Regional LCA in a global perspective

Wegener Sleeswijk, A.

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GLOBOX: a spatially differentiated global fate, intake and effect model for toxicity assessment in LCA*

Abstract

GLOBOX is a model for the calculation of spatially differentiated LCA toxicity characterisation factors on a global scale. It can also be used for human and environmental risk assessment. The GLOBOX model contains equations for the calculation of fate, intake and effect factors, and equations for the calculation of LCA characterisation factors for human toxicity and ecotoxicity. The model is differentiated on the level of 239 countries/territories and 50 seas/oceans. Each region has its own set of homogeneous compartments, and the regions are interconnected by atmospheric and aquatic flows. Multimedia transport and degradation calculations are largely based on the EUSES 2.0 multimedia model, and are supplemented by specific equations to account for the advective air and water transport between different countries and/or seas. Metal-specific equations are added to account for speciation in fresh and marine surface water. Distribution parameters for multimedia transport equations are differentiated per country or sea with respect to geographic features, hydrology, and climate. The model has been tested with nitrobenzene as a test chemical, for emissions to all countries in the world. Spatially differentiated characterisation factors turn out to show wide ranges of variation between countries, especially for releases to inland water and soil compartments. Geographic position, distribution of lakes and rivers and variations in environmental temperature and rain rate are decisive parameters for a number of different characterisation factors. Population density and dietary intake play central

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roles in the variation of characterisation factors for human toxicity. Among the countries that show substantial deviations from average values of the characterisation factors are not only small and remote islands, but also countries with a significant economic production rate, as indicated by their GDPs. It is concluded that spatial differentiation between countries is an important step forward with respect to the improvement of LCA toxicity characterisation factors.

Keywords

LCA, spatial differentiation, toxicity assessment, multimedia model, global model, actual impacts

5.1 Introduction

The life cycles of products 'from cradle to grave' comprise large numbers of economic processes, including mining activities, agricultural and industrial processes, product use activities, and waste processing. A product life cycle may consist of tens to hundreds of processes, taking place in many different parts of the world. In product life cycle assessment (LCA), the environmental effects of all these processes can be quantified, resulting in an environmental profile. This profile comprises different categories of environmental impacts (the so-called 'impact category indicators'), ranging from global warming to more regional and local effects like acidification and local toxicity-related impacts. A formalised framework for LCA has been defined by the International Organization for Standardization, in the 14040 series (ISO, 2006a,b). This framework offers guidelines for the overall structure and terminology of LCA methods. With respect to toxicity assessment, Pennington *et al.* (2006) contrast risk assessment for regulatory purposes, in which worst-case situations and safety factors are import elements, with comparative risk assessment and LCA, in which a more realistic and fair comparison is the aim. This chapter builds on the comparative paradigm.

Much progress has been made during the past ten years by the introduction of the multimedia modelling concept into LCA toxicity characterisation (*cf.* Guinée and Heijungs, 1993), which has been explicitly recommended by the Society of Environmental Toxicology and Chemistry (SETAC) Europe First Working Group on Life-Cycle Impact Assessment (WIA-1) (Hertwich *et al.*, 2002). Spatial differentiation of these models on a global scale can be considered as a natural next step.

Multimedia environmental models – as first proposed by Mackay (1991) are widely used for toxicity characterisation in LCA. Commonly used models include USES-LCA (Huijbregts *et al.*, 2000; Van Zelm *et al.*, 2009), CalTOX (Hertwich *et al.*, 2001), IMPACT 2002 (Pennington *et al.*, 2005) and USEtox (Rosenbaum *et al.*, 2008). In environmental risk assessment – for which most multimedia models have originally been designed – the spatial scope of the fate and intake model is

generally linked to the magnitude of the region for which it is to be applied, and its direct surroundings. Product life cycles, however, usually include processes from all over the world. For this reason, the spatial scope of regional models should be expanded for use in LCA. This implies that the ranges of model parameters, such as environmental temperature or meat consumption, will largely increase, which brings up the question whether spatial differentiation may be inescapable for fate and intake assessment in LCA.

The subject of spatial differentiation in LCA has been pioneered by Potting (2000) with practical applications for acidification (Potting *et al.*, 1998) and for human exposure to air emissions. Other early studies on the subject include those by Hertwich *et al.* (2001), McKone *et al.* (2000), Nigge (2000), Krewitt *et al.* (2001) and Schulze (2001).

Several authors have introduced spatial differentiation into comprehensive LCA impact assessment models (*cf.* Huijbregts *et al.*, 2003; Hauschild and Potting, 2005; Potting and Hauschild, 2005; Pennington *et al.*, 2005; Rochat *et al.*, 2006; Humbert *et al.*, 2009). In some spatially differentiated multimedia models, a difference is made between an evaluative region (for which emissions can be entered in the model) and a larger, encompassing region of dispersion, in which the emission region is nested. In the USES-LCA model (Huijbregts *et al.*, 2000; Van Zelm *et al.*, 2009), the evaluative region at the continental level (Western Europe) is not spatially differentiated, but the dispersion region (the northern hemisphere) is characterised by its own environmental parameters for three different climate zones. Huijbregts *et al.* (2003) evaluated the influence of spatial differentiation at the continental level by comparing three different versions of the USES-LCA model, with Western Europe, the United States and Australia as three alternative continental levels. Pennington *et al.* (2005) have introduced spatial differentiation in the IMPACT 2002 model at three levels: the level of Western European watersheds (for soil and surface water) and grid cells (for air and sea/ocean), the continental level of Western Europe, and the global level, in which the continental level is nested. Emissions can be entered at the watershed/grid cell or at the continental level. Rochat *et al.* (2006) have applied spatial differentiation at the level of continents to a global version of the IMPACT 2002 model with respect to both emission and dispersion. Another regionally differentiated multimedia model, that has not been designed specifically for LCA, but that has been used in the LCA-context, is BETR-North America (MacLeod *et al.*, 2001). This model comprises North America, differentiated at the level of ecological regions. Humbert *et al.* (2009) recently developed the IMPACT North America model, in which the evaluative region North America – which is nested into a global dispersion level – is differentiated at the level of several hundred zones.

Global, spatially differentiated fate models that are not specifically designed for LCA include Globo-POP (Wania and Mackay, 1995), BETR-World (Toose *et al.*,

2004) and BETR-Global (MacLeod *et al.*, 2005). These models are primarily designed for the analysis of the global distribution of persistent organic pollutants (POPs), *i.e.* the 'global fractionation' phenomenon. In principle, these models could also form a useful basis for LCA-directed global fate modelling.

The GLOBOX model for LCA toxicity assessment comprises the entire world. With respect to the basis of spatial differentiation, four types of possibilities had to be considered:

1. Differentiation on the basis of ecozones (*e.g.* Webster *et al.*, 2004). This type of regionalisation defines the region according to homogeneous ecological conditions. It is basically an effect-oriented differentiation.
2. Differentiation on the basis of watersheds (*e.g.* Pennington *et al.*, 2005). Here the regions are defined according to distribution-oriented features, with emphasis on the distribution of chemicals in the aquatic compartment.
3. Differentiation on the basis of grid cells (*e.g.* Prevedouros *et al.*, 2004). This method of defining regions easily connects with GIS-available data, *e.g.*, on vegetation, population, and wind.
4. Differentiation on the basis of political boundaries, *e.g.* continents and oceans or countries and seas. This is primarily an inventory-driven approach, for instance it connects with country-specific emission databases, like ecoinvent or the TRI. But of course, there are also intake-related parameters (like food consumption patterns) which are available at this level.

Since LCA studies often have to deal with large numbers of emissions to different environmental compartments, an inventory-driven approach is specifically interesting in the context of LCA. For this reason, spatial differentiation in the GLOBOX model is based on political boundaries. In order to account for the large differences in human intake characteristics that exist within continents, the level of countries and seas was chosen as the basis of differentiation. GLOBOX is a level 3 (or steady state) multimedia model (Mackay, 1991), based on the European Union model EUSES 2.0 (EC, 2004), and can be considered as an extended and more refined elaboration of this model.

The main goals of the GLOBOX model are:

1. Accounting for spatial variation in fate, intake and effect parameters at the level of countries/territories and seas/oceans.
2. Accounting for the global range of life cycles.
3. Accounting for life cycle processes outside the Euro-American and Japan regions.

The idea behind the model is that LCA requires region-specific characterisation factors (CFs) for releases of any toxic chemical at any location in the world. These

factors should account for the summed impacts of such an emission in all countries/territories (further referred to as 'countries') and seas/oceans (further referred to as 'seas') over which it is dispersed during its lifetime.

GLOBOX basically consists of three related parts. First, it is a mathematical model for fate, intake and effect. Second, the mathematical equations contain many regionalised parameters, such as temperature, lake depth and leaf crop consumption. This chapter describes some of the estimation routines. The regionalised parameters themselves are available as the GLOBACK data set. Third, the model equations and an interface with the GLOBACK data set have been implemented in software. The GLOBOX software program, a full list of model equations, and the GLOBACK parameter set are downloadable from cml.leiden.edu.

5.2 The GLOBOX model

5.2.1 Model structure

GLOBOX is based on the EUSES 2.0 model (EC, 2004). Apart from a higher level of spatial differentiation, the main difference between GLOBOX and EUSES 2.0 is a difference in model structure: while the EUSES 2.0 unit world consists of a number of scales that are nested into each other, the GLOBOX environmental assessment system consists of a series of interconnected regions at the same modelling level.

Like most general purpose fate-exposure-effect models (Rosenbaum *et al.*, 2007), the GLOBOX model consists of three main modules: an impact category independent fate module, a human intake module, applicable to all impact categories that are related to human intake of chemicals, and an effect module, in which effect-related parameters can be introduced for every separate impact category. The effect module is the only module that focuses on impact category specific processes and data. Both other modules are impact category independent. The impact category specific character of the characterisation factor – which is the product of fate-, intake and effect factor – is thus determined by the effect factor only.

A specific parameter set – GLOBACK – contains estimates on fate and intake-related parameters for each separate country and sea. All data can be overruled by the user's own estimates if desired. The model requires only substance-specific input of physico-chemical and toxicity data to calculate specific human and ecological toxicity characterisation factors for unit emissions to any compartment in any country of the world. The principal characteristics of the GLOBOX model are listed in Table 5.1.

Table 5.1 Summary table of principal characteristics of the GLOBOX model.

model quality	elaboration
basics	multimedia model based on EUSES 2.0
impact categories (toxicity related)	human toxicity (carcinogenic and non-carcinogenic) and ecotoxicity (aquatic (separate for river, lake and salt lake) and terrestrial)
fate, intake and effect	all included
emissions compartments	air; rivers, freshwater lakes, salt lakes; seawater; natural soil, agricultural soil; urban soil
distribution compartments	air, rivers, freshwater lakes, salt lakes, groundwater, seawater, freshwater lake sediment, salt lake sediment, sea sediment, natural soil, agricultural soil, urban soil
chemicals considered	organic chemicals and metals
spatial variation	distinction between 239 different countries and 50 different seas (global scale)
intake routes considered	air, drinking water, leaf crops, root crops, meat, dairy, freshwater fish, sea fish
additional options to basic model	taking into account above- and below-threshold concentrations separately. (Requires additional data input). Risk assessment calculations (steady state), *e.g.* emission scenarios.
chemical input and model parameters	model parameters included in model (can be changed by user); chemical input to be entered by user
distinct features of model	global range of model; high level of spatial differentiation; accounting specifically for cold regions and salt lakes; specific equations for metals in water

The combination of a global range and a high level of spatial differentiation is the central feature that distinguishes the GLOBOX model from other LCA multimedia models. 239 individual countries/territories (CIA, 2004) and 50 individual seas/oceans (IHO, 1986) form the basic level of spatial differentiation in the model.

At the level of effect assessment, the model contains specific equations for the distinction between potential and actual impacts. Two aspects are accounted for in this context: first the difference between sensitive and insensitive areas, and second the difference between areas with prevailing environmental concentrations above and below environmental threshold levels, as we proposed earlier (Wegener Sleeswijk, 2003).

The *Supporting information* provides further details on aspects such as a spatially differentiated version of the global hydrological cycle, constructed for the GLOBOX model, the treatment of cold regions, and metal-specific model features.

5.3 Fate

5.3.1 General

Multimedia transport and degradation calculations are largely based on the multimedia fate model in EUSES 2.0, and are supplemented by specific equations to account for environmental-flow-mediated (advective) air and water transport between different countries and/or seas. All distribution equations are solved simultaneously by matrix inversion, allowing for the calculation of global-scale multimedia transport. The model-internal output of the fate module, applied to a standard emission of a certain substance to a certain compartment, is a list of approximately 3000 fate factors describing the specific spatial distribution of this emission over all environmental boxes. These factors can be considered as the time-integrated amounts of substance in all individual boxes that result from the given standard emission (Heijungs, 1995).

5.3.2 Specific features of the GLOBOX fate module

The fate module is differentiated with respect to geographic, geophysical, climatological, intermedia transfer and water-balance-related parameters. In addition to the subjects covered by the EUSES 2.0 model, a number of fate-related subjects are explicitly addressed in the GLOBOX model:

- cold regions: permanent or temporal ice cover of land and sea areas
- rivers, freshwater lakes and internally drained (endorheic) salt lakes as three separate inland water compartments
- groundwater as a separate compartment
- irrigation of agricultural soil with surface water and groundwater, respectively
- metal-specific equations.

EUSES 2.0 is spatially differentiated on the basis of a limited number of spatially differentiated parameters. The GLOBOX fate module has a wider range of spatial differentiation. In Table 5.2, the spatially differentiated parameters in the fate module of the GLOBOX model are listed, together with the sources from which the values have been extracted or derived. Parameters that have not been spatially differentiated are not included in this table. The values of these last parameters have been set to the corresponding parameter values in the EUSES 2.0 model (*e.g.* 1000 m for the atmospheric mixing height).

Table 5.2 Spatially differentiated fate parameters in the GLOBOX model.

parameter	sources used (direct or for derivation)
area of country lengths of land boundaries[a] lengths of coastlines[a]	CIA (2000, 2004)
fraction natural, agricultural and urban soil	CIA (2000, 2004); UNSD (2007); WRI (2000)
land area, covered by freshwater lakes[b] land area, covered by endorheic salt lakes[a] total depths and mixing depths of lakes[b] water residence time in lakes[b]	Babkin (2003); CIA (2004); ILEC/UNEP (1994, 1995, 1996); LakeNet (2004); Statistics Finland (2004); SWCSMH (2004)
area of lake sediment that is part of the system	follows from assumption on maximum mixing depth
area of sea total depth of sea[b]	Statistics Finland (2004); Van der Leeden *et al.* (1990)
area of sea sediment that is part of the system	follows from assumption on maximum mixing depth
lengths of sea boundaries[a]	calculations, based on latitudes and longitudes at the border edges
permanent ice cover of land (glaciers and ice fields)*	CIA (2004); Statistics Finland (2004)
mixing depth of the natural and urban soil compartments[b] suspended matter concentration in inland waters and seawater[b] fraction of solids in soil runoff[b] fractions of air, water and solids in natural, agricultural and urban soil[b] fraction of organic carbon in soil, lake and sea suspended matter and lake and sea sediments[b] sea temperature[b] fraction of sea area covered by ice[b] deposition rate of aerosol particles[b]	Wania & Mackay (1995); Troll & Paffen (1966)
concentration of hydroxyl radicals in air[b]	Toose *et al.* (2004)
temperature (countries) wind speed (countries) temporal ice cover (lakes)[a]	Nellestijn & Dekker (1995); long year averages (30 years); values, measured at weather station in or near capitals used as approach for country-scale values
rain rate (countries)[b]	FAO (2004); Nellestijn and Dekker (1995); Korzoun *et al.* (1977) (for Antarctica)
rain rate (seas)[b]	Babkin (2003); Korzoun *et al.* (1977); Van der Leeden *et al.* (1990)
river inflows from other countries	FAO (2004)

parameter	sources used (direct or for derivation)
river outflows to other countries	
water runoff from soil	
water flow from soil to groundwater	
water flow from groundwater to river water	
groundwater inflows from other countries	
groundwater outflows to other countries	
use of surface water for sprinkling and irrigation[a]	FAO (2004); WRI (2004)
use of groundwater for sprinkling and irrigation[a]	
water flow from groundwater to sea	
volume of rivers[a]	Shiklomanov & Rodda (2003)
volume of ice shields[a]	
evaporation rate from lakes	climate-dependent assumptions
evaporation rate from salt lakes	
river outflows to sea	FAO (2004); Gleick (1993); balancing items

[a] Not included in EUSES 2.0.

[b] Not differentiated in EUSES 2.0.

5.3.3 Compartments

Twelve distribution compartments are distinguished: air, rivers, freshwater lakes, freshwater lake sediments, salt lakes, salt lake sediments, natural, agricultural and urban soil, groundwater, seawater, and seawater sediments. Compared to EUSES 2.0, salt lakes, salt lake sediments, and groundwater are additional compartments. The distinction of river and freshwater lake as two separate freshwater compartments is new as well. Only endorheic salt lakes are accounted for in the salt lake compartment – salt lakes with an open connection to the sea are excluded from the assessment. The separate distinction of endorheic salt lakes (and their sediments) makes it possible to pay attention to the vulnerability of the ecosystems in these compartments, from which pollutants cannot escape by water outflow. Groundwater is distinguished as a separate compartment in order to deal with the influence of groundwater pollution on the quality of drinking water and irrigation water. Finally, the distinction between rivers and lakes as separate freshwater compartments makes it possible to account for the large regional differences in residence time between freshwater compartments in different countries, caused by relative differences in lake volume. Reservoirs are considered to be part of the *freshwater lake* compartments.

5.3.4 Intramedium transport

The *Supporting information* contains further details on the transport of a chemical between the air compartments of adjacent regions, between different types of aquatic compartments within each region, into and between the seas, and into the

groundwater. It also provides information on the subdivision of the soil compartment regarding various types of soil use.

5.4 Human intake

5.4.1 General

Human intake characteristics are decisive for the eventual human exposure to pollutants in the environment. In Table 5.3, the spatially differentiated parameters in the human intake module of the GLOBOX model are listed, together with the sources from which their values have been extracted or derived.

Table 5.3 Spatially differentiated human intake parameters in the GLOBOX model.

parameter	sources used (direct or for derivation)
population	CIA (2004)
average human body weight[b]	CIA (2004); World Bank (2000)
air inhalation rate[b]	CIA (2004)
drinking water consumption[b]	CIA (2004)
origin of drinking water (groundwater or surface water)	WRI (2004)
purification of drinking water from surface water (fraction)[b]	UNSD (2004)
food consumption for each food category (leaf crops, root crops, dairy products, meat, freshwater fish, marine fish)[b]	FAO (2000)
food import for each food category[a]	FAO (2000)
food export for each food category[a]	FAO (2000)

[a] Not accounted for in EUSES 2.0.

[b] Not differentiated in EUSES 2.0.

A detailed description on how these parameters have been derived is given in the *Supporting information*.

5.4.2 Intake fraction versus characterisation factor

Although LCIA needs characterisation factors, many authors nowadays concentrate on the intake fraction (iF) in their publications on human toxicity assessment in LCIA (c.f. Pennington *et al.*, 2005; Huijbregts *et al.*, 2005; Rochat *et al.*, 2006; Rosenbaum *et al.*, 2008; Humbert *et al.*, 2009). There is a fundamental reason why we made another choice here. The intake fraction expresses the fraction of a chemical, wherever released, that enters the population, wherever the individuals live (Bennett *et al.*, 2002). The body weight of these individuals is not accounted for in this variable. In the GLOBOX model, values for the human body weight are regionally differentiated, ranging between 30 kg (for four African countries) and 70 kg (for Vatican City), depending on the degree of welfare and the relative number of children. Although it may be questioned whether the introduction of a body weight (with a variation of only a factor 2) makes enough difference to justify the introduction of an extra complication, we like to emphasise the more fundamental

side of it in this chapter, in which the relative role of different modelling parameters takes such a central place. It is in that context that we focus on the characterisation factor instead of the intake fraction.

For a related reason, we replaced the production-based approach for determining the intake by humans (Pennington *et al.*, 2006) by a consumption-based approach, accounting for the intake per kilogram of body weight. Thus, if polluted food is consumed by a population, characterised by a relatively low average body weight, the resulting impact will be relatively high, and vice versa. A drawback of this approach is the fact that the modelling of export and import of food requires data and assumptions that may introduce additional uncertainty. Nevertheless, we believe that the principle of using a consumption-based approach in combination with a regionalised body weight is an innovative feature of GLOBOX that deserves a further discussion as to its practical trade-off between accuracy and precision.

5.5 Toxic impacts

5.5.1 General

Region-specific values for toxicity data like EC_{50} and ED_{50} (median Effect Concentration and Dose, respectively) values are not available for most chemicals. This does not imply, however, that it should be impossible to add any region-specific information on toxicity as such. In an earlier publication, we advocated the introduction of the possibility to distinguish between sensitive and non-sensitive areas and between areas with prevailing concentrations above and below a certain environmental threshold (Wegener Sleeswijk, 2003). This distinction is sometimes also referred to as the difference between 'actual' and 'potential' impacts (White *et al.*, 1995). Although LCA traditionally focuses on potential impacts, we have introduced the possibility to choose for an assessment of 'above threshold' effects in the effect module of the GLOBOX model. To this end, two new, both chemical and impact category specific parameters are introduced: the *sensitivity factor* (SF) for ecotoxicity and the *threshold factor* (TF) for both ecotoxicity and human toxicity.

The sensitivity factor reflects the fraction of area that is sensitive to a certain chemical, *e.g.* the fraction of area covered by sensitive ecosystems, while the threshold factor reflects the fraction of sensitive area where a predefined no-effect level for the ecosystem concerned (*e.g.* the *hazardous concentration 5%* (HC_5): the concentration above which 5% of species are adversely affected) is exceeded by the existing background concentration of that chemical. A low value of the sensitivity factor reflects the situation that a large part of a region does not house sensitive species. This may be the case in highly urbanised regions, on bare rocks, where terrestrial ecosystems are almost lacking, or in regions where aquatic or terrestrial ecosystems do occur, but not the species that are sensitive to a specific chemical. A low value of the threshold factor reflects the situation that the background con-

centration exceeds the threshold value in only a limited part of the region, *e.g.* around a certain factory. Note that SF and TF reflect the presence of actual impacts of a chemical in a region. They do not concern the severity of an impact (Humbert *et al.*, 2009) or the safety margin in case of incomplete data for regulatory purposes (Pennington *et al.*, 2006).

As a default, SF and TF are both set to a value of 1. Available information on (in)sensitive areas or on a distinction between above and below-threshold situations can be added in a simple and consistent way by estimating the SF and/or the TF, reflecting the fraction of the area of each region that is sensitive to a certain chemical, or the fraction of area where a certain threshold is (almost) reached or exceeded.

5.5.2 Characterisation of toxic impacts

The GLOBOX model combines fate-, intake and effect factors for the calculation of region-specific toxicity characterisation factors. The physico-chemical and toxicity-related data, needed as input for the exemplifying calculations in this chapter, were taken from the USES-LCA 1.0 model (Huijbregts *et al.*, 2000), which includes substance-specific data sets. With respect to toxicity, the USES-LCA 1.0 data set lists DALYs (*disability adjusted life years*) for human toxicity and PDFs (*potentially disappeared fractions*) for aquatic and terrestrial ecotoxicity.

Toxicity-data may vary among different sources, not only with respect to their values, but also with respect to toxicity modelling. For use in LCA, it is important that the toxicity data for different chemicals reflect the best estimate of toxicity ratios between different chemicals as well as possible, without uncertainty-dependent safety margins that are sometimes used for risk assessment purposes.

Characterisation factors for the calculation of spatially differentiated potential impacts are calculated as the volume-weighted (for ecotoxicity) or population-weighted (for human toxicity) average RCR-values:

$$IPECRtox_{eco} = \frac{\sum_{j \in L} \left(RCRtox_{eco}(j) \times V_{comp}(j) \right)}{\sum_{j \in L} \left(V_{comp}(j) \right)}$$

for ecotoxicity and

$$IPECRtox_{hum} = \frac{\sum_{j \in L} \left(RCRtox_{hum}(j) \times Npop(j) \right)}{\sum_{j \in L} Npop(j)}$$

for human toxicity, where $IPECRtox_{eco}$ (–) and $IPECRtox_{hum}$ (–) are the *integrated potential effect characterisation ratios* for ecotoxicity (terrestrial, river, freshwater lake, salt lake or seawater) and human toxicity, respectively, $RCRtox_{eco}(j)$ (–) and $RCRtox_{hum}(j)$ (–) are the *risk characterisation ratios* for region j for ecotoxicity and human toxicity, $V_{comp}(j)$ (m^3) refers to the volume of the compartment in region j where the corresponding ecosystem dwells (soil, river, freshwater lake, salt lake or seawater), $N_{pop}(j)$ (–) is the human population in region j and L is the total set of regions. Human toxicity may refer to carcinogenic or to non-carcinogenic toxicity.

Multiplication of the (unweighted) characterisation factors for the potential effect by the sensitivity factor and the threshold factor yields the 'actual' impact score:

$$LAECRtox_{eco} = \frac{\sum_{j \in L} \left(RCRtox_{eco}(j) \times V_{comp} \times SF_{eco}(j) \times TF_{eco}(j) \right)}{\sum_{j \in L} \left(V_{comp}(j) \right)}$$

for ecotoxicity and

$$LAECRtox_{hum} = \frac{\sum_{j \in L} \left(RCRtox_{hum}(j) \times Npop(j) \times TF_{hum}(j) \right)}{\sum_{j \in L} Npop(j)}$$

for human toxicity, where $LAECRtox_{eco}$ (–) and $LAECRtox_{hum}$ (–) are the *integrated actual effect characterisation ratios* for ecotoxicity and human toxicity, $SF_{eco}(j)$ (–) is the sensitivity factor for ecotoxicity and $TF_{terr}(j)$ (–) and $TF_{hum}(j)$ (–) are the threshold factors for ecotoxicity and human toxicity, respectively. The sensitivity factor is not defined for human toxicity because all human populations are assumed to be equally sensitive to the chemicals to be assessed.

It is usual in LCA to define toxicity characterisation factors in relation to the emission of a reference substance to a certain reference compartment:

$$CF_{toxpot}(s) = \frac{IPECRtox(s)}{IPECRtox(ref)}$$

for potential impacts and

$$CF_{toxact}(s) = \frac{LAECRtox(s)}{LAECRtox(ref)}$$

for actual impacts, to be used for ecotoxicity ($CF_{tospoteco}$ (–) and $CF_{toxacteco}$ (–)) and human toxicity ($CF_{tospothum}$ (–) and $CF_{toxacthum}$ (–).

5.6 Results for nitrobenzene

5.6.1 General

In order to test the GLOBOX model, sets of characterisation factors have been calculated for nitrobenzene as a test chemical (see Table 5.4 for the physico-chemical properties and the degradation rate constants of nitrobenzene). Since there are 239 countries with each up to seven possible emission compartments, 50 seas with each two possible emission compartments, and seven toxicity-related impact categories, each set of characterisation factors for a certain chemical consists of up to ((239×7)+(50×2))×7=12411 numbers. In practice, this is somewhat less, since not all compartments exist in every country, and some substances may exert no effect for some impact categories.

Table 5.4 Physico-chemical properties and degradation rate constants of nitrobenzene.

physico-chemical properties			degradation rate constants at 25° C		
parameter	unity	value	parameter	unity	value
molecular weight	g/mol	123	rate constant for degradation in air	s^{-1}	7.00E–08
vapour pressure at 25 °C	Pa	33	rate constant for photolysis in water	s^{-1}	7.74E–08
solubility at 25 °C	mg/l	1 921	rate constant for biodegradation in bulk surface water	s^{-1}	1.56E–07
melting temperature	°C	6	rate constant for biodegradation in aerobic sediment	s^{-1}	1.56E–07
octanol-water partition coefficient (K_{ow})	–	69	rate constant for biodegradation in anaerobic sediment	s^{-1}	1.60E–06
organic carbon-water partition coefficient (K_{oc})	l/kg	156	rate constant for biodegradation in bulk soil	s^{-1}	1.56E–07

Source: Huijbregts *et al.* (2000).

The emission of nitrobenzene to Dutch air was chosen as a reference emission in CF calculations. Different calculations were performed in order to give an indication of the answers to the main research questions, related to spatial differentiation:

- Does spatial differentiation cause substantial differences between LCA toxicity characterisation factors?
- Which environmental parameters are primarily responsible for the spatial variation in characterisation factors?
- How can differences between extreme values of characterisation factors be explained?
- To which extent is spatial differentiation in LCA toxicity assessment necessary with respect to the reliability of LCA outcomes?

- For which countries is spatial differentiation – instead of the use of standard EU CFs – important?

5.6.2 Differences between characterisation factors

Differences between characterisation factors for a certain impact category and a certain chemical, released to a certain compartment, can be analysed and described in many different ways. In this chapter, we have chosen for the interquartile range (IQR), in relation to the median. Abbreviations, used for the emission compartments, are given in Table 5.5.

Table 5.5 Emission compartments and abbreviations, used throughout this chapter.

emission compartment[a]	abbreviation
air	air
agricultural soil	agr
urban soil	urb
river water	riv
fresh lake water	lake
salt lake water	salt

[a] *Natural soil* and *seawater* are also possible emission compartments in the GLOBOX model, but have not been used for the calculations in this chapter.

Since we are primarily interested in CFs that apply to emissions in inhabited countries, we excluded uninhabited territories and sea areas as emission regions (but not as effect regions) from the statistical calculations in this chapter. Greenland was excluded as well, because its extreme environmental circumstances causes CFs for emissions in Greenland to be so extreme that they would fully dominate CF ranges for most impact category/emission compartment combinations, and thus create a distorted image of these ranges, while in global perspective, Greenland is quite unimportant from an emission point of view.

Total ranges, interquartile ranges and median values of the CFs for inhabited countries excluding Greenland are represented in Figure 5.1.

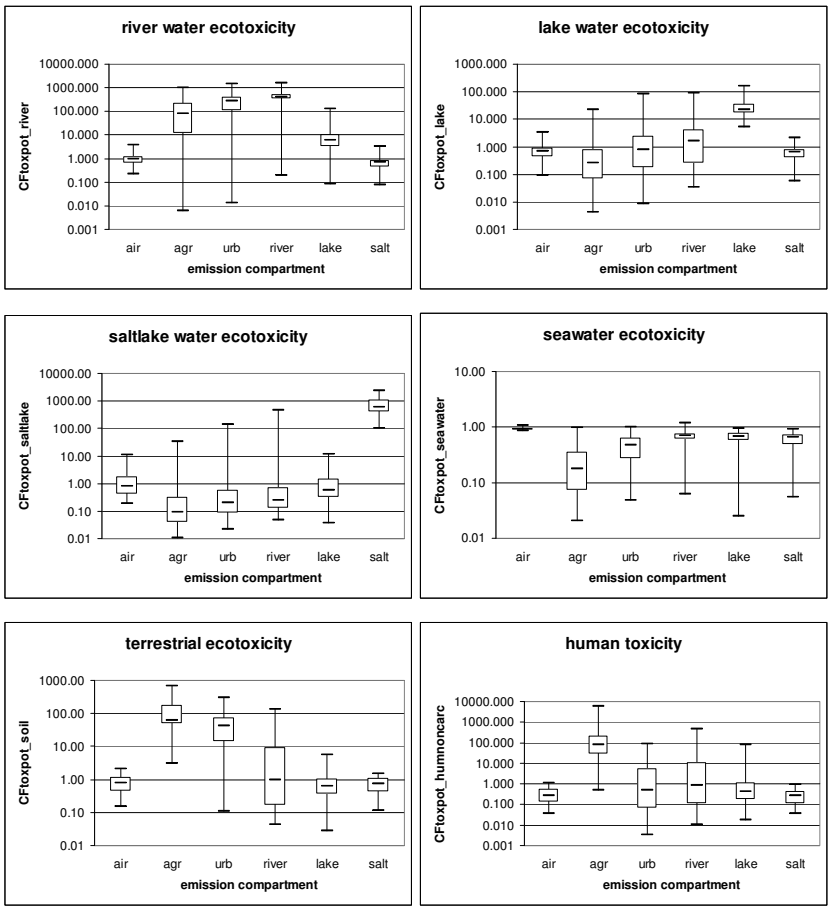

Figure 5.1 Total ranges, interquartile ranges and median values of the GLOBOX characterisation factors (potential impacts) for nitrobenzene for different impact categories and emission compartments.

Total ranges of characterisation factors turn out to vary by orders of magnitude for almost all impact categories and emission compartments. Interquartile ranges are much smaller, but may still be considerable, especially for the human-toxicity related impact categories. This implies that, at least for nitrobenzene emissions, spatial differentiation has a substantial influence on the magnitude of characterisation factors for a large number of countries.

5.6.3 Individual parameters and characterisation factors: a sensitivity analysis

Due to the complexity of the web of equations, embedded in the GLOBOX model, it is difficult to establish the relationship between individual parameters on the one hand and characterisation factors on the other directly from model equations. An analysis of the model outcomes may, however, deliver new insights into the relative importance of individual parameters for the resulting CFs. To evaluate which of the environmental parameters are primarily responsible for the large spatial variation in characterisation factors, a sensitivity analysis was performed. To this end, an alternative version of the GLOBACK parameter set was constructed, with minimal spatial variation, setting all easily adaptable parameters to (weighted) average values.* Full spatial differentiation was first compared to minimal spatial differentiation – with all easily adaptable parameters set to their average values for inhabited countries (except Greenland) and for seas. No average values could be applied to geographic parameters like *total surface area* and *border length*, or to flow parameters like *river inflow* and *river outflow*, since this would disturb the internal coherence of the geographic system and the global water balance, respectively. The total range of the characterisation factors, resulting from 'full differentiation' versus 'minimal differentiation', is represented in Figure 5.2. Relative ranges are especially large if characterisation factors are strongly dependent on emission locations, *e.g.* for river emissions in countries with salt lakes (where rivers may end up in a salt lake) versus river emissions in countries without salt lakes (which usually end up in seas). It is clear from Figure 5.2 that the influence of geographic and flow parameters alone on the range of characterisation factors is substantial for most impact category/emission compartment combinations. The additional range of the fully differentiated version reflects the influence of all other parameters together. Since each individual parameter may cause either an increase or a decrease of the characterisation factor, influences may (partly) balance each other. This implies that the difference in range between *full differentiation* and *minimal differentiation* is not automatically indicative for the influence of individual parameters. A large difference indicates that there should be at least one additional parameter that has a significant influence on the range of the characterisation factor. A small difference indicates either that geographic and flow parameters are dominant or that other influential parameters compensate each other's impact on the range of the characterisation factor.

* Some parameters had to be slightly adapted in order to have the switch-off of spatial differentiation make sense, *e.g.* in the case of population: equal population densities – not equal populations as such – were applied for the different countries.

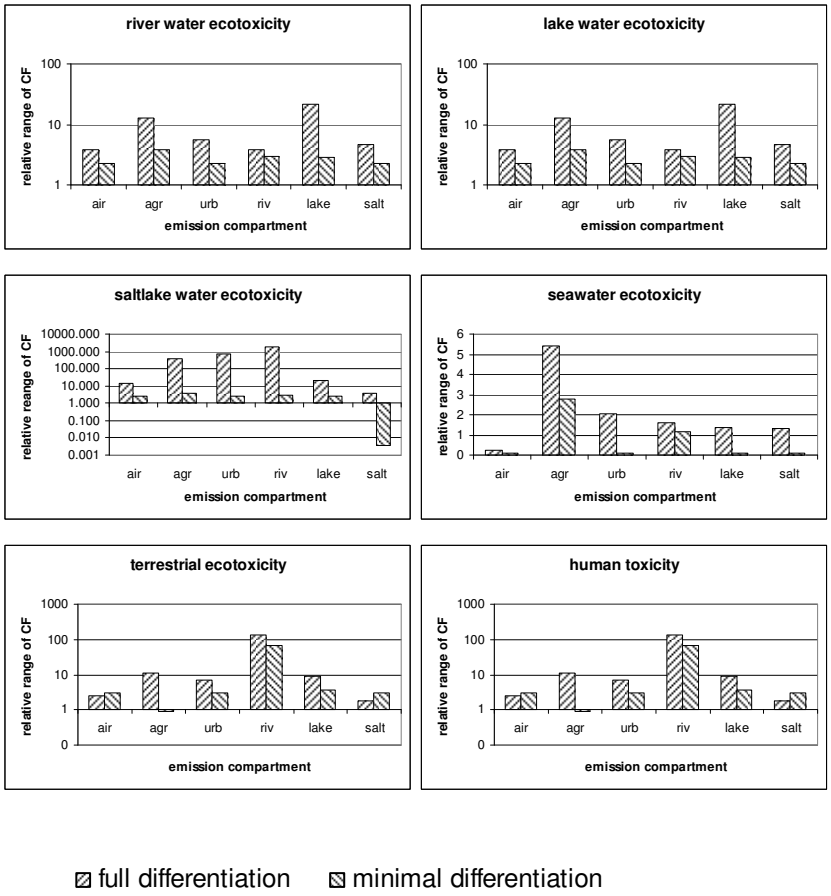

Figure 5.2 Relative range (range divided by mean) of GLOBOX characterisation factors for nitrobenzene, resulting from 'full differentiation' versus 'minimal differentiation'

To evaluate the influence of each single parameter, the entire set of characterisation factors was recalculated for 15 additional variant versions of the GLOBACK parameter set. For each set, minimal spatial variation was applied to all parameters or parameter categories but one. The differences in outcome between the version with minimal spatial variation and each of the variants with one spatially differentiated parameter indicate the relative contribution of this parameter to the total influence of spatial differentiation on the values and ranges of the characterisation factors. In Table 5.6, the parameters that cause a deviation of more than 25% from the median or the interquartile range of the variant with minimal spatial differentiation are indicated for each impact category and emission compartment.

Table 5.6 Parameters that cause a deviation of more than 25% from median nitrobenzene CFs (normal font), inter quartile range (italics) or both (bold), compared to the situation with minimal differentiation.

> 25% influence of parameter differentiation on CF for emission compartment mentioned[1]						
parameter	aquatic ecotoxicity (river)	aquatic ecotoxicity (lake)	aquatic ecotoxicity (salt lake)	aquatic ecotoxicity (seawater)	terrestrial ecotoxicity	human toxicity[5]
area division nat/agr/urb	–	–	–	agr	urb	urb
lake parameters[3]	*riv*, salt	air, agr, urb, riv, lake, *salt*	air, agr, urb, *riv*, *lake*, salt	lake, *salt*	lake, salt	urb, riv, lake, salt
river volume	*air*, *agr*, *urb*, riv, lake, *salt*	*riv*, *urb*	*riv*, *urb*	*riv*	riv	riv
ice cover	–	*lake*	–	*lake*	–	–
area sea sediment	–	–	–	–	–	–
geophysical parameters[4]	–	–	–	–	–	–
temperature	agr, *lake*	agr, urb, *riv*, *lake*	agr, urb, *salt*	*air*, lake, *salt*	agr, *urb*, lake	agr, urb, lake
wind speed	–	*lake*	*salt*	*lake*	–	–
rain rate	–	agr, *lake*	*agr*, *riv*		*air*, *lake*, *salt*	*air*, *salt*
population (density)	n.a.	n.a.	n.a.	n.a.	n.a.	agr, *urb*, *riv*, lake
mean body weight	n.a.	n.a.	n.a.	n.a.	n.a.	–
fraction of population with access to safe drinking water	n.a.	n.a.	n.a.	n.a.	n.a.	–
fraction of drinking water that is groundwater	n.a.	n.a.	n.a.	n.a.	n.a.	–
dietary intake	n.a.	n.a.	n.a.	n.a.	n.a.	air, *agr*, riv, salt
food import/export	n.a.	n.a.	n.a.	n.a.	n.a.	–
sea fish catch per sea	n.a.	n.a.	n.a.	n.a.	n.a.	–

[1] n.a.=not applicable.

[2] Variability of the relative coverage of the area by natural (nat), agricultural (agr) and urban (urb) soil respectively.

[3] Area, depth, mixing depth and sediment area of both freshwater lakes and salt lakes.

[4] Mixing depth of non-agricultural soil compartments, volume fraction of particles in inland water and soil runoff, volume fraction air and water in agricultural and non-agricultural soil compartments, organic carbon content of soil, suspended matter and sediment, deposition rate of aerosols, concentration of hydroxyl radicals in air.

[5] Non-carcinogenic and carcinogenic are taken together here, because of the reference substance nitrobenzene, these impact categories have equal CFs.

It can be concluded from Table 5.6 that most of the parameters that have been differentiated in the GLOBACK parameter set have an important influence on the characterisation factors of nitrobenzene for a number of impact category/emission compartment combinations. For nitrobenzene, spatial differentiation seems to be

more important for water and soil emissions than for emissions to air, which may be explained by the fact that air compartments are characterised by a relatively high mixing rate, resulting in a relatively fast convergence of environmental concentrations in air compartments. With respect to human intake, the differentiation of population and dietary intake turns out to play a crucial role.

5.6.4 Individual parameters and characterisation factors: correlations

From the sensitivity analysis elaborated above, a number of important combinations of model parameters and characterisation factors have been selected for correlation analysis. Two types of correlation coefficients have been determined: the Pearson correlation coefficient *r*, indicating the degree of correspondence to a linear function, and the Spearman rank order correlation coefficient, indicating the degree of monotony with respect to rise or descent as such. In Table 5.7, both types of correlation coefficients are reported for the selected parameters and characterisation factors.

Table 5.7 Correlation coefficients for linear correlation (Pearson (*r*)) and rank correlation (Spearman) between model parameters and nitrobenzene CFs for a selection of parameters and CFs.

parameter	CF	emission compartment	Pearson corr. coeff. (*r*)	Spearman rank order corr. coeff.	*N*
relative lake area	aquatic ecotoxicity (lake)	air	0.33	0.35	231
relative lake volume	aquatic ecotoxicity (lake)	air	0.37	0.38	231
relative salt lake area	aquatic ecotoxicity (salt lake)	air	0.55	0.37	231
relative alt lake volume	aquatic ecotoxicity (salt lake)	air	0.42	0.37	231
rain rate	terrestrial ecotoxicity	air	-0.61	-0.62	231
leaf crop consumption	human toxicity	air	0.24	0.27	231
temperature	aquatic ecotoxicity (lake)	agr	-0.38	-0.53	216
temperature	terrestrial ecotoxicity	agr	-0.73	-0.73	216
temperature	human toxicity	agr	-0.11	-0.37	216
population density	human toxicity	agr	0.59	0.65	216
relative urban area	terrestrial ecotoxicity	urb	0.33	0.44	223
relative river volume	aquatic ecotoxicity (river)	riv	0.37	0.42	223
freshwater fish consumption	human toxicity	riv	-0.02	0.20	223
frost period	aquatic ecotoxicity (lake)	lake	0.45	0.38	182
relative lake area	human toxicity	lake	-0.09	-0.40	182
relative lake volume	human toxicity	lake	-0.04	-0.46	182
wind speed	aquatic ecotoxicity (lake)	lake	-0.12	-0.21	182
relative salt lake area	aquatic ecotoxicity (salt lake)	salt	0.32	0.25	33

parameter	CF	emission compartment	Pearson corr. coeff. (r)	Spearman rank order corr. coeff.	N
relative salt lake volume	aquatic ecotoxicity (salt lake)	salt	0.63	0.63	33

It is clear from Table 5.7 that large correlations exist between most of the selected parameters and the corresponding characterisation factors. From the differences between Pearson and Spearman correlation coefficients it appears that the relationship between parameter and characterisation factor is probably non-linear in at least a number of cases. The strongest relationship ($r = -0.73$), exists between temperature on the one hand and the characterisation factor for terrestrial ecotoxicity in case of emission to agricultural soil on the other, at least for nitrobenzene.

5.6.5 Explanation of the influence of individual parameters

The influence of individual parameters on characterisation factors can be explained in a qualitative way on the basis of the general properties of multimedia distribution and human exposure. The GLOBOX model contains a number of possibilities for a quantitative check of the resulting suppositions. Model outcomes for pairs of countries with distinct values for certain parameters will be used to illustrate the explanations, elaborated in this paragraph. In Table 5.8, an overview of a number of key parameters is given for these countries.

Table 5.8 Key parameters for a selection of countries.

	Afghanistan	New Zealand	Mongolia	Mali
total area [km^2]	647 500	268 680	1 565 000	1 240 000
length of coastline [km]	0	15 134	0	0
area salt lakes [km^2]	2 020	0	5 098	0
average temperature in capital [°C]	12	14	-2	28
rain rate [mm/year]	327	1 732	241	282
river volume [km^3]	1.6	9.2	0.8	1.6
population density [inhab./km^2]	44	15	2	10
crop consumption per capita [g/day]	434	676	331	463
meat consumption per capita [g/day]	18	146	130	30

The fact that geographic and river flow parameters have a large influence on characterisation factors can be explained by the distribution of released substances between land and sea regions. Chemicals that are released on islands, on peninsulas and in areas with a comparatively long coastline will usually spend a relatively large part of their lifetime in sea areas, where seawater aquatic ecotoxicity is the only toxicity related impact category to which they contribute. This influence will be largest for emissions to isolated areas, areas with a high ratio between coastline length and surface area, and areas with a relatively large river outflow to the sea. In

Figure 5.3, the global distribution over land and sea areas of nitrobenzene is represented for emissions to air and agricultural soil in Afghanistan and New Zealand, respectively. Afghanistan and New Zealand are countries with comparable environmental temperatures. Afghanistan is fully enclosed by other countries, while New Zealand is fully surrounded by sea. Total amounts of nitrobenzene residing in the environment hardly differ between comparable emissions to both countries, but, although a remarkable part of air emission ends up in sea areas even for Afghanistan, the distribution ratio between land and sea areas turns out to be quite different.

a. emission to air

b. emission to agricultural soil

Figure 5.3 Distribution of nitrobenzene over land and sea areas for emissions in Afghanistan and New Zealand.

A decrease in environmental temperature is associated with lower volatility values – which can lead to 'cold condensation' (Wania and Mackay, 1993) – and with decreased environmental degradation rates. In Figure 5.4, the distribution of nitro-

benzene over the different environmental compartments after emissions to air and agricultural soil, respectively, is shown for Mali and Mongolia: two landlocked countries with comparable rain rates but distinctly different average environmental temperatures.

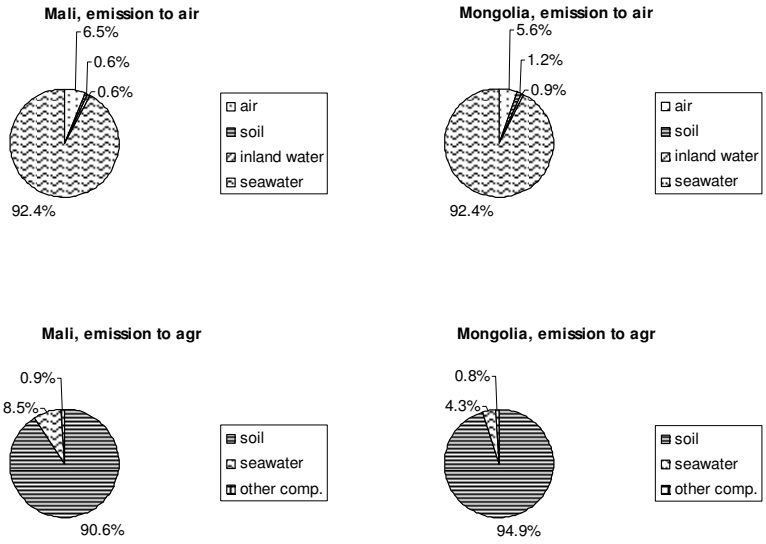

Figure 5.4 Distribution of nitrobenzene over different environmental compartments for emissions to air and agricultural soil in Mali and Mongolia.

If nitrobenzene is released to air, whether this happens in Mali or in Mongolia, more than 90% turns out to end up in seawater, despite the fact that both countries are landlocked. Total environmental amounts are almost equal: 9266 and 9258 t in the steady state situation for emissions of 1 kg/s nitrobenzene to air in Mali and Mongolia, respectively. The relative amounts, present in inland water and soil, are larger in Mongolia by factors 2 (1.2% versus 0.6%) and 1.5 (0.9% versus 0.6%), respectively. This may indicate either cold condensation or delayed degradation in these two compartments occurs in cold regions – or both. In case of emission to agricultural soil, the bulk part of the total environmental amount turns out to remain in the soil compartment, with a slightly larger part in Mongolia. The relative amount, ending up in seawater, is a factor 2 higher for Mali than for Mongolia. Remarkably, total environmental amounts differ by more than an order of magnitude: 3028 and 36692 t for emissions of 1 kg/s nitrobenzene to agricultural soil in Mali and Mongolia, respectively. This forms a strong indication for a substantial

influence of the temperature-dependent increase of soil residence times on accumulation of chemicals in cold regions – at least for nitrobenzene.

According to the data in Table 5.6, variations with respect to drinking water purification (fraction of population with access to safe drinking water) have hardly any influence on characterisation factors for human toxicity. Such a lack of influence of a certain parameter can have several reasons:

- the parameter plays a subordinate role in the model, compared to other parameters
- parameter variations between regions are relatively small
- the parameter refers to an exposure route that is relatively unimportant for the chemical under study.

In Figure 5.5, the relative magnitudes of human exposure routes are represented for the case of an emission to Dutch air, leading to exposure in all inhabited countries.

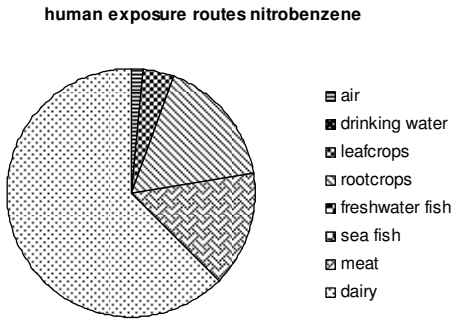

Figure 5.5 Relative magnitude of human exposure routes to nitrobenzene for Dutch air emissions.

Drinking water turns out to be only a minor exposure route for nitrobenzene. For other chemicals, this may be different, and the parameter that represents drinking water purification may be more important.

5.6.6 GDP weighting

From the analyses above, it is obvious that regional differentiation can have a large influence on the magnitude of characterisation factors. It is not yet clear, however, how important these differences are in the context of general LCA practice. The regions distinguished in the GLOBACK parameter set are very unequal, not only with respect to the magnitudes of areas and populations, but also regarding economic productivity, to which product life cycles are directly connected. In the

context of LCA, it is important to know whether the regions for which characterisation factors deviate substantially from 'standard' characterisation factors represent a significant part of the global economic system, resulting in a relatively large share of regional processes in global product life cycles. The relative importance of different countries with respect to their CFs has therefore been approached by the application of weighting with their economic productivity in terms of their GDP (CIA, 2009), arguing that GDP is a good measure for a country's influence on the world market, and thus of its contribution to global economic productivity reflected by summed product life cycles.

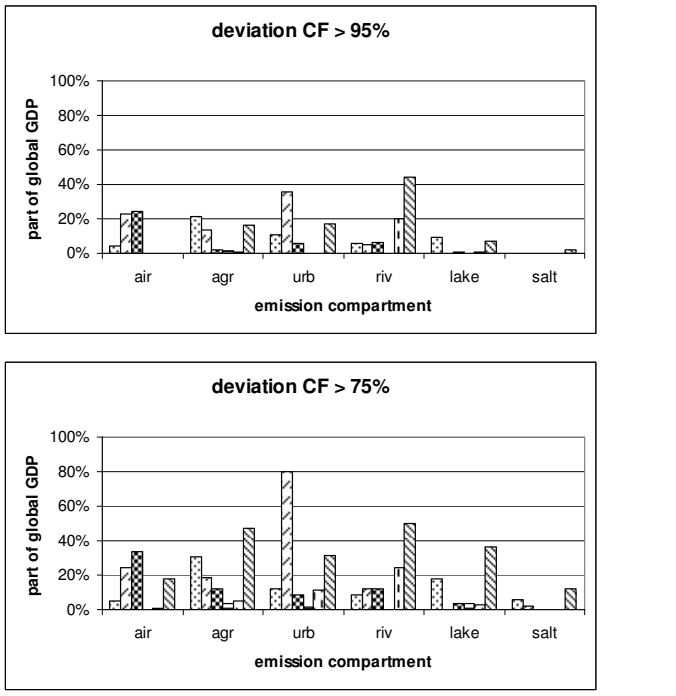

Figure 5.6 Part of the global GDP, produced in countries with a nitrobenzene CF that deviates more than 75 and 95 percent respectively from the European GDP-weighted mean nitrobenzene CF for different impact categories and emission compartments.

To test CFs for the relative importance of spatial deviation, GDP-weighted average values for the EU-27 were constructed for all characterisation factors, to act as European standard CFs. Subsequently, the countries for which individual CFs deviated more than 75% from the European standard CF were grouped, their GDPs were added, and the total GDP of these countries together was compared to the total world GDP. The same procedure was applied to countries with CFs that deviated more than 95% from the European standard CF. The results of this analysis are shown in Figure 5.6.

Figure 5.6 shows that regions with characterisation factors that deviate substantially from European standard CF values represent a substantial part of global GDP for several impact category/emission compartment combinations. This implies that the use of spatially differentiated characterisation factors may bring about an important refinement in the assessment, not just for processes that take place in small, 'exotic' countries, but for a substantial part of the global economic production.

5.7 Discussion

5.7.1 The influence of spatial differentiation

The main goal of the GLOBOX model is to increase the accuracy of LCA toxicity characterisation factors by accounting for spatial environmental differences. In a general sense, it has been clearly shown by the calculations on the test chemical nitrobenzene that the application of spatial differentiation can make a large difference between characterisation factors for different countries, especially for emissions to water and soil – with ranges of up to 5 orders of magnitude between different countries for nitrobenzene. Extremely low CF values are found for small, geographically isolated islands, such as Fiji. Extremely high CF values are found for countries for which emissions affect relatively large shares of certain ecosystems or large human populations, especially in combination with relatively high residence times in the country itself or in surrounding areas with equal environmental circumstances. A number of examples, referring to atmospheric emissions, are:

- The top-3 countries with respect to the CF for aquatic ecotoxicity for rivers are Colombia, Bolivia and Peru, all countries where the Amazon rises.
- The top-3 countries with respect to the CF for human toxicity are the Czech Republic, Slovakia and Poland, all centrally positioned in densely populated Europe.
- The top-3 countries with respect to the CF for aquatic ecotoxicity for salt lakes are Kyrgyzstan, Uzbekistan and Tajikistan, all countries of which river flows end up in the Aral Sea.

In general, the highest characterisation factors indicate which regions on earth are the most vulnerable to pollution with respect to a certain environmental impact. Important parameters with respect to spatial differentiation include geographic position, lake parameters (area, volume, and water residence time), environmental temperature, and rain rate. For human toxicity, population density and dietary intake play a major role as well.

The choice to differentiate at the level of countries and seas has advantages with respect to data availability, but a drawback of this choice is the fact that the regions distinguished are very different in size. Large countries like the United States, Canada, Russia and China, which are characterised by a wide variation in environmental parameters, need a refinement of this approach to decrease uncertainty in their CFs. Such refinement would be a desirable next step in the further development of the GLOBOX model. A further refinement could consist of making a distinction between urban and rural areas, which, according to other publications, could make a difference in human intake of a factor 3 (Huijbregts *et al.*, 2005) up to more than an order of magnitude (Humbert *et al.*, 2009).

The question can be posed whether the deviation of spatially differentiated characterisation factors (CF_i) from a reference value – *e.g.* a non-differentiated alternative characterisation factor (CF_{ref}) – should be expressed in terms of orders of magnitude deviation

$$\frac{CF_i}{CF_{ref}}$$

or in terms of percentage difference

$$\frac{CF_i - CF_{ref}}{CF_{ref}} \times 100\%$$

Since the difference in terms of percentage gives a better and more symmetrical reflection of the relative under- or overestimation of effects in absolute terms, this latter form has been chosen as the best option for this specific purpose, with deviations of 75 and 95 percent of the GDP-weighted average CF as a measure. For other purposes, such as the indication of ranges of CF values, orders of magnitude have been used.

The use of spatial differentiation makes sense if it results in a fairly large overlap of ranges of characterisation factors between different chemicals. An overlap in range between two chemicals indicates that the answer to the question which of the chemicals should be preferred in case of equal emissions is region-dependent. Since the environmental profiles of product alternatives are often dominated by a relatively small number of chemicals, an error with respect to the question which chemicals are to be preferred can potentially reverse the outcomes of comparative

LCA studies. In these cases, spatial differentiation is essential for the reliability of LCA results, in terms of the ranking of product alternatives. A small overlap indicates that a reverse ranking applies to extremes only, *e.g.* 'chemical A is preferred to chemical B, except for the situation that chemical A is released in Iceland and chemical B in Bolivia'. The larger the overlap, the more likely rank reversal will be. This can be illustrated by comparing the ranges of non-differentiated CFs for different chemicals to the ranges of spatially differentiated CFs for one chemical. The USES-LCA 2.0 spreadsheet program offers the possibility to calculate midpoint and endpoint characterisation factors for a number of different toxicity-related impact categories for 3396 chemicals (Van Zelm *et al.*, 2009). In Table 5.9, the ranges of a number of selected series of midpoint CFs, calculated with USES-LCA 2.0, are given.* In the third and fourth column, full CF ranges are compared to the ratio between the third and the first quartile of these CFs (Q_3/Q_1), spanning the central 50% of chemicals.

Table 5.9 Ranges of midpoint CFs in USES-LCA 2.0 (3396 chemicals) and GLOBOX (nitrobenzene).

		USES-LCA (3396 chem.)	USES-LCA (3396 chem.)	GLOBOX (nitrobenzene)
impact category	emission compartment	full range CFs	range CFs of central 50% (Q_3/Q_1)	range CFs of 50% with highest GDP[a]
terrestrial ecotoxicity	air	9.78×10^{14}	279	11
human toxicity	air	5.71×10^{10}	69	21
freshwater ecotoxicity	freshwater/lake	2.85×10^{12}	83	25
freshwater ecotoxicity	freshwater/river	2.85×10^{12}	83	86
soil ecotoxicity	agricultural soil	1.41×10^{14}	44	37
human toxicity	agricultural soil	8.01×10^{46}	72	3 815

[a] These countries are considered to cover the main part of world economic production, and thus, their CFs are considered to be of relative importance to LCA.

While the full range of the these series of characterisation factors spans between 10 and 46 orders of magnitude, the ratios between the third and the first quartiles (Q_3/Q_1) vary between 44 and 279, indicating that the characterisation factors for half the number of chemicals lie together within ranges of just one or two orders of magnitude, depending on impact category and emission compartment. In the fifth column, the ranges of spatially differentiated GLOBOX-CFs that represent

* Scenario options chosen: fate and exposure: emission compartments: air, freshwater and agricultural soil (respectively); time horizon: steady state; bioaccumulation metals: yes; include ocean for essential metals: yes; human effect and damage: effect endpoint: DALY; carcinogenic substances: all; ecological effect: method freshwater ecological effect: linear; minimum nr of tested species: 1.

the 50% of the countries with the highest GDP values (together representing 99% of the global GDP) are given. These factors vary between 11 and 3815. This means that for the central group of chemicals with CF values between Q_1 and Q_3, spatial characteristics have an influence on the CF that is comparable to the influence of chemical properties or larger, at least for these selected combinations of impact categories and emission compartments.

It can be concluded from this example that spatial differentiation is of large importance. The differences between spatially differentiated CFs for nitrobenzene of up to 5 orders of magnitude, as found in this study, are in good accordance with the findings of other authors, who report differences of up to 1 or 2 orders of magnitude for differentiation at the level of continents (Huijbregts *et al.*, 2003 for fate and exposure factors; Rochat *et al.*, 2006 for intake fractions), differences of up to around 3, 5 or 8 orders of magnitude between smaller regions within continents (MacLeod *et al.*, 2004; Pennington *et al.*, 2005; Humbert *et al.*, 2009; all for intake fractions), and a noticeable increase of differences with increasing level of spatial model resolution (Manneh *et al.*, 2009 for intake fractions).

Besides spatial model resolution, the range of calculated model results is also influenced by internal model properties:

The parameters to which spatial differentiation has been applied
In the GLOBOX model, spatial differentiation has been applied not only to fate parameters, population and food production areas, but also to intake parameters concerning dietary pattern, body weight, fraction of drinking water that has been purified, and fraction of drinking water that is ground- and surface water, respectively. Especially dietary patterns turned out to have a marked influence on CFs.

The inclusion of spatially differing parameters in the model equations
Model equations were largely taken from the EUSES 2.0 model, which, in contrast to the earlier model version EUSES 1.0, included temperature-dependency of environmental degradation rates. The relatively large influence of temperature variations on CFs found in this study may be largely due to this model property, which stresses the importance of model equations to adequately reflect possible spatial dependencies.

The number of environmental compartments distinguished
The number of aquatic environmental compartments in the GLOBOX model is larger than in most multimedia models. Rivers, freshwater lakes, salt lakes and groundwater are distinguished as separate compartments. Regarding the differences in characterisation factors of more than an order of magnitude on average between freshwater lake CFs on the one hand and salt lake and river water CFs on the other (for emissions to the respective compartments concerned), it can be concluded that a separate distinction of these compartments does make sense.

Hollander (2009), concludes from a European study that substance properties are more influential than environmental properties with respect to environmental concentrations. This author bases her conclusion on full concentration ranges. As mentioned above, however, there exists a large discrepancy between the full range (10 to 46 orders of magnitude) and the range of the central 50% Q_3/Q_1 (one or two orders of magnitude) of USES-LCA characterisation factors for the given combinations of impact category and emission compartment. While the full concentration ranges, found by Hollander (2009) may be largely due to chemical properties, it is very well possible that at the same time, the variation of the central 50% of these concentrations display a much smaller range as well. In other words: even though chemical properties may be more influential than environmental properties on average, it is very well possible that for a substantial number of chemicals, spatial characteristics of environmental properties have a comparable or larger influence on environmental concentrations than chemical properties.

More chemicals should be tested with the GLOBOX model in order to obtain a detailed image of the general influence of spatial differentiation on the range of characterisation factors. Yet, it can be concluded from the calculations with nitrobenzene that spatial variation in environmental and human exposure parameters on toxicity-related LCA characterisation factors can be substantial. Summarising, the following findings on nitrobenzene can be reported:

- Characterisation factors for nitrobenzene vary up to 5 orders of magnitude – with corresponding interquartile ranges up to 2 orders of magnitude – due to spatial differentiation.
- Geographic and flow parameters alone cause characterisation factors to vary up to 3 orders of magnitude.
- Environmental parameters or parameter categories that cause the characterisation factor for one or more impact category/emission compartment combinations to deviate more than 25 percent from the median value of the interquartile range include area division natural/agricultural/urban soil, lake parameters, river volume, ice cover, temperature, wind speed and rain rate. Intake parameters that cause human toxicity characterisation factors to show a similar deviation include population and dietary intake. Most of these parameters can be demonstrated to have a sound correlation with at least one impact category/emission compartment combination, either with the Pearson correlation coefficient (r) or with the Spearman rank order correlation coefficient, or with both.
- The distribution of nitrobenzene between land and sea areas after emission seems to depend largely on the geographic location of the country of emission, with emissions in landlocked countries tending to reside for a comparatively large part in land areas, especially after emission to soil.

- The environmental temperature seems to have an important influence on the distribution of nitrobenzene between the different compartments, with lower temperatures being in favour of lower air concentrations and higher concentrations in water and soil. This may be largely explained by the seemingly decisive influence of environmental temperature on soil residence times.
- Lack of spatial differentiation causes characterisation factors for nitrobenzene to be more than 95 percent wrong for countries that together represent up to 40 percent of world GDP, and for more than 75 percent wrong for countries representing up to 80 percent of world GDP, compared to GDP-weighted average EU characterisation factors.
- The range of the spatially differentiated characterisation factors for nitrobenzene has the same order of magnitude as the range of the central 50% of USES-LCA midpoint characterisation factors for 3396 chemicals (Van Zelm *et al.*, 2009) for at least a number of impact category/emission compartment combinations.

From these findings, it can be concluded that at least for nitrobenzene, spatial differentiation has an important influence on toxicity-related LCA characterisation factors, that a fair number of environmental and human intake parameters are involved, and that lack of spatial differentiation may cause rank reversal and thus erroneous outcomes of LCA characterisation.

5.7.2 Practical application of GLOBOX in life cycle impact assessment

For LCA practice, spatial differentiation has two important consequences: in the first place, the use of spatially differentiated characterisation factors requires region-specific emission data. And in the second place, spatial differentiation on this scale results in a dramatic increase in the number of characterisation factors. This increase is proportional to the number of regions distinguished in the characterisation model – 289 in the case of GLOBOX. Distinguishing between actual and potential impacts will cause a further increase by a factor 2. The requirement of region-specific emission data refers to the process data in the LCA inventory. Existing LCA databases may have to be adapted by the spatial specification of emissions. The format of the inventory table will have to be supplemented with the emission location. The current EcoSpold format that is used for the ecoinvent data, for instance, allows for a geographical tag to processes and to products, but not to emissions, and would thus have to be adapted. A summation of emissions that take place in different regions will no longer be appropriate. As a consequence, the inventory table will largely increase in size. Despite the increases in the number of inventory data and characterisation factors, spatial differentiation has no influence on the eventual number of category indicator results, since for each

impact category, all region-specific outcomes are summed to one overall impact score.

5.7.3 Broader application

Although the GLOBOX model has been primarily developed for the calculation of LCA toxicity characterisation factors, it is basically a multimedia model for fate, exposure and effect modelling, with a much broader range of application than just LCA. One of the possible applications is the analysis of distribution and exposure patterns, resulting from the emission profile of a certain chemical in a certain region. Different substances with specific chemical properties can thus be compared with respect to their distribution patterns. Distribution can be determined over different countries or over different environmental compartments. This type of analysis can for instance be used to calculate the distribution patterns of POPs on a global scale. With respect to cold areas, the findings about large differences in environmental amounts of nitrobenzene, following soil emissions to respectively Mali and Mongolia, are in good accordance with the global fractionation theory on accumulation of POPs in cold regions (Wania and Mackay, 1993), which is also confirmed by the findings of Toose *et al.* (2004). Another type of application is for supporting scenario studies, in which an entire emission profile for one or more regions is an intermediate result. For instance, the EXIOPOL project (Tukker *et al.*, 2009) provides a multi-region environmentally extended input-output table on the level of (more than forty) countries, easily connectible to the GLOBOX model when there is a need to move from the substance level to the impact level.

5.7.4 Future outlook

The current GLOBOX model is a steady state multimedia model. For the calculation of LCA characterisation factors, this is sufficient (Heijungs, 1995). A drawback of steady state models is, however, that they are more difficult to validate, since it requires well-documented steady state situations, caused by emissions of globally distributed chemicals. Developing a dynamic variant of the GLOBOX model would open up better possibilities for model validation, and is therefore considered as a desirable next step. This step would also broaden the applicability of the GLOBOX model for further environmental analyses of the global distribution patterns of chemicals. Yet, the main goal of the GLOBOX model remains the calculation of spatially differentiated LCA toxicity characterisation factors on a global scale. A logical next step would be the extension of the GLOBOX model with other impact categories such as global warming and acidification, including the specific environmental processes involved, and thus the creation of an integrated system for global spatial differentiation in life cycle impact assessment.

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Appendix: Supplementary data

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