Comparing priority setting in integrated hazardous substance assessment and in life cycle impact assessment

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Abstract

Purpose The purpose of the study was to compare three recent Life Cycle Impact Assessment (LCIA) models in prioritizing substances and products from national emission inventories. The focus was on ecotoxic and human toxic impacts. The aim was to test model output against expert judgment on chemical risk assessment.

Materials and methods An emission inventory was collected for Finland describing the year 2005. The inventory included publicly reported emissions to air and water and it was complemented by the emissions of tributyltin, benzene, and pesticides from research papers and statistics. The emissions were characterized with three LCIA models: IMPACT 2002+, ReCiPe, and USEtox and priority substances were identified. The results were connected to an environmentally extended input–output model to study priority products and supply chains. A comparison was made with two integrated assessments of the chemical status and human exposure in the Baltic region.

Results and discussion The three assessed models presented very different priorities. In ecotoxicity, IMPACT2002+ and USEtox highlighted heavy metals while ReCiPe focused on tributyltin. The integrated assessment identified both groups. In human toxicity, IMPACT2002+ and the integrated assessment focused on organic air pollutants while USEtox and ReCiPe identified mainly metals.

Conclusions LCIA models can be used for priority setting in chemical emission control and consumption based analyses. However the models give differing prioritizations so care must be taken in model selection. The studied models differed from expert assessment mostly in substances which are bioaccumulative. Further studies in including bioaccumulation to LCIA models of toxic impact are recommended.

Keywords Ecotoxic impact assessment · Emission inventory · Environmentally extended input–output analysis · Human toxic potential · Priority substance · Structural path analysis

1 Introduction

Life Cycle Impact Assessment (LCIA) of hazardous substances has progressed to the level, where the models used are fully comparable with those used in chemicals risk assessment (MacLeod et al. 2010; Rosenbaum et al. 2008).

The models are based on fugacity modeling, which has a long history in environmental chemistry and generally good correspondence with environmental monitoring (MacLeod et al. 2010). In the most recent models, the toxicological effect factors are based on statistical analysis of empirical measurements and population disease occurrences (Rosenbaum et al. 2008). In comparison, the integrated assessment of chemical risks in the environment and chemical management in environmental policy rely strongly on measured concentrations and agreed regulatory limits (e.g., EC 2004; EVIRA 2010; HELCOM 2010). The borderline between risk assessment and risk management is often blurred (Assmuth and Jalonen 2005).

LCA has been used more and more together with environmentally extended input–output analysis (EEIO; Suh 2009). In that context, LCIA models are used to analyze entire national economies with the aim of identi-

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fying the main sources of environmental impact in the production–consumption structures (UNEP 2010; EPA 2009). When entire national emission inventories are analyzed with LCIA, it offers an opportunity to test LCIA model output against other methods, especially national chemical integrated assessments and priority settings. This serves as a kind of a model validation, since ideally the prioritization from LCIA models should highlight the same issues as expert assessment (EVIRA 2010; HELCOM 2010) and perhaps point out targets for further enquiries.

In this study, we applied three recent LCIA models to an environmentally extended input–output analysis of the toxic emissions of Finland. The aim of this study was twofold: (1) to identify the main pathways causing ecotoxic and human toxic impacts in Finland and (2) test the applicability of impact assessment models against integrated assessments that were based on period from 1999 to 2007 (HELCOM 2010) and from 2002 to 2009 (EVIRA 2010).

2 Material and methods

2.1 Emission inventory and life cycle impact assessment

The emission inventory was based on public data sources and the emission registry of the Finnish Environmental Administration (VAHTI -database). The database includes reported toxic air and water emissions for industrial sites and estimated emissions for households, service production, and agriculture. In the Environmental Impacts of Material Flows Caused by the Finnish Economy (ENVIMAT) project, each site was connected to an industrial classification code to get the industry total emissions. The inventory included air emissions of arsenic, cadmium, cobalt, chromium, copper, mercury, nickel, lead, zinc, vanadium, PAH compounds, dioxins, and furans and hydrogen fluoride. Benzene emissions were estimated separately by combining fuel use (Statistics Finland 2006) and benzene emission factors (Pietarila et al. 2002). Since the exact distribution of PAH compounds was unknown and only the total was reported, it was assumed that the distribution would be similar to that estimated for EU (Sleeswijk et al. 2008).Water emissions of arsenic, cadmium, cobalt, chromium (III and VI), copper, mercury, nickel, lead, antimony, tin, vanadium, zinc, phenols, toluene, and vinyl chloride were included from the VAHTI database. Tributyltin emissions from ships were included based on expert assessment (Ministry of the Environment 2006).

Pesticide emissions to agricultural soil were included by using the sales statistics of 34 herbicides, insecticides, and growth regulators. Although detailed models for estimating air and water emissions from application rates were available (Birkved and Hauschild 2006), they were not applied. Instead the fate factors of the characterization models were used to transform the application rates (emission to soil) into water concentrations and human exposure. This was deliberately done to increase variability between characterization models and is an approach similar to that used in calculating recent LCIA normalization factors (Sleeswijk et al. 2008).

Three life cycle impact assessment (LCIA) methods were applied to characterize the emissions: IMPACT 2002+, ReCiPe, and USEtox. Only the freshwater ecotoxicological and human toxic midpoint impact categories were included in the comparison. All three models use fugacity-based multimedia environmental fate models to estimate environmental concentrations and exposure, but differ in the parameterization and detail of the models used. IMPACT2002+ is based on a spatially detailed multimedia-model of West Europe (Jolliet et al. 2003). ReCiPe 2008 (Goedkoop et al. 2009) uses a fugacity model based on the European System for the Evaluation of Substances; however, with better soil and air compartmentalization and other improvements (Huijbregts et al. 2005). USEtox is a consensus model, developed by an UNEP-SETAC group on toxic impact assessment (Rosenbaum et al. 2008). In USEtox, seven LCIA models were calibrated together and the simplified consensus model was used to produce characterization factors, which were recommended by the developers of the original LCIA methods.

Since the three models presented indicators in different units, the results were normalized for comparison. The normalization was done by calculating a reference impact based on European emissions in the year 2000 (Sleeswijk et al. 2008). In order to avoid bias in normalization (Heijungs et al. 2007), reference emissions were included only for substances, which were also present in the national inventory (e.g., emissions of atrazine were not included in the reference or in the national inventory, but emissions of copper were included in both).

2.2 Input–output model and analysis

Environmentally, EEIO was used to interpret the results of the emission inventory and to identify the main economic interactions causing the emissions. EEIO extends the input–output tables of the national accounts with the emission intensity per industry and with a characterization model (Suh and Huppes 2005). In this study, the ENVIMAT EEIO model with 150 sector resolution (Seppälä et al. 2009) was used, but only toxic emissions to air, water, and agricultural soil were included. The model equation was then:

$$q = CF(I - A)^{-1}f \qquad (1)$$

where q is the indicator result for human toxicity and ecotoxicity [kilogram reference substance], C is the matrix of characterization factors [kilogram reference substance/

kilogram] (2-by-62 matrix), F is the emission intensity matrix [kilogram per M€](62-by-150 matrix), I is the identity matrix, A is the input–output coefficient matrix [M€/M€ in basic prices] (150-by-150 matrix) and f is the final demand vector [M€] (150-by-1 vector).

Characteristics of the EEIO model were studied with structural path analysis (SPA). It is a method for extracting individual flows from the whole EEIO system described by Eq. 1 (Lenzen 2003). It can be used to identify the main economic interactions which contribute most to the studied overall environmental impact. SPA is commonly used in hybrid LCA to simplify input–output-based results and to focus the collection of more detailed emission inventories (Lenzen and Crawford 2009). Simplification is necessary to interpret the results, since the amount of economic interactions increases exponentially when more layers of supply chain are included in the analysis. For example, following the interactions of 150 industries for three production layers includes more than three million economic pathways. However, usually only a few dozen pathways cover the most of the environmental impact (Lenzen 2003). The SPA algorithm starts with the final demand supplied to consumption, investments, and export. It then follows the supply chain backwards until toxic emissions are encountered. The emissions are characterized and the path from final demand to the emission source is stored in a list. The list is finally sorted and the most important pathways are identified for further analysis.

3 Results and discussion

3.1 Comparing substance priority in LCIA and Baltic Sea integrated assessment for ecotoxicity

Both IMPACT2002+ and USEtox identified copper and zinc emissions to water and air causing a major part of ecotoxic impacts. In addition, USEtox identified vanadium air emissions as a priority and IMPACT2002+ highlighted also nickel emissions to air and water. In ReCiPe however, most of the ecotoxic potential was caused by water emissions of organic substances, especially tributyltin from ships (Fig. 1). Overall, the normalized results expressed as a share of the toxic pressure from European emissions varied over four orders of magnitude between models (0.5% in ReCiPe, 1.4% in IMPACT2002+, and 2.1% in USEtox). The small result in ReCiPe was caused by a small share of tributyltin (TBT) compounds in Finland compared to European emissions. If TBT was ignored, ReCiPe had similar results to the other models (i.e., 2.0% of European toxic impact).

The difference between ReCiPe and the two other models was caused by differences in the ecotoxic potential
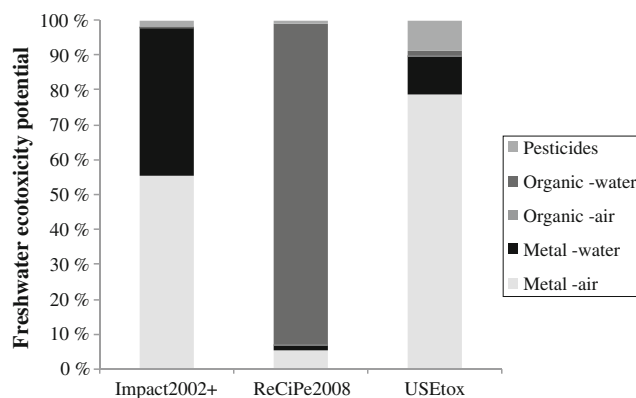

Fig. 1 Comparison of the share of toxic load from substance groups in the three assessed LCIA models

of TBT compounds. Although their use has been banned since 2003, emissions from leaching ship paints were still estimated to be 4,000 kg in 2005 (Ministry of the Environment 2006). In the latest integrated assessment of the Baltic Sea, TBT compounds were identified as a source of high concern, since their observed concentrations in biota exceeded quality limits in most parts of the Baltic (HELCOM 2010). Impact2002+ did not include TBT, but in ReCiPe, it was the main pollutant, amounting to 92% of the ecotoxic pressure. Using USEtox, TBT amounted to only 1.4% of the ecotoxic pressure. The difference between the impact models is caused to a large extent by the different chemical properties for TBT in USEtox and ReCiPe. The half-life of TBT in water is 9 days in the USEtox database, while in the ReCiPe database it is 128 days (Huijbregts et al. 2005; Hauschild et al. 2010). This reflects the considerable variability in the measured experimental degradation rates (ECHA 2008). As a consequence, the fate factor of TBT is significantly lower in USEtox than in ReCiPe. If USEtox were used in national prioritization of ecotoxic impacts, the importance of TBT would be ignored and a focus would be on controlling air emissions of heavy metals.

The share of agricultural pesticides varied considerably between models. In IMPACT2002+ pesticide emissions caused 1.7% of the ecotoxic impact mainly through the use of dimethoate, glyphosate, prochloraz, and propiconazole. In ReCiPe, impacts from pesticide emissions were only 0.6% of the total, caused mainly by linuron and dimethoate. In USEtox pesticide emissions caused 9% of the ecotoxic impact and caused by a broad scale of pesticides, but mainly prochloraz, mancozeb, and linuron. Therefore both the overall share and substance prioritization of agriculture varied between models used. The results of this study are in contrast with the recent materials and products prioritization studies, where agriculture was found to cause 82% of freshwater ecotoxic impact (UNEP 2010; EPA 2009). However this result was obtained using only one set of

characterization factors and due to the extreme variability between models cannot be considered to be reliable. In further prioritization studies model intercomparison is recommended to test the robustness of results.

In the development of normalization factors for the year 2000 using ReCiPe (Sleeswijk et al. 2008), pesticides were found to cause 24–30% of freshwater ecotoxic impacts. The impacts were mainly caused by atrazine use, which has been banned in the EU since 2004 (Sass and Colangelo 2006). This highlights the importance of using recent emission inventories in priority setting. Without atrazine emissions the share of pesticides would have been 9–20% of total, which is comparable to the results of this study.

The LCA included emissions to air, water, and soil and evaluated the effects over time and space. In contrast, the integrated assessment was based on measured concentrations and quality targets in the Baltic. The two approaches cannot be directly compared, but can be seen as complementary. In the integrated assessment PCBs, lead, mercury, cesium-137, DDT/DDE, TBT, benz[a]anthracene, cadmium, and dioxins/furans were identified as substances of high concern (HELCOM 2010). The prioritization was done based on the proximity of observed concentrations to environmental quality limits. In contrast to current best practices in chemical risk management, the limits were set mainly based on human exposure, and not on ecotoxicological dose–response data. This was done as a precautionary approach to protect consumers from secondary poisoning (HELCOM 2010). Of the highlighted substances, TBT was the only compound identified as a source of concern. PCBs, DDT/DDE, and cesium-137 were not identified as they have no current emissions and the observed concentrations are caused by chemical residence in sediments. Lead, mercury, and cadmium are bioaccumulative heavy metals (Hendriks and Heikens 2001). The current LCIA models do not include ecotoxicity from secondary poisoning through food web bioaccumulation, which results in the underestimation of the toxic potential of food web accumulative substances. To some extent, this explains also the lack of benz[a]anthracene and dioxins/furans in the LCIA results, while they are a source of concern in the integrated assessment (HELCOM 2010). Basing the toxicity endpoints to critical body residues instead of dissolved concentrations would possibly make the results of LCIA more closely comparable to those of chemicals risk management in general.

3.2 Human toxicity potential

Compared to the several orders of magnitude of difference in estimating ecotoxic impacts, the models estimated quite similar human toxicities (Fig. 2). The share of Finnish emissions of the European reference emissions ranged
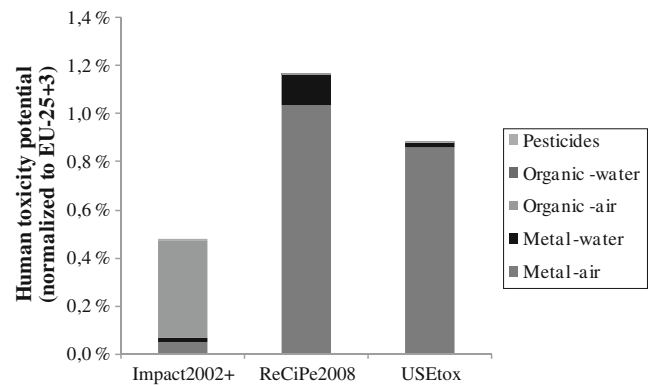
Fig. 2 Human toxicity potential of Finnish emissions in 2005 estimated with three life cycle impact assessment (LCIA) models. Impacts were reported as a share of the European reference emissions

between 0.5% and 1.2%. The main difference between models is the toxicity of polycyclic aromatic hydrocarbons (PAHs), dioxins/furans, and benzene. Dioxins, PAHs, and benzene amounted to 85% of the human toxic potential in IMPACT2002+, but less than 0.1% in ReCiPe and USETOX. Based on national air-quality assessments, PAH exposure is a cause for concern, since atmospheric concentrations of benzo[a]pyrene regularly exceed the regulatory limits (Alaviippola et al. 2007). The concentrations are the highest during wintertime and are caused by the incomplete combustion of wood. Also the exposure from food is at a high level (EVIRA 2010). In comparison, benzene levels are generally below regulatory limits (Pietarila et al. 2002). The results from ReCiPe and USEtox concerning PAHs were clearly in contradiction with the integrated assessments, while IMPACT2002+ captured the effect better.

The consumption of Baltic fish has been regulated due to high concentrations of dioxins and furans (EVIRA 2010), which have well known emission estimates, but lack characterization factors in USEtox. In IMPACT2002+ and ReCiPe, dioxins (2,3,7,8-TCDD) had the highest reported characterization factor for human toxicity, but dioxins were highlighted only using IMPACT2002+. The main exposure route to humans is through Baltic fish. Therefore it may be that the fugacity and food web models applied in the USEtox and ReCiPe do not represent the Baltic ecosystem and foodweb in sufficient detail. This could be tested by developing site-specific characterization factors for the Baltic, using the Popcycling Baltic model with foodweb components (Mattila and Verta 2008). As such, the IMPACT2002+ model represents the chemical risk management results for organics better than USEtox or ReCiPe.

Metal emissions dominated the results from ReCiPe and USEtox. Both models highlighted zinc, mercury, lead, arsenic, and cadmium, but ReCiPe identified vanadium as an additional priority pollutant. In IMPACT2002+ only arsenic zinc were identified. Cadmium, mercury, and

Table 1 The main economic pathways in the Finnish 2005 input–output table causing freshwater aquatic ecotoxicity calculated with IMPACT2002+, ReCiPe, and USEtox LCIA models

Emission	IMPACT2002+ industries	Share of impact
Copper–water	Non-ferrous metals	11.2%
Copper–air	Households	9.0%
Zinc–water	Artificial fibers	6.6%
Copper–air	Non-ferrous metals	4.2%
Zinc–air	Households	2.4%
Emission	**ReCiPe Industries**	**Share of impact**
TBT–water	Shipping	69.6%
TBT–water	Shipping → retail trade	3.5%
TBT–water	Shipping → pulp and paper	1.8%
TBT–water	Shipping → residential construction	1.1%
Vanadium–air	Oil refining	0.8%
Emission	**USEtox Industries**	**Share of impact**
Vanadium–air	Oil refining	13.3%
Zinc–air	Households	4.5%
Zinc–water	Artificial fibers	4.2%
Vanadium–air	Households	1.3%
TBT–water	Shipping	1.1%

arsenic were also identified as sources of concern in the recent integrated assessment on chemical exposure in Finland, since their uptake is close to the tolerable weekly intakes (EVIRA 2010). Zinc and vanadium were not included in the assessment and lead concentrations were found to be decreasing. Overall the results of the LCIA models are found to be in agreement with the integrated assessment in respect to heavy metals.

The integrated assessment included several substance groups, which were not included in the LCIA models. Nitrates and fungal toxins are included in food safety, but no characterization factors exist for converting agricultural

practices into human exposure. Polybrominated diphenyl ethers, perfluorooctanesulfonic acid, and diethylhexylphthalates are measured from food (EVIRA 2010), but they lack LCIA characterization factors and emission estimates.

3.3 Priority products and main emission pathways

Applying input–output analysis and structural path analysis to the emission inventory allowed a consumption-based perspective on the emissions of hazardous substances. This exercise could be used to identify the priority products, which have the highest embodied emissions in their supply

Table 2 The main structural paths in the Finnish input–output table causing human toxic impacts calculated with IMPACT2002+, ReCiPe, and USEtox LCIA models

Emission	IMPACT2002+ Industries	Share of impact
PAH–air	Households	19.1%
Benzene–air	Households	6.3%
Dioxins and furans–air	Electricity	5.6%
Dioxins and furans–air	Non-ferrous metals	3.7%
Dioxins and furans–air	Electricity → renting apartments	3.6%
Emission	**ReCiPe Industries**	**Share of impact**
Vanadium–air	Oil refining	8.2%
Arsenic–air	Non-ferrous metals	3.7%
Arsenic–air	Oil refining	3.4%
Mercury–air	Iron and steel manufacturing	2.6%
Arsenic–water	Non ferrous metals	2.6%
Emission	**USEtox Industries**	**Share of impact**
Zinc–air	Households	14.6%
Mercury–air	Iron and steel manufacturing	6.4%
Zinc–air	Non-ferrous metals	2.8%
Mercury–air	Pulp and paper industry	2.6%
Zinc–air	Iron and steel manufacturing	2.3%

chain (UNEP 2010). These would then be priority candidates for policy actions.

The models yielded overlapping results. IMPACT2002+ and USEtox highlighted zinc emissions from artificial fiber manufacture and household fuel use. Both USEtox and ReCiPe also identified vanadium from oil refining. IMPACT2002+ also identified copper emissions from metal industry and households. In contrast to other models, ReCiPe highlighted the importance of TBT from shipping, consumed either directly or through purchases of retail trade, pulp and paper, or residential construction.

Compared to ReCiPe the USEtox put less emphasis to TBT emissions from shipping. Vanadium emissions from oil refining are considered as the main priority, followed by zinc and vanadium emissions from domestic fuel use and zinc water emissions from artificial fiber production. Overall, the focus is moved from shipping to petrochemical manufacture and use (Table 1).

Compared to ecotoxic priority setting, the models had more differences in the human toxic priority pathways. IMPACT2002+ highlighted the main emission sources of PAHs, benzene, and dioxins, identifying among others the electricity use in apartments as a key pathway. The other two models focused more on direct emissions of zinc, mercury, and arsenic from oil refining, metals industry and pulp and paper industry (Table 2).

All models could be used to identify a set of priority products and pathways. However the prioritization differed considerably as did the evenness of pathways. Five top ranking pathways covered 70% of the ecotoxic impact in ReCiPe, 33% in IMPACT2002+, and only 24% in USEtox. Compared to earlier work using structural path analysis (Lenzen 2003), the identified paths were very short, indicating that toxic emissions are mainly released in the final stages of the supply chain. Using USEtox for policy recommendations would then result in a broader scope of measures, while based on the two other method a focus on few key pollutant sources would be recommended. In human toxicity, completely opposite focus would be obtained using IMPACT2002+ or the two other models, with IMPACT2002+ focusing on household energy use and the others controlling on heavy metal emissions from industry.

4 Conclusions

Based on the results, LCIA models can be used for priority setting in chemical emission control and consumption-based analyses. However, careful selection of the model is advised since the models provide very differing prioritizations. For ecotoxicity, ReCiPe provided the prioritization most consistent with the integrated assessment. For human toxicity, IMPACT2002+ provided the priorities most

similar to integrated assessment. A comparison of characterization model output to expert assessment is therefore recommended in further prioritization studies. Since the studied models differed from expert assessment mostly in substances which are bioaccumulative, further studies to include bioaccumulation to LCIA models is recommended.

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