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Geographical scenario uncertainty in generic fate and exposure factors of toxic pollutants for life-cycle impact assessment

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Abstract

In environmental life-cycle assessments (LCA), fate and exposure factors account for the general fate and exposure properties of chemicals under generic environmental conditions by means of 'evaluative' multi-media fate and exposure box models. To assess the effect of using different generic environmental conditions, fate and exposure factors of chemicals emitted under typical conditions of (1) Western Europe, (2) Australia and (3) the United States of America were compared with the multi-media fate and exposure box model USES-LCA. Comparing the results of the three evaluative environments, it was found that the uncertainty in fate and exposure factors for ecosystems and humans due to choice of an evaluative environment, as represented by the ratio of the 97.5th and 50th percentile, is between a factor 2 and 10. Particularly, fate and exposure factors of emissions causing effects in fresh water ecosystems and effects on human health have relatively high uncertainty. This uncertainty is mainly caused by the continental difference in the average soil erosion rate, the dimensions of the fresh water and agricultural soil compartment, and the fraction of drinking water coming from ground water.

Keywords: LCA; Fate and exposure factors; Ecotoxicity; Human toxicity; Evaluative environment

1. Introduction

Characterisation factors for toxic pollutants, also called toxicity potentials, are substance-specific, quantitative representations of potential impacts per unit emission of a toxic substance. In environmental life-cycle assessments of products (LCAs), these potentials are used to determine the relative contribution of a substance to toxicity related impact categories, such as human toxicity. Generally, generic characterisation factors are used for the comparison of toxic compounds in LCA case studies. These generic characterisation factors account for the general properties of the chemical, such as its toxicity and persistence under generic conditions by means of 'evaluative' multi-media fate and exposure box models (e.g. Hertwich et al., 2001, 2002).

Before generic characterisation factors can be calculated, the choices of the landscape and human

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characteristics have to be discussed. In the calculation of current toxicity potentials, 'evaluative' environments applied represent in most cases the continental scale, such as Western Europe or the United States of America (Huijbregts et al., 2000a; Hertwich et al., 2001). To assess the uncertainty on generic toxicity potentials of using different 'evaluative' environments, fate and exposure characteristics of toxic substances emitted under typical conditions of (1) Western Europe, (2) Australia and (3) the 48 contiguous States of the United States of America will be compared with the 'evaluative' multi-media fate and exposure box model USES-LCA, originally applied by Huijbregts et al. (2000a) for West European conditions. The present article briefly outlines the multi-media fate and exposure part of USES-LCA and the conversion of the model for Australia and the United States. For the three evaluative environments, fate and exposure factors for humans and ecosystems of 375 substances emitted to four different compartments are compared and the uncertainties found are discussed.

2. Method

2.1. Fate and exposure factors

USES-LCA, based on the Uniform System for the Evaluation of Substances 2.0 (USES 2.0) as developed by RIVM et al. (1998), is designed to calculate the potential impacts of toxic substances emitted in Western Europe on humans and ecosystems. USES-LCA models fate, exposure and effects for a time horizon of 20, 100, and 500 years and an infinite time horizon (Huijbregts et al., 2000a, 2001). The fate and exposure part of USES-LCA are briefly explained below. Huijbregts et al. (2000a) give a more extensive elaboration of the model.

The fate module of USES-LCA consists of the nested multi-media fate model Simplebox 2.0 (Brandes et al., 1996), which is used to calculate environmental concentrations in the various environmental media. "Nested" means that chemicals can be transported from one scale to a higher scale and vice versa. USES-LCA has two spatial scales (continental and hemispheric) and three climate zones, reflecting arctic, moderate and tropic climatic zones of the Northern hemisphere (Fig. 1). The continental scale is defined within the moderate climate zone and consists of six compartments: air, fresh water, seawater, natural soil, agricultural soil, and industrial soil. The hemispheric scale comprises the arctic, moderate and tropical climate zones. Every climatic zone is divided in three compartments: air, seawater and soil. Because the hemispheric scale is modelled as a closed system without transport across the system boundaries, emitted substances cannot escape.

The exposure module is used to calculate the change in exposure for the human population via air, drinking water and food products. For an ecosystem, the change in exposure equals the change in the dissolved environmental concentration predicted by the fate module. In formula this means that

$$F_{x,i\to j} = \sum_s N_{j,s} \frac{\mathrm{d}D_{x,j,s}}{\mathrm{d}M_{x,i}} \tag{1}$$

in which $F_{x,i\to j}$ represents the fate and exposure factor that accounts for transport of substance x from compartment i to target j (dimensionless for humans or days for ecosystems), $N_{j,s}$ is the total number of target j at scale s (in number of individuals for humans or in m^3 of water, sediment or soil for ecosystems) and $\mathrm{d}D_{x,j,s}/\mathrm{d}M_{x,i}$ is the marginal change in exposure of substance x by target j at scale s ($\mathrm{d}D_{x,j,s}$ in kg day^{-1} person^{-1} for humans or in kg m^{-3} for ecosystems) caused by a marginal change in the emission of substance x to compartment i ($\mathrm{d}M_{x,i}$ in kg day^{-1}).

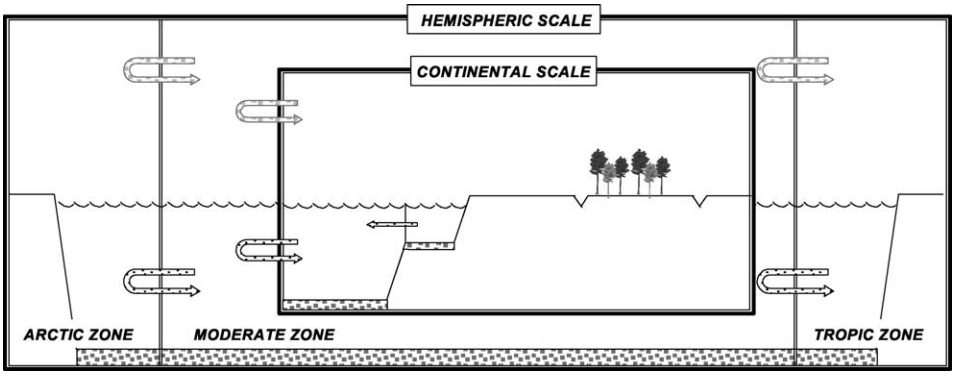

Fig. 1. Schematic representation of transportation routes between different compartments and geographical scales after Brandes et al. (1996).

2.2. Continental scenarios

The following geographical differences between Western Europe, Australia and the United States were taken into account (Table 1):

- environmental parameters and human exposure characteristics on the continental scale were set for conditions representative for the continents under consideration;
- Australia was nested in the tropical zone, while Western Europe and the United States were nested in the moderate zone of the hemispheric scale; and
- for the Australian calculations, environmental parameters on the three climate zones, such as land–sea ratio, were set for the Southern hemisphere, while for Western Europe and the United States environmental parameters on the three climate zones were set for the Northern hemisphere.

2.3. Calculations

The infinite time horizon setting was used in all the USES-LCA calculations. For each continental scenario, fate and exposure factors of 375 substances were calculated for the environmental endpoints (1) fresh water

Table 1
Input parameters for fate analysis and human exposure assessment

	Unit	Australia	US	W-Europe	References
Fate analysis					
Fresh water area [C]	km^2	6.0×10^4	3.5×10^5	1.1×10^5	a–c
Natural soil area [C]	km^2	3.0×10^6	3.9×10^6	2.1×10^6	b–d
Agricultural soil area [C]	km^2	4.5×10^6	3.8×10^6	9.6×10^5	b–e
Industrial/other soil area [C]	km^2	8.0×10^4	3.3×10^5	3.6×10^5	a–c, f
Sea area [C]	km^2	2.5×10^6	1.6×10^6	3.6×10^6	c, g, h
Suspended particles (fresh water) [C]	$kg\,m^{-3}$	3.0×10^{-2}	3.0×10^{-1}	2.5×10^{-2}	b, c, i, j
Settling velocity of suspended particles [C]	$m\,d^{-1}$	10	13	2.5	b,c
Depth (fresh water) [C]	m	1	5	3	b, c, k
Temperature [C]	°C	22	11.6	12	b, c, l
Rainrate [C]	$mm\,year^{-1}$	450	835	700	b, c, m
Soil erosion rate [C]	$mm\,year^{-1}$	2.0×10^{-1}	5.0×10^{-2}	3.0×10^{-2}	b, c, n
Wind speed [C]	$m\,s^{-1}$	3.2	4.4	3.0	b, c, l
Fraction of precipitation that runs off soil [C]	–	0.12	0.27	0.25	b, c, o
Fraction land [M]	–	0.04	0.50	0.50	p
Fraction sea [M]	–	0.96	0.50	0.50	p
Fraction land [T]	–	0.21	0.30	0.30	p
Fraction sea [T]	–	0.79	0.70	0.70	p
Fraction land [A]	–	0.33	0.40	0.40	p
Fraction sea [A]	–	0.67	0.60	0.60	p
OH-radicals [C, M, T, A]	$mol\,cm^{-3}$	6.0×10^5	5.0×10^5	5.0×10^5	q
Human exposure assessment					
Fraction drinking water from surface water [C]	–	0.8	0.5	0.4	b, c, r
Fraction drinking water from ground water [C]	–	0.2	0.5	0.6	b, c, r
Daily intake of drinking water [C]	$l\,day^{-1}$	1.8	1.4	1.4	c, s, t
Daily intake of fish [C]	$kg_{wwt}\,day^{-1}$	0.03	0.02	0.03	c, s, u
Daily intake of leaf crops [C]	$kg_{wwt}\,day^{-1}$	0.73	0.66	0.77	c, s, u
Daily intake of root crops	$kg_{wwt}\,day^{-1}$	0.25	0.08	0.18	c, s, u
Daily intake of meat [C]	$kg_{wwt}\,day^{-1}$	0.20	0.13	0.26	c, s, u
Daily intake of dairy products [C]	$kg_{wwt}\,day^{-1}$	0.39	0.50	0.28	c, s, u
Number of humans [C]	–	1.8×10^7	2.6×10^8	3.7×10^8	p
Number of humans [M]	–	6.1×10^7	2.5×10^9	2.5×10^9	p
Number of humans [T]	–	6.1×10^8	2.3×10^9	2.3×10^9	p
Number of humans [A]	–	1.0×10^4	9.9×10^7	9.9×10^7	p

[C]: continental scale, [M]: moderate zone, [A]: arctic zone, [T]: tropical zone; a: CWA (1980); b: McKone et al. (2000); c: Huijbregts (1999); d: ABS (2001); e: NASS (2001); f: US Census Bureau (2000); g: Bowen et al. (1996); h: NOAA (2001); i: VWRDW (2001); j: DLWC (2001); k: Roser (2001); l: BMA (2001); m: Beckman (1996); n: Noble et al. (1996); o: Crabb (1997); p: Anonymous (1997); q: Atkinson (1988); r: NLWRA (2001); s: USEPA (1999); t: ABS (1999); u: ABS (1998).

aquatic ecosystems, (2) marine aquatic ecosystems, (3) terrestrial ecosystems, and (4) humans after emission to respectively air, fresh water, seawater and agricultural soil. Sediment ecosystems and emissions to industrial soil were not included in the analysis, because Huijbregts (2001) found that fate and exposure factors for sediment and aquatic ecosystems, and fate and exposure factors after emissions to industrial soil and agricultural soil are highly correlated. These high correlations imply that the same type of geographical differences will be found for sediment and aquatic ecosystems and emissions to the two soil compartments, respectively. For humans, not only full fate and exposure factors, taking into account all exposure pathways, were calculated, but also separate fate and exposure factors for the pollutant exposure routes (a) food; (b) drinking water, and (c) inhalation. The datasets of Huijbregts et al. (2000a) and Hertwich et al. (2001) were used to specify substance-specific parameters.

For each human health and ecological endpoint, the fate and exposure factors of all 375 substances for all four emission compartments were used to derive linear regression equations of the form

$$\log F_{j,S_n} = a \log F_{j,S_m} + b \tag{2}$$

in which S_n and S_m are continental scenarios. We optimized the regression equations using a linear least squares fit to find appropriate values of the slope (a) and intercept (b) of the regressions. Apart from the regression parameters a and b, the correlation coefficient (r^2), the residual standard error (SE), and the uncertainty factor k were derived. The correlation coefficient (r^2) represents the fraction of explained variance by the regression equation, while the residual SE represents the average residual error between the calculated F with USES-LCA and the estimated F with the regression equation. The SE can be converted to an uncertainty factor k which is defined such that 95% of the values of a stochastic variable are within a factor k from the median of a lognormal distribution (Slob, 1994). The uncertainty factor k can be calculated from SE by

$$k = \sqrt{\frac{97.5p}{50p}} = 10^{1.96\mathrm{SE}} \tag{3}$$

in which $50p$ and $97.5p$ are respectively the 50th percentile and 97.5th percentile of the uncertainty distribution.

3. Results

Table 2 shows the statistics of the regression equations for the fate and exposure factors of the three continental scenarios. For all ecosystem fate and exposure regression equations it can be seen that the explained variance r^2 is high (>0.95), while the uncertainty factor k is always lower than 5, except for the fresh water fate and exposure factors (FwF) of the US versus Australia with a uncertainty factor k of 8. As the regression coefficient a approaches unity for all ecosystem regression equations, the relationship between the ecosystem fate and exposure factors of the three continental scenarios is approximately linear. Systematic differences between the ecosystem fate and exposure factors of the three continental scenarios, represented by the factor 10^b, remain within a factor 3, except for the comparison of Australian FwFs with the two other continental FwFs (a factor of 10).

For the human fate and exposure regression equations it can also be seen in Table 2 that the explained variance r^2 is high (>0.90). The uncertainty factor k in the human fate and exposure regression equations can be up to a factor of 25. The uncertainty in inhalation exposure regression equations do not largely contribute to the uncertainty in total human fate and exposure factors (HuFs), in contrast to the uncertainty in exposure via food and drinking water intake. Again, the regression coefficient a approaches unity for the human fate and exposure regression equations, resulting in an approximate linear relationship between the HuFs of the three continental scenarios. Systematic differences between the Australian HuFs and the HuFs of the two other continents are relatively high (up to a factor of 70).

4. Discussion

4.1. Evaluative environment

The uncertainty in fate and exposure factors for ecosystems and humans due to choice of an evaluative environment, represented by the uncertainty factor k, is between a factor 2 and 10. Previous investigations indicated that uncertainty in chemical-specific parameters, such as degradation rates, lead to uncertainty factors of 4–50 for human fate and exposure factors (Huijbregts et al., 2000b; Hertwich et al., 1999, 2000) and uncertainty factors of 2–500 for ecosystem fate and exposure factors (Huijbregts et al., 2000b), depending on the chemical and emission compartment considered. Comparing the influence of the choice of an evaluative environment and parameter uncertainty on fate and exposure factors, our results indicate that the influence of the choice of an arbitrary evaluative environment may only be relevant for the fresh water aquatic and human fate and exposure factors.

Uncertainty in the regression equations for fresh water aquatic and human fate and exposure factors can be clarified by the fact that the three evaluative environments do not change fate and exposure in the

Table 2
Regression characteristics of the fate and exposure factors for the three continental scenarios

Regression equation	Statistics				
	a	b	r^2	SE	k
Fresh water aquatic ecosystems					
$\log(\mathrm{FwF_{EU}}) = a\log(\mathrm{FwF_{US}}) + b$	1.02	−0.24	0.99	0.22	2.7
$\log(\mathrm{FwF_{AU}}) = a\log(\mathrm{FwF_{EU}}) + b$	1.03	−0.96	0.98	0.37	5.3
$\log(\mathrm{FwF_{AU}}) = a\log(\mathrm{FwF_{US}}) + b$	1.06	−1.21	0.97	0.47	8.4
Marine aquatic ecosystems					
$\log(\mathrm{MaF_{EU}}) = a\log(\mathrm{MaF_{US}}) + b$	0.97	0.14	0.99	0.19	2.3
$\log(\mathrm{MaF_{AU}}) = a\log(\mathrm{MaF_{EU}}) + b$	1.06	−0.49	0.99	0.30	3.8
$\log(\mathrm{MaF_{AU}}) = a\log(\mathrm{MaF_{US}}) + b$	1.03	−0.34	0.98	0.37	5.3
Terrestrial ecosystems					
$\log(\mathrm{TeF_{EU}}) = a\log(\mathrm{TeF_{US}}) + b$	1.01	−0.10	1.00	0.13	1.8
$\log(\mathrm{TeF_{AU}}) = a\log(\mathrm{TeF_{EU}}) + b$	1.00	−0.28	0.99	0.25	3.1
$\log(\mathrm{TeF_{AU}}) = a\log(\mathrm{TeF_{US}}) + b$	1.01	−0.38	0.99	0.30	3.9
Humans (total)					
$\log(\mathrm{HuF_{EU}}) = a\log(\mathrm{HuF_{US}}) + b$	1.03	0.62	0.95	0.38	5.5
$\log(\mathrm{HuF_{AU}}) = a\log(\mathrm{HuF_{EU}}) + b$	0.96	−1.63	0.91	0.52	10.3
$\log(\mathrm{HuF_{AU}}) = a\log(\mathrm{HuF_{US}}) + b$	1.01	−0.90	0.91	0.52	10.7
Humans (food)					
$\log(\mathrm{HuF_{EU}}) = a\log(\mathrm{HuF_{US}}) + b$	1.04	0.76	0.95	0.43	7.0
$\log(\mathrm{HuF_{AU}}) = a\log(\mathrm{HuF_{EU}}) + b$	0.96	−1.78	0.95	0.42	6.8
$\log(\mathrm{HuF_{AU}}) = a\log(\mathrm{HuF_{US}}) + b$	1.00	−0.96	0.93	0.50	9.6
Humans (drinking water)					
$\log(\mathrm{HuF_{EU}}) = a\log(\mathrm{HuF_{US}}) + b$	1.04	0.79	0.99	0.28	3.5
$\log(\mathrm{HuF_{AU}}) = a\log(\mathrm{HuF_{EU}}) + b$	0.97	−1.85	0.96	0.69	22.4
$\log(\mathrm{HuF_{AU}}) = a\log(\mathrm{HuF_{US}}) + b$	1.01	−1.07	0.95	0.71	24.4
Humans (air)					
$\log(\mathrm{HuF_{EU}}) = a\log(\mathrm{HuF_{US}}) + b$	0.99	0.20	1.00	0.16	2.1
$\log(\mathrm{HuF_{AU}}) = a\log(\mathrm{HuF_{EU}}) + b$	0.99	−1.23	1.00	0.29	3.6
$\log(\mathrm{HuF_{AU}}) = a\log(\mathrm{HuF_{US}}) + b$	0.98	−1.10	1.00	0.32	4.3

AU: Australia; US: United States of America; EU: Western Europe; FwF: fresh water fate and exposure factor; MaF: marine fate and exposure factor; TeF: terrestrial fate and exposure factor; HuF: human fate and exposure factor; r^2: explained variance; SE: residual standard error; k: uncertainty factor.

same way (1) for all substances and (2) for all initial emission compartments. A combination of the following factors cause substance-specific differences in the fate and exposure factors of the three evaluative environments:

- the lower Australian fresh water area compared to Western Europe and the United States results in lower chemical *residence times* in the fresh water compartment and thus lower FwFs, if burial of sediment or advection to the marine environment are the most important chemical removal processes from the fresh water compartment. The lower fresh water area also results in a higher dissolved *concentration* of chemicals in the fresh water compartment and thus higher HuFs for drinking water and fish intake, if degradation or volatilisation are the most important chemical removal processes from the fresh water compartment. Differences are up to a factor of 6.
- the lower depth of the Australian fresh water compartment compared to Western Europe and the United States results in lower chemical *residence times* and thus lower FwFs, if burial of sediment, volatilisation or advection to the marine environment are the most important removal processes for chemicals from the fresh water compartment. The lower depth also results in a higher dissolved *concentration* of chemicals and thus higher HuFs for drinking water and fish intake, if degradation is the most important chemical removal processes from the fresh water compartment. Differences are up to factor of 5.
- the lower rainrate and fraction of precipitation that runs off soil in Australia compared to Western Europe and the United States result in a smaller transfer

from soil to fresh water of substances with relatively low solid–water partition coefficients. Australian FwFs and HuFs for drinking water and fish intake of these of substances are up to factor of 4 lower; and
- the relatively high soil erosion rate in Australia results in a higher average net sedimentation and consequently a higher burial of sediment in the Australian fresh water compartment compared to Western Europe and the United States. As sedimentation is a significant removal route for water persistent, hydrophobic, low-volatility substances, and metals with relatively high solid–water partition coefficients, both the residence time and concentration will be lower for these type of substances in Australian surface waters, resulting in lower FwFs and HuFs for drinking water and fish intake of these of substances (up to a factor of 7).

The following differences in the evaluative environments cause emission compartment-specific deviations in the fate and exposure factors:

- the lower Australian fresh water area compared to Western Europe and the United States results in a smaller chemical transfer from air to fresh water, lowering the Australian FwFs and HuFs of substances emitted to air, seawater and agricultural soil (up to a factor 5), while the smaller chemical transfer from air to fresh water does not change the FwFs and HuFs of substances emitted to fresh water;
- the lower rainrate and fraction of precipitation that runs off soil in Australia compared to Western Europe and the United States results in smaller chemical transfer from soil to the fresh water, lowering the Australian FwFs and HuFs of substances emitted to agricultural soil, air and seawater (up to factor of 4). The smaller chemical transfer from soil to fresh water does not change the FwFs and HuFs of substances emitted to fresh water; and
- the Australian HuFs of substances emitted to agricultural soils are on average a factor of 160 lower compared to the Western European HuFs, while substances emitted to air, fresh water and seawater are on average a factor of 20 lower. The combination of the larger area of agricultural soil, the higher erosion rate and the smaller fraction of drinking water coming from ground water in Australia compared to Western Europe explain the lower Australian HuFs of substances emitted to agricultural soils. The larger area of agricultural soil and the higher erosion rate in Australia result in lower soil and ground water concentrations per unit emission and, as a result, lower concentrations in agricultural food products and drinking water coming from ground water. These differences lower the Australian HuFs of substances with agricultural food products and drinking water as dominant human exposure routes, which is the case for most substances emitted to the agricultural soil.

4.2. Site dependency

A number of studies addressed the potential importance of spatial differentiation in the impact assessment of toxic substances in LCA (Hertwich et al., 2000; McKone et al., 2000; Nigge, 2000; Potting, 2000; Krewitt et al., 2001; Schulze, 2001). In the context of spatial differentiation, the present study shows systematic differences between continental FwFs and total HuFs of Australia versus Western Europe and the United States between a factor of 9–16 and 8–43, respectively. These systematic differences in continental fate and exposure factors are caused by differences in, for instance, human population numbers, temperature and hydroxyl radical concentration at the different continents. The significance of these systematic continental differences will depend, however, on (1) the spatial range of the pollutant and (2) the spatial scale at which important regional parameters vary (Hertwich et al., 2002). According to Hertwich and McKone (2000), the spatial range of a pollutant is the mean distance that a pollutant travels once it is released and can be calculated by the product of persistence and average mobility. Following this definition, USES-LCA calculated that for about 90% of the substances the spatial range is smaller than the continental scales as used in our calculations. If the spatial scale at which important regional parameters vary is smaller than the continental scale, scenarios are required at a smaller geographical scale. The question whether scenarios are required at a smaller scale may be answered by comparing the state-to-state (or region-to-region) variability in fate and exposure characteristics *within* the three continents with the variation of fate and exposure characteristics *between* these three continents. Further research is required to reveal whether the systematic differences found between the three evaluative environments are of direct relevance for LCA purposes.

5. Conclusions

The model USES-LCA, suitable for Western European conditions, can be modified to reflect conditions of other geographical regions, such as Australia and the United States. For each of the three geographical regions, USES-LCA was applied to calculate fresh water, marine, terrestrial and human fate and exposure factors of 375 substances emitted to four different compartments. From the comparison of the three evaluative environments it was found that fate and exposure factors of emissions causing effects in fresh water ecosystems and human health effects have relatively high

uncertainty. In this respect, the choice of the average soil erosion rate, the dimensions of the fresh water and agricultural soil compartment, and the fraction of drinking water coming from ground water is considered important in the calculation of generic fate and exposure factor for toxic pollutants.

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