

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

3

4 **Running Title** – Ecotoxic Impacts on Warm-blooded Predators

5

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

8

9 † Radboud University Nijmegen, Institute for Water and Wetland Research, Department of
10 Environmental Science, PO Box 9010, 6500 GL, Nijmegen, The Netherlands

11 ‡ Norwegian University of Science and Technology, Industrial Ecology Programme, NO-
12 7491 Trondheim, Norway

13 * Email: L.Golsteijn@science.ru.nl

14

15 **This is an earlier version of the following article: Golsteijn L., Van Zelm R., Veltman K.,**
16 **Musters G., Hendriks A.J., Huijbregts M.A.J. 2011. Including ecotoxic effects on warm-**
17 **blooded predators in life cycle impact assessment, which has been published in final form**
18 **in Integr. Environ. Assess. Manag. 8 (2): 372–378.**

19

1 **ABSTRACT**

2 In current Life Cycle Impact Assessment (LCIA), the focus of ecotoxicity is on cold-blooded
3 species. We developed a method to calculate Characterization Factors (CFs) for the impact
4 assessment of chemical emissions on warm-blooded predators in freshwater food chains. The
5 method was applied to 329 organic chemicals. The CF for these predators was defined as a
6 multiplication of the Fate Factor (FF), Exposure Factor (XF), Bioaccumulation Factor (BF), and
7 Effect Factor (EF). FFs and XFs were calculated with the model USES-LCA 2.0. BFs were
8 calculated with the model OMEGA, for chemical uptake via fresh water, food and air. EFs were
9 calculated based on experimental, median lethal doses (LD50). The chemicals' Concentration
10 Buildup (CB, i.e. FF, XF, and BF over the 3 routes of exposure) showed a range of 7 to 9 orders
11 of magnitude, depending on the emission compartment. EFs displayed a range of 7 orders of
12 magnitude. CFs ranged 9 orders of magnitude. After emissions to fresh water, the relative
13 contribution of the uptake routes to CB were 1% (90%-CI: 0-2%) for uptake from air, 43% (11-
14 50%) for uptake from water, and 56% (50-87%) for uptake from food. After an emission to
15 agricultural soil, the contribution was 11% (0-80%) for uptake from air, 39% (5-50%) for uptake
16 from water, and 50% (11-83%) for uptake from food. Uptake from air was mainly relevant for
17 emissions to air (on average 42%, 90%-CI: 5-98%). CFs for cold-blooded species were typically
18 4 orders of magnitude higher than CFs for warm-blooded predators. The correlation between
19 both types of CFs was low, which means that a high relative impact on cold-blooded species
20 does not necessarily indicate a high relative impact on warm-blooded predators. Depending on
21 the weighing method to be considered, the inclusion of impacts on warm-blooded predators can
22 change the relative ranking of toxic chemicals in a life cycle assessment. **Keywords** – organic
23 chemicals, pesticides, bioaccumulation, aquatic food chain, warm-blooded predators

24

1 **INTRODUCTION**

2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31

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.

1

2 **METHODOLOGY**

3

4 Ecotoxicity Characterization Factors

5

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

10
$$CF_{x,i} = \underbrace{\sum_j (FF_{x,i,j} \cdot XF_{x,j} \cdot BF_{x,j})}_{CB_{x,i}} \cdot EF_x \quad (1)$$

11

12 where $CF_{x,i}$ is the ecotoxicological characterization factor of a chemical x emitted into an
13 environmental compartment of emission (i) ($yr \cdot kg^{-1}$). The fate factor describes the fraction of the
14 chemical x transferred from the emission compartment i to a compartment of reception (j), and
15 its subsequent residence time in compartment j ($yr \cdot m^{-3}$). The dimensionless exposure factor is the
16 fraction of the chemical x in the receiving compartment j that is bioavailable for uptake by
17 organisms. The bioaccumulation factor for substance x represents the predators' uptake potential
18 of the bioavailable concentration in fresh water, food and air (quantified as unit of environmental
19 volume per unit of wet weight, i.e. $m^3 \cdot kg_{wwt}^{-1}$). For the remainder of this paper, we will refer to
20 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
21 chemical's Concentration Buildup ($CB_{x,i}$ in $yr \cdot kg_{wwt}^{-1}$). $CB_{x,i}$ is the change in the internal
22 concentration of chemical x in warm-blooded predators, resulting from a change in emission of
23 chemical x of 1 kilogram per year. EF_x is the effect factor of chemical x describing the effects of
24 chemical x on warm-blooded predators per unit of internal concentration ($kg_{wwt} \cdot kg^{-1}$). It is based
25 on the assimilated dose that has lethal effects on 50 percent of the species (kg chemical per kg
26 wet weight, i.e. $kg \cdot kg_{wwt}^{-1}$)

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

1

2 Fate and Exposure

3

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

$$7 \quad FF_{x,i,j} = \frac{dC_{x,j,total}}{dM_{x,i}} \quad (2)$$

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

$$9 \quad XF_{x,w} = \frac{dC_{x,w,diss}}{dC_{x,w,total}} \quad (3)$$

10 where $dC_{x,w,diss}$ represents the change in concentration of chemical x dissolved in the freshwater
11 compartment due to a change in the total concentration of chemical x in the freshwater
12 compartment ($dC_{x,w,total}$ both in $kg \cdot L^{-1}$). For air, the exposure factor was set to 1, since both
13 chemical attached to aerosols and chemical in the gaseous phase contribute to the exposure.
14 USES-LCA 2.0 was used to calculate fate and exposure factors (van Zelm et al. 2009b).

15

16 Bioaccumulation

17

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

22 The bioaccumulation factor for uptake from water was defined as:

$$23 \quad BF_{x,w} = \frac{dC_{x,predator}}{dC_{x,w,diss}} = \frac{k_{x,w,in}}{\sum k_{x,out}} \quad (4)$$

24 where $BF_{x,w}$ is the bioaccumulation factor of chemical x in warm-blooded predators due to
25 uptake from fresh water ($m^3 \cdot kg_{wwt}^{-1}$), $dC_{x,predator}$ is the change in predators' internal concentration
26 of chemical x ($kg \cdot kg_{wwt}^{-1}$), $k_{x,w,in}$ is the influx rate constant for chemical x via water absorption
27 for warm-blooded predators ($L \cdot kg_{wwt}^{-1} \cdot yr^{-1}$), and $\sum k_{x,out}$ is the sum of the rate constants for the

1 different elimination routes in warm-blooded predators, i.e. excretion, egestion,
2 biotransformation, growth dilution, and exhalation (yr^{-1}).

3 For the bioaccumulation factor of uptake from food ($\text{BF}_{x,f}$), the concentration change in
4 predators results from a change in the dissolved chemical concentration in water, via a
5 concentration change in the predators' food:

$$6 \quad \text{BF}_{x,f} = \frac{dC_{x,\text{predator}}}{dC_{x,w,\text{diss}}} = \frac{k_{x,f,\text{in}} \cdot \text{BF}_{x,3}}{\sum k_{x,\text{out}}} \quad (5)$$

7 where $\text{BF}_{x,f}$ is the bioaccumulation factor of chemical x in warm-blooded predators attributable
8 to uptake from food ($\text{m}^3 \cdot \text{kg}_{\text{wwt}}^{-1}$), $k_{x,f,\text{in}}$ is the predators' influx rate constant for assimilation of
9 chemical x from food ($\text{L} \cdot \text{kg}_{\text{wwt}}^{-1} \cdot \text{yr}^{-1}$), and $\text{BF}_{x,3}$ ($\text{m}^3 \cdot \text{kg}_{\text{wwt}}^{-1}$) is the bioaccumulation factor of
10 trophic level 3 attributable to freshwater uptake which is both direct and indirect (i.e. via food).

11 The equation describing $\text{BF}_{x,3}$ can be found in the SI.

12 The concentration of a chemical in predators of trophic level 4 can further increase due to
13 uptake from the air via inhalation. The resulting bioaccumulation was described by $\text{BF}_{x,a}$
14 ($\text{m}^3 \cdot \text{kg}_{\text{wwt}}^{-1}$):

$$15 \quad \text{BF}_{x,a} = \frac{dC_{x,\text{predator}}}{dC_{x,a}} = \frac{k_{x,a,\text{in}}}{\sum k_{x,\text{out}}} \quad (6)$$

16 where $dC_{x,a}$ is the change in concentration of chemical x in the air ($\text{kg} \cdot \text{L}^{-1}$), and $k_{x,a,\text{in}}$ is the
17 predators' uptake rate constant for inhalation ($\text{L} \cdot \text{kg}_{\text{wwt}}^{-1} \cdot \text{yr}^{-1}$).

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

1

2 Effect

3

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

7
$$EF_x = \frac{dPAF}{dC_{x,predator}} = \frac{0.5}{BB50_x} \quad (7)$$

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

12
$$BB50_x = p_{x,ass} \cdot HD50_x \quad (8)$$

13 The fraction of the ingested dose that is assimilated by predators ($p_{x,ass}$) was obtained by dividing
14 the rate constant for assimilation by the rate constant for food ingestion as calculated in OMEGA
15 (Hendriks et al. 2001). The orally hazardous dose of chemical x ($HD50_x$ in $kg \cdot kg_{wwt}^{-1}$) represents
16 the oral dose that is lethal to 50 percent of the individuals in 50 percent of the species:

17
$$\log HD50_x = \frac{1}{n} \cdot \sum_n \log LD50_x \quad (9)$$

18 where n is the number of species tested and $LD50_x$ is the dose of chemical x lethal to 50 percent
19 of the individuals of a certain species ($kg \cdot kg_{wwt}^{-1}$).

20

21 Data Collection

22

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

27 For bioaccumulation modeling, the biotransformation rate constants ($k_{x,m,out}$) in fish of the
28 third trophic level were taken from EPI Suite™ 4.0 (Arnot et al. 2008). Arnot and colleagues
29 defined biotransformation as the change of a chemical to another molecule or a conjugated form

1 of that chemical. Experimental biotransformation rates were available for 69 out of the 329
2 chemicals modeled in this study (Arnot et al. 2008). We used model estimates for the
3 biotransformation rates of the remaining chemicals (see Electronic Supporting Information).
4 Biotransformation rates in warm-blooded predators were assumed to be five times faster than
5 biotransformation rates in fish of the third trophic level on a per body weight basis, based on the
6 work of Arnot and others (2010). We did not take elimination via biotransformation in algae and
7 invertebrates into account due to lack of data. For bioaccumulation modeling, the chemicals'
8 K_{ow} -values and K_{aw} -values were taken from USES-LCA 2.0 (van Zelm et al. 2009b).

9 For all 329 organic chemicals, experimental LD50-values for mammals and birds were
10 obtained from literature (ATSDR 2006; Gaines 1960; 1969; Hudson et al. 1979; Luttik and
11 Aldenberg 1997; Mineau et al. 2001; Schafer and Bowles 1985; Schafer et al. 1983; Vernot et al.
12 1977). We grouped the effect data available for mammals and birds in order to calculate effect
13 factors for warm-blooded predators.

14 15 Model Comparison

16
17 We compared our characterization factors for warm-blooded predators with
18 characterization factors for cold-blooded species calculated by USES-LCA 2.0 (van Zelm et al.
19 2009b).

20 21 22 RESULTS

23
24 Figure 2 shows that the chemicals' concentration buildup (the product of $FF_{x,i,j}$, $XF_{x,j}$, and
25 $BF_{x,j}$, summed for uptake from fresh water, food, and air) ranged 7 orders of magnitude for an
26 emission to air, and 9 orders of magnitude for an emission to fresh water or agricultural soil. For
27 illustrative purposes, Acephate, Aldicarb, Lindane, and DDT are highlighted in our figures. More
28 details on their physical and chemical properties can be found in Table S2 (Electronic Supporting
29 Information).

30 Figure 2 also shows that chemicals' CBs were positively correlated with the K_{ow} . Of the
31 highlighted chemicals, Acephate had the lowest concentration buildup for all three emission

1 scenarios. This can be attributed to a combination of a low K_{ow} and a high biotransformation
2 rate. The difference in CB between Lindane and DDT was mainly determined by a difference in
3 biotransformation rate of one order of magnitude. The contribution of uptake from air to a
4 chemical's CB was positively correlated with the chemical's K_{aw} for all emission scenarios.
5 These results are shown in the Electronic Supporting Information (Figure S2).

6 Table 1 displays the relative contribution of the three uptake routes to chemicals' CBs for
7 the three emission scenarios. After an emission to fresh water, the relative contribution was 1%
8 (90%-CI: 0-2%) for uptake from air, 43% (90%-CI: 11-50%) for uptake from water, and 56%
9 (90%-CI: 50-87%) for uptake from food. After an emission to agricultural soil, the relative
10 contribution was 11% (90%-CI: 0-80%) for uptake from air, 39% (90%-CI: 5-50%) for uptake
11 from water, and 50% (90%-CI: 11-83%) for uptake from food. Uptake from air was mainly
12 relevant for emissions to air (on average 42% with 90%-CI: 5-98%). Relative uptake from food
13 increased with increasing K_{ow} , at the expense of uptake from water. For chemicals with a high
14 K_{ow} , uptake from food was by far the most important uptake route. After an emission of DDT to
15 fresh water for example, on average 98% of the DDT uptake by warm-blooded predators was
16 from food.

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

25 To test the influence of biotransformation on CFs for warm-blooded predators, we
26 performed a model scenario in which biotransformation rates in trophic level 4 were set to zero.
27 We compared the CFs resulting from this scenario to the CFs from the default scenario, in which
28 biotransformation rates in warm-blooded predators were assumed to be five times faster than
29 biotransformation rates in fish of the third trophic level on a per body weight basis (Arnot et al.
30 2010) (see Methodology – Data Collection). Excluding biotransformation in warm-blooded

1 predators typically increased the CF with a factor of 140 (90%-CI: 2.2-8900). Figure 4 shows
2 that this factor decreased with increasing CF.

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

16 **DISCUSSION**

17
18 In this study, we calculated characterization factors for warm-blooded predators at the
19 end of freshwater food chains. Here, we discuss the uncertainties associated with our
20 methodology and the practical implications of our outcomes.

22 **Uncertainty**

23
24 To calculate CFs for warm-blooded predators, we estimated the fate of chemical
25 emissions, and subsequent exposure, bioaccumulation and resulting potential effects. As the
26 bioaccumulation factor was newly introduced in this type of modeling and the effect factor was
27 adjusted, this section discusses uncertainties in the BF and EF quantification.

28 Uncertainty in the estimation of the bioaccumulation factor was mainly caused by the
29 exposure routes included and the chemicals' biotransformation rates. Chemical exposure via
30 ingestion of sediment of sediment-dwelling organisms was not taken into account in the
31 calculations of the bioaccumulation factor of higher predators, but may be relevant for persistent,

1 bioaccumulative, organic chemicals (ECHA 2008). Therefore, for this type of chemicals,
2 excluding exposure via ingestion of sediment may have caused an underestimation of CFs.

3 The inclusion of biotransformation as an elimination route was highly relevant, but an
4 important source of uncertainty at the same time. Biotransformation rates in warm-blooded
5 predators were assumed to be five times faster than those in fish of the third trophic level on a
6 per body weight basis (Arnot et al. 2010), an assumption that is very uncertain. The use of model
7 estimates rather than experimental biotransformation rates for trophic level 3, for seventy-nine
8 percent of the chemicals modeled in this study, adds uncertainty as well. Excluding
9 biotransformation can cause excessive overestimation of critical body residues (Hendriks 2005).
10 The importance of biotransformation is further stressed by McLachlan et al (2010), who state
11 that the role of biotransformation exceeds the role of partitioning properties in cases with
12 multimedia perspective. In our study, the inclusion of biotransformation in trophic level 4
13 typically increased the CF with a factor of 140 (90%-CI: 2.2-8900). Due to lack of data,
14 elimination via biotransformation was not included for algae and invertebrates. Mostly,
15 biotransformation rates increase from low to high trophic levels, but there are some exceptions.
16 For example, biotransformation of DDT appears to be faster in invertebrates in comparison to
17 vertebrates, and small datasets suggest that nitrogen biocides are rapidly eliminated by plants and
18 slowly by animals (Hendriks et al. 2001). To test the importance of possible biotransformation
19 rates in trophic levels 1 and 2, we also performed a model scenario in which biotransformation
20 rates in algae and invertebrates were assumed to be equal to the biotransformation rate in trophic
21 level 3. The CBs were self-explanatory lower in the rerun scenario than in the original one, but
22 the change in CBs was <1%. Hence, we can conclude that for our dataset the lack of
23 biotransformation in trophic levels 1 and 2 had hardly any influence on the CBs for warm-
24 blooded predators.

25 Uncertainty in EF calculations was mainly caused by the small size of our dataset, and
26 the duration of the toxicity tests on which the EFs were based. The number of species tested per
27 chemical varied between 1 and 37. Average uncertainty in EFs decreases with increasing number
28 of species tested to around one order of magnitude for $n \geq 4$ (van Zelm et al. 2009a). In our study,
29 $n \geq 4$ for 36% of the chemicals.

30 Although a few studies are available on chronic toxicity data for warm-blooded species
31 (e.g. Haag et al (1950), Schafer et al. (1977) and Stomer (1970)), we used acute toxicity values

1 (LD50) to calculate effect factors, because the vast majority of the experimental data available is
2 based on short-term tests. However, chronic toxicity values are probably closer to the wild life
3 situation. Also, sub-lethal, chronic effects – such as inhibition of reproduction and migration –
4 may give more insight in possible damage at population level than lethal doses. These effects
5 occur mostly at doses that are a median factor of 2.5 lower than lethal doses (Hendriks 1995b).

6 7 Practical Implications

8
9 We found that CFs for cold-blooded species were typically four orders of magnitude higher than
10 CFs for warm-blooded species. The correlation between characterization factors for warm-
11 blooded predators and cold-blooded species was relatively low ($R^2 < 0.3$), which means that, in
12 terms of ranking of chemicals, a high relative impact on cold-blooded species does not
13 necessarily indicate a high relative impact on warm-blooded predators. In contrast with the
14 conservative approach of environmental risk assessment, LCIA aims at a best estimate for fate,
15 exposure and effect of chemicals (Hauschild 2005). Therefore, we recommend that the impact of
16 chemicals on both cold-blooded and warm-blooded species is taken into account in an LCA. We
17 suggest that CFs for cold-blooded and warm-blooded species are calculated separately. The
18 (normalized) characterization scores of cold-blooded and warm-blooded species can be further
19 weighed on the basis of e.g. the importance society attributes to the protection per trophic level.
20 Depending on the weighing method to be considered, the inclusion of impacts on warm-blooded
21 predators can change the relative ranking of toxic chemicals in a life cycle assessment.

22 23 24 **ACKNOWLEDGEMENTS**

25 The authors would like to thank Mara Hauck for assistance with bioaccumulation
26 modeling, and Edgar Hertwich and Stuart Marshall for their valuable comments on an earlier
27 version of the manuscript. This research was partly funded by the European Commission under
28 the 7th framework program on environment; ENV.2009.3.3.2.1: LC-IMPACT - Improved Life
29 Cycle Impact Assessment methods (LCIA) for better sustainability assessment of technologies,
30 grant agreement number 243827.

31

1 REFERENCES

- 2
- 3 Agency for Toxic Substances & Disease Registry (ATSDR). 2006. Toxicological Profile
4 Information Sheet [Internet]. [cited 2006, December 13]. Available from:
5 <http://www.atsdr.cdc.gov/toxpro2.html>.
- 6 Arnot JA, Mackay D, Bonnell M. 2008. Estimating metabolic biotransformation rates in fish
7 from laboratory data. *Environmental Toxicology and Chemistry* 27(2):341-351.
- 8 Arnot JA, Mackay D, Parkerton TF, Zaleski RT, Warren CS. 2010. Multimedia modeling of
9 human exposure to chemical substances: The roles of food web biomagnification and
10 biotransformation. *Environmental Toxicology and Chemistry* 29(1):45-55.
- 11 European Chemicals Agency (ECHA). 2008. Guidance on information requirements and
12 chemical safety assessment. Chapter R10.5 Sediments. p. 32-37.
- 13 Gaines TB. 1960. The Acute Toxicity of Pesticides to Rats. *Toxicology and Applied*
14 *Pharmacology* 2(1):88-99.
- 15 Gaines TB. 1969. Acute Toxicity of Pesticides. *Toxicology and Applied Pharmacology*
16 14(3):515-534.
- 17 Haag HB, Finnegan JK, Larson PS, Riese W, Dreyfuss ML. 1950. Comparative chronic toxicity
18 for warm-blooded animals of 2,2-bis-(para-chlorophenyl)-1,1,1-trichloroethane (DDT)
19 and 2,2-bis-(para-methoxyphenyl)-1,1,1-trichloroethane (DMDT, methoxychlor).
20 *Archives Internationales De Pharmacodynamie Et De Therapie* 83(4):491-504.
- 21 Hauschild M, Pennington DW. 2002. Indicators for ecotoxicity in life-cycle impact assessment.
22 In Udo de Haes, H. A., Finnveden, G., Goedkoop, M., Hauschild, M., Hertwich, E.,
23 Hofstetter, P., Jolliet, O., Klöpffer, W., Krewitt, W., Lindeijer, E., Müller-Wenk, R.,
24 Olsen, S., Pennington, D.W., Potting, J., Steen, B. eds. *Life-Cycle Impact Assessment:*
25 *Striving towards Best Practice*, © 2002 Society of Environmental Toxicology and
26 Chemistry (SETAC). Pensacola, FL, USA: ISBN 1-880611-54-6. p. 149-174.
- 27 Hauschild MZ. 2005. Assessing environmental impacts in a life-cycle perspective.
28 *Environmental Science & Technology* 39(4):81A-88A.
- 29 Hendriks AJ. 1995a. Modeling equilibrium concentrations of microcontaminants in organisms of
30 the rhine delta - can average field residues in the aquatic foodchain be predicted from
31 laboratory accumulation. *Aquatic Toxicology* 31(1):1-25.
- 32 Hendriks AJ. 1995b. Modeling response of species to microcontaminants - comparative
33 ecotoxicology by (sub)lethal body burdens as a function of species size and partition ratio
34 of chemicals. *Ecotoxicology and Environmental Safety* 32(2):103-130.
- 35 Hendriks AJ, Traas TP, Huijbregts MAJ. 2005. Critical body residues linked to octanol-water
36 partitioning, organism composition, and LC50 QSARs: Meta-analysis and model.
37 *Environmental Science & Technology* 39(9):3226-3236.
- 38 Hendriks AJ, van der Linde A, Cornelissen G, Sijm D. 2001. The power of size. 1. Rate
39 constants and equilibrium ratios for accumulation of organic substances related to
40 octanol-water partition ratio and species weight. *Environmental Toxicology and*
41 *Chemistry* 20(7):1399-1420.
- 42 Hudson RH, Haegele MA, Tucker RK. 1979. Acute oral and percutaneous toxicity of pesticides
43 to mallards: Correlations with mammalian toxicity data. *Toxicology and Applied*
44 *Pharmacology* 47(3):451-460.

- 1 Kelly BC, Ikonou MG, Blair JD, Morin AE, Gobas F. 2007. Food web-specific
2 biomagnification of persistent organic pollutants. *Science* 317(5835):236-239.
- 3 Luttik R, Aldenberg T. 1997. Extrapolation factors for small samples of pesticide toxicity data:
4 Special focus on LD50 values for birds and mammals. *Environmental Toxicology and*
5 *Chemistry* 16(9):1785-1788.
- 6 McKone TE. 1993. CalTOX, a multimedia total exposure model for hazardous-waste sites; Part
7 2, Multimedia Transport and Transformation Model. Livermore, CA: Lawrence
8 Livermore National Laboratory. No. UCRL-CR--111456-Pt.1.
- 9 McLachlan MS, Czub G, MacLeod M, Arnot JA. 2010. Bioaccumulation of Organic
10 Contaminants in Humans: A Multimedia Perspective and the Importance of
11 Biotransformation. *Environmental Science & Technology* 45(1):197-202.
- 12 Mineau P, Baril A, Collins BT, Duffe J, Joerman G, Luttik R. 2001. Pesticide acute toxicity
13 reference values for birds. *Reviews of Environmental Contamination and Toxicology*
14 170:13-74.
- 15 Pennington DW, Margni M, Ammann C, Jolliet O. 2005. Multimedia fate and human intake
16 modeling: Spatial versus nonspatial insights for chemical emissions in Western Europe.
17 *Environmental Science & Technology* 39(4):1119-1128.
- 18 Pennington DW, Payet J, Hauschild M. 2004a. Aquatic ecotoxicological indicators in life-cycle
19 assessment. *Environmental Toxicology and Chemistry* 23(7):1796-1807.
- 20 Pennington DW, Potting J, Finnveden G, Lindeijer E, Jolliet O, Rydberg T, Rebitzer G. 2004b.
21 Life cycle assessment Part 2: Current impact assessment practice. *Environment*
22 *International* 30(5):721-739.
- 23 Rosenbaum RK, Bachmann TM, Gold LS, Huijbregts MAJ, Jolliet O, Juraske R, Koehler A,
24 Larsen HF, MacLeod M, Margni M et al. . 2008. USEtox-the UNEP-SETAC toxicity
25 model: recommended characterisation factors for human toxicity and freshwater
26 ecotoxicity in life cycle impact assessment. *International Journal of Life Cycle*
27 *Assessment* 13(7):532-546.
- 28 Schafer EW, Bowles WA. 1985. Acute oral toxicity and repellency of 933 chemicals to house
29 and deer mice. *Archives of Environmental Contamination and Toxicology* 14(1):111-
30 129.
- 31 Schafer EW, Bowles WA, Hurlbut J. 1983. The acute oral toxicity, repellency, and hazard
32 potential of 998 chemicals to one or more species of wild and domestic birds. *Archives of*
33 *Environmental Contamination and Toxicology* 12(3):355-382.
- 34 Schafer EW, Brunton RB, Cunningham DJ, Lockyer NF. 1977. The chronic toxicity of 3-chloro-
35 4-methyl benzamine HCl to birds. *Archives of Environmental Contamination and*
36 *Toxicology* 6(1):241-248.
- 37 Stomer VV. 1970. Chronic and acute toxicity of chlorinated hydrocarbon insecticides in
38 mammals and birds. *Canadian Veterinary Journal* 11(11):215-226.
- 39 van Zelm R, Huijbregts M, Posthuma L, Wintersen A, van de Meent D. 2009a. Pesticide
40 ecotoxicological effect factors and their uncertainties for freshwater ecosystems. *The*
41 *International Journal of Life Cycle Assessment* 14(1):43-51.
- 42 van Zelm R, Huijbregts MAJ, van de Meent D. 2009b. USES-LCA 2.0-a global nested multi-
43 media fate, exposure, and effects model. *International Journal of Life Cycle Assessment*
44 14(3):282-284.
- 45 Veltman K, McKone TE, Huijbregts MAJ, Hendriks AJ. 2009. Bioaccumulation potential of air
46 contaminants: Combining biological allometry, chemical equilibrium and mass-balances

1 to predict accumulation of air pollutants in various mammals. *Toxicology and Applied*
2 *Pharmacology* 238(1):47-55.

3 Vernot EH, MacEwen JD, Haun CC, Kinkead ER. 1977. Acute toxicity and skin corrosion data
4 for some organic and inorganic compounds and aqueous solutions. *Toxicology and*
5 *Applied Pharmacology* 42(2):417-423.

6
7

1 LIST OF TABLES AND FIGURES

2

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

5

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

8

9 Figure 2: Correlation plot of the K_{ow} of chemicals and their CB for an emission to air (a,
10 $R^2=0.49$), fresh water (b, $R^2=0.68$), and agricultural soil (c, $R^2=0.28$),
11 respectively. Acephate (\diamond), Aldicarb (\square), Lindane (Δ), and DDT (O) are
12 highlighted. The dotted line is the accompanying linear fit for the data.

13

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

17

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

23

24 Figure 5: Correlation between our new CFs for warm-blooded predators and CFs for cold-
25 blooded species calculated according to existing methodologies, for an emission
26 to fresh water ($R^2=0.26$). Acephate (\diamond), Aldicarb (\square), Lindane (Δ), and DDT (O)
27 are highlighted. The dashed line indicates the 1:1 relation, whereas the dotted line
28 shows the linear fit for the data.

29

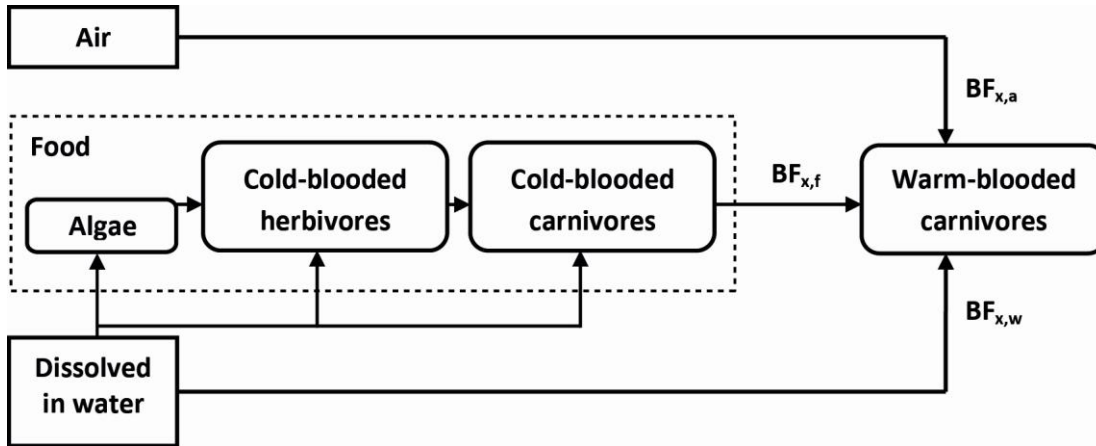
1 Table 1

Chemical	Uptake route	Emission to air	Emission to fresh water	Emission to agricultural soil
Acephate	Fresh water	11	50	50
	Food	11	50	50
	Air	79	0	0
Aldicarb	Fresh water	48	50	50
	Food	48	50	50
	Air	4	0	0
Lindane	Fresh water	35	40	39
	Food	53	60	59
	Air	11	0	2
DDT	Fresh water	2	2	2
	Food	72	98	90
	Air	26	0	8
All	Fresh water	25 (1-47)	43 (11-50)	39 (5-50)
	Food	33 (1-60)	56 (50-87)	50 (11-83)
	Air	42 (5-98)	1 (0-2)	11 (0-80)

2

3

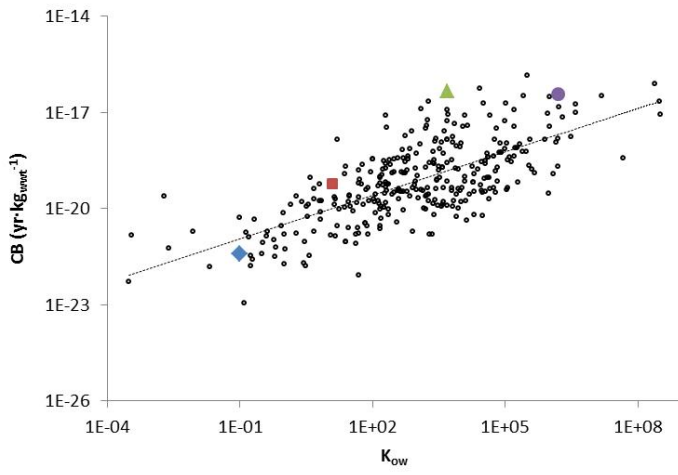
1 Figure 1



2

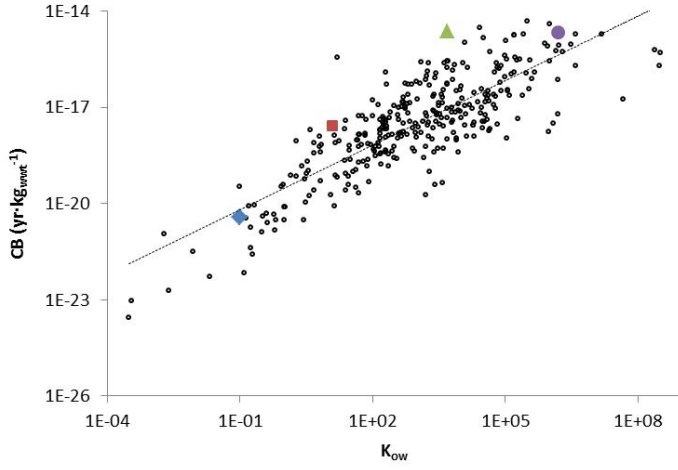
3

1 Figure 2



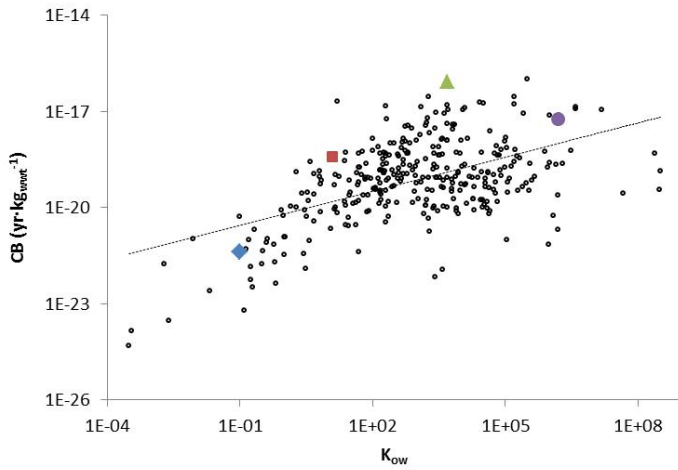
2

(a)



3

(b)

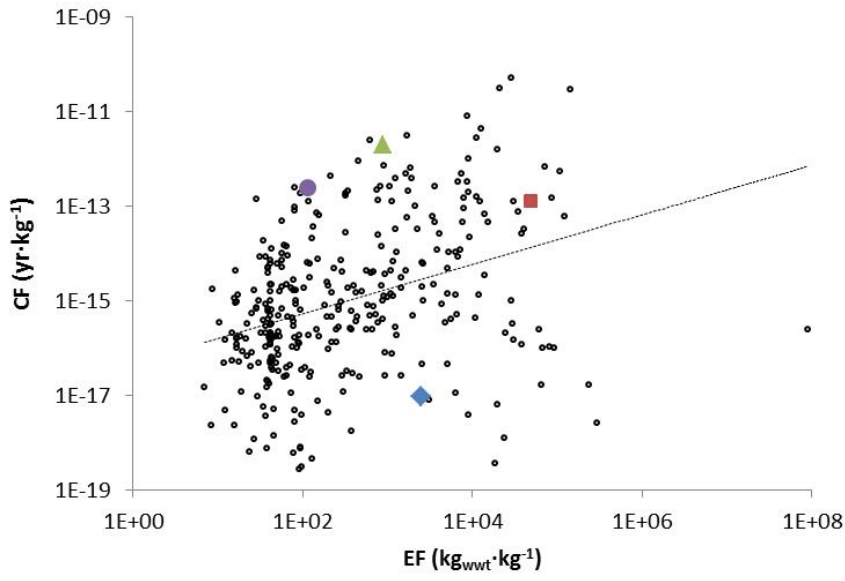


4

(c)

5

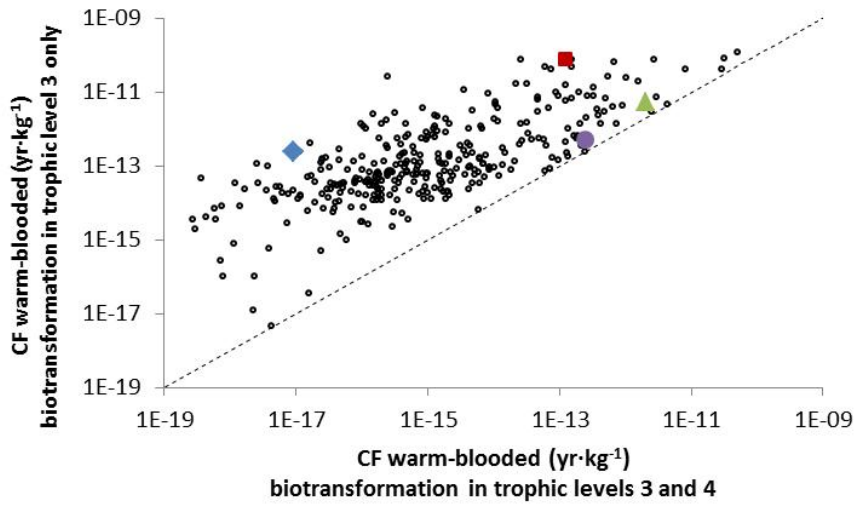
1 Figure 3



2

3

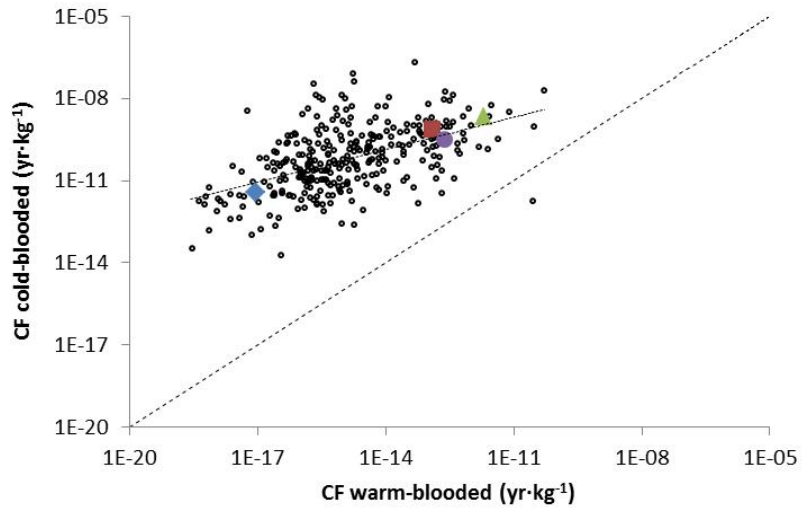
1 Figure 4



2

3

1 Figure 5



2