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Including the spatial variability of metal speciation in the effect factor in life cycle impact assessment: Limits of the equilibrium partitioning method

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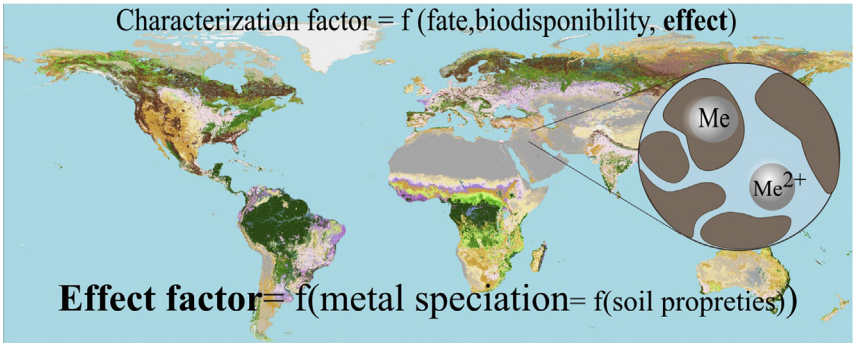
HIGHLIGHTS

- We calculate regionalized EFs for terrestrial ecotoxicity.
- The two regionalized methods show EF variability between 6 and 19 orders of magnitude.
- The effect and bioavailability factors are the major contributors to characterization factor variability.

GRAPHICAL ABSTRACT



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ABSTRACT

In life cycle assessment (LCA), the potential terrestrial ecotoxicity effect of metals, calculated as the effect factor (EF), is usually extrapolated from aquatic ecotoxicological data using the equilibrium partitioning method (EqP) as it is more readily available than terrestrial data. However, when following the AMI recommendations (i.e. with at least enough species that represents three different phyla), there are not enough terrestrial data for which soil properties or metal speciation during ecotoxicological testing are specified to account for the influence of soil property variations on metal speciation when using this approach. Alternatively, the TBLM (Terrestrial Biotic Ligand Model) has been used to determine an EF that accounts for speciation, but is not available for metals; hence it cannot be consistently applied to metals in an LCA context. This paper proposes an approach to include metal speciation by regionalizing the EqP method for Cu, Ni and Zn with a geochemical speciation model (the Windermere Humic Aqueous Model 7.0), for 5213 soils selected from the Harmonized World Soil Database. Results obtained by this approach ($EF^{EqP}_{regionalized}$) are compared to the EFs calculated with the conventional EqP method, to the EFs based on available terrestrial data and to the EFs calculated with the TBLM ($EF^{TBLM}_{regionalized}$) when available. The spatial variability contribution of the EF to the overall spatial variability of the characterization factor (CF) has been analyzed. It was found that the $EFs^{EqP}_{regionalized}$ show a significant spatial variability. The EFs

Abbreviations: LCA, life cycle assessment; LCI, life cycle inventory; LCIA, life cycle impact assessment; EF, effect factor; CF, characterization factor; BF, bioavailability factor; FF, fate factor; Cu, copper; Ni, nickel; Zn, zinc; TBLM, terrestrial biotic ligand model; EqP, equilibrium partitioning method; WHAM 7.0, Windermere Humic Aqueous Model 7.0; PAF, potentially affected fraction; ERA, ecotoxicological risk assessment; UNEP, United Nations Environmental Programme; SETAC, Society of Environmental Toxicology and Chemistry; HWSD, Harmonized World Soil Database.

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calculated with the two non-regionalized methods (EqP and terrestrial data) fall within the range of the EFs-$^{EqP}_{regionalized}$. The EFs$^{TBLM}_{regionalized}$ cover a larger range of values than the EFs$^{EqP}_{regionalized}$ but the two methods are not correlated. This paper highlights the importance of including speciation into the terrestrial EF and shows that using the regionalized EqP approach is not an acceptable proxy for terrestrial ecotoxicological data even if it can be applied to all metals.

1. Introduction

Life cycle assessment (LCA) is an environmental tool to quantify the potential environmental impacts of a product or service throughout its life cycle, from the extraction of raw materials to end of life (Hauschild, 2005). A product's environmental footprint may be assessed and improved or compared with others. An LCA study is divided into four parts: definition of the goal and scope, life cycle inventory (LCI) (amount of inputs and outputs in the product system), life cycle impact assessment (LCIA) and results' interpretation (de Haes et al., 2002). LCIA converts each environmental intervention in the life cycle inventory of the studied product into a potential impact for a specific impact category, such as global warming or ecotoxicity, via a characterization factor (CF).

Currently, CFs for ecotoxicological impacts in operational LCIA methodologies are calculated as the multiplication of a fate factor (FF) and an effect factor (EF). The FF accounts for the fraction of substance transported from the environmental compartment of the emission to the receiving compartment (soil, in the case of terrestrial ecotoxicity) and the metal's time of residence in this compartment. The EF models the effect of a substance on the ecosystem per concentration of exposure for the total fraction of the metal.

To estimate the EF, Payet and Jolliet (2004) recommend using the hazardous concentration affecting 50% of the species in the ecosystem ($HC50_{EC50}$) (Payet and Jolliet, 2004). The AMI method (Assessment of the Mean Impact) used in IMPACT 2002 + and USEtox determines this concentration as the geometric mean of the effect concentration affecting 50% of the individuals of each species (EC_{50}) for at least three phyla (Payet and Jolliet, 2004). Indeed, EC_{50} is more widely used in ecotoxicological testing for soil vertebrates, invertebrates and plants and has less variability than other ecotoxicological indicators such as NOEC/LOEC (no or low observed effect concentration) because it is determined by a curve considering all the empirical samples instead of being the sample with the highest concentration before observing an impact (NOEC) or the first with an observed impact, which may both vary significantly depending on the number of samples (Payet and Jolliet, 2004). For this reason, the indicator is more suitable for life cycle impact assessment studies since potential impacts are evaluated for comparative purposes and receptor protection is not the goal as it is in ecotoxicological risk assessment (ERA).

EC_{50} are estimated from ecotoxicological testing and can be found in databases. In the case of terrestrial organisms, EC_{50} values are scarce and terrestrial $HC50_{EC50}$ ($HC50_{EC50\text{-}soil}$) are usually extrapolated from aquatic ecotoxicological data with the equilibrium partitioning method (EqP) (Eq. (1)). Developed by Wenzel et al. (2000) this method is based on the $HC50_{EC50}$ of aquatic organisms ($HC50_{EC50\text{-}w}$), soil density (ρs (kg/L)), fraction of soil water (f_w (0.4 L/L)) and partition coefficient K_d (Wenzel et al., 2000) based on the underlying assumption that the terrestrial ecosystem has a similar sensitivity to toxic substances as the aquatic ecosystem and that it is only exposed to the dissolved fraction of the contaminant in the soil.

$$HC50_{EC50-soil} = HC50_{EC50-w} \times (K_d \times \rho s + f_w) \quad (1)$$

K_d (L/kg) is the ratio of the total metal bound to the solid phase (in mg/kg) and the metal found in the liquid phase (in mg/L). Currently, in LCIA, the EqP uses a generic K_d per substance to extrapolate an aquatic data to a terrestrial $HC50_{EC50\text{-}soil}$ ($mg_{total\ metal}/kg_{\ dry\ soil}$).

Modeling the available fraction of metal using a single K_d value assumes that the toxic fraction of a substance is independent of soil physicochemical conditions. However, metal toxicity is a function of speciation, which depends on the physicochemical conditions of the environment (Fairbrother et al., 2007). In the case of metallic substances, it is recognized that soil properties such as pH, organic matter and texture have a huge impact on a metal's speciation and, thus, on the K_d (Janssen et al., 1997; Sauvé et al., 2000). A study by the US EPA provides Kd values that vary by 3.4 orders of magnitude for copper, 3.8 for nickel and 6 for zinc (Allison and Allison, 2005). Generic K_d values should therefore not be used to extrapolate metal aquatic ecotoxicological data to terrestrial ecotoxicological data.

Haye et al. (2007) showed a strong discrepancy for metals between EFs based on extrapolated aquatic data and EFs based on terrestrial data and recommend avoiding such extrapolation and directly using terrestrial ecotoxicological data when available to determine the $HC50_{EC50\text{-}soil}$ (Haye et al., 2007). However, on one hand, not enough data are available on all metals to calculate a robust $HC50_{EC50\text{-}soil}$: Haye et al. (2007) stated that the three phyla were covered for only nine metals when using available ecotoxicological data. On the other hand, the available terrestrial data is rarely documented enough to determine metal speciation during testing. This means that a) terrestrial ecotoxicological data are valid only for the soil for which they were determined and cannot be extrapolated to other types of soil accounting for speciation and b) it is questionable to use different ecotoxicological data obtained from different tests performed in different types of soil to calculate a terrestrial EF.

The need to account for metal speciation was acknowledged under the Clearwater Consensus (M. Diamond et al., 2010), with the recommendation to express the CF as the multiplication of a fate factor (FF), a bioavailability factor (BF) and an effect factor (EF). The BF is the bioavailable fraction of the metal exposing the ecosystem. When using a BF, the EF must be expressed for the bioavailable metal forms. These three factors should take metal speciation into account and be regionalized. Some studies include the speciation of metals in soils in LCIA models but chiefly relate to the FF and BF (Plouffe et al., 2015a, 2015c). Plouffe et al., 2015a define the bioavailable fraction of zinc only for its fate by including a soil-specific BF but the terrestrial ecotoxicological data needed for soil-specific EF is lacking.

Another way to deal with the lack of terrestrial EC_{50} data in the literature and account for metal speciation in soil is to use the Terrestrial Biotic Ligand Model (TBLM). This approach was applied by Owsianiak et al. (2013) to estimate new regionalized and soil-specific EFs for copper and nickel. The TBLM is based on the assumption that the toxicity of a metal in a soil depends on the concentration of the metal bound to the biotic ligand (Di Toro et al., 2001). Biotic ligands are receptors that enable a substance to have an effect on an organism. The TBLM accounts for the competition between the free ion form of metals and the other soil's compounds such as the major cations (Ca^{2+}, Na^{+}, Mg^{2+}, H^{+}). This model is highly relevant to EF determination because the effect concentration of a metal on an organism is better represented by the fraction of metal bound to the biotic ligand of the organisms than by the labile of free ion form. However, the model was only developed for copper and nickel (Koster et al., 2006; Thakali, 2006) and cadmium and lead (An et al., 2012), meaning that it does not allow for consistent

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