Water use and quality in Life Cycle Assessment: identifying good practices and developing operational spatial approaches

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Abstract

Pressure on freshwater resources is increasingly covered by methodological developments addressing freshwater use in the field of Life Cycle Assessment (LCA). These developments ultimately lead to the publication of the ISO 14046 standard to define the principles, requirement and guidelines for a "water footprint" in August 2014. The objective of this thesis is to foster the application of water footprint by identifying good practices and developing operational approaches to assess and improve its discriminatory power. Indeed, academic development and LCA application by practitioners often evolve within distinct communities and with various practical constraints. For instance, newly developed methods are spatially differentiated to reduce model uncertainty, improve accuracy, precision and confidence in LCA results. However, models officially recommended by the European commission for product water footprints are generic (i.e., results do not depend on location), such as USEtox for the modelling of the human toxic and ecotoxic impacts. Furthermore, the practitioner knowledge of the location of emissions is often reduced to direct or first tier supplier emissions when information on emission location is available. The trade-off between theoretical variability reduction and decrease of uncertainty in practice considering the level of geographical information available to the practitioner requires solutions meeting the needs of both LCA actors. This thesis answers the need to bridge methodological development and water footprint application through three main specific objectives:

- Review existing and applicable inventory and impact assessment methods that address quantitative freshwater use in a life cycle perspective and provide preliminary application recommendations for practitioners
- Analyse spatial differentiation of toxic emissions to water applied to USEtox at the inter-continental and intra-continental level and explore simplified spatial differentiation approaches such as spatial archetypes
- Develop a fate and exposure characterization model and factors for toxic emissions into water with global coverage and fine resolution and test practical solutions to apply spatial characterization factors on a case study

The first part of this thesis (Chapter 2) focuses on quantitative aspects of water use by providing a comprehensive review of existing and applicable inventory and impact assessment methods that address freshwater use. It develops a detailed framework of cause-effect chains leading from water use inventory to impact on human health, ecosystem quality and resources, based on which a set of qualitative evaluation criteria were developed aligned with those used in the ILCD handbook. Methods and databases are evaluated against these criteria leading to recommendations of model components to be included in new methodological developments. This analysis proves that a water footprint is already applicable for experienced practitioners relying on the current state-of-the-art methods. It provides a basis to reference the state-of-the-art of methods addressing water use in LCA already used and cited by several authors in the field of water management and specific water footprint case studies.

Practical solutions to operationalize spatial differentiation are tested in Chapter 3 and 4, focusing on impact pathways generated by toxic emissions into water. This thesis validates the use of a nested spatially differentiated model and a sector-specific aggregation of characterization factors into a global average and discriminate the archetype approach for the evaluation of toxic emissions to water. Spatially differentiated landscapes and models are first developed as a reference to test simplified approaches: (1) landscape parameters are created for USEtox to develop continent and sub-continent specific boxes nested within the world and (2) a fate and exposure characterization model and factors are developed to assess toxic emissions into water with global coverage at 0.5°*0.5° resolution. The analysis of inter-continental and intra-continental variation in Chapter 3 leads to the conclusion that a sub-continental nested model such as USEtox, with continent-specific parameterization complemented with freshwater archetypes, can represent spatial variations whilst minimizing model complexity. However, when going one step further into spatial differentiation, i.e. at the 0.5*0.5° scale in chapter 4, the developed archetypes do not represent anymore an implementable and simplified approach of spatial differentiation as they do not follow a systematic geographical pattern (such as rural and urban). A characterization factors aggregation into a global average is shown to be relevant in the case of sector-specific emissions and aggregation for the case study of arsenic and chromium(VI) emissions from red mud disposal during alumina production. The results of this work prove that using a generic model is acceptable in the latter case study to cover a low resolution such a continent, country, or sector (when the detailed emission location is unknown) while a finer resolution is essential for a regional impact score at the watershed, grid cell or point source level. In any case, an uncertainty analysis is useful at a low resolution in order to consider the result variability during the interpretation of impact results.

The limitations and constraints related to water footprint practice are then discussed based on published case studies and personal consulting experience. As an outlook of this work, it is recommended to develop a detailed practitioner-oriented spatial differentiation implementation framework for a pragmatic and easily interpretable application of spatial differentiation in LCA.

Keywords

Life cycle assessment – Water footprint – Spatial differentiation – Human toxicity - Ecotoxicity

Résumé

La pression sur les ressources d'eau douce est traitée par de récents développements méthodologiques dans le domaine de l'analyse de cycle de vie (ACV). Ces développements ont mené à la publication du standard ISO 14046 pour définir les principes, exigences et règles pour l'évaluation d'une « empreinte eau » en août 2014. L'objectif de cette thèse est de favoriser l'application de l'empreinte eau en identifiant les bonnes pratiques et en développant des approches opérationnelles pour évaluer et améliorer son pouvoir discriminatoire. En effet, les développements académiques et l'application de l'ACV par les praticiens évoluent souvent dans des communautés distinctes avec des contraintes pratiques différentes. Par exemple, les méthodes récemment développées sont spatialement différentiées pour réduire les incertitudes du modèle, améliorer son exactitude, sa précision et la confiance dans les résultats de l'ACV. D'autre part, les modèles officiellement recommandés par la commission européenne pour l'empreinte eau d'un produit sont génériques (les résultats ne sont pas différentiés selon la localisation), tel que USEtox pour la modélisation des impacts liés à la toxicité humaine et l'écotoxicité. De plus, les connaissances du praticien sur la localisation des émissions se limitent souvent aux émissions directes ou au premier niveau de fournisseur lorsque l'information est disponible. Le compromis entre la réduction de variabilité théorique et la réduction d'incertitude en pratique considérant la disponibilité d'informations géographiques sur les différents lieux d'émission nécessite des solutions qui satisfassent les besoins de ces deux acteurs de l'ACV. Cette thèse répond au besoin de créer une passerelle entre développements méthodologiques et application de l'empreinte eau à travers les trois objectifs spécifiques suivants :

- Passer en revue les méthodes d'inventaire et d'impact existantes et applicables qui traitent de l'usage quantitatif de l'eau dans une perspective de cycle de vie et proposer des recommandations préliminaires pour les praticiens
- Analyser la différentiation spatiale des émissions toxiques dans l'eau appliquée à USEtox au niveau intercontinental et intracontinental et explorer des approches simplifiées pour la différentiation spatiale telles que les archétypes
- Développer un modèle de devenir et exposition ainsi que des facteurs de caractérisation pour les émissions toxiques dans l'eau avec une couverture globale et fine résolution ainsi que tester des solutions pratiques pour appliquer des facteurs de caractérisations spatiaux dans un cas d'étude

La première partie de cette thèse (chapitre 2) se focalise sur les aspects quantitatifs de l'usage de l'eau douce en réalisant une revue des méthodes d'inventaire et d'impact existantes et applicables. Un cadre de réflexion pour décrire la chaîne cause-à-effet menant de l'inventaire d'usage de l'eau à l'impact sur la santé humaine, la qualité des écosystèmes et les ressources est proposé, et sert ensuite comme base pour établir des critères d'évaluation qualitatifs alignés avec ceux du « ILCD handbook ». A partir de ces critères, les méthodes et bases de données sont évaluées, menant à des recommandations sur les éléments clé du modèle à inclure dans les nouveaux développements méthodologiques. Cette analyse prouve qu'une empreinte eau est déjà applicable pour des praticiens expérimentés en se basant sur l'état de l'art actuel. Cette étude fournit une base pour référencer l'état de l'art de méthodes qui traite de l'usage de l'eau en ACV, déjà utilisée et citée par différents auteurs dans le domaine de la gestion de l'eau et dans des cas d'étude effectuant une empreinte eau.

Des solutions pratiques pour opérationnaliser la différentiation spatiale sont testées dans les chapitres 3 et 4, se focalisant sur les chaînes cause-à-effet générées par des émissions toxiques dans l'eau. Cette thèse a validé l'utilisation d'un modèle imbriqué et d'une agrégation de facteurs de caractérisation en une moyenne pondérée spécifique à un secteur industriel et a discriminé l'approche des archétypes pour l'évaluation des émissions toxiques dans l'eau. Des paysages et modèles avec une différentiation spatiale sont créés comme référence pour tester les approches simplifiés: (1) des paramètres de paysage sont créés pour USEtox pour développer des boîtes continentales et sous-continentales imbriquées dans une boîte globale et (2) un modèle et facteurs de caractérisation sur le devenir et l'exposition des émissions toxiques dans l'eau sont développés avec une couverture globale et une résolution de 0.5*0.5°. L'analyse de variation intercontinentale et intracontinentale dans le chapitre 3 mène à la conclusion qu'un modèle imbriqué à l'échelle sous-continentale tel que USEtox, avec une paramétrisation propre à chaque contient complémentée d'archétypes basés sur le temps de résidence de l'eau jusqu'à la mer, peut représenter les variations spatiales tout en minimisant la complexité du modèle. Toutefois, lorsque l'on va plus loin dans la différentiation spatiale, tel qu'à la résolution de 0.5*0.5° dans le chapitre 4, les archétypes développés ne représentent plus une approche simplifiée et facilement applicable car ils ne suivent pas une disposition géographique systématique (tel qu'urbain et rural). L'agrégation des facteurs de caractérisation en une moyenne globale est néanmoins pertinente dans le cas d'émission et agrégation spécifique à un secteur industriel dans le cas d'étude d'émissions de chrome(VI) et arsenic percolant de la mise en décharge de boues rouges issues de la production d'alumine. Les résultats de ce travail démontrent que l'utilisation d'un modèle générique est acceptable pour couvrir une faible résolution telle qu'au niveau d'un continent, d'un pays ou d'un secteur industriel, tandis qu'un modèle à la résolution spatiale fine est essentiel pour une évaluation régionale au niveau du bassin versant, de la cellule ou d'un site particulier.

Les limites et contraintes à la pratique de l'empreinte eau sont finalement discutées sur la base de cas d'études publiés et de mon expérience personnelle de travail dans une entreprise de conseil. En perspective de ce travail, il est recommandé de développer une approche pragmatique et facilement interprétable pour l'application de la différentiation spatiale en ACV du point de vue du praticien.

Mots-clés

Analyse de cycle de vie – Empreinte eau – Différentiation spatiale – Toxicité humaine - Ecotoxicité

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Chapter 1

Introduction

1.1 Context

Human influence on the hydrological cycle has been increasing over the last decades and brings alterations to the water resources and their availability (Shiklomanov and Rodda 2003). These alterations in turn affect human well-being and ecosystem health (United Nations Environment Programme 2007).

Today, it has been recognized by the international community that potential impacts of water use involved in the production, consumption and end-of-life of manufactured goods and services (both being qualified as products) can be quantified through the tool Life Cycle Assessment (LCA) (ISO 2014). Impact from water use is one among several environmental problem accounted in LCA. Water use is defined according to ISO 14046 2014 as including "any water withdrawal, water release or other human activities within the drainage basin impacting water flows and/or quality, including in-stream uses such as fishing, recreation, transportation".

Nevertheless, the assessment of freshwater quantitative use has received a very limited attention in LCA (Koehler 2008) until the last five years, where it was recognized that there was a need to define a conceptual and normative framework to streamline the implementation of methods assessing freshwater quantitative use, especially based on a review of the developments in that field, and recommend a consensus method to cover this specific cause-effect chain. Two majors initiatives were launched at the end of the last decade to fulfill these needs and aiming at fostering application of water assessment in LCA (also called "water footprint"): the United Nations Environment Programme (UNEP)/Society of Environmental Toxicology and Chemistry (SETAC) Life Cycle Initiative's working group on the assessment of freshwater use and consumption in life cycle assessment (LCA) called "WULCA", and the International Organization for Standardization (ISO) water footprint working group (ISO/TC207/SC5/WG8) with the mission to draft the new ISO 14046 standard (ISO 2014).

Impacts related to water pollution have been traditionally fairly well covered by LCA, using impact categories such as aquatic eutrophication, aquatic acidification, aquatic ecotoxicity (sometimes considering ionizing radiations from decay of radioactive material) and human toxicity. Aquatic eutrophication can be defined as a nutrient enrichment of surface water generating increasing algal growth and changing species abundance and diversity (Smith 2003). Unlike eutrophication, aquatic acidification results from airborne emissions of acidifying substances and their subsequent direct or indirect deposition on aquatic environment. Acidity ultimately creates a number of consequences such as decreased availability of nutrients or an increased concentration of soluble toxic metallic substances that affect fish, aquatic plants and insects in addition to the deviation from their optimum pH living conditions (Roy et al. 2014). The human toxicity and aquatic ecotoxicity impact categories refer to an adverse change in the structure, or function of respectively humans and aquatic species as a result of exposure to a chemical (Pennington et al. 2004). These chemicals encompass both organic and inorganic substances emitted to air, soil or water. Both human toxicity and aquatic ecototoxicity categories are covered through models depicting the entire impact pathway from the emission to the effect on aquatic species or humans. The last decade saw the emergence of two different and complementary model development trends: while some models are sophisticated to increase their environmental relevance and reduce their uncertainty, an effort is provided in parallel by the scientific community to develop scientific consensus models with reduced components to enhance their implementation among all stakeholders. An example of sophistication is made through spatial differentiation, i.e. the discrimination of impacts in relation to the place of emission (Potting and Hauschild 2006; Manneh et al. 2010; Wegener Sleeswijk and Heijungs 2010). Recently developed impact assessment methods are covered by models spatially differentiated at different scales, e.g., from a continental (Lundie et al. 2007; Shaked 2011) to a grid cells resolution (Pistocchi et al. 2010; Roy et al. 2012) for the most refined ones. An example of scientific consensus model is USEtox (Rosenbaum et al. 2008), which aims at combining existing models into a unique one and integrating consensual modelling element by applying the principles of parsimony and mimetism. This consensual model aims at fulfilling the need expressed by LCA practitioners for recommended methods that increase LCA results comparability. These two parallel trends of model development both contribute to the LCA field following sometimes divergent directions by respectively (1) improving its results reliability, which is explored in this work by and (2) fostering and systematizing LCA application.

This thesis focuses on the impact of quantitative water use and qualitative water use. This research work has been performed through the participation in two international projects: the UNEP-SETAC project on water use in LCA (WULCA) and the Life Cycle Impact assessment Methods for imProved sustAinability Characterisation of Technologies (LC-IMPACT), a European Commission founded project under the Seventh Framework Programme. The papers of this PhD are direct contributions to the deliverables of the two projects. I performed this research whilst working as a LCA consultant at the company Quantis. Quantis performs and implements LCA to inform companies and public organisations on the environmental impact of their activities or their products and help them in their impact reduction strategies.

The first chapter of this thesis describes the context and defines the objectives of this thesis. The second chapter provides a systematic review of recently published methods addressing quantitative impacts of water use in LCA. The third and fourth chapters analyze different approaches for assessing spatial differentiation of toxic emissions into water (a specific case of degradative water use) at global scale, develop a set of relevant model landscape archetypes and provide recommendations on how to address spatial differentiation in LCA. The fifth chapter provides a critical appraisal of the thesis where the achievements are put in perspective as a contribution to LCA science and practice and concludes regarding the achievement of defined objectives.

1.2 Literature review

1.2.1 Pressure on water resources

Freshwater in all its states represents only 2.53% of the total hydrosphere of which 1.74% is in the ice sheets of the Antarctic, the Arctic and in mountain glaciers (Shiklomanov and Rodda 2003). However, this situation is not static, as water continuously moves from liquid, solid and gaseous forms as the hydrological cycle progresses under the effects of solar radiation, the energy released from the Earth's interior and gravitational forces (Shiklomanov and Rodda 2003). This cycle is more and more influenced by human freshwater withdrawal, which increased by more than 6 times between 1900 and 2010 (from 500 km^3 yr^{-1} to 3300 km^3 yr^{-1}) (Wada et al. 2014) to sustain growing population and food demand as well as increasing standard of living. Climate changes may also threaten water availability, bringing a warmer climate that decreases water availability, increases the chances of disastrous events, changes the level of water use and its quality, and creates changes in the food supply production through the need for more irrigation, increased productivity or dependence on other countries (Kulshreshtha 1993). Röckström et al. (2009) estimated that humanity's total surface and groundwater consumptive freshwater use is currently within a safe operating limit with an estimation of the current use of 2600 km^3 yr^{-1} and a proposed planetary boundary of 4000 km^3 yr^{-1}. However, when considering the regional nature of freshwater scarcity, the majority of global freshwater withdrawals currently takes place in watersheds already experiencing high water scarcity (Ridoutt and Pfister 2010a). Seckler et al. (1999) from the International Water Management Institute (IWMI) estimated that nearly 1.4 billion people live in regions that will experience severe water scarcity within the first quarter of the twenty-first century.

Water degradation arises from different types of sources and varies depending on a country level of development. Meybeck and Helmer (1989) sequenced chronologically water quality issues in industrialised countries as faecal pollution, organic pollution, salinization, metal pollution, eutrophication, radioactive waste, nitrate organic pollutants and acid rain. Thermal pollution was more recently studied and proved to significantly influence aquatic environments and their biota (Verones et al. 2010). Water quality degradation is proved to harm human and ecosystem health (United Nations Environment Programme 2007). It is estimated that three million people die each year from water-borne diseases mainly related to microbial pathogens and excessive nutrient load in developing countries (United Nations Environment Programme 2007). The impact related to the emission of toxic substances such as pesticides, pharmaceutical products or industrial waste is dramatically known through mass accidental or voluntary release, such as in the case of the Minamata disease, where methylmercury was released in the industrial wastewater, then ingested by population through fish and shellfish (Ui 1992). While human health effects of long term exposure from continuous diffuse emissions to water are unknown (Margot et al. 2013), toxic substances released to water can affect sensitive aquatic organisms even at very low concentrations (Santos et al. 2010). When substances are emitted to surface freshwater, the receiving body hydrology (e.g., freshwater residence time until the sea) plays an important role in the impact magnitude (Pennington et al. 2005; Henderson et al. 2011).

Awareness of humanitarian, social, environmental and economic stakes around water also grows in the private sector. Several initiatives were recently set up to provide tools to measure water use, assist companies in developing water sustainable management or certification programs, such as respectively Global Water Tool by the World Business Council for Sustainable Development (WBCSD 2010), the CEO water mandate (UN Global Compact Office 2011), and the Alliance for Water Stewardship (AWS) (Abdel Al et al. 2014).

A first step towards an integrated water management at the company, sector, community or political boundary level is to gain an adequate understanding of the extent of the inventory and impacts generated by all water uses related to the considered system. There is a need to develop rigorous and evaluated models to measure and compare the “water footprint” of a system, defined as “the metric(s) that quantifies the potential environmental impacts related to water” (ISO 2014).

1.2.2 Life cycle assessment

Life cycle assessment (LCA) is an approach to understand the relationships between human activities and their impact on the environment over the entire life cycle of a system, i.e., from raw material acquisition, via production and use phases, to waste management. Product LCAs are usually conducted to support corporate decision-making, such as for eco-design of products, process optimizations, supply-chain management, and marketing and strategic decisions (Hellweg and Milà i Canals 2014). It quantifies all relevant consumed resources, emissions and waste produced during products (goods or services) life cycle and relates them to associated environmental impacts (including health effects) as well as resource depletion issues. One of the powerful advantages of LCA is that it prevents “shifting” of the impacts from one life cycle stage to another or from one impact category to another. LCA is one of several existing environmental management techniques (others being, e.g., risk assessment, environmental performance evaluation, environmental auditing, and environmental impact assessment). Following the standards ISO 14040 and ISO 14044 (ISO 2006a; ISO 2006b), LCA is defined as “the compilation and evaluation of the inputs, outputs and the potential environmental impacts of a product system throughout its life cycle” and it does not address the economic or social aspects of a product. The general structure of life cycle assessment consists in four phases (ISO 2006a): goal and scope definition, life cycle inventory (LCI), life cycle impact assessment (LCIA) and interpretation that are presented in Figure 1.1.

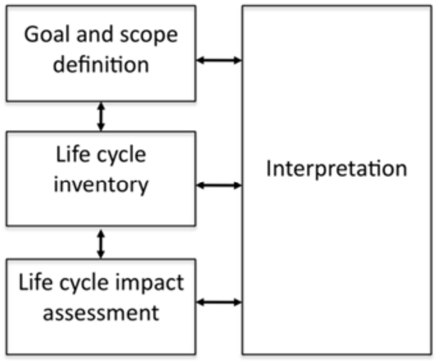

Figure 1.1: Life cycle assessment methodology structure based on ISO 14044 (ISO 2006a)

While the ISO normative framework details the requirements for conducting and reporting an LCA, it does not provide recommendations on LCIA methods that shall be applied (although some application examples are provided in ISO 14047 (2003)). Examples of existing LCIA methods are IMPACT 2002+ (Jolliet et al. 2003), TRACI (Bare et al. 2003), EDIP 2003 (Hauschild and Potting 2005) and ReCiPe (Goedkoop et al. 2009). The ILCD handbook provides recommendations on models that should be used for each impact category covered by LCIA (JRC-IES 2011). Based on these recommendations, the European commission also published a key reference document for LCA industrial actors: both a Product and an Organisation Environmental Footprint (PEF/OEF) guide with detailed recommendations that cover all LCA phases (European Commission 2013a), including a set of impact categories to be used by default in LCA that are PEF/OEF compatible. New methodologies, such as the recently developed IMPACT World+, provide regionalized characterization modeling approaches (Bulle et al. 2012; IMPACTWorld+ 2014). An example of structure of LCIA methodology is presented in Figure 1.2 (Jolliet et al. 2003). LCI results with similar cause-effect chains (e.g., all elementary flows influencing stratospheric ozone concentrations) are grouped into midpoint categories, which ultimately lead to an endpoint (damage) category. An impact indicator is a class representing environmental issues of concern to which life cycle inventory analysis results may be assigned. A category endpoint corresponds to an attribute or aspect of natural environment, human health, or resources, identifying an environmental issue giving cause for concern. The term “midpoint' expresses the fact that this point is located somewhere on an intermediate position between the LCI results and the damage (or endpoint) on the impact pathway.

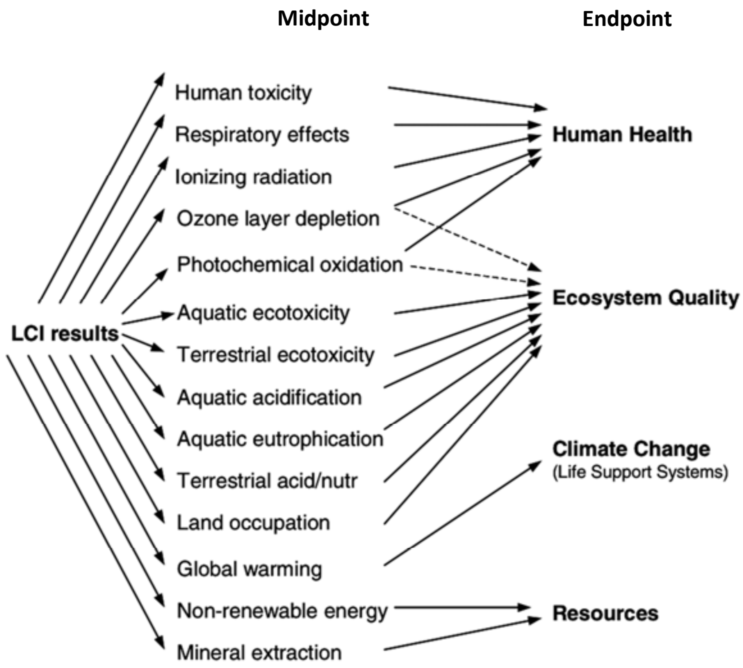

Figure 1.2: Overall LCIA scheme linking LCI results via the category midpoint and category endpoint when using the LCIA method IMPACT 2002+ (Jolliet et al. 2003)

LCIA is then done using a weighted summation of the releases of the substances related to a product system with the help of characterization factors, as illustrated in Equation 1.1:

$$IS = \sum_{i} \sum_{x} CF_{x,i} . M_{x,i}$$

Equation 1.1: Impact score calculation

Where IS is the impact score (e.g., in kg CO_2-eq for the category global warming), $CF_{x,i}$ is the characterization of the substance x released to compartment i (e.g., in kg CO_2-eq/kg) and $M_{x,i}$ is the emission of x to compartment i (e.g., in kg).

1.2.3 Water use in LCA

Quantitative water use is historically a poorly covered impact category in LCA (Koehler 2008). The ILCD handbook (JRC-IES 2011) classifies water use as a sub-category of resource depletion. It recommends the midpoint method of Frischknecht et al. (2006) but “to be used with caution” and considers that evaluated endpoint methods (as of 2008, the time of evaluation) are too immature to be recommended. However, in recent years, new midpoint and endpoint methods have been developed that propose different water use inventory schemes and impact assessment characterization models considering various cause-effect chain relationships (Pfister et al. 2009; Boulay et al. 2011b; Berger et al. 2014). Spatial differentiation is a key cross-cutting issue of these methods given the regional nature of water resource availability and quality.

The initiative WULCA launched by the UNEP/SETAC Life Cycle Initiative's working group on the assessment of water is a platform where the efforts on the development of methodologies related to water use in LCA are coordinated with the aim to build consensus and harmonize water accounting schemes and impact assessment methods. The first achievement of WULCA was the development of a general framework to assess freshwater resources within LCA (Bayart et al. 2010). This work provided recommendations on freshwater use modeling and relevant impact categories to be considered. The second chapter of my thesis represents a direct contribution to the second achievement of WULCA: a review of existing methods, including inventory, impact assessment methods and indices focused on water scarcity. The third achievement of WULCA consisted in broadening the understanding of existing water use impact assessment methods and their applicability within a water footprint study on a laundry product (Boulay et al. 2014a; Boulay et al. 2014b). In May 2013, WULCA received the mandate from the UNEP/SETAC Life Cycle Initiative to lead the harmonization and consensus building for the water use impact category in the context of the Project on Global Guidance on Environmental life cycle impact assessment indicators (Frischknecht and Jolliet 2014), involving key method developers and stakeholders through an international collaborative effort (UNEP SETAC Life Cycle Initiative 2014).

Parallel to this initiative, an ISO technical committee initiated a draft standard on water footprint in 2009. After a 5 year process, the final draft international standard ISO 14046 (ISO 2014) was accepted in May 2014 and published on August 1, 2014. Following the ISO 14040 and 14044 series (ISO 2006a; ISO 2006b), this new standard defines the principles, requirements and guidelines related to water footprint of products, processes and organizations based on LCA. It defines the scope, normative references, specific terms and definitions related to water, principles, methodological framework, reporting rules and critical review guidelines. The methodological framework distinguishes a "water footprint inventory assessment" from "a water footprint" that follows the four phases of LCA presented in Figure 1.1. According to this standard, a water footprint inventory assessment alone shall not be reported as a water footprint. The representation of a water footprint shall rather be one or more parameters which quantify the potential environmental impact(s) related to water, i.e. a water footprint unique indicator result related to one single impact category or the water footprint profile which comprises several indicator results. These indicator results can be related to three different types of assessments: (1) a water availability footprint (2) a water scarcity footprint, which corresponds to a water availability footprint considering only water quantity and (3) a water degradation footprint that corresponds to a water footprint that addresses water degradation. Two different impact pathways related to water quality degradation are considered in a water availability footprint and a water footprint addressing water degradation. The water availability footprint reflects the fact that water quality issues can be related to water quantity through the concept of functionality, introduced by

Boulay et al.'s (2011a). Boulay et al. (2011a) defined water as functional "if it can meet users' needs without generating adverse effects or a change in activities". A functionality loss can thus make water non-suitable for a defined type of user. On the other hand, a water footprint addressing water degradation covers environmental degradation associated with eutrophying, acidifying, and toxic emissions or thermal pollution but excludes the functionality loss.

Similarly to ISO 14040 and ISO 14044, ISO 14046 does not provide recommendations on which LCIA methods should be used for water assessment. The need to review and evaluate existing methods was fulfilled by the WULCA project, which succeeded to provide scientific arguments aiming at a future harmonization of the implementation of methods addressing water use in LCA. The work presented in chapter 2 of this thesis represents a direct contribution to the WULCA project, where the lack of comprehensive review and evaluation of existing state-of-the-art was addressed at the time of the study, from 2009 to 2013.

1.2.4 Water degradation through toxic emissions to water in LCA

Quality issues related to water degradation as mentioned in chapter 1.2.3 have been classified and streamlined in LCA through impact categories such as aquatic acidification, aquatic eutrophication, human toxicity and aquatic ecotoxicity (Jolliet et al. 2003). An overview of human toxicity and aquatic ecotoxicity is provided in the following paragraph.

As mentioned previously, the human toxicity and aquatic ecotoxicity impact categories refer to an adverse change in the structure, or function of respectively humans and aquatic species as a result of exposure to a chemical (Pennington et al. 2004). This impact is initiated by a natural or anthropogenic emission to air, water or soil of an organic or inorganic toxic substance. An emission inventory for the life cycle of a product can easily contain several hundred different substances, many of which have the potential to cause toxicity to humans or ecosystems when released to the environment (Henderson et al. 2011). The cause-effect chain following the emission was assessed alternatively by policy-based safe threshold data such as critical dilution volume (CDV) (Ecolabel EU 1995) and later by mechanistic model-based approaches such as the scientific consensus multimedia model USEtox (Rosenbaum et al. 2008). Policy-based approaches were developed to provide a conservative estimate of the impact to insure the safety of a receiving body and exposed population or ecosystem. Inversely, best estimate approach was adopted in LCA to compute more robust and stable metrics to perform product-based relative assessments. Figure 1.3 describes the 3-step assessment followed by both approaches to model the impact of toxic emissions. This model includes chemical fate, exposure and effect modeling steps (adapted from Rosenbaum (2008)). Multi-media models are traditionally used to simulate the fate of the chemical, i.e. the behavior of chemicals released from the technosphere, e.g., from a manufacturing facility or waste treatment plant, to the environment as the net result of mass flows between a suite of well-mixed, homogeneous compartments (Henderson et al. 2011). Once the substance is partitioned among different compartments, the substance exposure is modeled through the bioavailability of the chemicals to aquatic organisms for aquatic ecotoxicity and the transport from environmental compartments to the human population via inhalation and ingestion. Finally, the effect modelling then relies on dose or concentration response (eco)toxic data and expresses respectively the ultimate change in the affected fraction of an ecosystem or the change in disease probability affecting human lifetime.

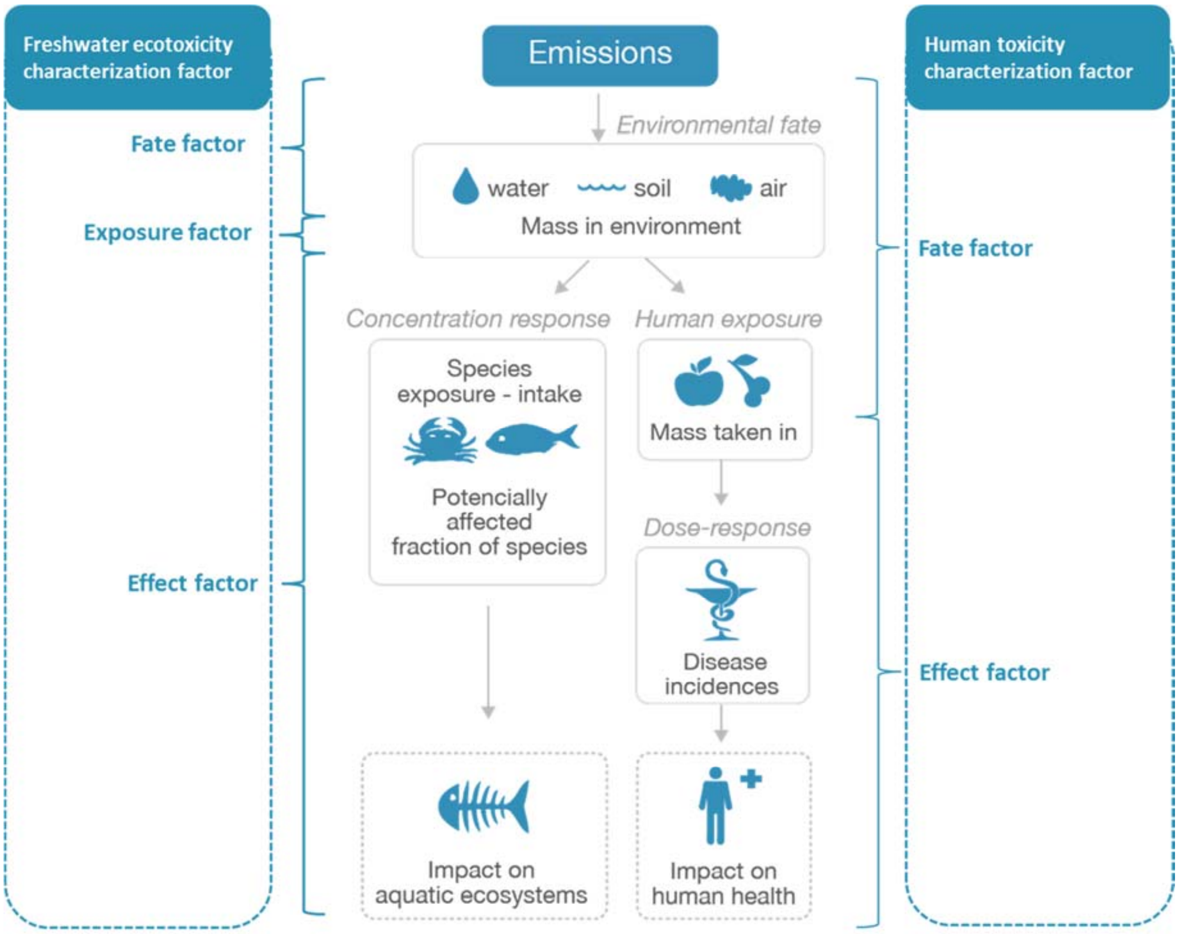

Figure 1.3: Framework of the scientific consensus USEtox model from emission to impact on aquatic ecosystems and human health, adapted from Rosenbaum et al. (2008)

The characterization factors for human toxicity and ecotoxicity encompass all steps of the cause-effect chain. It can be expressed in terms of metrics such as respectively Disability Ajusted Life Years (DALY) and the marginal change in potentially affected fraction (PAF) over a given area (m^2 or volume (m^3) and during a given period (day or year) per unit emission of a chemical (Goedkoop and Spriensma 2001; Huijbregts et al. 2001). The DALY is a measure of overall disease burden, expressed as the number of years lost due to ill-health, disability or early death. The DALY indicator was jointly developed by the World Bank, the World Health Organization (WHO) and the Harvard School of Public Health in the late 1980s (European Centre for

Disease Prevention and Control 2011) and is yet widely applied in the LCA field (Crettaz et al. 2002; Pennington et al. 2005; Goedkoop et al. 2009). On the other hand, the PAF indicator was developed within the LCA field as a measure of the toxic stress that substances put on ecosystems (Van De Meent 1999).

Equation 1.2 presents the characterization factor calculation through the multiplication of the fate FF, exposure XF and effect EF factors:

$$CF = FF.XF.EF$$

Equation 1.2: Human toxicity and ecotoxicity characterization factor calculation

Where:

FF is the fate factor that links the quantity released into the environment to the chemical masses (or concentrations) in a given compartment (Rosenbaum et al. 2011) through multimedia mass balance modeling (Mackay 2002). It accounts for multimedia and spatial transport between the environmental media (e.g., air, water, soil, etc.). For toxicity related categories, pollutants fate can be evaluated by multimedia and multi-pathway fate models e.g., BETR (Macleod et al. 2004), multimedia and multi-pathways fate and exposure models such as IMPACT Europe spatial and single zone (Pennington et al. 2005), USES-LCA (Huijbregts et al. 2000b) and GLOBOX (Wegener Sleeswijk and Heijungs 2010). A multimedia and multi-pathways exposure pathway model is needed because many chemicals are multimedia in nature, being transported from the medium of emission into another medium that either directly, or indirectly, results in the dominant exposure pathway of a species (Margni 2003). Compartment and place of emission, pollutant decay rate in different media, partitioning coefficients and bioaccumulation factors are important parameters considered in these models (Rosenbaum et al. 2011).

XF is the exposure factor that links the amount of chemical in a given environmental compartment to the chemical intake by human or chemical exposure by ecosystems. For human toxicity, exposure can be distinguished between direct intake (e.g., by breathing air and drinking water, etc.), indirect intake through bioconcentration processes in animal tissues (e.g., meat, milk and fish) and intake by dermal contact. For aquatic ecotoxicity, it is equal to the fraction of substance in dissolved form (Rosenbaum et al. 2008), the underlying hypothesis being that the ecosystem is exposed to the dissolved part of the chemicals reaching the freshwater. Although pollutant fate modeling is similar for human toxicity and aquatic ecotoxicity, these two cause-effect chains are influenced by different exposure parameters. For inhalation, the population density was identified as a key factor driving the intake, except for persistent and mobile chemicals that are taken in by the population independently from their place of emission (Rosenbaum et al. 2011). Inhalation, above-ground produce and fish were proved to be important exposure pathways for diffuse emissions driven by the key parameters population density, bioaccumulation /bioconcentration and dietary habits. The first two impact characterization steps, i.e. fate and exposure, can be expressed as a combined factor: the intake fraction as per the concept introduced by Bennett et al (2002).

EF is the effect factor that relates the level of exposure of a population or ecosystem to the (ultimate) damage. For human toxicity, it corresponds to the link between the quantity taken in via a given exposure route by a population to the adverse effects (or potential risk) generated by the chemical and the severity of disabilities caused by a disease in terms of affected life years. Cancer and non-cancer effects are considered separately as the amount of DALY as well as the related uncertainty caused by both types of diseases is different. The human toxicity effect factor estimation relies on toxicity dose-response tests extrapolated to humans. The aquatic ecotoxicity effect factor refers to the mean response of aquatic species to a chemical concentration increase in freshwater. It builds on available aquatic ecotoxicity of species belonging to different (typically three) phyla. In current methods, space is often neglected when assessing the effect factor for human toxicity and aquatic ecotoxicity factor (Pennington et al. 2006). The implications of this assumption are yet poorly evaluated, given needed data to assess this variability are lacking. It is therefore commonly accepted to neglect effects of spatial variability for human toxicity and aquatic ecotoxicity effect factors.

The USEtox model (Rosenbaum et al. 2008) is a scientific consensus and reference model for human toxicity and freshwater aquatic ecotoxicity. It is developed within the Life Cycle Initiative led by the UNEP/SETAC life cycle initiative, recognized as a state-of-the-art model by the European Commission (JRC-IES 2011)) and recommended in the Product/Organization Environmental Footprint guidelines (European Commission 2013a). This parsimonious and transparent model can screen about 3000 chemicals included in a published database and provides a freely accessible tool to calculate or revise characterisation factors for chemicals of interest.

However, the default version of USEtox is a generic model, i.e. there is no differentiation between various emission and impact locations. There is a need to evaluate the accuracy and precision of this consensus model while tackling the potential impact of emissions taking place in various locations with different hydrological properties. The next paragraph presents the state-of-the-art on spatial differentiation in LCA and more specifically of toxic impact modelling.

1.2.5 Spatial differentiation

In the present era of global trade and economy, product and organization life cycles usually include processes from all over the world. In LCI, spatial differentiation is relevant for most processes as habits and technologies are different around the world. In LCIA, spatial differentiation is relevant for all non-global impact categories, i.e., all categories for which the magnitude of impact depends on the location of the emission. Global warming and ozone depletion are global impact categories because their consequences are independent of the emission location. Other impact categories such as human and eco-toxicity, respiratory effects caused by inorganics, ionizing radiations, photochemical oxidation, terrestrial and aquatic acidification, eutrophication, land use, water use and noise often occur as regional or local impact (Potting and Hauschild 2006; Sedlbauer et al. 2007). Evaluating impacts in relation to the place of emission is important to reduce model uncertainty, improve accuracy, precision and confidence in LCA results. Spatial differentiation is recognized as a key step to improve its discriminatory power for comparative assessments (Potting and Hauschild 2006) and hot spot analysis.

Three levels of spatial differentiation were defined by Potting and Hauschild (2006): site-generic (generic receiving environment), site-dependent (distinguishing between classes of sources and determining their subsequent receiving environment) and site-specific (very detailed spatial differentiation is performed by considering sources at specific locations). Based on the same approach, Margni et al. (2008) added the distinction between archetype differentiation, geographic differentiation, and combined archetype-geographic approach. Humbert et al (2009) refined geographic differentiation based on distance from emission to location: “local” means within a few ten kilometers (e.g., the urban area), “regional” means within a few hundred kilometers (e.g., California, the Central valley), “continental” means within the continent (e.g., North America), and global means worldwide.

More recently developed impact assessment methods address the consequences of regional emissions for several impact categories. For example, regional characterization factors were developed to evaluate terrestrial acidification and eutrophication (Potting et al. 1998; Huijbregts et al. 2000a; Norris 2002; Seppälä et al. 2006; Posch et al. 2008; Roy et al. 2012), human toxicity (Macleod et al. 2004; Pennington et al. 2005; Shaked 2011), respiratory effects caused by primary and secondary particles (Humbert 2009; Humbert et al. 2011; Gronlund et al. 2014), and photochemical smog formation (Hauschild et al. 2006) as well as resources-related impact categories such as water use (Milà i Canals et al. 2008; Pfister et al. 2009; Motoshita et al. 2010; Boulay et al. 2011b; Berger et al. 2014). Depending on the impact category, the impact indicator variability ranges from two orders of magnitude when assessing generic emission inventories from different continents (Sedlbauer et al. 2007) up to 8 orders of magnitude at 2*2.5° (Humbert et al. 2009) or 10 orders of magnitude at the subwatershed resolution for emissions in water (Manneh et al. 2010). The project LC-IMPACT funded by the European Commission under the Seventh Framework Programme provided an important milestone regarding the coverage of spatially differentiated characterization factors for resources- and emission- related impact categories (Huijbregts 2010). IMPACT World+ is a new methodology released in 2014 that systematically integrates spatial differentiation for all covered impact categories (Bulle et al.

2012; IMPACTWorld+ 2014). The choice of resolution should consider both required differentiation from a scientific relevance standpoints as well as constraints from a practical standpoint (Sedlbauer et al. 2007). Indeed, the latter can be an issue given that (1) a large amount of geographical data is required e.g., meteorological, soil sensitivity, water scarcity, human development level data, etc. (2) storage of data and generation of characterization factors (CF) can create storage capacity issues (3) inventory databases and collected data need to offer the required level of details to make the methods applicable. Mutel and Hellweg (2009) address the latter point by proposing spatial methods to couple regionalized characterization factors with generic life cycle inventory databases in existing softwares. Currently, spatial differentiation starts to be implemented in commercial softwares, where the available geographical information of the Life Cycle Inventory (LCI) determines the required level of aggregation of characterization factors, e.g., the grid cell, country, continent of generic level. For instance, Pfister et al's (2009) water scarcity assessment method has been integrated in Simapro 8 (PRé consultants 2013). For the impact related to water use, the resolution of existing models can be down to the watershed scale (Hanafiah et al. 2011) or the 0.5°*0.5° grid cell scale (Pfister et al. 2009).

To assess human toxicity and aquatic ecotoxicity, multimedia and multi-pathway fate models are regionalized at different scales, from continental (Shaked 2011) to grid cells resolution, e.g., of 1*1° resolution (Pistocchi et al. 2010) with European coverage for the most refined ones. Several publications have quantified the variability linked to spatial inhomogeneity in multimedia modeling at national or regional scale. Impact indicators can vary by a factor of 5 to 10 between large geographical regions such as continents (Rochat et al. 2006) or up to 3 orders of magnitude for a higher spatial resolution e.g., ecological zones using a spatially-differentiated model at a continental level (Macleod et al. 2004) or 10 orders of magnitude at the subwatershed resolution for emissions to water (Manneh et al. 2010). Some publications recognized the necessity of including an archetype approach through an urban environment (Rosenbaum et al. 2008; Humbert et al. 2009) and indoor emissions (Hellweg et al. 2009) to better account for spatial fate and exposure inhomogeneities.

Although several approaches have been explored to model the (eco)toxicity cause-effect chain, there exists so far no model with global coverage and lower resolution than the sub-continental scale. Furthermore, simplified spatial differentiation approaches such as regional archetypes have not yet been explored for emissions to freshwater compartments with various hydrological key characteristics. Indeed, a simplified approach could prevent from shifting modeling uncertainty into inaccuracy in case regionalized methods are applied while geographical information is lacking.

1.3 Scope of work of this dissertation

1.3.1 Problem statement

During the last five years, the field of water use in LCA has experienced important development, both in terms of method (Pfister et al. 2009; Boulay et al. 2011b; Berger et al. 2014) and normative framework (ISO 2014). In light of these developments, the recommendations of the European Commission to evaluate the impact related to “water resource depletion” are outdated (European Commission 2013a) since they date from the state-of-the-art of 2008. For instance, the impact from water use is addressed through the impact category “resource depletion” that does not consider potential impact on human health and ecosystems. Furthermore, it provides impact scores in ecopoints, a unit developed specifically for the Swiss context given that it is normalized based on Swiss emissions, resource use and emission targets (Frischknecht et al. 2006) and was not dimensioned to be applied on a global scale. Existing case studies (Van Hoof et al. 2013; Boulay et al. 2014a) showed that different impact assessment methods could lead to results with opposite trends, leading to the need of a harmonized framework to evaluate the impact of water use in a consistent way. At the corporate level, several initiatives were recently set up to provide tools to measure water use, assist companies in developing water sustainable management or certification programs, such as respectively Global Water Tool by the World Business Council for Sustainable Development (WBCSD 2010), the CEO water mandate (UN Global Compact Office 2011), and the Alliance for Water Stewardship (AWS) (Abdel Al et al. 2014). Their scope is often reduced to inventory level reporting, while a wider fostering of impact assessment practices outside of the academic sphere would enhance the outreach of this type of assessment. There is a need to review, evaluate and organize the knowledge generated by existing methods in order to bridge recent academic developments and their implementation. Recommendations from the academic field on consensual methods for a water footprint are needed to support a sound normative framework and method harmonization relying on scientific arguments.

The methodological development of toxic emissions into water in a LCA context has been addressed since twenty years. The academic effort of the last decade focused on creating a scientific consensus model as a mimetic, parsimonious, transparent and evaluated combination of existing models. The outcome of this work was published in 2008 as the USEtox model (Rosenbaum et al. 2008), that is publically available and increasingly recommended (European Commission 2013a). Nevertheless, there is yet a need to improve the accuracy, precision and confidence in LCA result by addressing spatial differentiation, i.e. the differentiation of the impact magnitude for different locations. The latter practice increases model sophistication and thus raises several new questions and constraints regarding the implementation of spatially differentiated methods in LCA practice. Indeed, two key actors of the LCA field have stakes in new methodological development: method developers and practitioners, both following diverging objectives and constraints. On one hand, model developers aim to provide environmentally relevant models to depict a specific impact pathway from emission or resource extraction to the impact on human health, ecosystems or resources. This modeling is constrained by the availability of data of involved environmental, political and geographical parameters and results in a characterization factor that can be applied in a LCA study. On the other hand, practitioners use characterization factors that are implemented in commercial softwares where they are combined with generic inventory data as well as potential specific processes. The practitioner thus requires characterization factors that are compatible with existing and available inventory data, and that holds limited complexity to be able to interpret, explain and justify LCA results in the light of his knowledge of the inventory and impact characterization factor modeling. These two complementary roles meet on the fine line and trade-off between the model sophistication required for scientific relevance, the improvement of LCA discriminatory power if scientific relevance is increased, the applicability of developed characterization factors and the interpretability of LCA results. In the case of spatial differentiation, there is a need to test the relevance of the resolutions recommended by USEtox (urban, continent and global level) to evaluate emissions from global supply chains. Furthermore, exploring the likelihood to develop simplified approaches such as regional archetypes could prevent from complexifying LCA practice through the implementation of fully regionalized life cycle inventories and impact assessment without reducing the overall uncertainty originating from the handling of complex globalized product supply chains. A fully connected model with global coverage and higher resolution needs to be developed as a reference to test the validity of simplified approaches.

1.3.2 Research hypothesis

This thesis analyses the following research hypothesis:

The water footprint practice can be operationalized through (1) the definition of robust recommendations based on a scientific analysis of the current water use state-of-the-art methods that support a consensual normative framework and method harmonization and (2) the evaluation of the importance of spatial differentiation in state-of-the-art water degradation methods (USEtox in our case). The first point is addressed by performing a systematic review of the existing literature and the search for a consensus building. This second point is addressed by analyzing the performance of the scientific consensus USEtox model (using a generic or spatially differentiated continental landscape) compared to highly differentiated models assessing the impact of water degradation through toxic emissions to freshwater and integrating spatial differentiation.

1.3.3 Objective

The main objective of this work is to foster the application of water footprint by identifying good practices and improve its discriminatory power by developing spatially differentiated operational approaches. This research work focuses on the impact of quantitative water use as well as degradative water use of toxic emissions into water.

Specific objectives of this thesis are to:

1. Review existing and applicable inventory and impact assessment methods that address quantitative freshwater use in a life cycle perspective and provide preliminary recommendations for practitioners and for method harmonization:
    - Provide a comprehensive overview of existing and applicable inventory and impact assessment methods that address freshwater use in a life cycle perspective,
    - Analyse each method with a set of pre-defined criteria in order to highlight and understand similarities and differences,
    - Analyse key parameters to be considered in a consensus-based operational characterization method encompassing the WULCA framework (Bayart et al. 2010),
    - Formulate preliminary application recommendations for method developers and practitioners given current state-of-the-art, and
    - Discuss the implementation of spatial differentiation in methods addressing the impact of water use
2. Evaluate an appropriate model architecture (nested vs. spatially differentiated) and spatial resolution for the freshwater eco-toxicity and human toxicity impact categories in order to maintain environmental relevance while limiting model sophistication in terms of landscape data requirements:
    - Develop landscape parameters for USEtox to develop continent-specific boxes nested within the world,
    - Analyse the inter-continental variation of chemical fate and intake fractions among continents and examine the influence of the region(s) surrounding the considered sub-continent (for this, results from a nested USEtox model with continent-specific parameterization are compared to a fully connected model), and
    - Study intra-continental variation and develop archetypes for freshwater eco-toxicity and human toxicity exposure to ingestion of fresh water and fish, as a parsimonious surrogate to higher spatial resolution;

3. Explore further the importance and applicability of spatial resolution for toxic emissions into water by analysing and comparing the variability of characterization factors at the highly resolved grid cell level and different aggregation methods:
    - Develop a spatially resolved fate and exposure characterization model and factors for toxic emissions into water with global coverage at 0.5°*0.5° resolution,
    - Analyse the variation of fate and exposure factors for water ingestion as well as the main factors of influence on ecosystems and human exposure by focusing on five selected substances,
    - Develop archetypes based on the developed characterization model and identified key parameters, and
    - Compare practical solutions to apply spatial characterization factors aggregated at different scales in LCA for two emission patterns: population related emissions and sector-specific emissions into water related to global aluminium production.

1.3.4 Outline

Each of the following chapter of this thesis answers a specific objective in chronological order.

Chapter 2 presents the review of existing and applicable inventory and impact assessment methods that address freshwater use in a life cycle perspective performed as a deliverable for the WULCA project. Each method is analysed with a set of predefined criteria in order to highlight and understand similarities and differences. Key parameters to be considered in a consensus-based operational characterization method are identified to orient future LCA developments.

Chapter 3 investigates the spatial differentiation of toxic emissions to freshwater at the inter-continental and intra-continental scale. Landscape parameters for USEtox are developed at the continental and sub-continental level as specific boxes nested within the world. Inter-continental variation of chemical fate and intake fractions are then analysed among continents and the influence of the region(s) surrounding the considered sub-continent are examined. For this, results from a nested USEtox model with continent-specific parameterization are compared to a fully connected model. Moving to the watershed scale, intra-continental variation is analysed with a European coverage. As a parsimonious surrogate to higher spatial resolution, an archetype model is developed to discriminate between intra-continental emissions at the watershed level in Europe and evaluate freshwater eco-toxicity and human toxicity exposure by ingestion of freshwater and fish. Regionalized landscapes and characterization factors at the continental scale developed in this chapter will be integrated in the next release of the USEtox model as well as in the spatially differentiated methodology IMPACT World+.

Based on the outlook of this study, chapter 4 explores the spatial differentiation at a higher resolution through the development of a model at the 0.5°*0.5° resolution and global coverage. The variations of fate and exposure factors for water ingestion as well as the main factors of influence on ecosystems and human exposure are analysed. Archetypes are developed based on identified key parameters for fate and intake fraction. Practical solutions to apply spatial characterization factors are tested in a case study related to global aluminium production to discuss advantages and limitation of pushing further spatial differentiation in LCA: population-based and sector-specific emissions are evaluated and compared.

Chapter 5 concludes this thesis through a critical appraisal where the achievements are put in perspective as a contribution to LCA science and practitioners. In particular, limitations and constraints related to water footprint practice are discussed based on published case studies and personal experience. The outcome of chapters 3 and 4 are highlighted and examined compared to the current spatial differentiation implementation state-of-the-art. A summary of achieved results is then provided.

Chapter 2

Review of methods addressing freshwater use in life cycle inventory and impact assessment

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Abstract

In recent years several methods have been developed that propose different freshwater use inventory schemes and impact assessment characterization models considering various cause-effect chain relationships.

This work reviewed a multitude of methods and indicators for freshwater use potentially applicable in life cycle assessment (LCA). This review is used as a basis to identify the key elements to build a scientific consensus for operational characterization methods for LCA. This evaluation builds on the criteria and procedure developed within the International Reference Life Cycle Data System Handbook and has been adapted for the purpose of this project. It therefore includes: (1) description of relevant cause-effect chains, (2) definition of criteria to evaluate the existing methods, (3) development of sub- criteria specific to freshwater use, and (4) description and review of existing methods addressing freshwater in LCA.

No single method is available that comprehensively describes all potential impacts derived from freshwater use. However, this review highlights several key findings to design a characterization method encompassing all the impact pathways of the “WULCA” framework:

- In most of databases and methods, consistent freshwater balances are not reported, either because output is not considered, or because polluted freshwater is recalculated based on a critical dilution approach.
- At the midpoint level, most methods are related to water scarcity index, and correspond to the methodological choice of an indicator simplified in terms of number of parameters (scarcity) and freshwater uses (freshwater consumption or freshwater withdrawal) considered. More comprehensive scarcity indices distinguish different freshwater types and functionalities.
- At the endpoint level, several methods already exist that report results in units compatible with traditional human health and ecosystem quality damage, and cover various cause-effect chains, e.g., the decrease of terrestrial biodiversity due to freshwater consumption.
- Midpoint and endpoint indicators have various levels of spatial differentiation, i.e., generic factors with no differentiation at all, or country, watershed and grid cell differentiation.

Existing databases should be (1) completed with input and output freshwater flow differentiated according to water types based on its origin (surface water, groundwater and precipitation water stored as soil moisture), (2) regionalized and (3) if possible characterized with a set of quality parameters. The assessment of impacts related to freshwater use is possible by assembling methods in a comprehensive methodology to characterize each use adequately.

Keywords

Life cycle assessment - freshwater use – method review – human health – ecosystem quality – resources

2.1 Introduction

Water is a vital natural resource for all ecosystems, human wellbeing and many economic activities. Because of the combination of population growth and economic development leading to increasing human freshwater use (Vörösmarty et al. 2000b) and enhanced climate change effects on the global water cycle, water scarcity is becoming an increasing environmental concern. Although freshwater is a local resource, water scarcity is leading to the threat of a global water crisis, with a large share of global population being affected (World Water Assessment Programme UN 2009). Given actual estimates of global freshwater consumption around 2600 km3/y and a proposed planetary boundary of 4000 km3/y consumptive surface and groundwater use (Rockström et al. 2009), it appears that humanity's freshwater use is currently within the safe operating limit (Alcamo et al. 2007; Shen et al. 2008). Other sources estimate the actual water withdrawal as less than 10% of the maximum available renewable freshwater resource (Oki and Kanae 2006). However, when considering the regional nature of freshwater scarcity, the majority of global freshwater withdrawals currently takes place in watersheds already experiencing high water scarcity (Ridoutt and Pfister 2010a). According to Ridoutt and Pfister (2010a), humanity's water footprint (referred as the sum of withdrawals multiplied by local water stress indices) must be globally reduced by approximately 50% to achieve a sustainable water use. The strong bond between water use and other global environmental and societal systems at various spatial scales such as land use, climate change and demographic developments justifies both global and regional perspectives for water management to tackle water related problems (Hoff 2009; Hoekstra 2010).

To tackle this major environmental concern, various initiatives were recently launched in order to develop and standardize analytical tools to measure and assess freshwater use at regional and global scale and to improve the overall management of freshwater resources as well as the overall environmental performance of products and operations. Among these initiatives are the Water Footprint Network (WFN) (Hoekstra et al. 2011), the International Organization for Standardization (ISO) water footprint working group (ISO 2014), and the World Business Council for Sustainable Development (WBCSD 2010) who launched the Global Water Tool and the UNEP/SETAC Life Cycle Initiative's working group on the assessment of freshwater use and consumption in life cycle assessment (LCA) called "WULCA" (Koehler and Aoustin 2008). These initiatives also aim to set up public-private partnerships to assist companies in the implementation of water sustainability policies (UN Global Compact Office 2011) and to develop certification programs (Abdel Al et al. 2014).

The authors of this article are part of the UNEP/SETAC Life Cycle Initiative's WULCA working group (Koehler and Aoustin 2008), which involves academic and industrial partners from around the globe that cooperate on the development of methodologies related to freshwater use from a life-cycle perspective, including both appropriate freshwater accounting schemes and impact assessment methods. Guidance is provided as scientific consensus regarding the consideration of freshwater in life cycle inventory (LCI) and the choice of life cycle impact assessment (LCIA) methods. Quantitative comparison of LCIA methods will be provided as next step of this work. The working group's deliverables are also used as methodological input to the ISO 14046 water footprint standardization process. A prominent achievement of WULCA was a general framework for the consideration of freshwater resources within LCA (Bayart et al. 2010). This work provided recommendations on freshwater use modeling and relevant impact categories building on the achievements of Phase 1 of the UNEP/SETAC Life Cycle Initiative (Bauer et al. 2007) and the conceptual framework including first indicators introduced by Owens (2002).

In the past, most LCA studies did not consider freshwater use and LCI databases reported freshwater use inventory by determining the total freshwater input from nature or respective technical systems (e.g., drinking water networks) while generally neglecting the water outputs from the LCA system under study (Koehler 2008). LCIA methods applied the amount of freshwater used without characterization factor to address the impacts. Recently, new methodologies were developed that propose freshwater use inventory schemes (WBCSD 2010; Peters et al. 2010; Boulay et al. 2011a) and assess the potential environmental impacts of freshwater use considering various cause-effect relationships (Milà i Canals et al. 2008; Pfister et al. 2009; Motoshita et al. 2010).

A selection of scientific methods for freshwater use assessment in LCA was evaluated by Berger and Finkbeiner (2010) regarding the methods' scope, input data requirements, and the ISO compliance summarizing the methodological differences. Considering the latter study, the WULCA working group has performed an extensive analysis of a broader variety of freshwater use assessment schemes and metrics applied both in the field of life cycle assessment and water management. In contrast to Berger and Finkbeiner (2010), this work employs a detailed and systematic analysis to understand differences and similarities in modeling choices using a comprehensive set of evaluation criteria including scientific robustness, transparency and reproducibility, applicability, the level of documentation, and stakeholder acceptance. It is based on the International Reference Life Cycle Data System (ILCD) (JRC-IES 2011).

The goal of the current method review is to provide:

1. A comprehensive overview of existing and applicable inventory and impact assessment methods that address freshwater use in a life cycle perspective

2. An analysis of each method with a set of pre-defined criteria in order to highlight and understand similarities and differences
3. An analysis of key parameters to be considered in a consensus-based operational characterization method encompassing the WULCA framework (Bayart et al. 2010) and
4. Preliminary application recommendations for practitioners given current state-of-the-art.

This study comprises methods for inventorying the use of different freshwater resources as well as for assessing the associated impacts. Methods assessing specific impact of pollutants, i.e., aquatic ecotoxicity, human toxicity, aquatic eutrophication and aquatic acidification, as well as the recent method dealing with impacts of changed freshwater temperatures due to cooling freshwater discharges (Verones et al. 2010) are not included in this work as they are generally assessed in conventional impact categories of LCA or oriented towards quality related impact.

2.2 Method

The review scheme adopted relies on the approach taken by the European Commission within the International Reference Life Cycle Data System (ILCD) defining the "Framework and requirements for LCIA models and indicators" (JRC-IES 2011). The following procedure was followed for the methods review: (1) description of relevant cause-effect chains, (2) definition of criteria to evaluate the existing methods, (3) development of sub-criteria specific to freshwater use, and (4) description and review of existing freshwater use assessment methods.

2.2.1 Description of relevant cause-effect chains

Figure 2.1 depicts the cause-effect chains that link freshwater type and use to potential impacts at the midpoint and endpoint level and ultimately to the related area of protection human health, ecosystem quality, and resources (Jolliet et al. 2003). The identified cause-effect chains serve as basis for the development of specific criteria linked to freshwater use. In nature, precipitation water (liquid or solid) is differentiated in three types of water that are interconnected: (1) surface water (river, lake and sea) (2) groundwater (renewable, shallow and deep) that is only reached through surface water and soil moisture and (3) precipitation water stored as soil moisture (also called "green water") (Falkenmark and Rockström 2006). Fossil groundwater compartment is not connected to other freshwater compartments. Freshwater is characterized by less than 1,000 milligrams per liter of dissolved solids (United Stated Geological Survey (USGS) 2012) and encompasses all previously mentioned three types. The impact of freshwater use is related to (1) consumption of one of these water types and (2) withdrawal of one of these water types and release of surface water. Impact of degradative use is considered, as withdrawal of surface or groundwater at given quality followed by release at another quality. However, impact of direct pollutant release in freshwater and resulting cause-effect chains are excluded from the scope of this study, in which there is no value judgment regarding the inclusion of degradative use in considered methods. Related impact assessment approaches are assessed in the ILCD handbook (JRC-IES 2011). Land occupation and transformation, as well as rainwater harvesting is a driver for change in surface water and precipitation water stored as soil moisture. The availability of the latter water type leads to debated potential impacts that are not considered in this work. However, the modification of the hydrological balance following land transformation or occupation is accounted for in the present framework as it corresponds to a modification of the amount of water that reaches groundwater and surface water (equivalent to a consumption of the corresponding water).

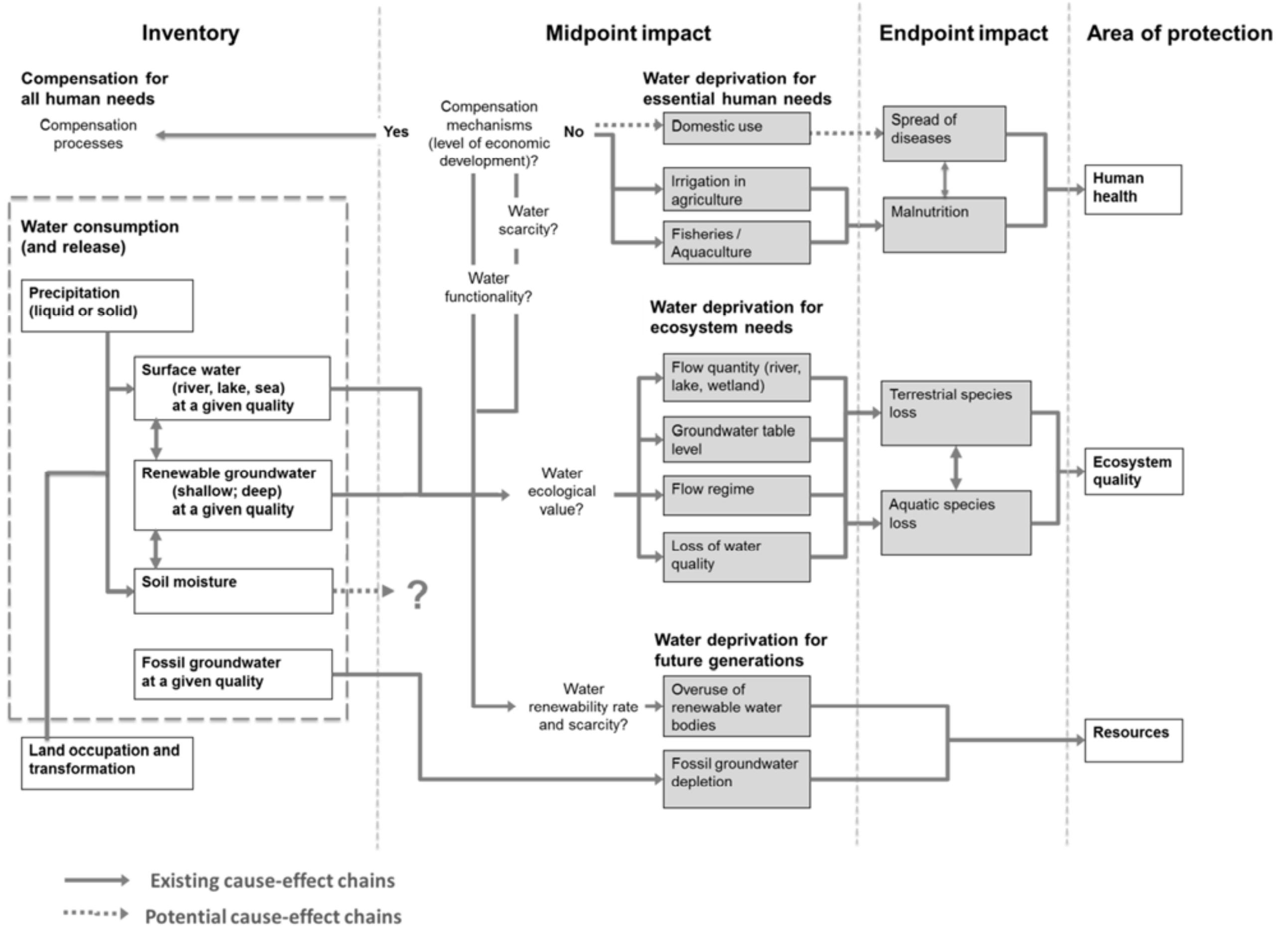

Figure 2.1: Cause-effect chains leading from the inventory to the areas of protection human health, ecosystem quality and resources (adapted from Bayart et al.(2010))

The use of freshwater can generate potential impacts to humans, ecosystems and resources. These impacts can be related to water scarcity, water functionality, water ecological value and water renewability rate and are influenced by the possibility to develop compensation mechanisms. Water scarcity is defined in this work being the water use approaching or exceeding the natural regeneration of water in a given area, e.g., a drainage basin. In this article, water scarcity is considered as a parameter leading to freshwater deprivation by limiting freshwater availability. Freshwater quality is defined as a set of parameters considered to characterize the chemical, physical and biological properties of freshwater. It is related to a functionality approach, which assesses to which users the freshwater withdrawn and released is functional (Bayart et al. 2010) and can also lead to water deprivation when water of a given quality is not available anymore for specific users. Water ecological value describes the physical relation to and dependency of ecosystems on freshwater (Bayart et al. 2010). Water renewability rate is the natural rate at which the resource is re-charged. Compensation mechanisms refer to the use of backup technologies by human users deprived of “functional” freshwater to meet their needs (Boulay et al. 2011b).

Human health: The way human health is affected by freshwater use depends on the level of economic development and welfare (Bayart 2008; Boulay et al. 2011b). If this is sufficient, the lack of freshwater will be compensated by the development of back-up technologies (such as desalination or the import of water-intensive goods as virtual water (Allan 1996)). These compensation activities need to be assessed with a new inventory and can in turn lead to environmental impacts via other interventions involved in the compensation activities (e.g., climate change impacts caused by energy consumption for desalination). If the level of economic development is not sufficient to cover these costs, freshwater use will lead to water deprivation for primarily three functions which fulfill essential human needs depending on local conditions: domestic use (hygiene and ingestion), agriculture and aquaculture/fisheries. Industrial functions of freshwater close to human essential needs (e.g., house building, provision of pharmaceuticals) are not considered in this framework, because they are more likely to consider compensation strategies rather than suffering from freshwater deprivation (Boulay et al. 2011b). Water quality degradation leads to water deprivation when it creates a loss of functionality for users who need water at a higher quality level than the released one. Users that are able to use freshwater at that or a lower quality level won’t be deprived. The extent of water quality degradation depends on the amount and intensity of chemical, biological and thermal pollution withdrawn and is related to the sanitation capacity. The withdrawn freshwater represents an adverse impact depriving users from a given amount of water at ambient water quality; the released freshwater (negative LCI flow) results in a burden reduction by making available the same amount of water for users capable to use water at that quality. Current models agree that the way human health is affected by water use depends on the level of economic development and welfare. They acknowledge that under given conditions, water use can lead to deprivation for essential human needs such as agriculture, fisheries and domestic use and ultimately to malnutrition and spread of diseases. However, there is currently not sufficient information to determine whether freshwater use in a low-income water-stressed region would lower water availability for domestic users or rather only affect other users (e.g., agricultural, fisheries or industries (Boulay et al. 2011b).

Malnutrition and spread of diseases are interconnected, i.e., malnutrition could for example make a person more vulnerable to the spread of diseases, and reciprocally, some enteric diseases could affect the ability to absorb nutrients and thus contribute to malnutrition. Freshwater use ultimately leads to an aggregated impact on human health, generally expressed in disability-adjusted life years (DALY) (Pfister et al. 2009; Motoshita et al. 2010; Motoshita et al. 2011; Boulay et al. 2011b).

Ecosystem quality: Water use can also affect the ecosystem, for instance by changes in the river, lake or wetland flow quantity (e.g., due to surface water withdrawals), changes in the level of the groundwater table (e.g., due to groundwater withdrawal), changes in flow regimes (e.g., due to turbined water use) and loss of freshwater quality. Similarly to human health, degradation corresponds to the consumption of freshwater of a higher quality (with a higher ecological value, or ecological functionality) and the release of freshwater of lower quality (with a lower ecological value, thus affecting all the ecological users needing a better water quality but not the users able to deal with a lower quality).

It should be noted that the latter cause-effect chain is related to the deprivation of freshwater of a given quality and not to the aquatic ecotoxicity, aquatic eutrophication and aquatic acidification impact of this degradation. The midpoint impacts related to freshwater deprivation, which depend on water scarcity and water quality, eventually lead to species diversity change in aquatic and terrestrial ecosystems. The extent of these changes depends on the ecological value of water in the considered ecosystem. Ultimate impacts on ecosystem quality are commonly expressed in potentially disappeared fraction of species (PDF) on given surface or volume during a given time (PDF·m2·y or PDF·m3·y) (van Zelm et al. 2011; Hanafiah et al. 2011).

Milà i Canals et al. (2008) suggest that changes caused by production systems on the amount of rainwater available to other users (ecosystems) through changes in the fractions of rainwater that follow infiltration, evapotranspiration and runoff should be included as impacts on ecosystem quality. This is closely linked to the impact of land occupation and transformation on green water availability through the variation of stock of water stored as soil moisture available for plant uptake (“green water”).

Resources: Consumption of all freshwater types as well as withdrawal and release of fossil groundwater can respectively lead to overuse of renewable water bodies or exhaustion of non-renewable fossil groundwater. Overuse of renewable water bodies depends on the water renewability rate. These midpoint impacts affect water flows and funds and ultimately have an effect on the resources stock. This reduction of available water affects other cause-effect chains by increasing local water scarcity. Different approaches exist to characterize the impact on resources encompassing the abiotic depletion potential given in antimony equivalents (Sb-eq) (Milà i Canals et al. 2008) at the midpoint level, and the backup-technology concept expressing the resource damage in megajoules (MJ) surplus energy (Pfister et al. 2009) or exergy-based methods given in megajoules of exergy (MJex) (Bösch et al. 2007) at the endpoint level.

2.2.2 Definition of criteria to evaluate the existing methods

Five scientific (1-5) criteria and one potential stakeholder acceptance (6) criterion based on the ILCD Handbook (JRC-IES 2011) were adopted within this review: (1) completeness of scope, (2) environmental relevance, (3) scientific robustness and certainty, (4) documentation, transparency and reproducibility, (5) applicability and (6) degree of potential stakeholder acceptance and suitability for communication in business and policy contexts. They are further described in Table A.5 in the appendix.

2.2.3 Development of sub-criteria specific to freshwater

Additionally to the six criteria mentioned above, sub-criteria specific to freshwater use were added in the criteria “completeness of scope” and ”environmental relevance” as described in Table 2.1. For the former, sub-criteria were needed to identify which areas of protection are considered by the existing methods and which midpoints and endpoints are modeled. For the latter, sub-criteria were needed to evaluate the coverage of relevant freshwater-specific cause-effect chains as depicted in Figure 2.1. The level of coverage was assessed without weighting the relative importance of different cause-effect chains and related parameters, but rather by exploring how far and with which method this coverage has been performed.

Table 2.1: Specific sub-criteria used to characterize inventory, midpoint and endpoint modeling

Criteria		Sub-criteria	Relevant modelling aspect
Completeness of scope		Midpoint: which impact mechanisms are covered by the impact indicators for the midpoint affecting the area of protection human health?	Water deprivation for: - Domestic use - Irrigation in agriculture (agricultural use) - Fisheries / aquaculture
		Midpoint: which impact mechanisms are covered by the impact indicator for the midpoint affecting the area of protection ecosystem quality?	- Changes in flow quantity (river, lake, wetland) - Changes in groundwater table level - Change in flow regimes - Loss of water quality
		Midpoint: which impact mechanisms are covered by the impact indicator for the midpoint affecting the area of protection resources?	- Overuse of renewable water bodies - Fossil groundwater exhaustion
		Endpoint: which impact mechanisms are covered by the endpoint indicator affecting the area of protection human health?	- Spread of diseases due to midpoint impact on domestic use - Malnutrition due to midpoint impact on irrigation and fisheries /aquaculture
		Endpoint: which impact mechanisms are covered by the endpoint indicator affecting the area of protection ecosystem quality?	- Terrestrial species loss - Aquatic species loss
		Endpoint: is the endpoint indicator affecting the area of protection resources covered?	
Environmental relevance	**Water type in nature**	What types of water are considered?	- Surface water (river, lake, sea) - Groundwater (renewable, fossil, shallow, deep) - Precipitation water stored as soil moisture
		Are consumption and water release considered?	
		Inventory: is intake and released water quality considered?	
	Cause-effect chain	Midpoint/endpoint cause-effect chain affecting area of protection human health: is **water scarcity** taken in account?	
		Midpoint/endpoint cause-effect chain affecting area of protection human health: are **water functionalities** of the water resource taken in account?	
		Midpoint/endpoint cause-effect chain affecting area of protection human health: are **economic development level** and **compensation mechanisms** taken in account?	
		Midpoint/endpoint cause-effect chain affecting area of protection ecosystem quality: is **water ecological** value taken in account?	
		Midpoint/endpoint cause-effect chain affecting area of protection resources: is **water scarcity** taken in account?	
		Midpoint/endpoint cause-effect chain affecting area of protection resources: is **water renewability rate** taken in account?	

2.2.4 Description and review of existing freshwater use assessment methods

Various methods have been developed to evaluate freshwater use in LCA. Many of them were already published or in the process of being published. All methods addressing freshwater use supported by sufficient documentation to be analyzed, i.e., a draft article, a report, etc. were considered in this paper. Unpublished methods were assessed regarding the latest information available in June 2012. Figure 2.2 summarizes the reviewed methods and classifies them at the inventory level, at the water index level, or at the impact assessment level, distinguishing between midpoint and endpoint assessments. It identifies those specifically addressing one area of protection or more comprehensive methods that address more than one area of protection. Databases are called according to the database name and methods according to the name of the developer for academic work, e.g., Boulay et al. (Boulay et al. 2011b) or the industry for methodology developed within a company e.g., Veolia (Bayart et al. 2014).

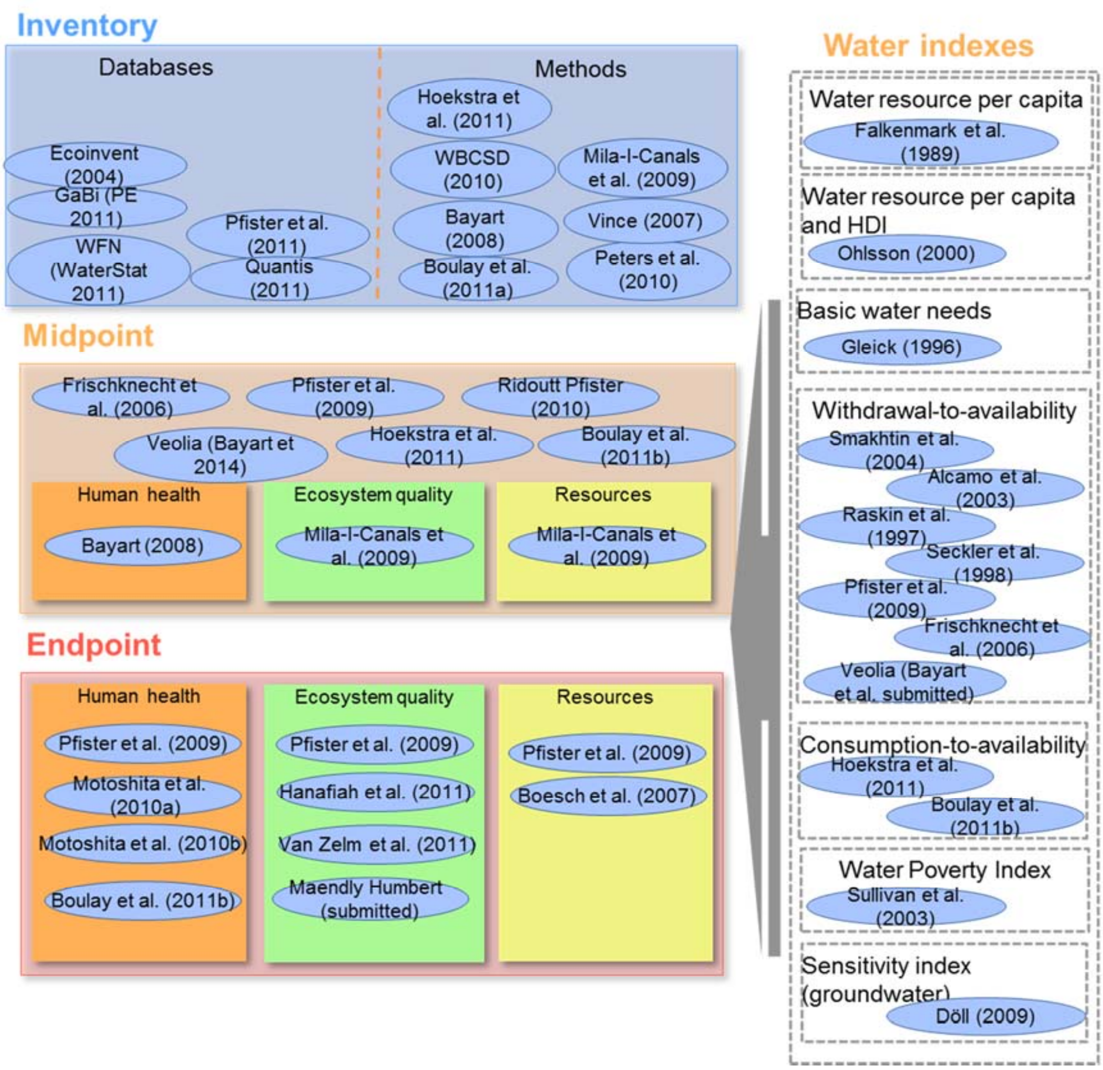

Figure 2.2: Scope of and relationship between the available freshwater use inventory, and impact assessment methods with classification for the three areas of protection

A short description of assessed methods is provided in the appendix A.2.

Inventory databases: The inventory section contains both inventory databases and inventory methods. The ecoinvent database (Frischknecht et al. 2005) and GaBi database (PE 2011) are widely used databases and contain elementary flows for freshwater withdrawal and turbined water. The WFN database (Water Footprint Network 2011) assesses the inventory consumptive and degradative flows of crops and derived crop products, farm animals and animal products, biofuels, national consumption and production as well as trade in crop, animal and industrial products according to the WFN method (Hoekstra et al. 2011). Pfister et al.'s database (Pfister et al. 2011) assesses the freshwater consumption for the production of 160 crops. An additional source of data for consumptive and evapo-transpirative use can be found for five crops and three livestock products (Hanasaki et al. 2010). The Quantis water Database (Quantis 2012) is a database of water uses based on ecoinvent 2.2 developed in the aim of providing industrial stakeholders with datasets required to apply all existing impact assessment methods.

Inventory methods: Inventory methods generally suggest concepts for a systematic classification of fresh-water elementary flows according to their type (surface water, groundwater, precipitation water stored as soil moisture, whether intake water quality is considered, etc.) without providing respective data. Inventory methods also describe technical water flows such as cooling water and irrigation water. The reviewed inventory methods differ widely in their objective and level of detail. Some focus on defining water categories to allow quality to be considered (Vince 2007; Bayart 2008; Boulay et al. 2011a)), others on providing inventory tools for organizations (WBCSD 2010; Hoekstra et al. 2011), integrating the effects of direct water use and of land occupation and transformation on water availability in a comprehensive methodology (Milà i Canals et al. 2008) or providing detailed hydrological modeling and classification of freshwater use data in specific sectors (e.g., Australian red meat sector) (Peters et al. 2010). Boulay et al. (2011a) is built on Vince's (2007) and Bayart's (2008) method.

Midpoint assessment methods: Midpoint impact assessment methods give either an indicator common to all areas of protection or an indicator specific to a defined area of protection. Methods covering all area of protections giving a single index related to water scarcity include the Swiss ecological scarcity (Frischknecht et al. 2006), Pfister et al. (2009), Ridoutt and Pfister (2010b), Water Impact Index of Veolia (Bayart et al. 2014), Boulay et al. (2011b) methods and Water Footprint impact indices (Hoekstra et al. 2011). Area of protection specific midpoint indicators describe the impact pathway leading to a decrease in freshwater availability for contemporary human users (Bayart 2008), as well as changes in freshwater availability for ecosystems leading to freshwater ecosystem impacts (Milà i Canals et al. 2008) and changes in groundwater availability causing freshwater depletion (Milà i Canals et al. 2008). Milà i Canals et al. (2008) suggest to use different types of water indices (Falkenmark et al. 1989; Raskin et al. 1997; Smakhtin et al. 2004) to assess freshwater ecosystem impacts. Falkenmark et al.'s (1989) index focuses on human use by evaluating the fraction of the total annual runoff available for human use. Raskin et al. (1997) use a water use per resource refined by Smakhtin et al. (2004) by subtracting environmental freshwater requirements from the available resources to derive a water index focused on freshwater resources available for human use.

The overall "blue-green-grey water" footprint concept of Hoekstra et al. (2011) was generally classified as an inventory metric given that precipitation water stored as soil moisture evapotranspirated by plants ("green water footprint") and consumptive use of surface and groundwater ("blue water footprint") represent physical metrics and are not further characterized. However, the "grey water footprint" can also be evaluated as a midpoint approach as grey water footprint denotes degradative freshwater use by characterizing the chemical pollution in water similar to "the critical dilution volumes approach" , i.e., an equivalent amount of water needed to dilute an emission below an acceptable threshold. This method thus juxtaposes measurable inventory results of "blue" and "green water footprint" with a theoretical volume of "grey water" which corresponds to a characterized inventory results. Using the term "grey water" also creates the problem of having two competing definitions of this term circulating in the water industry (Henriques and Louis 2011).

Endpoint assessment methods: Endpoint impact assessment methods provide specific indicators for potential damages on the areas of protection human health (Pfister et al. 2009; Motoshita et al. 2010; Motoshita et al. 2011; Boulay et al. 2011b), ecosystem quality (Maendly and Humbert 2009; Pfister et al. 2009; van Zelm et al. 2011; Hanafiah et al. 2011) and resources (Bösch et al. 2007; Pfister et al. 2009).

Other approaches exist to estimate impact on resources that attempt to account for the emergy flows put into place by natural processes to make available a given resource at a given state (Zhang et al. 2010; Rugani et al. 2011) but are not evaluated in this review because they are not specific to the characteristics of freshwater resource. Emergy is defined as the measure of both the work of nature and that of humans in generating products and services, i.e., a record of previously used-up available energy that is a property of the smaller amount of available energy in a transformed product (Odum 1996).

Water indices: Water indices are originally non LCA-based indicators that express a measure of human and environmental water needs or of the fraction of resource available to meet these needs. Water indices can be used as characterization factors for midpoint (Falkenmark et al. 1989; Raskin et al. 1997; Smakhtin et al. 2004) and endpoint (Sullivan 2002; Döll 2009) impact assessment methods when applied to freshwater consumptive or degradative use. Such indices can be considered as human use oriented (Falkenmark et al. 1989; Gleick 1996; Seckler et al. 1999; Ohlsson 2000; Sullivan 2002; Döll 2009), ecosystem use oriented

(Smakhtin et al. 2004), or cover all three areas of protection (Raskin et al. 1997; Frischknecht et al. 2006; Alcamo et al. 2007; Pfister et al. 2009; Hoekstra et al. 2011; Boulay et al. 2011b; Bayart et al. 2014). In this work, the terminology “water scarcity index” is related solely to withdrawal-to-availability ratio (Raskin et al. 1997; Seckler et al. 1999; Smakhtin et al. 2004; Frischknecht et al. 2006; Alcamo et al. 2007; Pfister et al. 2009; Bayart et al. 2014) or consumption-to-availability ratio (Hoekstra et al. 2011; Boulay et al. 2011b). Water scarcity indices can be based solely on a measure of water scarcity or include additionally a measure of water quality (Boulay et al. 2011b; Bayart et al. 2014). The details of the implementation of water indices in an LCA context, i.e., the water type to be considered in the inventory phase, needs to be specified in order to make water indices applicable in a method.

Uncertainty: Uncertainties are generally large in life cycle impact assessment, especially on the endpoint level and are yet generally not quantified in most of methods. Only a few authors, i.e., Pfister and Hellweg (2011) reported uncertainties for human health and WSI indicators on watershed and country level.

2.3 Results and discussion on method evaluation and cross comparison

The methods were evaluated and compared according to the selected criteria and sub-criteria displayed in Table 2.1 and Table A.5 at the inventory, midpoint, and endpoint level and key differences were identified. Table A.1, Table A.2 and Table A.3 in the appendix provide a summary of the review for each method.

Inventory databases

While the ecoinvent (Frischknecht et al. 2005), GaBi (PE 2011) and Quantis (Quantis 2012) databases give the opportunity to distinguish freshwater input as water withdrawal according to its natural source (surface water (river, lake) or groundwater (renewable, fossil)), in the WFN database (Water Footprint Network 2011) and Pfister et al.’s data sets (Pfister et al. 2011), water input is restricted to consumption of precipitation water stored as soil moisture evapotranspirated by plants (so-called “green water footprint”) as well as consumption of surface and groundwater combined (so-called “blue water footprint”). All datasets consider water outputs and global water balances in a different manner. The ecoinvent datasets in their current version 2.2 do not allow the determination of water balances because water releases are not reported, water consumption being thus an unknown part of the withdrawal. In contrast, GaBi and Quantis databases contain water inputs and outputs for all foreground and background processes. The WFN database (Water Footprint Network 2011) considers volumetric estimations of water consumption through “blue” and “green” water footprint, while degradative use is expressed through the “grey water” concept, where pollutant persistence, inter-compartment transfer and bioaccumulation properties are only implicitly included in water quality standard definitions which exist for a reduced set of substances. Only the Quantis water

Database considers water evaporated from reservoirs. The WFN database, the GaBi database, the Quantis water Database, and Pfister et al.'s datasets provide regionalized data per country where appropriate in regards to the product (global commodities or region-specific products).

Inventory methods

The water flow classification of Boulay et al. (2011a), the Global Water Tool of the WBCSD (2010) distinguish water according to its origin (e.g., surface, groundwater) and account for water balances by using input-output inventories. Milà i Canals et al. (2008), Peters et al.'s (2010), and the WFN method (2011) account only for consumptive water use of soil moisture lost by evapotranspiration ("green water") as well as evaporated surface and groundwater flows. Boulay et al.'s method, which is an upgraded version of Vince's (2007) and Bayart's (2008) method is more comprehensive, as it enables to classify 11 input and output water inventory flows by using corresponding water quality classes based on 137 parameters.

Midpoint assessment methods

Water indices used in midpoint methods are based on a withdrawal-to-availability ratio (Frischknecht et al. 2006; Milà i Canals et al. 2008; Pfister et al. 2009; Ridoutt and Pfister 2010b; Bayart et al. 2014) or a consumption-to-availability ratio (Hoekstra et al. 2011; Boulay et al. 2011b). They are used as a characterization factor (CF) for freshwater use in life cycle impact assessment to assess the impact of water consumption (Frischknecht et al. 2006; Milà i Canals et al. 2008; Pfister et al. 2009; Ridoutt and Pfister 2010b; Hoekstra et al. 2011; Boulay et al. 2011b; Bayart et al. 2014) and water degradation (Ridoutt and Pfister 2010b; Hoekstra et al. 2011; Boulay et al. 2011b; Bayart et al. 2014). Ridoutt and Pfister's (2010b) index is an extended version of Pfister et al.'s approach (2009) given that degradative water use ("grey water") is included additionally to consumptive use ("blue water consumption"). The Water Footprint Network's impact indices ("green", "blue" and "grey water" footprint impact index) (Hoekstra et al. 2011) follow the same concept by applying "blue", "grey" and "green" water scarcity indices to corresponding water categories. The Water Impact Index (Bayart et al. 2014) and Boulay et al.'s (2011b) index both include water quality as a parameter additionally to water scarcity considering that water quality parameters could restrict its use by humans and the natural environment as defined in Figure 1. Storage capacity has been considered in Pfister et al. (2009) as it is strongly related to water deprivation (deprivation occurs only if storage capacity is insufficient or if much of the stored water is evaporated).

Most of the methods provide characterization factors differentiated by country (Frischknecht et al. 2006; Bayart 2008; Milà i Canals et al. 2008; Pfister et al. 2009; Ridoutt and Pfister 2010b; Boulay et al. 2011b), watershed (Frischknecht et al. 2006; Pfister et al. 2009; Hoekstra et al. 2011; Boulay et al. 2011b) or grid cell (Pfister et al. 2009; Ridoutt and Pfister 2010b). The Water Footprint Network blue water footprint impact indices provide characterization factors with monthly temporal differentiation (Hoekstra et al. 2011) and thus offer more temporal precision for impact evaluation. However, storage of water is not included. Bayart et al.'s (2014) and Mila I Canals et al.'s (2008) methods on freshwater depletion do not provide regionalized characterization factors.

Endpoint assessment methods

Human health: The impact pathways covered by current methods regarding human health include the lack of freshwater for hygiene and ingestion resulting in the spread of communicable diseases (Motoshita et al. 2010; Boulay et al. 2011b), water deprivation for irrigation causing in malnutrition (Pfister et al. 2009; Motoshita et al. 2011; Boulay et al. 2011b) and water deprivation for freshwater aquaculture and fisheries resulting in loss of productivity and food supply (Boulay et al. 2011b). Indirect impact of freshwater use, i.e., impact on human health and conflict creation is not covered by existing methods. The cause-effect chain modeling is based on hydrological and socio-economical data (Pfister et al. 2009; Motoshita et al. 2010; Motoshita et al. 2011; Boulay et al. 2011b). Some of them consider the water scarcity index used at the midpoint level (Pfister et al. 2009; Boulay et al. 2011b). The level of economic development is considered in studied methods through parameters such as Human Development Index (HDI) (Pfister et al. 2009), house connection to water supply (Motoshita et al. 2010) or adaptation capacity based on gross national income (Boulay et al. 2011b). All methods consider the reduction of human health impacts in case the level of economic development is sufficient to cover compensation mechanism costs, but none of them includes the impact of the development and functioning of compensation mechanisms. Not expanding the system boundary is a common approach in attributional LCA. Some of the cause-effect chains relationships have been calculated based on empirical data, e.g., malnutrition rate and human development index (Pfister et al. 2009), water scarcity and accessibility to safe water (Motoshita et al. 2010). Other cause-effect chains rely on the multiplication of key parameters (Boulay et al. 2011b). Both approaches are relevant, but need to be further characterized by a measure of uncertainty to assess the deviation of potential impacts estimation.

Endpoint indicators are generally regionalized on a country (Pfister et al. 2009; Motoshita et al. 2010; Boulay et al. 2011b) or watershed level (Pfister et al. 2009; Boulay et al. 2011b).

Ecosystem quality: Methods addressing ecosystem quality cover different parts of the cause-effect chains relevant to ecosystem services and biodiversity. The cause-effect chains that current methods cover regarding damages to ecosystem quality are the decrease of terrestrial biodiversity due to freshwater consumption (Pfister et al. 2009), decrease of aquatic biodiversity due to turbined water use (Maendly and Humbert

2009), disappearance of terrestrial plant species due to groundwater withdrawal and related lowering of the water table (van Zelm et al. 2011), and the effects of freshwater consumption on freshwater fish species (Hanafiah et al. 2011). These endpoint methods do not use water scarcity indices as elements of the modeling equations. Rather, they are applied to different water types and uses, and should be used complementarily. Most methods consider the ecological value of freshwater resources through an empirical observation of decreased biodiversity or of other proxy data such as net primary production (Maendly and Humbert 2009; Pfister et al. 2009; van Zelm et al. 2011) and from a mechanistic perspective, e.g., by relating fish species richness to river discharge (Hanafiah et al. 2011).

Some cause-effect chains e.g., the impact due to water deprivation related to water quality degradation on aquatic ecosystems still need to be covered by additional methods.

Endpoint methods addressing ecosystem quality have different levels of spatial differentiation: no differentiation, generic or for a specific region (van Zelm et al. 2011), archetype (e.g., alpine and non-alpine dam (Maendly and Humbert 2009), country (Pfister et al. 2009), or watershed (Pfister et al. 2009; Hanafiah et al. 2011). This variability of the differentiation level reflects the diversity of the parameters considered in the cause-effect chain.

Resources: Methods addressing the area of protection resources quantify the impact on future freshwater availability through a backup-technology approach to evaluate the impact of freshwater consumption above their renewability rate (Pfister et al. 2009) or through the exergy content of the freshwater resource (Bösch et al. 2007). In contrast to the Pfister et al.'s method (2009), Bösch et al. (2007) is not specific to water resources and does not consider water scarcity.

None of the evaluated endpoint methods covers the cause-effect chain comprehensively: the pathway addressing impact due to fossil groundwater depletion is poorly known and is not covered by available methods. Furthermore, estimation of impact of consumption over the renewability rates lacks differentiation between different water types and change in green water availability is not covered.

Pfister et al.' (2009) is a spatially differentiated method on a watershed and a country level whereas Bösch et al.'s (2007) method is not differentiated.

2.4 Recommendations

2.4.1 Description and review of existing freshwater use assessment methods

The previously described findings can guide future consideration of freshwater use in LCA.

Inventory databases

From a business and industry perspective, data availability on freshwater use as well as harmonized reporting formats are limiting factors for establishing meaningful water footprints of products, processes and organizations (Koehler 2008). A balanced approach between LCIA methods and business data requirements is therefore needed to make characterization methods broadly applicable and meaningful. In order to link up with emerging LCI and LCIA methods, inventory databases should preserve the maximum freedom to provide necessary flows for application of different impact methods. The following recommendations for inventory database developments were drawn based on existing LCI and LCIA methods and are evaluated as necessary:

- Differentiate consumptive freshwater use from withdrawal (abstraction) through consistent water balances for foreground and background processes and do not mix physical flows with assessment units such as m^3-equivalents of polluted water.
- Distinguish between different water types based on origin (surface freshwater, including river, lake and sea, groundwater, including renewable, shallow and deep and precipitation freshwater stored as soil moisture and fossil groundwater) and freshwater quality (and thus functionality). This can be done by applying the systematic classification proposed by Boulay and colleagues (2011a) according to quality data that could be collected, e.g., data on general parameters (which include microbial parameters), inorganic compounds and organic compounds.
- Include freshwater evaporation from water reservoirs as consumptive use, as it makes freshwater locally/regionally no more available.

Additional optional guidelines could be integrated:

- For the assessment of groundwater withdrawals and associated impacts differentiate shallow (<2.3 meters) and deep water tables (van Zelm et al. 2011) or estimate regional average fractions of areas of each type.
- Differentiate withdrawal of fossil groundwater from renewable groundwater based on regionally available resources as far as possible.

Inventory methods

General recommendations for inventory methods are:

- Include only measurable freshwater types, e.g., surface water and groundwater, or a method to estimate those flows shall be provided, water stored as soil moisture evapo-transpirated by plants, so called “green water”, which can be estimated with a crop model suitable, based on input data on climate, soil and crop characteristics (Hanasaki et al. 2010; Hoekstra et al. 2011).

- Use water quality parameters to characterize freshwater flows that are available in existing databases.

Impact assessment methods

In order to ease their applicability, LCIA shall in general show robust examples linking the inventory of freshwater types with all needed calculation steps to apply characterization factors and aggregate results for obtaining related midpoint or endpoint indicator.

Midpoint assessment methods

The water consumption or withdrawal to availability ratio has been recognized as a representative proxy for scarcity, in comparison to other indices, e.g., water use per capita, which reflects rather a socio-economic situation. Midpoint methods addressing water scarcity shall:

- Include water storage capacity in the modelling of total water availability within a geographical unit.
- Be quantitatively compared to evaluate the trade-off between easiness of application and cause-effect chain coverage and related uncertainty between indicators based solely on water scarcity (Frischknecht et al. 2006; Milà i Canals et al. 2008; Pfister et al. 2009; Ridoutt and Pfister 2010b) and more comprehensive midpoint indicators (Boulay et al. 2011b; Bayart et al. 2014).
- Provide further empirical evidence of the link between water scarcity, water deprivation, and impact on different areas of protection to evaluate the relevance of midpoint versus endpoint indicators. In an LCA perspective, water scarcity indicator does not refer to any potential impact. This does not necessarily mean that an endpoint is ultimately affected. Water stress index is for example involved in Pfister et al.’s and Boulay et al.’s endpoint models for human health, but human health is not affected if the economic development level is sufficient. Clear evidence of the link between water scarcity, water deprivation, and impact on different areas of protection would be needed to evaluate the relevance of midpoint versus endpoint indicators.

Endpoint assessment methods

Next steps towards a consistent framework for application of endpoint methods are as follows.

For the area of protection human health:

- Provide a quantitative comparison of existing methods as well as an evaluation against empirical figures.
- Assess the relevance and uncertainty of modelling indirect impacts related to water deprivation, e.g., human health impact due to conflict creation, population displacement.
- Develop new approaches for modelling of compensation mechanisms to prevent water loss in functionality throughout impact categories, knowing that technical means can also be used to cope with other impacts such as climate change.

For the area of protection ecosystem quality:

- Identify extensively missing cause-effect chain.
- Provide global coverage and appropriate spatial resolution (e.g., watershed scale) for methods developed for a single country (van Zelm et al. 2010) or with partial basin coverage (Hanafiah et al. 2011).

For the area of protection resources:

- Cover the cause-effect chain leading to impact of fossil groundwater exhaustion, as well as include it in the inventory.
- Distinguish impact related to different freshwater types consumption, given they have different renewability rates and functionalities.
- Quantifying the link between green water use and resources. Although Heuvelmans et al. (2005) developed a method to quantify impact of land use concerned with changes in hydrological response of the land, no characterization factors yet exist to quantify this relationship.

For all midpoint and endpoint methods, uncertainties of input data as well as model uncertainty still need to be evaluated and documented. Midpoint and endpoint methods covering human health and ecosystem quality impact shall provide characterization factors with monthly differentiation to reflect variability related to meteorological conditions and associated ecosystem changes.

2.4.2 Application recommendations for practitioners given current state-of-the-art

The evaluation of freshwater use is possible by assembling methods in a comprehensive methodology to characterize each use adequately. Current state-of-the-art can already provide a preliminary understanding of water uses and associated impacts, especially on human health and ecosystem quality.

In this respect, a detailed inventory including freshwater withdrawal and release, water consumption, and turbined water constitutes a first step towards understanding the various flows related to the system. Inventory results can be used as an indicator as such (WBCSD 2010; Hoekstra et al. 2011) but the interrelation between inventory results and impact linked to freshwater use is not yet proven and can be in some cases misleading (Ridoutt et al. 2010; Ridoutt 2011). Clarity of LCI scope demands clear communication regarding whether an attributional or consequential LCI approach has been taken.

For midpoint level assessment, it is not yet possible to draw conclusions on method preference given that case studies to test the significance of each method are under development. It is recommended to use the existing midpoint methods most relevant for the study under elaboration to ensure an extensive sensitivity analysis on the methodological choice, keeping in mind their cause-effect chain overlaps. If possible, the information given by scarcity indices should be interpreted in parallel with damage oriented impact assessment indicators to provide a comprehensive picture of impacts related to freshwater use.

For endpoint level assessment it is recommended to combine indicators of all cause-effect chains, i.e., malnutrition or infectious diseases related to water deprivation of a defined quality class for agriculture, fisheries and domestic use for human health. For ecosystem quality, the scopes of methods developed could so far be considered as complementary. All ecosystem quality indicators could therefore be used simultaneously and summed up into a single metric. However results should be interpreted with caution as not all the indicators are addressing the same endpoints. The resource area of protection is considered not being sufficiently developed to provide significant results.

This assessment needs to be completed by emission to all compartments ultimately affecting water (e.g., aquatic acidification, eutrophication, human toxicity, ecotoxicity as well as heat release to water) to provide a complete picture of water related impacts.

2.5 Conclusion

This is the first state-of-the-art assessment of freshwater use related methods. This review assesses relevant tools to make an assessment from a product or site perspective, extending the analysis beyond the water flow inventory and encompassing impact from indirect water use in the system limits. Although some cause-effect chains still needs to be covered, spatial differentiation refined and uncertainty assessed, the set of methods presented can already help to grasp water-related challenges and risks which humans face and serve as a first base for strategic decisions. Water assessment is a fast progressing field, and this review will need to be regularly updated to include new developments.

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Appendix

The appendix includes the detailed results of the review of each method against general and specific criteria, a short description of reviewed methods and a summary of general assessment criteria.

Chapter 3

Spatial analysis of toxic emissions in LCA: A sub-continental nested USEtox model with freshwater archetypes

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Abstract

This paper develops continent-specific factors for the USEtox model and analyses the accuracy of different model architectures, spatial scales and archetypes in evaluating toxic impacts, with a focus on freshwater pathways.

Inter-continental variation is analysed by comparing chemical fate and intake fractions between sub-continental zones of two life cycle impact assessment models: (1) the nested USEtox model parameterized with sub-continental zones and (2) the spatially differentiated IMPACTWorld model with 17 interconnected sub-continental regions. Substance residence time in water varies by up to two orders of magnitude among the 17 zones assessed with IMPACTWorld and USEtox, and intake fraction varies by up to three orders of magnitude. Despite this variation, the nested USEtox model succeeds in mimicking the results of the spatially differentiated model, with the exception of very persistent volatile pollutants that can be transported to polar regions.

Intra-continental variation is analysed by comparing fate and intake fractions modelled with the a-spatial (one box) IMPACT Europe continental model vs. the spatially differentiated version of the same model. Results show that the one box model might overestimate chemical fate and characterization factors for freshwater eco-toxicity of persistent pollutants by up to three orders of magnitude for point source emissions. Subdividing Europe into three archetypes, based on freshwater residence time (how long it takes water to reach the sea), improves the prediction of fate and intake fractions for point source emissions, bringing them within a factor five compared to the spatial model.

We demonstrated that a sub-continental nested model such as USEtox, with continent-specific parameterization complemented with freshwater archetypes, can thus represent inter and intra-continental spatial variation, while minimizing model complexity.

Keywords

Spatial differentiation; USEtox; life cycle assessment; ecotoxicity; human toxicity; archetypes

3.1 Introduction

Decision-making in green chemistry and chemical screening needs adapted tools to assess fate, exposure and risks of chemicals on human health and ecosystems. In a global economy, where products are manufactured and used in various continents over their life cycle, we specifically need tools able to assess and differentiate pollutants emitted on different continents and in meaningful geographical units within a continent and related potential impacts. Life Cycle Assessment (LCA) is a useful approach for such decisions, with its multimedia and multi-pathway exposure models recognized as particularly well-suited to assess eco-toxicity and human toxicity impacts (Udo de Haes et al. 2002; Pennington et al. 2004; Finnveden et al. 2009).

USEtox is a consensus model developed within the Life Cycle Initiative led by the United Nations Environmental Program and the Society of Environmental Toxicology and Chemistry (UNEP-SETAC) (Hauschild et al. 2008; Rosenbaum et al. 2008; Rosenbaum et al. 2011; Henderson et al. 2011). This parsimonious and transparent model can screen thousands of chemicals and is widely used, but only provides continent-generic characterization factors and impact scores for a generic unknown continent. Since human and eco-toxicity occur as regional or local impacts (Potting and Hauschild 2006; Sedlbauer et al. 2007), recently developed multimedia and multi-pathway models are spatially differentiated in order to provide different impact scores for each regional zone. Spatial differentiation reduces model uncertainty and improves accuracy, precision and confidence in LCA results (Potting and Hauschild 2006; Manneh et al. 2010; Wegener Sleeswijk and Heijungs 2010). There is therefore a need to customize USEtox for different specific regions of the world in addition to the existing generic continent.

Inter-continental variation has been investigated using a nested parameterization of the IMPACT 2002 model (Rochat et al. 2006), with continent-specific boxes nested within the world. The Australian adaptation of the USES-LCA 2.0 model has been similarly investigated by Lundie et al. (2007). Rochat et al. (2006) found a factor 1.7 to 25 variation in human health impacts among continents. The variation is especially high for short-lived pollutants, e.g., the ingestion intake fraction of aldrin varies by a factor 25 between emissions in Europe and Oceania. These studies, however, do not address whether region-specific nested models accurately capture results obtained by spatially resolved models that include advection between continents for a set of chemicals covering a wide set of physico-chemical properties. There is a need to evaluate how far this full advection modifies the assessment of fate and exposure and whether a nested individual sub-continental model is sufficient for chemical screening.

Intra-continental variation has been investigated at several resolutions, including 1*1 km grid cells for the MAPPE Europe model (Pistocchi et al. 2010; Vizcaíno and Pistocchi 2010), ecological zones with a continental coverage for the BETR North America model (Macleod et al. 2004), and watershed or sub-watershed resolution for freshwater emissions in various parameterizations of the IMPACT 2002 model (Humbert et al. 2009; Manneh et al. 2010). Depending on the emission location within a given continent, intake fractions vary by 2 to 3 orders of magnitude for emissions to air (Macleod et al. 2004) and up to 10 orders of magnitude for emissions to water (Manneh et al. 2010), highlighting the necessity of high resolution to reduce intake fraction variability.

Assuming continent-level homogeneity may therefore lead to systematic errors, spatial differentiation is necessary. The choice of spatial resolution should account for scientific needs, as well as more practical data and computational constraints (Sedlbauer et al. 2007). Dividing a region into sub-regions with specific characteristics provides one way of limiting the geographical data requirements (e.g., meteorological, population, and agricultural zones) while maintaining sufficient accuracy. Humbert et al. (2011) showed that intake fractions from inhalation of primary particulate matter can be modelled based on emission release height and "archetypal" environment (indoor versus outdoor; urban, rural, or remote locations) and vary by orders of magnitude among conditions considered. Several other authors have used the archetype approach to estimate human toxicity impacts from air emissions, including Hellweg et al. (2009) and Wenger et al. (2012) for indoor air and Rosenbaum et al. (2011) for urban emissions by continent. However, a similar archetypal approach has not yet been developed for related fate and exposure for water emissions. There is a need to explore the relevance of the archetype approach for emissions to freshwater compartments with various hydrological key characteristics.

This work aims to evaluate an appropriate model architecture (nested vs. spatially differentiated) and spatial resolution for the freshwater eco-toxicity and human toxicity impact categories in order to maintain environmental relevance while limiting model sophistication in terms of landscape data requirements. This paper primarily focuses on freshwater related pathways affecting human health and ecosystem quality by analysing pollutant fate in fresh water, as well as ecosystem and human exposure, aiming to:

1. Develop landscape parameters for USEtox to develop continent-specific boxes nested within the world.
2. Analyse the *inter-continental* variation of chemical fate and intake fractions among continents and examine the influence of the region(s) surrounding the considered sub-continent. For this, results from a nested USEtox model with continent-specific parameterization are compared to a fully connected model.
3. Study *intra-continental* variation and develop archetypes for freshwater eco-toxicity and human toxicity exposure to ingestion of fresh water and fish, as a parsimonious surrogate to higher spatial resolution.

3.2 Materials and methods

We selected IMPACTWorld (Shaked 2011) to create and parameterize USEtox continents nested within a global box and analyse intra-continental variation on the sub-continental level. IMPACTWorld is the only global interconnected model of pollutant fate and exposure modelling atmospheric air transport, while the only other interconnected global model GLOBOX (Wegener Sleeswijk and Heijungs 2010) is based on average measured wind speeds at ground level (independent of direction) in capital cities. It models media-specific concentrations and intake fractions in 17 sub-continental regions fully interconnected by advective air and freshwater flows, which offers an interesting element of comparison with nested model, but results in an increased level of complexity. The concentrations of PCB-118 in the environment and food were compared to measured empirical concentrations (Shaked 2011). The comparison showed that the accuracy of IMPACTWorld in predicting environmental concentrations is generally within an order of magnitude, compared to 12 orders of magnitude of variability among impact characterization factors among different substances (Rosenbaum et al. 2008).

We selected the IMPACT 2002 model on the watershed scale for a European resolution to analyse inter-continental variation. This model was compared and evaluated against monitored data (except for the freshwater fish ingestion pathway) (Margni et al. 2004; Pennington et al. 2005; Humbert et al. 2009). The advantage of this model is that it is resolved on a watershed scale, which corresponds to an adequate definition of flow patterns at the regional scale to study intra-continental variation for freshwater eco-toxicity and human toxicity exposure to ingestion of fresh water and fish. The water runoff data has been compiled in this model based on empirical data from the Global Run Off Data Centre (2002). Hydrological datasets are recently available at a higher resolution at 0.5° (Vörösmarty et al. 2000a; Jolliet et al. 2012) and 15’ (Lehner et al. 2006) but are not yet implemented in multimedia models.

3.2.1 Parameterization of USEtox landscape data

IMPACTWorld is a spatially differentiated multimedia model that divides the world into 17 sub-continental regions, 9 ocean regions, and 33 coastal regions (Figure 3.1). The regions of the IMPACTWorld model are similar to those chosen for the Input-Output model (Peters and Hertwich 2007; Friot and Antille 2009; Miller and Blair 2009) with some key differences to (1) put less emphasis on geographical boundaries and (2) represent the best trade-off between continental or sub-continental resolution and the representation of population densities and meteorological conditions (Shaked 2011). As in previous IMPACT versions (Pennington et al. 2005), each continental region consists of an air zone (containing an air compartment) and a terrestrial zone (containing water, soil, above-ground leaf crops, roots, and sediment), and each ocean region consists of an air zone and an ocean zone (containing surface ocean, deep ocean, and ocean sediment). Each region is characterized by environmental and demographic parameters, such as rainfall rate, vegetation fraction, and, most importantly for estimating population intake, vegetable and animal production intensity and population density.

In a first step, we developed parameterized sub-continental and continental specific landscapes in USEtox based on the 17 zones of the IMPACTWorld model (Shaked 2011). To achieve this, we successively consider the IMPACTWorld parameterization of each sub-continent, grouping the rest of the world into the USEtox global box. Special care was taken to define advection rates between each sub-continent and the surrounding global box, based on average wind speed over the height of the continental air box (for example, the US region has an average wind speed of 7.0 m/s over the lowest 1000 m of air). This advective wind speed over 1000 m is typically higher than the 3 m/s used in USEtox, which is the default wind speed at 10 m height used to determine exchange rates between air, soil and water surface compartments. In a second step, we grouped these 17 zones into 8 continental zones delimited by Humbert et al. (2011) to reduce data collection needs for LCA practitioners while still meeting the need of continent-specific characterization factors for LCA studies.

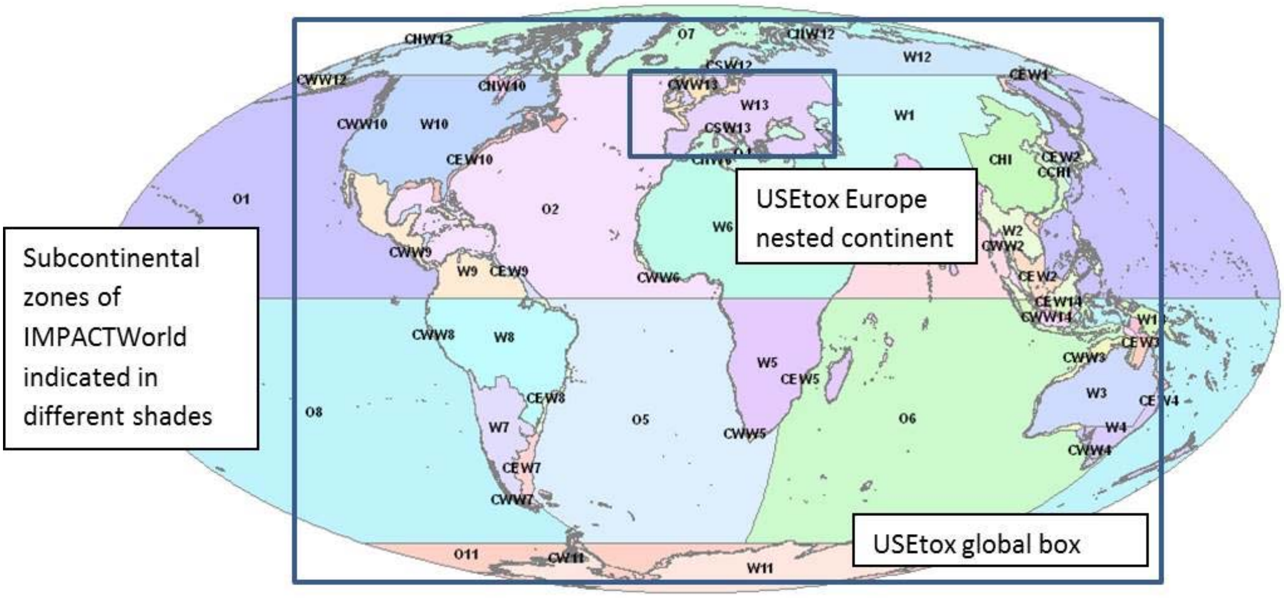


Figure 3.1: Depiction of how the IMPACTWorld model (Shaked 2011) is used to parameterize the Europe box of USEtox, nested within the global box

Table 3.1 shows selected key landscape parameters of the USEtox parameterization for each of the 17 zones. The full set of parameters is provided for these sub-continental zones and for the 8 continental zones in Table B.1, Table B.2, Table B.3 and Table B.4 in the appendix. The key physical parameters which influence exposure through the aquatic environment are the mean freshwater depth, which varies by a factor 15 across continents, and the freshwater residence time of water to the sea (Pennington et al. 2005; Henderson et al. 2011), which varies by a factor 83. Freshwater residence time of water to the sea $\tau_{\mathrm{sea}\,i}$ [day] is calculated by summing the residence time in sub-continental zone i with the transfer fraction to all sub-continental zones j downstream of i $f_{j\,\mathrm{downstream}}$ [-] multiplied by their freshwater residence times τ_j [day] as follows:

$$\tau_{\text{sea } i} = \tau_i + \sum_{f_{j \text{ downstream}}} \tau_j$$

Equation 3.1: Residence time to the sea calculation

Exposure data are based on regional populations and food production statistics from FAO (2001), and they vary by up to a factor 1850 for marine fish production per capita.

Table 3.1: Key parameters for each of the 17 sub-continental zones in the USEtox parameterization

ID #	Name	Land area	Sea area	Freshwater fraction of land area	Precipitation rate	Mean freshwater depth	Continental human population	Urban human population	Exposed produce	Unexposed produce	Meat	Dairy products	Fresh-water fish	Marine coastal fish	Water residence time to the sea
		km^2	km^2	[-]	$mm \cdot y^{-1}$	M	[-]	[-]	kg/(day-capita)	kg/(day-capita)	kg/(day-capita)	kg/(day-capita)	kg/(day-capita)	kg/(day-capita)	day
W1	**West Asia**	1.7E+07	7.4E+05	1.7E-02	2.2E+02	1.3E+01	2.35E+08	1.47E+06	1.71	0.33	0.08	0.26	0.011	0.05	1300
W2	**Indochina**	3.3E+06	2.2E+06	3.6E-02	2.4E+03	1.3E+01	4.65E+08	1.30E+06	2.57	0.22	0.05	0.01	0.008	0.06	260
W3	**N. Australia**	6.6E+06	1.6E+06	9.9E-03	1.5E+03	3.0E+00	3.20E+06	8.24E+05	10.45	0.18	0.51	1.39	0.006	6.65	28
W4	**S. Australia+**	1.5E+06	6.4E+05	1.2E-02	5.1E+02	3.0E+00	2.12E+07	1.03E+06	9.02	0.21	0.60	2.84	0.004	0.61	98
W5	**S. Africa**	1.0E+07	6.2E+05	2.2E-02	1.0E+03	4.6E+01	3.24E+08	1.25E+06	1.03	0.44	0.03	0.06	0.006	0.03	1400
W6	**N. Africa**	2.4E+07	9.7E+05	1.9E-02	5.1E+02	4.6E+01	7.89E+08	2.30E+06	0.98	0.46	0.04	0.10	0.006	0.01	2400
W7	**Argentina+**	4.2E+06	1.1E+06	1.5E-02	7.0E+02	8.0E+00	6.67E+07	2.89E+06	4.92	0.46	0.23	0.57	0.002	0.31	240
W8	**Brazil+**	1.1E+07	5.8E+05	8.3E-03	1.8E+03	8.0E+00	2.42E+08	2.62E+06	6.08	0.36	0.20	0.28	0.004	0.05	54
W9	**Central America**	5.9E+06	1.3E+06	3.6E-02	2.0E+03	2.0E+01	3.05E+08	2.76E+06	2.62	0.10	0.09	0.20	0.003	0.04	480
W10	**US+**	1.4E+07	1.8E+06	3.4E-02	7.1E+02	2.0E+01	3.28E+08	1.32E+06	4.82	0.42	0.35	0.69	0.003	0.04	1300
W12	**N. Eur. + N. Canada**	1.8E+07	5.6E+06	4.9E-02	4.9E+02	1.7E+01	1.67E+07	6.56E+05	1.74	0.47	0.15	0.75	0.008	1.43	2100
W13	**Europe+**	8.6E+06	1.7E+06	1.6E-02	5.5E+02	1.5E+01	7.59E+08	1.41E+06	2.57	1.12	0.19	0.80	0.003	0.02	610
W14	**East Indies**	2.0E+06	1.4E+06	3.0E-02	1.5E+03	3.0E+00	2.07E+08	1.30E+06	1.21	0.16	0.02	0.01	0.013	0.13	80
IND	**India**	4.6E+06	4.6E+05	4.2E-02	1.2E+03	1.3E+01	1.57E+09	1.76E+06	1.52	0.07	0.01	0.20	0.008	0.003	580
CHI	**China**	6.4E+06	8.4E+05	4.6E-02	1.2E+03	1.3E+01	1.33E+09	1.47E+06	1.90	0.38	0.12	0.03	0.029	0.01	620
JAP	**Japan**	6.0E+05	4.2E+05	4.4E-02	2.4E+03	1.3E+01	1.51E+08	4.56E+06	1.16	0.21	0.09	0.19	0.027	0.06	310
Source		Based on GIS computation			GEOS-Chem model	UNEP freshwater depth (http://www.unep.org/vitalwater/freshwater.htm)	GIS analysis of 2010 UNEP GEO 1°x1° population data (http://geodata.grid.unep.ch/)	Regional calculation based on city data	FAO production data from 2001				FAO FishSTAT		Recalculated based on model algorithm

3.2.2 Inter-continental variation and influence of surrounding regions

Landscape parameters such as sub-continental land area, mean freshwater depth, freshwater residence time to sea, and population density influence chemical fate in environment and human intake. We will first analyse the variation of fate and human exposure within each of the 17 sub-continents and thus the relevance of using a specific sub-continent rather than generic continental parameters. The analysis is performed for a set of 36 non-dissociating and non-amphiphilic organic chemicals selected from the OMNIITOX project (Margni et al. 2002; Margni 2003). It represents well the variability of physicochemical properties of organic substances as reported in Table B5 and Table B6 in the appendix. This set covers all relevant combinations in terms of environmental partitioning and exposure routes, overall persistence, long-range transport and feedback fraction.

In parallel, we use this chemical set to examine the influence of surrounding region(s) on the fate and exposure of emissions within the considered sub-continent. For this, results from the nested USEtox model with continent-specific parameterization are compared to the fully connected IMPACTWorld model. The 17 zones of the nested USEtox model have the same resolution and landscape parameters as those in the interconnected IMPACTWorld model. Beyond this commonality, the two models calculate fate and intake fraction differently in two key ways: (1) USEtox embeds the sub-continent in a single global box, whereas IMPACTWorld explicitly connects the sub-continental zone to specific adjacent zones, and (2) the model algorithms for exposure and particularly fate are somewhat different. The latter difference can be illustrated by the modelling of advective outflow from a sub-continental zone. It is based on river discharges out of sub-continental zones taken from external references in IMPACTWorld and on a mass balance based on precipitation, evaporation and advection in USEtox. Between these two models, we compared the fate and inhalation intake fractions, as well as ingestion intake fractions through drinking fresh water and eating fish, exposed produce (above-ground leaf crops, including fruit and cereals), unexposed produce (below-ground root crops), dairy and meat products.

3.2.3 Intra-continental variation and identification of key spatial variation parameters

Intra-continental variation was analysed on a finer resolution by comparing USEtox for Europe to the following versions of the IMPACT Europe model: the IMPACT Europe single zone model without spatial distinction (i.e., with one homogeneous compartment per environmental medium), and the IMPACT Europe spatial model accounting for spatial differentiation of 135 watersheds and land zones and 156 air zones on a 2*2.5 degree grid. Both spatial and a-spatial versions are nested into an a-spatial global zone.

This comparison was carried out first assuming uniformly distributed emissions (i.e., emissions distributed in each watershed proportionally to its land surface area). We then compared results for emissions occurring entirely in one of three selected watersheds being representative of three very different landscape characteristics: a near coast emission in Brittany, an emission into a long river (Danube) and an emission upstream of a large lake (Lake Geneva). We then analysed the interaction between chemical properties and spatially differentiated landscape properties of each watershed to identify the key parameters influencing the fate factors.

Previous observations (Pennington et al. 2005) show that within an open system (1) the spatial differentiation for aquatic eco-toxicity is only relevant for persistent pollutants (i.e., pollutants with a degradation rate higher than the advection rate); and (2) for these persistent pollutants, one important factor affecting fate is the freshwater residence time until reaching the landscape boundary (i.e., until the sea or any other advection into the global system). The mean freshwater depth also affects elimination rates by volatilization and sedimentation and might also play a role. Exposure factors such as the fraction of freshwater volume ingested by the population as drinking water or indirectly through fish ingestion may also influence the freshwater-mediated intake fraction. We therefore analyse the influence of water residence time, water depth and population intake rates and their auto-correlation across the 135 watersheds.

3.2.4 Development of freshwater archetypes based on freshwater residence time

Based on the identified key parameters, we ultimately developed a method to create a limited number of watershed archetypes and test how well these archetypes reflect major variations in fate and exposure across watersheds.

Due to the importance of freshwater residence time until reaching the sea (or the model boundary) found in previous work (Pennington et al. 2005), this parameter could be used in a first step as the main variable to define these archetypes. To keep the number of archetypes manageable for common practice in Life Cycle Inventory, we define three freshwater residence time archetypes, corresponding to A1) coastal zones with short freshwater residence times, A2) zones with medium freshwater residence times and A3) zones with high freshwater residence times. We define the mean residence times in each zone as $\tau_{\text{sea}\,A1}$, $\tau_{\text{sea}\,A2}$ and $\tau_{\text{sea}\,A3}$ and the upper threshold residence times for zones A1 and A2 as τ_{12} and τ_{23} (upper threshold is infinite for A3). These upper thresholds were defined by minimizing the standard deviation variation between the log of the freshwater residence time for each watershed i in the spatial model and the log of the mean residence time for that watershed's archetype using the Excel solver tool. The total standard deviation SD_{tot}, is the sum of the standard deviations for each of the three residence time archetypes:

$$SD_{tot} = \{\frac{1}{n} * \sum_{\tau_{\text{sea}\,i}=0}^{\tau_{12}} [(\log(\tau_{\text{sea}\,i}) - \log(\tau_{\text{sea}\,A1})]^2 + \sum_{\tau_{\text{sea}\,i}=\tau_{12}}^{\tau_{23}} [\log(\tau_{\text{sea}\,i}) - \log(\tau_{\text{sea}\,A2})]^2 + \sum_{\tau_{\text{sea}\,i}=\tau_{23}}^{\infty} [\log(\tau_{\text{sea}\,i}) - \log(\tau_{\text{sea}\,A3})]^2\}^{1/2}$$

Equation 3.2: Residence time to the sea calculation

where

$\tau_i = \frac{V_i}{Q_i}$ [d] is the water residence time in watershed i (1≤i<135 for the spatial IMPACT Europe model), calculated as the watershed volume V_i [m3] divided by the advection flow out of the watershed Q_i [m3/d];

$\tau_{\text{sea}\,i} = \tau_i + \sum_{f_{j\,\text{downstream}}} \tau_j$ [d] is the water residence time until reaching the sea, calculated by summing the residence time in watershed i with the transfer fraction to all watersheds j downstream of i multiplied by their freshwater residence times;

$\tau_{\text{sea }A1}$, $\tau_{\text{sea }A2}$ and $\tau_{\text{sea }A3}$ are the residence times for each archetype, calculated based on the total volume of all watersheds corresponding to this archetype and the total advective flow out of all watershed flows corresponding to this archetype.

The mean freshwater residence time to sea is calculated as a surface weighted average of the water residence times of each watershed classified in one of the three archetypes. The calculated mean freshwater residence time of each watershed is presented in section 3.3.3. We test the relevance of these archetypes by determining the variability in fate and intake fraction across emissions in each of the 135 watersheds described by these three archetypes.

The practitioner may choose the archetype based on the emission location by finding the archetype corresponding to the place of emission according to Figure B.6 in appendix.

3.3 Results and discussion

3.3.1 Inter-continental variation: comparison between spatially differentiated IMPACT-World and nested USEtox model

Residence times: Figure 3.2a presents the range of freshwater residence times to sea, comparing the nested continent-specific USEtox model to the fully connected IMPACTWorld model. These residence times vary by up to two orders of magnitude among sub-continental zones, with North Australia having one of the shortest times and North Africa having the longest. Values in the two models are similar for all sub-continental zones, with the highest difference being a factor 4 for the East Indies (W14). These differences in freshwater residence times to sea are due to different ways of calculating total water advection. IMPACTWorld outflows are advective flows based on river discharges out of the sub-continental zone (Global Runoff Data Centre 2002), whereas USEtox uses a water balance approach based on rainfall, evapotranspiration, infiltration and runoff.

Fate factors: Figure 3.2b compares the fate factors in fresh water for chemical emissions to fresh water for each sub-continental zone in each of the two models. These fate factors represent the chemical mass in the freshwater environment per unit flow emission (units of kg per kg/d), which corresponds to the residence time of each substance in fresh water (in days). For persistent pollutants that have a long degradation half-life in water ($t_{1/2}$water), such as gamma-HCH (lindane) ($t_{1/2}$water = 1.9 y) or aldrin ($t_{1/2}$water = 2.0 y), their fate is more sensitive to the zone's freshwater residence time to sea. The fate factors of these persistent pollutants can thus vary by more than one order of magnitude among sub-continental zones, with aldrin ranging from 10 days in North Australia to 199 days in North Africa in the USEtox parameterization (Figure B.1 in appendix). For non-persistent substances, the emission location has little influence, and the fate factor is identical for all regions. N-nitrosodiethylamine has $_{t1/2}$water = 6 hours, and thus has a fate factor of 0.36 days in all continents (Figure B.1).

Discrepancies in fate factors between the two models are limited but observable, particularly for the sub-continental zones that have different freshwater residence times, such as W14 East Indies in Figure 3.2.a. W14 has a freshwater residence time of 19 days in IMPACTWorld and 80 days in USEtox. Thus persistent pollutants whose disappearance is not driven by degradation, evaporation or sedimentation (e.g., methomyl) have fate factors limited to 19 days in IMPACTWorld (and 80 days in USEtox). Yet Figure 3.2b shows one outlier that exceeds this maximum fate factor of 19 days – hexachlorobenzene. This is due to a dynamic that can only be captured by an interconnected spatial model like IMPACTWorld, in which a pollutant can be transferred from one sub-continental zone into an adjacent zone with a higher freshwater residence time to sea. The fate factor also depends on the product of the inter-continental transfer fraction from water to air (Figure B.2 in appendix) with the water residence time in the receiving compartment. The fate factor can therefore exceed the water residence time to sea of the emission compartment if it is transferred to the freshwater compartment of another sub-continental zone with higher freshwater residence time. In our case, the fate factor of hexachlorobenzene emitted in W14 (42 days) is driven by the fraction of the pollutant transferred through air to Antarctica (W11) (1.2%). Hexachlorobenzene is highly volatile (K_H=170 Pa.m3.mol-1) and persistent ($t_{1/2\ air}$=0.84 y and $t_{1/2\ water}$=6.3 y), and the IMPACTWorld model freshwater residence time in Antarctica (W11) is much higher than the hexachlorobenzene half-life. The fate of hexachlorobenzene in W11 is thus not limited by the freshwater residence time of the emitting compartment.

Intake fractions: Figure 3.2c and 3.2d display the range of human intake fractions through freshwater ingestion and fish ingestion, respectively, due to freshwater emissions of various substances. In addition to variation of water residence time in each sub-continental zone (influenced by water surface and depth), inter-continental variation also depends on landscape parameters related to exposure, such as population density and various food products intake rates. These parameters lead to variation in intake fractions for the same substance in different sub-continental zones of more than three orders of magnitude, not only for persistent substances, but also for some easily degradable chemicals. Given the many parameters that influence intake fraction, neither difference in spatialization nor in model algorithms causes substantial deviation of intake fractions between the two models, which generally remain within two orders of magnitude. Intake fraction results for other pathways from freshwater emissions, as well as intake fraction for all pathways from air emissions, are presented in Figure B.3 and Figure B.4 the appendix.

The previous results show that considered zones cover a wide range of landscape parameters corresponding to a comprehensive overview of sub-continental fate and intake fraction variability.

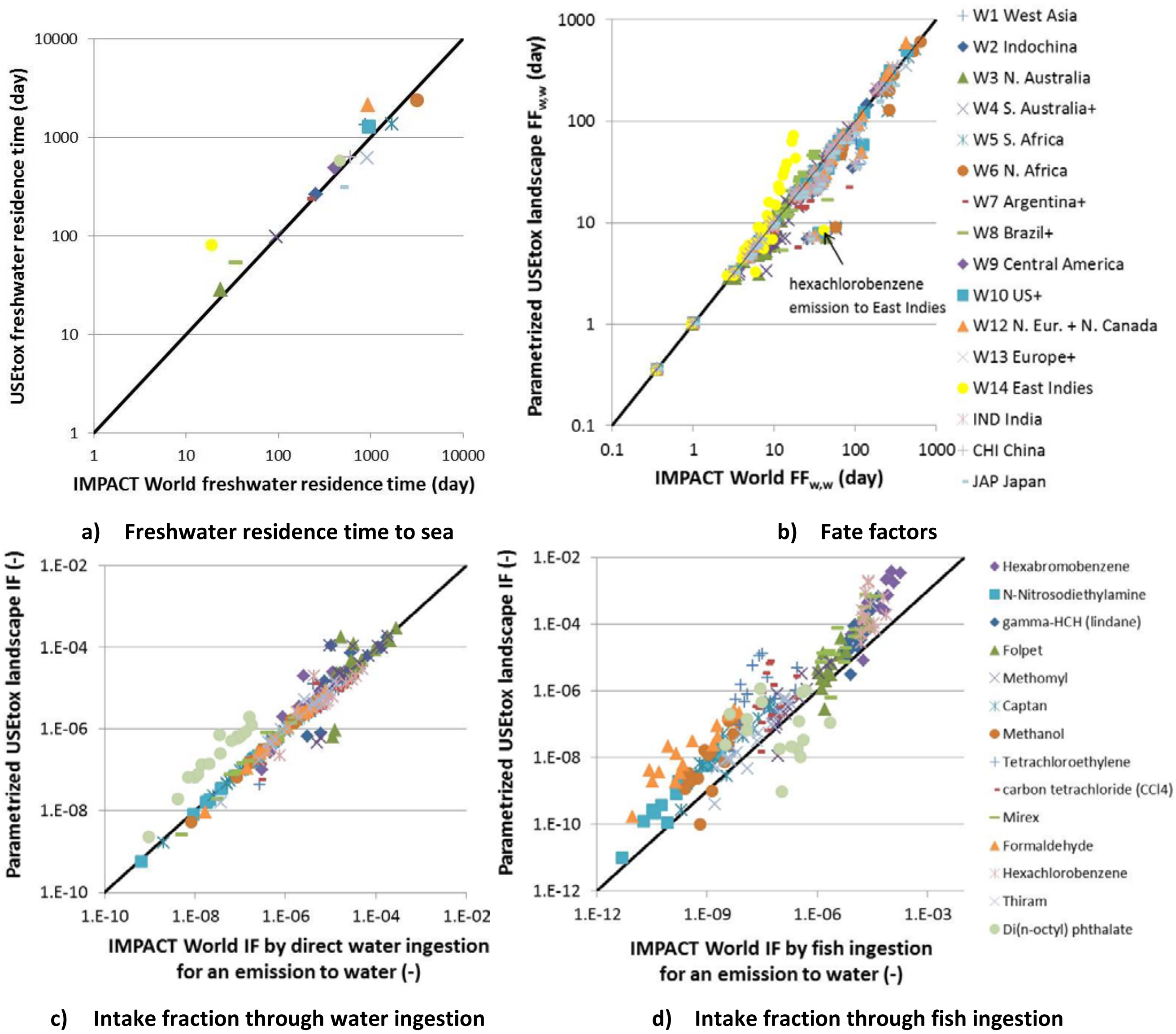


Figure 3.2 Comparison between nested USEtox model and the spatially differentiated IMPACTWorld model regarding: a) Freshwater residence time to sea in each sub-continent, b) Residence time in fresh water of 36 representative chemicals emitted in each sub-continent c) Intake fraction through freshwater ingestion and d) Intake fraction through fish ingestion

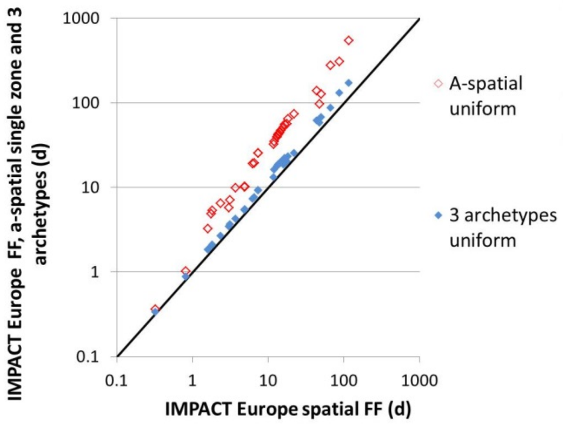
3.3.2 Intra-continental variation and importance of spatialization: Europe a-spatial and Europe spatial IMPACT model

This section analyses the intra-continental variation of fate and intake fractions in Europe on a watershed scale. Figure 3.3 uses a red symbol to compare the fate factors (a,b) and intake fractions (c-f) of the a-spatial with those of the spatial IMPACT model, for both uniform and point-source emissions (i.e., respectively for surface weighted emissions and emissions in specific European watersheds). Results for the three watershed archetypes are displayed in blue and discussed in section 3.3.3.

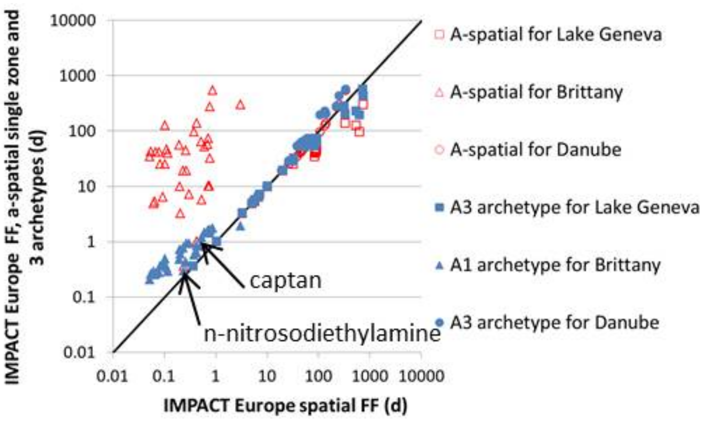
Fate factors: For a uniform emission (when a substance is uniformly emitted on the surface covered by the model), Figure 3.3a shows that fate factors smaller than 3 days (lower left portion of the graph) are similar for the spatial and a-spatial versions of the model. For more persistent pollutants and regions with higher freshwater residence times to sea, the a-spatial single box model overestimates the fate factor by up to a factor 5 when compared to the spatially differentiated model due to the high freshwater residence time to sea in the a-spatial model (4.1 y).

For point-source emissions (when a substance is emitted in a defined single geographical location covered by the model) in locations with increasing freshwater residence time to sea (Brittany is 0.81 days (corresponds to archetype A1), Danube is 1.4 y (corresponds to archetype A3) and Lake Geneva is 13.6 y to the sea (corresponds to archetype A3)), Figure 3.3b shows that the a-spatial model accuracy depends on the watershed in which the pollutant is emitted and on the pollutant persistence in fresh water. For highly degradable pollutants with degradation half-lives less than 3 days, such as n-nitrosodiethylamine and captan, a-spatial fate factors are aligned with the spatial ones for all watersheds. This is not the case for more persistent pollutants for which the freshwater residence time to sea of the emitting compartment is a key factor. For an emission to Lake Geneva, the a-spatial single box model only slightly underestimates the residence time, since the a-spatial freshwater residence time to sea (4.1 y) is three times higher than Lake Geneva's freshwater residence time to sea (13.6 y in the spatial model). For the same reason, fate factors for an emission into the Danube are slightly overestimated in the a-spatial model (freshwater residence time in the Danube watershed is 1.4 y). Brittany a-spatial fate factors are overestimated by about 3 orders of magnitude because of the short freshwater residence time to sea in this coastal region (0.81 day = 0.002 year). When an emission location is known, a spatially differentiated model can thus improve the model accuracy by up to 2- to 3 orders of magnitude.

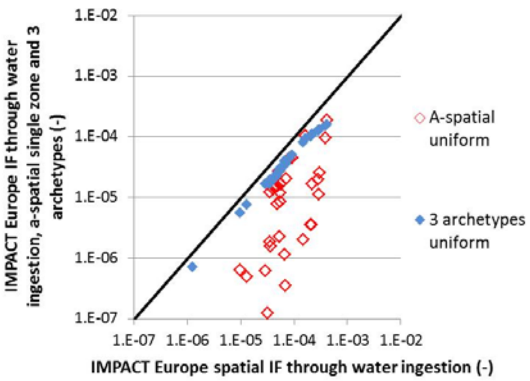
Intake fractions: Human intake fractions for freshwater and fish ingestion are represented in Figure 3.3c to f. For a uniform emission, the a-spatial model underestimates both these intake fractions by up to 2 orders of magnitude (Figure 3.3c and Figure 3.3e). This trend is different from that observed for fate prediction due to differences in exposure estimation between these two models. Despite the low freshwater residence time in coastal zones, exposure factors tend to be higher in these zones, compared to inland watersheds with high freshwater residence times. These variations in exposure also influence the intake fraction for a point source emission into fresh water (Figure 3.3d and Figure 3.3f), such that the a-spatial model underestimates the intake fraction for an emission in Brittany. The model overestimates intake for emissions into a large body of water such as the Lake Geneva watershed.



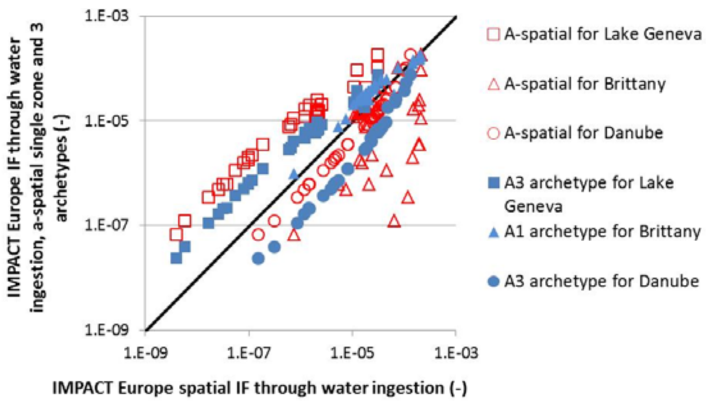
a) Fate factor for a uniform emission



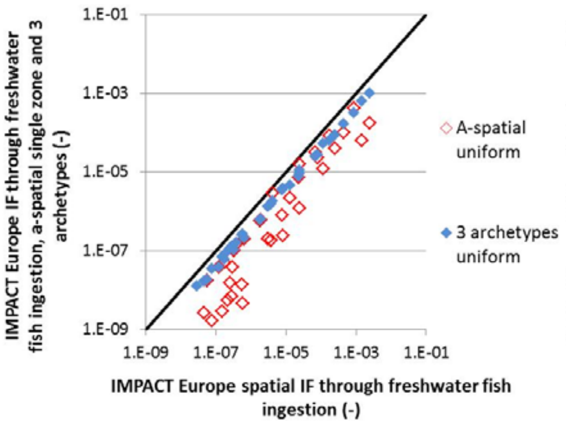
b) Fate factor for a point source emission



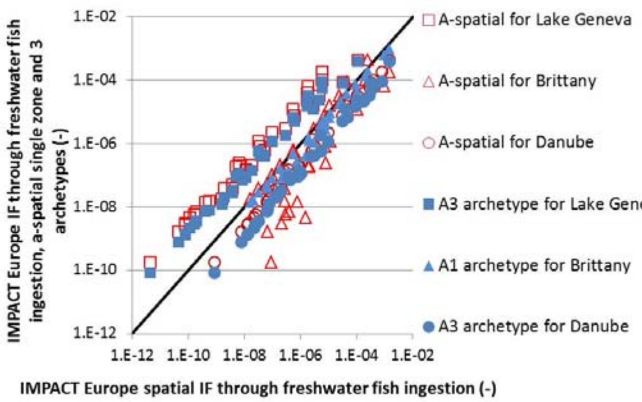
c) Intake fraction through freshwater ingestion for a uniform emission



d) Intake fraction through freshwater ingestion for a point source



e) Intake fraction through freshwater fish ingestion for a uniform emission

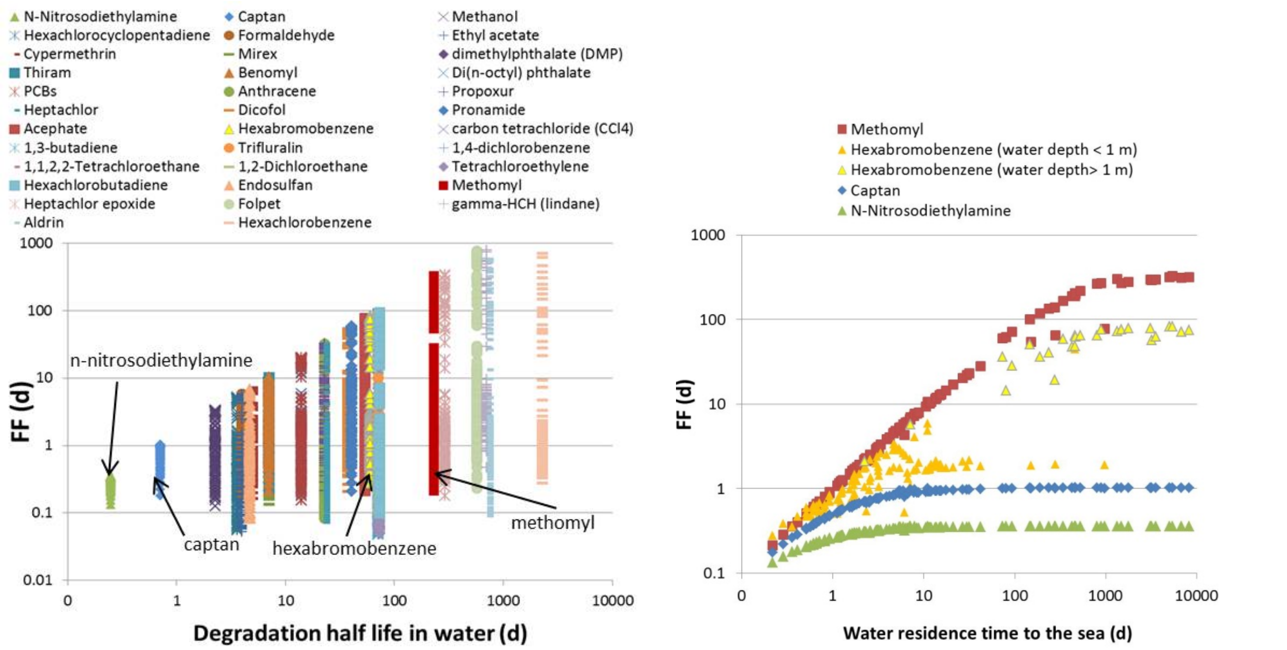


f) Intake fraction through freshwater fish ingestion for a point source

Figure 3.3: Fate factors and ingestion intake fractions of 36 organic chemicals through freshwater and fish, calculated for the spatial IMPACT Europe model, and compared to the a-spatial single zone IMPACT Europe model (red) and for the three watershed archetypes A1 and A3 (blue). a) Fate, c) intake by water ingestion and e) intake by fish ingestion for a uniform emission. b) Fate, d) intake by water ingestion and f) intake by fish ingestion for point source emissions in Lake Geneva (□), Brittany (Δ), and Danube (○)

Key parameters affecting fate: To further analyse the main parameters responsible for the spatial variation of fate factors, Figure 3.4a presents the variability in chemical fate for emissions in different European watersheds, as calculated by the spatially differentiated European IMPACT model. For easily degraded pollutants with a half-life lower than a day, such as n-nitrosodiethylamine ($t_{1/2}$=6 h in freshwater), fate factors vary by less than one order of magnitude across European watersheds. Such highly degradable or volatile chemicals disappear before being advected out of the watershed and their residence times do not vary much across watersheds. On the contrary, fate can vary up to four orders of magnitude for persistent pollutants with half-lives larger than 100 days, such as methomyl ($t_{1/2}$=230 days in fresh water).

By displaying the fate factor as a function of the freshwater residence time for four pollutants with different levels of persistence in fresh water (n-nitrosodiethylamine has $t_{1/2}$=0.25 d, captan has $t_{1/2}$=0.71 d, hexabromobenzene has $t_{1/2}$=73 d, and methomyl has $t_{1/2}$=230 d), Figure 3.4b shows that the variability across watersheds is mostly explained by the freshwater residence time to sea. While the fate factor of the short-lived n-nitrosodiethylamine is not influenced by freshwater residence time, fate factors for methomyl show a strong linear dependence on freshwater residence time. The distinction between emissions of hexabromobenzene in watersheds with a water depth below and above 1 m shows that outliers (watersheds for which fate factors are not limited by freshwater residence time) depend on the mean depth in the considered geographical unit that influences other loss processes, such as volatilization or deposition rates. The higher the freshwater depth, the lower is the volatilization and deposition rates and the greater the fate factor. Figure B.5a and b show the same analysis applied to intake fractions through water ingestion.



a) b)

Figure 3.4: Main factors affecting fate. a) Fate factors of all test substances as a function of their degradation half-life in water for each of the 136 watersheds of the European spatial model and b) Fate factors of n-nitrosodiethylamine, captan, hexabromobenzene (for emissions to watersheds with water depth below and above 1m) and methomyl as a function of water residence time to sea for each of the 136 watersheds of the European spatial model

Key parameters affecting exposure: To understand the main factors affecting the variability of the exposure factors XP_i [1/day] across Europe, we calculate for each watershed i an equivalent exposure factor through freshwater ingestion and fish ingestion by dividing the intake fraction IF_i by its fate factor FF_i:

$$XP_i = \frac{IF_i}{FF_i}$$

Equation 3.3: Exposure factor calculation

The resulting exposure factor can be interpreted as the equivalent fraction of the overall volume of water available that is taken in every day by the population.

Figure 3.5 presents the exposure factors by freshwater ingestion as a function of the freshwater residence time to sea for all watersheds of the spatial model, for the three archetypes and for the a-spatial model. We observe a clear inverse relationship between the exposure factor and the freshwater residence time to sea; for watersheds with a short residence time to sea, such as Brittany, the available volume of water is limited and thus the equivalent fraction of the water volume taken in every day by the population is high. As the freshwater residence time to sea increases, the fraction of water taken in is reduced by more than two (Danube) to three (Lake Geneva) orders of magnitude.

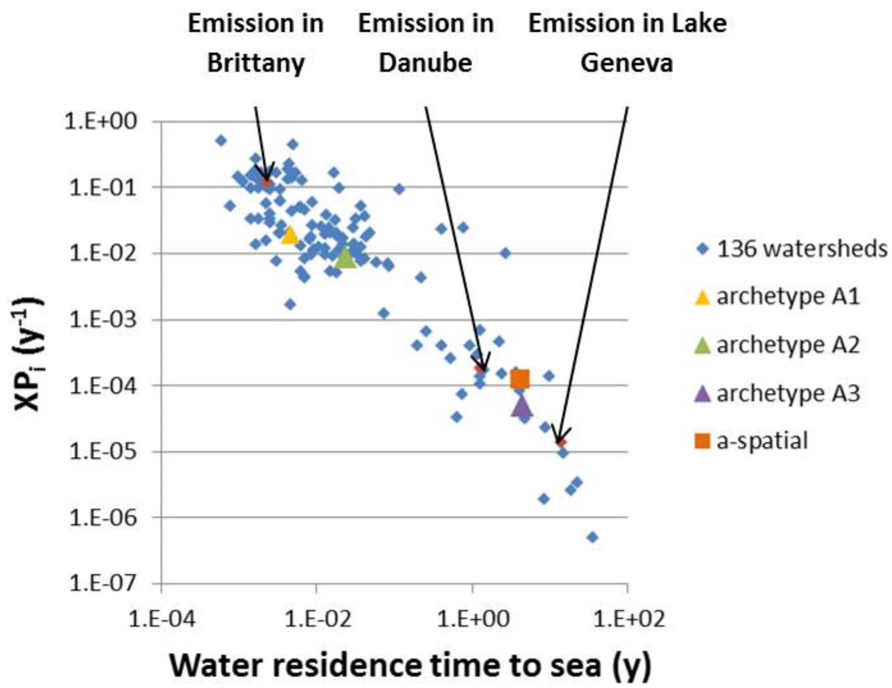


Figure 3.5: Variation in methomyl exposure factor from freshwater ingestion for the spatial, archetype and a-spatial models, where XPi is the exposure factor for an emission in watershed, archetype or a-spatial zone i

3.3.3 Freshwater archetype

Development of the archetype watersheds: The previous section has shown that the freshwater residence time to sea is a major determinant of both fate and exposure. This residence time to sea (or the model boundary) has thus been used to define three water archetypes by minimizing the variation between the log of freshwater residence times of the spatial model and the log of the mean value of the archetype watershed, as described in section 3.2.4. The resulting three archetypes have the following key characteristics. Archetype A1 covers coastal zones with short freshwater residence times, reaching the sea in less than 2 days, with an average value of 1.7 days, and a total volume of water of 0.073 m^3 per m^2 land area (also equal to the mean depth). Archetype A2 covers watersheds of medium freshwater residence times between 2 and 60 days, with an average of 8.6 days and a water volume of 0.27 m^3 per m^2 land area. Archetype A3 covers watersheds with freshwater residence times longer than 60 days, with an average residence time of 1600 days and a volume of water of 39 m^3 per m^2 land area.

Figure B.6 in the appendix presents the archetype classification of each of the 136 watersheds of the IMPACT Europe spatial model, as well as the geographical delimitation of the three newly developed archetypes.

Evaluation of the archetype model: We evaluated the performance of this watershed archetype model by comparing the resulting fate and intake fraction of each test pollutant in each archetype to the IMPACT Europe spatial model results for uniform and point source emissions (blue markers in Figure 3.3).

For a uniform emission, chemical fate factors of the archetype model are aligned with those of the Europe spatial model (Figure 3.3.a). For intake fractions through freshwater and freshwater fish ingestion, the archetype model also improves upon the a-spatial model estimation by substantially reducing the difference in results to within a factor two (Figure 3.3c and e). This reflects the improvement in both fate and exposure modelling related to population density compared to the a-spatial model, given that the archetype A1 is composed of coastal areas and thus mimics more adequately the intake fraction related to coastal zones modelled with the Europe spatial model.

For a point source emission, the archetype model represents a substantial improvement over the a-spatial model, with fate factors generally within a factor five of the spatial model (Figure 3.3b). This improvement is particularly stark for Brittany, because its freshwater residence time of 0.81 day is well approximated by the average residence time of 1.7 day in the archetype model.

Figure 3.3d and f show that the intake fractions are also improved in the archetype model, especially for a source emission in Brittany due to the modelling of higher exposure in coastal zones.

3.4 Conclusions

This paper develops continent-specific factors for the USEtox model and provides a first evaluation of the variability of fate and exposure induced by simplified approaches addressing spatial differentiation of toxic impacts such as: (1) region-specific nested models with a global surrounding box and (2) spatial archetypes based on key landscape parameters. It shows that simplifying models to either a nested model of continent-specific landscapes or to a model with a limited number of watershed archetypes still captures the main variability in impacts and may represent an efficient solution to account for spatial variations while limiting the complexity of the analysis.

Inter-continental variations in water residence time, fate and intake fraction are greater than one order of magnitude among the 17 zones assessed with the IMPACTWorld and USEtox models, due to differences in continent-specific landscape and population parameters. However, the model architecture of the surrounding box(es) (i.e., the single global box of USEtox or the interconnected continents of IMPACT) generally does not have a significant influence on results, with the exception of volatile and persistent pollutants in both air and water. A nested model, such as USEtox with a specific sub-continental parameterization, is thus well suited to model inter-continental variations in fate and exposure for most substances.

For *intra-continental* variation, an a-spatial model might substantially overestimate the chemical fate and characterization factors for freshwater eco-toxicity, by up to 3 orders of magnitude when compared to a spatially regionalized multimedia model representing the variations in hydrology and water use between watersheds. We identified freshwater residence time to sea as a key parameter affecting the variation in fate and exposure of persistent chemicals in water, and thus developed a set of three watershed archetypes based on this residence time as an alternative to a spatially differentiated model. This archetype model predicts aquatic fate and intake fractions within a factor 5 of those predicted by a detailed spatial model, while decreasing model complexity.

This work is an important step towards a regionalized assessment of toxic impact in the context of LCA and chemical screening. Further research work should be pursued to evaluate the robustness and the accuracy of the proposed archetypes applying a model with finer spatial resolution capabilities, whilst maintaining global coverage and multimedia modelling capacities. Models running on recently available hydrological datasets at 0.5° and 15’ should provide better basis to determine the optimal number of archetypes and further explore landscape key characteristics influencing fate and exposure across continents.

Acknowledgements

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Appendix

Appendix includes the detail of the developed sub-continental and continental landscapes, physic-chemical and toxicological properties of the selected set of substances, additional USEtox parameterization results, additional intake fraction variation results and the classification of IMPACT Europe spatial model watersheds into archetype categories.

Chapter 4

Spatial analysis of toxic emissions to freshwater in LCA: operationalization at global scale

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In preparation (soon to be submitted)

Abstract

There is an increasing interest to discriminate the impacts in relation to the place of emission in chemical fate and exposure models depicting the toxic emissions impact pathway. This work aims at exploring the operationalization of spatially differentiated models addressing toxic emissions into freshwater by analysing and comparing the variability of characterization factors at high resolution with aggregated factors at different levels of lower resolution.

We developed models to analyse the variation of fate and exposure factors for water ingestion as well as the main factors of influence: (1) a reference spatially resolved characterization model covering the freshwater media with global coverage at 0.5°*0.5° resolution and (2) two archetype models based on the main landscape key influential parameters addressing respectively impact on ecosystems and human health. We tested the validity of the latter models as well as different aggregation approaches on a case study of emissions from red mud disposal as a waste from alumina production.

The freshwater residence time to the sea was the most influential parameter with the equivalent depth, defined for a specific emission as the freshwater depth cumulative over all downstream cells, in particular for substances that sediment or complex with suspended solids or sediments. Results show that there are up to 3 orders of magnitude variation for fate and intake fraction across all 0.5°*0.5° grid cells in the case study of chromiumVI and arsenic emissions from alumina production. We propose four landscape archetypes defined as a function of their water residence time to the sea and their cumulative equivalent depth. These archetypes reflect well variation in fate, but are not able to reflect variation in exposure and in intake fraction. A population and sector-specific approaches were also tested to aggregate characterization factors from their native resolution to a coarser scale such as country, continent or global average. In practice, population weighted and sector-specific average characterization factors may represent the most operational way to account for specific distribution patterns of emissions.

Operationalization approaches exist that simplify the implementation of spatially differentiated methods addressing the impact of toxic emissions to water. Unlike air emissions that affect their direct surrounding, the influence of downstream cells prevents the efficient use of urban and rural archetypes. The alumina production case study shows that the determination of industry-specific weighted average represents a pragmatic way to account for sector specific location of emissions for goods that are traded at continental or global level. Population weighted approach is applicable to e.g. household emissions that are directly related to population density. This work also enables to evaluate the representativeness of the USEtox default generic characterization factor, where the

USEtox fate and intake fraction default values are found within one order of magnitude compared to the global value calculated by our sophisticated model, showing the relevance of this generic model for average situations.

Keywords

Life cycle assessment – human toxicity – ecotoxicity – spatial differentiation - freshwater

4.1 Introduction

Emissions of toxic substances can impact environment to various extents depending on their geographical location in addition to their intrinsic physico-chemical properties. When substances are emitted to surface freshwater, such as pharmaceutical substances released from wastewater treatment plants, or pesticides and fertilizers leaching from agricultural operations, the receiving body hydrology plays an important role in determining the magnitude of the impact (Pennington et al. 2005; Henderson et al. 2011). Substances removed from freshwater by advection to the sea are, for example, influenced by freshwater residence time to the sea (Kounina et al. 2014). When substances are adsorbed on sediments and settle at the bottom of the freshwater body, sedimentation occurs at various rates in lakes and rivers (Alexander et al. 2004). Evaporation depends on the exchange processes at the air-water interface area (Schwarzenbach et al. 2003). Furthermore, human exposure through water ingestion depends on the availability and type of wastewater treatment technology, as well as human population density, which varies substantially from coastal zones to desert areas.

Models predicting chemical fate and exposure are traditionally used in Life Cycle Assessment (LCA) to assess toxic impacts, with an increasing interest in including spatial differentiation in these models. The SimpleBox 3.0 (Den Hollander et al. 2004) and BETR North America models (Macleod et al. 2004) are multimedia and multi-pathway models that cover emissions to freshwater at a 50 km resolution on a European scale, and ecological zones on a North American scale (24 ecological zones cover the North American continent). The model GREAT-ER (Koormann et al. 2006) simulates the fate of substances in a single media, i.e. surface water, emitted to sewer systems and lakes or rivers for several watersheds in Europe. Models on finer resolutions provide precision up to 1*1 km grid cells in Europe for the MAPPE Europe model (Pistocchi et al. 2010) and sub-watershed resolution for freshwater emissions in Canada (Manneh et al. 2010). The latter model shows up to 10 orders of magnitude variability in human intake fraction (Manneh et al. 2010). A recent work focusing on the assessment of terrestrial ecotoxicity of copper and nickel differentiates among 760 noncalcareous soils from around the world spanning a wide range of properties, based on the scientific consensus model USEtox (Owsianiak et al. 2013). Kounina et al. (2014) analyzed the physico-chemical properties of substances emitted to water influencing regionalization and demonstrated that only persistent substances are influenced by regional parameters. Fate and exposure models developed so far for the assessment of toxic potential impacts from emissions to freshwater in LCA (1) lack differentiating between lake and river dynamics, where sedimentation and evaporation transfer processes significantly differ (the GREAT-ER model does not include lake dynamics) and (2) lack a global coverage as they traditionally are restricted to a continental scope. There is therefore a need to evaluate the variability of impact results from models with higher resolution, global coverage and distinguishing between river and lake. Hydrological datasets with global coverage exist at 0.5° x 0.5°(Vörösmarty et al. 2000a; Jolliet et al. 2012) and 15' resolution (Lehner et al. 2006), but are not yet implemented in LCA multimedia models.

The implementation of regionalized impact assessment methods for LCA practitioners raises several new constraints and dilemmas compared to a generic impact assessment. From the perspective of the modeller, model sophistication supporting higher spatial resolution allows quantification and minimization of the uncertainty related to spatial variability. However, this sophistication involves large computation times (Sedlbauer et al. 2007), higher model uncertainty and, for inclusion in LCA work, the knowledge about the geographical location of life cycle inventory flows. The following dilemmas are thus raised: (1) How can modellers apply detailed spatialized models to then provide approaches easily applicable for LCA practitioners? (2) How can the geographical resolution of impact assessment be adapted to the practitioner level of information for process location?

Two alternative approaches have been proposed so far to address several degrees of model sophistication: a) a top down approach proposing a set of simplified archetypes for different emission situations versus b) a bottom up approach proposing spatially resolved CFs and aggregation schemes to e.g. country or continental levels, based on emission weighting.

a) The top down archetype-based approaches that differentiate urban, rural, or remote emission locations, emission height, and indoor environment has previously been explored to estimate impacts of particulate matter inhalation (Humbert et al. 2011) and toxic substances inhalation (Hellweg et al. 2009). Kounina et al. (2014) developed watershed archetypes based on freshwater residence time to the sea to analyse intra-continental variation for emissions to freshwater at the watershed level in Europe. Although the watershed archetypes model captures the main variability within a factor 5, the authors recommend to evaluate the robustness and the accuracy of these findings with a model with finer spatial resolution and a global coverage.

b) The bottom-up approach can be illustrated by the IMPACT World+ methodology (Bulle et al. 2012), which represents a first attempt to consistently implement regionalized methods within a full LCIA framework operationalized in LCA software. CFs are aggregated from their native spatial resolution up to a global generic factor through national and continental resolution levels. Different aggregation schemes might be adopted. Emission weighting aggregates actual point source emissions within a given spatial unit when geographical location is known. Alternatively, an emission proxy can be adopted based on a default aggregation scheme, such as population (Bulle et al. 2012). Bourgault (2014) evaluated the variability of characterization factors at different levels of aggregation such as the country and archetype. He also evaluated the relevance of developing sector-specific aggregation schemes and argued in favour of the creation of industry-specific CFs to reduce the spatial variability. To consider specific geographic location of technosphere processes without aggregating characterization factors at a lower resolution, Mutel and Hellweg (2009) propose spatial methods to couple regionalized characterization factors with spatial life cycle inventory databases at the country level in existing software. In this approach, the available geographical information of the Life Cycle Inventory (LCI) determines the required level of aggregation of characterization factors, e.g., the grid cell, country, continent of generic level. There is now a need to develop and compare such approaches for toxic emitted to surface water and to test their applicability and operationalization at global scale on a case study.

This paper therefore aims at exploring further the importance and applicability of spatial resolution for toxic emissions into freshwater, by analysing and comparing the variability of CFs at the highly resolved grid cell level with different methods and levels of aggregation. The specific objectives are the following:

1. Develop a spatially resolved fate and exposure characterization model and factors for toxic emissions into water with global coverage at 0.5°*0.5° resolution;
2. Analyse the variation of fate and exposure factors for water ingestion as well as the main factors of influence on ecosystems and human exposure by focusing on five selected substances;
3. Develop archetypes based on the developed characterization model and identified key parameters;
4. Compare practical solutions to apply spatial characterization factors aggregated at different resolution scales in LCA and apply the different approaches to a case study of global aluminium production

4.2 Material and methods

4.2.1 Development of a fate and exposure model for toxic emissions with global coverage and at 0.5°*0.5° resolution

A human toxicity and ecotoxicity CF_i for a substance emitted in location *i* is calculated by multiplying the fate factor FF_i [year], with the exposure factor XF_j [year^{-1}] that varies depending on the receiving location *j* and the effect factor EF [cases.kg$_{intake}^{-1}$] independent from the emission location (Rosenbaum et al. 2008):

$$CF_i = \sum_j (FF_{i,j} * XF_j) * EF = iF_i * EF$$

Equation 4.1: Calculation of the characterisation factor in an emission cell *i*

In current methods, the influence of location on population sensitivity to toxicants (including genetics, age and gender) is often neglected when assessing the effect factor for human toxicity and aquatic ecotoxicity factor (Pennington et al. 2006), we thus consider EF without geographical differentiation. The largest spatial variation stems thus from FF_i and XF_j. iF_i [kg$_{intake}$.kg$_{emitted}^{-1}$] is the human intake fraction through inhalation and ingestion and is calculated as the product between FF_i and XF_j.

Fate factor

FF_i is the multiplication of the substance persistence FF_j [year] in freshwater in a receptor cell *j* and the substance fraction transferred from the source grid cell *i* to receptor grid cell *j*, $f_{i,j}$ [-]:

$$FF_i = \sum_j f_{i,j} * FF_j$$

Equation 4.2: Calculation of the fate factor in an emission cell *i*

Vörösmarty et al. (2000a) provide a 0.5*0.5° global hydrological model that was used by Helmes et al. (2012) to assess freshwater eutrophication by phosphorus emissions. Helmes et al. (2012) provided worldwide fate factors for phosphorus emissions to freshwater from point sources to the ocean on the same resolution. Their model includes substance advection from grid cell upstream to downstream cells, as well as differentiation between phosphorus removal rates (by sedimentation and biological uptake) between lakes and rivers. We adapted and extended the approach of Helmes et al. (2012) to develop a toxicity model using the 0.5°*0.5° gridded river network from Vörösmarty et al. (2000a). We modelled the chemical fate for toxic emissions based on the freshwater advection and particle settling rates in rivers modelled by Helmes et al. (2012), and we introduced the removal processes relevant for toxics, namely chemical and biological degradation and volatilization. We calculated the overall persistence FF_j [year] of a substance in freshwater in a receptor cell *j* as the inverse sum of the removal rates by advection $k_{adv,j}$ [year^{-1}], sedimentation $k_{sed,j}$ [year^{-1}], evaporation $k_{evap,j}$ [year^{-1}] and degradation $k_{deg,j}$ [year^{-1}]:

$$FF_j = \frac{1}{k_{adv,j} + k_{sed,j} + k_{evap,j} + k_{deg}}$$

Equation 4.3: Calculation of the substance persistence in a receptor cell *j*

$f_{i,j}$ is calculated as per the original model as the product of all substance fractions transported from the emission grid cell i to the receptor cell j (Helmes et al. 2012).

Table 4.1 presents the model characteristics and assumptions used to calculate each removal rate used in Equation 4.3.

Table 4.1: Description of fate parameters: advection rate, sedimentation rate, evaporation rate and degradation model, assumptions and references

Removal rate	Advection rate	Sedimentation rate	Evaporation rate	Degradation rate
Variable name	$k_{adv,j}$ [year^{-1}]	$k_{sed,j}$ [year^{-1}]	$k_{evap,j}$ [year^{-1}]	k_{deg} [year^{-1}]
General equation	$k_{adv,j} = \frac{Q_{grid,j}}{V_{grid,j}}$ **Equation T4.1.1**	$k_{sed,j} = \frac{V_{riv,j} * k_{sed\ riv,j} + A_{lak,j} * v_{sed\ lake}}{V_{grid,j}}$ **Equation T4.1.2**	$k_{evap,j} = \frac{A_{grid,j} * v_{evap}}{V_{grid,j}}$ **Equation T4.1.4**	$k_{deg} = \frac{ln2}{t_{1/2}}$ **Equation T4.1.5**
Variables used in the equation	$Q_{grid,j}$: Total freshwater discharge from grid cell j [km^3.year^{-1}] $V_{grid,j}$: Total freshwater volume in grid cell j [km^3]	$V_{riv,j}$: river volume in grid cell j [km^3] $k_{sed\ riv,j}$: sedimentation rate in rivers [year^{-1}] $A_{lak,j}$: lake area in grid cell j [km^2] $v_{sed\ lake}$: sedimentation speed in lakes [km.year^{-1}]	$A_{grid,j}$: freshwater area in grid cell j [km^3] v_{evap}: evaporation speed in freshwater [km.year^{-1}]	k_{deg}: degradation constant [year^{-1}]
Details	$Q_{grid,j}$ and $V_{grid,j}$ are based on the hydrological model of Vörösmarty et al. (2000a) integrated in Helmes et al's (2012) eutrophication model.	Following Helmes et al.'s (2012) model, we differentiate sedimentation in lakes and rivers by using specific sedimentation rates in rivers $k_{sed\ riv,j}$ [year^{-1}] and sedimentation speed in lakes $v_{sed\ lake}$. While sedimentation in lakes can be described as completely mixed boxes, transport processes in rivers and streams are dominated by the unidirectional flow of the water which creates resuspension and desorption (Schwarzenbach et al. 2003). ***Rivers*** $k_{sed\ riv,j} = k_{sed\ riv\ phos,j} * \frac{1-f_{diss\ subs}}{1-f_{diss\ phos}}$ **Equation T4.1.3** $k_{sed\ riv\ phos,j}$: sedimentation rates for phosphorus in rivers based on Alexander et al. (2004) and calculated by the SPARROW model. It includes both loss from biological uptake and from sedimentation and vary from 71.2 [year^{-1}] for small streams (flow < 2.8 m^3s^{-1}) to 4.38 [year^{-1}] for large streams (flow > 14.2 m^3s^{-1}). We use $k_{sed\ riv\ phos,j}$ as a reference for the removal rate of all substances, applied to the particulate fraction of the substance according to Schwarzenbach (2003) . $(1 - f_{diss\ phos})$: fraction of phosphorus sorbed on the sediment surface . $f_{diss\ phos}$ is the fraction of phosphorus dissolved in water, i.e. 0.19 based on the average of free PO_4^{2-} compared to dissolved organic matter bound and particulate bound phosphorus sampled in the Vansjø catchment, Norway (Parekh 2012). Details of the raw data and calculation to get the average of phosphorus dissolved in water 0.19 are detailed in appendix C.1. $1 - f_{diss\ subs}$: fraction of substance sorbed on the sediment surface ($f_{diss\ subs}$ being the fraction of the assessed substance dissolved in water based on the USEtox database (Rosenbaum et al. 2008)) ***Lakes*** $v_{sed\ lake}$ is based on the substance specific sedimentation speed calculated in USEtox (Rosenbaum et al. 2008), taken for most substances from the SimpleBox model (Den Hollander et al. 2004)	v_{evap} is based on the substance specific evaporation speed calculated in the USEtox model (Rosenbaum et al. 2008), taken for most substances from the SimpleBox model (Den Hollander et al. 2004)	k_{deg} is based on substance specific degradation rate calculated in the USEtox model (Rosenbaum et al. 2008), taken for most substances from EPIsuite (U.S. Environmental Protection Agency 2012)

Exposure factor and intake fraction

The intake fraction iF_i [$kg_{intake}.kg_{emitted}^{-1}$] is calculated for water ingestion by the multiplication of the freshwater fate factor FF_i (i.e. by substance persistence FF_j [year]) in a receptor cell *j* multiplied by the substance fraction transferred from the source grid cell *i* to receptor grid cell *j*, $f_{i,j}$, by the exposure factor XP_j [year^{-1}] in receptor cell *j* as follows:

$$iF_i = \sum_j (f_{i,j} * FF_j * XP_j)$$

Equation 4.4: Calculation of the intake fraction in an emission cell *i*

Focusing on the drinking water ingestion pathway, XP_j [year^{-1}] represents the yearly equivalent fraction of the chemical mass in water media ingested by the population. It is calculated based on the fraction of substance dissolved in freshwater $f_{diss\,subs}$ [-] (tap water is assumed to be treated to remove the particulate fraction), the volume of water ingestion per person $V_{ing\,wat}$ equal to 511 [l/(person*year)], the population in the receptor cell *j* $N_{pop,j}$ [person], the ratio of surface water over the total water withdrawal r_{surf}, the water density (equal to 1000 [kg/m^3]), and the volume of the freshwater compartment $V_{grid,j}$ [m^3)] (Rosenbaum et al. 2008) as:

$$XP_j = \frac{f_{diss\,subs} * V_{ing\,wat} * N_{pop,j} * f_{surf,j}}{\rho_{wat} * V_{grid,j}}$$

Equation 4.5: Calculation of the exposure factor in a receptor cell *j*

The population data $N_{pop,j}$ per 0.5°*0.5° grid cell is based on a population density grid provided by the Center for International Earth Science Information Network (CIESIN 2005). The fraction of surface water over the total water withdrawal $f_{surf,j}$ per country is based on the Quantis water database (Quantis 2012) compiling Aquastat data with the most recent data available between 1975 and 2010 (FAO 2014).

The exposure factor through drinking water XP_j is thus calculated based on the water requirement of the receptor cell, disregarding local water exploitability. In case of a water scarce area, the amount of water consumed for drinking water can exceed the amount of water available in a given grid cell *j*, and IF_j could thus potentially reach or exceed 1. In reality, drinking water will be supplied by a neighboring grid cell or from a more distant location through bottled water. To avoid the model artefact of artificially high iF values, we introduced a threshold value that cannot be exceeded within a single grid cell j ($IF_{max,j}$) to account for water-scarce grid cells. $IF_{max,j}$ is determined by considering the fate factor $FF_{j,modified}$ in a receptor cell *j* that includes freshwater removal rate $k_{withdrawn,j}$ [year^{-1}] in addition to other removal processes defined in Equation 4.3 $k_{adv,j}$, $k_{sed,j}$, $k_{evap,j}$ and k_{deg} [year^{-1}]. This addition reflects the fact that drinking water withdrawn is not available in the same grid cell, while it can be reused in grid cells downstream (drinking water is withdrawn in a centralized system for large cities). $XP_{j,max}$ [year] is the maximum exposure factor calculated using modelled industrial, domestic and agricultural withdrawal. $IF_{max,j}$ is defined as:

$$IF_{max,j} = FF_{j,modified} * XP_{j,max} = \frac{1}{\frac{1}{FF_j * k_{withdrawn,j}} + 1} * f_{diss\,subs} * f_{drink\_dom,j} * f_{dom\_tot,j}$$

Equation 4.6: Calculation of the maximum intake fraction in a receptor cell *j*

The derivation of Equation 4.6 is explained in appendix C.2.

4.2.2 Variation of fate and exposure factors and main factors of influence on ecosystems and human exposure

Substance selection: We selected five organic and inorganic substances to represent the following types of substances: a) highly degradable substances - represented by the pharmaceutical substances mannitol, b) persistent substances that have low evaporation and low sedimentation rates - the fungicide captafol and c) persistent substances with higher sedimentation removal rates - the detergent tinopal. We additionally selected chromiumVI and arsenic as they are the most important contributors to toxicity impact of aluminium production of the case study presented in section 4.2.4. Table 4.2 shows the selected substances as well as their degradation, sedimentation, evaporation rates and their dissolved fraction. Figure C.2 in appendix C.3 shows the distribution of degradation, sedimentation and evaporation rates of all organic and inorganic substances covered by the USEtox organic and inorganic database (Rosenbaum et al. 2008). Since there can be significant variations of metal mobility and solid/liquid partitioning over very small geographic scales depending on pH, dissolved organic carbon, water hardness and dissolved mineral concentration (Gandhi et al. 2010), we tested the influence of the variability of the partitioning coefficients that may vary substantially, namely the partition coefficient between dissolved organic carbon and water K_{DOC} and between suspended solids and water $K_{P_{SS}}$ on the sedimentation rate (Allison and Allison 2005). Indeed, the more a metal will combine with organic carbon or suspended solids, the more it will tend to sediment.

Table 4.2: Physico-chemical properties of the five selected substances in USEtox

CAS	Chemical substance	k_{deg} (year^{-1}) (values below 1E-10 were considered as 0)	$k_{sed\ lake,j}$ (year^{-1})[1]	$k_{evap,j}$ (year^{-1})[1] (values below 1E-10 were considered as 0)	Dissolved fraction (-)
69-65-8	**Mannitol**	108 (less than 3 [days] for the chemical half-life)	0.26	3.7E-10	1.0
242-50-61	**Captafol**	1.4	0.16	4.3E-3	0.99
13863-31-5	**Tinopal**	1.4	60	0	1.1E-3
18540-29-9	**Chromium(IV)**	0	1.4	0	0.81
7440-38-2	**Arsenic**	0	2.0	0	0.89

Analysis of fate factors, exposure factors and intake fractions: Based on influential spatial parameters identified in previous work (Pennington et al. 2005; Kounina et al. 2014), we analysed the variation of fate factors FF_i and intake fractions IF_i through water ingestion for the five selected substances depending on water residence time to sea, water depth and cumulative exposure factor. We introduce the cumulative exposure factor XP_i for an emission cell *i* (summing over the downstream *j* cells). XP_i is specifically developed to analyse the relative contribution of exposure factors to the IF_i and is calculated as:

$$XP_i = \frac{IF_i}{\sum_j FF_i}$$

Equation 4.7: Calculation of the cumulative exposure factor in an emission cell *i*

With FF_i and IF_i defined in Equation 4.2 and Equation 4.4. XP_i strongly depends on the substance persistence and sedimentation, as it considers all the XP_j values for the cells through which the substance passes.

Development of the cumulative equivalent depth parameter: Given the importance of the cells' water depth grid cell to substance fate, demonstrated in section 4.3.2, we defined a new parameter $d_{i\ equ}$ [m] being the cumulative equivalent depth for an emission in cell *i* for a given substance.

The overall cumulative residence time of a substance for an emission in cell *i* can be approximated as:

$$FF_i = \frac{1}{k_{adv,i} + k_{deg} + k_{sed,lake,i} + k_{evap,i}} = \frac{1}{k_{adv,i} + k_{deg} + \frac{v_{sed,lake} + v_{evap}}{d_{i\ equ}}}$$

Equation 4.8: Approximation to calculate the substance residence time in an emission cell *i*

[1] This value is cell dependent and calculated here for a water depth of 2.5m.

The inversion of this equation solved for $d_{i\ equ}$ gives:

$$d_{i\ equ} = \frac{v_{sed,lake} + v_{evap}}{k_{tot} - k_{adv,i} - k_{deg}} = \frac{v_{sed,lake} + v_{evap}}{\frac{1}{FF_i} - \frac{1}{\tau_{adv,i}} - k_{deg}}$$

Equation 4.9: Cumulative equivalent depth in an emission cell *i*

$d_{i\ equ}$ is determined to grasp emissions cells *i* with an important sedimentation speed, $v_{sed\ lake}$ [m.year^{-1}] is thus used as a constant proxy that does not depend on landscape parameters to determine the overall sedimentation speed. Using Equation 4.9 and having already determined the cumulative substance residence time for an emission in cell i FF_i [year] and the water residence time to the sea $\tau_{adv,i}$ [year] by running the model, it becomes possible to calculate for each emission cell i its cumulative equivalent depth for a given substance as a function of its degradation rate k_{deg}, the sedimentation speed $v_{sed,lake}$ and the evaporation speed v_{evap}.

4.2.3 Development of grid cell based archetype landscapes

Four archetypes are defined based on the identified influential parameters for chemical fate and two for human exposure. An archetype specific box model is parameterized for each archetype. To do so, each 0.5*0.5° grid cell of the characterization model has been attributed to its corresponding fate of exposure archetype. The freshwater volume, area, depth and freshwater residence time to sea of each archetype box are then calculated based on the sum or average of the characteristics of 0.5*0.5° grid cells included in a defined archetype. Advective flows between two archetype boxes were calculated by summing the advections between 0.5*0.5° grid cells classified in respective boxes.

4.2.4 Case study on aluminium production and aggregation approaches

Aluminium is the most widely used non-ferrous metal, with over 40 million tons of pure aluminium metal produced annually for the transport, construction, packaging and electrical sectors (Tabereaux and Peterson 2014). It is produced through two main processes: the Bayer process for alumina production (used for over 90% of the world alumina production (Tabereaux and Peterson 2014)) and the Hall-Héroult electrochemical process to produce pure aluminium. The dominant toxic impacts of primary aluminium production stem from heavy metals leaching during red mud disposal to residual material landfill from bauxite digestion in the Bayer process. Analyzing the process "Aluminium, primary at plant" for Europe from the ecoinvent v2.2 database (Frischknecht et al. 2005) with the USEtox v.1.01 method (Rosenbaum et al. 2008) shows that 64% of the total (cancer and non-cancer) human health impact cancer and 77% of the ecotoxic impact are due to chromiumVI and arsenic ion emissions to water during red mud disposal. The complete results of the contribution to toxicity are detailed in appendix C.4. This disposal takes place close to alumina refineries, whose coordinates were identified based on global data from the International Aluminium Institute (IAI 2012) and (Alcor Technology 2013). The coordinates and annual alumina production (up to 6'300 kt/year) are presented in Figure 4.1.a and detailed in appendix C.5.

We determined fate factors and intake fractions at different levels of resolution: (1) 0.5°*0.5° grid cell (when the location is known), (2) fate or exposure archetype, (3) country (when only the country is known), (4) continental level (when only the continent is known) and (5) global level (when the location is unknown). For these various resolutions, we explored two approaches to aggregate CFs from their native resolution to a coarser lever: a production and population based approach. The production based approach relies on point source emissions and relies on the assumption that chromiumVI and arsenic emissions from the red mud disposal is proportional to alumina production (assuming similar technologies across the world for the Bayer process) and takes place in the same location as the alumina refinery. The population-based approach as per the regionalized method IMPACTWorld+ (Bulle et al. 2012) is based on the assumption that emissions occur where humans live and are proportional to human population.

We aggregated the FF_i and iF_i results for each 0.5°*0.5° grid cell on different geographical units X of each resolution level: country, archetype and continental level as follows:

$$FF_{i,X} = \frac{\sum_{i \in X}^{n}(FF_{i,0.5} * N_{prod\ or\ pop\ ,i,0.5})}{\sum_{i \in X}^{n} N_{prod\ or\ pop,i,0.5}}$$

Equation 4.10: Aggregation of the fate factors on a higher resolution

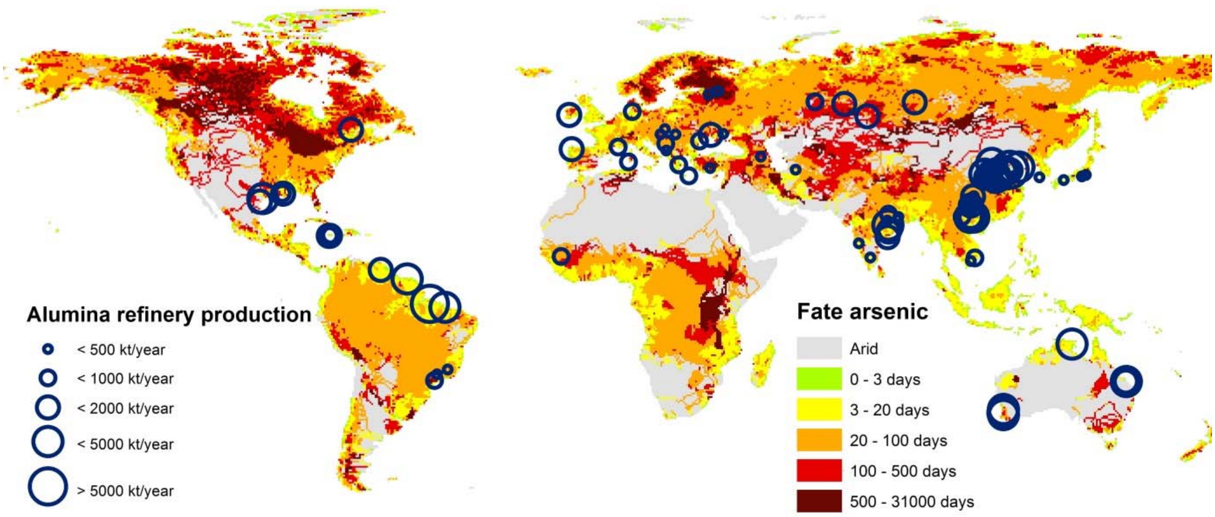
Where $FF_{i,0.5}$ represent the grid cell fate factor within a defined geographical unit X and $N_{prod\ or\ pop,i,0.5}$ corresponds to the alumina production or the population within this grid cell. $\sum_{i \in X}^{n} N_{prod\ or\ pop,i,0.5}$ represents thus the total production or population within the defined geographical unit X.

4.3 Results and discussion

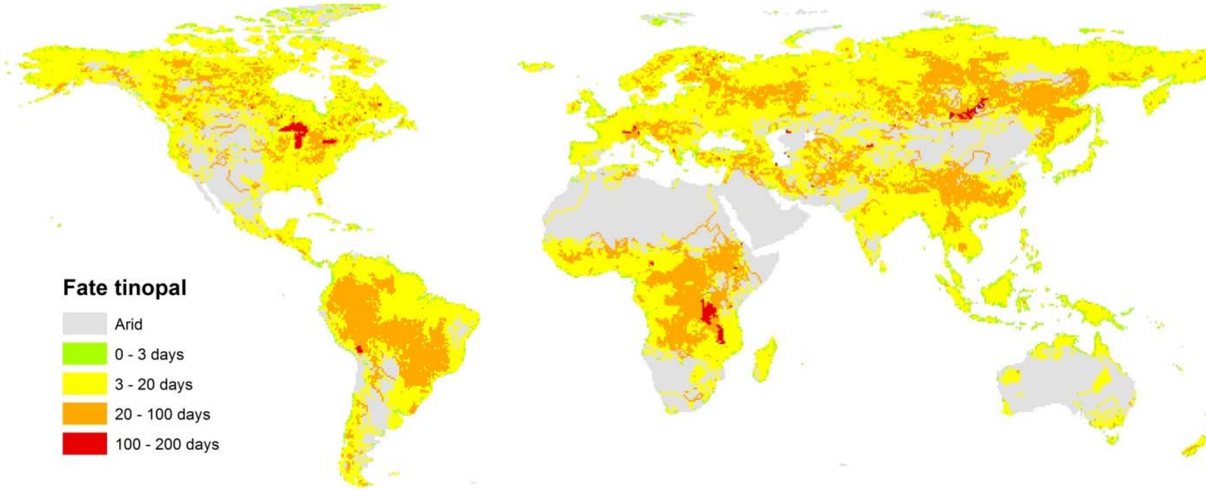
4.3.1 Fate and exposure of toxic emissions to water with global coverage at 0.5°*0.5° resolution

Fate: Figure 4.1.c characterizes the world water hydrology at the 0.5°*0.5° resolution, presenting for each cell its freshwater residence time until it reaches the sea, with arid zone in grey for which yearly potential evaporation exceeds rainfall. As expected low water residence time to the sea are found in coastal areas. Regions with high residence times (>500 days) cover 17% of overall none-arid cells and correspond to regions with lakes and large river water basins. We also calculated the equivalent depth for all chemicals. Figure 4.1.d shows the equivalent depth of tinopal on a 0.5°*0.5° resolution. It shows that a large part of the non-arid territory has an equivalent depth greater than 3 m. Equivalent depths superior to 100 m occur in grid cells with large lakes where fate values are superior to 100 days.

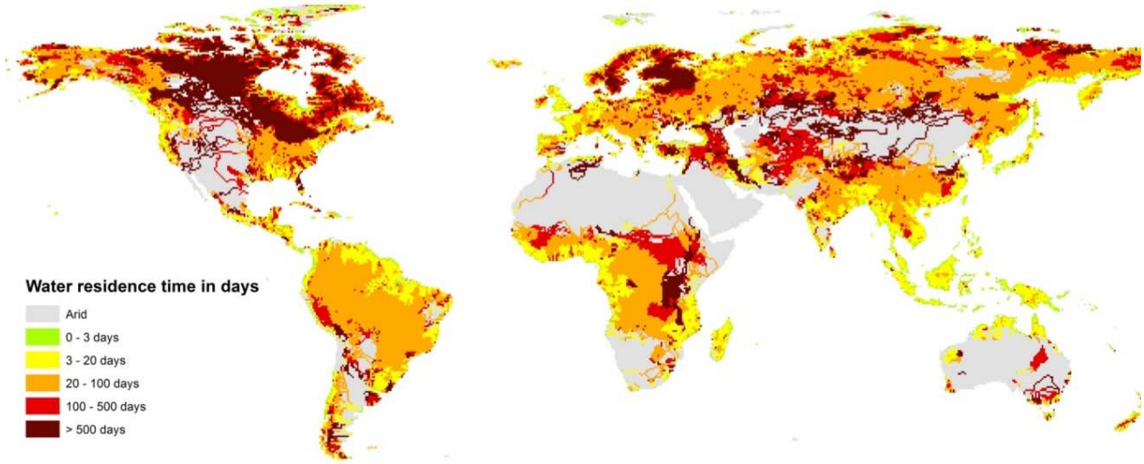
Fate factors, i.e. the substance residence time in the freshwater compartment are determined for the five selected substances. Figure 4.1.a maps the fate factors of arsenic for an emission into water in each of the 0.5°*0.5° cell worldwide. Fate values less than 3 days occur in coastal areas throughout the world. The regional patterns of the fate factors mostly reflect the freshwater residence time map (Figure 4.1.c). As a rather persistent substance with high sedimentation removal rates, tinopal tends to have shorter residence time than arsenic, due to its substantially higher sedimentation rate (Table 4.1). Fate values larger than 100 days take place in large and deep lakes such as the large North American and African lakes, European lakes in the vicinity of the Alps and the Baikal lake, suggesting that not only advection but also water depth plays an important role. Indeed the map for tinopal is very similar to the map of equivalent depth provided in Figure 4.1.d. Results for mannitol, captafol and chromiumVI are presented in appendix C.6, showing that the fate values of quickly degradable mannitol do not exceed 4 days, while about 30% of the fate values of captafol and chromiumVI are higher than 100 days.



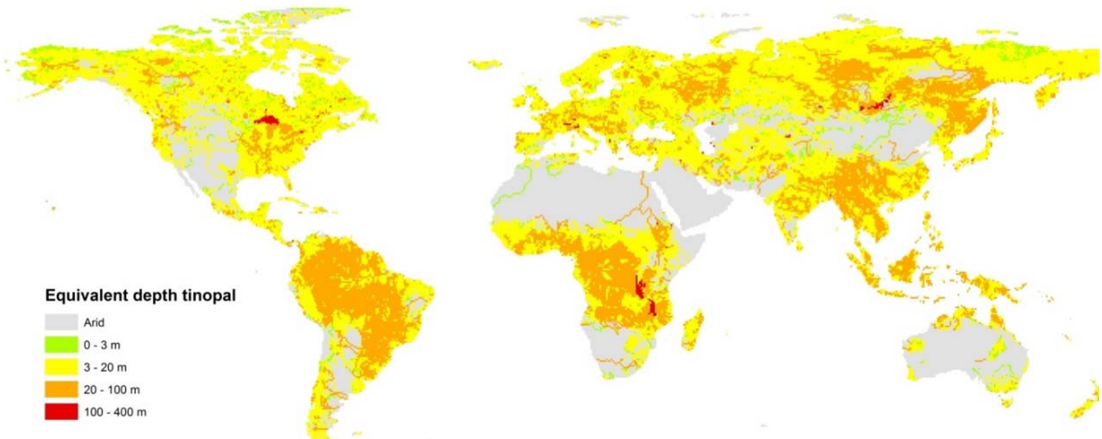
a) Fate of arsenic and location of alumina refineries



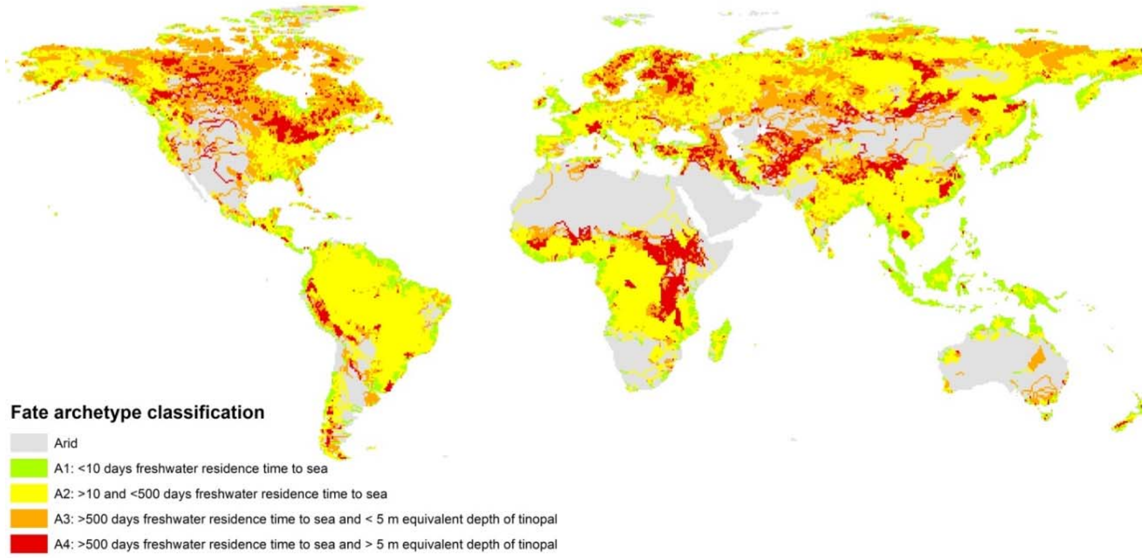
b) Fate of tinopal



c) Water residence time to the sea



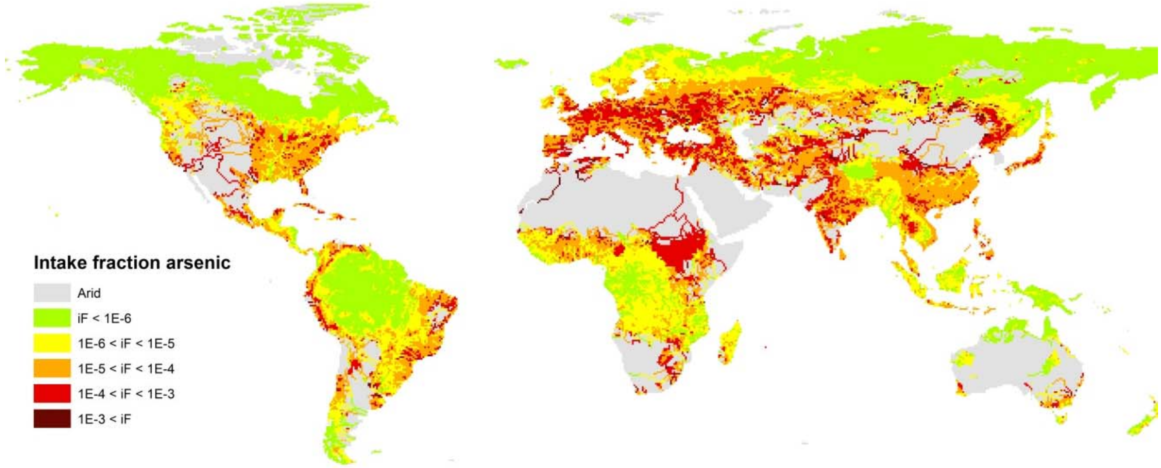
d) Equivalent depth of tinopal



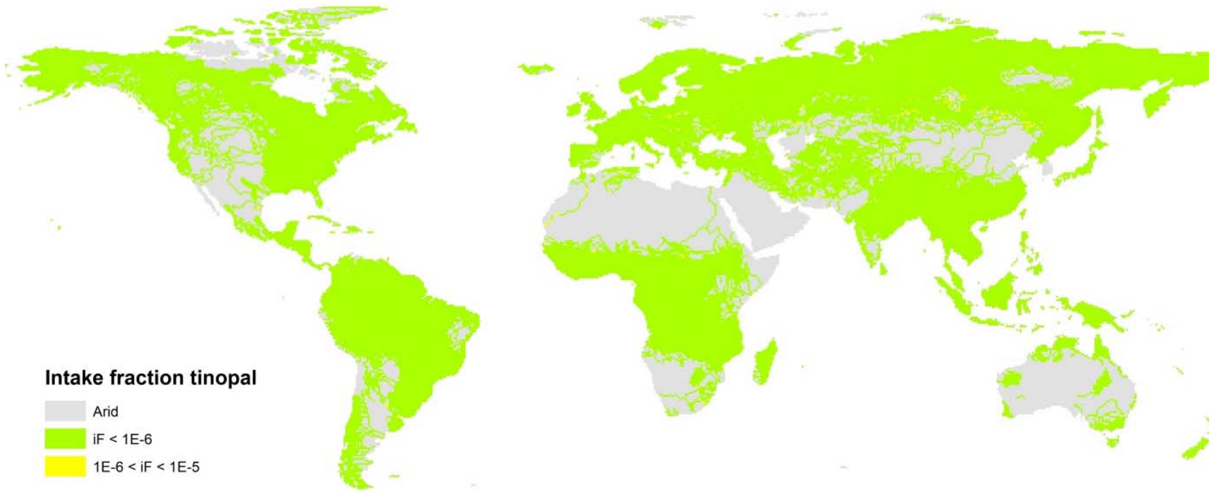
e) Fate archetype classification

Figure 4.1: Maps of fate factor in freshwater, i.e. substance residence time, and of main parameters of influence for (a) arsenic with location of alumina refineries sites and their production weight (b) for tinopal, (c) for the water residence time the sea, (d) for tinopal equivalent depth and (e) for freshwater fate archetype classification

Intake fractions: Figure 4.2.a and b present the intake fraction of arsenic and tinopal respectively. Given that the dissolved fraction of tinopal is as low as 0.001 (see Table 4.2) and that its fate is limited by sedimentation, the intake fraction calculated according to Equation 4.4 and Equation 4.6 rarely exceed 1E-6 [$kg_{ingested}/kg_{emitted}$] and never exceeds 1E-5. The intake fraction of arsenic is substantially higher and go up to 0.01 (as well as mannitol, captafol and chromiunVI), limited by $IF_{max,j}$ defined in Equation 4.6 based on the figures in appendix C.6.



a) Arsenic intake fraction



b) Tinopal intake fraction

Figure 4.2: Arsenic intake fraction by water ingestion (a) and tinopal intake fraction by water ingestion (dimensionless) (b), in each 0.5°*0.5° grid cell modeled on a global resolution

4.3.2 Analysis of the variation of fate and exposure factors as well as the main factors of influence on ecosystems and human exposure

Fate: We analyzed the wide variation in fate of the reference substances observed in the maps of the previous section in plotting them as a function of the freshwater residence time to the sea, for equivalent water depths higher and lower than 10 m (Figure 4.3.a and b).

Figure 4.3.a shows that the fate of mannitol is limited to 0.01 [year] due to its high degradability. The fate of captafol linearly increases with the water residence time to the sea (which can be interpreted as a limitation by the advection rate) until it reaches 1 [year] where it becomes constant, limited by the degradation rate. This value of 1 [year] is due to its high persistence (k=1.41 year^{-1}, that corresponds to a degradation half-life of $t_{1/2}$=0.49 [year]) and to low sedimentation and evaporation rate. The fate of tinopal and chromiumVI are also limited by advection to the sea as show the upper values linearly dependent on the freshwater residence time. Chemical fate values are lower in some grid cells due to the removal though other mechanisms than advection such as complexation with suspended solids, organic matter or sediments and ultimately sedimentation. Low fate factors are associated with cells with a water depth less than 10 m. We performed a complementary analysis of the variation of fate depending on freshwater depth in appendix C.7.

We tested the sensitivity of the model to a range of partitioning coefficients between dissolved organic carbon as well as water suspended solids and water, K_{DOC} and $K_{P_{SS}}$, that directly affect the sedimentation rates of chromiumVI and arsenic. K_{DOC} values available in the literature vary between 1 and 1'000 L.kg^{-1} with a mean value of 100 and $K_{P_{SS}}$ from 100 to 1'000'000 with a mean value of 7'940 L.kg^{-1} for arsenic (Allison and Allison 2005). These means correspond to the reference values used in the USEtox inorganic database (Huijbregts et al. 2010). On the one hand, K_{DOC} variability is responsible for less than 0.01% change in the reference sedimentation rate of 1.99 year^{-1}, only leading to minor changes lower than 0.1% in the fate factor. On the other hand, $K_{P_{SS}}$ strongly influences the sedimentation rate, more than a factor of 200, with sedimentation rates ranging from 0.077 to 17 year^{-1}. This variation in sedimentation rate also affects the fate factor by up to a factor 100, a high $K_{P_{SS}}$ corresponding to a high sedimentation rate and thus to a lower fate factor down to 1% of its reference value.

Intake fraction: The intake fraction is plotted as a function of the cumulative exposure factor XP_i (Figure 4.3.c and d) of each substance. For each of the considered substance, IF_i via drinking water varies by more than 20 orders of magnitude, primarily due to the high variations of the exposure factor and in a lesser extent related to the fate.

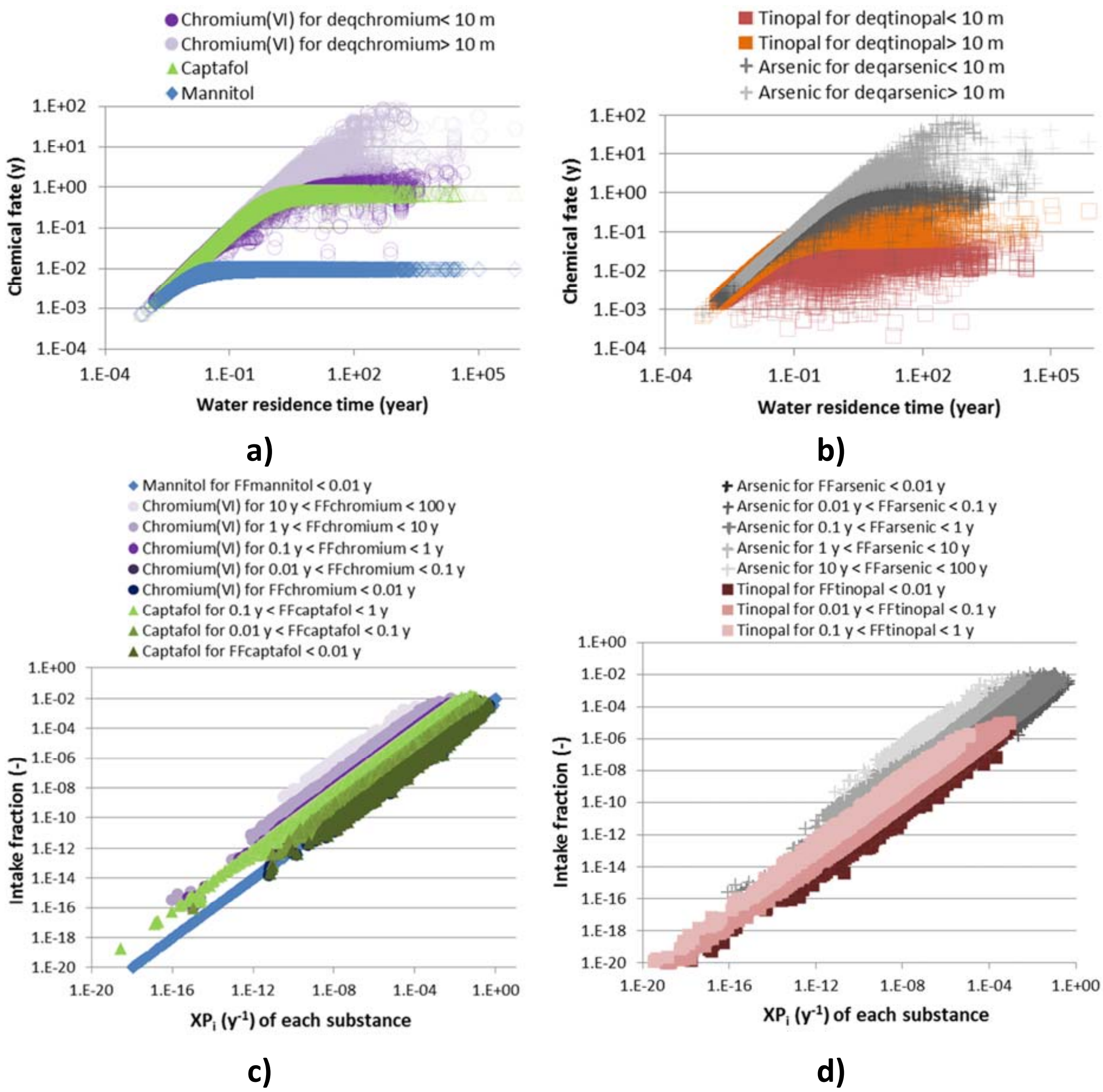


Figure 4.3: Fate (a and b) and intake fraction (c and d) of five selected chemicals (mannitol, chromiumVI, captafol, arsenic and tinopal, vs. water residence time until the sea (a and b) and reference exposure factor (c and d) in each 0.5°*0.5° grid cell

In appendix C.8, we also analyse the variation of fate versus equivalent water depth of tinopal for tinopal, captafol, mannitol, arsenic and chromiumVI. Tinopal was selected as a reference substance to estimate equivalent depth for the representativeness of its physico-chemical properties: a low degradation and high sedimentation rate according to Table 4.2. We recalculate the fate of selected substances based on the equivalent depth of tinopal and conclude that this substance-specific parameter can be used as a generic proxy for fate estimation.

4.3.3 Grid cell based archetypes

Fate: Based on the previous analysis, water residence time and equivalent depth were found to be influential parameters for fate. We used these parameters to develop a model with four archetypes: A1 from 0 to

10 days of freshwater residence time, A2 from 10 to 80 days of freshwater residence time and A3 and A4 with more than 80 days of freshwater residence time and respectively less and more than 10 m equivalent depth of tinopal. Four archetypes were selected as a trade-off between precision and applicability, according to the following considerations. Coastal grid cells classified in A1 are dominated by advection and are thus grouped according to their freshwater residence time to the sea, inferior to 10 days. A2 was delimited based on the geometric mean of freshwater residence time to the sea in all grid cells, equal to 81.8 m and approximated to a threshold of 80 m. A3 and A4 with a water residence time superior to 80 days cover fate values that vary up to 3 orders of magnitude depending on the equivalent depth. The threshold to delimit A3 and A4 based on the equivalent depth of tinopal was set to 10 m as a rounding of tinopal equivalent depth geometric mean over all 0.5*0.5° grid cells equal to 11.3 m.

We recalculated the fate and intake fraction for selected substances in these two archetype models. Fate results are based on Equation 4.8 applying the equivalent depth of tinopal and substance specific evaporation, sedimentation and degradation rates. Intake fraction results were generated based on Equation 4.4 and Equation 4.5.

Table 4.3 shows hydrological parameters for the six developed archetypes: freshwater volume, surface, depth and residence time to the sea. Water depth for A1, A2 and A3 is between 4.8 and 5.6 m while it reaches 77.3 m for A4. A large set of water residence time to the sea are also represented: between 7.5 days for A1 and almost 20 years for A4.

Table 4.3 : Fate and intake fraction archetype models hydrological and exposure parameters

	Freshwater volume	Freshwater surface	Freshwater depth	Freshwater residence time	$\frac{XP_i}{f_{diss\ subs}}$
Unit	km^3	km^2	m	day	day^{-1}
A1	7.24E+02	1.29E+05	5.6	7.5	Not calculated
A2	2.24E+03	4.68E+05	4.8	29.3	Not calculated
A3	3.74E+03	7.55E+05	5.0	292.3	Not calculated
A4	9.09E+04	1.18E+06	77.3	7.27E+03	Not calculated
Rural	9.64E+04	2.47E+06	39.0	760.4	4.30E-08
Urban	1.16E+03	5.51E+04	21.0	29.0	2.63E-06

Figure 4.1.e maps the geographic location of each of these fate archetypes. A1 is located on coastal areas, and A3 and A4 correspond to the locations with high fate factors.

The performance of these archetypes is tested and compared to the spatial model in appendix C.9. These fate archetype results succeed in reducing fate variability to 2 orders of magnitude compared to 5 orders of magnitude of variability among all spatial results. They are however not easier to link to a life cycle inventory than the high resolution 0.5°*0.5° model , since in both case the location of emission needs to be accurately known.

Intake fraction: We also generated archetypes for influential parameters on the intake fraction. We proved in part 4.3.2 that XP_i is a key cause-effect chain modeling driver of iF_i, that depends on regional parameters $N_{pop,j}$ and V_{grid} as shown in Equation 4.5. Humbert et al. (2011) set up archetypes to distinguish rural and urban landscapes for particulate matter emissions based on the threshold of 390 people.km^{-1}, defined by the US Census Bureau (U.S. Census Bureau 2014). We use the same reference to create a simplified two urban and rural archetype landscapes following the same modelling principles as for the fate archetypes. The $\frac{XP_i}{f_{diss\ subs}}$ score varies by 2 orders of magnitude for the rural and urban landscapes, i.e. respectively 4.30E-08 day and 2.63E-06 day and the archetype map is provided in Figure C.5 of the appendix.

We recalculated the intake fraction for selected substances in these two archetype models. The distinction between rural and urban intake fraction poorly represents the variation of the exposure factor, leading to a difference of up to 10 orders of magnitude compared to the spatial model results. These differences are due to the strong hydrological connection between urban and rural cells, with 91% of freshwater advected from rural to urban landscapes and 60% in the other direction. Persistent substances can thus pass through both compartments if their fate exceeds the compartment residence time, i.e. 760 days for the rural box and 29 days for the urban one. These observations support the conclusion that the relevance of the urban and rural archetypes is limited and not applicable in practice.

4.3.4 Operationalization at global scale: Evaluating practical solutions to apply characterization factors in LCA

The developed toxicity model on a 0.5*°0.5° grid cell and the archetype delimitation are explored through the application to the case study of arsenic emissions from alumina production, comparing different methods and level of aggregation.

Figure 4.4.a and b compare respectively FF_i and iF_i on the global, continent, archetype and country level aggregated through a production (left hand side, specific to alumina refineries) vs. population based approach (right hand side, valid for population related emissions such as soap ingredients). The default USEtox value is also represented. The plots show weighted average values and 5th and 95th weighted centiles represented by whiskers. The weighted centiles represent the total percentage regarding production or population instead of the number of values. 23% of arid and sea cells for the production weighted average were not included in the representation of the 0.5°*0.5° weighted percentile as they do not have advection data according to Vorosmarty et al. (2000a) and thus result in null fate and intake fraction results. Accord-

ing to Figure 4.1.a, the majority of existing alumina refineries are located in Asia (58%), followed by Europe (18%). All other continents represent less than 10% of the amount of existing refineries.

Figure 4.4.a and b show no variability for Africa, where the weighted average actually represents the score of the single refinery on the continent.

Figure 4.4.a shows that the global value based on the population weighted approach is 3.7 times higher than the production weighted one, reflecting the fact that large fate values take place in more densely populated areas. Archetype fate values spread over 3 orders of magnitude from the lower to the higher mean value, with a variability that does not exceed 1 order of magnitude compared to the mean value except for the A4 archetype in the population-based approach. The latter trend supports their relevance to represent the variability of fate results at the global scale. The intra-continental variability is up to 3 orders of magnitude, while inter-continental variability of average values barely exceeds 1 order of magnitude. A similar trend is visible on the country scale showing that the continental and country scales do not represent relevant solutions to reduce variability compared to the global scale.

iF_i results in Figure 4.4.b show that on a 0.5°*0.5° grid cell level iF_i spread over 3 orders of magnitude, up to nearly 1E-3 [$kg_{ingested}/kg_{emitted}$] in the case of a population weighted aggregation. The global value for the population based average is 3.3 times higher than the production based average which represents a pragmatic way to account for sector specific location of emissions for goods that are traded at continental or global level. The population weighted approach provides a clear emphasis on high iF_i values, and is valid for substances used in human daily activities such as pharmaceutical substances, detergents and health care products. In practice, this assumption is not appropriate in the case of chromiumVI and arsenic emissions to water from alumina production given that these emissions do not take place in a domestic context or during daily human related activities. Appendix C.10 provides additional information on the intake fraction repartition, showing iF_i of arsenic versus alumina refineries emissions. Appendix C.11 shows a case study where aggregation of fate and intake fraction results are tested for different resolutions on two specific alumina refinery sites.

The continental, country, or urban and rural archetype aggregation do not show a substantial reduction in variability compared to the global value. The urban iF_i production weighted average is surprisingly only 22% higher than the rural one. This is due to the presence of high iF_i values among alumina refineries located in rural cells. For instance, the site of Achinsk has the highest intake fraction value, 0.0014 [$kg_{ingested}/kg_{emitted}$], while it is classified in the rural archetype due to low population density in the grid cell of emission, although there is a high population density in downstream grid cells.

The USEtox default fate and intake fraction values are both within one order of magnitude compared to the global value calculated with our model, showing the relevance of this generic model for this specific case study with emissions occurring all around the globe.

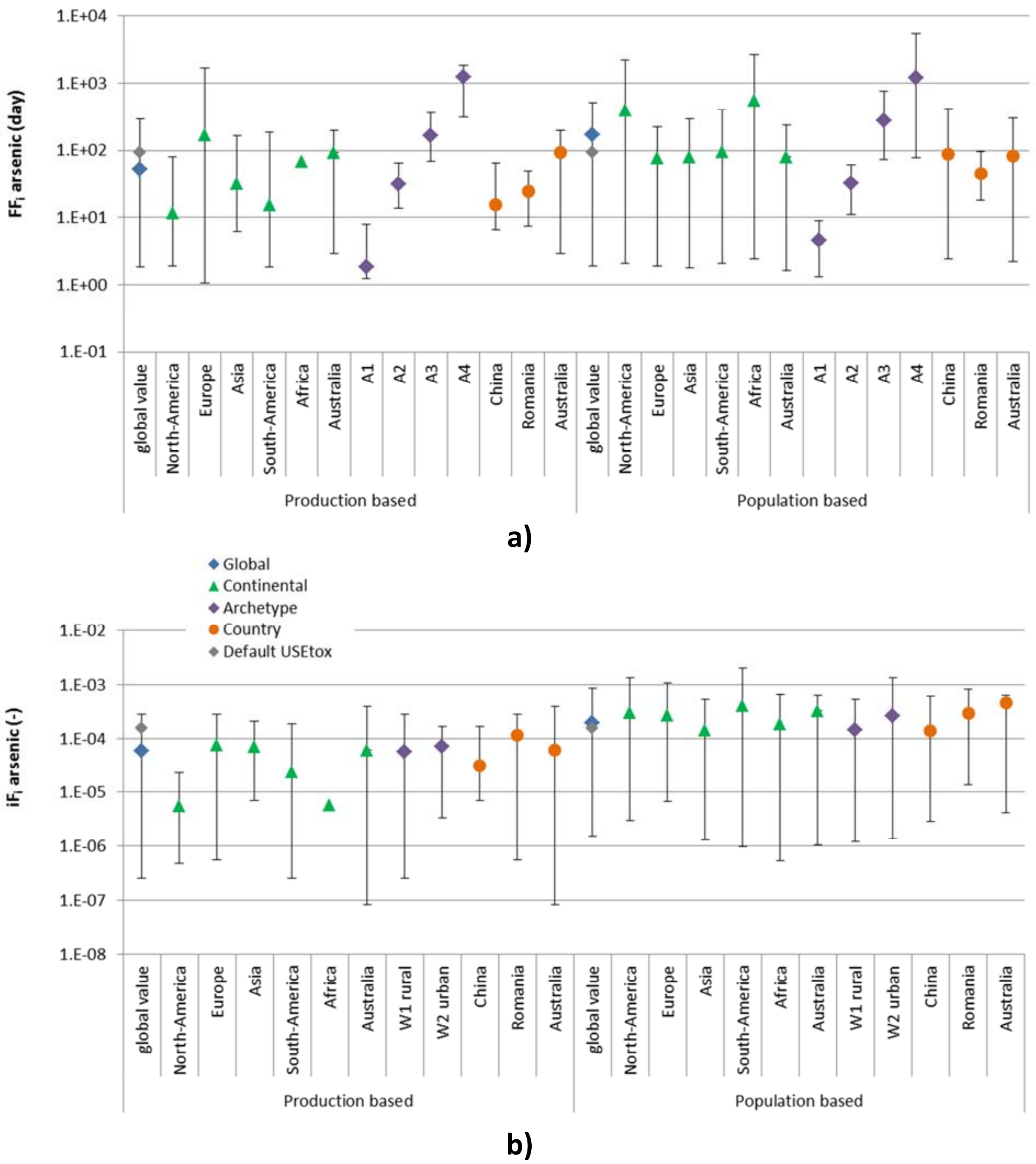


Figure 4.4: Distribution of arsenic FF_i (a) and arsenic iF_i (b) at the global, continent, archetype and country using a production-based and a population-based emission and aggregation. The whiskers represent the 5th and 95th population weighted centiles.

4.4 Conclusion

This work explores the importance of spatial resolution and its applicability to assess toxic emissions to freshwater within LCA. Operationalization approaches exist that simplify the implementation of spatially differentiated methods addressing the impact of toxic emissions to water.

Fate archetypes provide consistent fate estimations. However they do not follow a systematic geographical pattern (such as rural and urban) that an LCA practitioner can deduce without the support of detailed geographical information.

Unlike air emissions that affect their direct surrounding, e.g., in urban zones within 49 km resolution (Humbert et al. 2011), the influence of downstream cells prevents from obtaining easily applicable exposure archetype classifications based on the landscape characteristics of the emission cell. This means that the urban and rural human exposure archetypes do not show a significant and systematic improvement for the intake fraction estimation, nor do they have a common basis with the fate assessment. This finding indicates the difficulty of devising a common archetype encompassing both ecotoxicity and human toxicity impact categories.

We evaluated approaches to aggregate characterization factors from their native resolution to a coarser scale such as country, continent or global average. Calculating sector-specific weighted averages represents a pragmatic and easily applicable approach to account for sector-specific distribution of emissions. We recommend the production weighted approach in applications where the distribution of production location and related emissions is known. The results of this study therefore need to be applied in a systematic way to a wide range of relevant industry sectors. The population-based weighting should be reserved to cases for which higher emissions occur in densely populated areas.

The USEtox fate and intake fraction default values are found within one order of magnitude compared to the global value calculated by our sophisticated model, showing the relevance and adequate representativeness of this generic model. These conclusions need to be extended to a wider set of substances by comparing the averaged results of the fully regionalized model to generic results for other industry sectors.

An alternative option than aggregation to operationalize spatial differentiation in LCA would be to use a spatialized life cycle inventory and apply site-dependent impact assessment factors to assess emissions where they take place before calculating the aggregated results (as proposed by Mutel and Hellweg (2009)). When point source emission are known, e.g. for power plant or aluminum production sites, an impact score can be calculated by summing the impact scores generated by each characterized point source emission. This approach, which prevents from losing information on impact variability through LCIA aggregation, would need to be further explored and tested in parallel with the development of systematically regionalized inventory datasets.

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Chapter 5

Conclusion and outlooks

5.1 Discussion on water footprint practice

Chapter 2 provides a comprehensive review of existing methods that cover various impact pathways generated by quantitative water use. This review leads to recommendations for method developers on key model components to build a scientific consensus. Preliminary application recommendations are also formulated for practitioners to perform a water footprint using the current state-of-the-art methods and databases. The outreach of these recommendations published in 2013 in the academic field, public institutions and private companies was ensured by the participation of the panel of LCA experts in the WULCA working group as the publication co-authors. These recommendations are thus presented as consensual findings from the LCA community and have, to date (September 2014), been applied and cited in scientific reviews (Hoekstra and Wiedmann 2014; Laurent et al. 2014), methodological developments (Ridoutt and Pfister 2014; Pfister and Bayer 2014) and in several water case footprint studies (Van Hoof et al. 2013; Boulay et al. 2014a; Feng et al. 2014). One of the latter case studies resulted from a collaborative work where I perfomed the water footprint analysis and the environmental experts from the contracting company wrote the scientific publication (Van Hoof et al. 2013). This work presents a water footprint of a hand dishwashing product. Other case studies were published on agro-food products (Milà i Canals et al. 2010), complex industrial product systems (Berger et al. 2012) or house care products (Boulay et al. 2014a) in parallel to the development of the ISO 14046 standard (ISO 2014). The outcome of these studies varies depending on sectors and geographical locations: while the water footprint of broccolis produced in scarce areas such as Spain is dominated by the consumption of irrigation water (Milà i Canals et al. 2010), the study on hand dish washing product highlighted the importance of the consumer use stage contribution, which can be up to >90% (Van Hoof et al. 2013). In the latter case study, the impact is driven by both direct water use (used in the dishwashing process) as well as indirect water use (used to generate the electricity to heat the water) (Van Hoof et al. 2013). The following paragraphs aim at discussing the application recommendations formulated in chapter 2 for practitioners in the light of my personal experience as life cycle analyst and of available published case studies.

5.1.1 Databases and specific inventory data

There are two main requirements for LCA inventory databases to be used in a water footprint: (1) cover all water flows addressed by LCIA methods in a consistent balance between input and output water flows and (2) provide regionalized inventories, especially for agricultural processes with a potentially large variation in irrigation (Stoessel et al. 2012). Chapter 2 emphasized the current limitations regarding the water use coverage in existing databases, e.g., the need to differentiate consumptive freshwater use from withdrawal, the need to distinguish between different water types based on origin as well as to include freshwater evaporation from water reservoirs as consumptive use. Some of the latter recommendations were integrated in the new version of existing inventory databases. For instance, only ecoinvent v2.2 was available at the time of the publication of the article presented in chapter 2, while the updated version ecoinvent v3 released in May 2013 includes both input and output water flows. As explained by Pfister (2014), ecoinvent v3 provides balanced water uses integrating the processes from the Quantis Water database (Quantis 2012), developed for the purpose of bridging the gap in water use coverage in ecoinvent v2.2. The GaBi database was also updated in 2013 (PE International 2013) with new water modelling principles. However, although water quality parameters as pollution flows are well covered in ecoinvent v2.2, v3 or GaBi, the water quality classes to implement water availability footprint methods as defined by Boulay et al. (2011a) are not covered in these databases. Furthermore, practitioners complemented the recommendations formulated in chapter 2 by highlighting the lack of detailed inventory data with spatially differentiated water use information (Berger et al. 2012; Van Hoof et al. 2013). Given the large influence of climatic and hydrological conditions on the amount of water withdrawn for an agricultural or industrial process in a given location (also depending on the irrigation or industrial technology), the field of a water scarcity and availability footprint raises high expectations in term of inventory data requirements to perform a meaningful assessment. These requirements could be fulfilled through a wide cooperation among LCA communities throughout the world to determine data requirements for globalized goods as well as national initiatives to gather LCI data. A successful example of development of a national database compatible with other databases such as ecoinvent v3 is AusLCI, that gathers more than 150 datasets (AusLCI Committee 2014).

Additionally to the use of generic databases, practitioners stress the need to identify and collect primary data for foreground processes and whenever possible also for sensitive generic background processes for a more accurate assessment (Boulay et al. 2014a). The systematization of on-site specific industrial water use data collection is not an easy task since industrial water flows are not necessarily collected comprehensively regarding LCIA needs. For instance, the Global Reporting Initiative (GRI) requires the reporting of three indicators related to water: the total water withdrawal (EN8), water sources significantly affected by withdrawal of water (EN9) and percentage and total volume of water recycled and reuse (EN10) (Global Reporting Initiative 2011).

5.1.2 Representativeness of characterization models

Some impact methods provide a characterization factor for a single local application. Chapter 2 highlights this limitation especially for endpoint methods addressing impact on ecosystem quality. Existing case studies confirm that in case a single characterization factor is applied generically for any locations, additional uncertainty in the method interpretation and applicability is created (Van Hoof et al. 2013) given that no testing of parameters sensitivities to other areas has been performed. For example, characterization factors for groundwater extraction impacts (van Zelm et al. 2011) and effects of thermal release to water (Verones et al. 2010) are only available respectively for specific cases in the Netherlands and Switzerland. Global coverage is a key requirement for LCIA method applicability.

5.1.3 Spatial differentiation

Chapter 2 concludes that spatial differentiation of LCIA methods could be refined for generic or with low resolution models in order to increase LCA results discriminating power. This recommendation is applied with increasing systematism in recently published methods (Berger et al. 2014; Pfister and Bayer 2014). However, the implementation of finely regionalized methods in existing case studies raises several constraints regarding their applicability when using available generic life cycle inventory databases. Case study authors highlight the difficulty of implementing spatial differentiation for background processes due to the lack of inventory information (Van Hoof et al. 2013; Boulay et al. 2014a). Indeed, global brands and retailers rely on complex supply-and-demand networks that span the globe, extend five or six tiers deep, and can reconfigure overnight in response to changes in consumer demands, commodity prices, currency fluctuations and political risks (O'Rourke 2014). The origin of materials is thus not always well identified by companies performing a LCA, given that the most important water flows do not necessary take place at the company site or at the first-tier supplier location, which are the level where the location information is potentially accessible. For example, a company will probably know the location of its chemical supplier, but although the chemical may have been blended and packed on-site, it may have been formulated, by a supplier, somewhere else in a region with different water scarcity (Boulay et al. 2014a). The characterization factor can vary for instance from 0.092 m^3 deprived per m^3 consumed in Switzerland to 0.97 in Egypt for the method of Pfister et al. (2009), which also provides scarcity indexes at the 0.5*0.5° level. Van Hoof et al. (2013) also emphasized the challenge in communicating the results for a product used under exactly the same consumer use conditions in various locations generating very different impact scores. Exploring solutions to operationalize spatial differentiation in a water availability or a water scarcity footprint would be useful, based on the conclusions of chapters 2 and 3 on water degradation through toxic emissions.

5.1.4 Software implementation

Chapter 2 does not mention purely practical constraints related to the implementation of available LCIA methods in commercial softwares. However, the application literature stresses that broad application of water footprint requires further development and integration in commercial life cycle assessment software in a user friendly way (Van Hoof et al. 2013). The integration in a commercial software of a comprehensive framework covering all existing impact pathways expressing an LCA profile as the sum of different footprints would be a welcome step to answer the practitioner need to perform comprehensive and easily applicable LCA. The latter framework shall make sure to avoid any double counting, as proposed by the IMPACT World+ methodology (Bulle et al. 2012).

5.1.5 Proposed outlooks

The analysis of recommendations formulated in chapter 2 in the light of performed case studies provided a snapshot of current practical constraints and limitations to perform a water footprint. The following outlooks aim at proposing broad perspectives and food for thought for practitioners and method developers for future developments in the water footprint field.

From a practitioner perspective:

From midpoint to endpoint assessment: My personal experience as a consultant for companies in the agro-food and home-, personal- and health-care products showed me that the novelty and complexity of impact pathways leading to endpoint assessment tend to refrain companies from communicating the results of comprehensive water footprints outside of scientific publications. On the other hand, water scarcity footprint indicators are widely used as a communication tool, for instance in the French environmental labeling experiment which aims at providing environmental labels for all consumer goods commercialized in France and was in the experimental phase until 2013 (AFNOR 2014). Many recent methodological developments attempt to fulfill the need for a simple and environmentally relevant scarcity indicator (Ridoutt and Pfister 2010b; UNEP SETAC Life Cycle Initiative 2014; Berger et al. 2014). The field of risk assessment also relies on water-consumption-to-availability or water-withdrawal-to-availability ratios to map risks related to water (Gassert et al. 2013). The ISO 14046 (ISO 2014) standard provides the possibility to perform water footprints with different scopes and levels of complexity. This flexible framework thus leaves the possibility for industries to move, once they are ready to do so, from inventory and water scarcity footprints to more comprehensive evaluations including water availability and degradation. The current and coming development of water footprint related methods, databases and softwares is expected to enhance and build confidence in water footprint practice in the coming decade.

Sector-specific approach: The rise of initiatives such as Environmental Product Declaration (EPD) or Single Market for Green Products Initiative (European Commission 2013b) aims at streamlining the application of LCA by respectively following ISO 14025 and product- / sector-specific rules at the European level. For instance, the Single Market for Green Products initiative (European Commission 2013b) was launched by the European commission in 2013 to establish product- and sector-specific rules to measure the environmental performance at the product (PEF) and organization (OEF) level. However, theses attempts to fulfill the harmonization gap through a normative framework are not focused on water use and do not provide state-of-the-art recommendations in this respect. Such a sector-specific approach could thus be followed to establish more specifically water footprint profiles for defined product categories. So far, a few specific case studies were covered and published in scientific articles to evaluate a product water footprint (Milà i Canals et al. 2010; Berger et al. 2012; Van Hoof et al. 2013; Boulay et al. 2014a). There is a need to further test and apply the current state-of-the-art methods to gain a deeper understanding of the dominant impact pathways for a specific product at different life cycle stages. By fostering a quantitative comparison of endpoint impact scores, the relevance of the information conveyed by widely used scarcity indexes could be verified. A review of dominant impact pathways specific to water related to specific product categories would be key information to provide guidance to practitioners for efficient and relevant inventory modeling.

From a method developer perspective:

Exploring spatial differentiation operationalization approaches: The modeling of the impacts related to water use and degradation rely on some common parameters such as hydrological data on water availability in a given geographical unit. However other parameters might differ, such as the human development index, used in the impact pathway leading to impact on human health (Boulay et al. 2011b). The exploration of spatial differentiation approaches specific to methods addressing water use would be useful to explore, based on the findings of chapters 3 and 4.

Product-oriented approach: All methods reviewed in chapter 2 are dimensioned to evaluate environmental impacts related to water use from a LCA perspective following a product-oriented approach, where water is just one area of attention among other impact categories such as global warming. On the other hand, water management is typically territorial-based and focused on increasing water-use efficiency. It is not related to absolute thresholds for the total volume of consumption per unit area (Hoekstra 2013). While water efficiency might increase on a product-based assessment, demand for water intensive commodities such as meat and biofuels is rapidly rising (Hoekstra and Wiedmann 2014), leading to a potential overall increase in the global water footprint and exceedance of water consumption threshold in sensitive areas. Combining product and territorial-based approaches is thus a priority for integrated water management measures and policies.

Assessment of the water "carrying capacity": As mentioned in chapter 2, most water scarcity indexes are based on consumption-to-availability or withdrawal-to-availability ratios. This approach is not related to absolute thresholds for the total volume of consumption per unit area (Hoekstra 2013) to guarantee human and ecosystem well-being. It thus does not account for the dependence on freshwater of a certain geographical area, given that water consumption in a water scarce area induces an impact if the water use is in competition with other users. In the case of a desert without human occupation and with reduced ecosystems with low net production, although water availability is low, water consumption might have less impact than in a scarce and densely populated area or in a sensitive ecosystem. Although newly developed models attempt to account for water "vulnerability" (Berger et al. 2014) through the consideration of the effective water consumption within a river basin, this indicator follows a user-oriented approach and does not consider local water needs for humans and ecosystems. The concept of "carrying capacity" is defined as the environment's maximal load in the ecological field (Hui 2006). This concept could be explored for an application in the field of water footprint, in order to create a locally-grounded water scarcity or availability indicator.

Explore the complementarity with the field of risk assessment: A complementary approach to water footprint is the field of risk assessment, where the physical (related to water quantity or quality), regulatory and reputational risks (related to political aspects such as media coverage) are quantified and mapped with a global coverage. Strategic risks can be defined as the threats or opportunities that materially affect the ability of an organization to survive (Allan and Beer 2006). The scope of this kind of assessment strongly differs from a water footprint, where the potential impacts on environment are quantified rather than the influence of environmental and political conditions on an organisation's activity. Furthermore, the visualization of key parameters influencing risks related to water, such as proposed in the Aqueduct tool (Gassert et al. 2013), do not include impact modeling but only a snapshot of the value of a given parameter. A water risk assessment could nevertheless provide a complementary insight that prevents from using implicit modelling value choices. Complexity shall indeed be introduced gradually within organizations that do not yet have a confirmed practice in the environmental field. For instance, carbon footprint is still commonly used as a global proxy for overall impact although it was proved to be a poor representative (Laurent et al. 2010).

5.2 Discussion on operationalization of spatial differentiation

While one of the conclusions of chapter 2 is the recommendation to explore further spatial differentiation for methods addressing water use, chapters 3 and 4 take one step back from model sophistications by testing the acceptability of using a generic model and exploring various approaches for the operationalization of spatially differentiated approaches, applied to potential impacts of toxic emissions into water. Several operationalization approaches were explored to fulfill two method developers- and practitioner- oriented goals: (1) decrease computation time and data requirements from a modeling perspective and (2) ease the application of spatially differentiated characterization factors for practitioners when detailed geographical information is partially available or unknown (particularly in case of background processes). The following paragraphs discuss the findings on operationalization technics analysed in chapter 3 and 4.

5.2.1 Addressing spatial differentiation through a nested model

We found in Chapter 3 that a nested approach, such as the USEtox model parameterized with sub-continental landscapes, succeeded in mimicking the results of a spatially differentiated model with interconnected compartment on the same resolution, with the exception of very persistent volatile pollutants that can be transported to Polar Regions. Landscape parameters at the continental or sub-continental level developed in chapter 3 can thus be easily implemented in USEtox without changing its inherent structure of a single continental box nested in a global one. While emissions into water of non-volatile substances remain in the hydrological network of the sub-continental zones of emission, emissions to other media do not necessarily follow the same trend. Persistent substances such as PCBs emitted to air were proven to be very mobile and remain as low as 3% of the emitted quantity in the region of emission (Shaked 2011). A nested model is thus relevant in a media where the residence time before advection to another zone is higher than the fate of the substance. In the opposite case, a substance will be quickly advected to surrounding zones where it will spend the largest part of its residence time. The geographical properties of the emission zone will thus not have a large influence on its fate in the latter case. The relevance of a nested model has been validated for emissions to water but needs to be studied and tested for emissions to air.

5.2.2 Aggregation of characterization factors on lower resolution

Chapters 3 and 4 explore different aggregation methods to provide characterization factors on lower resolution to relate them to existing generic inventory databases or to the level of knowledge of practitioners for specific processes. Additionally to these implementation constraints related to water footprint impact assessment, impact score interpretation by the practitioner is also influenced by the inventory and characterization factors resolution. Indeed, the practitioner needs to know characterization factors values or order of magnitude in order to justify a specific impact score. The finer the LCIA method resolution, the more data the practitioner will have to process to interpret and justify the impact results. Furthermore, the practitioner would require to be informed of the parameters used for the aggregation weighting in order to interpret results correctly. The following paragraphs discuss two aggregation solutions: surface-weighted aggregation into landscape archetypes and production- or population- weighted aggregation into political or geographical units.

Surface-weighted aggregation into landscape archetypes: Simplified models based on regional archetypes were created in chapters 3 and 4. An archetype model based on water residence time to the sea was developed in chapter 3. It improved the prediction of fate and intake fractions for point source emissions, within a factor five compared to the spatial model at the watershed resolution (e.g., the Rhine watershed covers more than 160’000 km^2 (Pennington et al. 2005)). As a comparison, the one-box model is off by a factor of 30 for coastal watersheds such as Brittany. By pushing further spatial resolution to a fine grid of 0.5*0.5° (about 2’000 km^2 at temperate latitudes), chapter 4 shows that an additional parameter influences the substance fate: the cumulated equivalent depth of the grid cells in the path of a specific substance. This parameter is defined as the mean depth of the rivers and lakes across which a substance travels until it is removed by sedimentation, evaporation or degradation, in a given hydrological network. Although cumulated equivalent depth depends on substance-specific removal rates from water, we proved that a reference substance could be used as an acceptable proxy to re-calculate the fate of other substances based on its equivalent depth. The developed fate archetypes that consider cumulated equivalent depth reduce fate variability to 2 orders of magnitude difference compared to 5 orders of magnitude of variability among all spatial results for persistent substances such as chromium(VI) or arsenic. However, the archetype model does not represent an implementable and simplified approach of spatial differentiation. Indeed, archetypes do not follow a systematic geographical pattern (such as rural and urban, or coastal and non-coastal) that an LCA practitioner can readily deduce without having to gather detailed geographical information. Developed intake fraction archetypes based on population density did not show a significant and systematic improvement in the intake fraction estimation. These analysis lead to the conclusion that the archetype approach to assess degradative water use of toxic emissions into water does not provide anymore clear benefits (regarding the two goals mentioned above) when the sophistication of spatially differentiated models is further increased.

Production- or population-weighted aggregation into political / geographical units: Population- and production based emissions approaches can be used to characterize emissions of a process occurring at different locations across the globe. For instance, the IMPACT World+ method currently uses population as a proxy for all emission categories (Bulle et al. 2012). A case study on degradative water use from toxic emissions of global aluminium production was evaluated in chapter 4 applying a highly spatially resolved characterization model. Overall, intake fraction from emissions of chromium(VI) and arsenic from the aluminium production process obtained by the population-based emission approach over-estimates the intake fraction calculated with the production-based emission approach, due to the unappropriate artefact of considering high emissions occurrence in densely populated areas. Chapter 4 also showed that generic fate and intake fraction of the scientific consensus model USEtox are within one order of magnitude compared to the production-based emission approach calculated from the spatially differentiated model. Current USEtox results can thus be considered as validated and appropriate in the case of chromium(VI) emissions from alumina production. However, results from other industrial or agricultural applications that require a specific hydrological setting, e.g., emissions from a desalination plant that is located on the sea shore, or a specific demographic setting, e.g., pharmaceutical substance emissions that are proportional to human density, could provide a different trend. Indeed, if a specific pattern influences fate or intake fraction, the production- or population- based emission average could for instance result in a reduced fate in the case of the desalination plant or increased intake fraction in case of substances used in daily activities. Nonetheless, substances are often manufactured for several uses. For instance, the optical brightener for detergents and cleaners tinopal, studied in chapter 4, is used in home and personal care products such as detergents, plastic product manufacturing such as bottle as well as the pulp and paper industry (BASF 2014). Differentiation of characterization factors for a single substance according to the associated applications would bring additional important complexity and is thus not advisable from a practical point of view. Furthermore, the field of chemical production and type of application follows a fast evolution pace and would require constant updates and adjustments in case application-specific characterization factors are developed. The precision of the USEtox generic results for the case study performed in chapter 4 thus suggests further exploring the validity of the consensus model results before moving to the complexity of aggregation in daily LCA practice.

5.2.3 Proposed outlooks

Two options of operationalization were explored in chapter 3 and 4: the development of a nested model and the aggregation of characterization factors at a lower resolution. The following outlooks suggest additional operationalization options that could be explored in the future:

Life cycle inventory disaggregation: There are alternative options to aggregation in order to operationalize spatial differentiation in LCA. The option of life cycle inventory disaggregation relies on using a sophisticated and highly (spatially) resolved model to assess emissions where they take place by applying site-dependent impact assessment factors before calculating the aggregated results (as proposed by Mutel and Hellweg (2009)). When point source emissions are known, e.g. for power plant or aluminum production sites, an impact score can be calculated by summing the impact scores generated by each characterized point source emission (so called “production-based emission” approach). When this is unknown, the emission occurring in a given continent, country or region (such as the information available in currently existing LCI databases) could be allocated into each grid cell of the spatially differentiated model using an emission proxy such as population (so called “population emission” approach). In other words, one could disaggregate the LCI to adapt to the resolution of the LCIA method rather than aggregating the LCIA method on the available LCI resolution. This approach prevents from losing information on impact variability through LCIA aggregation. However, the choice of proxy to disaggregate the LCI as well as its sensitivity needs to be analysed.

Partial spatial differentiation: Another pragmatic and practitioner-oriented approach would be to perform a “partial spatial differentiation” by implementing the principle of parsimony, summarized by the following quote attributed to A. Einstein: “Everything should be made as simple as possible, but not simpler”. This principle can be applied to spatial differentiation in LCA: model complexity should be considered if it reduces the impact scores variability, defined as stemming from inherent variations in the real world (Huijbregts 1998), without increasing its uncertainty, defined as coming from inaccurate measurements, lack of data or model assumption that “convert” the real world into LCA outcomes (Huijbregts 1998). A complementary approach could thus be to establish key principles for spatial differentiation from a practitioner perspective given the current methodological state-of-the-art. Rather than applying spatial differentiation systematically to an entire system, it can be operationalized through the following suggested iterative process for each impact category: (1) identifying if main impact contributions take place in the foreground or at the level of the first tier-supplier (2) if this is the case, gathering location specific information for these processes and regionalizing the inventory database and impact on a fine resolution (3) for all other process, using meaningful global aggregated averages. For instance, the process of cotton production in the United States in ecoinvent v3 called “Cotton fiber {US}| cotton production | Alloc Def, U” has a water scarcity footprint of 3.23 m^3 according to the method of Pfister et al. (2009). Dominant processes are the production of hydro-electricity involved in the irrigation that contributes to 34% of the water scarcity footprint and the direct water use for irrigation that contributes to 25% of the impact score. The impact from hydro-electricity production is due to water evaporated from reservoirs in the United States, which is captured as a water scarcity index of 0.5 m^3 deprived per m^3 turbined as a national average. For this process, a simplified regionalization approach would be to provide a finely regionalized characterization factor, i.e. at the 0.5*0.5° grid cell for the irrigation water at the cotton production location. Then a national characterization factor could be applied for the hydro-electricity production given that electrical grid mixes are determined at the country level. It would be useful to provide a weighted average of the scarcity factor of hydro-electric power plants in the US or to disaggregate the inventory suggested previously. The same approach could be followed for other contributions from imported hydro-electricity that contributes in total to 32% of the total impact score. The remaining processes contribute to 9% of the water scarcity impact score and could be rely on a global water scarcity index given the potential difficulty to track their location.

In order to operationalize this approach, a large sectorial analysis would be needed to provide consistent global aggregated characterization factors for substances that are not emitted in human daily activities or to disaggregate the inventory data as suggested previously to tackle the lack of spatial differentiation in existing inventory databases. As mentioned in part 5.2.2, a sector-specific average is valid in case a substance is emitted mainly during a single industrial activity. In substances with various uses, we recommend to use more complex models to estimate overall cross-sectorial emissions. As a first tier approach, characterization factors from generic models such as USEtox can be used until meaningful aggregated averages are developed.

This integrated approach could be operationalized in commercial softwares, where the user would rely on default population- and sector-based global or generic characterization factors that could be changed for specific geographical location and related characterization factors for relevant processes.

Operationization of a fully spatially differentiated multimedia model: The 0.5*0.5° model developed in chapter 4 including hydrological data shall be integrated in a multimedia model integrating fate, exposure and effect in all media through trans-boundary transport and multi-pathway exposure. The latter model could be used as a reference to validate or discriminate the relevance of suggested operationalization approaches on a sound basis for emissions to all media. The water module developed within this thesis could be directly integrated in such a model.

5.3 Achieved results

This thesis explores good practices and operational approaches to assess a water footprint and improve its discriminatory power. In addition to the release of the ISO 14046 standard, the review of existing methods evaluating a water footprint performed in this thesis is shown to improve the dissemination of water footprint practice, based on the recent scientific publications citing this work. The recommendations formulated from the analysis of the current state-of-the-art are already integrated in the new release of existing inventory databases.

For the impact of water degradative use from (eco)toxic emissions to water, the acceptability of using a generic model, for example USEtox, to evaluate point source emissions was tested at different resolutions and for different type of emissions (point source, uniform and sectorial). Uniform and sectorial emissions respectively at the watershed and grid cell resolution miss less than one order of magnitude variability. On the other hand, more than three orders of magnitude of fate and intake fraction variability are covered by a single number for point source emissions at the watershed and grid cell resolution. Using a generic model is thus acceptable to cover a low resolution such a continent, country, or sector while a finer resolution is essential for a regional impact score at the watershed, grid cell or point source level.

Practical solutions to operationalize spatial differentiation were tested and some of them validated to decomplexify the implementation of regionalized assessments for both method developers and practitioners. These operationalization approaches aim at minimizing the trade-off between theoretical variability reduction and decrease of uncertainty in practice considering the level of geographical information known by the practitioner. This thesis demonstrated that implementable pragmatic solution meeting both LCA method developer and practitioner needs exist and vary depending on the spatial differentiation resolution. A nested approach such as proposed in the scientific consensus USEtox model was proved to be valid at the continental scale for the evaluation of toxic emissions to water. Characterization factors aggregation techniques were explored and the relevance of a production-weighted geographical average could be acknowledged for the case study of alumina production. However, most of substances have several industrial applications and require a more complex model to determine a production weighted result representative of the substance overall emissions. The archetype approach to evaluate toxic emissions to water was proved to be useful at the watershed scale, while it was discriminated at the grid cell scale because of its complexity and lack of applicability. Indeed, despite fate archetypes providing consistent fate estimations, they do not follow a systematic geographical pattern (such as rural and urban) that a LCA practitioner can deduce without the support of detailed geographical information. Unlike air emissions that affect their direct surrounding, e.g., in urban zones within 49 km resolution (Humbert et al. 2011), the influence of downstream cells prevents from validating simplified archetype classifications based on the landscape characteristics of the emission cell.

Based on these findings, I can formulate four recommendations for the implementation of a spatially differentiated water footprint based on currently available methods, databases and softwares (as in September 2014):

- **Apply spatially differentiated methods in addition to generic ones**: Practitioners yet acquired a high level of knowledge of generic available methodologies such as IMPACT2002+ (Jolliet et al. 2003), EDIP (Hauschild and Potting 2005) and ReCiPe (Goedkoop et al. 2009). This knowledge could be used as bridge to explore the new field of spatially differentiated results interpretation.
- **Test the interpretability of the disaggregated spatial differentiation approach**: As mentioned in part 5.2.3, Mutel and Hellweg (2009) propose an interesting approach to prevent loosing information by aggregating impact characterization factors. When a point source emission location is unknown, the allocation of the spatially differentiated model into each grid cell using an emission proxy such as population could be explored. The choice of allocation proxy should however be further studied.
- **Further evaluate the representativeness of generic USEtox results:** This work supported the representativeness of the USEtox generic characterization factor at the sector level. These conclusions need to be extended to a wider set of substances by the comparison of the results of a fully regionalized model compared to generic results in other case studies.
- **Explore semi-systematized tiered spatial differentiation approach:** As suggested in part 5.2.3, the operationalization of spatial differentiation could further rely on the LCA practitioner expertise through a semi-systematized approach where spatial differentiation can be performed for direct and first tier dominant processes. Indeed, the practitioner needs to be able to track and interpret LCA results by having a deep understanding of inventory data and LCIA modeling. Increased modelling complexity in LCIA methods might refrain interpretation capacity and thus discredit confidence in LCA results.

References

References

Abdel Al I, Ahmad S, Ballestero M, Bezbaroa S, Cookey P, Dourojeanni A, Langford J, Mensink M, Opondo G, Park JM, Pinero E, Ruffier P, Witmer L, Xin H (2014) The AWS International Water Stewardship Standard. Available at: http://www.allianceforwaterstewardship.org/Beta%20AWS%20Standard%2004_03_2013.pdf.

AFNOR (2014) Affichage environnemental de produits de grande consommation. Available at: http://affichage-environnemental.afnor.org/.

Alcamo J, Doll P, Henrichs T, Kaspar F, Lehner B, Rosch T, Siebert S (2003) Global estimates of water withdrawals and availability under current and future “business-as-usual” conditions. Hydrol Sci J 48:339–348.

Alcamo J, Flörke M, Märker M (2007) Future long-term changes in global water resources driven by socio-economic and climatic changes. Hydrol Sci J 52:247–275.

Alcor Technology (2013) Alcor technology. Available at: Http://www.alcortechnology.com/GlobalOperations.html.

Alexander RB, Smith RA, Schwarz GE (2004) Estimates of diffuse phosphorus sources in surface waters of the United States using a spatially referenced watershed model. Water Sci Technol 49:1–10.

Allan J (1996) Policy responses to the closure of water resources: regional and global issues. Water Policy Alloc Manag Pract 228–234.

Allan N, Beer L (2006) Strategic risk: it’s all in your head. Working paper series. School of management. University of Bath. United Kingdom. 1–32.

Allison JD, Allison TL (2005) Partition coefficients for metals in surface water, soil, and waste. US EPA. Washington, United States of America.

AusLCI Committee (2014) Requirements for the development of AusLCI Data sets. Australia. 1–54.

Bare JC, Norris GA, Pennington DW, McKone T (2003) TRACI: the tool for the reduction and assessment of chemical and other environmental impacts. J Ind Ecol 6:49–78.

BASF (2014) Description ofTinopal. Available at http://www.basf.com/group/corporate/en_GB/brand/TINOPAL.

Bauer C, Warren F, Barton K, Llorenc M, Bo W, Assumpcio A (2007) Impact category natural resource use: freshwater. UNEP/ SETAC LC initiative report.

Bayart J (2008) Quantification des impacts reliés à l’utilisation de la ressource eau en analyse de cycle de vie: définition d’un cadre d’étude et dévelopement de facteurs de caractérisation. Master thesis from: Ecole Polytechnique de Montréal, Canada

Bayart J-B, Bulle C, Deschênes L, Margni M, Pfister S, Vince F, Koehler A (2010) A framework for assessing off-stream freshwater use in LCA. Int J Life Cycle Assess 15:439–453.

Bayart J-B, Worbe S, Grimaud J, Aoustin E (2014) The Water Impact Index: a simplified single-indicator approach for water footprinting. Int J Life Cycle Assess 19:1336–1344.

Bennett DH, Margni MD, McKone TE, Jolliet O (2002) Intake fraction for multimedia pollutants: a tool for life cycle analysis and comparative risk assessment. Risk Anal 22:905–18.

Berger M, Finkbeiner M (2010) Water footprinting: how to address water use in Life Cycle Assessment? Sustainability 2:919–944.

Berger M, van der Ent R, Eisner S, Bach V, Finkbeiner M (2014) Water accounting and vulnerability evaluation (WAVE): considering atmospheric evaporation recycling and the risk of freshwater depletion in water footprinting. Environ Sci Technol 48:4521–8.

Berger M, Warsen J, Krinke S, Bach V, Finkbeiner M (2012) Water footprint of European cars: potential impacts of water consumption along automobile life cycles. Environ Sci Technol 46:4091–9.

Bösch ME, Hellweg S, Huijbregts MAJ, Frischknecht R (2007) Applying Cumulative Exergy Demand (CExD) Indicators to the ecoinvent Database. Int J LCA 12:181–190.

Boulay A-M, Bayart J-B, Bulle C, Franceschini H, Motoshita M, Muñoz I, Pfister S, Margni M (2014a) Analysis of water use impact assessment methods (Part B): Applicability for 15 water footprinting and decision making with a laundry case study. submitted

Boulay A-M, Bouchard C, Bulle C, Deschênes L, Margni M (2011a) Categorizing water for LCA inventory. Int J Life Cycle Assess 16:639–651.

Boulay A-M, Bulle C, Bayart J-B, Deschênes L, Margni M (2011b) Regional characterization of freshwater Use in LCA: modeling direct impacts on human health. Environ Sci Technol 45:8948–57.

Boulay A-M, Motoshita M, Pfister S, Bulle C, Muñoz I, Franceschini H, Margni M (2014b) Analysis of water use impact assessment methods (Part A): Evaluation of modeling choices based 15 on a quantitative comparison of scarcity and human health indicators. submitted

Bourgault G (2014) Gestion de l'incertitude causée par l'incohérence d'échelle spatiale à l'interface de l'inventaire et de l'analyse des impacts en ACV. PhD thesis at Ecole Polytechnique de Montréal. 1–243.

Bulle C, Jolliet O, Humbert S, Rosenbaum R, Margni M (2012) IMPACT World +: a new global regionalized life cycle impact assessment method. Conference proceedings of Ecobalance. Yokohama, Japan

Chapagain AK, Hoekstra AY (2004) Water footprints of nations. Value of water research report series No. 16. 1:1–80.

CIESIN (2005) Gridded Population of the World (GPW), v3. Population density grid. University. Columbio University. Available at: http://sedac.ciesin.columbia.edu/data/collection/gpw-v3. Columbia, USA

Crettaz P, Pennington D, Rhomberg L, Brand K, Jolliet O (2002) Assessing human health response in life cycle assessment using ED10s and DALYs: part 1—Cancer effects. Risk Anal 22:931–46.

Den Hollander H, Van Eijkeren J, Van de Meent D (2004) SimpleBox 3.0: Multimedia mass balance model for evaluating the fate of chemicals in the environment. National Institute for Public Health and the Environment, Bilthoven. Report no. 601200003. Bilthoven, The Netherlands

Dewulf J, Van Langenhove H, Van De Velde B (2005) Exergy-based efficiency and renewability assessment of biofuel production. Environ Sci Technol 39:3878–82.

Döll P (2009) Vulnerability to the impact of climate change on renewable groundwater resources: a global-scale assessment. Environ Res Lett 4:1–12.

Ecolabel EU (1995) Commission decision of 25 July 1995 establishing the ecological criteria for the award of the community ecolabel to laundry detergents. Off J Eur Communities, L217:14–30.

European Centre for Disease Prevention and Control (2011) Current and future burden of communicable diseases in the European Union and EEA / EFTA countries – Methodology protocol. Stockholm, Sweden

European Commission (2013a) Commission recommendation on the use of common methods to measure and communicate the life cycle environmental performance of products and organisations. Off J Eur Union 56:1–216.

European Commission (2013b) Single Market for Green Products Initiative. http://ec.europa.eu/environment/euss.

Falkenmark M, Lundqvist J, Widstrand C (1989) Macro-scale water scarcity requires micro-scale approaches. Aspects of vulnerability in semi-arid development. Nat Resour Forum 13:258–267.

Falkenmark M, Rockström J (2006) The new blue and green water paradigm : breaking new ground for water resources planning and management. J Water Resour Plann Manag 132:129–132.

FAO (2001) Food Balance Sheets. Available at: http://faostat.fao.org/site/368/default.aspx#ancor.

FAO (2014) AQUASTAT database, Food and Agriculture Organization of the United Nations (FAO). Available at: http://www.fao.org/nr/water/aquastat/main/indexfra.stm.

Feng K, Hubacek K, Pfister S, Yu Y, Sun L (2014) Virtual scarce water in China. Environ Sci Technol 48:7704–7713.

Finnveden G, Hauschild MZ, Ekvall T, Guinée J, Heijungs R, Hellweg S, Koehler A, Pennington D, Suh S (2009) Recent developments in Life Cycle Assessment. J Environ Manage 91:1–21.

Friot D, Antille G (2009) Carbon tariffs & sharing scheme of de-carbonisation costs: analytical tools based on multi-regional Input-Output models. submitted

Frischknecht R, Jolliet O (2014) Project on global guidance on environmental life cycle impact assessment indicators. Available at: http://www.lifecycleinitiative.org/.

Frischknecht R, Jungbluth N, Althaus H, Doka G, Dones R, Heck T, Hellweg S, Hischier R, Nemecek T, Rebitzer G, Spielmann M (2005) The ecoinvent database : overview and methodological framework. Int J LCA 10:3–9.

Frischknecht R, Jungbluth N, Althaus H, Doka G, Dones R, Heck T, Hellweg S, Hischier R, Nemecek T, Rebitzer G, Spielmann M, Wernet G (2007) Overview and methodology - ecoinvent report No. 1. 1–77.

Frischknecht R, Steiner R, Jungbluth N (2006) The Ecological Scarcity method eco-Factors 2006. Report for the Swiss Federal Office for the Environment (FOEN). 1–4.

Gandhi N, Diamond ML, van de Meent D, Huijbregts M a J, Peijnenburg WJGM, Guinée J (2010) New method for calculating comparative toxicity potential of cationic metals in freshwater: application to copper, nickel, and zinc. Environ Sci Technol 44:5195–201.

Gassert F, Landis M, Luck M, Reig P, Shiao T (2013) Aqueduct global maps 2.0. Available at: http://www.wri.org/publication/aqueduct-global-maps-20. 1–20.

Gleick PH (1996) Basic water requirements for human activities: meeting basic needs. Water Int 21:83–92.

Global Reporting Initiative (2011) Indicator Protocols Set Environment (EN). Available at: https://www.globalreporting.org/resourcelibrary/G3.1-Environment-Indicator-Protocols.pdf.

Global Runoff Data Centre (2002) Global Runoff Data Centre. Available at: http://www.bafg.de/GRDC/EN/Home/homepage_node.html.

Goedkoop M, Heijungs R, Huijbregts M, Schryver A De, Struijs J, Zelm R Van (2009) ReCiPe 2008: A life cycle impact assessment method which comprises harmonised category indicators at the midpoint and endpoint level. Report for the Dutch ministry of environment. 1–132.

Goedkoop M, Spriensma R (2001) The Eco-indicator 99: A damage oriented method for Life Cycle Impact Assessment. Methodology Annex. Amersfoort, The Netherlands

Grafe M, Eick MJ, Grossl PR (2001) Adsorption of arsenate (V) and arsenite (III) on goethite in the presence and absence of dissolved organic carbon. Soil Sci Soc Am J 65:1680–1687.

Gronlund CJ, Humbert S, Shaked S, O'Neill MS, Jolliet O (2014) Characterizing the burden of disease of particulate matter for life cycle impact assessment. Published online. Air Qual. Atmos. Heal.

Guinee JB (2002) Handbook on life cycle assessment operational guide to the ISO standards. Int J Life Cycle Assess 7:311–313.

Hanafiah MM, Xenopoulos MA, Pfister S, Leuven RSEW, Huijbregts M a J (2011) Characterization factors for water consumption and greenhouse gas emissions based on freshwater fish species extinction. Environ Sci Technol 45:5272–5278.

Hanasaki N, Inuzuka T, Kanae S, Oki T (2010) An estimation of global virtual water flow and sources of water withdrawal for major crops and livestock products using a global hydrological model. J Hydrol 384:232–244.

Hauschild M, Potting J (2005) Spatial differentiation in Life Cycle impact assessment - The EDIP2003 methodology. Report for the Danish Environmental Protection Agency. 1–195.

Hauschild MZ, Hertel O, Bastrup-birk A (2006) Special issue honouring Helias A . Udo de Haes : LCA methodology spatial differentiation in the characterisation of photochemical ozone formation the EDIP2003 methodology. Int J LCA 1:72–81.

Hauschild MZ, Huijbregts MAJ, Jolliet O, Macleod M, Margni M, Meent D van de, Rosenbaum RK, McKone TE (2008) Building a model based on scientific consensus for Life Cycle Impact Assessment of chemicals : the search for harmony and parsimony. Environ Sci Technol 42:7032–7037.

Hellweg S, Demou E, Bruzzi R, Meijer A, Rosenbaum R, Huijbregts MA, McKone T (2009) Integrating human indoor air pollutant exposure with life cycle impact assessment. Environ Sci Technol 43:1670–1679.

Hellweg S, Milà i Canals L (2014) Emerging approaches, challenges and opportunities in life cycle assessment. Science 344:1109–13.

Helmes RJK, Huijbregts M a. J, Henderson AD, Jolliet O (2012) Spatially explicit fate factors of phosphorous emissions to freshwater at the global scale. Int J Life Cycle Assess 17:646–654.

Henderson AD, Hauschild MZ, Meent D, Huijbregts M a. J, Larsen HF, Margni M, McKone TE, Payet J, Rosenbaum RK, Jolliet O (2011) USEtox fate and ecotoxicity factors for comparative assessment of toxic emissions in life cycle analysis: sensitivity to key chemical properties. Int J Life Cycle Assess 16:701–709.

Henriques JJ, Louis GE (2011) A decision model for selecting sustainable drinking water supply and greywater reuse systems for developing communities with a case study in Cimahi, Indonesia. J Environ Manage 92:214–22.

Heuvelmans G, Muys B, Feyen J (2005) Extending the life cycle methodology to cover impacts of land use systems on the water balance. Int J LCA 10:113–119.

Hoekstra AY (2013) The water footprint of modern consumer society. Routledge. New York, United States of America. 1–224.

Hoekstra AY (2010) The global dimension of water governance: why the river basin approach is no longer sufficient and why cooperative action at global level is needed. Water 3:21–46.

Hoekstra AY, Chapagain AK, Aldaya MM, Mekonnen MM (2011) The water footprint assessment manual: setting the global standard. Eathscan, UK. 1–228.

Hoekstra AY, Wiedmann TO (2014) Humanity's unsustainable environmental footprint. Science 344:1114–1117.

Hoff H (2009) Global water resources and their management. Curr Opin Environ Sustain 1:141–147.

Hui C (2006) Carrying capacity, population equilibrium, and environment's maximal load. Ecol Modell 192:317–320.

Huijbregts M (2010) Kick-off meeting LC-IMPACT. Meeting slides. Nijmegen, The Netherlands

Huijbregts M, Margni M, Jolliet O, McKone T, van de Meent D, Rosenbaum RK, Hauschild M (2010) USEtoxTM Chemical database: inorganics. USEtoxTM team publication. 1–11.

Huijbregts MAJ (1998) Application of uncertainty and variability in LCA. Int J Life Cycle Assess 3:273–280.

Huijbregts MAJ, Meent D van de, Goedkoop M, Spriensma R (2001) Including ecotoxicological impacts in life-cycle assessment. In: Posthuma L, Traas TP and Suter GW. The use of species sensitivity distributions in ecotoxicology. CRC Press, Boca Raton, USA.

Huijbregts MAJ, Schöpp W, Verkuijlen E, Heijungs R, Reijnders L (2000a) Spatially explicit characterization of acidifying and eutrophying air pollution in Life-Cycle Assessment. J Ind Ecol 4:75–92.

Huijbregts MAJ, Thissen U, Guin JB, Jager T, Kalf D, Meent D Van De, Ragas AMJ, Sleeswijk AW, Reijnders L (2000b) Priority assessment of toxic substances in life cycle assessment. Part I: Calculation of toxicity potentials for 181 substances with the nested multi-media fate, exposure and effects model USES-LCA. Chemosphere 41:541–573.

Humbert S (2009) Geographically differentiated life-cycle impact assessment of human health. PhD thesis at University of California, Berkeley. 265.

Humbert S, Manneh R, Shaked S, Wannaz C, Horvath A, Deschênes L, Jolliet O, Margni M (2009) Assessing regional intake fractions in North America. Sci Total Environ 407:4812–20.

Humbert S, Marshall JD, Shaked S, Spadaro J V, Nishioka Y, Preiss P, McKone TE, Horvath A, Jolliet O (2011) Intake fraction for particulate matter: recommendations for life cycle impact assessment. Environ Sci Technol 45:4808–16.

IAI (2012) Personal communication from IAI (Chris Bayliss to Olivier Jolliet).

IMPACTWorld+ (2014) http://www.impactworldplus.org/en/presentation.php. Available at: http://www.impactworldplus.org/en/presentation.php.

ISO (2014) ISO 14046:2014(E) Environmental management — Water footprint — Principles , requirements and guidelines. 42.

ISO (2006a) ISO 14044:2006(E) Environmental management — Life cycle assessment — Requirements and guidelines. 46.

ISO (2006b) ISO 14040:2006(E) Environmental management — Life cycle assessment — Principles and framework. 20.

ISO (2003) ISO/TR 14047:2003(E) Environmental management — Life cycle impact assessment — Examples of application of ISO 14042. 87.

Jolliet O, Margni M, Charles R, Humbert S, Payet J, Rebitzer G, Rosenbaum R (2003) IMPACT 2002+: A new life cycle impact assessment methodology. Int J Life Cycle Assess 8:324–330.

Jolliet O, Wannaz C, Fantke P, Shaked S (2012) Multi-scale, multimedia modeling with Pangea – local to global human health impacts of emissions in multiple continents. Conference proceedings of Ecobalance. Yokohama, Japan

JRC-IES (2011) ILCD handbook - recommendations for Life Cycle Impact Assessment in the European context. European Union report. 1–159.

Koehler A (2008) Water use in LCA: managing the planet's freshwater resources. Int J Life Cycle Assess 13:451–455.

Koehler A, Aoustin E (2008) UNEP/SETAC Life Cycle Initiative. Conference proceeding of SETAC Europe Annual Meeting, Project group on assessment of use and depletion of water resources within LCA. Warsaw, Poland

Koormann F, Rominger J, Schowanek D, Wagner J-O, Schröder R, Wind T, Silvani M, Whelan MJ (2006) Modeling the fate of down-the-drain chemicals in rivers: An improved software for GREAT-ER. Environ Model Softw 21:925–936.

Kounina A, Margni M, Shaked S, Bulle C, Jolliet O (2014) Spatial analysis of toxic emissions in LCA: A sub-continental nested USEtox model with freshwater archetypes. Environ Int 69:67–89.

Kulshreshtha SN (1993) World water resources and regional vulnerability : impact of future changes. International Institute for Applied Systems Analysis report. 1–138.

Laurent A, Clavreul J, Bernstad A, Bakas I, Niero M, Gentil E, Christensen TH, Hauschild MZ (2014) Review of LCA studies of solid waste management systems--part II: methodological guidance for a better practice. Waste Manag 34:589–606.

Laurent A, Olsen S, Hauschild M (2010) Carbon footprint as environmental performance indicator for the manufacturing industry. CIRP Ann - Manuf Technol 59:37–40.

Lehner B, Verdin K, Jarvis A (2006) HydroSHEDS technical documentation. Available at: http://hydrosheds.cr.usgs.gov/HydroSHEDS_TechDoc_v10.doc.

Lundie S, Huijbregts M a. J, Rowley H V., Mohr NJ, Feitz AJ (2007) Australian characterisation factors and normalisation figures for human toxicity and ecotoxicity. J Clean Prod 15:819–832.

Mackay D (2002) Multimedia environmental models: the fugacity approach. 1–257.

Macleod M, Bennett DH, Perem M, Maddalena RL, Mckone TE, Mackay D (2004) Dependence of intake fraction on release location in a multimedia framework a case study of four contaminants in North America. J Ind Ecol 8:89–102.

Maendly R, Humbert S (2009) Empirical characterization model and factors assessing aquatic biodiversity damages of hydropower water use. submitted

Manneh R, Margni M, Deschênes L (2010) Spatial variability of intake fractions for Canadian emission scenarios: a comparison between three resolution scales. Environ Sci Technol 44:4217–24.

Margni M (2003) Source to intake modeling in life cycle impact assessement. Thesis from Ecole Polytechnique Fédérale de Lausanne (EPFL). 1–157.

Margni M, Gloria T, Bare J, Seppälä J, Steen B, Struijs J, Toffoletto L, Jolliet O (2008) Guidance on how to move from current practice to recommended practice in Life Cycle Impact Assessment. UNEP/SETAC Life Cycle Initiative Life Cycle Impact Assessment Programme report. 1–33.

Margni M, Pennington D, Birkved M, Larsen HF, Hauschild MZ (2002) Test set of organic chemicals for life cycle impact assessment characterisation method comparison. OMNITOX project report.

Margni M, Pennington DW, Amman C, Jolliet O (2004) Evaluating multimedia/multipathway model intake fraction estimates using POP emission and monitoring data. Environ Pollut 128:263–277.

Margot J, Kienle C, Magnet A, Weil M, Rossi L, Felippe L, Alencastro D, Abegglen C, Thonney D, Chèvre N, Schärer M, Barry DA (2013) Treatment of micropollutants in municipal wastewater : ozone or powdered activated carbon ? Sci Total Environ 462:480–498.

Meybeck M, Chapman D V, Helmer R (1989) Global freshwater quality : a first assessment, Oxford. 1–306.

Milà i Canals L, Chapagain A, Orr S, Chenoweth J, Anton A, Clift R (2010) Assessing freshwater use impacts in LCA, part 2: case study of broccoli production in the UK and Spain. Int J Life Cycle Assess 15:598–607.

Milà i Canals L, Chenoweth J, Chapagain A, Orr S, Antón A, Clift R (2008) Assessing freshwater use impacts in LCA: Part I—inventory modelling and characterisation factors for the main impact pathways. Int J Life Cycle Assess 14:28–42.

Miller RE, Blair PD (2009) Input-Output analysis: foundations and extensions, Cambridge . 1–784.

Motoshita M, Itsubo N, Inaba A (2010) Development of impact factors on damage to health by infectious diseases caused by domestic water scarcity. Int J Life Cycle Assess 16:65–73.

Motoshita M, Itsubo N, Tahara K, Inaba A (2011) Damage assessment model for freshwater consumption and a case study on PET bottle production applied new technology for water footprint reduction in: Towards life cycle sustainability management. 399–410.

Mutel CL, Hellweg S (2009) Regionalized life cycle assessment: computational methodology and application to inventory databases. Environ Sci Technol 43:5797–803.

Norris G a. (2002) Impact characterization in the tool for the reduction and assessment of chemical and other environmental impacts. J Ind Ecol 6:79–101.

O'Rourke D (2014) The science of sustainable supply chains. Science 344:1124–7.

Odum H (1996) Environmental accounting: emergy and environmental decision making, Wiley. 1–384.

Ohlsson L (2000) Water conflicts and social resource scarcity. Phys Chem Earth 25:213–220.

Oki T, Kanae S (2006) Global hydrological cycles and world water resources. Science 313:1068–72.

Owens JW (2002) Water resources in Life-Cycle Impact Assessment: considerations in choosing category indicators. J Ind Ecol 5:37–54.

Owsianiak M, Rosenbaum RK, Huijbregts MA, Hauschild MZ (2013) Addressing Geographic Variability in the Comparative Toxicity Potential of Copper and Nickel in Soils. Environ Sci Technol 47:3241−3250.

Parekh NA (2012) Assessment of phosphorus fractions in streams draining different land use and development of new monitoring method. Master thesis at the University of Oslo. 1–155.

PE (2011) GaBi software and database. Available at: http://www.gabi-software.com/.

PE International (2013) GaBi database & modelling principles 2013. Leinfelden – Echterdingen, Germany. 1–162.

Pennington DW, Margni M, Ammann C, Jolliet O (2005) Multimedia fate and human intake modeling: spatial versus nonspatial insights for chemical emissions in Western Europe. Environ Sci Technol 39:1119–28.

Pennington DW, Margni M, Payet J, Jolliet O (2006) Risk and regulatory hazard-based toxicological effect indicators in Life-Cycle Assessment (LCA). Hum Ecol Risk Assess An Int J 12:450–475.

Pennington DW, Potting J, Finnveden G, Lindeijer E, Jolliet O, Rydberg T, Rebitzer G (2004) Life cycle assessment part 2: current impact assessment practice. Environ Int 30:721–39.

Peters G, Hertwich EG (2007) The application of multi-regional input-output analysis to industrial ecology: evaluating trans-boundary environmental impacts. in: Handbook of input-output analysis for industrial ecology. Dordrecht, The Netherlands

Peters GM, Wiedemann SG, Rowley H V., Tucker RW (2010) Accounting for water use in Australian red meat production. Int J Life Cycle Assess 15:311–320.

Pfister S (2014) New water data in ecoinvent slides for the 348th LCA DF Forum. Dübendorf, Switzerland

Pfister S, Bayer P (2014) Monthly water stress: spatially and temporally explicit consumptive water footprint of global crop production. J Clean Prod 73:52–62.

Pfister S, Bayer P, Koehler A, Hellweg S (2011) Environmental impacts of water use in global crop production: hotspots and trade-offs with land use. Environ Sci Technol 45:5761–8.

Pfister S, Hellweg S (2011) Surface water use—human health impacts. Report for the LC-IMPACT project (FP7 European commission project).

Pfister S, Hellweg S (2009) The water "' shoesize '" vs . footprint of bioenergy. Proc Natl Acad Sci U S A 106:93–94.

Pfister S, Koehler A, Hellweg S (2009) Assessing the environmental impacts of freshwater consumption in LCA. Environ Sci Technol 43:4098–4104.

Pistocchi A, Zulian G, Vizcaino P, Marinov D (2010) Multimedia assessment of pollutant pathways in the environment, European scale model. JRC scientific and technical reports. 1–55.

Posch M, Seppälä J, Hettelingh J-P, Johansson M, Margni M, Jolliet O (2008) The role of atmospheric dispersion models and ecosystem sensitivity in the determination of characterisation factors for acidifying and eutrophying emissions in LCIA. Int J Life Cycle Assess 13:477–486.

Potting J, Hauschild MZ (2006) Special issue honouring Helias A . Udo de Haes : Spatial differentiation in Life Cycle Impact Assessment: a decade of method development to increase the environmental realism of LCIA. Int J Life Cycle Assess 1:11–13.

Potting J, Schöpp W, Blok K, Hauschild M (1998) Site-dependent Life-Cycle Impact Assessment of acidification. J Ind Ecol 2:63–87.

PRé consultants (2013) Simapro 8. Available at: http://www.pre-sustainability.com/simapro8.

Quantis (2012) Quantis Water Database v1.33. Available at: http://www.quantis-intl.com/microsites/waterdatabase.php.

Raskin P, Gleick P, Kirshen P, Pontius G, Strzepek K (1997) Water futures: assessment of long-range patterns and prospects. 1–77.

Ridoutt B (2011) Development and application of a water footprint metric for agricultural products and food industry. In: Towards life cycle sustainability management. Springer, Netherlands

Ridoutt BG, Pfister S (2010a) Reducing humanity's water footprint. Environ Sci Technol 44:6019–21.

Ridoutt BG, Pfister S (2014) Water footprinting using a water stress index (WSI) that integrates stress on humans and ecosystems. Proceedings of 4th International Conference on Green and Sustainable Innovation (ICGSI). 1–4.

Ridoutt BG, Pfister S (2010b) A revised approach to water footprinting to make transparent the impacts of consumption and production on global freshwater scarcity. Glob Environ Chang 20:113–120.

Ridoutt BG, Williams SRO, Baud S, Fraval S, Marks N (2010) Short communication: The water footprint of dairy products: case study involving skim milk powder. J Dairy Sci 93:5114–7.

Rochat D, Margni M, Jolliet O (2006) Special Issue Honouring Helias A . Udo de Haes Continent-specific Intake Fractions and Characterization Factors for Toxic Emissions : Does it make a Difference ? Int J Life Cycle Assess 11:1–9.

Rockström J, Steffen W, Noone K, et al. (2009) A safe operating space for humanity. Nature 461:472–475.

Rosenbaum RK, Bachmann TM, Gold LS, Huijbregts MA, Jolliet O, Juraske R, Koehler A, Larsen HF, MacLeod M, Margni M, McKone TE, Payet J, Schuhmacher M, van de Meent D, Hauschild MZ (2008) USEtox—the UNEP-SETAC toxicity model: recommended characterisation factors for human toxicity and freshwater ecotoxicity in life cycle impact assessment. Int J Life Cycle Assess 13:532–546.

Rosenbaum RK, Huijbregts MAJ, Henderson AD, Margni M, McKone TE, van de Meent D, Hauschild MZ, Shaked S, Li D, Swirsky Gold L, Jolliet O (2011) USEtox human exposure and toxicity factors for comparative assessment of toxic emissions in Life Cycle Analysis: Sensitivity to key chemical properties. Int J Life Cycle Assess 15:710–727.

Roy P, Deschenes L, Margni M (2014) Uncertainty and spatial variability in characterization factors for aquatic acidification at the global scale. Int J Life Cycle Assess 19:882–890.

Roy P-O, Huijbregts M, Deschênes L, Margni M (2012) Spatially-differentiated atmospheric source–receptor relationships for nitrogen oxides, sulfur oxides and ammonia emissions at the global scale for life cycle impact assessment. Atmos Environ 62:74–81.

Rugani B, Huijbregts M a J, Mutel C, Bastianoni S, Hellweg S (2011) Solar energy demand (SED) of commodity life cycles. Environ Sci Technol 45:5426–33.

Santos LHMLM, Araújo a N, Fachini A, Pena A, Delerue-Matos C, Montenegro MCBSM (2010) Ecotoxicological aspects related to the presence of pharmaceuticals in the aquatic environment. J Hazard Mater 175:45–95.

Schwarzenbach RP, Gschwend PM, Imboden DM (2003) Environmental organic chemistry, John Wiley. 1327.

Seckler D, Barker R, Amarasinghe U (1999) Water scarcity in the twenty-first century. Water Resour Dev 15:29–42.

Sedlbauer VK, Braune A, Humbert S, Margni M, Schuller O, Fischer M (2007) Spatial differentiation in LCA: moving forward to more operational sustainability. Tech Theor und Prax 3:24–31.

Seppälä J, Posch M, Johansson M, Hettelingh J (2006) LCA Methodology country-dependent characterisation factors for acidification and terrestrial eutrophication based on accumulated exceedance as an impact category indicator. Int J Life Cycle Assess 11:403–416.

Shaked S (2011) Multi-Continental Multimedia Model of Pollutant Intake and Application to Impacts of Global Emissions and Globally Traded Goods. Thesis from University of Michigan. 1–177.

Shen Y, Oki T, Utsumi N, Kanae S, Hanasaki N (2008) Projection of future world water resources under SRES scenarios: water withdrawal. Hydrol Sci J 53:11–33.

Shiklomanov I, Rodda J (2003) World water resources at the beginning of the twenty-first century. Cambridge, United Kingdom. 1–25.

Smakhtin V, Revenga C, Döll P (2004) A pilot global assessment of environmental water requirements and scarcity. Water Int 29:307–317.

Smith VH (2003) Eutrophication of freshwater and coastal marine ecosystems a global problem. Environ Sci Pollut Res 10:126–139.

Stoessel F, Juraske R, Pfister S, Hellweg S (2012) Life cycle inventory and carbon and water FoodPrint of fruits and vegetables: application to a Swiss retailer. Environ Sci Technol 46:3253–62.

Sullivan C (2002) Calculating a water poverty index. World Dev 30:1195–1210.

Tabereaux AT, Peterson RD (2014) Aluminum production. In: Treatise on process metallurgy: industrial processes ,volume 3: industrial processes. 839–917.

U.S. Census Bureau (2014) https://www.census.gov/geo/reference/ua/urban-rural-2000.html. In: Census 2000 Urban Rural Classif. https://www.census.gov/geo/reference.

U.S. Environmental Protection Agency (2012) EPI SuiteTM v4.11. Available at: http://www.epa.gov/oppt/exposure/pubs/episuitedl.htm.

Udo de Haes HA, Finnveden G, Goedkoop M, Hauschild M, Hertwich E, Hofstetter P, Jolliet O, Klöpffer W, Krewitt W, Lindeijer E, Müller-Wenk R, Olsen S, Pennington D, Potting J, Steen B (2002) Life-Cycle Impact Assessment: striving towards best practice. 1–272.

Ui J (1992) Industrial pollution in Japan. Available at: http://archive.unu.edu/unupress/unupbooks/uu35ie/uu35ie00.htm.

UN Global Compact Office (2011) The CEO Water Mandate: an initiative by business leaders in partnership with the international community. 1–16.

UNEP SETAC Life Cycle Initiative (2014) WULCA. Available at: http://www.wulca-waterlca.org/projec.

United Nations Environment Programme (2007) Fourth Global Environment Outlook: environment for development. 1–572.

United Stated Geological Survey (USGS) (2012) The water cycle: freshwater storage. Available at http://ga.water.usgs.gov/edu/; watercyclefreshstorage.html.

Van De Meent D (1999) Potentially Affected Fraction as measure of toxic stress on ecosystems (Potentieel Aangetaste Fractie als maatlat voor toxische druk op ecosystemen). RIVM rapport 607504007. 1–66.

Van Hoof G, Buyle B, Kounina A, Humbert S (2013) Life cycle-based water assessment of a hand dishwashing product: opportunities and limitations. Integr Environ Assess Manag 9:633–44.

Van Zelm R, Schipper AM, Rombouts M, Snepvangers J, Huijbregts M a J (2011) Implementing groundwater extraction in life cycle impact assessment: characterization factors based on plant species richness for The Netherlands. Environ Sci Technol 45:629–35.

Verones F, Hanafiah MM, Pfister S, Huijbregts M a J, Pelletier GJ, Koehler A (2010) Characterization factors for thermal pollution in freshwater aquatic environments. Environ Sci Technol 44:9364–9.

Vince F (2007) UNEP/SETAC Life Cycle Initiative working group: assessment of water use and consumption within LCA. Report from Véolia Environment.

Vizcaíno P, Pistocchi A (2010) A GIS model-based assessment of the environmental distribution of gamma-hexachlorocyclohexane in European soils and waters. Environ Pollut 158:3017–27.

Vörösmarty C, Fekete B, Meybeck M, Lammers R (2000a) Geomorphometric attributes of the global system of rivers at 30-minute spatial resolution. J Hydrol 237:17–39.

Vörösmarty CJ, Green P, Salisbury J, Lammers RB (2000b) Global water resources: vulnerability from climate change and population growth. Science 289:284–288.

Wada Y, Wisser D, Bierkens MFP (2014) Global modeling of withdrawal, allocation and consumptive use of surface water and groundwater resources. Earth Syst Dyn 5:15–40.

Water Footprint Network (2011) WaterStat. Available at: http://www.waterfootprint.org/?page=files/WaterStat.

Water systems analysis group (2014) World water development report II. Indicators for world water assessment programme. Available at: http://wwdrii.sr.unh.edu/index.html.

WBCSD (2010) The global water tool. Available at: http://www.wbcsd.org. Geneva, Switzerland

Wegener Sleeswijk A, Heijungs R (2010) GLOBOX: A spatially differentiated global fate, intake and effect model for toxicity assessment in LCA. Sci Total Environ 408:2817–32.

Wenger Y, Li D, Jolliet O (2012) Indoor intake fraction considering surface sorption of air organic compounds for life cycle assessment. Int J Life Cycle Assess 17:919–931.

World Water Assessment Programme UN (2009) The United Nations World Water Development report 3: water in a changing world. 1–429.

Xenopoulos M a, Lodge D m, Alcamo J, Märker M, Schulze K, van Vuuren DP (2005) Scenarios of freshwater fish extinctions from climate change and water withdrawal. Glob Chang Biol 11:1557–1564.

Zhang Y, Baral A, Bakshi BR (2010) Accounting for ecosystem services in Life Cycle Assessment, Part II: toward an ecologically based LCA. Environ Sci Technol 44:2624–31.

Appendix

Part A: Review of methods addressing freshwater availability in life cycle inventory and impact assessment

Part B: Spatial analysis of toxic emissions in LCA: A sub-continental nested USEtox model with freshwater archetypes

Part C: Spatial analysis of toxic emissions to freshwater in LCA: operationalization at global scale

Appendix A

Review of methods addressing freshwater availability in life cycle inventory and impact assessment

A.1 Method evaluation

Table A.1, A.2 and A.3 summarize the evaluation against all criteria for all databases and methods.

Table A.1: Results of overall evaluations for inventory methods

Inventory databases					
Criteria	**Ecoinvent (Frischknecht et al. 2005)**	**GaBi (PE 2011)**	**WFN WaterStat (Water Footprint Network 2011)**	**Pfister et al. (2011)**	**Quantis Water Database** (2012)
Complete-ness of scope	• Detailed inventory database in terms of number of process-es presented • Spatial differentiation on a country scale where appropri-ate in regards to the product (global commodities, e.g., in-dustrial process, or region-specific products, e.g., electrici-ty mix).	• Detailed inventory database focused on industrial and agricul-tural water use • Spatial differentiation on a country scale for electricity mixes, but not for industrial or agricul-tural technologies and climatic conditions	• Inventory database focused on crops and derived crop products, farm animals and animal products biofuels, national consumption and production, trade in crop, animal and industrial products • Spatial differentiation for all countries for all products	• Inventory of consumed water for 160 crops and crop groups • Spatial differentiation for all countries	• Detailed inventory and impact data-base based on ecoinvent 2.2 list of processes • Spatial differentiation of processes on a country level • Spatial differentiation of impact assessment methods on a country level
Environ-mental relevance	• Surface water (river, lake, sea), groundwater (renewable, fossil), considered. Precipitation water stored as soil moisture water not considered • Consumptive use is an un-known part of the withdrawal. Degradative use considered through emissions to water, but released water not considered • Intake and released water quality not considered	• Surface water (river, lake, sea), groundwater (renewable, fossil), considered. Precipitation water stored as soil moisture water not considered • Consumptive use can be recal-culated (difference between water withdrawal and release). Degradative use considered through emissions to water and released water considered • Intake and released water quality not considered	• Surface water (river, lake) and groundwater (renewable, fossil) are not distinguished. Precipita-tion water stored as soil moisture considered. Seawater not consid-ered • Consumptive use considered. Degradative use considered through grey water • Intake water quality not consid-ered. Released water quality indirectly considered through grey water	• Surface water (river, lake) and groundwater (renewable, fossil) are not distinguished. Only irrigation and precipitation water consumed considered • Consumptive use considered. Degradative use not considered • Intake and released water quality not considered	• Surface water (river, lake, sea), groundwater (shallow and deep re-newable, fossil), precipitation water stored as soil moisture considered • Consumptive use and degradative use (released water) are distinguished as different types of outputs. Degradative use is also estimated according to WFN method for chemically polluted water • Intake water quality considered based on Boulay et al.’s classification (2011b). Released water quality indi-rectly considered through grey water

Scientific robustness and certainty	• Data uncertainty provided	• Data uncertainty not provided (lack of a meaningful measure)	• Data uncertainty not provided (lack of a meaningful measure)	• For each crop and country, two estimates are provided which can be used to evaluate model robustness/uncertainty • For 17 crops and 27 countries the uncertainty due to aggregation on country level is quantified by the coefficient of variation • Overall data uncertainty not quantified	• Data uncertainty provided based on various references used for the database
Documentation, transparency and reproductibility	• Data is generally disaggregated and therefore available • Accessible but not free • Method reviewed • Data reviewed • Data can be purchased • Published	• Data available on an aggregated and disaggregated level; some data are available on request only (data warehouse concept) • Accessible but not free • Method reviewed • Data reviewed • Data can be purchased • Published	• Data available on an aggregated level only • Accessible • Method reviewed • Data reviewed (compilation from various reviewed sources) • Published (open access)	• Data available on an aggregated level only • Accessible • Method reviewed • Data reviewed • Published (open access)	• Data is generally disaggregated and therefore available • Accessible but not free • Method under review • Data under review • Data can be purchased • Not published
Applicability	• Inventory data are straightforward to apply • Units are comparable with other inventory methods	• Inventory data are straightforward to apply • Units are comparable with other inventory methods	• Inventory data are straightforward to apply • Units (m^3) are comparable with other inventory method for “blue” and “green water” but not “grey water” (critical dilution volume)	• Inventory data are straightforward to apply • Units are comparable with other inventory methods	• Inventory data are straightforward to apply • Units are comparable with other inventory methods
Potential stakeholder acceptance	• Database already extensively used	• Database already extensively used	• Database already extensively used for agricultural and breeding purposes	• New database which acceptance still needs to be proven among industrial stakeholders due to the 'young age' of the method	• New database supported by industrial partners

Inventory methods							
Criteria	**Water footprint (Hoekstra et al. 2011)**	**WBCSD (2010)**	**Bayart (2008)**	**Boulay et al. (Boulay et al. 2011a)**	**Milà i Canals et al. (2008)**	**Vince (2007)**	**Peters et al. (2010)**
Completeness of scope	• Inventory method which distinguishes three water types: “green”, “blue”, “grey” • Countries where water is consumed can be distinguished at three levels (global, regional, local) • Not developed specifically for LCA, but can be made compatible	• Inventory method focused on industrial and agricultural water use • Output data can be linked to country and watershed water and sanitation availability information • Not developed specifically for LCA, but can be made compatible	• Inventory methodol integrating Vince's detailed description of water quality	• Classification which defines water by its source, quality parameters and functionality for downstream users, with an emphasis on definition of water quality	• Single method (besides Water Footprint Network method) which considers effect of land occupation and transformation on water availability and distinguishes fossil and renewable underground water	• First lines of an inventory method which brings a detailed description of water quality based on electricity required to achieve the upper quality level, through the distinction of 9 water flux types • Not developed specifically for LCA, but can be made compatible	• Inventory method developed to be applied to the meat production sector • Specific climate file is used for each site
Environmental relevance	• Surface water (river, lake), groundwater (renewable, fossil), precipitation water stored as soil moisture considered. Seawater not considered • Consumptive use considered. Degradative use considered through grey water • Intake and released water quality not considered	• Surface water (river, lake, sea), groundwater (renewable), precipitation water stored as soil moisture considered. Fossil groundwater not considered • Consumptive use considered. Degradative use not considered • Intake and released water quality not considered	• Surface water (river, lake, sea), groundwater (renewable, fossil), precipitation water stored as soil moisture water considered • Consumptive use considered. Degradative use considered through released water at a given quality • Intake and released water quality considered through eight water flux types	• Surface water (river, lake, sea), groundwater (renewable, fossil), precipitation water stored as soil moisture considered. Fossil groundwater is considered indirectly as being groundwater with a maximal scarcity • Consumptive use considered. Degradative use considered through released water at a given quality • Intake and released water quality considered through 137 quality parameters	• Surface water (river, lake), groundwater (renewable, fossil), precipitation water stored as soil moisture considered. Seawater not considered • Consumptive use considered. Degradative not considered • Intake and released water quality not considered	• Surface water (river, lake, sea), groundwater (renewable), considered. Precipitation water stored as soil moisture and fossil groundwater not considered • Consumptive use considered. Degradative use considered through released water at a given quality • Intake and released water quality considered through nine water flux types	• Surface water (river, lake), groundwater (renewable, fossil), precipitation water stored as soil moisture considered. Seawater not considered • Consumptive use considered. Degradative use considered through water released • Intake and released water quality considered implicitly in source sustainability characterization

Scientific robust-ness and certainty	• No data	• No data	• No data	• No data	• No data	• No data	• Underlying data for a unique application in 3 regions of Australia • Climatic model for precipitation water stored as soil moisture estimation uncertainty provided
Docu-menta-tion, trans-parency and repro-ductibil-ity	• Accessible • Reviewed through different scientific journal papers • Published	• Accessible • Partly reviewed by the GRI and The Nature Conservancy • Published	• Accessible online on databases such as Proques • Not reviewed • Published as a master thesis	• Accessible • Reviewed • Published	• Accessible • Reviewed • Published	• Not accessible • Not reviewed • Not published	• Accessible • Reviewed • Published
Applica-bility	• Easy applicability in general, but not in the LCA context. Not adapted to LCA software tools • Units (m^3) are not all com-parable with other inventory methods as volume of “grey” water cannot be estimated as equivalent to volume of “green” and “blue” water	• Easy applicability in general, but not in the LCA context. Ready-made life cycle inven-tory figures are directly available in the tool, but not adapted to LCA software tools • Units are comparable with other inventory methods	• Units are comparable with other inventory methods	• Units are comparable with other inventory methods	• Units are comparable with other inventory methods	• Units are comparable with other inventory methods	• Units are comparable with other inventory methods
Potential stake-holder ac-ceptance	• Principles of the method easily understandable and widely used by companies	• Principles of the method easily under-standable and widely used by companies	• The acceptance might be difficult as it is not reviewed	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method	• Acceptance still to be proven among industri-al stakeholders due to the method 'young age'	• Not published	• Acceptance still to be proven among industrial stakeholders due to the method 'young age'

Table A.2: Results of overall evaluations for midpoint impact assessment methods

Midpoint impact assessment methods				
Criteria	**Swiss ecological scarcity** (Frischknecht et al. 2006)	**Water impact index (Bayart et al. 2014)**	**Pfister et al. (2009)**	**Water footprint impact indexes (Hoekstra et al. 2011)**
Complete-ness of scope	• Single indicator which does not cover a specific AoP • Focus on water scarcity quantification through ways in which the availability of freshwater is impacted • Spatial differentiation at the country and watershed level	• Single indicator which does not cover a specific AoP • Focus on modification of freshwater availa-bility generated by a human activity. It allows evaluating how other water users (both humans and ecosystems) would potentially be deprived from this resource • Spatial differentiation based on scarcity index chosen by the practitioner (e.g., Pfister et al. (2009))	• Single indicator which does not cover a specific AoP • Focus on water scarcity quantification • Spatial differentiation at the country, watershed (11050 watersheds) or 0.5° grid cell level	• Three indicators, which do not cover a specific AoP. “Green”, “blue” and “grey” water are charac-terized respectively by “green”, “blue” and “grey” water scarcity index in three disaggregated results • Focus on water scarcity quantification through ways in which the availability of freshwater is im-pacted • Spatial differentiation possible • Not developed specifically for LCA but compatible
Environ-mental relevance	• Surface water (river, lake), groundwater (renewable, fossil), considered. Precipita-tion water stored as soil moisture and seawater not considered • Application on consumptive use rec-ommended • Intake and released water quality not considered • Water scarcity and water renewability rate (fossil water gets the same eco-factor like water used in regions with "extreme water scarcity”) considered. Economic development level, water functionalities not considered • Compensation mechanisms not consid-ered	• Surface water (river, lake), groundwater (renewable, fossil), precipitation water stored as soil moisture considered. Seawater not considered. Additionally, precipitation water over cities considered • Degradative and consumptive use consid-ered • Intake and released water quality consid-ered • Water scarcity, water renewability rate and water quality are considered through the water scarcity index and water quality index. Water functionalities and economic devel-opment level not considered • Compensation mechanisms not considered	• Surface water (river, lake), groundwa-ter (renewable, fossil), considered. Pre-cipitation water stored as soil moisture and seawater not considered • Consumptive uses considered as water consumption can be characterized by the WSI. Degradative use not considered • Intake and released water quality not considered • Water scarcity and water renewability rate considered through the water scarci-ty index. Water quality, economic devel-opment level, water functionalities not considered • Compensation mechanisms not consid-ered	• Surface water (river, lake), groundwater (renewa-ble, fossil), precipitation water stored as soil mois-ture considered. Seawater not considered • Degradative and consumptive use considered through “blue”, “green” and “grey” water • Intake water quality not considered, released water quality indirectly considered through grey water • Water scarcity and water renewability rate consid-ered through the water scarcity index. Water quali-ty, economic development level, water functionali-ties not considered. • Compensation mechanisms not considered

Scientific robustness and certainty	• Indicator uncertainty provided by a semi-quantitative indicator related to the eco-factors • Based on a scarcity indicator which does not reflect a particular cause-effect chain • Indicator cannot be verified against monitoring data (expressed in eco-point units)	• Model uncertainty not provided • Based on a scarcity indicator which does not reflect a cause-effect chain • Indicators cannot be verified against monitoring data (expressed in m^3 equivalent)	• Model uncertainty not provided • Based on a scarcity indicator which does not reflect a cause-effect chain • Indicator cannot be verified against monitoring data	• Model uncertainty not provided • Based on a scarcity indicator which does not reflect a cause-effect chain • Indicators cannot be verified against monitoring data (expressed in m^3 equivalent)
Documentation, transparency and reproducibility	• Accessible • Reviewed by a panel of industry and governmental experts • Published	• Not accessible • Under review • Not published	• Accessible • Reviewed • Published	• Accessible • Peer reviewed by different scientific journal papers • Published
Applicability	• CF available for all countries and watersheds • Units cannot be compared with other methods (eco-points), but within distance to target methods	• CF not available • Units cannot be compared with other methods (m^3eq)	• CF available at the country and grid cell level on a Google Earth layer • Units cannot be compared with other methods (m^3eq)	• CF available only for blue water scarcity for major watersheds • Units cannot be compared with other methods (m^3eq) • Grey water impact index leads to double counting of emissions to water already covered by other impact categories (such as eutrophication)
Potential stakeholder acceptance	• Indicator easily understood as eco-points have a long tradition in LCA • Endorsed at a national level by the Swiss FOEN	• Not published	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method	• These indicators derived from the water footprint concept give an opportunity to make a parallel with the LCA impact modeling concept. However, they do not aim at being applied as such as the author explicitly refrains from recommending these impact indices to be applied as characterization factors in LCA.

Midpoint impact assessment methods				
Criteria	**Ridoutt and Pfister (2010b)**	**Boulay et al. (2011b)**	**Bayart et al. (2008)**	**Milà i Canals et al. (2008)**
Completeness of scope	• Single indicator based on Pfister et al.'s (2009) midpoint method which does not cover a specific AoP • Focus on water scarcity quantification through ways in which the availability of freshwater is impacted • Spatial differentiation at the watershed and country level	• Single indicator which does not cover a specific AoP • Focus on water scarcity and quality quantification • Spatial differentiation at the country, watershed or grid cell (808 grid cells) level • Not a specific midpoint oriented method	• Covers midpoint for AoP *human health* • Focus on the quantification of impact of water quantity decrease on "sufficiency of freshwater resource for contemporary human users. Water deprivation for different users (domestic use, irrigation, fisheries and aquaculture) distinguished • Spatial differentiation at the country level (CF available for 17 countries)	• Two indicators which cover the impact pathways: *ecosystem quality* (freshwater ecosystem impact (FEI)) and *resources* (freshwater depletion(FD)). • Focus on the quantification of impact of water quantity decrease on *ecosystem quality* and *resources*. *Human health* is not covered as the method only considers water quantity • Spatial differentiation at watershed and country level for FEI and aquifer level for FD
Environmental relevance	• Surface water (river, lake), groundwater (renewable, fossil), considered. Precipitation water stored as soil moisture and seawater not considered • Degradative and consumptive use considered through blue and grey water • Intake water quality not considered, released water quality indirectly considered through grey water • Water scarcity and water renewability rate considered through the water scarcity index. Water quality, economic development level, water functionalities not considered • Compensation mechanisms not considered	• Surface water (river, lake, sea), groundwater (renewable, fossil), precipitation water stored as soil moisture considered. Fossil groundwater is considered indirectly as being groundwater with a maximal scarcity • Consumptive use and degradative use considered • Intake and released water quality considered • Water scarcity, water quality, water renewability rate considered through the water scarcity index. Economic development level and water functionalities are not considered • Compensation mechanisms not considered	• Surface water (river, lake), groundwater (renewable, fossil), considered. Precipitation water stored as soil moisture and seawater not considered • Consumptive use considered. Degradative use considered through released water at a given quality • Intake and released water quality considered • Water scarcity, water quality, water functionalities and economic development level considered • Compensation mechanisms considered	• Surface water (river, lake), groundwater (renewable, fossil), net precipitation water stored as soil moisture considered. Seawater not considered • Consumptive use considered. Degradative use not considered as water quality not taken into account • Intake and released water quality not considered • FEI considers water scarcity but not water quality. FD takes into account renewability rate through the abiotic depletion potential (ADP) • Compensation mechanisms not considered

Scientific robustness and certainty	• Model uncertainty not provided • Based on a scarcity indicator which does not reflect a cause-effect chain • Indicators cannot be verified against monitoring data (expressed in m^3 equivalent)	• Model uncertainty not provided • Based on a scarcity indicator which does not reflect a cause-effect chain • Indicator cannot be verified against monitoring data	• Model uncertainty not provided • The model reflects a part of the cause-effect chain • Indicator cannot be verified against monitoring data	• Model uncertainty not provided • The model reflects a part of the cause-effect chain • Indicators can only partially be verified against monitoring data
Documentation, transparency and reproducibility	• Accessible • Peer reviewed • Published	• Accessible • Reviewed • Published	• Accessible online on databases such as Proquest • Not reviewed in a scientific journal • Published as a master thesis	• Accessible • Reviewed • Published
Applicability	• Pfister et al's CF available at the 0.5° grid cell and country level • Units cannot be compared with other methods (m^3eq)	• CF available at the country and watershed level • Units cannot be compared with other methods (m^3eq)	• CF available for 17 countries • Units cannot be compared with other methods (not a common midpoint)	• CF available for FEI for all countries and partially for watersheds (no global coverage), but not for FD • FEI units cannot be compared with other methods (m^3 of "ecosystem equivalent" water). FD units can be compared (Sb-eq/kg, CML compatible)
Potential stakeholder acceptance	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method. The acceptance might be difficult as it is not reviewed	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method

Table A.3: Results of overall evaluations for endpoint impact assessment methods

Endpoint impact assessment methods				
Criteria	**Pfister et al. (2009)**	**Motoshita et al. (2010): infectious diseases arising from domestic water consumption**	**Motoshita et al. (2011): health damages of undernourishment related to agricultural water scarcity**	**Boulay et al. (2011b)**
Completeness of scope	• Covers all AoPs • Spatial differentiation at the watershed and country level	• Covers partially AoP *human health* • Focus on the relationship between the available quantity of domestic water and health damages of infectious diseases by conducting multiple regression analyses based on statistical data • Spatially differentiation at the country level	• Covers partially AoP *human health* • Focus on the health damages of undernourishment related to agricul-tural water scarcity (not domestic and fisheries) based on statistical analysis • Spatially differentiation at the country level	• Covers AoP *human health* • Focus on the impact mechanism due to agriculture, domestic use and fisher-ies • Spatial differentiation at the country and watershed level
Environmental relevance	• Surface water (river, lake), groundwater (renewable), precipitation water stored as soil moisture considered. Seawater and fossil groundwater not considered. Net precipita-tion water stored as soil moisture can be considered through change in blue resulting of land occupation and transformation in-ventory. • Consumptive use considered. Degradative use not considered • Intake and released water quality not considered • Covers AoP *human health* by considering impact mechanism due to agriculture use (not domestic water and fisheries), including scarcity and economic development level as parameters (not water functionalities). Com-pensation mechanisms are not taken into account unless the system is expanded	• Surface water (river, lake), groundwater (renewable), considered. Precipitation water stored as soil moisture, seawater and fossil groundwater not considered • Consumptive use considered. Degradative use not considered • Intake and released water quality not considered • Water functionality (only domestic) and economic development level (through acces-sibility to safe water) considered (not water quality). Water scarcity is only considered through “domestic scarcity” which corre-sponds to domestic water availability • Compensation mechanisms are partly taken in account through house connection rate to water supply and sanitation. The method does not include the impact of the compensation mechanisms	• Surface water (river, lake), groundwater (renewable), consid-ered. Precipitation water stored as soil moisture, seawater and fossil groundwater not considered • Consumptive use considered. Degradative use not considered • Intake and released water quality not considered • Water scarcity, water functionality (only agricultural) and economic development level (through dietary energy consumption and gross na-tional income) considered. Water quality not considered • Compensation mechanisms are not taken into account	• Surface water (river, lake, sea), groundwater (renewable, fossil), pre-cipitation water stored as soil moisture considered. Fossil groundwater is considered indirectly as being ground-water with a maximal scarcity • Consumptive and degradative use considered • Intake and released water quality considered • Water scarcity, water functionalities, economic development level and water quality considered • Compensation mechanisms are taken in account through the adaptation capacity

	• Covers AoP *ecosystem quality* including water resource scarcity and water ecological value (through vegetation/net primary production dependence on water) as parameters (not water quality) • Covers AoP *resources* including water renewability rate through fraction of freshwater consumption that contributes to depletion			
Scientific robustness and certainty	• Model uncertainty not provided • The model reflects a part of the cause-effect chain • The indicator for AoP *human health* (DALY) and AoP *ecosystem quality* (PDF•m^2•y) can be verified against monitoring data whereas indicator for AoP *resources* cannot (MJ)	• Model uncertainty not provided • The model reflects a part of the cause-effect chain • Indicator (DALY) can be compared with monitoring data	• Model uncertainties provided through R square and t-value of regression analysis • The model reflects a part of the cause-effect chain • Indicator (DALY) can be compared with monitoring data	• Model uncertainties provided through the p-value and confidence interval • The model reflects a part of the cause-effect chain • Indicator (DALY) can be compared with monitoring data
Documentation, transparency and reproducibility	• Accessible • Reviewed • Published	• Accessible • Reviewed • Published	• Partly accessible • Not reviewed yet • Not published	• Accessible • Reviewed • Published
Applicability	• CF available at the country level and watershed level and can be integrated in Eco-indicator-99 LCIA method • All endpoints are in commonly used units (DALY, PDF•m^2•y, MJ)	• CF available at the country level • Endpoint is in commonly used units (DALY)	• CF not available • Endpoint is in commonly used units (DALY)	• CF available at the country and watershed level • Endpoint is in commonly used units (DALY)
Potential stakeholder acceptance	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method	• Not published	• Acceptance still to be proven among industrial stakeholders due to the method 'young age'

Criteria	Hanafiah et al. (2011)	Van Zelm et al. (2011)	Maendly and Humbert (Maendly and Humbert 2009)	Bösch et al. (2007)
Completeness of scope	• Covers partially AoP *ecosystem quality* • Focus on impact of consumption on aquatic ecosystems • Spatial differentiation at the watershed level (214 watershed covered)	• Covers partially AoP *ecosystem quality* • Focus on impact of groundwater extraction on occurence of soil plant species • No spatial differentiation (CF for Netherlands only)	• Covers partially AoP *ecosystem quality* • Focus on the assessment of the impact of turbined water on aquatic biodiversity • Limited spatial differentiation through distinction between alpine and non-alpine dam	• Covers partly AoP *resources* • Not specific to water, so it needs further development • No spatial differentiation
Environmental relevance	• Surface (river, lake) and groundwater (renewable) considered. Precipitation water stored as soil moisture, fossil groundwater and seawater not considered • Consumptive use considered. Degradative use not considered • Intake and released water quality not considered • Water ecological value considered through the global species-discharge model developed by Xenopoulos et al. (2005) for 214 global river basins. Water scarcity and intake water quality not considered	• Only effects on renewable shallow (<3.5 m depth) groundwater considered. Precipitation water stored as soil moisture, surface water (river, lake, sea), fossil groundwater not considered • Consumptive use considered. Degradative use not considered • Intake and released water quality not considered • Water ecological value considered through empirical observation of decreased biodiversity (multiple regression equation). Water scarcity and water quality not considered	• Only surface water (river, lake) considered. Precipitation water stored as soil moisture, groundwater (renewable, fossil) and seawater not considered • Degradative use (turbined water) considered. Consumptive use not considered • Intake and released water quality not considered • Water ecological value considered through empirical observation of decreased biodiversity (multiple regression equation) as parameter. Water scarcity and water quality not considered	• Surface water (river, lake, sea), groundwater (renewable, fossil), considered. Precipitation water stored as soil moisture water not considered (same classification as ecoinvent, except seawater is taken as reference) • Consumptive use considered. Degradative use not considered • Intake and released water quality not considered • Water renewability rate implicitly considered through notion of exergy
Scientific robustness and certainty	• Model uncertainty not provided • The model reflects a part of the cause-effect chain (fate part by considering that change in water consumption is fully reflected in a change in water discharge, effect part with global species-discharge model). • Indicator (PDF) can be compared with monitoring data	• Model uncertainties provided through R square value of regression analysis • The model reflects a part of the cause-effect chain (fate part with the MODFLOW model, effect part with a statistical model) • Indicator (PNOF) can be compared with monitoring data	• Indicator uncertainty provided through upper and lower threshold • The method simplifies the cause-effect chain by a simple statistical regression • Indicator (PDF•m^2•y) can be verified against monitoring data	• Model uncertainty provided through semi-quantitative information • The method does not really reflect the cause effect chain • Indicator (exergy MJ) cannot be verified against monitoring data
Documentation, transparency	• Accessible • Reviewed	• Accessible • Reviewed	• Accessible • Under review	• Accessible • Reviewed

and reproducti-bility	• Published	• Published	• Not published	• Published
Applicability	• CF available • Endpoint is in commonly used units (PDF·m^3·y)	• CF available for the Netherlands • Endpoint is in commonly used units (PNOF•m^2•y)	• CF available • Endpoint is in commonly used units (PDF•m^2•y).	• CF available • Endpoint is not in commonly used unit (MJ exergy)
Potential stake-holder ac-ceptance	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method	• Acceptance still to be proven among industrial stakeholders due to the 'young age' of the method	• Not published	• As the method is not specific to water, its acceptance as a water indicator is fair

A.2 Short description of each method

A.2.1 Inventory databases

Ecoinvent (Frischknecht et al. 2007)

The ecoinvent database contains elementary flows for water. These flows are for water withdrawal (sometimes referred as non-turbined water) as well as turbined water and salt water, all expressed in m^3 of water. The ecoinvent database distinguishes between water from lake; river; ground (well); unspecified natural origin; turbined water; and other non-elementary flows such as salt water from sole (e.g., produced water in oil and gas extraction); and salt water from ocean. It also has a flow referred as cooling water from unspecified natural origin. Because each unit process dataset contains standardized information about its location (extended list of ISO 3166 two letter codes), spatial differentiation is supported. Water outputs and input water quality are not considered.

Gabi (PE 2011)

The following flows are currently used in the GaBi software and database: feed, river, ground water, lake, sea water, well water, water salt sole, water (used in turbine), surface water (unspecified) and water. (Sven Lundie, personal communication, 21 December 2009, 8 January 2010). Water outputs are considered and input water quality is not considered.

The WFN database (Water Footprint Network 2011)

The WFN assesses the inventory flows of crops and derived crop products, biofuels, farm animals and animal products, national consumption and national production, virtual-water flows related to international trade in crop, animal and industrial products, as well as national and global water savings related to trade in agricultural and industrial products according to Hoekstra et al.'s method (Hoekstra et al. 2011). Databases considers "green" water, consumptive use of surface and groundwater ("blue water") and degradative use of water ("grey water") for all countries. Water outputs and input water quality are not considered.

Pfister et al.'s database (2011)

Pfister et al.'s database assesses the water consumption for the production of 160 crops covering most harvested mass on global cropland, including full-irrigation water consumption, deficit water consumption and expected water consumption.

The Quantis Water Database (2012)

The Quantis Water Database is based on ecoinvent data and aims at filling the gap and providing practitioners of water footprint the data and structure they need to apply the latest methodologies. The project is supported by industrial partners (Danone, Kraft, L'Oréal, Molson Coors, Natura, Steelcase and Veolia Environnement) and by ecoinvent.

A.2.2 Inventory models

WFN (Hoekstra et al. 2011)

The water footprint method developed by Hoekstra et al. calculates the amount of water consumed or polluted during the production of a product. The method can also be applied to compute the water consumed to generate the goods and services that are produced by a country (i.e., the water footprint of a nation - (Chapagain and Hoekstra 2004)), which is equivalent to the entire amount of water used per country. This method is closely linked to the virtual water concept. Virtual water is the amount of water that is required to produce a certain good, commodity or service (Allan 1996) and does not contain spatial and temporal information. It is termed virtual because the largest part of the water is not incorporated into the product itself.

The water footprint method distinguishes among "green," "blue" and "grey" water. The green water footprint refers to consumption of green water resources (rainwater stored in the soil as soil moisture) by evapotranspiration (Hoekstra et al. 2011). The blue water footprint refers to consumption of blue water resources (surface and ground water) along the supply chain of a product (Hoekstra et al. 2011). 'Consumption' refers to loss of water from the available ground-surface water body in a catchment area, which happens when water evaporates, returns to another catchment area or the sea or is incorporated into a product. The grey water footprint refers to pollution and is defined as the volume of freshwater that is required to assimilate the load of pollutants based on existing ambient water quality standards (Hoekstra et al. 2011). Grey water can also contain the amount of water used for cooling and returned to the watershed with a temperature higher than the receiving body. However, cooling water is, at presents still rarely considered in grey water. Water withdrawal that is returned, unpolluted, to the watershed or turbined water (in-stream use) are not included in blue water. The water footprint method in itself is an inventory (in kg, liters or m3 of water consumed). Spatial differentiation is possible for the inventory by indicating the location of water consumption (for example, on a map). The water footprint method is developed and promoted by the water footprint network.

Although this method provides a simple and visual inventory indicator of consumptive water through blue and green water, and degradative use through grey water, other communication difficulties emerge with the use of an increasingly large palette of water colors and the conflict between the well-established definition of grey water in the water industry (e.g., wastewater generated from activities such as laundry, dishwashing, and bathing (Henriques and Louis 2011)).

Note that the term "water footprint", as used by Hoekstra et al. (2011) and the Water Footprint Network is currently debated. For example, Ridoutt and Pfister (2010b) suggest that the term water footprint should be reserved for assessments which describe the impact of consumptive water use on freshwater availability and not solely for the amount of water consumed.

WBCSD (2010)

The global water tool is a free tool of the World Business Council for Sustainable Development (WBCSD) launched in 2009. It inventories the water use of companies and organizations, providing a map of water use and water consumption combined with a map of water scarcity at the watershed level. By mapping the water use on water scarcity maps, a visual and qualitative assessment of potential risks relative to their global operations and supply chains is possible. The tool consists of an Excel work book and online mapping tool and addresses questions, like 'How many of the sites lie in water scarce areas, and how many will in the future?', or 'How many employees live in countries that lack access to improved water and sanitation?'. The model also attempts to evaluate the number of the company's suppliers that are located in water scarce areas and how many will be in the future. However, the tool does not provide any specific guidance on local situations; this would require more in-depth systematic analysis. The tool also allows companies to compare their water use with validated water and sanitation availability (Total Renewable Water Resources per person, Access to Improved Water, Access to Improved Sanitation) both on country and watershed levels. It allows the calculation of Global Reporting Initiative (GRI) indicators linked to water (EN8, EN10, EN21).

Bayart (2008)

Bayart developed an inventory method for assessing off-stream freshwater use in life cycle assessment. It distinguishes eight water flux types (elementary flows of underground water of drinkable, good and bad quality, surface water of drinkable, good and bad quality, wastewater of bad quality and saltwater).

The inventory method deals only with off-stream water uses (e.g., water for irrigation, industrialized processes, cooling water) (e.g., water for irrigation, industrialized processes, cooling water). It assesses the modification of water availability for further uses. This modification can be generated by: (1) freshwater consumptive use (a reduction of the net volume of water within the watershed) and (2) degradative use as it provokes a loss of water functionality.

Boulay et al. (2011a)

This inventory method is an operational version of Bayart's method. They suggest classifying water into 8 different quality level, which can be from surface or ground, as well as one rain water category. They represent different combinations of low, medium or high for toxic and microbial contamination. These classes were elaborated based on the quality required for a water to be functional for specific users. These threshold values were determined based on international standards, national regulations and industry data. The final version is published.

Milà I Canals et al. (2008)

Milà I Canals et al. propose a method to assess impacts of freshwater use in LCA. It takes in account all off stream and in-stream uses (except aquaculture) and considers the land use occupation and transformation through the change in evapotranspiration and runoff. In this method, the word "evaporative" refers to consumptive water use. The types of water use considered in the inventory are (i) surface and groundwater evaporative use, (ii) evaporative and non-evaporative uses of water stocks (groundwater—fossil water) and over extracted water funds (groundwater—aquifers), and differences in rainwater availability caused by land use and land use change.

Vince (2007)

The inventory framework proposed by Vince is a preliminary approach for an inventory method based on a detailed description of water quality and electricity required to achieve an upper quality level (e.g., from drinking to ultrapure water, or from average quality river water to high quality) through the distinction of nine types of water flux. The nine water flux types are as follows: ultrapure water/desalinated water, drinking water, groundwater, high quality river water, low quality aquifer/average quality river water/treated wastewater, brackish water/low quality river water (in emerging countries), municipal wastewater, industrial wastewater, and seawater. This method considers degradative, consumptive and borrowing uses but only for off-stream use. This approach can contain spatial information. This inventory method is yet neither published nor reviewed.

Peters et al. (2010)

Peters et al focus on the link between LCI reporting and its use in public debate and propose that as an interim measure while international consensus on impact mechanisms is developed, water should be considered 'used' in the production of goods when it is delivered by unnatural means or it leaves the production site at a lower quality. They apply detailed local hydrological modeling with field data to calculate all natural and engineered water inputs and outputs. Inputs of rain and outputs of water vapour are considered sustainable and high quality, and in some agricultural systems may not have changed significantly since before the development of agricultural society. These are therefore reported separately, while transferred flows and funds are grouped as water used from less sustainable water sources, and water which is excreted, drains, runs off, is discharged to sewer or alienated in a product is grouped as used by virtue of the quality change.

A.2.3 Impact assessment, at midpoint level

Swiss Ecological Scarcity (Frischknecht et al. 2006)

The Swiss ecological scarcity method is a "distance-to-target" method. Indeed, the calculation of the eco-factor is determined by setting the current flow (corresponding to the actual situation) into relation with the critical flow (deduced from legislative guidelines or political goal). The unit given by this method is eco-points (EP, or UBP = Umweltbelastungspunkte, meaning eco-points in German) for various impacts on the environment: the fewer eco-points, the better. It only considers total freshwater withdrawal. First, the method performs a normalization step in which 1 eco-point is assigned to the total water use in the reference region. Then a the weighting step assigns Eco-points to Eco-factors calculated depending on total water use and critical water use (i.e., the amount of water use at which scarcity starts to be experienced, set by default to 20 % of water renewable rate) of a water shed area, a country or region. Ecological water scarcity is defined for each individual watershed area. Six scarcity classes are proposed to simplify life cycle inventory modeling. The water scarcity value of each individual watershed area can be assigned to one of these six scarcity classes. A temporal or spatial differentiation can be performed with the version of 2006 depending on available data.

Veolia (Bayart et al. 2014)

The Water Impact index is a simplified metric for assessing impacts on water use. This indicator aims to address the modification of freshwater resource availability due to human activities. It allows evaluating how other water users (both humans and ecosystems) would potentially be deprived from this resource. It allows to cover implicitly all three midpoints areas of protection (human health, ecosystem quality and resources).

Water flows abstracted from, or released into the environment are weighted by (1) a water scarcity index of the location where the water is used (e.g. Pfister's Water Stress Index); and (2) by a quality index. The latter is calculated as a ratio between a reference concentration based on Environmental Quality Standards and the actual concentration in the flow, for a specific pollutant. In the case of multiple pollutants, the quality index is calculated according to the most penalizing pollutant. If, for all pollutants considered, the concentration is above the reference concentration, the Quality Index is set to 1 (water quality reaching environmental requirements).

Water withdrawal is accounted positively (increase of Water Impact Index: reduction of water availability). Water discharge is accounted negatively (decrease of Water Impact Index: increase of water availability).

Pfister et al. (2009)

The method by Pfister et al. assesses the impacts of freshwater consumptive use on human health, ecosystem quality and resources. At the midpoint level, the characterization factor is the adapted water scarcity index (WSI, see below). This index uses a modified withdrawal to hydrological availability factor (WTA^{*}) (calculated as a criticality ratio), which differentiates watersheds with strongly regulated flows (SRF). WTA^{*} introduces a variation factor (VF) which takes into account insufficient water storage capacities or lack of stored water in case of increased water scarcity during specific periods due to both monthly and annual variability of precipitation.

$$WTA^{*} = \sqrt{VF} \times WTA \text{ for SRF}$$

$$WTA^{*} = VF \times WTA \text{ for non-SRF}$$

Equation A.1: Pfister et al.'s withdrawal to hydrological availability factor

The formula proposed by Pfister et al. (2009) to evaluate the modified annual freshwater withdrawals to hydrological availability of a specific watershed (WTA^{*}))) is presented in Equation A.1 (see Pfister et al. (2009).

$$WSI = \frac{1}{1 + e^{-6.4 \times WTA} \times (\frac{1}{0.01} - 1)}$$

Equation A.2: Pfister et al.'s water scarcity index

The adapted water scarcity index calculated based on Equation A.2 can serve as a characterization factor for water consumption in life cycle impact assessment for, for example, screening assessments for human health and ecosystems but not for resources. The results can be expressed in scarcity-characterized water footprint (m^3) (when the amount of water is multiplied by the WSI) (Ridoutt and Pfister 2010b) or scarcity-weighted water footprint for a certain region-equivalent (m^3-eq) (when the scarcity-characterized water footprint is divided by the WSI of a reference region) (2010b). The result is defined as water footprint by Pfister and Hellweg (Pfister and Hellweg 2009).

See Pfister et al. (2009) or "Water_Methods_Flows&CF_RESULTS" for the list of country-based characterization factors. Watershed values can be downloaded as kmz-file (for use in Google Earth) at http://www.ifu.ethz.ch/staff/stpfiste.

Hoekstra (2011)

Green, blue and grey water are characterised respectively by green, blue and grey water scarcity index in three disaggregated results. It allows to cover all three midpoints areas of protection (human health, ecosystem quality and resources) implicitly through each of these 3 indicators. The green and the blue water scarcity indexes focus respectively on green and blue water scarcity defined as the ratio of the total green water footprint in the catchment to the green water availability and the ratio of the total blue water footprint in the catchment to the blue water availability. The water pollution level index focuses on the fraction of the pollution assimilation capacity consumed, i.e. by taking the ratio of the total grey water footprint in a catchment (WFgrey) to the runoff from that catchment.

Ridoutt and Pfister (2010b)

Pfister et al.'s characterisation factors (Pfister et al. 2009) are applied on blue water consumption and gray water.

Boulay et al. (2011b)

The midpoint proposed by Boulay et al. is the scarcity parameter of their endpoint model for human health. This scarcity is distinct for different water categories, and is null for water of quality as low as seawater. For surface water, the parameter is based on the CUs/Q90 proposed by Döll (2009) where CUs is the surface water consumed and Q90 is a "statistical low flow" accounting for seasonal variation. For groundwater, it is similarly CUg/GWR, where CUg is the groundwater consumed and GWR is the availability of groundwater resource. These ratios are then adapted to include the local water quality availability based on available data from GEMStat database. This midpoint is calculated at the watershed scale and can be used for all three endpoint categories; human health, ecosystems and resources.

Bayart (2008)

Bayart, as part of his master thesis, developed a method to evaluate the impact of water deprivation for human use. He developed characterization factors which vary according to repartition of human use among possible functionalities, water scarcity and water quality. Compensation scenarios are also taken in account but their impact is not explicitly calculated. The modeling phase is implemented in two softwares: Excel and Analytica

Milà I Canals et al. (2008)

The method proposed by Milà I Canals et al. introduces two midpoint impact categories: the freshwater ecosystem impact (FEI) and the freshwater depletion (FD). It focuses on impact from surface and groundwater evaporative use and land use transformation. The water uses considered are all evaporative uses of freshwater (including evaporated irrigation water, cooling water, evaporated water from dams and reservoirs, etc.). Milà I Canals et al. (2008) acknowledge that it can lead to an underestimation of local effects, when non-evaporative uses are considered to have no impact on freshwater ecosystem impact. The FEI impact indicator is calculated with a water scarcity indicator, to be chosen between Falkenmark et al.'s water resources per capita (Falkenmark et al. 1989), Raskin et al.'s water use per resource (WUPR) (Raskin et al. 1997) or Smakhtin et al.'s environmental water scarcity (2004). The FD impact indicator accounts for the depletion of water stocks and funds. FD is calculated via an abiotic resource depletion potential (ADP) formula, suggested as a baseline method for abiotic resources depletion in the CML 2001 guide (Guinee 2002). It is applied to evaporative and non-evaporative uses of groundwater from over-abstracted aquifers.

A.2.4 Impact assessment, at damage level

Pfister et al. (2009)

At the damage level, the method by Pfister et al. assesses the impacts of freshwater consumptive use on human health, ecosystem quality and resources. It is an impact assessment for water use based on the Eco-indicator 99 (Goedkoop and Spriensma 2001) impact assessment method and it can be added to the hierarchist version of the Eco-indicator 99 method. Impact on human health are calculated based on Pfister et al.'s WSI which also serves as midpoint indicator. It models the cause-effect chain covering the water deprivation for agriculture use (lack of agriculture water) leading to malnutrition, translated as an impact in DALY. For ecosystem quality, net primary production (NPP) which is limited by water availability is modeled through the dependancy of vascular plant species biodiversity (VPBD) on water resource, as NPP and VPBD are assumed to be significantly correlated. Damages to resources are calculated based on WTA and evaluate the amount of water withdrawned above water availability. The original method provides characterization factors only for consumptive water use. Based on this type of water use, impacts on human health, ecosystem quality and resources are assessed. Spatial differentiation is possible at the watershed, country and supranational level. Pfister et al. (2009) provide characterization factors at the country level but also at the watershed level. The units with which the impacts are assessed are DALY, PDF·m^2·y and MJ surplus energy for human health, ecosystem quality and resources respectively. The aggregated results are expressed in Eco-indicator 99 points per kg product or m3 water use.

See Pfister et al. (2009) or "Water_Methods_Flows&CF_RESULTS" for the list of country-based characterization factors. Watershed values can be downloaded as kmz-file (for use in Google Earth) at http://www.ifu.ethz.ch/staff/stpfiste.

Motoshita et al. (2011)

Motoshita et al. propose a damage assessment model for health damages of undernourishment related to agricultural water scarcity. The modeling uses the relationship between agricultural water use, crop productivity and the undernourishment damage related to the change of food consumption. The method provides country-based characterization factors, expressed in DALY per m3 of water consumed, with the consideration of the rate of agricultural water use in each country.

Motoshita et al. (2010)

Motoshita et al. propose a damage assessment model for infectious diseases arising from domestic water consumption. Health impacts are evaluated by correlating oral intake of unsafe water with water scarcity. Indeed, the method assumes that water resource scarcity caused by water consumption will lead to a loss of access to safe water, and subsequently drinking unsafe water will result in use of infectious sources and health impairment by diseases. The method provides country-based characterization factors, expressed in DALY per m3 of domestic water consumed.

Boulay et al. (2011b)

Boulay et al. evaluate impact from water consumption on human health. Direct and indirect impacts can be obtained from a system expansion based on the water to be compensated. Indeed, the main innovation introduced by this method regarding the LCA field is the consideration of the adaptation capacity and the partition of freshwater use impacts between the impact pathways leading to human health impacts, and the impact pathways leading to compensation. Human health impacts from water deprivation for agriculture, aquaculture and for domestic uses are evaluated by the change in water availability leading to a loss of functionality for each water user. This loss of functionality is evaluated based on the quality of water used and released by a process, and to which extent the consumption and degradation of the resource will affect other users. Boulay et al. provide country and watershed-based characterization factors, expressed in DALY per m3 of water used for the impacts from malnutrition, aquaculture and domestic uses.

Hanafiah et al. (2011)

This method focuses on the effect of water consumption and global warming based on freshwater fish species loss. Calculation of characterization factors for potential freshwater fish losses from water consumption was estimated using a generic species-river discharge curve for 214 global river basins. Characterization factors were also derived for potential freshwater fish species losses per unit of greenhouse gas emission. Based on five global climate scenarios, characterization factors for 63 greenhouse gas emissions were calculated. Depending on the river considered, characterization factors for water consumption can differ up to 3 orders of magnitude. Characterization factors for greenhouse gas emissions can vary up to 5 orders of magnitude, depending on the atmospheric residence time and radiative forcing efficiency of greenhouse gas emissions. An emission of 1 ton of CO_2 emission is expected to cause an equal impact on potential fish species disappearance compared to 10-1000 m^3 of water consumption, depending on the river basin considered. This method gives the opportunity to compare the impact of water consumption with greenhouse gas emissions.

Van Zelm et al. (2011)

Van Zelm et al. propose a method to calculate the terrestrial biodiversity reduction due to lowering of the groundwater table caused by groundwater withdrawal (dQ). The cause-effect chain is based on the fact that groundwater withdrawal causes the lowering of groundwater tables, which implies ultimately the disappearance of terrestrial plant species, expressed as potentially not occurring fraction (PNOF). The different plant species are not differentiated. The model is based on a fate factor (Area*dASG/dQ, in m^2drop in the soil * msoil per m^3water/y, and therefore in years, with ASG being the average spring groundwater level), which expresses the amount of time that is needed in order to replenish the groundwater balance, multiplied by an effect factor (dPDF/dASG, in PNOF of terrestrial plant species per drop of ground water table). This method focuses on total plant species occurrence, not differentiating between the species, i.e. whether one species is reduced, and another one gets a higher occurrence.

The characterization factors express therefore the reduction of biodiversity (in PDF·m^2·y) per m3 of ground water extracted. The results are not yet published.

Maendly and Humbert (Maendly and Humbert 2009)

The method developed by Maendly and Humbert assesses the impact of dams used for hydropower generation on aquatic biodiversity. It assesses the impact as the potentially disappeared fraction (PDF) of species of an aquatic ecosystem over a certain affected area per amount of water that passes the dam per year or per yearly-generated electricity (PDF·m2·y/m^3 or PDF·m^2·y/kWh). Using the approach 'per m^3' is more in line with the traditional approach of life cycle assessment because water reported in life cycle inventories are quantified in m^3. The use of 'per kWh' allows to be directly usable with electricity output when no information is known about the amount of water turbined to produce one kWh. The use of the unit PDF·m^2·y is a commonly used unit in other impacts assessment methods, thus facilitating comparison with other ecosystem impact categories. At present, only characterization factors for hydropower dams exist. Results show that for hydroelectric dams, damage to aquatic biodiversity are of the order of 0.004 PDF·m^2·y/m3 of water turbined or 0.03 PDF·m^2·y/kWh produced for run-of-river and non-alpine dams, and of 0.001 PDF·m2·y/m^3 of water turbined or 0.0005 PDF·m^2·y/kWh produced for alpine dams. The method can be extended to other types of dams (e.g., for irrigation). The method submitted for publication contains only characterization factors for damage to aquatic ecosystems. However, the present report proposes an approach to also evaluate damage to human health from dams (see below). The proposed approach aims at quantifying the amount of DALY caused by social stress (e.g., increase in smoking, drinking or suicides) experienced by population that is displaced by a new reservoir.

Cumulative exergy demand (Bösch et al. 2007)

The cumulative exergy demand (CExD) method uses the principle of exergy, which is "the amount of work that can be obtained from a system when it is brought in equilibrium with the environmental state, the so-called dead state" (Dewulf et al. 2005).

As water represents an exergy stock in the natural environment, CExD quantifies the exergy of water that is taken away from natural ecosystems. It calculates the amount of exergy of all resources required to provide a process or product (transferred to a technical system). For water, only chemical exergy (for all material resources) and potential exergy (potential energy of water in hydropower generation) are of relevance. Water types considered are freshwater resources and turbined water. Cooling water is not mentioned. As the exergy approach requires a baseline, seawater is used as reference species for water and thus has no exergy. However, it does not consider further aspects of resource scarcity or quality. Spatial differentiation is theoretically possible. This method is not well suited for assessing water use as it does not allow for easy spatial differentiation as well as does not address impacts on human health and ecosystems.

A.2.5 Water indexes

Indexes are mostly developed and given at the country level. However, if the information are available, they can be adapted at the watershed level.

Falkenmark et al. (1989): water resource per capita (WRPC)

The Falkenmark et al's (1989) water index measures per capita water availability and considers that a per capita water availability of between 1'000 and 1'600 m^3 indicates water scarcity, a per capita water availability between 500 and 1'000 m^3 indicates chronic water scarcity, while a per capita water availability below 500 m^3 indicates a country or region beyond the 'water barrier' of manageable capability.

Ohlsson (2000): social water scarcity index (SWSI)

The social water scarcity index is constructed by dividing the widely used first-generation water scarcity index of per capita availability of renewable fresh water (Falkenmark index) by the human development index (HDI) as shown in Equation A.3. It is useful to highlight the importance of a society's social adaptive capacity facing the challenges of water scarcity. For example, according to the SWSI, Israel is "merely" water-scarce due to its high level of social adaptive capacity whereas it was classified as "beyond the barrier" according to Falkenmark index.

$$SWSI = \frac{WRPC}{HDI}$$

Equation A.3: Ohlsson's SWSI

Gleick (1996): basic water requirement (BWR)

Gleick developed an indicator that shifted the focus from measuring water availability to measuring some aspects of water use. In this assessment, he quantifies a basic water requirement (BWR) for drinking, cooking, bathing, and sanitation and hygiene at 50 liters per person per day and then presents estimates of the population by country, without access to this BWR. Limitations of this indicator include highly inadequate country data on domestic water use and an inability to distinguish regional water problems hidden by country-level aggregation. This measure, like most single-factor indicators, includes no information on water quality issues.

If BWR<50 L/person/d, large-scale human misery and suffering will continue and grow in the future, contributing to the risk of social and military conflict.

Smakhtin et al. (2004): environmental water requirements (EWR)

Smakhtin makes a first attempt at estimating the environmental water requirements (EWR) for all world river basins. They then combine EWR with the water resources available and their use (i.e. water use per resource (WUPR) defined per river basin) by subtracting EWR from the available water resources (WR) to derive a water scarcity indicator as shown in Equation A.4.

$$WSI = \frac{WU}{WR - EWR}$$

Equation A.4: Smakhin et al's WSI

Alcamo et al. (2003): criticality ratio

The criticality ratio (CR) is the ratio of water use to water availability in a watershed or country calculated based on Equation A.5. Water availability refers to the renewable water resources generated inside the entity of interest (river discharge and the groundwater recharge). Values for this ratio range from near zero in sparsely-inhabited watersheds (where water use is small compared to water availability) to greater than one in arid watersheds (where water use is computed to exceed its availability).

$$CRi = \frac{\sum_j WU}{WAi}$$

Equation A.5: Alcamo et al.'s criticality ratio

CRi is the criticality ratio in watershed i and user groups j are industry, agriculture, and households.

Alcamo et al. (2003): criticality index (CI)

The criticality index combines two factors – the criticality ratio and the water availability per capita – into a single indicator of water vulnerability in a watershed and country as shown in Table A.4.

Table A.4: Alcamo et al.'s criticality index

	criticality ratio (use/availability)			
Per capita water availability [m3/(cap.yr)]	< 0.4	0.4 - 0.6	0.6 - 0.8	> 0.8
< 2,000	2	3	4	4
2,000-10,000	1	2	3	4
> 10,000	1	1	2	4

1: water surplus; 2: marginally vulnerable; 3: water scarcity; 4. severe water scarcity

Raskin et al. (1997): water resources vulnerability index (WRVI)

The water resources vulnerability index (WRVI) put forward by Raskin et al. is made up of three sub-indices as shown in Figure A.1: (i) a use-to-resource ratio sub-index, which measures the average water-related scarcity that both ecological and socioeconomic systems place on a country's usable resources (similar to the criticality ratio); (ii) a coping capacity sub-index, which measures the economic and institutional ability of countries to endure water-related scarcity, and (iii) a reliability sub-index, composed of three factors that examine different aspects of uncertainty of water supply (storage-to-flow indicator, coefficient of variation of precipitation indicator and import dependence indicator). Each of these indicators and sub-indices is divided into four classes in which values denote a level of scarcity: no scarcity, low scarcity, scarcity, and high scarcity. Indicator scores are then averaged to produce the WRVI. A variant of the WRVI relies not on averages but on the highest value of any of the three sub-indices. This produces a stronger signal of vulnerability, reasoning that if a country is vulnerable in any one of these areas, it is considered "vulnerable".

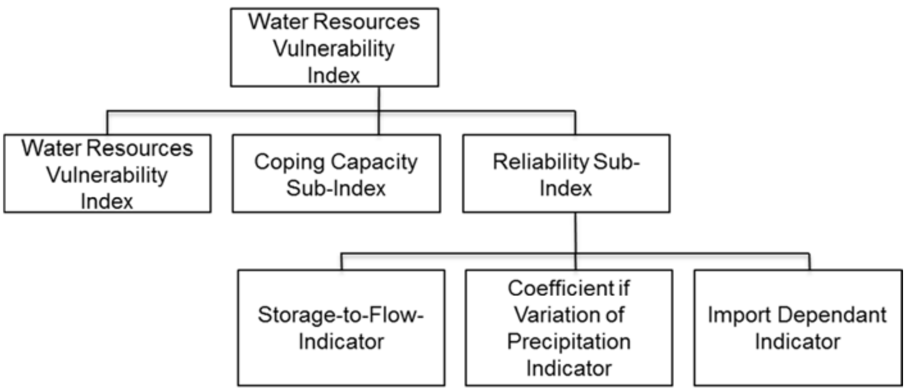


Figure A.1: Water Resources Vulnerability Index

Seckler et al. (1999): index of relative water scarcity (IRWS)

The index of relative water scarcity (IRWS) is based on two criteria: (i) percentage increase in water "withdrawals" over the 1990-2025 period; (ii) water withdrawals in 2025 as a percentage of the annual water resources (AWR) of the country (similar to criticality ratio). Thus, the IRWS measures both how fast a country's water use is growing and how close it is to its total available water limit. The IRWS analysis takes into account the share of the renewable water resources available for human needs (accounting for existing water infrastructure) and the primary water supply. Its analysis of demands is based on consumptive use (evapotranspiration) and the remainder of water withdrawn is accounted for as return flows.

Pfister et al. (2009)

Same as Pfister et al.'s midpoint.

Swiss Ecological Scarcity (Frischknecht et al. 2006)

Same as Frischknecht et al.'s midpoint.

Veolia (2014)

Same as Bayart et al.'s midpoint.

Hoekstra et al. (2011)

Same as Hoekstra et al.'s midpoint.

Boulay et al. (2011b)

Same as Boulay et al.'s midpoint.

Sullivan et al. (2002): water poverty index (WPI)

The water poverty index (WPI) was developed by the Centre for Ecology and Hydrology in the United Kingdom and aims to reflect both the physical availability of water and the degree to which human populations are served by that water, subject to constraints imposed by the maintenance of ecological integrity. The WPI can be constrfucted in a number of ways. The two most comprehensive are either a conventional method, made up of relevant components that are weighted by importance. This is a relative method that would allow the comparison of the index from one year to the next, thereby measuring progress over time. The latter is an absolute method that would consider how much water provision and use in a given country or a region deviate from pre-determined standards (such as discussed in Gleick P., The World's Water, The biennial report on freshwater resources 2002-2003) of ecosystem and human health, economic welfare, and community well-being. However, one obstacle to formulate of such an absolute index is expert disagreement over where and how much standards should be set.

The WPI can also be constructed as either a time-analysis function or as a part of a larger measure of general environmental sustainability.

Döll P (2009)

The water scarcity indicator developed by Döll is the ratio of consumptive water use to statistical low flow Q90 in each grid cell in 0.5*0.5° grid cells on a monthly base. This index is then used in addition to an indicator for dependence of water supply on groundwater and the Human Development Index to form a sensitivity index. Global maps of vulnerability to the impact of decreased groundwater recharge in the 2050s are derived combining this sensitivity index with per cent groundwater recharge decrease.

A.3 General criteria adapted from the ILCD handbook (JRC-IES 2011)

Table A.5: General criteria adapted from the ILCD handbook and used in the review of methods addressing water

Criteria	Sub-Criteria
Completeness of scope	The characterization model is adaptable to spatial and temporal explicit evaluation
	Global geographical validity
	The method is compatible with, or developed specifically for, the comparative assessment scope of LCA (e.g., factors do not include security factors / precautionary principle)
	When empirical data is used, double counting is avoided
Overall evaluation	
Overall relevance	Described in Table 2.1
Overall evaluation	
Scientific robustness and certainty	The critical part of the model including the input data have been peer reviewed (journal, panel, book, etc.)
	The model reflects the latest knowledge for the cause-effect chain (the critical links are covered)
	The model including the underlying data have a good potential for being consistently improved and further developed including regarding geographical/emission situation and temporal differentiation
	Indicators can be confirmed and verified against monitoring data, if available
	Uncertainty estimates of the indicators are provided, motivated and reported in statistical terms
	Scenario and model uncertainty as well as substance data and parameter uncertainty are taken into account
Overall evaluation: the category indicator and characterization models fulfill the requirements of being science based	
Documentation and Transparency and Reproducibility	The model documentation is published and accessible (incl. description of the mechanism, the model, temporal and spatial scale, etc.)?
	The set of characterization factors/models is published and accessible
	The input data are published and accessible
	The characterization model is published and accessible
	Ability for third parties to freely generate additional, consistent factors and to further develop models e.g. incorporating further geographical/emission situation, temporal and speciation differentiation
	Value choices are explicitly stated
Overall evaluation	
Applicability	The characterisation factors are straightforward to apply for general LCA practitioners and in most market-relevant LCA software tools
	Life cycle inventory figures for the distinguished emission compartments or resource types can be made directly available by producing industry
	Unit comparable with other impact categories
Overall evaluation	
Overall evaluation of science based criteria	
Stakeholder acceptance criteria	The indicator is easily understood
	There is an authoritative body behind the general model principles like the IPCC model (consensus/international endorsement)
	The principles of the model are easily understood by non-LCIA experts and preferably also by the general public
	The covered elementary flows and impact models do not inappropriately favour or disfavour specific industries, processes, or products
Overall evaluation of stakeholders acceptance criteria	
Final recommendation	

Appendix B

Spatial analysis of toxic emissions in LCA: A sub-continental nested USEtox model with freshwater archetypes

B.1 Landscape data for USEtox parameterization

Table B.1: USEtox landscape parameters for the 17 sub-continental and the 8 zone continental resolution (the title terminology is the same as in the USEtox tool)

ID #	Name	Continental scale														
		Land area	Sea Area	Areafrac	Areafrac	Areafrac	Areafrac	Temp	Wind speed	Rain rate	Depth	River-Flow	Fraction	Frac-tion	Soil erosion	Water resi-dence time to the sea
		land	Sea	fresh-water	nat soil	agr soil	other soil				freshwa-ter	reg-cont	run off	infiltra-tion		conti-nent
		km^2	km^2	[-]	[-]	[-]	[-]	^oC	$m.s^{-1}$	$mm.y^{-1}$	M	[-]	[-]	[-]	$mm.y^{-1}$	day
W1	West Asia	1.7E+07	7.4E+05	1.7E-02	0.88	1.0E-01	1.0E-20	1.2E+01	7.3E+00	2.2E+02	1.3E+01	0.0E+00	4.6E-01	2.7E-01	3.0E-02	1300
W2	Indo-china	3.3E+06	2.2E+06	3.6E-02	0.86	1.0E-01	1.0E-20	1.2E+01	4.9E+00	2.4E+03	1.3E+01	0.0E+00	2.7E-01	2.7E-01	3.0E-02	260
W3	N. Aus-tralia	6.6E+06	1.6E+06	9.9E-03	0.89	1.0E-01	1.0E-20	1.2E+01	4.4E+00	1.5E+03	3.0E+00	0.0E+00	2.0E-01	2.7E-01	3.0E-02	28
W4	S. Aus-tralia+	1.5E+06	6.4E+05	1.2E-02	0.89	1.0E-01	1.0E-20	1.2E+01	1.0E+01	5.1E+02	3.0E+00	0.0E+00	1.0E-01	2.7E-01	3.0E-02	98
W5	S. Africa	1.0E+07	6.2E+05	2.2E-02	0.88	1.0E-01	1.0E-20	1.2E+01	3.5E+00	1.0E+03	4.6E+01	0.0E+00	1.9E-01	2.7E-01	3.0E-02	1400
W6	N. Afri-ca	2.4E+07	9.7E+05	1.9E-02	0.88	1.0E-01	1.0E-20	1.2E+01	5.1E+00	5.1E+02	4.6E+01	0.0E+00	1.8E-01	2.7E-01	3.0E-02	2400
W7	Argen-tina+	4.2E+06	1.1E+06	1.5E-02	0.89	1.0E-01	1.0E-20	1.2E+01	7.4E+00	7.0E+02	8.0E+00	0.0E+00	3.0E-01	2.7E-01	3.0E-02	240
W8	Brazil+	1.1E+07	5.8E+05	8.3E-03	0.89	1.0E-01	1.0E-20	1.2E+01	4.9E+00	1.8E+03	8.0E+00	0.0E+00	4.0E-01	2.7E-01	3.0E-02	54
W9	Central America	5.9E+06	1.3E+06	3.6E-02	0.86	1.0E-01	1.0E-20	1.2E+01	7.3E+00	2.0E+03	2.0E+01	0.0E+00	3.8E-01	2.7E-01	3.0E-02	480
W10	US+	1.4E+07	1.8E+06	3.4E-02	0.87	1.0E-01	1.0E-20	1.2E+01	7.0E+00	7.1E+02	2.0E+01	0.0E+00	3.7E-01	2.7E-01	3.0E-02	1300
W12	N. Eur. + N. Can-ada	1.8E+07	5.6E+06	4.9E-02	0.85	1.0E-01	1.0E-20	1.2E+01	8.8E+00	4.9E+02	1.7E+01	0.0E+00	3.6E-01	2.7E-01	3.0E-02	2100
W13	Europe+	8.6E+06	1.7E+06	1.6E-02	0.88	1.0E-01	1.0E-20	1.2E+01	6.8E+00	5.5E+02	1.5E+01	0.0E+00	1.7E-01	2.7E-01	3.0E-02	610
W14	East Indies	2.0E+06	1.4E+06	3.0E-02	0.87	1.0E-01	1.0E-20	1.2E+01	8.0E+00	1.5E+03	3.0E+00	0.0E+00	2.0E-01	2.7E-01	3.0E-02	80
IND	India	4.6E+06	4.6E+05	4.2E-02	0.86	1.0E-01	1.0E-20	1.2E+01	5.0E+00	1.2E+03	1.3E+01	0.0E+00	2.7E-01	2.7E-01	3.0E-02	580
CHI	China	6.4E+06	8.4E+05	4.6E-02	0.85	1.0E-01	1.0E-20	1.2E+01	6.1E+00	1.2E+03	1.3E+01	0.0E+00	2.7E-01	2.7E-01	3.0E-02	620
JAP	Japan	6.0E+05	4.2E+05	4.4E-02	0.86	1.0E-01	1.0E-20	1.2E+01	8.3E+00	2.4E+03	1.3E+01	0.0E+00	2.7E-01	2.7E-01	3.0E-02	310

North America	**North America**	1.4E+07	1.8E+06	3.4E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	7.0E+00	7.1E+02	2.0E+01	0.0E+00	3.7E-01	2.7E-01	3.0E-02	1.3E+03
Latin America	**Latin America**	2.1E+07	3.0E+06	1.8E-02	8.8E-01	1.0E-01	1.0E-20	1.2E+01	6.5E+00	1.6E+03	1.5E+01	0.0E+00	3.8E-01	2.7E-01	3.0E-02	2.3E+02
Europe	**Europe**	8.6E+06	1.7E+06	1.6E-02	8.8E-01	1.0E-01	1.0E-20	1.2E+01	6.8E+00	5.5E+02	1.5E+01	0.0E+00	1.7E-01	2.7E-01	3.0E-02	6.1E+02
Africa+Middle East	**Africa+Middle East**	3.4E+07	1.6E+06	2.0E-02	8.8E-01	1.0E-01	1.0E-20	1.2E+01	4.3E+00	6.6E+02	4.6E+01	0.0E+00	1.8E-01	2.7E-01	3.0E-02	1.9E+03
Central Asia	**Central Asia**	1.7E+07	7.4E+05	1.7E-02	8.8E-01	1.0E-01	1.0E-20	1.2E+01	7.3E+00	2.2E+02	1.3E+01	0.0E+00	4.6E-01	2.7E-01	3.0E-02	1.3E+03
South east Asia	**South-east Asia**	1.7E+07	5.3E+06	4.1E-02	8.6E-01	1.0E-01	1.0E-20	1.2E+01	6.5E+00	1.5E+03	1.2E+01	0.0E+00	2.6E-01	2.7E-01	3.0E-02	4.3E+02
Northern regions	**Northern regions**	1.8E+07	5.6E+06	4.9E-02	8.5E-01	1.0E-01	1.0E-20	1.2E+01	8.8E+00	4.9E+02	1.7E+01	0.0E+00	3.6E-01	2.7E-01	3.0E-02	2.1E+03
Oceania	**Oceania**	8.1E+06	2.2E+06	1.0E-02	8.9E-01	1.0E-01	1.0E-20	1.2E+01	7.4E+00	1.3E+03	3.0E+00	0.0E+00	1.9E-01	2.7E-01	3.0E-02	3.4E+01
Source		Based on GIS computation			Calculated based on freshwater, agr soil and other soil ratios	Set at 0.1	Same as default USEtox landscape		Based on GEOS-Chem wind speeds	Same as default USEtox landscape	Same as default USEtox landscape	Same as default USEtox landscape	Based on GIS computation		Same as default USEtox landscape	Recalculated based on model algorithm

Table B.2: USEtox landscape parameters for the 17 sub-continental and the 8 zone continental resolution (the title terminology is the same as in the USEtox tool) (continued)

ID #	Name	Global scale													
		Area	Area	Areafrac	Areafrac	Areafrac	Areafrac	Temp	Wind speed	Rain rate	Depth	River-Flow	Fraction	Fraction	Soil erosion
		land	sea	freshwa-ter	nat soil	agr soil	other soil				freshwa-ter	cont-reg	run off	infiltration	
		km^2	km^2	[-]	[-]	[-]	[-]	°C	$m.s^{-1}$	$mm.y^{-1}$	m	[-]	[-]	[-]	$mm.y^{-1}$
W1	West Asia	1.2E+08	3.6E+08	3.0E-02	8.7E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W2	Indochina	1.4E+08	3.6E+08	2.7E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W3	N. Australia	1.3E+08	3.6E+08	2.8E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W4	S. Australia+	1.4E+08	3.6E+08	2.7E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W5	S. Africa	1.3E+08	3.6E+08	2.8E-02	8.7E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W6	N. Africa	1.1E+08	3.6E+08	3.2E-02	8.7E-01	1.0E-01	1.2E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W7	Argentina+	1.3E+08	3.6E+08	2.7E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W8	Brazil+	1.3E+08	3.6E+08	2.8E-02	8.7E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W9	Central Ameri-ca	1.3E+08	3.6E+08	2.7E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W10	US+	1.2E+08	3.6E+08	2.9E-02	8.7E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W12	N. Eur. + N. Canada	1.2E+08	3.6E+08	3.0E-02	8.8E-01	1.0E-01	1.2E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W13	Europe+	1.3E+08	3.6E+08	2.8E-02	8.7E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
W14	East Indies	1.4E+08	3.6E+08	2.7E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
IND	India	1.3E+08	3.6E+08	2.7E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
CHI	China	1.3E+08	3.6E+08	2.8E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
JAP	Japan	1.4E+08	3.6E+08	2.6E-02	8.7E-01	1.0E-01	1.0E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
North America	North America	1.2E+08	3.6E+08	2.9E-02	8.7E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
Latin America	Latin America	1.2E+08	3.6E+08	3.1E-02	8.7E-01	1.0E-01	1.2E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
Europe	Europe	1.3E+08	3.6E+08	2.8E-02	8.7E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
Afri-ca+Middl e East	Africa+Middle East	1.0E+08	3.6E+08	3.5E-02	8.7E-01	1.0E-01	1.3E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
Central	Central Asia	1.2E+08	3.6E+08	3.0E-02	8.7E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02

Asia															
Southeast Asia	**Southeast Asia**	1.2E+08	3.6E+08	3.0E-02	8.8E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
Northern regions	**Northern regions**	1.2E+08	3.6E+08	3.0E-02	8.8E-01	1.0E-01	1.2E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
Oceania	**Oceania**	1.3E+08	3.6E+08	2.8E-02	8.7E-01	1.0E-01	1.1E-20	1.2E+01	3.0E+00	7.0E+02	2.5E+00	0.0E+00	2.5E-01	2.5E-01	3.0E-02
Source		Recalculated based on continental data						Same as default USEtox landscape							

Table B.3: USEtox landscape parameters for the 17 sub-continental and the 8 zone continental resolution (the title terminology is the same as in the USEtox tool) (continued)

ID #	Name	Urban scale			Human Population			Exposure	
		Area	Areafrac	Areafrac	Human pop	Human pop	Human pop	Human breathing rate	Water ingestion
		land	nat soil	other soil	World	Continent	urban	world + cont + urban	world + cont
		km^2	[-]	[-]	[-]	[-]	[-]	m^3/(person*day)	l/(person*day)
W1	West Asia	240	0.67	0.33	6.58E+09	2.35E+08	1.47E+06	13	1.4
W2	Indochina	240	0.67	0.33	6.35E+09	4.65E+08	1.30E+06	13	1.4
W3	N. Australia	240	0.67	0.33	6.82E+09	3.20E+06	8.24E+05	13	1.4
W4	S. Australia+	240	0.67	0.33	6.80E+09	2.12E+07	1.03E+06	13	1.4
W5	S. Africa	240	0.67	0.33	6.50E+09	3.24E+08	1.25E+06	13	1.4
W6	N. Africa	240	0.67	0.33	6.03E+09	7.89E+08	2.30E+06	13	1.4
W7	Argentina+	240	0.67	0.33	6.75E+09	6.67E+07	2.89E+06	13	1.4
W8	Brazil+	240	0.67	0.33	6.58E+09	2.42E+08	2.62E+06	13	1.4
W9	Central America	240	0.67	0.33	6.51E+09	3.05E+08	2.76E+06	13	1.4
W10	US+	240	0.67	0.33	6.49E+09	3.28E+08	1.32E+06	13	1.4
W12	N. Eur. + N. Canada	240	0.67	0.33	6.80E+09	1.67E+07	6.56E+05	13	1.4
W13	Europe+	240	0.67	0.33	6.06E+09	7.59E+08	1.41E+06	13	1.4
W14	East Indies	240	0.67	0.33	6.61E+09	2.07E+08	1.30E+06	13	1.4
IND	India	240	0.67	0.33	5.25E+09	1.57E+09	1.76E+06	13	1.4
CHI	China	240	0.67	0.33	5.49E+09	1.33E+09	1.47E+06	13	1.4
JAP	Japan	240	0.67	0.33	6.67E+09	1.51E+08	4.56E+06	13	1.4
North America	North America	240	0.67	0.33	6.5E+09	3.3E+08	1.3E+06	13	1.4
Latin America	Latin America	240	0.67	0.33	6.2E+09	6.1E+08	8.3E+06	13	1.4
Europe	Europe	240	0.67	0.33	6.1E+09	7.6E+08	1.4E+06	13	1.4
Africa+Middle East	Africa+Middle East	240	0.67	0.33	5.7E+09	1.1E+09	3.6E+06	13	1.4
Central Asia	Central Asia	240	0.67	0.33	6.6E+09	2.4E+08	1.5E+06	13	1.4
Southeast Asia	Southeast Asia	240	0.67	0.33	3.1E+09	3.7E+09	1.0E+07	13	1.4
Northern re-gions	Northern regions	240	0.67	0.33	6.8E+09	1.7E+07	6.6E+05	13	1.4
Oceania	Oceania	240	0.67	0.33	6.8E+09	2.4E+07	1.9E+06	13	1.4
Source		Same as default USEtox landscape			Recalculated based on continental data			Same as default USEtox landscape	

Table B.4: USEtox landscape parameters for the 17 sub-continental and the 8 zone continental resolution (the title terminology is the same as in the USEtox tool) (continued)

ID #	Name	Production-based intake rates											
		Exposed produce	Exposed produce	Unexposed produce	Unexposed produce	Meat	Meat	Dairy products	Dairy products	Fish freshwater	Fish freshwater	Fish coastal marine freshwater	Fish coastal marine freshwater
		world	continent	world	continent	World	continent	World	continent	World	continent	world	continent
		kg/(day*capita)	kg/(day*capita)	kg/(day*capita)	kg/(day*capita)	kg/(day*capita)	kg/(day*capita)	kg/(day*capita)	kg/(day*capita)	kg/(day*capita)	kg/(day*capita)	kg/(day*capita)	kg/(day*capita)
W1	**West Asia**	2.13	1.71	0.37	0.33	0.10	0.08	0.24	0.26	0.01	0.011	0.04	0.05
W2	**Indochina**	2.08	2.57	0.38	0.22	0.10	0.05	0.25	0.01	0.01	0.008	0.03	0.06
W3	**N. Australia**	2.11	10.45	0.37	0.18	0.09	0.51	0.24	1.39	0.01	0.006	0.03	6.65
W4	**S. Australia+**	2.10	9.02	0.37	0.21	0.09	0.60	0.23	2.84	0.01	0.004	0.03	0.61
W5	**S. Africa**	2.17	1.03	0.37	0.44	0.10	0.03	0.25	0.06	0.01	0.006	0.04	0.03
W6	**N. Africa**	2.27	0.98	0.36	0.46	0.10	0.04	0.26	0.10	0.01	0.006	0.04	0.01
W7	**Argentina+**	2.09	4.92	0.37	0.46	0.09	0.23	0.23	0.57	0.01	0.002	0.03	0.31
W8	**Brazil+**	1.97	6.08	0.37	0.36	0.09	0.20	0.24	0.28	0.01	0.004	0.04	0.05
W9	**Central America**	2.09	2.62	0.38	0.10	0.10	0.09	0.24	0.20	0.01	0.003	0.04	0.04
W10	**US+**	1.98	4.82	0.37	0.42	0.08	0.35	0.21	0.69	0.01	0.003	0.04	0.04
W12	**N. Eur. + N. Canada**	2.12	1.74	0.37	0.47	0.09	0.15	0.24	0.75	0.01	0.008	0.03	1.43
W13	**Europe+**	2.06	2.57	0.27	1.12	0.08	0.19	0.17	0.80	0.01	0.003	0.04	0.02
W14	**East Indies**	2.15	1.21	0.38	0.16	0.10	0.02	0.24	0.01	0.01	0.013	0.03	0.13
IND	**India**	2.30	1.52	0.46	0.07	0.12	0.01	0.25	0.20	0.01	0.008	0.05	0.00
CHI	**China**	2.17	1.90	0.37	0.38	0.09	0.12	0.29	0.03	0.01	0.029	0.04	0.01
JAP	**Japan**	2.14	1.16	0.37	0.21	0.10	0.09	0.24	0.19	0.01	0.027	0.04	0.06
North America	**North America**	1.98	4.82	0.37	0.42	0.08	0.35	0.21	0.69	0.012	0.003	0.04	0.04
Latin America	**Latin America**	1.91	4.24	0.38	0.24	0.09	0.15	0.23	0.27	0.012	0.003	0.03	0.08
Europe	**Europe**	2.06	2.57	0.27	1.12	0.08	0.19	0.17	0.80	0.012	0.003	0.04	0.02
Africa+Middle	**Africa+Middle**	2.34	0.99	0.35	0.45	0.11	0.03	0.27	0.09	0.012	0.006	0.04	0.02

East	**East**												
Central Asia	**Central Asia**	2.13	1.71	0.37	0.33	0.10	0.08	0.24	0.26	0.011	0.011	0.04	0.05
Southeast Asia	**Southeast Asia**	2.56	1.76	0.56	0.21	0.14	0.06	0.40	0.10	0.005	0.017	0.05	0.02
Northern regions	**Northern regions**	2.12	1.74	0.37	0.47	0.09	0.15	0.24	0.75	0.011	0.008	0.03	1.43
Oceania	**Oceania**	2.09	9.21	0.37	0.21	0.09	0.59	0.23	2.65	0.011	0.005	0.03	1.40
		Recalcu-lated based on conti-nental data	FAO pro-duction data from 2001	Recalcu-lated based on continen-tal data	FAO pro-duction data from 2001	Recalcu-lated based on continen-tal data	FAO pro-duction data from 2001	Recalculated based on continental data	FAO pro-duction data from 2001	Recalcu-lated based on continen-tal data	FAO FishSTAT	Recalcu-lated based on continen-tal data	FAO FishSTAT

B.2 Selected set of pollutants

Table B.5: Chemical data of the set of 36 pollutants of the OMNIITOX set (Margni et al. 2002)

Name	CAS	Degradable with H and Kow = 0; Non-degradable or specification of partitioning coefficients = 1	Molecular Mass (g/mole)	Henry's Constant (Pa m3 mol-1) or Kaw	Log Kow	tropospheric degradation half life (hours)	water-column degradation half life (hours)	Soil surface layer degradation half life (hours)	Source	sediment degradation half life (hours)	vegetation degradation half life (hours)	Soil root zone degradation half life (hours)	Soil vadose layer degradation half life (hours)	Source
Tetrachloroethylene	**127-18-4**	0	166	1.77E+03	2.88	5.50E+02	1.75E+03	1.70E+03	OM-NITOX	5.50E+03	1.70E+03	1.70E+03	1.70E+03	MACKAY
Carbon tetrachloride (CCl4)	**56-23-5**	0	154	2.76E+03	2.64	1.70E+04	1.70E+03	6.04E+03	OM-NITOX	1.70E+04	5.50E+03	5.50E+03	5.50E+03	MACKAY
1,3-butadiene	**106-99-0**	0	54	7.36E+03	1.99	1.70E+04	1.70E+02	5.50E+02	OM-NITOX	1.70E+03	5.50E+02	5.50E+02	5.50E+02	MACKAY
Methomyl	**16752-77-5**	0	162	1.84E-05	0.60	5.80E+01	5.52E+03	5.03E+02	OM-NITOX	5.04E+02	5.04E+02	5.04E+02	5.04E+02	USES
Acephate	**30560-19-1**	0	183	5.01E-08	-0.85	3.44E+01	1.28E+03	5.29E+01	OM-NITOX	5.28E+01	5.28E+01	5.28E+01	5.28E+01	USES
Formaldehyde	**50-00-0**	0	30	3.37E-02	0.35	3.63E+00	9.58E+01	5.50E+01	OM-NITOX	3.84E+02	9.60E+01	9.60E+01	9.60E+01	HOWARD
PCBs	**1336-36-3**	0	292	4.15E+01	7.10	4.73E+02	3.38E+02	9.00E+02	OM-NITOX	1.34E+03	3.36E+02	3.36E+02	3.36E+02	Estimate
Di(n-octyl) phthalate	**117-84-0**	0	391	2.57E-01	8.10	1.87E+01	3.36E+02	3.37E+02	OM-NITOX	6.54E+03	3.36E+02	3.36E+02	3.36E+02	USES
Hexabromobenzene	**87-82-1**	0	551	2.81E+00	6.07	3.36E+04	1.44E+03	1.44E+03	OM-NITOX	5.76E+03	1.44E+03	1.44E+03	1.44E+03	Estimate
Cypermethrin	**52315-07-8**	0	416	1.92E-02	6.60	1.80E+01	1.20E+02	1.25E+03	OM-NITOX	1.25E+03	1.25E+03	1.25E+03	1.25E+03	USES
Mirex	**2385-85-5**	0	546	8.11E+01	6.90	1.70E+02	1.70E+02	5.50E+04	OM-NITOX	5.50E+04	5.50E+04	5.50E+04	5.50E+04	MACKAY
Trifluralin	**1582-09-8**	0	336	1.03E+01	5.34	1.70E+02	1.70E+03	1.70E+03	OM-NITOX	5.50E+03	1.70E+03	1.70E+03	1.70E+03	MACKAY
Dicofol	**115-32-2**	0	370	2.42E-02	5.02	1.12E+02	9.00E+02	1.46E+03	OM-NITOX	3.84E+02	1.46E+03	1.46E+03	1.46E+03	CALTOX
1,4-dichlorobenzene	**106-46-7**	0	147	2.41E+02	3.40	5.50E+02	1.70E+03	5.50E+03	OM-NITOX	1.70E+04	5.50E+03	5.50E+03	5.50E+03	MACKAY
Aldrin	**309-00-2**	0	365	4.40E+00	3.01	4.99E+00	1.75E+04	1.70E+04	OM-NITOX	5.50E+04	1.70E+04	1.70E+04	1.70E+04	MACKAY

Name	CAS	Degradable with H and Kow = 0; Non-degradable or specification of partitioning coefficients = 1	Molecular Mass (g/mole)	Henry's Constant (Pa m3 mol-1) or Kaw	Log Kow	tropospheric degradation half life (hours)	water-column degradation half life (hours)	Soil surface layer degradation half life (hours)	Source	sediment degradation half life (hours)	vegetation degradation half life (hours)	Soil root zone degradation half life (hours)	Soil vadose layer degradation half life (hours)	Source
1,1,2,2-Tetrachloroethane	**79-34-5**	0	168	3.67E+01	2.39	1.70E+04	1.70E+03	5.50E+03	OM-NITOX	1.70E+04	5.50E+03	5.50E+03	5.50E+03	MACKAY
Captan	**133-06-2**	0	301	6.48E-04	2.30	1.70E+01	1.70E+01	5.50E+02	OM-NITOX	5.50E+02	5.50E+02	5.50E+02	5.50E+02	MACKAY
Pronamide	**23950-58-5**	0	256	9.77E-04	3.43	2.91E+01	9.77E+02	1.93E+03	OM-NITOX	1.80E+02	1.93E+03	1.93E+03	1.93E+03	CALTOX
Anthracene	**120-12-7**	0	178	5.56E+00	4.54	5.50E+01	5.50E+02	5.50E+03	OM-NITOX	1.70E+04	5.50E+03	5.50E+03	5.50E+03	MACKAY
Gamma-HCH (lindane)	**58-89-9**	0	291	5.14E-01	3.70	1.04E+03	1.70E+04	1.70E+04	OM-NITOX	5.50E+04	1.70E+04	1.70E+04	1.70E+04	MACKAY
Dimethylphthalate (DMP)	**131-11-3**	0	194	1.05E-02	2.12	1.70E+02	1.70E+02	5.50E+02	OM-NITOX	1.70E+03	5.50E+02	5.50E+02	5.50E+02	MACKAY
Methanol	**67-56-1**	0	32	4.55E-01	-0.77	3.92E+02	5.50E+01	5.50E+01	OM-NITOX	7.20E+01	9.60E+01	9.60E+01	9.60E+01	HOWARD
1,2-Dichloroethane	**107-06-2**	0	99	1.18E+02	1.48	1.70E+03	1.70E+03	5.50E+03	OM-NITOX	1.70E+04	5.50E+03	5.50E+03	5.50E+03	MACKAY
Ethyl acetate	**141-78-6**	0	88	1.34E+01	0.73	1.94E+02	9.58E+01	1.70E+02	OM-NITOX	3.84E+02	9.60E+01	9.60E+01	9.60E+01	HOWARD
N-Nitrosodiethylamine	**55-18-5**	0	102	3.63E-01	0.48	6.00E+00	6.00E+00	1.70E+03	OM-NITOX	2.40E+01	2.40E+03	2.40E+03	2.40E+03	HOWARD
Thiram	**137-26-8**	0	240	3.04E-02	1.73	1.70E+02	1.70E+02	5.50E+02	OM-NITOX	1.70E+03	5.50E+02	5.50E+02	5.50E+02	MACKAY
Propoxur	**114-26-1**	0	209	1.43E-04	1.50	5.00E+00	5.50E+02	5.50E+02	OM-NITOX	1.70E+03	5.50E+02	5.50E+02	5.50E+02	MACKAY
Folpet	**133-07-3**	0	297	7.66E-03	2.85	2.45E+01	1.38E+04	1.38E+04	OM-NITOX	1.38E+04	1.38E+04	1.38E+04	1.38E+04	USES
Benomyl	**17804-35-2**	0	290	4.93E-07	2.30	4.99E+00	1.70E+02	1.70E+03	OM-NITOX	5.50E+03	1.70E+03	1.70E+03	1.70E+03	MACKAY
Hexachlorobutadiene	**87-68-3**	0	261	1.03E+03	4.78	1.28E+04	1.75E+03	1.70E+03	OM-NITOX	1.70E+03	1.70E+03	1.70E+03	1.70E+03	USES
Hexachlorocyclopentadiene	**77-47-4**	0	273	2.70E+03	5.04	9.77E+02	8.63E+01	4.20E+02	OM-NITOX	1.68E+03	4.20E+02	4.20E+02	4.20E+02	HOWARD
Heptachlor epoxide	**1024-57-3**	0	389	2.10E+00	4.98	7.43E+01	7.03E+03	7.03E+03	OM-NITOX	9.60E+01	7.02E+03	7.02E+03	7.02E+03	HOWARD
Hexachlorobenzene	**118-74-1**	0	285	1.70E+02	5.50	7.35E+03	5.50E+04	5.50E+04	OM-	5.50E+04	5.50E+04	5.50E+04	5.50E+04	MACKAY

Name	CAS	Degradable with H and Kow = 0; Non-degradable or specification of partitioning coefficients = 1	Molecular Mass (g/mole)	Henry's Constant (Pa m3 mol-1) or Kaw	Log Kow	tropospheric degradation half life (hours)	water-column degradation half life (hours)	Soil surface layer degradation half life (hours)	Source	sediment degradation half life (hours)	vegetation degradation half life (hours)	Soil root zone degradation half life (hours)	Soil vadose layer degradation half life (hours)	Source
									NITOX					
Heptachlor	**76-44-8**	0	373	2.94E+01	5.27	5.50E+01	5.50E+02	1.70E+03	OM-NITOX	5.50E+03	1.70E+03	1.70E+03	1.70E+03	MACKAY
Nitrobenzene	**98-95-3**	0	123	2.4	1.85	4.94E+00	1.75E+03	1.23E+03	OM-NITOX	5.50E+03	1.70E+03	1.70E+03	1.70E+03	MACKAY
Endosulfan	**115-29-7**	0	407	6.5	3.83	3.85E+01	1.13E+02	2.11E+02	OM-NITOX	7.68E+02	1.11E+02	1.11E+02	1.11E+02	HOWARD

Table B.6: Chemical data of the set of 36 pollutants of the OMNIITOX set (Margni et al. 2002)

Name	PKa	BCF (kg-water/kg-fish)	Chemical Class (optional)	water - top - surface layer degradation half life (hours)	water - bottom - deep sea degradation half life (hours)	sediment (anaerobic) degradation half life (hours)	BCF (kg-water/kg-fish)	ED10 - oral- non cancer (mg / kg body weight - day, median estimate)	ED10 - inhalation - non cancer (mg / kg body weight - day, median estimate)	DALY/Incidence - oral- non cancer	DALY/Incidence - inhalation - non cancer	ED10 - oral - cancer (mg / kg body weight - day, median estimate)	ED10 - inhalation - cancer (mg / kg body weight - day, median estimate)	Aquatic Ecotoxicological Effect Factor (PAF per kg/m3, median estimate)
Tetrachloroethylene		8.28E+01	Non dissociating compound	5.50E+02	5.50E+03	5.50E+03	8.28E+01	5.26E-01	2.08E+00	1.30E+00	1.30E+00	4.00E+00		6.13E+02
Carbon tetrachloride (CCl4)		3.01E+01	Non dissociating compound	1.70E+03	1.70E+04	1.70E+04	3.01E+01	5.38E-02	4.09E+00	1.30E+00	1.30E+00	1.19E+00	1.19E+00	6.73E+01
1,3-butadiene		6.80E+00	Non dissociating compound	1.70E+02	1.70E+03	1.70E+03	6.80E+00	6.76E-01	6.76E-01	1.30E+00	1.30E+00		1.04E+01	2.01E+02
Methomyl		3.16E+00	Non dissociating compound	5.52E+03	5.04E+02	5.04E+02	3.16E+00	2.50E+00		1.30E+00	1.30E+00			7.01E+03
Acephate		3.16E+00	Non dissociating compound	1.26E+03	5.28E+01	5.28E+01	3.16E+00	2.04E-03		1.30E+00	1.30E+00	1.00E+01		7.26E+01
Formaldehyde	1.33E+01	3.16E+00	Non dissociating compound	9.60E+01	3.84E+02	3.84E+02	3.16E+00	3.75E+00	2.52E-03	1.30E+00	1.30E+00		9.09E-02	7.42E+01
PCBs		5.80E+04	Mixture (of non dissociating compounds)	3.36E+02	1.34E+03	1.34E+03	5.80E+04			1.30E+00	1.30E+00			7.50E+05
Di(n-octyl) phthalate		6.35E+01	Non dissociating compound	3.36E+02	6.54E+03	6.54E+03	6.35E+01	3.09E+00		1.30E+00	1.30E+00			1.50E+01
Hexabromobenzene		9.42E+03	Non dissociating compound	1.44E+03	5.76E+03	5.76E+03	9.42E+03	1.61E-01		1.30E+00	1.30E+00			1.45E+06
Cypermethrin		2.07E+02	Non dissociating compound	1.20E+02	1.25E+03	1.25E+03	2.07E+02	1.00E+00		1.30E+00	1.30E+00			6.47E+06
Mirex		4.03E+04	Non dissociating compound	1.70E+02	5.50E+04	5.50E+04	4.03E+04	1.85E-02		1.30E+00	1.30E+00	7.14E-02		3.33E+03
Trifluralin		2.58E+03	Non dissociating compound	1.70E+03	5.50E+03	5.50E+03	2.58E+03	7.69E-01		1.30E+00	1.30E+00	2.94E+01		1.13E+04

Name	PKa	BCF (kg-water/kg-fish)	Chemical Class (optional)	water - top - surface layer degradation half life (hours)	water - bottom - deep sea degradation half life (hours)	sediment (anaerobic) degradation half life (hours)	BCF (kg-water/kg-fish)	ED10 - oral- non cancer (mg / kg body weight - day, median estimate)	ED10 - inhalation - non cancer (mg / kg body weight - day, median estimate)	DALY/Incidence - oral- non cancer	DALY/Incidence - inhalation - non cancer	ED10 - oral - cancer (mg / kg body weight - day, median estimate)	ED10 - inhalation - cancer (mg / kg body weight - day, median estimate)	Aquatic Ecotoxicological Effect Factor (PAF per kg/m3, median estimate)
Dicofol		1.46E+03	Hydrolyses app $t_{1/2}$: 120 h (Bulle et al.)	8.99E+02	3.84E+02	3.84E+02	1.46E+03			1.30E+00	1.30E+00	1.32E+00		1.03E+04
1,4-dichlorobenzene		8.89E+01	Non dissociating compound	1.70E+03	1.70E+04	1.70E+04	8.89E+01	2.03E+00	7.69E+01	1.30E+00	1.30E+00	2.56E+01		7.63E+02
Aldrin		2.02E+04	Non dissociating compound	1.70E+04	5.50E+04	5.50E+04	2.02E+04	1.25E-03		1.30E+00	1.30E+00	1.18E-02	1.18E-02	8.27E+04
1,1,2,2-Tetrachloroethane		1.38E+01	Non dissociating compound	1.70E+03	1.70E+04	1.70E+04	1.38E+01	2.15E+00	2.32E+01	1.30E+00	1.30E+00	1.54E+00	9.09E-01	2.83E+02
Captan		2.86E+01	Non dissociating compound	1.70E+01	5.50E+02	5.50E+02	2.86E+01	3.33E+00		1.30E+00	1.30E+00			7.21E+03
Pronamide		8.73E+01	Non dissociating compound	9.79E+02	1.80E+02	1.80E+02	8.73E+01	7.69E+00		1.30E+00	1.30E+00	4.76E+00		3.66E+02
Anthracene		5.33E+02	Non dissociating compound	5.50E+02	1.70E+04	1.70E+04	5.33E+02	3.70E+01		1.30E+00	1.30E+00			3.92E+04
Gamma-HCH (lindane)		3.08E+02	Non dissociating compound	1.70E+04	5.50E+04	5.50E+04	3.08E+02	2.63E-02	1.16E-01	1.30E+00	1.30E+00	1.23E+00		2.85E+04
Dimethylphthalate (DMP)		3.40E+00	Non dissociating compound	1.70E+02	1.70E+03	1.70E+03	3.40E+00			1.30E+00	1.30E+00			4.14E+01
Methanol	1.53E+01	3.16E+00		5.50E+01	7.20E+01	7.20E+01	3.16E+00	3.79E+01		1.30E+00	1.30E+00			1.50E+00
1,2-Dichloroethane		2.75E+00	Non dissociating compound	1.70E+03	1.70E+04	1.70E+04	2.75E+00	8.70E-01	8.67E+01	1.30E+00	1.30E+00	3.23E-01	2.17E+00	2.57E+01
Ethyl acetate		3.16E+00	Non dissociating compound	9.60E+01	3.84E+02	3.84E+02	3.16E+00	7.14E+01		1.30E+00	1.30E+00			7.42E+00
N-Nitrosodiethylamine	3.89E+00	3.16E+00	Non dissociating compound	6.00E+00	2.40E+01	2.40E+01	3.16E+00			1.30E+00	1.30E+00	9.09E-04	1.33E-03	6.00E+00

Name	PKa	BCF (kg-water/kg-fish)	Chemical Class (optional)	water - top - surface layer degradation half life (hours)	water - bottom - deep sea degradation half life (hours)	sediment (anaerobic) degradation half life (hours)	BCF (kg-water/kg-fish)	ED10 - oral- non cancer (mg / kg body weight - day, median estimate)	ED10 - inhalation - non cancer (mg / kg body weight - day, median estimate)	DALY/Incidence - oral- non cancer	DALY/Incidence - inhalation - non cancer	ED10 - oral - cancer (mg / kg body weight - day, median estimate)	ED10 - inhalation - cancer (mg / kg body weight - day, median estimate)	Aquatic Ecotoxicological Effect Factor (PAF per kg/m3, median estimate)
Thiram	8.70E-01	4.29E+00		1.70E+02	1.70E+03	1.70E+03	4.29E+00	1.33E+00		1.30E+00	1.30E+00			4.00E+04
Propoxur	1.19E+01	2.95E+00		5.50E+02	1.70E+03	1.70E+03	2.95E+00	3.23E-02		1.30E+00	1.30E+00			3.21E+03
Folpet		3.12E+01	Non dissociating compound	1.38E+04	1.38E+04	1.38E+04	3.12E+01	1.00E+01		1.30E+00	1.30E+00	4.17E+01		8.07E+03
Benomyl		8.56E+00	Significant hydrolysis	1.70E+02	5.50E+03	5.50E+03	8.56E+00	1.33E+00		1.30E+00	1.30E+00			1.31E+03
Hexachlorobutadiene		9.56E+02	Non dissociating compound	1.70E+03	1.70E+03	1.70E+03	9.56E+02	1.40E-03	1.14E+00	1.30E+00	1.30E+00	2.94E+00	2.94E+00	5.82E+03
Hexachlorocyclopentadiene		1.52E+03	Hydrolyses app $t_{1/2}$: 173 h (Bulle et al.)	8.65E+01	1.68E+03	1.68E+03	1.52E+03	5.56E-01	1.80E-03	1.30E+00	1.30E+00			4.38E+04
Heptachlor epoxide		1.36E+03	Non dissociating compound	7.02E+03	9.60E+01	9.60E+01	1.36E+03	2.33E-03		1.30E+00	1.30E+00	2.17E-02	2.17E-02	1.66E+05
Hexachlorobenzene		5.15E+03	Non dissociating compound	5.50E+04	5.50E+04	5.50E+04	5.15E+03	2.13E-02	1.50E-05	1.30E+00	1.30E+00	7.69E-02	7.69E-02	2.16E+04
Heptachlor		9.93E+03	Hydrolyses app $t_{1/2}$: 23.1 h (Bulle et al.)	5.50E+02	5.50E+03	5.50E+03	9.93E+03	4.00E-02		1.30E+00	1.30E+00	4.76E-02	4.35E-02	1.02E+05
Nitrobenzene		5.30E+00		1.70E+03	5.50E+03	5.50E+03	5.30E+00	3.23E-02		1.30E+00	1.30E+00			
Endosulfan		1.78E+02		1.11E+02	7.68E+02	7.68E+02	1.78E+02	1.59E-01		1.30E+00	1.30E+00			2.65E+05

B.3 Additional USEtox parameterization results

Figure B.1 compares the freshwater residence times (fate factors) for 14 chemicals emitted in each sub-continent, in the a-spatial USEtox model and the spatially differentiated IMPACTWorld model. It is similar to Figure 3.2.b, but with results differentiated by substance and for a restricted number of substances.

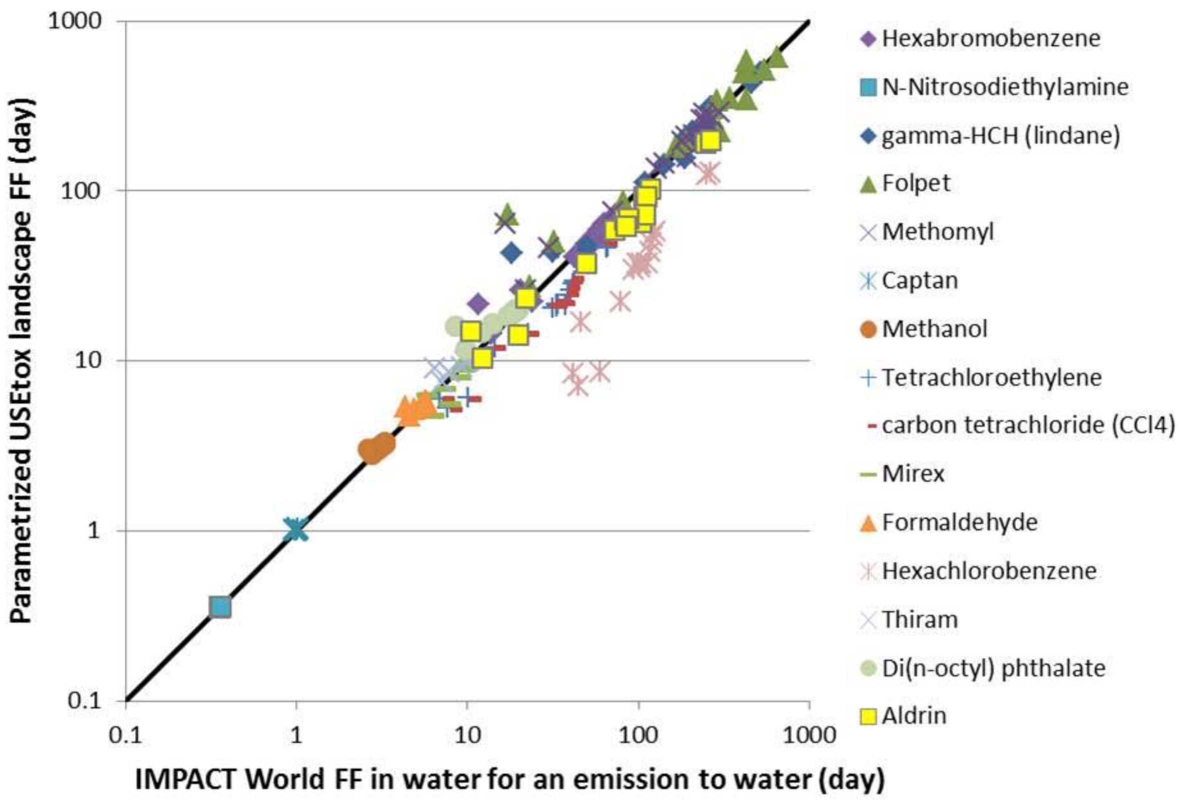


Figure B.1: Comparison between a-spatial USEtox model and the spatially differentiated IMPACTWorld model predictions of freshwater residence time (fate factor, FF) for 14 chemicals emitted in each sub-continent

We calculated transfer fractions from air to freshwater ($TF_{a,w}$, unitless) and from fresh water to air ($TF_{w,a}$, unitless) based on the following fate factors:

$$TF_{a,w} = \frac{FF_{a,w}}{FF_{w,w}}$$

$$TF_{w,a} = \frac{FF_{w,a}}{FF_{a,a}}$$

Equation B.1: Transfer fraction from air to freshwater

where all the fate factors (FF) are in units of days and equal to the steady state substance mass in the second subscript (in kg) for an emission flow of 1 kg/day to the compartment indicated by the first subscript. $FF_{a,w}$ is the fate factor in fresh water for an emission to air $FF_{w,a}$ is the fate factor in air for an emission to fresh water (days), $FF_{w,w}$ is the fate for an emission to fresh water in fresh water (days), and $FF_{a,a}$ is the fate factor in air for an emission to air (days).

Figure B.2a shows that for many pollutants, IMPACTWorld overestimates the transfer factor from air to fresh water by about one order of magnitude compared to USEtox. The dominant disappearance pathway of pollutants with high K_H (e.g., hexachlorobenzene K_H =170 Pa.m3.mol-1, carbon tetrachloride K_H =2760 Pa.m3.mol-1, and n-nitrosodiethylamine KH=0.362 Pa.m^3.mol^{-1}) emitted to continental air is transfer to global air.

The transfer factor from air to fresh water is higher than 1.0 for hexachlorobenzene in IMPACTWorld for emission to Brazil (W8) and East Indies (W14). This result is in line with Figure 3.2.b results, where the fate of hexachlorobenzene in W14 is observed to exceed the freshwater residence time in this sub-continental zone due to a transfer to Antarctica (W11). In the same way, when hexachlorobenzene is emitted to air in East Indies and Brazil, it is transported to Antarctica where the freshwater residence time is higher than in East Indies and Brazil (>8000 y compared to 19 days and 34 days). Hexachlorobenzene fate in water when emitted to air is thus higher than when emitted in water in W8 and W14, due to a transfer to Antarctica and important substance residence time there.

Figure B.2.b shows that when emitted to fresh water, the thiram and n-nitrosodiethylamine transfer fractions to air are under-estimated by USEtox by six and three orders of magnitude, respectively.

Based on these observations, deviations in fate results are more due to model algorithm differences (i.e., modelling of freshwater outflow and the volatilization algorithm) than to the influence of surrounding global or continental zones.

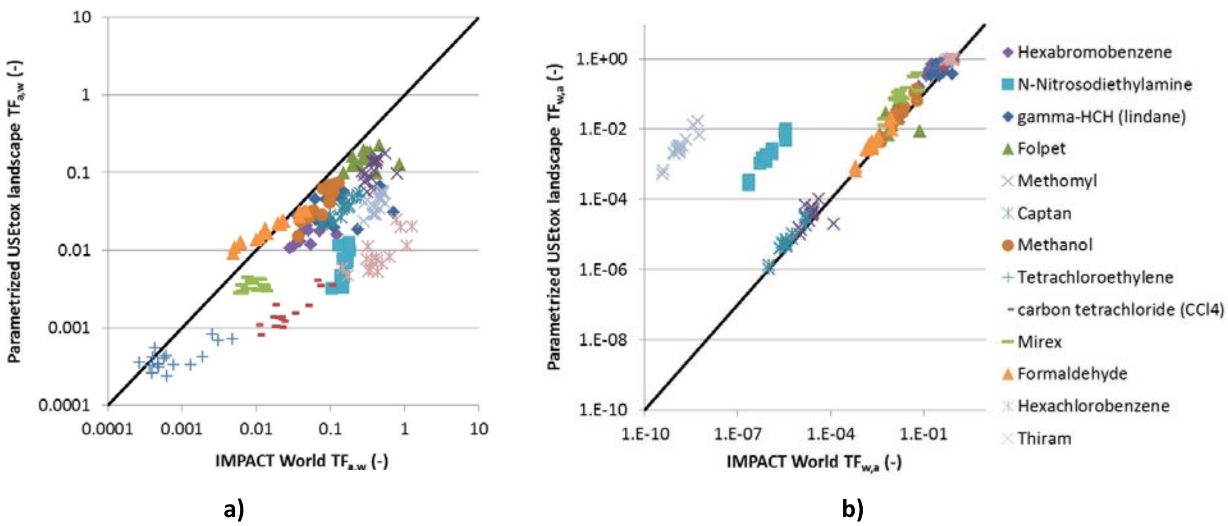


Figure B.2: Comparison between a-spatial USEtox model and the spatially differentiated IMPACTWorld model regarding: a) Transfer fraction from air to fresh water and b) Transfer fraction from freshwater to air

Figure B.3 and Figure B.4 present respectively intake fractions for an emission to fresh water and for an emission to air. Figure B.3 a shows that results for both models are aligned except for thiram and n-nitrosodiethylamine. This difference is correlated with the discrepancies observed for transfer fraction from fresh water to air (see Figure B.2.b), and thus due to a difference in fate factor $FF_{w,a}$.

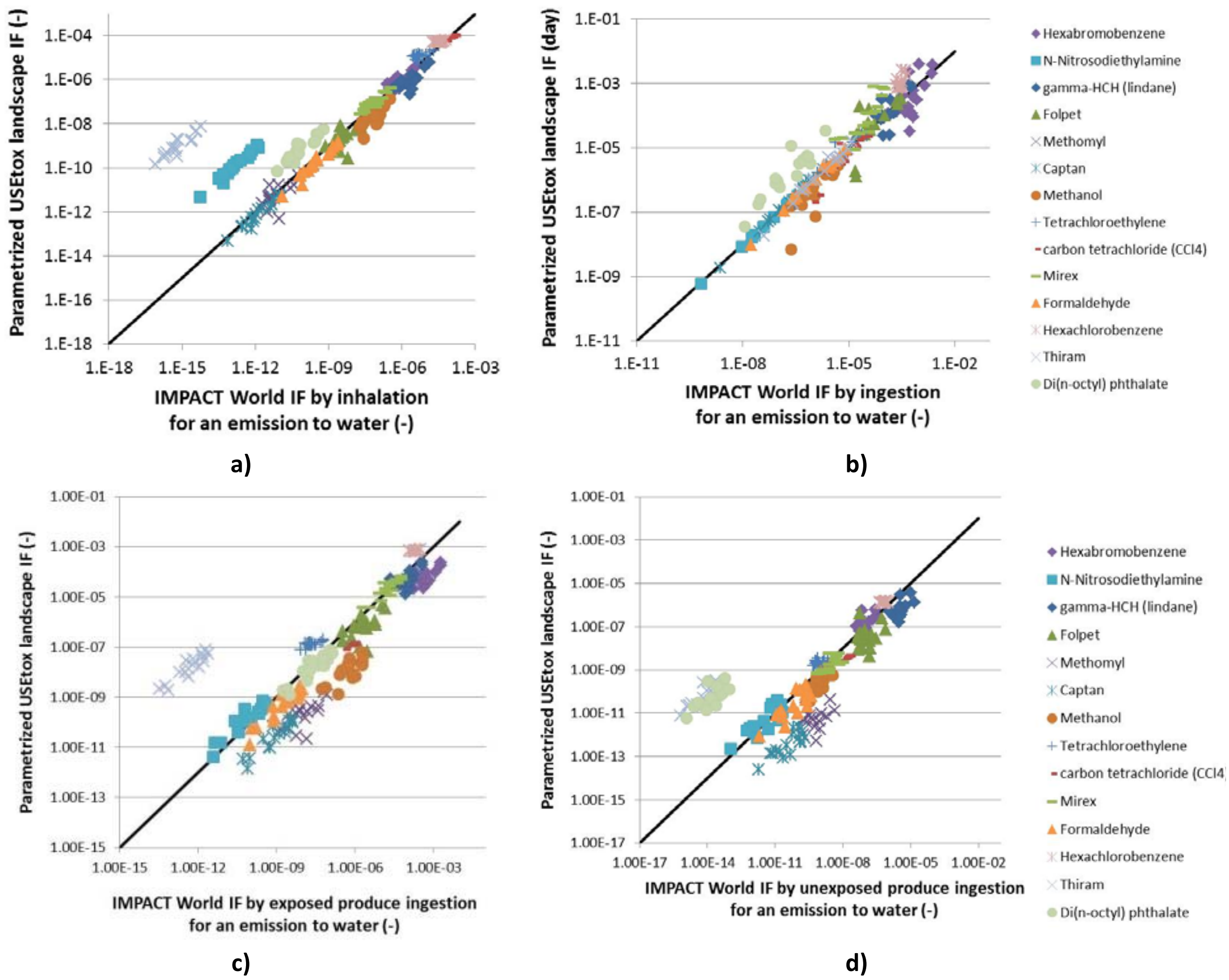


Figure B.3: Comparison between a-spatial USEtox model and the spatially differentiated IMPACTWorld model regarding the intake fraction for an emission to water: a) by inhalation, b) by total ingestion, c) by exposed produce ingestion, d) by unexposed produce ingestion

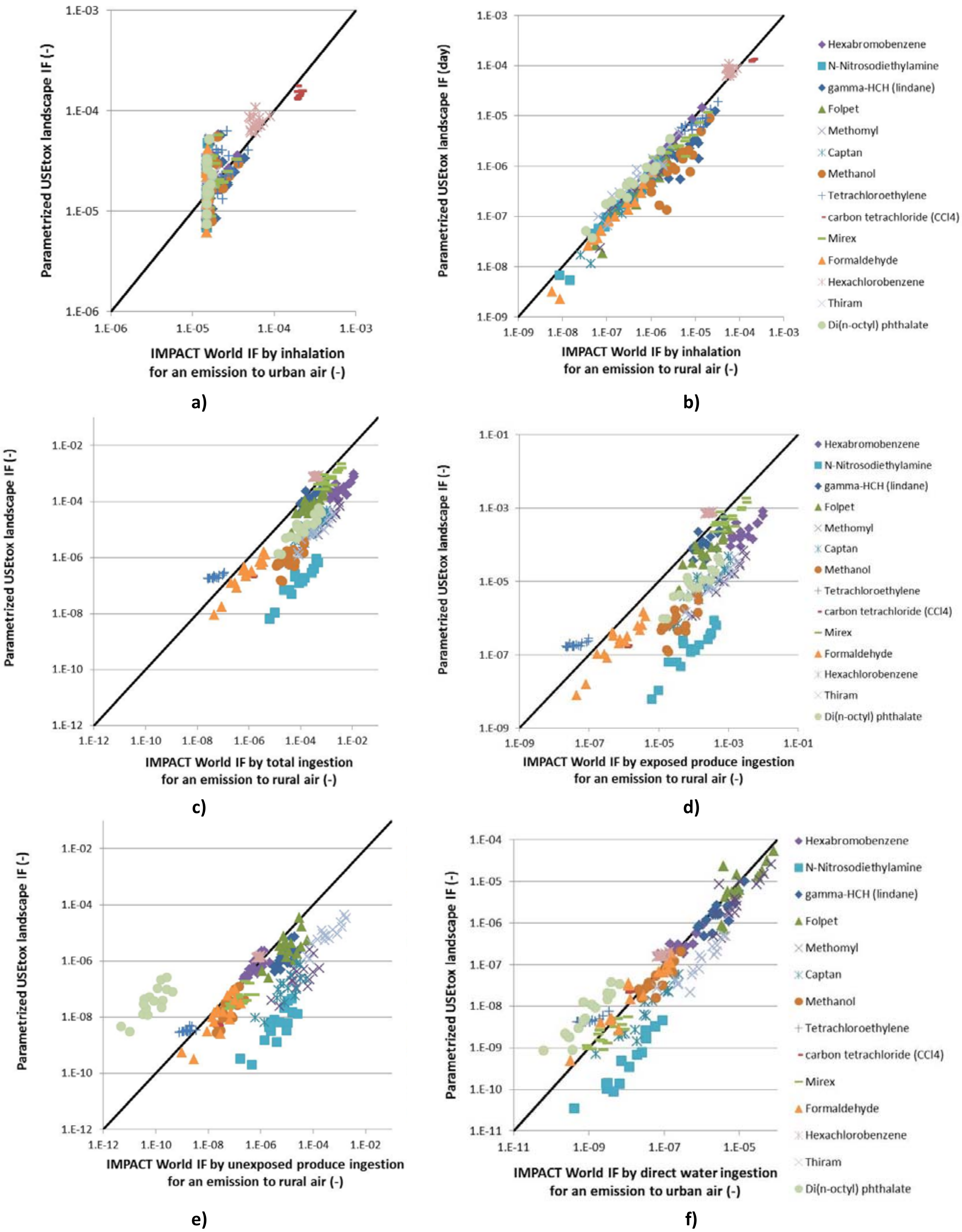
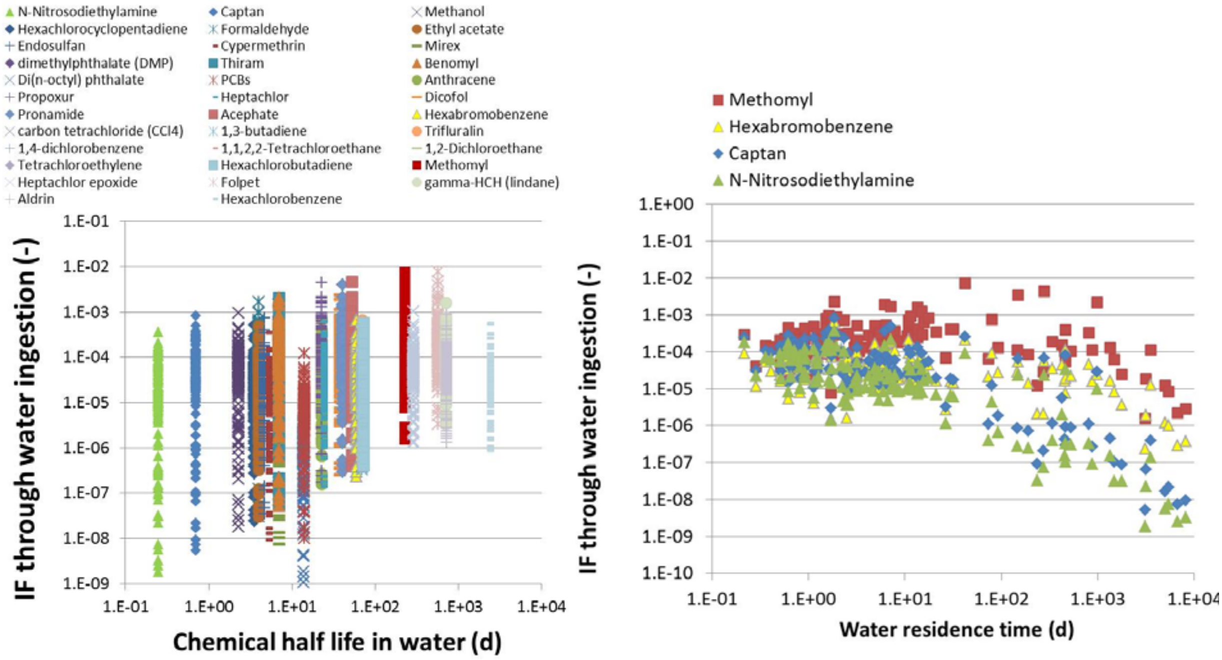


Figure B.4: Comparison between a-spatial USEtox model and the spatially differentiated IMPACTWorld model regarding the intake fraction for an emission to air: a) by inhalation for an emission in an urban zone, b) by inhalation for an emission in an rural zone c) by total ingestion, d) by exposed produce ingestion, e) by unexposed produce ingestion, f) by freshwater ingestion

B.4 Additional intake fraction variation analysis

Figure B5.a presents the variability in intake fraction through water ingestion across European watersheds, as calculated by the spatially differentiated European model for emissions in each of the European watersheds, as a function of the chemical degradation half-life in fresh water. In contrast to the variation of fate, the intake fraction varies by up to five orders of magnitude for quickly degraded pollutants with half-lives shorter than a day, such as n-nitrosodiethylamine ($t_{1/2}$=6 h in fresh water). On the contrary, the intake fraction through freshwater ingestion varies by three to four orders of magnitude for persistent pollutants with half-lives larger than 100 days, such as methomyl ($t_{1/2}$=230 days in fresh water).

By displaying the intake fraction through water ingestion as a function of the freshwater residence time for four pollutants with different persistences in fresh water (n-nitrosodiethylamine $t_{1/2}$=0.25 d, captan: $t_{1/2}$=0.71 d, hexabromobenzene: $t_{1/2}$=73 d, and methomyl: $t_{1/2}$=230 d), Figure B.**5**b shows that the variability across watershed decreases as water residence time increases, more prominently for quickly degraded pollutants. In short-residence-time watersheds, all substances are equally ingested through drinking water independently of their persistence (persistent substances are removed from the system by advection). In long-residence-time watersheds, the intake fraction of n-nitrosodiethylamine is up to 4 orders of magnitude lower than in short-residence-time watersheds. This is explained by the fact that low persistence substances are degraded in watersheds where they have been emitted, where the population is generally lower than for watersheds close to the coast (population is the only spatially differentiated parameter affecting exposure by water ingestion). This variability is reduced to three orders of magnitude for methomyl, given that this substance crosses watersheds with various population patterns due to its high persistence.



a) b)

Figure B.5: Main factors affecting intake fraction through water ingestion. a) Intake fractions through water ingestion of all test substances as a function of their degradation half-life in water for each of the 136 watersheds of the European spatial model and b) Intake fractions through water ingestion of n-nitrosodiethylamine, captan, hexabromobenzene and methomyl as a function of water residence time to sea for each of the 136 watershed of the European spatial model

B.5 Classification of IMPACT Europe spatial model watersheds into archetype categories

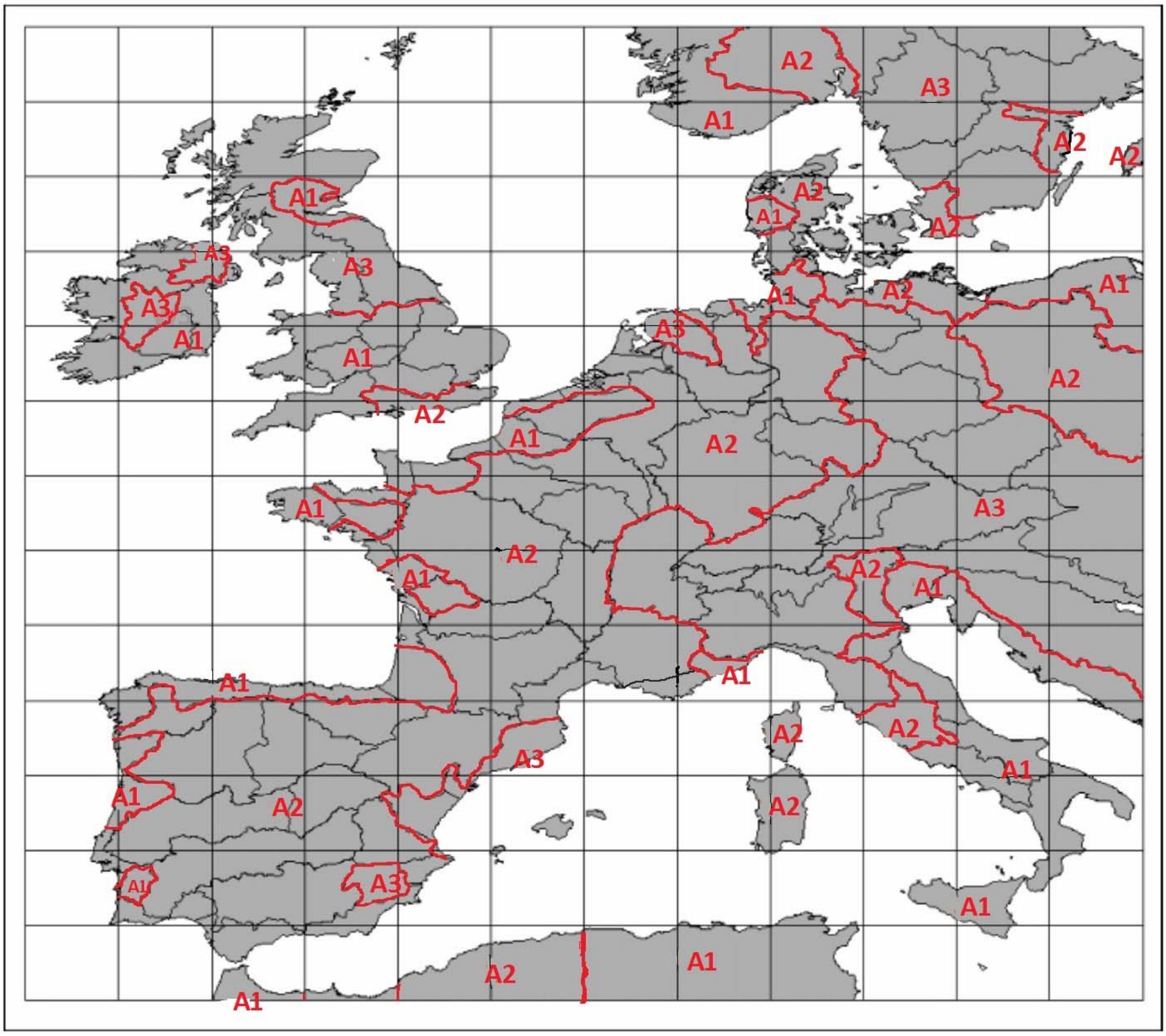


Figure B.6: Boundaries of the three archetypical watershed A1, A2 and A3 model based on IMPACT Europe spatial model (Pennington et al. 2005)

Table B.7: Classification of IMPACT Europe spatial model watersheds into A1, A2 and A3 archetype categories

Region No	Volume (m3)	Advection rate (m3/h)	Is there a watershed after this one	Retention in watershed (d)	Retention after watershed (d)	Retention time until the sea (d)	Archetype watershed classification
W0	4.00E+13	4.15E+09		4.01E+02		4.01E+02	A0
W2	1.00E+08	1.24E+06		3.39E+00		3.39E+00	A2
W3	1.04E+07	2.49E+05		1.74E+00		1.74E+00	A1
W4	8.79E+09	8.15E+05	W4	4.50E+02	8.83E+02	1.33E+03	A3
W5	7.46E+10	6.78E+06	W5	4.58E+02	4.24E+02	8.83E+02	A3
W6	6.63E+07	9.28E+05		2.98E+00		2.98E+00	A2
W7	9.76E+07	1.47E+06	W7	2.77E+00	3.15E+00	5.92E+00	A2
W8	1.43E+08	1.88E+06		3.15E+00		3.15E+00	A2
W9	2.80E+07	5.28E+05		2.21E+00		2.21E+00	A2
W10	1.90E+08	2.04E+06		3.90E+00		3.90E+00	A2
W11	8.89E+07	8.80E+05		4.21E+00		4.21E+00	A2
W12	9.41E+07	5.88E+05		6.67E+00		6.67E+00	A2
W13	5.66E+06	9.99E+04		2.36E+00		2.36E+00	A2
W14	9.66E+07	1.23E+06	W14	3.28E+00	4.62E+00	7.90E+00	A2
W15	3.49E+08	3.15E+06		4.62E+00		4.62E+00	A2
W16	1.22E+08	1.04E+06		4.93E+00		4.93E+00	A2
W17	7.92E+07	1.43E+06		2.31E+00		2.31E+00	A2
W18	1.39E+11	1.60E+06	W18	3.62E+03	1.76E+03	5.38E+03	A3
W19	2.09E+11	4.94E+06		1.76E+03		1.76E+03	A3
W20	1.31E+11	3.71E+06	W20	1.48E+03	7.18E+00	1.48E+03	A3
W21	5.23E+07	5.74E+05	W21	3.80E+00	7.18E+00	1.10E+01	A2
W22	9.12E+08	7.72E+06	W22	4.93E+00	2.26E+00	7.18E+00	A2
W23	4.71E+08	8.69E+06		2.26E+00		2.26E+00	A2
W24	1.06E+11	9.15E+05	W24	4.83E+03	1.45E+02	4.97E+03	A3
W25	1.39E+10	4.04E+06	W25	1.43E+02	2.57E+00	1.45E+02	A3
W26	3.72E+08	6.05E+06		2.57E+00		2.57E+00	A2
W27	1.90E+07	4.81E+05		1.64E+00		1.64E+00	A1
W28	1.65E+07	4.59E+03		1.50E+02		1.50E+02	A3
W29	4.60E+07	8.49E+05	W29	2.26E+00	3.08E+00	5.34E+00	A2
W30	3.20E+07	5.53E+05	W30	2.41E+00	3.08E+00	5.49E+00	A2
W31	1.20E+08	1.63E+06		3.08E+00		3.08E+00	A2
W32	1.23E+07	2.08E+05	W32	2.46E+00	4.72E+00	7.18E+00	A2
W33	1.96E+08	1.73E+06		4.72E+00		4.72E+00	A2
W34	3.07E+06	2.08E+05		6.16E-01		6.16E-01	A1
W35	1.95E+08	1.72E+06		4.72E+00		4.72E+00	A2
W36	2.97E+06	1.34E+05		9.24E-01		9.24E-01	A1
W37	4.01E+07	4.40E+05	W37	3.80E+00	3.18E+00	6.98E+00	A2
W38	1.74E+08	2.28E+06		3.18E+00		3.18E+00	A2
W39	7.71E+06	3.91E+05		8.21E-01		8.21E-01	A1
W40	8.74E+07	3.18E+05		1.14E+01		1.14E+01	A2
W41	1.28E+08	4.53E+05		1.17E+01		1.17E+01	A2
W42	1.95E+10	2.30E+05		3.54E+03		3.54E+03	A3
W43	6.52E+07	2.79E+03		9.75E+02		9.75E+02	A3
W44	3.27E+07	1.18E+05		1.15E+01		1.15E+01	A2
W45	7.70E+07	1.51E+05		2.12E+01		2.12E+01	A2

W46	7.86E+07	2.09E+05		1.56E+01		1.56E+01	A2
W47	1.24E+07	2.93E+04		1.76E+01		1.76E+01	A2
W48	1.70E+06	1.15E+04		6.16E+00		6.16E+00	A2
W49	3.31E+07	1.19E+05		1.16E+01		1.16E+01	A2
W50	3.01E+07	9.79E+05		1.28E+00		1.28E+00	A1
W51	1.58E+07	7.11E+05		9.24E-01		9.24E-01	A1
W52	3.02E+07	1.06E+06		1.18E+00		1.18E+00	A1
W53	2.19E+07	1.37E+06		6.67E-01		6.67E-01	A1
W54	6.36E+06	7.38E+05		3.59E-01		3.59E-01	A1
W55	4.86E+07	1.23E+06		1.64E+00		1.64E+00	A1
W56	3.03E+07	1.60E+05		7.87E+00		7.87E+00	A2
W57	3.45E+06	1.75E+05		8.21E-01		8.21E-01	A1
W58	3.68E+07	1.11E+05		1.38E+01		1.38E+01	A2
W59	6.32E+06	3.21E+05		8.21E-01		8.21E-01	A1
W60	1.99E+07	9.87E+04		8.42E+00		8.42E+00	A2
W61	2.04E+06	2.96E+05		2.87E-01		2.87E-01	A1
W62	5.26E+06	2.67E+05		8.21E-01		8.21E-01	A1
W63	1.07E+06	1.08E+05		4.10E-01		4.10E-01	A1
W64	6.56E+06	2.22E+05		1.23E+00		1.23E+00	A1
W65	1.00E+07	1.30E+05		3.21E+00		3.21E+00	A2
W66	6.78E+07	2.10E+05		1.35E+01		1.35E+01	A2
W67	8.96E+08	4.67E+05		7.99E+01		7.99E+01	A3
W68	9.46E+06	1.51E+05		2.61E+00		2.61E+00	A2
W69	4.90E+06	2.21E+05		9.24E-01		9.24E-01	A1
W70	2.60E+06	1.51E+05		7.18E-01		7.18E-01	A1
W71	5.53E+06	3.46E+05		6.67E-01		6.67E-01	A1
W72	4.56E+07	3.50E+05		5.43E+00		5.43E+00	A2
W73	8.58E+06	3.87E+05		9.24E-01		9.24E-01	A1
W74	1.46E+08	4.50E+05		1.35E+01		1.35E+01	A2
W75	7.17E+07	3.39E+05		8.83E+00		8.83E+00	A2
W76	1.26E+08	1.66E+05		3.16E+01		3.16E+01	A2
W77	9.29E+07	3.18E+05		1.22E+01		1.22E+01	A2
W78	9.47E+07	2.58E+05		1.53E+01		1.53E+01	A2
W79	7.64E+09	9.19E+05	W79	3.46E+02	4.59E+02	8.05E+02	A3
W80	1.46E+08	2.20E+06	W80	2.77E+00	4.56E+02	4.59E+02	A3
W81	2.23E+10	2.42E+06	W81	3.83E+02	7.31E+01	4.56E+02	A3
W82	4.79E+09	2.73E+06		7.31E+01		7.31E+01	A3
W83	4.73E+07	1.60E+06		1.23E+00		1.23E+00	A1
W84	9.42E+06	3.19E+05		1.23E+00		1.23E+00	A1
W85	3.49E+09	1.57E+06		9.27E+01		9.27E+01	A3
W86	1.60E+07	7.23E+05		9.24E-01		9.24E-01	A1
W87	4.68E+07	8.64E+05		2.26E+00		2.26E+00	A2
W88	1.34E+07	4.96E+05		1.13E+00		1.13E+00	A1
W89	2.40E+07	1.39E+06		7.18E-01		7.18E-01	A1
W90	2.29E+07	1.55E+06		6.16E-01		6.16E-01	A1
W91	1.24E+07	8.40E+05		6.16E-01		6.16E-01	A1
W92	1.54E+08	6.08E+05		1.06E+01		1.06E+01	A2
W93	1.08E+08	6.75E+05		6.66E+00		6.66E+00	A2
W94	4.16E+07	5.83E+04		2.97E+01		2.97E+01	A2
W95	3.00E+07	1.52E+06		8.21E-01		8.21E-01	A1

W96	2.42E+07	5.79E+05		1.74E+00		1.74E+00	A1
W97	9.04E+06	7.34E+05		5.13E-01		5.13E-01	A1
W98	4.02E+09	7.11E+05		2.35E+02		2.35E+02	A3
W99	5.01E+07	1.85E+06		1.13E+00		1.13E+00	A1
W100	5.48E+09	4.96E+05		4.61E+02		4.61E+02	A3
W101	1.42E+07	9.59E+05		6.16E-01		6.16E-01	A1
W102	4.12E+11	5.55E+06		3.09E+03		3.09E+03	A3
W103	2.27E+07	1.03E+06		9.24E-01		9.24E-01	A1
W104	1.45E+10	3.20E+06		1.89E+02		1.89E+02	A3
W105	8.44E+09	1.04E+06		3.39E+02		3.39E+02	A3
W106	1.85E+07	1.25E+06		6.16E-01		6.16E-01	A1
W107	1.65E+07	4.47E+05		1.54E+00		1.54E+00	A1
W108	2.01E+07	4.29E+05		1.95E+00		1.95E+00	A1
W109	1.59E+07	4.04E+05		1.64E+00		1.64E+00	A1
W110	1.19E+07	2.69E+05		1.85E+00		1.85E+00	A1
W111	1.08E+08	3.01E+05		1.50E+01		1.50E+01	A2
W112	1.27E+07	1.03E+06		5.13E-01		5.13E-01	A1
W113	2.84E+08	6.94E+06		1.71E+00		1.71E+00	A1
W114	1.36E+08	2.20E+06		2.57E+00		2.57E+00	A2
W115	1.88E+11	6.18E+05		1.27E+04		1.27E+04	A3
W116	2.88E+11	1.79E+06		6.71E+03		6.71E+03	A3
W117	1.69E+11	8.55E+05		8.21E+03		8.21E+03	A3
W118	1.64E+07	2.13E+05		3.20E+00		3.20E+00	A2
W119	4.87E+09	7.42E+05		2.74E+02		2.74E+02	A3
W120	1.44E+08	2.25E+05		2.66E+01		2.66E+01	A2
W121	5.92E+10	7.79E+05		3.17E+03		3.17E+03	A3
W122	3.23E+07	5.34E+05		2.52E+00		2.52E+00	A2
W123	7.72E+06	5.63E+05		5.72E-01		5.72E-01	A1
W124	3.43E+08	2.24E+06		6.37E+00		6.37E+00	A2
W125	9.30E+06	2.52E+05		1.54E+00		1.54E+00	A1
W126	4.62E+09	8.42E+06	W126	2.28E+01	4.01E+02	4.24E+02	A3
W127	7.40E+09	2.50E+06	W127	1.23E+02	4.01E+02	5.24E+02	A3
W128	7.37E+09	5.03E+06	W128	6.11E+01	4.01E+02	4.62E+02	A3
W129	6.24E+07	5.03E+06		5.16E-01		5.16E-01	A1
W130	1.77E+06	3.40E+05		2.17E-01		2.17E-01	A1
W131	1.04E+07	1.56E+03		2.78E+02		2.78E+02	A3
W132	1.16E+08	4.43E+05		1.10E+01		1.10E+01	A2
W133	2.39E+07	2.36E+04		4.21E+01		4.21E+01	A2
W134	1.81E+07	4.31E+05		1.74E+00		1.74E+00	A1
W135	1.44E+07	3.39E+05		1.77E+00		1.77E+00	A1
W136	4.46E+07	1.47E+06		1.26E+00		1.26E+00	A1

Appendix C

Spatial analysis of toxic emissions in LCA: A sub-continental nested USEtox model with freshwater archetypes

C.1 Determination of P dissolved ratio

Table C.1: Total-P concentrations under high flow and low flow regimes in the Vansjø catchment, Norway

Streams		Total-P concentrations under high-flow regimes in (μg/L)					Total- P concentrations under low flow regime (µg/L)			
	Sampling stream	Number of samples	P bound to dissolved organic matter (DOM-P)	Free-PO_4^{3-}	Particulate bound phosphorus (PP)	Total	P bound to dissolved organic matter (DOM-P)	Free-PO_4^{3-}	Particulate bound phosphorus (PP)	
STO1	**Stoa1**	7	14.79	104.6	154.8	274.19	2.39	3.81	16.5	22.7
ORE	**Orejordet**	1	3.1	1.12	10.3	14.52	2.55	1.16	5.5	9.21
HUG	**Huggenes**	17	8.6	11.4	91.6	111.6	6.56	2.9	20.52	29.98
VAS	**Vaskeberget**	4	6.1	26.17	41.7	73.97	13.7	13.2	18.83	45.73
ARV	**Arvold**	1	2	7.58	14.8	24.38	5.2	1.2	7.9	14.3
SPE	**Sperrebotn**	1	13.6	21.7	46	81.3	6.49	5.89	6.56	18.94
AUG	**Augerod**	4	4.2	2.7	13.8	20.7	3.99	2.98	7.06	14.03
GUT	**Guthus**	1	14.4	5.9	32.9	53.2	4.8	2.4	7.3	14.5
DALEN	**Dalen**	25	5.2	1	2.9	9.1	4.5	1.34	3.8	9.64

Table C.2: Total-P fractions under high flow and low flow regimes in the Vansjø catchment, Norway

Streams		Total-P fractions under high- flow regimes in (μg/L)				Total- P fractions under low flow regime (µg/L)		
	Sampling stream	Number of samples	P bound to dissolved organic matter (DOM-P)	Free-PO_4^{3-}	Particulate bound phosphorus (PP)	P bound to dissolved organic matter (DOM-P)	Free-PO_4^{3-}	Particulate bound phosphorus (PP)
STO1	Stoa1	7	5%	38%	56%	11%	17%	73%
ORE	Orejordet	1	21%	8%	71%	28%	13%	60%
HUG	Huggenes	17	8%	10%	82%	22%	10%	68%
VAS	Vaskeberget	4	8%	35%	56%	30%	29%	41%
ARV	Arvold	1	8%	31%	61%	36%	8%	55%
SPE	Sperrebotn	1	17%	27%	57%	34%	31%	35%
AUG	Augerod	4	20%	13%	67%	28%	21%	50%
GUT	Guthus	1	27%	11%	62%	33%	17%	50%
DALEN	Dalen	25	57%	11%	32%	47%	14%	39%
Total free-PO_4^{3-} average of all sites under high and low flow regimes								**19%**

C.2 Derivation of $IF_{max,j}$

We introduced a threshold intake fraction value within a single grid cell j $IF_{max,j}$ to limit the IF_j value for water-scarce grid cells. $IF_{max,j}$ is determined by considering the fate factor $FF_{j,modified}$ in a receptor cell *j* that includes freshwater removal rate $k_{withdrawn,j}$ [year^{-1}] in addition to other removal processes $k_{adv,j}$, $k_{sed,j}$, $k_{evap,j}$ and k_{deg} [year^{-1}]. This addition reflects the fact that drinking water withdrawn is not available in the same grid cell, while it can be reused in grid cells downstream (drinking water is withdrawn in a centralized system for large cities). $XP_{j,max}$ [year] is the maximum exposure factor calculated using modelled industrial, domestic and agricultural withdrawal, while XP_j is calculated based on the water requirement of the receptor cell. $IF_{max,j}$ is defined as:

$$IF_{max,j} = FF_{j,modified} * XP_{j,max} = \frac{1}{k_{adv,j} + k_{sed,j} + k_{evap,j} + k_{deg} + k_{withdrawn,j}} * XP_{j,max}$$

$$= \frac{1}{\frac{1}{FF_j} + k_{withdrawn,j}} * XP_{j,max}$$

Equation C.1: Definition of the maximal intake fraction in a receptor cell *j*

The withdrawal rate $k_{withdrawn,j}$ [year^{-1}] is calculated based on the withdrawal values for domestic, industrial and agricultural use from the World Water Development Report II (Water systems analysis group 2014) respectively named $Q_{dom,j}$, $Q_{ind,j}$ and $Q_{agr,j}$ [km^3.y^{-1}], the volume of water $V_{grid,j}$ [km^3] in the receptor cell *j* defined in equation T4.1.1 and the fraction of surface water $f_{surf,j}$ [-] defined in equation 5 . $k_{withdrawn,j}$ is calculated as:

$$k_{withdrawn,j} = \frac{f_{surf,j} * (Q_{dom,j} + Q_{ind,j} + Q_{agr,j})}{V_{grid,j}}$$

Equation C.2: Definition of the withdrawal rate in a receptor cell *j*

When $Q_{dom,j}$, $Q_{ind,j}$ and $Q_{agr,j}$ are null according to the World Water Development Report II model, we estimate $Q_{dom,j}$ based on the water requirement of the receptor cell as in equation A2.c. The parameters involved are $V_{ing\,wat}$ [km^3], $N_{pop,j}$ [-] and $f_{surf,j}$ [-] defined in equation 5 and the fraction of domestic water used for a drinking purpose $f_{drink\_dom,j}$ [-]. f_{drink_dom} is estimated as 1%, calculated as the ratio of drinking water 511 [l/(person*year)] over the population weighted average of drinking water withdrawal $Q_{dom,j}$ attributed per capita 50'000 [l/(person*year)].

$$Q_{dom,j} = V_{ing\,wat} * N_{pop,j} * f_{surf,j} * \frac{1}{f_{drink\_dom}}$$

Equation C.3: Calculation the domestic withdrawal in a receptor cell *j* in case modelled data is unavailable

$XP_{j,max}$ [year] is the maximum exposure factor calculated involving $f_{diss\,subs}$, f_{drink_dom}, $f_{surf,j}$, $Q_{dom,j}$, $Q_{ind,j}$, $Q_{agr,j}$ and $V_{grid,j}$ defined previously. The fraction of water withdrawn for a domestic purpose over total withdrawal $f_{dom\_tot,j}$ is calculated using previously introduced withdrawal data. $XP_{j,max}$ [year] is calculated as follows:

$$XP_{j,max} = \frac{f_{diss\,subs} * f_{drink\_dom} * f_{dom\_tot,j} * f_{surf,j} * (Q_{dom,j} + Q_{ind,j} + Q_{agr,j})}{V_{grid,j}} = f_{diss\,subs} * f_{drink\_dom} * f_{dom\_tot,j} * k_{withdrawn,j}$$

Equation C.4: Calculation of the maximum exposure factor in a receptor cell *j*

By introducing both $XP_{j,max}$ and $k_{withdrawn,j}$ values in $IF_{max,j}$ calculation, one can obtain the following expression:

$$IF_{max,j} = \frac{1}{\frac{1}{FF_j * k_{withdrawn,j}} + 1} * f_{diss\,subs} * f_{drink\_dom,j} * f_{dom\_tot,j}$$

Equation C.5: Final resolution for the maximal intake fraction in a receptor cell *j*

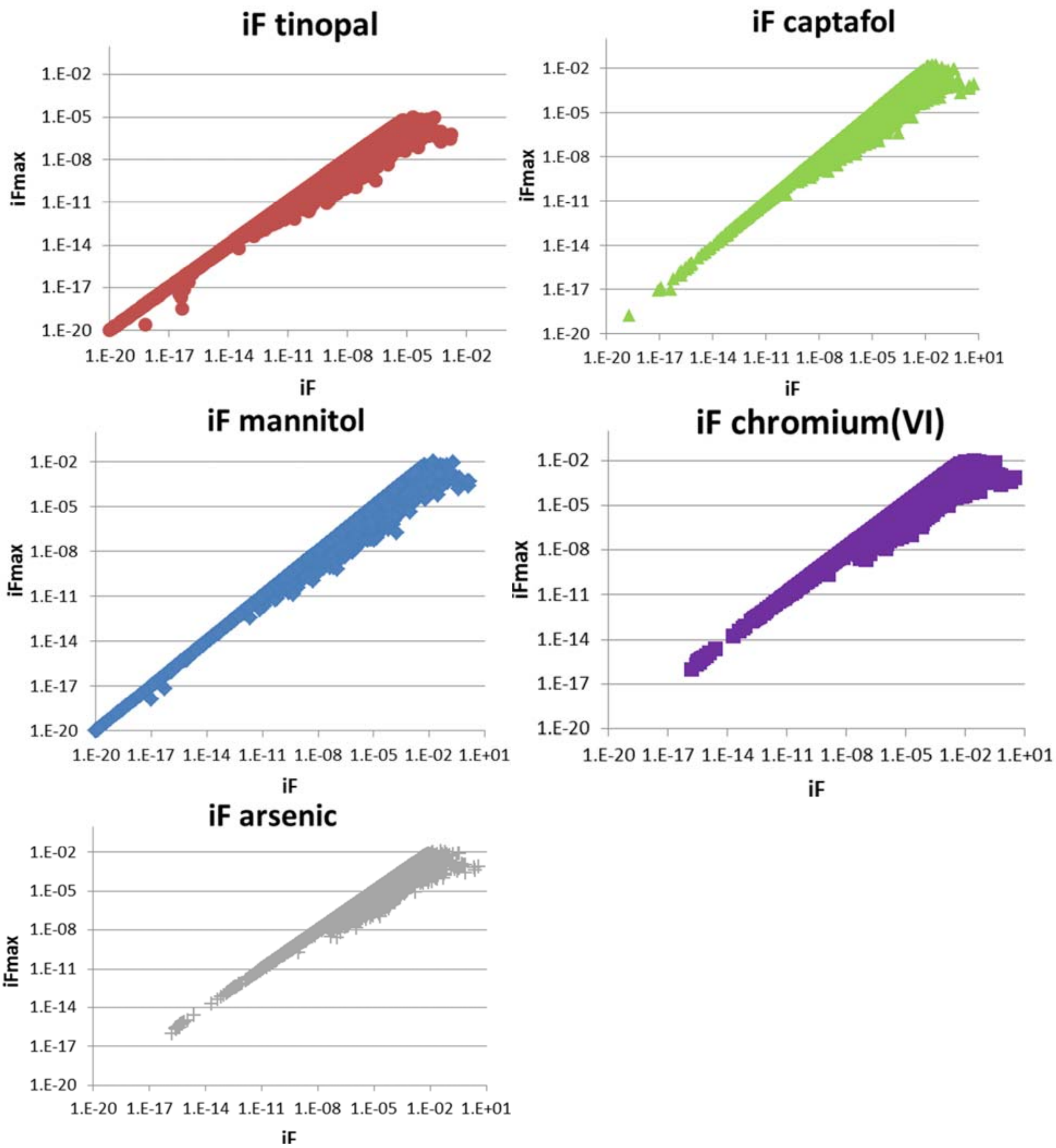


Figure C.1: Comparison between the intake fraction limited by the maximum threshold (defined in Equation 4.6) versus the intake fraction (defined in Equation 4.4)

C.3 Choice of representative substances

Figure C.2 shows the distribution of degradation, sedimentation and evaporation rates of respectively all organic and inorganic substances covered by the USEtox database (Rosenbaum et al. 2008) including recommended and interim characterization factors (3073 organics and 21 inorganics). The boxplots show the 5th centile, 1st quartile, mediane, 3rd quartile, 95th centile minimum for each parameter. For organic substances, the degradation rate varies by 1 order of magnitude while the sedimentation and evaporation vary by respectively 3 and 13 orders of magnitude between the 5th and the 95th value. For inorganic substances, the sedimentation rate varies by more than 1 order of magnitude.

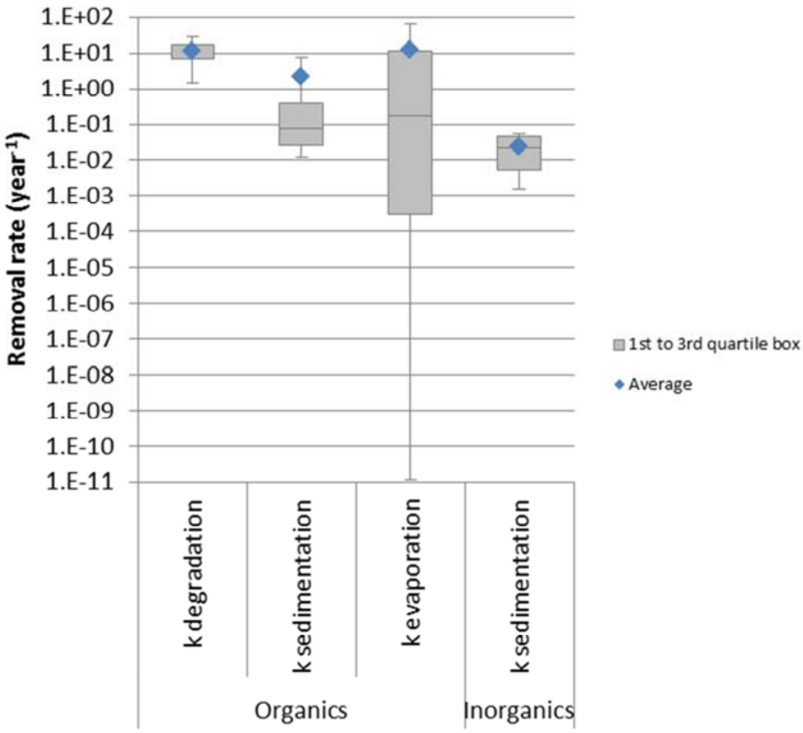


Figure C.2: Variation of the sedimentation, degradation and evaporation rate of a. organic and inorganic substances covered by the USEtox database (Rosenbaum et al. 2008)

C.4 Contribution to toxicity of the ecoinvent v2.2 process “Aluminium, primary, at plant/RER U” and “Disposal, red-mud from bauxite digestion, 0% water, to residual material landfill/CH U”

Arsenic exists in two main oxidation states: arsenic(III) and arsenic(V) (Grafe et al. 2001), that are not specified in the ecoinvent v2.2 dataset where it is called “arsenic ion”. However, according to the USEtox database (Rosenbaum et al. 2008), the two oxidation states have the same physico-chemical data.

Table C.3: Contribution to toxicity of primary aluminium production

	Aluminium, primary, at plant/RER U					**Disposal, redmud from bauxite digestion, 0% water, to residual material landfill/CH U**				
	Substance	**Compartment**		**Impact score**	**Unit**	**Substance**	**Compartment**		**Impact score**	**Unit**
Functional unit		**1 kg**					**1 kg**			
Toxicity cancer	Total of all compartments			3.36088E-06	CTUh	Total of all compartments			2.00115E-06	CTUh
	Remaining substances			4.03E-08	CTUh	Remaining substances			1.07E-08	CTUh
	ChromiumVI	Water	groundwater, long-term	2.52E-06	CTUh	ChromiumVI	Water	groundwater, long-term	1.51E-06	CTUh
	ChromiumVI	Water	river	6.96E-07	CTUh	ChromiumVI	Water	river	4.79E-07	CTUh
	ChromiumVI	Soil		1.00E-07	CTUh					
Toxicity non-cancer	Total of all compartments			2.64751E-06	CTUh	Total of all compartments			7.96976E-07	CTUh
	Remaining substances			1.25E-07	CTUh	Remaining substances			5.24E-09	CTUh
	Arsenic, ion	Water	river	1.15E-06	CTUh	Arsenic, ion	Water	river	7.92E-07	CTUh
	Arsenic, ion	Water	groundwater, long-term	7.27E-07	CTUh					
	Zinc	Air	high. pop.	1.76E-07	CTUh					
	Mercury	Air	low. pop.	1.61E-07	CTUh					
	Mercury	Air	high. pop.	6.97E-08	CTUh					

	Zinc	Air	low. pop.	6.30E-08	CTUh					
	Arsenic, ion	Water	groundwater	5.30E-08	CTUh					
	Lead	Air	low. pop.	4.83E-08	CTUh					
	Mercury	Air		4.45E-08	CTUh					
	Arsenic	Air	low. pop.	2.98E-08	CTUh					
Ecotoxicity	Total of all compartments			37.084368	CTUe	Total of all compartments			20.562342	CTUe
	Remaining substances			2.2857781	CTUe	Remaining substances			0.039480732	CTUe
	ChromiumVI	Water	groundwater, long-term	25.00202	CTUe	ChromiumVI	Water	groundwater, long-term	14.97586	CTUe
	ChromiumVI	Water	river	6.895605	CTUe	ChromiumVI	Water	river	4.7407686	CTUe
	Arsenic, ion	Water	river	1.1720861	CTUe	Arsenic, ion	Water	river	0.80623207	CTUe
	ChromiumVI	Soil		0.98889909	CTUe					
	Arsenic, ion	Water	groundwater, long-term	0.73997987	CTUe					

C.5 Coordinates and production of worldwide alumina production sites

Table C.4: Alumina production sites location

Name	Country	Latitude	Longitude	Alumina production in kt/year	Reference 1	Reference 2	Additional reference for production data	Additional reference for coordinates
Gladstone QAL	Australia	-23.866	151.290	3954	IAI	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	-
Gladstone Yarwun	Australia	-23.826	151.154	1400	IAI	-	http://sales.riotintoaluminium.com/freedom.aspx?pid=205	-
Gove	Australia	-13.069	135.703	3000	IAI	Alcor	-	-
Kwinana	Australia	-31.825	116.017	2200	IAI	Alcor	-	-
Pinjarra	Australia	-32.629	115.875	4200	IAI	Alcor	-	-
Wagerup	Australia	-32.908	115.898	2600	IAI	Alcor	-	-
Worsley	Australia	-33.308	116.006	3600	IAI	Alcor	-	-
Ganja	Azerbaijan	40.683	46.361	450	-	Alcor	-	http://en.wikipedia.org/wiki/Ganja,_Azerbaijan
Zvornic	Bosnia	44.419	19.110	600	-	Alcor	-	http://wikimapia.org/12413536/Alumina-Factory-Bira%C4%8D
Alumínio - SP	Brazil	-23.535	-47.261	920	IAI	Alcor	-	-
Barcarena	Brazil	-1.519	-48.617	6300	IAI	Alcor	-	-
Pocos de Caldas	Brazil	-21.788	-46.563	390	IAI	Alcor	-	-
Sao Luis	Brazil	-2.531	-44.307	3600	IAI	Alcor	-	-
Saramenha/Ouro Preto	Brazil	-20.386	-43.503	150	IAI	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	-
Vaudreuil (Jonquiere)	Canada	48.423	-71.243	1600	IAI	Alcor	-	-

Binzhou	China	38.088	117.727	1000	-	Alcor	-	http://lubeilvye.alu.cn/
Chiping	China	36.620	116.259	2000	-	Alcor	-	http://club.kdnet.net/dispbbs.asp?id=8995886&boardid=1
Dengfeng	China	34.386	113.186	400	-	Alcor	-	https://plus.google.com/100685232189069422582/about?gl=us&hl=en
Guiyang	China	26.691	106.668	1200	IAI	Alcor	-	http://www.chalco-gzfgs.com/
Hejin	China	35.651	110.671	2200	IAI	Alcor	-	http://www.sx.chalco.com.cn/sxweb/jsp/listAndView.jsp?ColumnID=3
Jiaokou	China	36.896	111.473	800	-	Alcor	-	https://plus.google.com/114576354773624279561/about?gl=us&hl=en
Jingxi	China	23.259	106.387	2400	-	Alcor	-	https://plus.google.com/113113383380473566881/about?gl=us&hl=en
Kaili	China	26.574	108.037	50	-	Alcor	-	https://plus.google.com/117004229960121443006/photos
Liaocheng	China	36.482	115.998	2400	IAI	Alcor	-	https://plus.google.com/101138781200094429621/about?gl=us&hl=en
Longkou	China	37.709	120.448	2400	-	Alcor	-	https://plus.google.com/108421795884736828549/about?hl=en
Mayi / Baise	China	23.388	106.599	1650	-	Alcor	-	https://plus.google.com/100462057856818094035/about?gl=us&hl=en
Nanchuan - Bosai	China	29.160	107.165	250	-	Alcor	-	https://plus.google.com/113637119505693573705/about?hl=en&gl=us&review=1
Nanchuan - Chalco	China	29.261	107.299	1600	-	Alcor	-	http://huagong.dxddcx.com/19017.htm
Pingdingshan	China	33.820	112.923	400	-	Alcor	-	http://www.1688.com/company/cjdn.html?fromSite=company_site&tab=companyWeb_contact
Pingguo	China	23.337	107.503	2500	IAI	Alcor	-	http://baike.baidu.com/view/20296.htm
Pinglu	China	34.826	111.276	800	-	Alcor	-	https://plus.google.com/110815237559030537050/about?gl=us&hl=en
Qingzhen	China	26.624	106.361	800	-	Alcor	-	https://plus.google.com/107679751352997882354/about?gl=us&hl=en
Sanmenxia - Easthope	China	34.804	111.798	1260	-	Alcor	-	https://plus.google.com/112733981361482672316/about?gl=us&hl=en
Sanmenxia - Kaiman -	China	34.720	111.056	1200	-	Alcor	-	https://plus.google.com/105811248076940074734/about?gl=us&hl=en

Sanmenxia - Yixiang	China	34.789	111.650	500	-	Alcor	-	http://yxlygs.cnal.com/
Wenshan	China	23.454	104.125	800	-	Alcor	-	http://www.ynwsly.net/contact/
Wulong	China	29.374	107.520	150	-	Alcor	-	http://www.cnpre.com/web/search/pre/index.php?modules=show&id=10041
Xiangjiang	China	34.720	112.075	1200	-	Alcor	-	http://www.heungkongwanji.com/wanji/qyjj/lxwm.html
Xiaoyi Huaqing	China	37.151	111.839	200	-	Alcor	-	http://huagong.dxddcx.com/43647.htm
Xiaoyi Xingan	China	37.087	111.850	1000	-	Alcor	-	https://plus.google.com/111281631664886368608/about?gl=us&hl=en
Yangquan	China	37.848	113.660	800	-	Alcor	-	https://plus.google.com/105673295277861199828/about?hl=en
Yuanping	China	38.756	112.675	2600	-	Alcor	-	http://www.mep.gov.cn/gkml/hbb/qt/201012/t20101224_199108.htm
Zhengzhou	China	34.720	113.788	2490	IAI	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	https://plus.google.com/104546525257852725848/about?hl=en
Zhengzhou - Chalco	China	34.720	113.788	2400	-	Alcor	-	https://plus.google.com/104546525257852725848/about?hl=en
Zhengzhou - Longshenxiang	China	34.720	113.788	100	-	Alcor	-	-
Zhongzhou	China	35.375	113.445	2200	IAI	Alcor	-	-
Zibo	China	36.755	118.050	1600	IAI	Alcor	-	-
Zouping	China	36.905	117.768	6000	-	Alcor	-	-
Zunyi	China	27.542	106.855	800	-	Alcor	-	-
Gardanne	France	43.454	5.469	630	IAI	Alcor	-	-
Stade	Germany	53.599	9.474	900	IAI	Alcor	-	-
Aghios Nikolaos	Greece	35.183	25.717	775	-	Alcor	-	http://www.mining-atlas.com/operation/Agios_Athanasios_Bauxite_Mine.php
Distomon (St-Nicolas/Agios Nikolaos)	Greece	38.333	22.833	830	IAI	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	-
Fria	Guinea	12.050	-10.933	640	-	Alcor	-	http://en.wikipedia.org/wiki/Fria

Ajka	Hungary	47.107	17.564	320	IAI	Alcor	-	-
Visakhapatnam, Andhra Pradesh	India	17.593	82.752	1500	-	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	http://en.wikipedia.org/wiki/List_of_alumina_refineries
Belgaum	India	15.850	74.505	380	IAI	Alcor	-	-
Damanjodi	India	21.032	82.837	2100	IAI	Alcor	-	-
Korba	India	22.349	82.698	205	IAI	-	http://en.wikipedia.org/wiki/Fria	-
Lanjigarh	India	19.708	83.368	1400	IAI	Alcor	-	-
Mettur	India	11.783	77.801	100	IAI	-	http://en.wikipedia.org/wiki/Fria	-
Muri Bihar	India	23.379	85.872	440	IAI	Alcor	-	-
Renukoot	India	24.200	83.030	700	IAI	Alcor	-	-
Utkal Alumina Project, Kashipur, Orissa	India	19.190	83.029	1500	-	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	http://en.wikipedia.org/wiki/List_of_alumina_refineries
Jajarm	Iran	36.95	56.38	280	-	Alcor	-	http://tools.wmflabs.org/geohack/geohack.php?pagename=Jajarm&params=36_57_00_N_56_22_48_E_type:city(25205)_region:IR
Aughinish	Ireland	52.669	-8.623	1927	IAI	Alcor	-	-
Porto Vesme	Italy	39.200	8.390	1000	IAI	Alcor	-	-
Claren-don/Woodside	Jamaica	17.971	-77.298	1420	IAI	Alcor	-	-
Ewarton	Jamaica	18.177	-77.088	650	IAI	Alcor	-	-
Kirkvine	Jamaica	18.081	-77.475	560	IAI	Alcor	-	-
Nain St Elizabeth	Jamaica	17.967	-77.608	1600	IAI	Alcor	-	-
Kikumoto	Japan	33.97206	133.28526	200	-	Alcor	-	http://postalcodedb.com/792-0801_Kikumoto_Japan.html
Shimizu	Japan	35.018	138.494	280	IAI	Alcor	-	-
Yokohama	Japan	35.444	139.638	250	-	Alcor	-	http://tools.wmflabs.org/geohack/geohack.php?pagename=Yokohama&params=35_26_39_N_139_38_17_E_type:city(3697894)_region:JP
Pavlodar	Kazakhstan	52.273	76.967	1600	IAI	Alcor	-	-

Podgorica	Montenegro	42.460	19.260	0	IAI	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	-
Tulcea	Romania	45.175	28.803	580	-	Alcor	-	http://tools.wmflabs.org/geohack/geohack.php?language=fr&pagename=Tulcea&params=45.175_N_28.802778_E_type:city_region:ro_scale:100000_globe:earth&title=
Oradea	Romania	47.072	21.921	400	-	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	http://tools.wmflabs.org/geohack/geohack.php?language=fr&pagename=Oradea&params=47.07222_N_21.92111_E_type:city_region:ro_scale:100000_globe:earth&title=
Kamensk-Uralsk	Russia	56.400	61.933	730	-	Alcor	-	http://tools.wmflabs.org/geohack/geohack.php?pagename=Kamensk-Uralsky&params=56_24_N_61_56_E_region:RU-SVE_type:city(174,689)
Achinsk	Russian Federation	56.282	90.517	1100	IAI	Alcor	-	-
Bogoslovsk	Russian Federation	55.790	70.349	1100	IAI	Alcor	-	-
Boksitogorsk	Russian Federation	59.469	33.860	120	IAI	Alcor	-	-
Pikalevo	Russian Federation	59.513	34.177	268	IAI	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	-
Uralsky	Russian Federation	56.426	61.908	750	IAI	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	-
Volkhovskiy	Russian Federation	58.578	31.334	400	IAI	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	-
Mopko	South Korea	34.767	126.350	180	-	Alcor	-	http://tools.wmflabs.org/geohack/geohack.php?language=fr&pagename=Mok-po&params=34.767_N_126.35_E_type:city_region:kr_globe:earth&title=
San Ciprian	Spain	42.852	-7.915	1500	IAI	Alcor	-	-
Slovalco	Slovakia	48.563	18.848	180	-	-	http://en.wikipedia.org/wiki/List_of_alumina_refineries	http://en.wikipedia.org/wiki/List_of_alumina_refineries
Paranam	Suriname	5.613	-55.099	2200	IAI	Alcor	-	-
Seydisehir	Turkey	37.419	31.845	230	IAI	Alcor	-	-

Nikolaev	Ukraine	46.958	32.018	1600	IAI	Alcor	-	-
Zaporozhye	Ukraine	47.227	35.592	230	IAI	Alcor	-	-
Burnside	United States of America	30.139	-90.924	540	IAI	Alcor	-	-
Corpus Christi	United States of America	27.802	-97.392	1600	IAI	Alcor	-	-
Gramercy	United States of America	30.061	-90.698	1200	IAI	Alcor	-	-
Point Comfort	United States of America	28.679	-96.558	2300	IAI	Alcor	-	-
Ciudad Guayana/Puerto Ordaz	Venezuela (Bolivarian Republic of)	8.210	-62.697	2000	IAI	Alcor	-	-
Ho Chi Minh City	Vietnam	10.767	106.667	12	-	Alcor	-	http://tools.wmflabs.org/geohack/geohack.php?language=fr&pagename=H%C3%B4-Chi-Minh-Ville&params=10.767_N_106.667_E_type:city_region:vn_globe:earth&title=
Tan Rai	Vietnam	11.633	107.833	650	-	Alcor	-	http://www.maplandia.com/vietnam/lam-dong/bao-loc/xa-tan-rai/

C.6 Fate and intake fraction of selected substances in water in each 0.5°*0.5° grid cell modeled on a global resolution

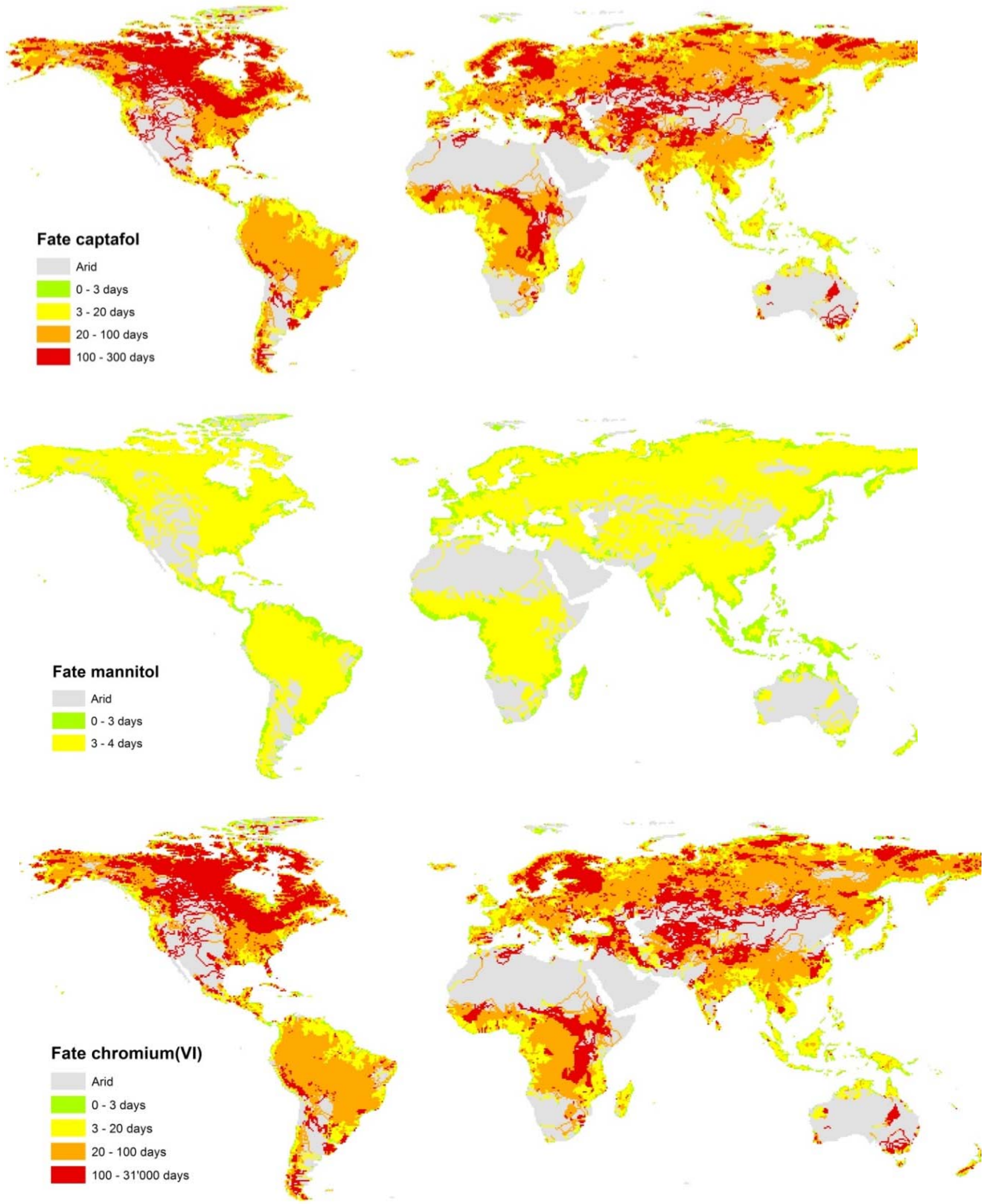
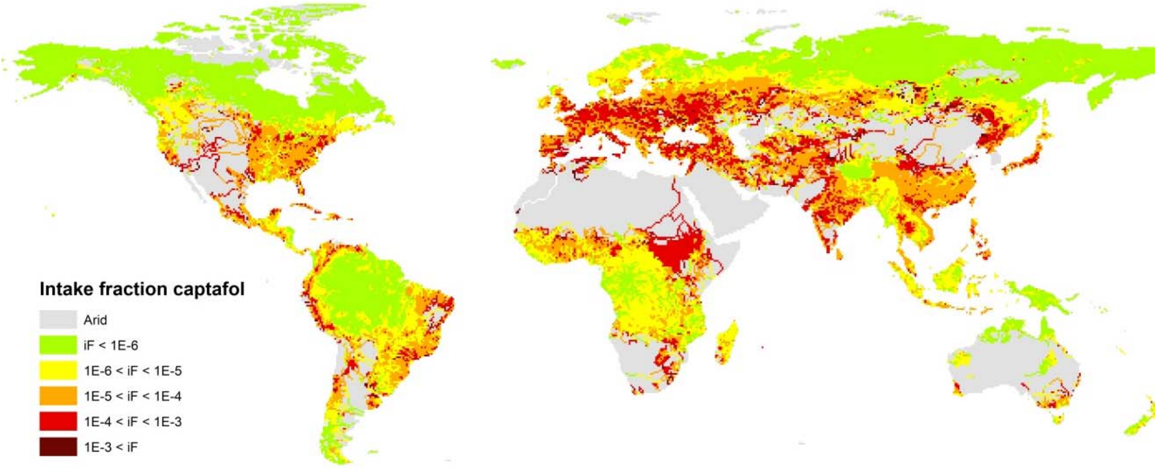
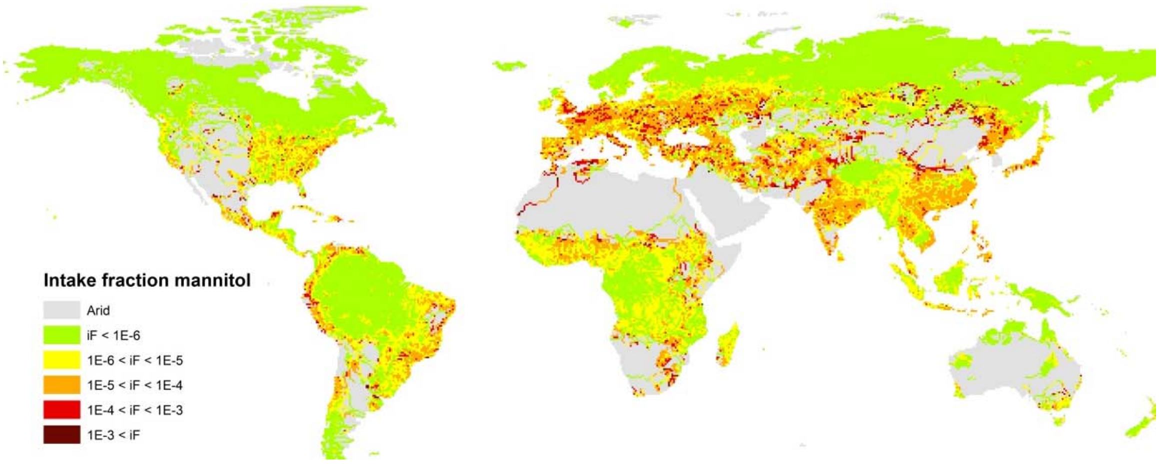


Figure C.3: Fate of selected substances in water in each 0.5°*0.5° grid cell modeled on a global resolution





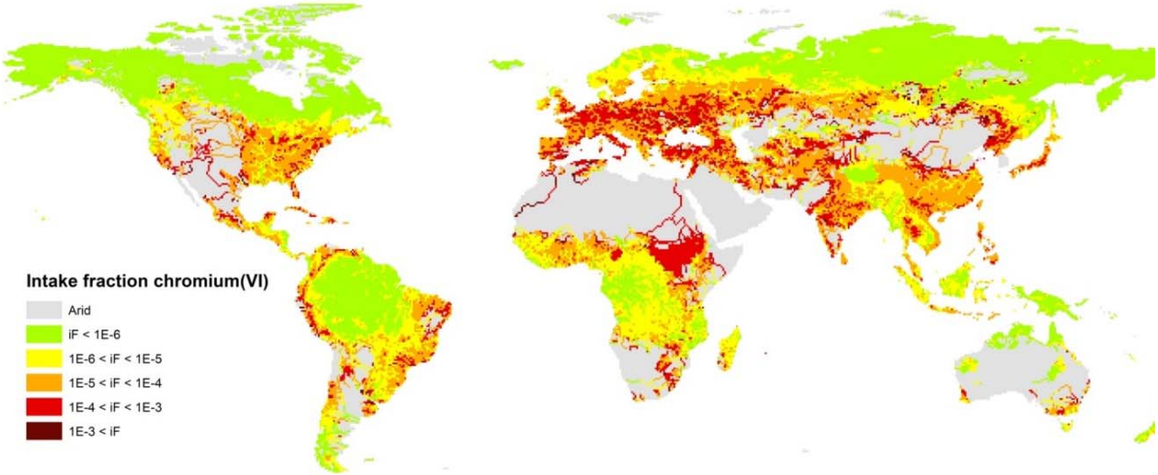


Figure C.4: Intake fraction of selected substances in water in each 0.5°*0.5° grid cell modeled on a global resolution

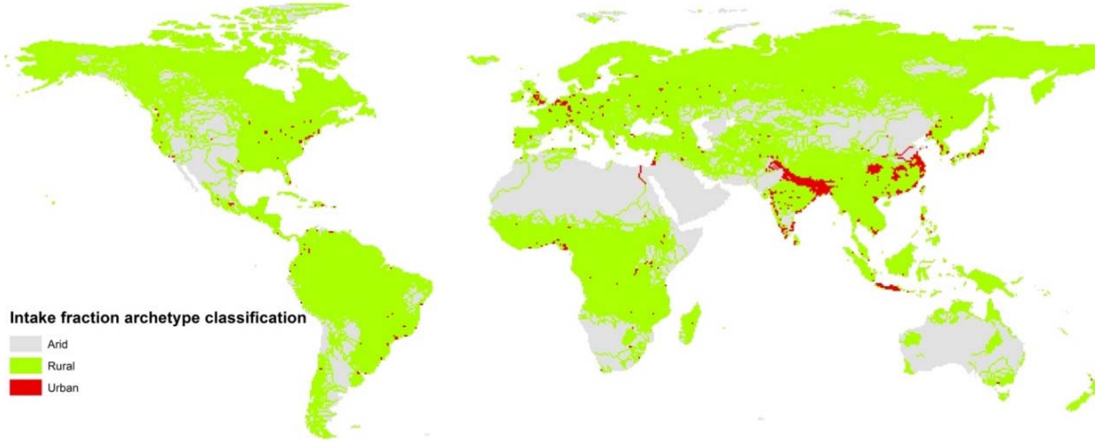


Figure C.5: Intake fraction archetype classification in each 0.5°*0.5° grid cell modeled on a global resolution

C.7 Fate variability complementary analysis

Figure C.6 shows the variation of chemical fate depending on water depth. Only chromiumVI, arsenic and tinopal show a correlation with water depth. The fate of tinopal follows the inverse of its evaporation and sedimentation rate in cells with depth greater than 1 m, i.e. cells that include or are dominated by lake sedimentation process. Captafol and mannitol are not influenced given that their disappearance pathways are respectively driven by advection and degradation rather than sedimentation.

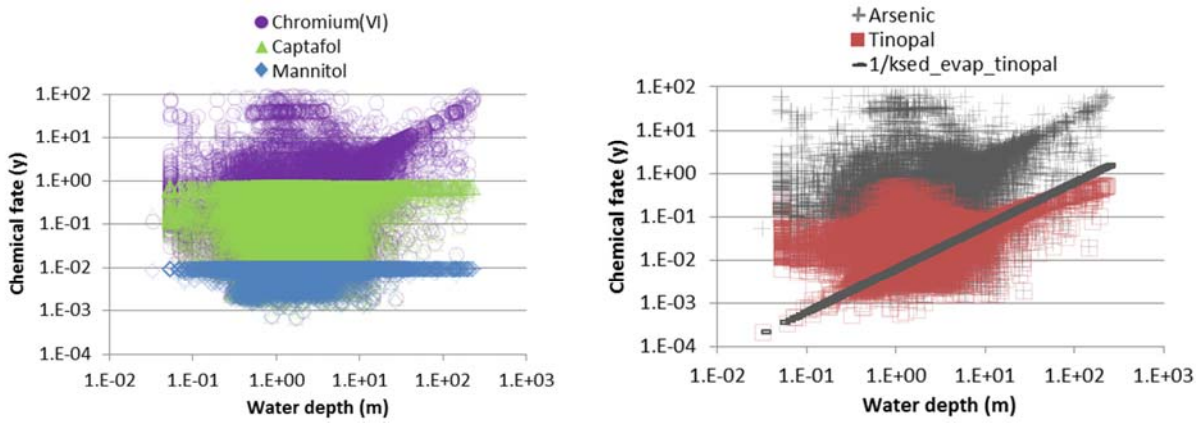


Figure C.6: Fate of five selected chemicals (mannitol, chromiumVI, captafol, arsenic and tinopal) vs. water depth in each 0.5°*0.5° grid cell of the model

C.8 Test of equivalent depth

Figure C.7 shows the variation of fate versus equivalent water depth of tinopal for tinopal, captafol, mannitol, chromiumVI and arsenic. The fate of tinopal is proportional to its equivalent depth within two orders of magnitude.

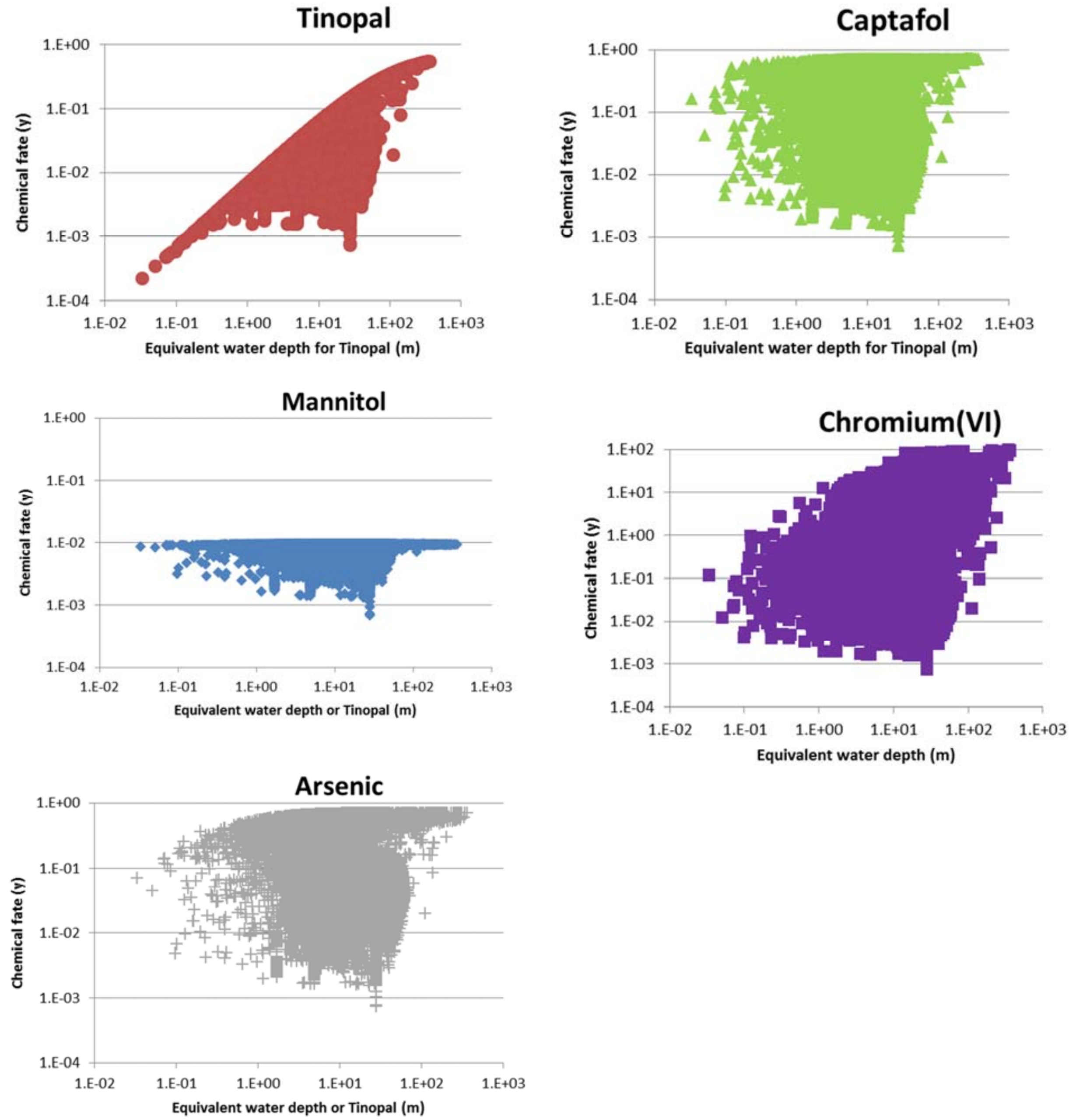


Figure C.7: Fate of 5 selected chemicals (Tinopal, Folpet, ZnII and PbII) not totally driven by water residence time vs. equivalent water depth for each specific pollutant in each 0.5°*0.5° grid cell

We recalculated the fate of all selected substances based on equation 8 using $d_{i\ equ}$ of tinopal and $k_{adv,\ sea\ i}$ for each cell to analyse whether it is can be used as a generic proxy for fate estimation.

Figure C.8 shows the fate of selected substances recalculated with $d_{i\ equ}$ of Tinopal. The variation with the fate modelled on a 0.5°*0.5° resolution is less than one order of magnitude for all substances.

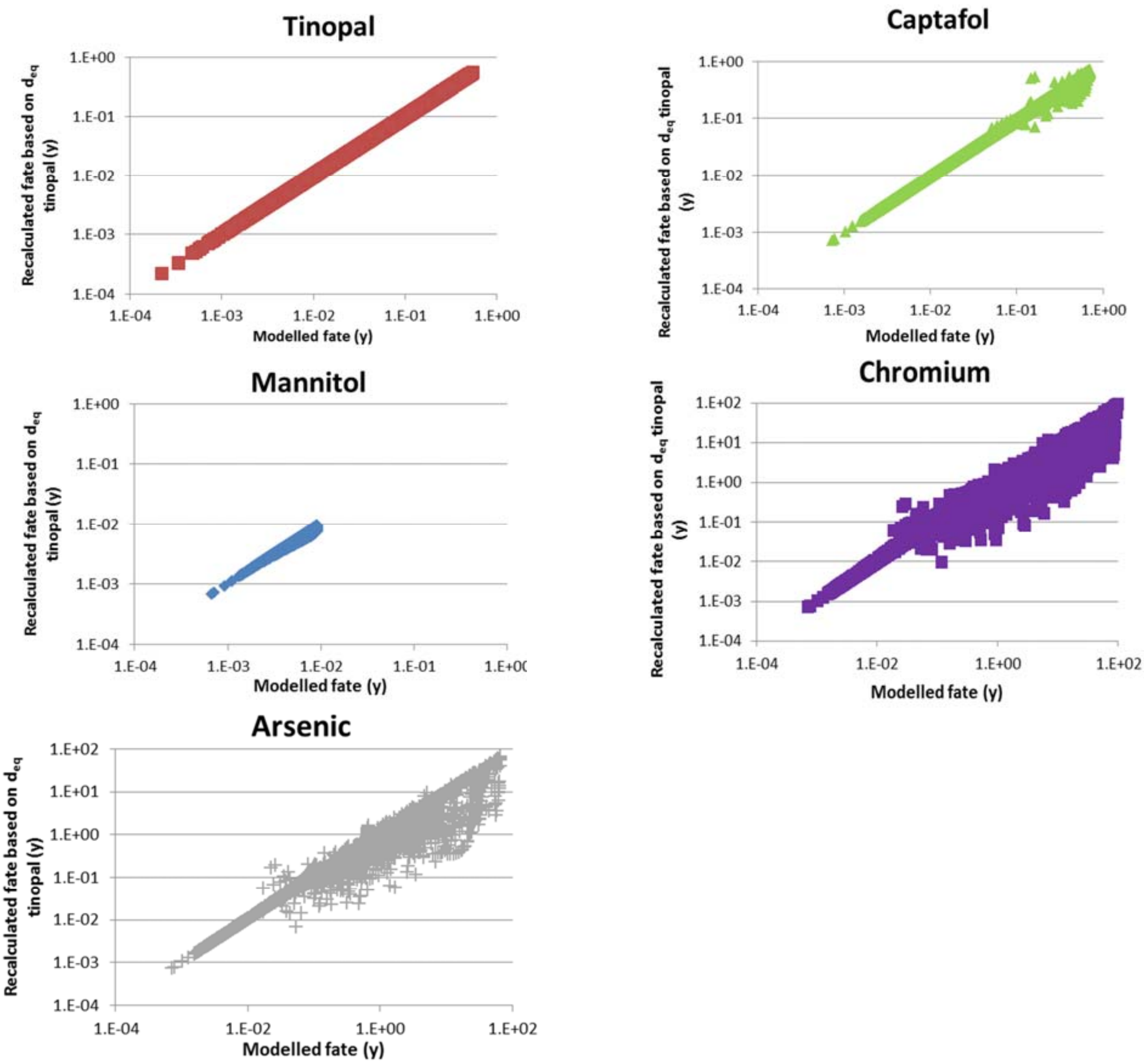
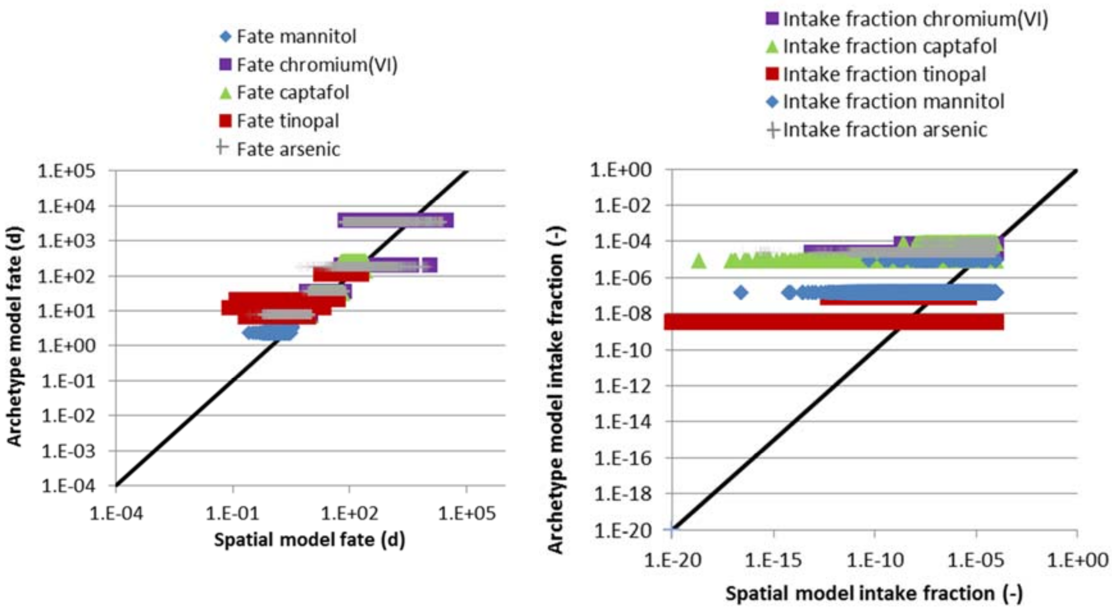


Figure C.8: Fate of 6 selected chemicals (tinopal, captafol, mannitol, chromiumVI) calculated with the model vs. fate recalculated based on equivalent depth of tinopal in each 0.5°*0.5° grid cell

C.9 Test of archetype performance

Figure C.9.a shows the comparison between the archetype model fate and the spatial results on a 0.5°*0.5° resolution. Results are within 2 orders of magnitude between archetype and spatial fate. The fate of tinopal for archetypes A1, A2 and A3 varies between 6 and 20 days while in A4 it is 109 days. Given that these archetypes do not follow a systematic regional pattern such as urban or rural landscapes, the practitioner needs to know the geographical location of an emission to deduce to which archetype category it can be attributed. This approach thus does not bring an important simplification from a data collection point of view, while it keeps up to two additional orders of magnitude variability compared to a 0.5°*0.5° approach.

Figure C.9.b presents an analysis of the correlation of the spatial model intake fraction with the rural and urban archetype two box model. It shows that there are up to 10 orders of magnitude difference between the two models for the rural area, while up to 3 for the urban landscape. The archetype model results are within the same order of magnitude for rural and urban landscapes for persistence substances such as chromiumVI and arsenic. This is due to the fact that urban and rural landscapes are hydrologically strongly connected, with 91% of freshwater advected from rural to urban landscapes and 60% in the other direction. Persistent substances can thus pass through both compartments if their fate exceeds the compartment residence time, i.e. 760 days for the rural box and 29 days for the urban one. These observations support the conclusion that the relevance of this model is limited.



a) **b)**

Figure C.9: Fate (a) and intake fraction (b) of the five selected substances modeled with (a) the four archetype model based on water residence time and equivalent depth vs. the spatial model and (b) the urban and rural archetype model

C.10 Intake fraction of arsenic vs arsenic emissions for alumina refinery

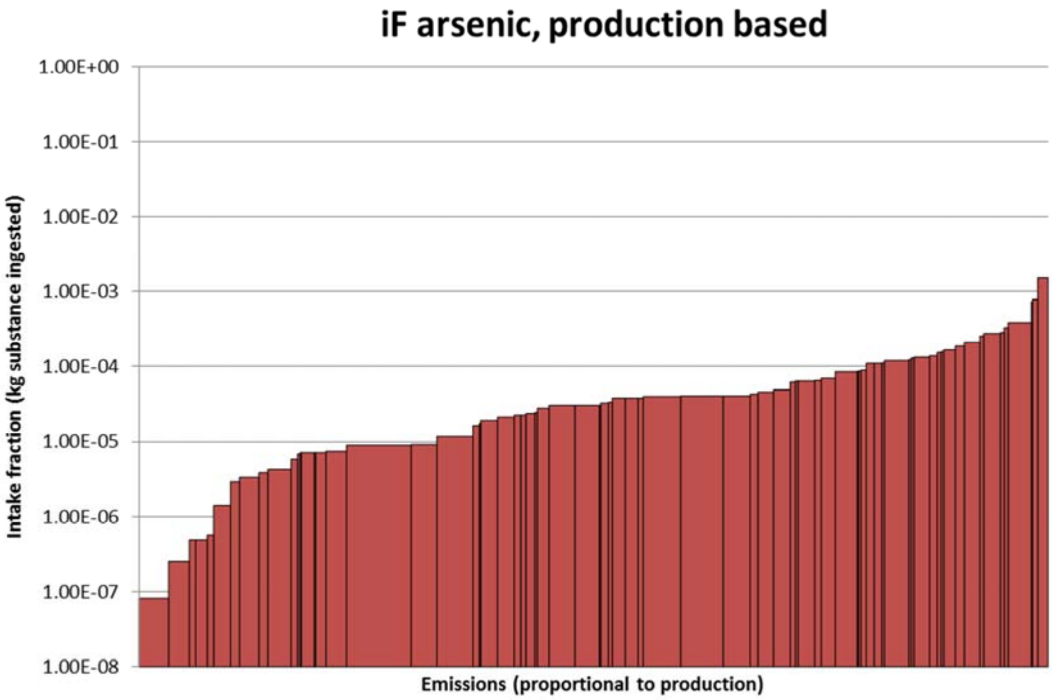


Figure C.10: Intake fraction of arsenic vs relative emissions for all alumina refinery sites

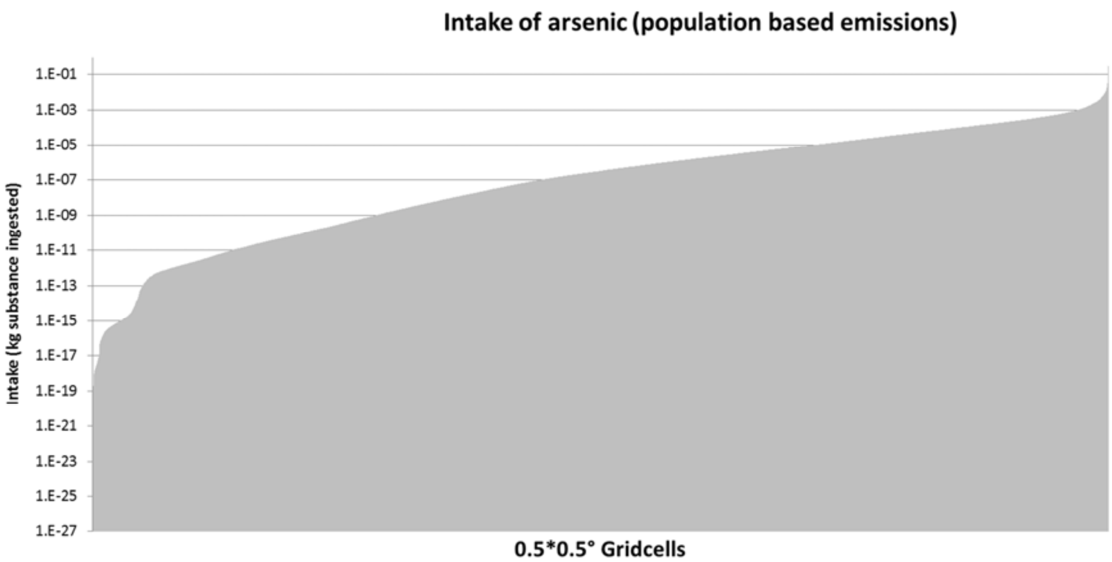


Figure C.11: Intake fraction for all 0.5*0.5° grid cells of the model

C.11 Analysis of production- and population-weighted aggregations at the country, continental, generic and archetype level for two alumina refinery sites

FigureC.12 presents the map of arsenic fate and alumina industries worldwide, that produce up to 6300 $kt_{alumina}$ per year. We analysed the fate and intake fraction values for the alumina refineries in Boksitogorsk (RUS) and Achinsk (RUS).

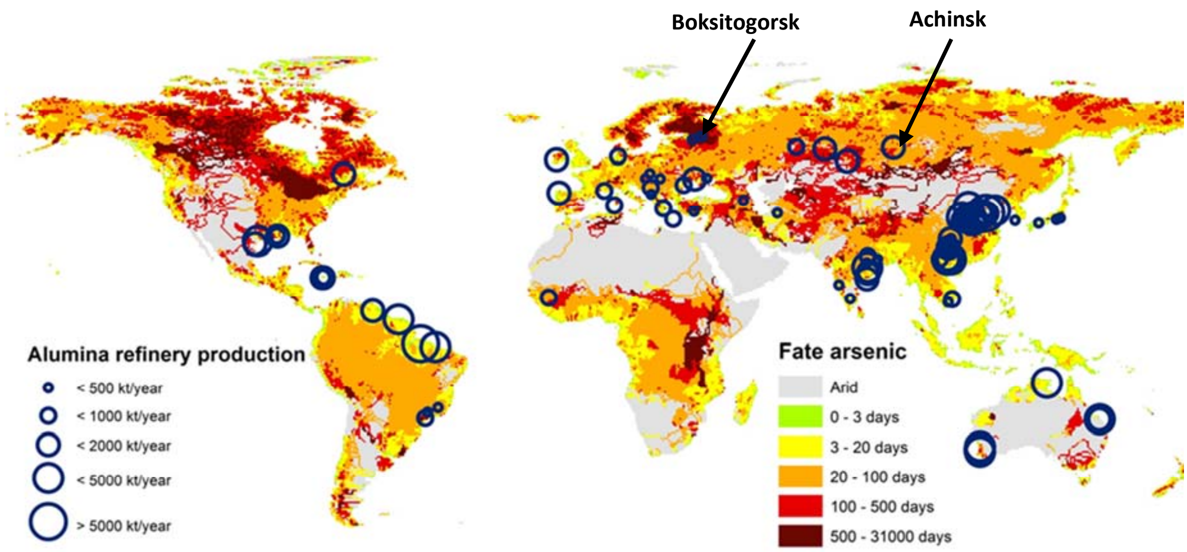


Figure C.12: Aluminium refineries sites and arsenic fate at a 0.5°*0.5° grid cell resolution

Figure C.13.a and b show the fate of arsenic for these three refineries calculated with (a) a production weighted and (b) a population weighted aggregation at a country, archetype, continental and generic level compared to the fate on a 0.5°*0.5° grid cell. While the archetype value always shows less than one order of magnitude difference with the grid cell scale result for all fate results, the country, continent and generic value can have a larger under- or overestimation of grid results. For example the production weighted results show that the fate grid cell value 5.3 years for Boksitogorsk production is underestimated by a factor 0.03 in the case of generic value and the Achinsk value 0.21 year is overestimated by a factor 8 in the case of the country value. Production weighted aggregation do not show a clear advantage compared to population weighted values in the case of these specific refineries. For single sites, only the archetype value is systematically relevant for the fate estimation.

Figure C.13.c and d show the intake fraction of arsenic for the same sites. There is more than 1 orders of magnitude of difference between the value on the country scale and the 0.5°*0.5° grid cell Boksitogorsk results.

From this analysis, a simplified regionalization approach does not seem appropriate to estimate the fate or intake fraction of single sites. Indeed, keeping the regionalized value seems preferable.

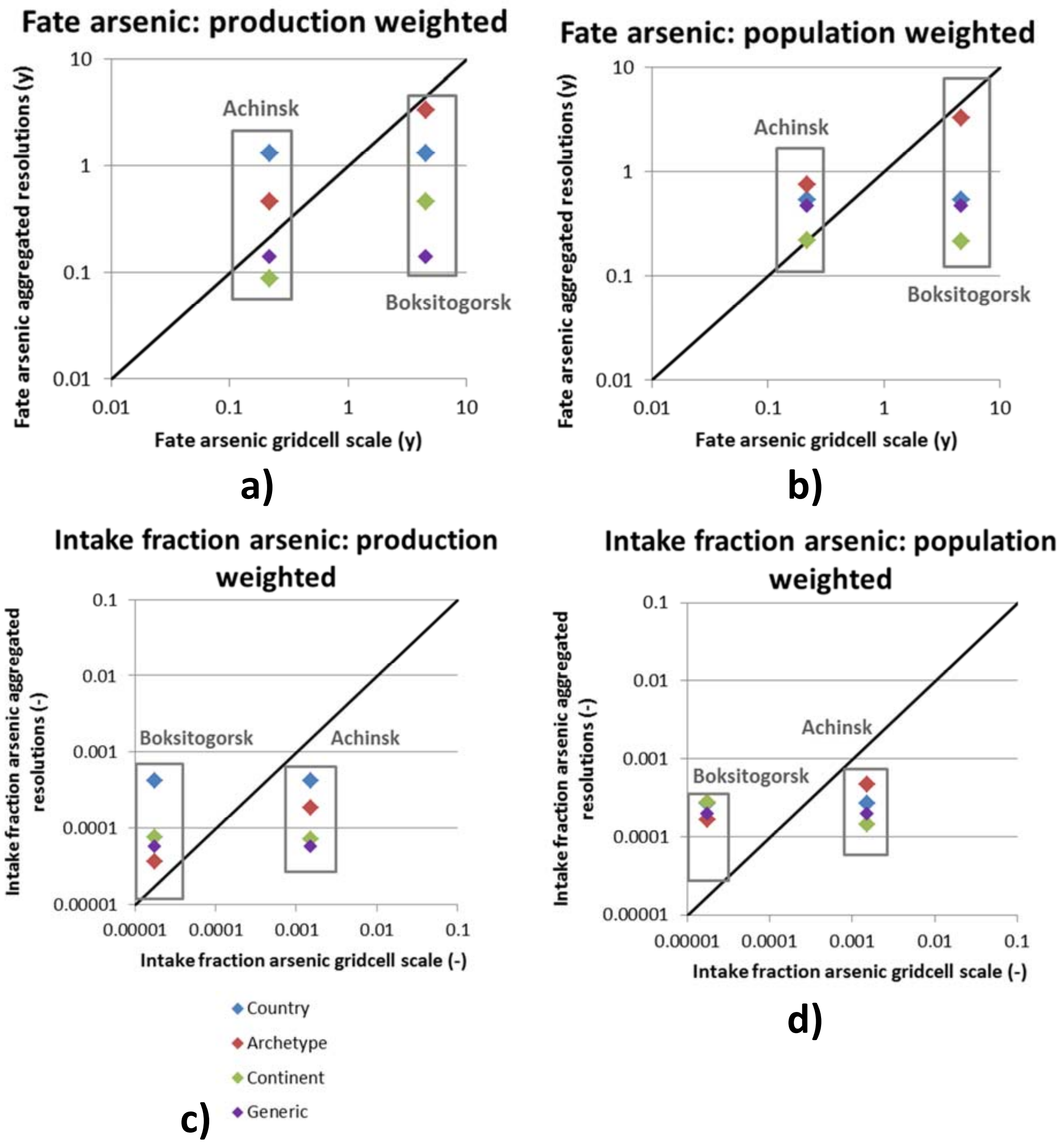


Figure C.13: Comparison of the fate (a and b) and intake fraction (c and d) of arsenic at the Achinsk and Boksitogorsk (Russia) alumina refinery modelled at the 0.5°*0.5° grid cell resolution with the country, archetype, continent and generic scale with a production weighted (a and c) and population weighted (b and d) aggregation

Acknowledgements

I would like to express all my gratitude to Prof. Manuele Margni who introduced me to the world of LCA research back in 2009 during my master thesis and who took the challenge to supervise my Phd thesis from the other end of the Atlantic Ocean. Thanks for his constant scientific support, optimism and for conveying an open-minded and dynamic vision of academic research, which favours human interation, and is strongly bonded with industry and real-case LCA applications. Many thanks also for the early timezone-dependent wake-ups! My second unofficial but nonetheless essential supervisor was Prof. Olivier Jolliet, who provided me substantial guidance and scientific support for the work on toxic emissions into water. Up to five-hour-long over-sea discussions gave me a unique dive in the world of toxic impact modelling and into quality academic research. Thanks for his patience, scientific enthusiasm and for sharing some of his great knowledge in this field. A special thanks to Prof. Philippe Thalmann, who gave me the opportunity to perform my thesis in EPFL under the atypical combination of consulting work in the private field and academic research, for being encouraging and very responsive to make sure my research was on the right track.

This work took place physically in the private company Quantis, where I had the opportunity to work in the field of LCA consulting. I would first like to thank Dr. Yves Loerincik and Dr. Sebastien Humbert for creating this special opportunity and allowing me flexible working hours. Many thanks to all my colleagues in Lausanne and in other Quantis offices, for their great expertise, the stimulating exchange of ideas, philosophico-technico-environmental discussions and moral support during my thesis. Also for the great times at lunch, drinks, running and climbing sessions, it is a real pleasure to work in this atmosphere!

Thank you to all researchers I interacted with during this 4 year-period: 1) Prof. Cécile Bulle, Prof. Andrew Henderson, Dr. Cédric Wannaz, Dr. Shanna Shaked who shared with me their experience in LCIA and were of great assistance in particular with IMPACTWorld, Matlab and ArcGIS, 2) the WULCA group who provided me substantial feedback on the review of water methods and helped me compiling my first article and (3) the QSA group in DTU who hosted me for overall 3 months during the Toxtrain project where I got insight on the academic environment.

To end, many thanks to my friends and family for their constant support, my ten-year-long flatmate Arianna for sharing a great part of this journey and my beloved life companion Kunal for making these years so fun, enjoyable and adventurous.

Lausanne, September 2014

Curriculum Vitae

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- Nationality: Russian
- Languages: French and Russian (native), English (fluent)

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Education

Since 2010	Ecole Polytechnique Fédérale de Lausanne (EPFL) / Switzerland Ph.D. thesis in Life Cycle Assessment *Thesis title*: Water use and quality in Life Cycle Assessment: identifying good practices and developing operational spatial approaches *Director and co-director*: Prof. Philippe Thalmann and Prof. Manuele Margni
2004-2010	Ecole Polytechnique Fédérale de Lausanne (EPFL) / Switzerland Bachelor and master in Environmental Science and Engineering
2009-2010	Ecole Polytechnique de Montreal/CIRAIG / Canada Master project on business risks related to water
2007-2008	Indian Institute of Engineering, Chennai / India Exchange year in Environmental Engineering

Work experience

Since 2010	Quantis / Switzerland Life Cycle Analyst, trainer in the topic of water footprinting and (eco-)toxicity assessment using USEtox Several projects on water footprinting and LCA using multi-indicators Coordinator of the review of methods addressing water use for the UNEP-SETAC Life Cycle Initiative Participation in European FP7 funded projects Participation to the drafting of the ISO norm 14046 on water footprint, including acting as secretary during the plenary meeting of working group 8 in charge of it in Léon, Mexico, June 2010

List of projects (non-exhaustive)

2013-to present	DEMEAU (Demonstration and exploitation of most promising prototypes and tools derived from European research activities): European FP7 funded project Life cycle assessment of the ozonation system for micropollutant removal in Neugut, Switzerland (on-going)
2013-to present	Nanovalid (Development of reference methods for hazard identification, risk assessment and LCA of engineered nanomaterials): European FP7 funded project Development of a life cycle impact assessment method for the human toxicity and ecotoxicity impact pathway of nanoparticles (on-going)
2011-to present	Toxtrain (Development and implementation of a tool box to assess toxicological impacts related to the life-cycle of technologies) Dissemination of the (eco-)toxicity impact category in LCA through USEtox courses
2010-2013	LC-IMPACT (Life Cycle Impact assessment Methods for imProved sustAinability Characterisation of Technologies): European FP7 funded project Development of regionalized characterization factors for the human toxicity, ecotoxicity and acidification impact categories
2013	Confidential client: consulting project Excel tool development on total cost of ownership from a life-cycle perspective
2012-2013	Alliance for beverage carton and the environment (ACE): consulting project Water footprint inventory evaluation
2012	Confidential client: consulting project

	A review of methods and existing data to evaluate the water footprint of a kWh
2012	Unilever: consulting project Water footprint of a laundry product
2011-2012	European Aluminium Foil Association: consulting project Scientific article on the importance of considering product loss rates in life cycle assessment: the example of closure systems for bottled wine
2010-2011	Procter & Gamble: consulting project Water footprint of a hand dishwashing product

Scientific publications and conference presentations

Publications included in this thesis

- Kounina A, Margni M, Hendersen A, Wannaz C, Jolliet O (in preparation) Spatial analysis of toxic emissions to freshwater in LCA: operationalization at global scale
- Kounina A, Margni M, Shaked S, Bulle C, Jolliet O (2014) Spatial analysis of toxic emissions in LCA: a sub-continental nested USEtox model with freshwater archetypes. Environment International 69:67-89
- Kounina A, Bayart J-B, Boulay A-M, Berger M, Bulle C, Frischknecht R, Koehler A, Margni M, Mila-i-Canals L, Motoshita M, Nunez M, Peters G, Pfister S, Ridoutt B, Van Zelm R, Verones F, Humbert S (2012) Review of methods addressing freshwater use in life cycle inventory and impact assessment. The International Journal of Life Cycle Assessment 18:707-721

Additional publications

- Kounina A, Tatti E, Humbert S, Pfister R, Pike A, Ménard JF, Loerincik Y, Jolliet O (2012) The importance of considering product loss rates in Life Cycle Assessment: the example of closure systems for bottled wine. Sustainability 4:2673-2706
- Van Hoof G, Buyle B, Kounina A, Humbert S (2013) Life cycle based water assessment of a hand dishwashing product: Opportunities and limitations. Integrated Environmental Assessment Management. 9:633-644
- Sevigné Itoiz E, Fantke P, Juraske R, Kounina A, Antón Vallejo (2012) Deposition and residues of azoxystrobin and imidacloprid on greenhouse lettuce with implications for human consumption. Chemosphere 9:1034-1041

Platform and poster presentations

- Getting USEtox out of the lab. Platform presentation at SETAC EU 2014: Basel, Switzerland, May 14th 2014
- Spatial differentiation for toxic emissions in LCA: paths toward the operationalization at global scale. Poster corner at SETAC EU 2014. Basel, Switzerland, May 14th 2014
- Spatial analysis of toxic emissions in LCA: A sub-continental nested USEtox model with freshwater archetypes. Platform presentation at SETAC EU 2013: Glasgow, United Kingdom, May 14th 2013
- Application of IMPACT World+ uncertainty assessment method on aquatic ecotoxicity impact category. Platform presentation the LC-IMPACT workshop on uncertainty: Zurich, Switzerland, January 20th 2012
- The importance of considering loss rates in LCAs: an example of bottled wine closure systems. Poster at LCM 2011: Berlin, Germany, August 28th -31th 2011
- Review of methods addressing freshwater use in life cycle inventory and impact assessment. Platform presentation at LCA IX 2009: Boston, United States of America, October 1st 2009

Trainings provided on water footprint and toxicity

- FHNW, Basel, Switzerland, February 2014: USEtox toxicity model
- Malaysian palm oil research institute, Lausanne, Switzerland, February 2014: Water footprint
- HEIG-VD, Yverdon, Switzerland, December 2013: Water footprint
- LCM conference, Göteborg, Sweden, September 2013: USEtox toxicity model
- Lausanne, Switzerland, July 2013: Water footprint
- Bern university of applied sciences, Bern, Switzerland, March 2013: Water footprint
- Paris, France, February 2013: Water footprint
- HEIG-VD, Yverdon, Switzerland, January 2013: Water footprint
- Novartis, Basel, Switzerland, November 2012: Water footprint
- University of Padova, Padova, Italy, October 2012: Water footprint
- Lausanne, Switzerland, October 2012: Water footprint
- Danish Technical University, Lyngby, Danemark, June 2012: Water footprint and the Quantis SUITE 2.0 software
- Paris, France, February 2012: Water footprint