Workshop on Environmental and Human Toxicity of metals in LCA:
Status, limitations and new developments

3-4 September 2014

Final report

Background

Metal emissions often come out as strong contributors to the indicator results for the impact categories covering eco- and human toxicity in LCA, and the appropriateness of the characterization models for these impact categories for metals has been contested. This was the background of the Montreal workshop organized in 2002 (Dubreuil, 2002) and of the Apeldoorn workshop organized in 2004 (Ligthart et al., 2004) under the auspices of the UNEP-SETAC Life Cycle Initiative with funding from the ICMM to discuss the current practices and complications of Life Cycle Impact Assessment (LCIA) methodologies for non-ferrous metals and agree on recommendations for development of the practice in the field. In the Clearwater consensus workshop, organized under the Life Cycle Initiative in 2009, experts in characterization modelling and metal risk assessment agreed on more detailed recommendations for good characterization modelling practice for ecotoxic impacts of metals in freshwater systems (Diamond et al., 2010). The recommendations were meant to inspire the development of the metal assessment in the UNEP-SETAC scientific consensus model USEtox® which was developed in an international collaboration of LCIA and chemical assessment experts over the years 2006-2010.

Recently, USEtox® model and characterization factors have entered the product policy context through the European Commission communication published in April 2013 on "*Building the Single Market for Green Products*… COM/2013/0196". This communication recommends using the Product Environmental Footprint methodology which includes USEtox® as recommended model to assess toxicity aspects of products.

Considering this potential use in a product policy context, the metal industry organized a scientific workshop to clarify the status and appropriateness of USEtox® for assessing the toxicity of metals. This workshop took place at the Leuven University, Belgium (Katholieke Universiteit Leuven) on 3 and 4 Sept 2014. It was attended by more than 40 invited participants composed of metal industry experts as well as USEtox® developers and users.

This document summarizes the main discussions and conclusions issued from that workshop.

Introductory session

After a welcome introduction by Jo Van Caneghem on behalf of KULeuven, Staf Laget presented, on behalf of Eurometaux, the context of toxicity assessment in LCA as well as the general objective of the workshop as:

- Communications on the state of the art of LCA methodologies for predicting or incorporating metal toxicity considerations, focusing on the USEtox® methodology.
- Identifying potential issues related to input data and methodologies for modelling eco-toxicity and human toxicity; identifying possible improvements

- Assessing progress in the LCIA eco-toxicity field since the "Apeldoorn Declaration (2004)[1]" and the "Clearwater Consensus (2009) [2]"
- Obtaining consensus regarding progress, limitations and research needs from academic and industrial practitioners in risk assessment and LCIA, e.g. from acknowledged USEtox® methodology experts.
- Reinforcing a constructive dialogue and synergies between these two communities, i.e. risk assessment and LCIA, to facilitate further developments

1. LCA vs. risk assessment: similarities and differences (Frank Van Assche)

Risk assessment is used worldwide to predict the risk a substance may pose in the environment. On the other hand, LCA toxicology impact categories aim to compare average impacts from emissions, rather than predict the effects of substance emissions individually in the environment.

However there are important similarities between both approaches, for instance, the use of the same data sources; or the use of factors to model the effects, i.e. characterization factor (CFs) in the case of LCA, and Risk Characterization ratios, in the risk assessment case. Final goal, while different among both approaches, has also similarities, such as to assess the potential effects of the substance in the receiving environment.

One core difference among the two types of approaches is the way regionalization is addressed. Risk assessment methodology integrates the characteristics of one specific local receiving environment to assess the potential impact of the substance on this local environment, while LCIA starts to consider local specificities but on a global scale and thus typically with less resolution (due to supply chains being global with their emissions occurring all over the planet). Regional consideration is of key importance for metals, since their bioavailability is highly depending on local conditions like water composition.

In addition, essentiality and deficiency are considered in Risk assessment and should be addressed as well in the LCA context. Currently, in LCIA, any metal emission is considered to result in a negative impact even when emissions of essential metals result in concentrations that are within their window of essentiality (for human health and the environment) Such "optimal range" concentrations are regulated by the organisms and contribute to their optimal growth and development. The "any emission is negative" principle is applied even if metal deficiencies occur. It has been proposed that emissions of essential metals shall be considered as negative impact in LCA only above emission levels which generate concentrations exceeding the upper boundary concentration of their window of essentiality.

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[1] See Apeldoorn declaration (http://media.leidenuniv.nl/legacy/declaration_of_apeldoorn.pdf)
[2] The clearwater consensus: the estimation of metal hazard in fresh water, Miriam L. Diamond & Nilima Gandhi & William J. Adams & John Atherton & Satyendra P. Bhavsar & Cécile Bulle & Peter G. C. Campbell & Alain Dubreuil & Anne Fairbrother & Kevin Farley & Andrew Green & Jeroen Guinee & Michael Z. Hauschild & Mark A. J. Huijbregts & Sébastien Humbert & Karen S. Jensen & Olivier Jolliet & Manuele Margni & James C. McGeer & Willie J. G. M. Peijnenbu g & Ralph K. Rosenbaum & Dik van de Meent & Martina G. Vijver, Int (2010), Int J Life Cycle Assess (2010) 15:143–147

2. Ecotoxicity in LCA: From Apeldoorn to 2014: where are we? (Katrien Delbeke and Ralph Rosenbaum)

Already in 2004, experts in the field of risk assessment and life cycle assessment met to discuss the issue of eco-toxicity of metals in the LCA context, which resulted in the Apeldoorn declaration.

The basis of this collaboration was to integrate the risk assessment knowledge into the LCA methods to provide a more meaningful result in ecotoxicity assessment, while recognizing that LCA was designed to compare different products and systems, and not to predict the actual risks associated with single substances.

At that time, the core weaknesses of toxicity assessment of metals effects in the environment were identified as the following:

- Speciation: Determines bioavailability and toxicity and is a highly desirable extension of LCIA, both in fate and effect modeling
- Persistence: Infinite time horizons in steady state effect models could only be appropriate if bio-availability is properly considered
- Essentiality: Within the essentiality window of essential metals the LCA general principle of “less is better” does not apply and the possibility of adverse biological effects should be set at zero
- Bioavailability: The Biotic Ligand Model (BLM) should be used preferentially, with the Free-Ion Activity Model (FIAM) used in cases where the BLM has not been fully developed.
- Effect characterisation: The effect factor should be chosen at the HC50 (geometric mean of EC50) level rather than the HC5 or the NOEC level.
- Compartments in the multimedia model: If effects are negligible in a given effect compartment, there is no need to consider effects. This may well be the case for essential metals in the ocean.
- Spatial aspects: The consequences of regional differences in bioavailability, background concentrations, and therefore toxicity need to be further elaborated

Since then, researchers have addressed most of the above issues, proposing a way forward to improve the metal characterization factors, by addressing speciation, compartment sections, spatial aspects, HC50-based effect characterization and bioavailability. Some issues still require further consideration regarding their actual importance:

Persistence: The residence time of freshwater in USEtox® is <100 days and a few years in coastal water. This essentially leads to metal accumulation in the ocean compartment, which does not result in freshwater (or coastal marine water) ecotoxicity impacts attributable to the influence of the choice of time horizon. The latter is hence only of importance in the context of terrestrial and deep-ocean ecotoxicity, both not yet mature enough and thus not implemented in USEtox®.

Essentiality/Compartments in the multimedia model: While its relevance for freshwater ecosystems remains to be studied, this is important to consider for terrestrial and marine ecotoxicity (both not yet mature enough/implemented in USEtox®).

Consideration of long-term (thousands of years) emissions and impacts from landfills and mine tailings is an ongoing discussion and research subject in the LCA community, thus the importance to differentiate effect within e.g. 100 years from longer term potential effects.

3. Human health and human toxicity in LCA: challenges and progresses (Scott Baker and Olivier Jolliet)

Scott Baker explained that the use of USEtox® to assess human health impact for metals has a shorter history of collaboration between human health experts and LCA experts compared to the situation for ecotoxicity.

The main challenges identified when assessing human health are the following:

- Include as appropriate quantitative expressions of all critical health endpoints (non cancer NOAEL, cancer risk probability) as identified in the hazard identification
- Toxicity analysis for life cycle application and risk assessment must be comprehensive
- Must consider dose-response and exposure.
- It may distinguish the risk in between individuals, population, global scope, regional.

Olivier Jolliet showed that human health impact are important to consider in Life Cycle Impact Assessment, since pollution is responsible for more than 6 million annual deaths worldwide according to the global burden of disease study from 2010. Substantial progress has been made in the Life Cycle Impact Assessment of metals on human health since the 2004 Apeldoorn workshop. Intake fractions (the fraction of an air emission taken in by the population) are now available for atmospheric emission in every city of the world.

4. Reference database: going for more harmonization (Ruth Danzeisen by teleconference)

One of the main weaknesses in metal characterization identified prior the workshop is that several new data sources are not used to determine characterization factors. During the workshop, Ruth Danzeisen presented criteria to assess the quality and reliability of data bases, such as:

- Time scale of the data, for instance, including the latest development in research
- Origin of the research, assuring that the data comprises different research groups, and in the case of industrial emissions, inclusion of industry based data.
- When comparing chemicals, it is of core importance to agree on common testing methods and procedures. Thus, studies should be consistent in group of organisms, trophic levels and data collection and processing methods.
- When possible only include peer-review of data by expert panels

To better address the questions posed during the introduction section, two different parallel sessions were held, the human health session and the environmental toxicity session.

Ecotoxicity Session

In the ecotoxicity session, an in-depth discussion of relevant aspects for metals in freshwater systems concerning the latest developments in USEtox® and the relation between LCA and Risk Assessment took place. The discussion mainly focused on the harmonization, quality, and transparency of data sets, background concentrations, essentiality, bioavailability, spatial variability, and the applicability of the existing USEtox® model for metals.

Two presentations helped to steer the discussion: *Latest developments in USEtox®* (by Nilima Gandhi), and *Comparison between LCA and Risk Assessment* (by Frederik Verdonck)

Gandhi[3] presented the new method of calculating characterization factors for metals. This method incorporates metal speciation/complexation into fate calculations by loosely coupling a commercial geochemical metal speciation model, such as WHAM, with the multimedia fate model, USEtox®. Another major improvement was to incorporate a biotic uptake model, such as Biotic Ligand Model (BLM) or Free Ion Activity Model (FIAM), to calculate the bioavailability-corrected adverse toxic effects of metals (Effect Factor). The latter is especially important for metals since the bioavailability and toxic effects of metal vary as per metal speciation, which is controlled by the environmental chemistry. These improvements are mainly guided by the Clearwater Consensus (Diamond et al. 2010). The new method has been applied to calculate revised CFs for several cationic metals (Gandhi et al. 2010, Dong et al. 2014). Next, Gandhi showed the use of various archetypes to consider regional aspects in the ecotoxicity assessment of metals. This new approach shows that it is possible to include local or regional background concentrations and bioavailability factors. This approach has been considered as very relevant for the various metals addressed and it has been recommended to continue research work to apply this method to other locations, other metals as well as for considering essentiality aspects.

Verdonck flagged a confusion regarding the relationship between USEtox® and the AIIDA[4] data base which uses the USEtox® model to calculate characterization factors. Indeed AIIDA is not always consistent and up-to-date. It was made clear that AIIDA is not part of the official USEtox® and that the calculated characterization factors are provided by a consultant who applies USEtox® as part of his business but they are not officially endorsed,. The calculation methodology of characterization factors were discussed considering that USEtox® metal CFs are mainly built up on HC 50 of EC 50s (preferring chronic data but also using some acute values where chronic data were not available) while most research focuses on EC10 values (chronic). The way extrapolation is applied was discussed. It was agreed that USEtox® should use adequate extrapolation factors for each substance, attending to the differences among components. It was also mentioned that the way extrapolation factors are calculated in USEtox® should be communicated with more transparency.

It was discussed how time horizon should be considered in LCA context. Typical approaches recommend calculations in two ways: a) limit calculation to 100 years (no

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[4] http://aiida.tools4env.com/

infinity) and include uncertainty of estimate, and b) calculation in steady state (provided steady state is reached) and include uncertainty of estimate. To address long-term behavior including persistence, the integration of long-term changes in metal speciation (so called ageing) was recommended. Where ageing models or factors are available, these should be taken into account in both calculations (100 years and steady-state).

Main concerns expressed by metal experts were also a perceived lack of transparency and missing documentation of the USEtox® methodology, the need to address the essentiality of some metals for freshwater ecosystems, the large variability among archetypes for the proposed coefficients, and the fact that the work of Gandhi et al. has not yet been integrated in LCA DB and software.

Human Health session

Addressing Human Health issues in RA and LCA (Scott Baker)

One of the key subjects discussed was the need to account for latest available data in the data bases. The REACH data sets were pointed as important input in this context that need to be incorporated.

The main metal specificities agreed for ecotoxicology (such as, background value, speciation in groundwater and freshwater) of metals should be taken into account when assessing metal fate in the environment also in human health characterization factors.

The model of human health should consider dose-response reaction and exposure as a driver for model delimitation.

Olivier Jolliet presented an aluminium case study illustrating how to address Human Health of metals building on USEtox®. He emphasized the latest work from Kounina et al. (2014), which provides the possibility to further spatialize fate and intake fraction for metal emissions to water worldwide at a 50km scale and provide sector specific fate factors and intake fractions for use in LCIA.

Preliminary results obtained on aluminum show that the following challenges can and should be addressed:

1) For long-term emission from landfills, the assumption of 100% transferred to surface water is not valid. A specific transfer fraction from groundwater to surface water needs to be determined, considering all removal processes in groundwater, such as precipitation for aluminum that reduces transfer by a factor 10,000.

2) It may be of high interest to present emissions and impacts that occur in the first 100 years separately from longer term emissions.

3) Several USEtox® input data based on EPIsuite need to be updated, accounting for improved data made available in latest publications or in ECHA.

4) Work on fate in surface water carried out by Dong and Gandhi requires to be extended to As and Cr VI, with specific parameterization of e.g. WHAM for these metals.

5) It is important to consider water treatment processes in determining exposure to drinking water.

6) Near field exposure of consumers to compounds within a product may be a dominant exposure pathway.

7) Bioavailability to humans and metal essentiality are important to address, especially for primarily deficient essential metals, such as Zinc. The framework is now available to address these challenges and improve the life cycle assessment of human health impacts of metals, in collaboration with the metal industry.

Uncertainly and variability of the measures was then one of the major drivers for the discussion, and it is recognized as a major challenge to overcome. It has been proposed to compare the relative values of human health results in log normal between substances to account for the typically two to three orders of magnitude uncertainty compared to 12 orders of magnitude variations between substances.

Concluding session: recommendations and priorities for further work

The last session, common to all participants, took place in the morning of the second day and focused on the main recommendations and conclusions.

Two sets of recommendations have been derived. The first set of recommendations is short term oriented and not directly linked to new research activities, e.g. like the use of the appropriate reference database, the update of USEtox® in LCA software or communication to USEtox® users. The second set of recommendations is directly linked to research or model development needs which are then more long-term objectives.

1) Non-research based recommendations (short term to medium term):

- **Implementing latest USEtox® results in LCA DB and software**: Latest development on USEtox® metal characterization factors, related to the work of Gandhi and Dong, are not yet included in LCA databases and software. Hence, it is strongly recommended to update the USEtox® model and CFs in LCA software and DB to consider such new developments.

- **Using state-of-the-art databases:** The USEtox® characterisation factors should be calculated considering a broader range of newer data bases, and as much as possible involving peer review. Such a review of data should cover substance-specific as well as landscape data, also including sub-model (e.g. WHAM/FIAM) input data. Extrapolations, e.g. acute-chronic, NOEC-EC50, etc. should be checked regarding the importance of their substance-specific variability. With regard to human health data on metals, the latest available quality screened data should be used. The REACH data base has been identified as a relevant reference data base at this stage. Metal industry experts recommended to check the consistency of the USEtox® database used for calculating the CFs and to use as far as possible REACH as the main source of information. It is suggested to develop a common case study between the metal industry and the USEtox® development team to understand USEtox® in further details and to explore how latest available data on metals may help improving the assessment quality.

- **Promoting transparency and facilitating verification of CF calculation**: It is currently difficult to verify and trace data used to calculate USEtox® CFs. Hence, it is recommended to implement a more transparent DB to allow a verification of the data choices for CF calculation by external experts.

- **Communication of uncertainty**: Within any life-cycle based toxicity assessment, the overall level of uncertainty should be quantitatively calculated and systematically communicated to LCA practitioners and other users of life cycle information who make critical policy and materials selection decisions.

- **Communicating adequately with the LCA community**: The potential misinterpretation and misuse of USEtox® results for product comparison and benchmarking activities are main concerns of metal industry experts. Hence, it is recommended to develop **a best practice guidance** explaining how to interpret USEtox® results, focusing on the limitations and potentialities of the USEtox® results, when assessing metals.

The advances in the characterization modelling of metals in USEtox® relative to the Apeldoorn recommendations are summarized in Table 1. At the time of the Leuven Workshop, the discussion referred to USEtox® 1.0 only. the publication of the new USEtox® 2.0 was not yet confirmed at that time. USEtox® 2.0 should be released early 2015 and will address several of the above-described issues, in particular by incorporating the work of Ghandi and Dong for metal speciation and bioavailability to ecosystems, by providing a detailed documentation of all processes, data and equations and by differentiating between 100 years characterization factor and longer term effects.

Table 1: Advances in the characterisation modelling of metals in USEtox® ("-" = not considered, "☑" = considered, "%" = partially considered, "+" = research needed "n.a." = not applicable).

Recommendations from Apeldoorn declaration (2004)	USEtox® 1.01 (2008) = Status at WS		USEtox® 2.0 (2015) = Future release*	
	Freshw. Ecotox.	Human toxicity	Freshw. ecotox.	Human toxicity
Speciation determines bioavailability and toxicity and is a highly desirable extension of LCIA, both in fate and effect modelling	-	-	% to ☑	-
Persistence Infinite time horizons in steady state effect models could only be appropriate if bio-availability is properly considered	-	-	% to ☑	100 years horizon also available
Essentiality Within the essentiality window of essential metals the LCA general principle of "less is better" does not apply and the possibility of adverse biological effects should be set at zero	-	+	+	+
Bioavailability The Biotic Ligand Model (BLM) should be used preferentially, with the Free-Ion Activity Model (FIAM) used in cases where the BLM has not been fully developed.	-	n.a.	%	n.a.
Characterisation The characterisation factor should be chosen at the HC50 (geometric mean of EC50) level rather than the HC5 or the NOEC level.	☑	n.a.	☑	n.a.
Compartments in the multimedia model If effects are negligible in a given compartment, there is no need to consider effects. This may well be the case for essential metals in the ocean.	☑	☑	☑	☑
Spatial aspects The consequences of regional differences in bioavailability, background concentrations, and therefore toxicity need to be further elaborated	-	%	% to ☑	%

*Once released, the level of completion of the various recommendations will be assessed by metal experts

2) Research-based recommendations (medium-term):

- **Essentiality**: Addressing essentiality aspects of metals is crucial for some metals for ecosystems and for human health. The approach developed by Gandhi and Dong should allow considering this specificity of some metals. Hence, it is recommended to perform further research work in this area to address metal essentiality both for ecotoxicity and human toxicity

- **Speciation:** Building on the approach developed for ecotoxicity, it is now important to account for essentiality in the human toxicity assessment of metals.

- **Persistence and time scale:** Infinite time horizons in steady state effect models could only be appropriate if bio-availability is properly considered. To address long term behaviour and persistence, it is recommended to integrate long term changes in metal speciation (so called ageing). This is potentially relevant for human exposure to – and ecotoxic impacts in - terrestrial and deep ocean compartments where substance residence times can be very long. Where ageing models or factors are available, these should be taken into account. In the context of terrestrial and deep-ocean ecotoxicity it is recommended to calculate CFs for a limited time horizon of 100 years and an additional CF based on a time scale of >100 years to infinite.

- **Variability and regionalisation (spatial aspects):** The works of Gandhi, Dong and Kounina have demonstrated the relevance and the need to differentiate CFs of metals according to local specificities. Further research work regarding their consideration in LCA (i.e. which scales, which archetypes, etc.) should be done in this promising area, keeping in mind however that appropriate regional LCI datasets should then also be developed to ensure a proper matching of regional CFs with emission data in the LCI datasets.

- **Uncertainty**: Assessing and tracking the level of uncertainty along the modelling steps from emission to the toxicity end point is seen as an important area for further research work.

- **Compartments in the multimedia model**: the groundwater compartment is a relevant pathway to be considered for metals. Terrestrial and marine ecosystems should also be developed along the same lines as the freshwater ecosystem. For human health, indoor exposure (home and occupational) and near-field exposure (occupational) may be relevant exposure pathways to be considered.

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