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Regional LCA in a global perspective

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Metals in the ocean: an adapted LCA fate and exposure model*

4.1 Introduction

One of the main output categories of human economic activity is generally characterised as products. The fact that we produce and consume products is the major cause of most anthropogenic environmental impacts. Every product may thus be held responsible for a certain amount of environmental damage. This damage is generally not only caused by the product as such or by its direct use, but also by processes, linked to production and waste management. In order to control or assess the overall environmental damage for which a product is responsible, the entire product life cycle should be accounted for.

Individual processes within the product life cycle may be controlled by operating with environmental constraints, and also by environmental monitoring, combined with the use of RA techniques. These techniques may be very specifically attuned to individual process circumstances and may give a detailed image of what is going on at a specific location.

Although environmental control of individual processes is necessary for minimising the environmental effects of each of these processes, it is generally not best for minimising the overall environmental burden of a product. Individual processes may be optimised, but it is difficult to use RA to account for the effects of substituting an entire process by another process, taking place at a different location (*e.g.* when metal parts of the product are replaced by plastic parts), let alone for the trade-off effects of such choices (*e.g.* on product durability, and thus on long-range material use and waste production). For this type of questions, product LCA is a more suitable tool.

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While RA is thus used for control in the context of individual (production) processes, LCA is generally the preferred instrument for overall product assessment. Product assessment may be necessary for the environmental optimisation of a certain product, but also for the choice between product alternatives. In this last case, process locations are often undetermined by definition. Questions on environmental comparisons between product alternatives – for example between glass and plastic bottles, or between paper and cotton nappies – often apply to a national or even international context, thus going beyond the level of individual processes at individual locations.

The life cycle of a product may easily cover tens of processes, causing hundreds of emissions, spread over a large part of the world. In order to keep the assessment manageable and feasible, these large numbers of emissions and emission locations make it necessary to use more simplified methods for LCA than the more constrained RA methods that are usually applied for the assessment of chemicals emitted from a particular process. RA looks in detail at toxic chemical impacts, for example.

For the abovementioned reasons, LCA is by necessity a less specific instrument than RA, producing results that apply to a more general context.

One of the core parts of LCA is LCIA. In LCIA, the emissions and subtractions, caused by the product to be assessed, are quantified in the context of a number of environmental impact categories, such as resource depletion, global warming, ozone layer depletion, acidification, eutrophication, ecotoxicity and human toxicity. The toxicity-related impact categories – to which metal emissions contribute – are considered to be the most complex ones, and the preliminary attempts to tackle them belong to the most criticised parts of LCA.

One of the central characteristics of LCA is the fact that it produces results in the form of 1 quantified environmental impact score for each of the environmental impacts assessed and for each product alternative involved in the assessment. This implies that the effects of different substances not only should be quantified, but also should be added up somehow. This is accomplished by ascribing different characterisation factors to individual substances, that serve as weighting factors for the eventual relative harm in the context of each impact category, caused by the emission of a standard amount of substance. In these characterisation factors, not only environmental hazard but also the environmental residence time and exposure characteristics are discounted. Very different effect types of different substances are quantified in such a way that they become comparable.

Generally applicable models are needed in order to handle large numbers of substances in an equivalent manner. It is common practice in LCIA to make use of Mackay-type multimedia models to account for substance fate. Although these models have originally been designed for organic chemicals, Diamond *et al.* (1990, 1994) applied and validated their use for metals in an early stage. Later on, this

type of application was officially recommended in the EC Technical Guidance Documents (EC 1996). The LCA characterisation factors for metals in the Handbook on Life Cycle Assessment (Guinée 2002) are based on the work of Huijbregts *et al.* (2000). These authors used the suggestions of Crommentuijn *et al.* (1997) on how to handle metals in the SimpleBox model (Brandes *et al.* 1996) underlying the fate part of their model USES-LCA. Metal-specific elimination processes in the marine environment and metal-specific exposure characteristics were not accounted for, however. This may have led to important deviations in modelling results, causing the relative effects of metals to be possibly overestimated by orders of magnitude.

The fact that metals are relatively sensitive to such over-estimations seems to be largely caused by two facts. In the first place, metals are non-degradable, and thus remain in the environment for a relatively long time, thereby spending a relatively large part of their environmental lifetime in the ocean compartment. In the second place, metal speciation in the marine environment reduces their availability to marine organisms. The first priority for the improvement of LCIA for metals is therefore to refine the LCA fate and exposure models with respect to the behaviour of metals in the ocean. The goal of this chapter is to set a modest and tentative first step in this direction, in order to investigate possible directions for improvement within the existing context of global-scale multimedia modelling.

4.2 The GLOBOX model

The adaptations described in this model have been introduced in the GLOBOX fate and exposure model for LCA. GLOBOX is a model under construction for the calculation of spatially differentiated LCA characterisation factors on a global scale (Wegener Sleeswijk *in prep.*). GLOBOX consists of 3 main modules: an impact-category independent fate module, a human-exposure module applicable to all impact categories that are related to human exposure to chemicals, and an effect module, which contains effect-related parameters for every separate impact category. The fate and exposure modules are differentiated on the level of separate countries and oceans, which means that there are about 250 regions. Each country or ocean has its own set of homogeneous compartments, and all countries and oceans are interconnected by atmospheric and aquatic flows. Oceanic water bodies are handled as a 2-compartment system: the surface mixed layer and the deep ocean. Multimedia transport and degradation equations are taken from the European Union System for the Evaluation of Substances (EUSES) multimedia model SimpleBox 2.0 (Brandes *et al.* 1996; ECB 1997), supplemented by specific equations to account for advective air and water transport between different countries or oceans. Distribution parameters for multimedia transport are differentiated per country or ocean with respect to climatic features, water coverage, and land use. All distribution equations are solved simultaneously by matrix inversion (matrix

dimension 3000×3000), allowing for the calculation of global-scale multimedia transport. Human exposure calculations are based on intake by food and drinking water consumption and inhalation of the exposed population. Exposure parameters are differentiated per country with respect to population density, estimated mean body weight, consumption pattern, fraction of drinking water derived from surface water and groundwater, respectively, and fraction of drinking water that has been purified. GLOBOX requires only substance-specific input of physico-chemical and toxicity data to calculate specific human and ecological toxicity characterisation factors for unit emissions to any compartment in any country of the world.

4.3 Adaptations

In order to adapt the GLOBOX model for metals, a small number of adaptations has been introduced. The adaptations that have been introduced so far cover the following subjects:

- speciation (with specific attention for Hg)
- adsorption to suspended particles, and
- sedimentation and vertical mixing.

Moreover, the model-dependency on parameters that make no sense in the context of metals, such as octanol-water partition coefficient, water solubility, and vapour pressure, has been eliminated.

4.3.1 Speciation

One of the most well-known metal-specific processes is speciation. Ratios between the different species depend on regional environmental properties such as temperature, acidity, the presence of complexing agents, and the medium considered.

Speciation influences both fate and exposure. With respect to fate as such, Diamond *et al.* (1992) developed an approach to account for species interconversion between species by use of specific rate constants. As they state, however: ‘The practical difficulty is that the rate constants for species interconversion ... are rarely known’. We therefore accounted for the influence of speciation on fate only in an implicit way, by using overall environmental transport rates for individual metals, as will be elaborated in section 4.3.4. With respect to exposure, however, we tried to account for speciation in a more explicit way.

Mainly due to the presence of complexing agents in marine waters, speciation tends to differ largely between fresh and marine waters. Because most of these complexing agents are not present in artificial test media, this factor is generally not accounted for by marine toxicity data. In order to get a good picture of the

relative toxicity of metals in marine surface waters, it should be accounted for separately.

Different metal species differ with respect to their chemical and toxic properties. The bio-availability of metals is believed to be largely determined by the free ion activity, rather than by the total aqueous concentration of a metal (Morel 1983).

In the GLOBOX model, only the fraction of free ions is considered to be potentially toxic, except for Hg, for which the (highly bioavailable) organic species are taken into account separately. Relative concentrations of free ions in seawater were derived from Byrne *et al.* (1988). These values refer the ocean as a whole. Specific values for the relative amount of free ions in the mixing layer of the ocean could only be found for Cd and Zn (Nolting *et al.* 1999). Although the difference between relative free ion concentrations in the ocean as a whole on the one side and in the surface layer on the other side can be enormous – mainly due to complexation by dissolved organic ligands in the surface layer – we applied the overall values to the oceanic surface layer as well for all other metals, for lack of better data. In the test medium, we assumed the relative free ion concentrations to equal the values reported by McGloskey (1996). An overview of all values is given in Table 4.1. For the calculation of the relative bioavailability of metal ions in seawater, we used the quotient of the relative concentration seawater (surface layer if available) and test medium, respectively.

Table 4.1 Relative concentrations of free ions in seawater (derived from Byrne *et al.* at T = 5°C and pH = 8.2) in the surface layer of the ocean (Nolting *et al.* 1999), and as assumed in the test medium (McGloskey 1996).

metal	free ions in seawater (%)	free ions in surface layer (%)	free ions in test medium (%)
Cd	3.2	0.8	71
Cr	0.0011		91
Cu	12		76
Pb	5		40
Ni	68		77
Zn	73	0.9	80

4.3.2 Mercury speciation

Mercury takes a special place among metals, first because of the volatile character of its elemental species, and second because of the environmentally important role of organometallic forms (largely methylmercury). Mackay *et al.* (1995) and Diamond (1999) use a ‘three species model’ for Hg, in which they make a distinction between elemental, inorganic, and organic mercury. For each of these groups of species, they present partition coefficients as well as concentration fractions in each environmental medium. In order to deal with temperature dependency, Mackay *et al.* (1995) present values for the enthalpy of phase change (ΔH) along with the

partition coefficients. Thus, partition coefficients (K) can be deduced at any desired temperature (T) expressed in absolute degree as

$$d(\ln K) / dT = \Delta H^0 / RT^2 \quad (4.1)$$

where R is the gas constant (Van 't Hoff equation).

For inorganic Hg (and other metals), partition coefficients (among which are specific values for marine waters) are provided by Crommentuijn *et al.* (1997). For elemental and organic Hg species, we calculated partition coefficients on the basis of the values provided by Mackay *et al.* (1995). To correct for the environmental temperature of 282° K, we used the Van 't Hoff equation. The partition coefficients for partitioning between wet sediment and water, and between wet suspended matter and water, were calculated from the respective solid-water partition coefficients by assuming the volume ratio between water and solids to be 4 for sediment and 9 for suspended matter. The solid-water partition coefficient for soil was assumed to equal the corresponding sediment value. The volume ratio between solids and water was assumed to be 1 for soil, allowing for the calculation of separate soil-water partition coefficients. The aerosol-air partition coefficients were calculated from the ratio between aerosol-water and air-water partition coefficients. The fraction of the chemical associated with aerosols (F_{aer}) was calculated from these values to be 1.0 for each of the 3 Hg species.

In order to make it possible to introduce Hg into the GLOBOX model without changing the model as such more than strictly necessary, a weighted mean of the partition coefficients for the three separate species was calculated for each relevant phase change. As weighting factors, the ratios in which the 3 species occur in the 'departure medium' were used.

4.3.3 Adsorption to suspended particles

Most substances in the water compartment will be adsorbed to a greater or lesser extent to suspended particles. For aquatic toxicity assessment, we leave the adsorbed fraction out of consideration in our model. The ratio between adsorbed and dissolved species of metal ions in seawater depends on the concentration and nature of suspended particles, on the mutual forces between these particles on the one hand and each individual metal on the other, and on qualities of the solvent (*e.g.* fresh or salt water). Crommentuijn *et al.* (1997) gives an overview of empirical partition coefficients between suspended matter and seawater, established for a number of metals in water of the North Sea and Wadden Sea. We have used these values as best available estimates.

4.3.4 Sedimentation and vertical mixing

In this chapter, the term 'sedimentation' is understood to refer to the gravitation-driven sinking of particles and adsorbed substances, whether or not these particles

and substances eventually reach the sediment. Sedimentation in the surface mixed ocean layer thus refers to particle transport to the deep ocean layer, and not necessarily to the ocean sediment.

In the EUSES model, the sedimentation rate of organics is modelled as a function of the suspended matter-water partition coefficient, the sedimentation rate of these particles as such, and the concentration of suspended particles. For metal sedimentation in the surface mixed ocean layer, we developed a specific approach.

For metals sedimentation in the surface mixed ocean layer is a key process. The sedimentation rate of metals in the surface mixed ocean layer thus largely determines the residence times of metals in the environmental system, and therewith eventually their relative environmental impacts as assessed by LCA.

Deriving sedimentation rates from the combination of particle sedimentation rates and suspended matter-water partition coefficients – the standard procedure for organics in the EUSES model – is not satisfactory for the purpose of modelling sedimentation of metals in the surface layer of the ocean. Suspended matter is a heterogeneous mixture of particles that largely differ in size and content, and therefore also in sedimentation rate and metal adhesion. Partition coefficients are average values that do not reflect the specific adsorption of a metal to a specific kind of suspended matter. The particle sedimentation rate in the EUSES model is an average value that does not reflect the specific sedimentation rate of different kinds of particles. Combining these average values may cause deviations between the calculated and actual sedimentation rates that – if applied to the ocean surface mixed layer – may eventually produce unrealistic LCA results for metals. If possible, the use of empirical, metal-specific residence times of metals in the surface mixed layer for the estimation of sedimentation rates would be largely preferable.

Bruland (1980) gives an overview of mean residence times with respect to particulate removal of metals in a 80 m surface mixed layer of the North Pacific, estimated from measured concentrations and inputs by atmospheric fluxes and vertical mixing (Table 4.2). Other sources of in- and output, such as eddy diffusion (or convection), and horizontal advection and diffusion, are stated to play a minor role within this oceanic region, and have been left out of consideration. We adapted these residence times for a 100 m surface mixed layer, assuming a constant mean net deposition rate, and used these adapted residence times for the calculation of advective flows from the surface mixed layer to the deep ocean.

Table 4.2 Estimated atmospheric fluxes, net vertical mixing input fluxes, and mean residence times in surface mixed layer (80 m) with respect to particular removal of metals in the North Pacific (Bruland 1980).

metal	atmospheric input ($\text{nmol} \times \text{cm}^{-2} \times \text{y}^{-1}$)	net vertical mixing input fluxes ($\text{nmol} \times \text{cm}^{-2} \times \text{y}^{-1}$)	mean residence time (part. removal) (y)
Cd	0.0018	0.14	0.1
Cu	0.16	0.21	10
Pb	0.18	-0.01	3
Ni	0.043	0.8	20
Zn	0.40	0.15	1.0

4.4 Results and discussion

In current LCIA models, the characterisation factors for metals – used to quantify their relative environmental harmfulness – are very high in comparison to characterisation factors for organic pollutants. This phenomenon is largely caused by the fact that metals are nondegradable. According to commonly used multimedia models (*e.g.*, the EUSES model), the only ways by which metals can leave the environmental system are sedimentation and sediment burial. This makes their residence times in the environmental system – especially in the world oceans – comparatively high. Because characterisation factors are not only proportional to toxicity measures and concentrations but also to environmental residence times, the relative harmfulness of metals according to these factors may easily go against intuition.

It may be questioned whether the magnitude of the differences between present characterisation factors for metals on the one hand and organic pollutants on the other are not too high. By taking some metal-specific processes into account we have tried to set a first step towards the improvement of metal-specific LCA characterisation factors.

In Table 4.3, the results of the application of a preliminary version of our metal-adapted GLOBOX model (Wegener Sleeswijk *in prep.*) are compared to the corresponding results of the USES-LCA model (Guinée 2002). In order to avoid the biasing influence of differences in toxicity, which we will not discuss here, we focussed on exposure as such. Table 4.3 therefore lists (relative) exposure magnitudes, not characterisation factors. (The characterisation factors have been divided by their composing EFs.) It should be remarked that the comparison between GLOBOX and USES-LCA should be handled with care, first because GLOBOX and USES-LCA are different models, and second because we could only use a very preliminary version of GLOBOX in this stage. In order to make results of both models comparable, we normalised both to toluene as a reference.

Table 4.3 Relative exposure magnitudes of human population and marine ecosystems to metals, compared to toluene, caused by equally large standard emissions to the ocean compartment.

metal	relative human exposure		relative marine ecosystem exposure	
	GLOBOX	USES-LCA	GLOBOX	USES-LCA
Cd	1.88E-04	1.13E+01	7.63E+00	1.64E+04
Cr	4.80E-08	5.48E+00	1.95E-03	7.49E+03
Cu	6.57E-04	9.00E+01	2.73E+01	4.43E+04
Pb	4.97E-04	3.13E+01	2.03E+01	3.25E+03
Hg	4.15E-03	3.66E+03	1.69E+02	2.93E+04
Ni	3.67E-03	3.99E+02	1.50E+02	2.80E+05
Zn	4.63E-05	1.04E+02	1.89E+00	1.95E+04
geometric means	1.54E-04	8.27E+01	6.31E+00	2.24E+04
toluene	1.00E+00	1.00E+00	1.00E+00	1.00E+00

It appears that the metal-specific adaptations, made in the GLOBOX model, result in a complete reversal of the original image: For human exposure, the relative exposure magnitudes are now lower for all metals than for toluene, while for marine ecosystem exposure, they are in the same order of magnitude as toluene on average. For some metals, a difference with toluene of 1 or 2 orders of magnitude remains. For human toxicity we see a difference of 5 orders of magnitude, while for marine ecotoxicity, a difference of 4 orders of magnitude shows up between GLOBOX and USES-LCA results. Although the adaptations in this preliminary version of GLOBOX are only a very first step towards adequate LCA modelling of metal-specific processes in the marine environment, these results may be considered an indication that the present chasm between LCA characterisation factors for metals and organic pollutants will most probably largely diminish if metal-specific processes are introduced into current LCA models.

In order to get a more reliable image of the relative harmfulness of metals, further research and model development are needed. A large number of possibilities to extend current LCA fate and exposure models with respect to metals remain to be explored, among which are the following:

- a more detailed and specific quantitative elaboration of the processes that we tried to cover here: metal speciation, adsorption to suspended particles, sedimentation, and vertical mixing;
- accounting for the specific organic ligand complexation in the upper mixed ocean layer for all metals;
- accounting for the kinetic aspects of metal speciation;
- accounting for the toxic aspects of complexed metals and particle-adsorbed chemicals;
- accounting for geochemical effects on environmental residence times of chemicals; and

- distinguishing between the open ocean and coastal zones as separate modelling regions.

Despite all efforts that have been made and that may be made in the future, we expect that metal emissions will still easily dominate LCA emission profiles, while this may be against intuition and knowledge of the actual relative damage caused by these substances. This phenomenon raises the question of whether the LCA approach to assess the anthropogenic environmental presence of substances merely on the basis of amounts, independently of the environmental concentrations caused by these amounts, always makes sense. It may be argued that as long as total anthropogenic emission levels of a substance do not considerably add to the natural background in a certain compartment, or alternatively, as long as concentrations remain far below environmental no effect levels, direct or indirect emissions to this compartment could be left out of consideration. Although this seems to be an attractive and sensible option, it should, in practice, be handled with great care. Multimedia models presume homogeneous concentrations and instantaneous dilution of emitted substances. Increased concentrations around emission sources remain invisible, and even increased concentrations in large areas may be averaged out by lower concentrations elsewhere in the same compartment. The fact that a certain environmental threshold is not surpassed on an average basis, as pointed out by the result of multimedia modelling application, does not imply that this threshold is not surpassed on a local level. Considerable effects may occur without being recognised by multimedia modelling alone. Further research is needed in order to enable a more specific approach that leaves out those emissions that cause no harm at all, but that maintains – possibly in an adapted form – the assessment of emissions that cause local effects.

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