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Spatial variability versus parameter uncertainty in freshwater fate and exposure factors of chemicals

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HIGHLIGHTS

- We compared parameter uncertainty and spatial variability in freshwater fate and exposure factors.
- Depending on partitioning behaviour, either variability or uncertainty dominates.
- Variability and uncertainty factors were up to 2 and 3 orders of magnitude, respectively.
- Both spatial variability and parameter uncertainty should be accounted for in freshwater fate factors.

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ABSTRACT

We compared the influence of spatial variability in environmental characteristics and the uncertainty in measured substance properties of seven chemicals on freshwater fate factors (FFs), representing the residence time in the freshwater environment, and on exposure factors (XFs), representing the dissolved fraction of a chemical. The influence of spatial variability was quantified using the SimpleBox model in which Europe was divided in 100 × 100 km regions, nested in a regional (300 × 300 km) and supra-regional (500 × 500 km) scale. Uncertainty in substance properties was quantified by means of probabilistic modelling. Spatial variability and parameter uncertainty were expressed by the ratio k of the 95%ile and 5%ile of the FF and XF. Our analysis shows that spatial variability ranges in FFs of persistent chemicals that partition predominantly into one environmental compartment was up to 2 orders of magnitude larger compared to uncertainty. For the other (less persistent) chemicals, uncertainty in the FF was up to 1 order of magnitude larger than spatial variability. Variability and uncertainty in freshwater XFs of the seven chemicals was negligible ($k < 1.5$). We found that, depending on the chemical and emission scenario, accounting for region-specific environmental characteristics in multimedia fate modelling, as well as accounting for parameter uncertainty, can have a significant influence on freshwater fate factor predictions. Therefore, we conclude that it is important that fate factors should not only account for parameter uncertainty, but for spatial variability as well, as this further increases the reliability of ecotoxicological impacts in LCA.

1. Introduction

Life Cycle Assessment (LCA) focuses on the assessment of impacts of products and services on human health and environment. The impact of a product or service for the impact categories of interest, e.g. ecotoxicity, is determined in the Impact Assessment (LCIA) phase, and quantified with Characterisation factors (CF) of individual chemicals. Characterisation factors for ecotoxicity express the magnitude of the impact of a unit mass released and depend, besides on the toxicity, on the environmental fate and exposure of a chemical. Fate and exposure is quantified with fate factors (FF [days]), which represents the compartment-specific residence time of the chemical in the environment, and exposure factors (XF [-]), which is depended on the dissolved fraction of the chemical (Huijbregts et al., 2000a; Pennington et al., 2004; Rosenbaum et al., 2008). The assessment of a chemical's fate and exposure can be done with multimedia fate models (Den Hollander et al., 2004; Pennington et al., 2005; Rosenbaum et al., 2008; Van

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Zelm et al., 2009). These models require information on landscape parameters, such as the fraction of organic carbon in soil, as well as physicochemical properties, such as the degradation rate of a chemical in water.

Fate and exposure predictions are subject to spatial variability, i.e. randomness of nature (Walker et al., 2003). Whereas uncertainty can be reduced with additional measurements, variability is inherent in the environment and cannot be reduced by additional research. Hollander et al. (2009) showed that variation in substances' environmental fate mainly depends on chemical properties (partition coefficients and degradation rates) and less on spatial variability.

Hertwich et al. (1999) investigated the influence of parameter uncertainty and spatial variability on potential human dose calculations for multiple chemicals, but this was never done for the environmental fate and exposure of chemicals in an LCA context. Several environmental fate analyses focused on either parameter uncertainty (e.g. Huijbregts et al., 2000b; Luo and Yang, 2007; MacLeod et al., 2002; Van Zelm et al., 2010) or spatial variability (e.g. Ciuffo and Sala, 2013; Hauck et al., 2010; Hauschild et al., 2006; Hollander et al., 2012; Manneh et al., 2010; Oldenkamp et al., 2014; Sleeswijk, 2011; Wania, 1996). The goal of this study was to compare the influence of spatial variability and parameter uncertainty on freshwater fate and exposure predictions. The freshwater fate and exposure of 2,4-dichlorophenol (2,4-DCP), aniline, dichlorodiphenyltrichloroethane (DDT), glyphosate, heptachlor, propiconazole and trichloroethylene were modelled with the nested multimedia fate model Simplebox (Den Hollander et al., 2004; Hollander et al., 2007). These chemicals were selected according to their physical–chemical equilibrium partitioning in air, water, and soil, so that every partitioning region as described by Gouin et al. (2000) was represented. The influence of spatial variability in environmental characteristics was addressed with a scenario analysis of Europe divided into regional grids. The influence of uncertainty in physicochemical properties was quantified by means of probabilistic modelling.

2. Materials and methods

2.1. Fate and exposure factor

Freshwater fate factors (FFs) were predicted with the nested multimedia model SimpleBox (version 3.31) (Den Hollander et al., 2004; Hollander et al., 2007). SimpleBox is an environmental multimedia model, which can be used to simulate a chemical's emission to one of the homogenous environmental compartments. Through a set of linear equations, the model subsequently determines the chemical's environmental fate throughout all the compartments, based on its physicochemical properties and the environmental parameters. The model thereby accounts for inter-compartmental chemical transfer and degradation. SimpleBox forms the basis of the European Union System for the Evaluation of Substances (EUSES) as a regional distribution model (Vermeire et al., 1997) as well as the USEtox consensus model (Rosenbaum et al., 2008), and was shown to predict in line with comparable multimedia fate models (see e.g. Hollander et al., 2007; Kounina et al., 2014; Rosenbaum et al., 2008). SimpleBox 3.31 includes default values for the enthalpy of phase change. Since chemical partitioning properties are temperature dependent, the enthalpy of vaporization was changed from the default value to a temperature and vapour pressure dependent regression following MacLeod et al. (2007).

Here, the FF was calculated by the sum of the chemical steady state mass in the freshwater compartment on the local, regional and supra-regional scale per unit of emission:

$$FF_{x,w,i} = \frac{(\Delta C_{i\rightarrow i} \times V_l) + (\Delta C_{i\rightarrow r} \times V_r) + (\Delta C_{i\rightarrow sr} \times V_{sr})}{\Delta M_w} \quad (1)$$

Where V_l, V_r and V_{sr} are the volumes of the receiving compartment on the local (grid i), regional and supra-regional scale respectively; $\Delta C_{i\rightarrow i}$, $\Delta C_{i\rightarrow r}$ and $\Delta C_{i\rightarrow sr}$ are the steady-state total concentration changes in the receiving compartment on each scale, as a result of the change in emission M to compartment w in local grid i; $FF_{x,w,i}$ is the grid-specific fate factor [day] for substance x emitted to compartment w.

The exposure factor (XF) represents the average dissolved fraction in the freshwater compartment on the local, regional, and supra-regional scale.

$$XF_{x,i} = \frac{(\Delta C_{d,i\rightarrow i} \times V_l) + (\Delta C_{d,i\rightarrow r} \times V_r) + (\Delta C_{d,i\rightarrow sr} \times V_{sr})}{(\Delta C_{i\rightarrow i} \times V_l) + (\Delta C_{i\rightarrow r} \times V_r) + (\Delta C_{i\rightarrow sr} \times V_{sr})} \quad (2)$$

Where $\Delta C_{d,i\rightarrow i}$, $\Delta C_{d,i\rightarrow r}$ and $\Delta C_{d,i\rightarrow sr}$ are the steady-state concentration changes of the dissolved fractions of the substance in freshwater on each scale.

2.2. Spatial variability

To quantify spatial variability in fate and exposure factors, the SimpleBox model was modified following Hauck et al. (2010) and Hollander et al. (2012). Europe was divided into 100 × 100 km grids (local scale), each with its own environmental characteristics. This model was then nested into a regional scale (300 × 300 km), a supra-regional scale (500 × 500 km) and the continental scale. We used this spatially adapted SimpleBox model to simulate the freshwater fate and exposure of an emission to air, freshwater, and agricultural soil on a local scale. The resulting range of the FFs and XFs per emission compartment over all grids was presented as the ratio between its 95%ile and 5%ile, here named variability factor k_{var}.

$$k_{var,x} = \frac{95\%\text{ile of the grid} - \text{specific FFs or XFs}}{5\%\text{ile of the grid} - \text{specific FFs or XFs}} \quad (3)$$

2.3. Parameter uncertainty

To account for the uncertainty in the FFs and XFs, Monte Carlo simulations were performed applying Latin Hypercube sampling by using the spreadsheet-based application Crystal Ball (Oracle©, Release 11.1.2.0.00) in MS Excel 2010 with 10,000 iterations per run. Results were presented as the ratios of the 95%ile and 5%ile of the uncertainty ranges, the latter referred to as the uncertainty factor $k_{unc,i,x}$.

$$k_{unc,i,x} = \frac{95\%\text{ile of the FF or XF uncertainty per grid}}{5\%\text{ile of the FF or XF uncertainty per grid}} \quad (4)$$

2.4. Sensitivity analysis

A sensitivity analysis was conducted to determine which chemical-specific input parameters contribute the most to the uncertainty in FFs and XFs and which environmental parameters contribute the most to the variability in FFs and XFs respectively. The sensitivity analysis was performed by using the Spearman's