Ecotoxicity

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The figures, see Fig. 8.2 and 8.4, should be printed in black and white

Chapter 8
Ecotoxicity

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Abstract
Ecotoxicity impact assessment of chemicals in life cycle assessment (LCA) adheres to a number of underlying principles and boundary conditions: 1) a large number of emitted substances to cover (at least 100,000 potentially relevant elementary flows with current models covering around 2500), 2) linearity of characterisation models, 3) conservation of mass and mass balance, 4) infinite time horizon, 5) additivity of toxicity, 6) assuming average conditions as best estimates to avoid bias in the comparison (including consideration of generic/average ecosystems and impacts). The cause-effect mechanism for ecotoxicity impacts of chemicals can be divided into four parts: 1) chemical fate (i.e. chemical behaviour/distribution in the environment), 2) exposure (i.e. bioavailability), 3) effects (i.e. affected species), and 4) severity (i.e. disappeared species). In terms of species represented, a freshwater ecosystem is described in this chapter by three trophic levels: 1) primary producers (e.g. algae), 2) primary consumers (i.e. invertebrates), and 3) secondary consumers (e.g. fish). Model uncertainty was estimated at about three orders of magnitude on top of important sources of parameter uncertainty such as degradation rates and effect factors. Current

midpoint LCIA methodologies covering ecotoxicity include TRACI 2.0, and the ILCD recommended methodology, both employing the USEtox factors. Current LCIA methodologies covering midpoint and endpoint characterisation are ReCiPe, LIME, IMPACT 2002+, and IMPACT World+. Important research needs are 1) increasing substance coverage, 2) further developing marine and terrestrial ecotoxicity modelling for midpoint, 3) improving endpoint modelling for ecotoxicity towards biodiversity, 4) consideration of long-term emissions and impacts of metals, 5) importance of spatial and temporal variability, 6) mixture toxicity, and 7) decreasing model and parameter uncertainty.

Keywords
Chemical fate, comparative ecotoxicity, concentration-response relationship, ecosystem, LCA, LCIA, life cycle assessment, life cycle impact assessment, potentially affected fraction of species (PAF)

1 Principles, fundamentals, and recommended practice of characterisation modelling

In the early days of LCA, impact assessment was generally considered as unfeasible and only with the development of the first LCIA methodologies did this perception slowly start to change. Ecotoxicity is among those impact categories that, only within the last decade, started to be considered as becoming mature enough for application, although with considerable reservations by many practitioners until today. The origin for today's approaches in characterising this impact category is clearly in the field of environmental hazard and risk assessment (Pennington et al. 2006).

Environmental Risk Assessment (ERA) quantifies risks due to environmental changes (e.g. a chemical emission into an environmental compartment) affecting biological systems (e.g. animals, plants, or entire ecosystems). This risk depends on the toxicity of the substance and the level of exposure of living organisms. The toxicity is generally estimated based on toxicological tests relating adverse effects to concentrations of a substance, so called concentration-effect relationships. The level of exposure can be measured, or it can be estimated by modelling the fate of the substance from the emission to the relevant environmental media, resulting in environmental concentrations.
While LCA and ERA both aim at the evaluation of potentially toxic impacts on the environment, partly using common data and assumptions, there are also important differences between them (Pennington et al. 2006, Udo de Haes et al. 2002, Olsen et al. 2001, Barnthouse et al. 1997, Owens 1997). Some of those are:

1) ERA usually applies conservative estimates of toxicity and several other properties of a compound (e.g. biodegradability) and realistic conservative (worst-case) scenarios for the modelled environmental system while LCIA aims at best estimates for all parameters for a comparative assessment.
2) ERA is generally performed in a regulatory context ensuring that an emission at a given site poses no risk to the protection targets. LCIA aims to address all relevant environmental impacts anywhere in the world due to a product or service while currently not necessarily considering time and localisation of the emissions (Hauschild and Pennington 2003).
3) ERA only considers the potential impact, in terms of risk, of one compound or mixture on the environment. LCIA by definition assesses several, sometimes large sets of chemicals and has to ensure the compatibility of the toxicity impact indicator for each chemical and with indicators for other impact categories.
4) LCA considers impacts integrated over time and space at the ecosystem level, while regulatory ERA typically focuses on peak exposures to individual (most sensitive) species.

As opposed to ERA where actual risks are calculated, comparative assessments aim to estimate the impact of a chemical relative to other substances, typically represented by rankings of chemicals by a certain indicator, e.g. toxicity and/or persistence in the environment. These rankings are then used as the basis for decisions, e.g. regarding choices of chemicals as product compounds with the least environmental impact, or in the context of chemical policy identifying priority substances for regulation, etc. In ERA acceptable exposure levels in terms of regulatory thresholds are used to e.g. evaluate an emission or the acceptability of an industrial installation. It thus estimates the potential impact of a compound on ecosystem stability for a specific part of the life cycle and at a local scale, yielding results

which are not necessarily comparable across different release sites or chemicals. Thus, risk assessment addresses different objectives, spatial scales and process chains.
Several important principles are common practice and required when developing a method for ecotoxicity impact assessment of chemicals in the framework of LCA:

- Large number of relevant substances emitted: Accounting for the often large number of potentially toxic elementary flows in a life cycle inventory requires coverage of a large number of substances in terms of available characterisation factors. For ecotoxicity this may range to hundreds of thousands potentially emitted substances.
- Linearity: As life cycle inventory (LCI) data are typically not spatially and/or temporally differentiated, integration of the impact over time and space is required. In ecotoxicity modelling in LCIA this leads to the use of characterisation models assuming steady-state conditions (i.e. no change in the relative distribution of a chemical between all environmental compartments over time), which implies a linear relationship between the increase in chemical emission into the environment and the consequent increase in chemical concentration in each environmental compartment. The effect model itself also assumes linearity (either for the full range of exposure or via different slopes for different ranges of exposure when considering marginal changes) between an increase in chemical exposure of an organism or ecosystem and observed toxic effects.
- Conservation of mass and mass balance: Mass cannot be created or disappear, it can only be transferred. Following this principle, the transport and transformation of a substance in the environment is modelled assuming that mass is conserved at all times.
- Time horizons: As another consequence of the steady-state assumption, most current ecotoxicity characterisation models essentially account for all potential ecotoxicity impacts, independently of their time and place of occurrence in the short or long-term future, which equals integration of the mass-balance differential equation system over infinity. A few exceptions exist, as some methods (e.g. ReCiPe and IMPACT World+), allow considering defined time horizons for metals.
- Additivity of toxicity: Current ecotoxicity models only characterise single substances, generally assuming that the toxicity of each substance can be added together. With three hypotheses possible, the toxicity of substances in mixtures may be 1) additive (i.e. response additive if independent toxicity mechanism, or concentration additive for chemicals with the same toxicity mechanism), 2) synergistic, or 3) antagonistic. Most likely, all three situations may occur to varying degrees, depending on which substances in which proportions are in the mixture and virtually endless combinations of substances are possible. Therefore, current research has no clear answer, and additive toxicity is commonly assumed in generic situations.
- Best estimates: A fundamental value choice in LCA is not to be conservative or protective, but to focus on avoiding any bias between compared scenarios by assuming average conditions, also referred to as best estimates. Products or services assessed in LCA are typically not representing one specific example (e.g. with a serial number or from a specific date), but an average, normally disregarding whether a specific life cycle process took place in summer or winter, during the day or night, etc. As discussed by Pennington et al. (2004b), LCA is a comparative assessment methodology. Direct adoption of regulatory methodology and data is often not appropriate. A conservative estimate of the ecotoxic effect of a substance is unwanted in a relative comparison. Best-estimates are desirable in LCA, with the need to account for uncertainties when making distinction amongst results. Furthermore, LCA is mostly used to compare competitive products (including services) for the same function. Avoiding a bias in the comparison, a best estimate of the potential risk of toxic releases associated with a product is needed, while risk assessment typically assumes conservative values in order to fulfil protective goals in line with prudent health or environmental policy thresholds.

- Generic/average ecosystems and impacts: Due to the limited information available on the sensitivity of species to toxic effects of substances and the local composition of an ecosystem (i.e. species present), the ecosystems assessed in LCIA are of a generic nature and currently do not consider variations in the composition of species present, or variability in their tolerance to toxic stressors. This may partly change with the introduction of terrestrial ecosystem assessment in LCA, where highly variable soil parameters can influence the toxicity of some substances. This level of detail (i.e. precision) may also not be required for each elementary flow in an LCA, making the generic/average character of models and data both a current methodological limitation (e.g. for very sensitive and uncertain elementary flows and impact indicators) as well as a principle (e.g. for the many elementary flows that are not sensitive to results/conclusions of an LCA).

Several working groups and initiatives have established criteria for good characterisation modelling for different impact categories. Of current relevance for ecotoxicity characterisation are notably the recommendations from the UNEP/SETAC Life Cycle Initiative and related activities:

1) The Lausanne review workshop in 2003 aimed to establish a framework for Life Cycle Toxicity Assessment and recommended a number of modelling elements and choices based on input from ecotoxicity experts outside the LCA community (Jolliet et al. 2006).
2) The declaration of Apeldoorn from 2004 on LCIA of Non-Ferrous Metals (Ligthart et al. 2004) underlined the relevance of a number of aspects for a correct modelling of the fate and toxicity of essential elements.
3) The Clearwater consensus workshop in 2008 for the estimation of metal hazard in fresh water (Diamond et al. 2010) addressed 'inconsistencies in assumptions and approaches for organic substances and nonferrous metals'.
4) The recommendations from these workshops were implemented as far as possible in the UNEP/SETAC toxicity consensus model USEtox. Additionally, a large number of scientifically consensual model elements were identified by the USEtox team and hence became further recommended elements, also included in USEtox (Henderson et al. 2011, Rosenbaum et al. 2008).
5) Based on pre-defined criteria and requirements for good characterisation modelling practice (EC-JRC 2010) and supported by a team of experts (Hauschild et al. 2013), the European Commission established recommendations for LCIA as described in the ILCD Handbook on Recommendations for Life Cycle Impact Assessment in the European context (EC-JRC 2011).

2 Impact pathway and affected Areas of Protection

2.1 Overview

As shown in **Error! Reference source not found.**8.1, the mechanism of toxic impacts of chemicals in LCA can be divided into four parts.

1) Fate modelling estimates the increase in concentration in a given medium due to an emission quantified in the life cycle inventory.
2) The exposure model quantifies the chemical's bioavailability in the different media by quantifying the bioavailable fraction of the total concentration.
3) The effect model relates the amount available to an effect on the ecosystem. This is typically considered a midpoint indicator in LCA, as no distinction between the severity of observed effects is made (e.g. a temporary/reversible decrease in mobility and death are given the same importance).

4) Finally, the severity (or damage) model translates the effects on the ecosystem into an ecosystem population (i.e. biodiversity) change integrated over time and space.

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All four parts of this environmental mechanism are accounted for in the definition of the substance-specific and emission-compartment-specific characterisation factor *CF*:

$$CF = FF * XF * EF * SF \quad \text{(Eq. 8.1)}$$

Where *FF* is the fate factor, *XF* the exposure factor, *EF* the effect factor, and *SF* the severity factor. Each of these four elements of the environmental mechanism of ecotoxicity, and thus its characterisation factor, is described in the following sections.

2.2 Fate

The fate model predicts the chemical behaviour/distribution in the environment accounting for multimedia (i.e. between environmental media and compartments) and spatial (i.e. between different zones but within the same compartment or medium) transport between environmental compartments (e.g. air, water, soil, etc.). This is accomplished via mass-balance-based modelling of (thermodynamic) exchange processes such as partitioning, diffusion, sorption, advection, convection – represented as arrows in **Error! Reference source not found.**8.2 for the USEtox model – as well as biotic and abiotic degradation (e.g. biodegradation, hydrolysis, or photolysis), or burial in sediments. These processes are quantified in rate coefficients which are used to construct a differential equation system for all compartments. This system is solved assuming steady-state by employing matrix algebra (see (Rosenbaum et al. 2007). Further details on fate modelling principles in the USEtox model can be found in Henderson et al. (2011) and Rosenbaum et al. (2008).

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2.3 Exposure

Exposure is the contact between a target and a pollutant via an exposure boundary for a specific duration and frequency (see detailed discussion by Duan et al. (1990)). The exposure model accounts for the fact that not necessarily the total ('bulk') chemical concentration present in the environment is available for exposure of organisms. Several factors and processes such as sorption, dissolution, dissociation and speciation may influence (i.e. reduce) the amount of chemical available for ecosystem exposure. According to Semple et al. (2004) such phenomena can be defined as bioavailability ('freely available to cross an organism's cellular membrane from the medium the organism inhabits at a given time'), and bioaccessibility ('what is actually bioavailable now plus what is potentially bioavailable'). Current LCIA methods consider exposure by calculating the dissolved concentration (Henderson et al. 2011, Rosenbaum et al. 2008), or the bioavailable fraction (Owsianiak et al. 2013, Gandhi et al. 2011a, Gandhi et al. 2010).

2.4 Effects

The effects model characterises the fraction of species within an ecosystem that will be affected by chemical exposure. Effects are described by lab-test derived concentration-response curves relating the concentration of a chemical to the fraction of a test group that is

affected (e.g. 50% of a group of individuals of the same species compared to a control situation). Affected can mean various things, such as mortality, reduced mobility, reduced growth or reproduction rate, mutations, behavioural changes, changes in biomass or photosynthesis, etc. The toxicity tests are standardised and the results are specific for each substance and species. Toxic effects are further distinguished into acute, sub-chronic, and chronic toxicity (including further sub-groups like sub-acute, etc.). Acute toxicity describes an adverse effect after a short period of exposure, relative to the life-time of the animal (e.g. < 7 d for vertebrates, invertebrates, or plants, and < 3 d for algae). Chronic toxicity is based on exposure over a prolonged period of time covering at least one life cycle or one sensitive period (e.g. ≥ 32 d for vertebrates, ≥ 21 d for invertebrates, ≥ 7 d for plants, and ≥ 3 d for algae). A simplified (i.e. illustrative) dose-response curve for a single species is shown in **Error! Reference source not found.**8.3 (left). Important toxicity measures typically determined and reported from the tests are the NOEC – No Observed Effect Concentration (highest tested concentration without any observable effect), the LOEC – Lowest Observed Effect Concentration (lowest tested concentration at which an effect is observed), and the EC50 – Effect Concentration affecting 50% of the individuals above background (if the observed effect is death, the reported parameter may be the LC50 – Lethal Concentration killing50% more of the individuals than in the background sample).

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Between different species a large variation of sensitivity to a given substance can usually be observed. This is described by a species-sensitivity-distribution (SSD) curve, which hence represents the sensitivity of the entire ecosystem to a substance. The SSD is constructed using the respective geometric mean of all available and representative EC50 values for each species. This curve represents the range of sensitivity to exposure to a given substance among the different species from the most sensitive to the most robust species. As discussed above, LCA requires linear models, therefore the SSD curve is generally simplified to a linear regression between the origin (i.e. where x-axis and y-axis cross in their 0 values) along the concentration-response relationship up to the point where the PAF is 0.5 (**Error! Reference source not found.**8.4). The ecotoxicity effect factor is then calculated using the HC50 – Hazardous Concentration at which 50% of the species (in an aquatic ecosystem) are exposed to a concentration above their EC50. Based on Traas et al. (2002) and Klepper et al. (1998), the dimension of the effect factor is PAF – Potentially Affected Fraction of species, while the unit is typically m^3/kg. As visible from **Error! Reference source not found.**8.3 (right), the HC50 employed in LCA is an average value (best estimate) and significantly different from the conservative choice of the PNEC – Predicted No Effect Concentration used in Environmental Risk Assessment, which is based on the most sensitive species and thus on the lower end of the SSD curve. The use of the geometric mean to represent average toxicity for a population or an ecosystem is a standard approach in both ERA (Aldenberg et al. 2002, Forbes and Calow 2002, Versteeg et al. 1999, Newman and Dixon 1996) and LCA (Henderson et al. 2011, Larsen and Hauschild 2007b, Pennington et al. 2006, Pennington et al. 2004a, Payet 2004, Payet and Jolliet 2004). An important argument for its use in LCA is to avoid a bias in the comparative assessment of substances. As discussed by Henderson et al. (2011) as well as Larsen and Hauschild (2007b), the geometric mean is less sensitive to extremely high toxicity (low EC50) values and thus more representative for average toxicity to an ecosystem. In LCA this is very important, since a very well-studied substance, tested on many species, will sooner or later have been tested on a very sensitive species, whereas a substance that has only been tested on one or a few species will likely come out less toxic when using the PNEC approach (most sensitive species), simply because it has not yet been tested on a very sensitive species

(the more sensitive species will differ among substances). Such a bias needs to be avoided in a comparative assessment.
According to the current scientific consensus, the ecotoxicological effect factor of a chemical is calculated as (Henderson et al. 2011):

$$EF = \frac{0.5}{HC50} \qquad \text{(Eq. 8.2)}$$

The log HC50 can be calculated as follows using the EC50 per species respectively:

$$\log HC50 = \frac{1}{n_s} \cdot \sum_s \log EC50_s \qquad \text{(Eq. 8.3)}$$

where n_s is the number of species.

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2.5 Severity

A damage model, incorporating the severity of the effect, goes even further along the cause-effect chain and quantifies how many species are disappearing from a given ecosystem. Disappearance may be caused by mortality, reduced proliferation, or migration, for example. Currently, various approaches exist but none is sufficiently accepted by the scientific community to reach the status of recommendation. Several LCIA methodologies are available expressing the damage on ecosystems in PDF – Potentially Disappeared Fraction. IMPACT 2002+, for example, assumes a relation between PAF and PDF as: PDF = PAF/2, based on the assumption that 50% of the affected species will disappear from the ecosystem (Jolliet et al. 2003). ReCiPe assumes that PAF(EC50) = PDF based on limited evidence from Posthuma and De Zwart(2006) that species loss due to mixture toxicity matches predicted risk with a maximum observed PDF equal to the EC50-based ecotoxicity predictor variable. Larsen and Hauschild (2007a) observed that ‘the recovery time approach used as media recovery has been used in some attempts to include damage modelling in the Eco-indicator 99 method (Goedkoop et al. 2000, Goedkoop et al. 1998), and most recently in IMPACT 2002+ (AMI) (Jolliet et al. 2003)’. Further details and a discussion on freshwater ecotoxicity damage modelling can be found in (Larsen and Hauschild 2007a).

2.6 Affected Areas of Protection

When relating to freshwater ecosystems, the question arises what exactly we mean by that. In LCIA, a freshwater ecosystem is typically described by at least the first three of the trophic levels (Henderson et al. 2011, Rosenbaum et al. 2008, Larsen and Hauschild 2007b, Pennington et al. 2006, Pennington et al. 2004a, Payet 2004, Payet and Jolliet 2004):

1. Primary producers, converting sunlight into biomass via photosynthesis (i.e. phytoplankton, algae),
2. Primary consumers, living off primary producers (i.e. zooplankton, invertebrates, planktivorous fish),
3. Secondary consumers at the upper end of the aquatic food chain (i.e. piscivorous fish).

It should be noted that only impacts on cold-blooded species in freshwater ecosystems are currently considered. The latest state-of-the-art methods available in scientific literature,

though only partially included in LCIA methodologies, also explore modelling the toxicity for warm-blooded freshwater predators (Golsteijn et al. 2012), as well as toxic impacts on terrestrial ecosystems (Owsianiak et al. 2013, Haye et al. 2007, Huijbregts 1999). However, there is no minimum requirement established, which trophic levels should be covered by a characterisation factor for terrestrial and marine ecosystems and available methods usually extrapolate from freshwater data or use the relatively few data available directly for these ecosystems.
Quantifying a potential reduction in species present in an ecosystem, ecotoxic impacts may contribute to damage to the Area of Protection sometimes called natural environment (e.g. ILCD Handbook (EC-JRC 2011)), and sometimes called ecosystem quality (e.g. IMPACT 2002+ (Jolliet et al. 2003), or ecosystems (e.g. ReCiPe (Goedkoop et al. 2012)).

3 Contributing substances (classification)

About 500 years ago Paracelsus stated that 'All substances are poisons; there is none which is not a poison. The right dose differentiates a poison and a remedy'. Today's toxicology science still agrees and adheres to this principle and in consequence any substance emitted may lead to toxic impacts depending on a number of driving factors:

1. Emitted quantity (as determined in the LCI)
2. Mobility (as determined by in the fate factor)
3. Persistence (as determined by the fate factor)
4. Exposure patterns and bioavailability (as determined by the exposure factor)
5. Toxicity (as determined by the effect factor)

This shows that toxicity is not the only parameter that determines the potential ecotoxic impact of a chemical in the environment as it first has to reach a potential target organism. For example, a substance may be very toxic, but never reach any organism due to its short lifetime in the environment (e.g. fast degradation) or because it is not mobile enough to be transported to a target organism and ends up bound to soil matrix or buried in sediment, in which case it contributes little to ecotoxic impacts. On the other hand, a substance may not be very toxic, but if it is emitted in large quantities and over prolonged periods of time or has a strong environmental persistence, it may still cause an ecotoxic impact.

Ecotoxity is very different from any other (non-toxicity) impact category when it comes to the number of potentially relevant elementary flows. Whereas no other (non-toxicity) impact category – with the exception of photochemical ozone formation – exceeds 100 contributing elementary flows (characterisation factors), both toxicity categories are facing the challenge of having to characterise several tens of thousands of chemicals. The CAS registry currently contains more than 70 million unique organic and inorganic substances (www.cas.org/about-cas/cas-fact-sheets) of which roughly 100,000 may play an important industrial role as reflected by the more than 90,000 substances registered in the European Classification and Labelling Inventory Database which contains REACH (Registration, Evaluation, Authorisation and Restriction of Chemical substances) registrations and CLP (Classification, Labelling and Packaging of substances and mixtures) notifications so far received by the European Chemicals Agency (ECHA: http://ihcp.jrc.ec.europa.eu/our_databases/esis). Current LCIA models cover around 2500 substances for aquatic ecotoxicity (Rosenbaum et al. 2008).

4 Scale, spatial variability, temporal variability

Ecotoxicity is often considered as a local impact category (i.e. potential impacts will be taking place relatively close – within a few hundred kilometres – to the source of emissions).

However, this is not fully true as this may vary greatly depending on the local conditions, the persistence, and the physico-chemical properties of the substance emitted. A very persistent, mobile and bioaccumulating substance may travel many thousands of kilometres and accumulate in food chains around the entire globe, thereby being diluted in the transport medium. Spatial variability is therefore a non-negligible source of uncertainty and requires further insights. With some exceptions (Owsianiak et al. 2013, Gandhi et al. 2011b, Gandhi et al. 2010), literature mostly focuses on spatial variability in chemical fate and human exposure. Temporal variability is a largely unexplored issue, which may have potentially important influence on the characterisation for some substances and/or ecosystems. Concerning the time scale, most LCIA methodologies employ an infinite time horizon, except ReCiPe and IMPACT World+. ReCiPe provides an extra scenario with characterisation factors for a 100-year time horizon for metals. IMPACT World+ provides characterisation factors for 100 years and >100 years respectively for metals. Both methodologies thus allow considering the influence of the time scale on the impact score. For metals this is very important because metals do not degrade and their impacts occur over a very long period of time, which leads to very high (typically dominating) ecotoxicity impact scores when integrating impacts over infinity. In such a case the choice of time horizon represents a trade-off between representing all impacts (when integrating over infinity) on the one hand, and representing impacts that may be large for current generations (when integrating over 100 years), but which are 'diluted' when integrating over infinity on the other hand.

Model uncertainty observed in model variability between harmonised versions of IMPACT 2002, USES-LCA, EDIP, and USEtox was estimated by Rosenbaum et al. (2008) as about three orders of magnitude. Important sources of parameter uncertainty are degradation rates for organic substances (Rosenbaum et al. 2008), neglecting bioavailability due to speciation in metals (Chapman 2008, Chapman et al. 2003), the effect factors due to the use of chronic and acute data as well as a linear dose–response curve (Henderson et al. 2011, Rosenbaum et al. 2008), and the lack of toxicity data for species from various trophic levels (van Zelm et al. 2007). As discussed in (Rosenbaum et al. 2008), despite their uncertainty, ecotoxicity impact scores can still be usefully interpreted when seen in the context of 12 (and in fact up to 17) orders of magnitude difference between the lowest and the highest possible (known and characterised) chemical impacts per unit emission. This means that for the LCA practitioner, these CFs can help identify the 10 or 20 most important chemicals (i.e. dominating the impact by contributing together more than 99% of the impact score) for a given application, and, perhaps more importantly, to disregard hundreds of other substances whose impact is not significant for the considered application. Tørsløv et al. (2005) discussed that excluding spatial variability may be less influential on overall uncertainty than parameter uncertainty. IMPACT World+ (impactworldplus.org) is the first fully spatially resolved LCIA methodology that provides quantified uncertainty estimates for all CFs together with separate estimates of spatial variability contributing to overall uncertainty depending on the level of spatial resolution applied.

5 Midpoint methodologies

In the 1990's several early models aiming at comparatively assessing toxicity have been published (Steen 1999, Krewitt et al. 1998, Hauschild and Wenzel, 1998, Jolliet and Crettaz 1997, Walz et al. 1996, Guinée and Heijungs 1993, Braunschweig and Müller-Wenk 1993). More details and overview of many of the early models is given by Udo de Haes et al. (2002).

Several early LCIA methodologies, such as Eco-indicator 99 (Goedkoop et al. 1998) or TRACI 1.0 (Bare et al. 2003) essentially adopted models and measures coming from the assessment of chemical risks to a local environment (e.g. EUSES 1.0 (EC 1996) and CalTOX 4.0 (Hertwich et al. 2001, McKone 2001) respectively), a methodology that was already well established and applied by then. Over time, however, a growing community of researchers started to adapt these models and redefine measures more suitable for comparative assessment of chemical impacts. Both communities are closely linked and collaborating but have evolved individually in the last decade, resulting in a number of specialised LCIA methodologies for characterisation of ecotoxicity impacts that employ well distinguishable approaches relative to regulatory risk assessment (Pennington et al. 2006).

Characterisation methods like EDIP (Hauschild and Potting 2003, Hauschild and Wenzel 1998) account for fate and exposure relying on key properties of the chemical applied to empirical models. For example, the octanol-water partitioning coefficient is used to determine the accumulation of the compound in the food chain. Mechanistic models and methodologies have been published accounting for fate, exposure, and effects providing cardinal impact measures. Among these methods are IMPACT 2002 (Pennington et al. 2005, Jolliet et al. 2003), USES-LCA (van Zelm et al. 2009, Huijbregts et al. 2000), Eco-Indicator 99 (Goedkoop et al. 1998) and ecotoxicity potentials provided by Hertwich et al. (2001) and McKone (2001) using the CalTOX model (McKone et al. 2001). All these methods adopt environmental multimedia, multipathway models employing mechanistic cause-effect chains to account for the environmental fate, exposure, and effects processes. However, they do not necessarily agree on how these processes are to be modelled, leading to variations in results of LCA studies related to the choice of LCIA methodology (Pant et al. 2004, Dreyer et al. 2003). The scientific consensus model USEtox (UNEP/SETAC toxicity consensus model) was developed with the intention to solve this situation by representing a scientifically agreed consensus approach to the characterisation of human and freshwater ecotoxicity (Henderson et al. 2011, Rosenbaum et al. 2008, Hauschild et al. 2008a). **Error! Reference source not found.** and **Error! Reference source not found.** give an overview of a number of LCIA methodologies and the ecotoxicity characterisation models they employ.

Among the existing LCIA methodologies on midpoint level, three main groups can be distinguished: 1) mechanistic, multimedia fate, exposure and effects models, 2) key property-based partial fate models, and 3) non-fate models (EC-JRC 2011). According to ISO 14044 (2006) “Characterisation models reflect the environmental mechanism by describing the relationship between the LCI results, category indicators and, in some cases, category endpoints. [...] The environmental mechanism is the total of environmental processes related to the characterisation of the impacts.” Therefore, ecotoxicity characterisation models falling into categories 2) and 3), do not completely fulfil this criterion. Caution is advised regarding their use and most importantly the interpretation of their results, which should not be employed without prior in-depth study of their respective documentation. Having said that, depending on the goal and scope of the LCA, they may still be an adequate choice in some applications, and indeed agree quite well with the more sophisticated multimedia-based models as demonstrated for a harmonized version of the EDIP model (Rosenbaum et al. 2008). When deciding which LCIA methodology to use in an LCA, an overview of selected properties of these methodologies helps to identify suitable options. As a general recommendation for selecting an LCIA methodology (for ecotoxicity), the priority should be on the following three criteria: 1) substance coverage, 2) state-of-the-art mechanistic modelling, and 3) which ecosystems are considered. As science (and tools) is advancing visibly in this field, it is recommendable to choose a methodology not older than 5-10 years. Unless a methodology provides spatially variable characterisation factors, the regional focus

is secondary and should not be a priority criterion for methodology selection. **Error! Reference source not found.** provides an overview of current midpoint LCIA methodologies and several of their properties. A good overview and many further details can be found in the ILCD Handbook on LCIA recommendations (EC-JRC 2011).

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6 Endpoint methodologies

Ecotoxicity endpoint modelling is still in an early state and much research needs to be performed before maturity is reached. The authors of the ILCD LCIA handbook concluded that "For all the three evaluated endpoint methods (EPS2000, ReCiPe, IMPACT2002+), there is little or no compliance with the scientific and stakeholder acceptance criteria, as the overall concept of the endpoint effect factors is hardly validated and the endpoint part of the methods is not endorsed by an authoritative body. [...] No method is recommended for the endpoint assessment of ecotoxicity, as no method is mature enough." (EC-JRC 2011). **Error! Reference source not found.** provides an overview of current endpoint LCIA methodologies and the ecotoxicity models employed, respectively.

< Insert Table 8.2 >

7 New developments and research needs

The principal objective and motivation behind any further research and development is reduction of uncertainty, notably parameter and model uncertainty, and ecotoxicity characterisation in LCIA is certainly no exception. As can be seen from the discussion above, substance coverage is an important area of further research needs (Hauschild et al. 2013, Finnveden et al. 2009, Rosenbaum et al. 2008). With at least 100,000 substances of potential industrial application and importance and current models covering some 2500 substances, much remains to be done to improve the situation. The currently limiting factor is effect data availability, mainly driven by funding priorities focusing on substances '"of highest political concern, [... while ...] coverage may not be for the most important chemical emissions in the life cycle of a specific product" (Finnveden et al. 2009). Even if a substance is already included in the list of available CFs, it may only be with significant uncertainties for some substances as reflected by the distinction between interim (i.e. with higher uncertainty) and recommended (i.e. with acceptable uncertainty) CFs in USEtox (Rosenbaum et al. 2008). Many substances are currently not characterised or only with insufficient accuracy. Important examples are ionic and amphiphilic substances, persistent bioaccumulating chemicals, persistent surface active compounds, pesticides and biocides, substituted musks/fragrances, biochemicals (i.e. antibiotics, nucleotides, proteins (including enzymes), peptides, polyamino acids, buffers, lipids, carbohydrates, antibodies), and metals. The same can be said for the robustness of the effect factor, which requires a minimum amount of ecotoxicological data for several species being available. These data should ideally represent chronic toxicity tests, but in reality most effect factors are based on acute effect data due to a lack of chronic data (which are significantly more expensive to obtain). Currently, availability and quality of ecotoxicological effect data are the most limiting factors for increasing substance coverage, as the physico-chemical properties required for the fate modelling are already available for several tens of thousands of substances.

Including marine and terrestrial ecosystems, in a scientifically more adequate way than currently, is another very important research need as they contain a significant biodiversity.

Although some LCIA methodologies propose CFs for marine and terrestrial ecotoxicity, their application is not recommended neither at midpoint nor at endpoint level by EC-JRC (2011) and Hauschild et al. (2013). The same authors in accord with Rosenbaum et al. (2008) also conclude that for freshwater ecotoxicity, no viable approach is available to model impacts from midpoint to endpoint, corresponding to the step from accounting for the number of species affected by any kind of toxic impact to the number of species potentially disappearing from the ecosystem.

An important problem for some emissions is the time horizon of their potential impacts and how to meaningfully consider these in LCA and LCIA (Doka 2009, Zhao et al. 2009, Reid et al. 2009, Hauschild et al. 2008b, Doka and Hischier 2005, Hellweg and Frischknecht 2004, Hellweg et al. 2003, Finnveden and Nielsen 1999). The most prominent examples are long-term emissions of metals from landfills or mine tailings. Small amounts of leachate or run-off containing very small concentrations of pollutants (especially heavy metals) are emitted from the landfill to the surrounding soil, aquifer, and eventually the surface water during thousands of years. In LCIA, impacts are modelled using steady-state conditions, applying integration over a defined time horizon. Integrating the impacts of long-term emissions over a relatively short time horizon like 500 years (as done for e.g. Global Warming Potentials), and thus neglecting impacts occurring later, leads to a strong underestimation of their impacts. On the other hand, their full consideration via integration over large or even infinite time horizons would lead to a strong overestimation, as the (perhaps small) impacts occurring over a long period of time would be fully attributed to the product as if they were occurring right now (as one large impact) and without considering the possibility of future technological solutions to the problem. While the latter approach does not account for the ‘dilution of the impact over time’, the first approach completely neglects these impacts. These two extremes represent a dilemma for which a meaningful solution is needed in LCIA.

Further insights into spatial variability – the influence of the place of an emission on the impact – will help reducing uncertainty due to neglecting locally specific ecotoxicity problems and thus help increase accuracy for this impact category. The influence of temporal variability, such as seasonal behaviour of species or weather patterns, remains to be examined. Mixture toxicity is a research area of high importance and complexity. One prominent example, potentially relevant for many LCA studies, concerns effluents from industrial and waste water treatment processes, which are mixtures of varying composition containing many different substances. These further research needs are thereby not a matter of increasing the complexity of characterisation models and their application to an impractical level, but rather about establishing a parsimonious balance between necessary complexity and maximised simplicity. In order to simplify, scientists have to first explore complexity, which will lead to finding a meaningful balance including the consideration of practical needs on the level of application and with full conscience about the uncertainties introduced.

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Tables

Table 8.1: Current midpoint characterisation methods (extended from ILCD handbook on LCIA (EC-JRC 2011))

LCIA methodology	Characterisation model	Fate/exposure modelling	Effect modelling	Ecosystems considered	Time horizon	Region modelled	No. of substances
CML 2002 (Guinée et al. 2002)	USES-LCA 1.0 (Huijbregts et al. 2000)	Mechanistic, nested, multimedia LCA model	Most sensitive species	Freshwater, marine, terrestrial	Infinite	Europe	~170
EDIP 2003 (Hauschild and Potting 2003; Hauschild and Wenzel 1998)	EDIP1997, combined with site dependent factors (Tørsløv et al. 2005)	Key property, partial fate	Most sensitive species	Freshwater, terrestrial	Infinite	Generic	~190
TRACI 1.0 (Bare et al. 2003)	CalTOX 4.0 (McKone et al. 2001)	Mechanistic, closed, multimedia ERA model	Most sensitive species	Freshwater, terrestrial	Infinite	USA	~160
IMPACT 2002+ (Jolliet et al. 2003)	IMPACT 2002 (Pennington et al. 2005)	Mechanistic, nested, multimedia LCA model	Average toxicity	Freshwater, marine, terrestrial	Infinite	Europe	~430
LUCAS (Toffoletto et al. 2007)				Freshwater, terrestrial		Canada	
MEEuP (Kemna et al. 2005)	None (policy-based target emission limits)	None	None	n/a	n/a	Europe	30
Swiss Ecoscarcity 2006 (Frischknecht et al. 2009)	None (policy-based target emission limits)	None	None	n/a	n/a	Switzerland	25
ReCiPe 2008 (updating CML2002) (Goedkoop et al. 2012)	USES-LCA 2.0 (van Zelm et al. 2009)	Mechanistic, nested, multimedia LCA model	Average toxicity	Freshwater, marine, terrestrial	Infinite, 100 years for metals	Europe	~2650
TRACI 2.0 (Bare 2011)	USEtox (Rosenbaum et al. 2008)	Mechanistic, nested, multimedia LCA model	Average toxicity	Freshwater	Infinite	Global generic	~2550
ILCD (EC-JRC 2011)							
IMPACT World+ (impactworldplus.org) updating IMPACT 2002+, LUCAS, EDIP				Freshwater, marine, terrestrial	Infinite, ≤100 y, >100 y for metals	Global generic + 9 sub-continents	

Table 8.2: Current endpoint characterisation methods (extended from ILCD handbook on LCIA (EC-JRC 2011)).

LCIA methodology	**Characterisation model**	**Fate/exposure modelling**	**Effect modelling**	**Ecosystems considered**	**Time horizon**	**Region modelled**	**No. of substances**
EPS 2000 (Steen 1999)	None (red list species pot. threatened by chemicals)	None	None	n/a	n/a	Generic	~45
Eco-Indicator99 (Goedkoop et al. 1998)	EUSES 1.0 (EC 1996)	Mechanistic, nested, multimedia ERA model	Most sensitive species	Freshwater, terrestrial	Infinite	Europe	~45
IMPACT 2002+ (Jolliet et al. 2003)	IMPACT 2002 (Pennington et al. 2005)	Mechanistic, nested, multimedia LCA model	Average toxicity	Freshwater, marine, terrestrial	Infinite	Europe	~430
LIME 1.0 (Itsubo and Inaba 2003)				Freshwater, terrestrial		Japan	
ReCiPe 2008 (updating EI99) (Goedkoop et al. 2012)	USES-LCA 2.0 (van Zelm et al. 2009)	Mechanistic, nested, multimedia LCA model	Average toxicity	Freshwater, marine, terrestrial	Infinite, 100 years for metals	Europe	~2650
IMPACT World+ (impactworldplus.org) updating IMPACT 2002+, LUCAS, and EDIP	USEtox (Rosenbaum et al. 2008)	Mechanistic, nested, multimedia LCA model	Average toxicity	Freshwater, marine, terrestrial	Infinite, ≤100 y, >100 y for metals	Generic + 9 parameterised sub-continents	~2550

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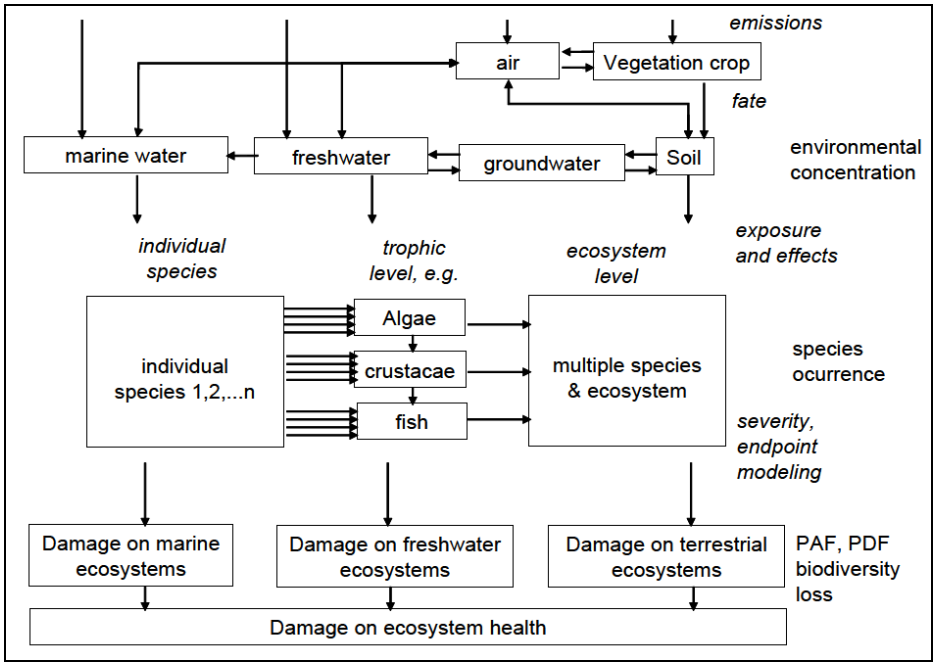

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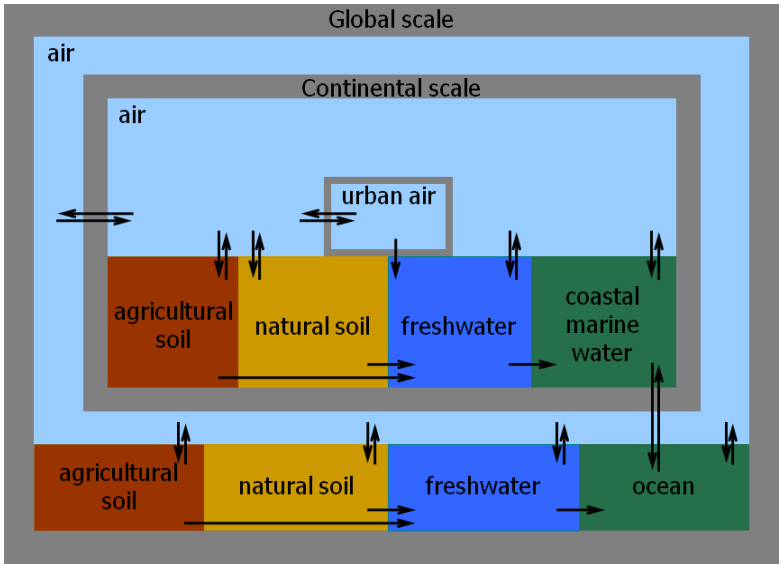

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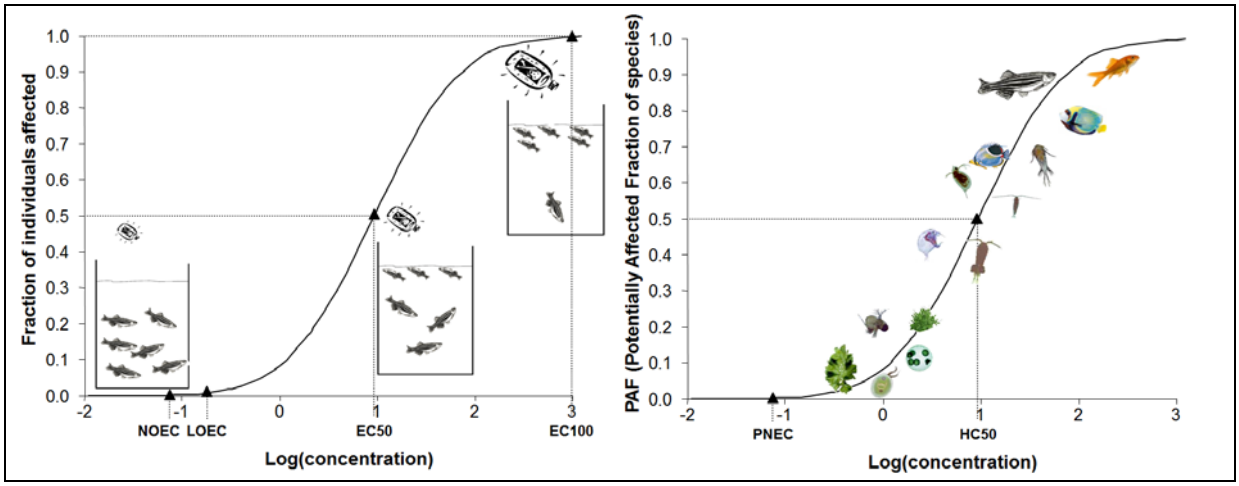

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