrative purposes, Acephate, Aldicarb, Lindane, and DDT are highlighted in our figures. More details on their physical and chemical properties can be found in Table S2 (Electronic Supporting Information).

Figure 2 also shows that chemicals’ CBs were positively correlated with the $K_{ow}$. Of the highlighted chemicals, Acephate had the lowest concentration buildup for all three emission
scenarios. This can be attributed to a combination of a low $K_{ow}$ and a high biotransformation rate. The difference in CB between Lindane and DDT was mainly determined by a difference in biotransformation rate of one order of magnitude. The contribution of uptake from air to a chemical’s CB was positively correlated with the chemical’s $K_{aw}$ for all emission scenarios. These results are shown in the Electronic Supporting Information (Figure S2).

Table 1 displays the relative contribution of the three uptake routes to chemicals’ CBs for the three emission scenarios. After an emission to fresh water, the relative contribution was 1% (90%-CI: 0-2%) for uptake from air, 43% (90%-CI: 11-50%) for uptake from water, and 56% (90%-CI: 50-87%) for uptake from food. After an emission to agricultural soil, the relative contribution was 11% (90%-CI: 0-80%) for uptake from air, 39% (90%-CI: 5-50%) for uptake from water, and 50% (90%-CI: 11-83%) for uptake from food. Uptake from air was mainly relevant for emissions to air (on average 42% with 90%-CI: 5-98%). Relative uptake from food increased with increasing $K_{ow}$, at the expense of uptake from water. For chemicals with a high $K_{ow}$, uptake from food was by far the most important uptake route. After an emission of DDT to fresh water for example, on average 98% of the DDT uptake by warm-blooded predators was from food.

Figure 3 shows that effect factors ranged 7 orders of magnitude, and characterization factors 9 orders of magnitude, irrespective of the emission compartment. It also shows that the correlation between EFs and CFs was low ($R^2$=0.13 for an emission fresh water). The correlation between EFs and CBs was also low ($R^2$=0.11 for an emission to fresh water, figure not shown). This low correlation was illustrated by, for example, Aldicarb and DDT: the EF of Aldicarb was more than two orders of magnitude higher than the EF of DDT, whereas the CF of Aldicarb was a little lower due to the fact was that its CB was three orders of magnitude lower. Hence, EFs and CBs are equally important to include in CF calculations.

To test the influence of biotransformation on CFs for warm-blooded predators, we performed a model scenario in which biotransformation rates in trophic level 4 were set to zero. We compared the CFs resulting from this scenario to the CFs from the default scenario, in which biotransformation rates in warm-blooded predators were assumed to be five times faster than biotransformation rates in fish of the third trophic level on a per body weight basis (Arnot et al. 2010) (see Methodology – Data Collection). Excluding biotransformation in warm-blooded
predators typically increased the CF with a factor of 140 (90%-CI: 2.2-8900). Figure 4 shows that this factor decreased with increasing CF.

Figure 5 shows the comparison of our characterization factors for warm-blooded predators and characterization factors for cold-blooded species currently applied for freshwater ecotoxicity in LCIA, for an emission to fresh water. Figure S3 (Electronic Supporting Information) shows this comparison for an emission to air and agricultural soil. CFs for cold-blooded species were median four orders of magnitude higher than the CFs for warm-blooded species (90%-CI: two to six orders of magnitude for emissions to fresh water or agricultural soil, and one to six orders of magnitude for emission to air). The chemicals approaching the 1:1 line in Figures 5 and S3 have a high $K_{ow}$ and a low biotransformation rate, e.g. Mirex, Pentac, and Brodifacoum. The correlation between the CFs of both methods was relatively low ($R^2=0.16$ for an emission to air, $R^2=0.18$ for an emission to agricultural soil, and $R^2=0.26$ for an emission to fresh water, respectively).

DISCUSSION

In this study, we calculated characterization factors for warm-blooded predators at the end of freshwater food chains. Here, we discuss the uncertainties associated with our methodology and the practical implications of our outcomes.

Uncertainty

To calculate CFs for warm-blooded predators, we estimated the fate of chemical emissions, and subsequent exposure, bioaccumulation and resulting potential effects. As the bioaccumulation factor was newly introduced in this type of modeling and the effect factor was adjusted, this section discusses uncertainties in the BF and EF quantification.

Uncertainty in the estimation of the bioaccumulation factor was mainly caused by the exposure routes included and the chemicals’ biotransformation rates. Chemical exposure via ingestion of sediment of sediment-dwelling organisms was not taken into account in the calculations of the bioaccumulation factor of higher predators, but may be relevant for persistent,
bioaccumulative, organic chemicals (ECHA 2008). Therefore, for this type of chemicals, excluding exposure via ingestion of sediment may have caused an underestimation of CFs.

The inclusion of biotransformation as an elimination route was highly relevant, but an important source of uncertainty at the same time. Biotransformation rates in warm-blooded predators were assumed to be five times faster than those in fish of the third trophic level on a per body weight basis (Arnot et al. 2010), an assumption that is very uncertain. The use of model estimates rather than experimental biotransformation rates for trophic level 3, for seventy-nine percent of the chemicals modeled in this study, adds uncertainty as well. Excluding biotransformation can cause excessive overestimation of critical body residues (Hendriks 2005).

The importance of biotransformation is further stressed by McLachlan et al (2010), who state that the role of biotransformation exceeds the role of partitioning properties in cases with multimedia perspective. In our study, the inclusion of biotransformation in trophic level 4 typically increased the CF with a factor of 140 (90%-CI: 2.2-8900). Due to lack of data, elimination via biotransformation was not included for algae and invertebrates. Mostly, biotransformation rates increase from low to high trophic levels, but there are some exceptions. For example, biotransformation of DDT appears to be faster in invertebrates in comparison to vertebrates, and small datasets suggest that nitrogen biocides are rapidly eliminated by plants and slowly by animals (Hendriks et al. 2001). To test the importance of possible biotransformation rates in trophic levels 1 and 2, we also performed a model scenario in which biotransformation rates in algae and invertebrates were assumed to be equal to the biotransformation rate in trophic level 3. The CBs were self-explanatory lower in the rerun scenario than in the original one, but the change in CBs was <1%. Hence, we can conclude that for our dataset the lack of biotransformation in trophic levels 1 and 2 had hardly any influence on the CBs for warm-blooded predators.

Uncertainty in EF calculations was mainly caused by the small size of our dataset, and the duration of the toxicity tests on which the EFs were based. The number of species tested per chemical varied between 1 and 37. Average uncertainty in EFs decreases with increasing number of species tested to around one order of magnitude for $n \geq 4$ (van Zelm et al. 2009a). In our study, $n \geq 4$ for 36% of the chemicals.

Although a few studies are available on chronic toxicity data for warm-blooded species (e.g. Haag et al (1950), Schafer et al. (1977) and Stomer (1970)), we used acute toxicity values
(LD50) to calculate effect factors, because the vast majority of the experimental data available is based on short-term tests. However, chronic toxicity values are probably closer to the wild life situation. Also, sub-lethal, chronic effects – such as inhibition of reproduction and migration – may give more insight in possible damage at population level than lethal doses. These effects occur mostly at doses that are a median factor of 2.5 lower than lethal doses (Hendriks 1995b).

Practical Implications

We found that CFs for cold-blooded species were typically four orders of magnitude higher than CFs for warm-blooded species. The correlation between characterization factors for warm-blooded predators and cold-blooded species was relatively low ($R^2<0.3$), which means that, in terms of ranking of chemicals, a high relative impact on cold-blooded species does not necessarily indicate a high relative impact on warm-blooded predators. In contrast with the conservative approach of environmental risk assessment, LCIA aims at a best estimate for fate, exposure and effect of chemicals (Hauschild 2005). Therefore, we recommend that the impact of chemicals on both cold-blooded and warm-blooded species is taken into account in an LCA. We suggest that CFs for cold-blooded and warm-blooded species are calculated separately. The (normalized) characterization scores of cold-blooded and warm-blooded species can be further weighed on the basis of e.g. the importance society attributes to the protection per trophic level. Depending on the weighing method to be considered, the inclusion of impacts on warm-blooded predators can change the relative ranking of toxic chemicals in a life cycle assessment.

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LIST OF TABLES AND FIGURES

Table 1: Relative contribution of different uptake routes to the CB. Numbers shown are average percentages. The values between brackets show the 90%-CI.

Figure 1: Scheme of the freshwater food chain applied in the bioaccumulation calculations of this study.

Figure 2: Correlation plot of the $K_{ow}$ of chemicals and their CB for an emission to air (a, $R^2=0.49$), fresh water (b, $R^2=0.68$), and agricultural soil (c, $R^2=0.28$), respectively. Acephate (◊), Aldicarb (□), Lindane (Δ), and DDT (O) are highlighted. The dotted line is the accompanying linear fit for the data.

Figure 3: Correlation plot of the EFs and CFs for warm-blooded predators for an emission to fresh water ($R^2=0.13$). Acephate (◊), Aldicarb (□), Lindane (Δ), and DDT (O) are highlighted. The dotted line is the accompanying linear fit for the data.

Figure 4: Correlation between CFs based on biotransformation in trophic level 3 and 4 (biotransformation in level 4 being five times higher than biotransformation in level 3) compared to CFs based on biotransformation in trophic level 3 only, for an emission fresh water. Acephate (◊), Aldicarb (□), Lindane (Δ), and DDT (O) are highlighted. The dashed line indicates the 1:1 relation.

Figure 5: Correlation between our new CFs for warm-blooded predators and CFs for cold-blooded species calculated according to existing methodologies, for an emission to fresh water ($R^2=0.26$). 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.
Table 1

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Figure 1

Air

Food
Algae

Cold-blooded herbivores
Cold-blooded carnivores

BF_{x,a}

BF_{x,f}

BF_{x,w}

Warm-blooded carnivores

Dissolved in water
Figure 2

(a) 

(b) 

(c)
Figure 3
Figure 4

Figure 5