



Including metal atmospheric fate and speciation in soils for terrestrial ecotoxicity in life cycle impact assessment

Lycia Aziz¹ · Louise Deschênes² · Rifat-Ara Karim² · Laure Patouillard² · Cécile Bulle³ 

Received: 16 January 2017 / Accepted: 5 January 2018 / Published online: 12 January 2018
© Springer-Verlag GmbH Germany, part of Springer Nature 2018

Abstract

Purpose The aim of the study is to calculate regionalized characterization factors for the atmospheric emissions of metals transferred to soil for zinc, copper, and nickel taking into account the atmospheric fate and speciation.

Methods In order to calculate characterization factors for all possible atmospheric emission locations around the world, the link between atmospheric deposition with regionalized soil fate factors and bioavailability factors accounting for the metal's speciation was established. The methodology to develop the regionalized fate factors and characterization factors is threefold. First, the emitted metal fraction that is deposited on soils is calculated from atmospheric source-receptor matrices providing for each emission location the fraction of an emission that is deposited on each worldwide receiving cell ($2^\circ \times 2.5^\circ$ resolution). Second, the fraction of metal deposited in different soil types is determined by overlapping the deposition map with a soil map, based on the 4513 different soil types from the Harmonized World Soil Database. Third, bioavailability factors are calculated for each soil type, which allows determining the bioavailable fraction of the deposited metal depending on the soil properties. Combining these steps with the effect factors results in a series of terrestrial ecotoxicological characterization factors. These characterization factors are then applied in an illustrative example and compared to results obtained with generic characterization factors. The case study focuses on the electricity production process in Québec, whose ecosystem impacts are currently dominated by metal ecotoxicity impacts. The uncertainty due to the spatial variability of the impact is quantified.

Results and discussion Our results show that regionalized characterization factors are over three orders of magnitude lower than generic characterization factors. They are presented on maps and their spatial variability was evaluated at different regional scales (region, country, world). The use of regionalized characterization factors with their spatial variability at different geographic resolution scales in the case study gives a result more or less precise depending on the level of resolution of the characterization factor applied (country or global-default). The impact scores of the three metals in the case study are three orders of magnitude lower when compared to the scores obtained with generic characterization factors.

Conclusions The development of those regionalized characterization factors improves the terrestrial ecotoxicity assessment in life cycle impact assessment by taking into account the atmospheric fate and the speciation of the metal for new 3 metals for the different soil types in the world and by documenting their spatial variability.

Keywords Atmospheric fate · Characterization factor · Life cycle assessment · Metals · Regionalization · Speciation · Terrestrial ecotoxicity

Responsible editor: Miguel Brandão

Electronic supplementary material The online version of this article (<https://doi.org/10.1007/s11367-018-1438-8>) contains supplementary material, which is available to authorized users.

✉ Cécile Bulle
bulle.cecile@uqam.ca

¹ Institute of Environmental Sciences, CIRAIQ, Université du Québec à Montréal, C.P. 8888, succ. Centre ville, Montréal, QC H3C 3P8, Canada

² Department of Chemical Engineering, CIRAIQ, Polytechnique Montréal, C.P. 6079, succ. Centre-Ville, Montréal, QC H3C 3A7, Canada

³ Department of Strategy and Corporate Social Responsibility, CIRAIQ, ESG UQAM, C.P. 8888, succ. Centre ville, Montréal, C.P. 8888, succ. Centre ville, Montréal, QC H3C 3P8, Canada

1 Introduction

This paper focuses on the characterization of the ecotoxicological impact of terrestrial airborne metals in life cycle assessment (LCA) considering a regionalized atmospheric fate and speciation in receiving soils.

Industry, raw material extraction, agriculture, and many other activities have a significant impact on the environment, some of which is due to toxic metal emissions. LCA is a comparative tool to assess the potential environmental impact of products, services, or processes considering their entire life cycle, from resource extraction to end of life (Jolliet et al. 2005). The ISO 14040 standard sets out four phases to perform an LCA study: goal and scope definition, life cycle inventory, life cycle impact assessment, and interpretation (ISO 2006). There are many impact categories assessed in LCA, including climate change, acidification, and eutrophication. In this study, we focus on terrestrial ecotoxicity. In LCA, the impact score is calculated by multiplying the total quantity of elementary flows throughout the entire life cycle of the product by a characterization factor (Eq. (1)).

$$S_j = \sum_i CF_{ij} * M_i \quad (1)$$

S_j , impact score for impact category j ; CF_{ij} , characterization factor of the elementary flow i for impact category j ; M_i , quantity of the elementary flow i

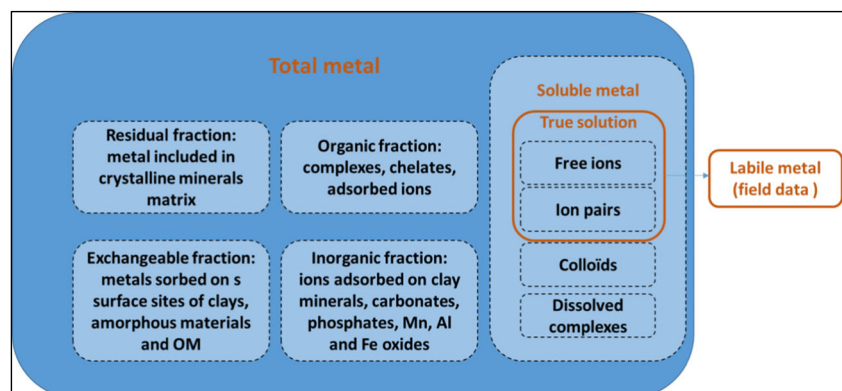
To assess the toxicity and ecotoxicity impacts, the current consensus in LCA is to use the USEtox model, a multimedia model resulting from an expert working group of the United Nations Environment Programme and Society of Environmental Toxicology and Chemistry (UNEP/SETAC) Life Cycle Initiative (Hauschild et al. 2010). The original characterization factor (CF) for ecotoxicological impact in USEtox used to be the product of a fate factor (FF) and an effect factor (EF). The FF gives the distribution of the substance between environmental compartments combined with the residence time in the receiving compartment. The EF calculates the effect of the transferred fraction of substance in the receiving environmental compartment on the ecosystems.

USEtox was developed for organic compounds and did not take into account speciation (i.e., the fact that the metal can take different forms) nor persistence in environment which are key properties of metals (Haye et al. 2007); hence, it used to be inappropriate for metals (Hauschild 2007; Strandesen et al. 2007). In fact, a metal may take different forms according to the physicochemical properties of the receiving environment (Fairbrother et al. 2007) meaning that the ecotoxicological impact of metals should be regionalized based on the receiving environment's properties. This problem was highlighted by a group of LCA and metal ecotoxicity modeling experts in the Clearwater Consensus, under the aegis of the UNEP/SETAC Life Cycle Initiative (Diamond et al. 2010).

Following this consensus, a lot of progress was made to improve metal ecotoxicological impact assessment in LCA. In the aquatic ecotoxicity impact category (Dong et al. 2014; Gandhi et al. 2010, 2011a, b), the bioavailability factor (BF) was integrated into the CF. The BF represents the fraction of the total metal that is considered available to the ecosystem. In the Clearwater Consensus, it was recommended to use the true soluble fraction of metal to determine the BFs (Diamond et al. 2010), a fraction that is composed of free ions and ion pairs as shown in Fig. 1. Experimentally, this fraction corresponds to the labile fraction of metal (Nolan et al. 2005). The Clearwater Consensus recommended the Windermere Humic Aqueous Model (WHAM) to determine the metal speciation to calculate the BFs (Diamond et al. 2010), which was done by Gandhi et al. (2010, 2011a, b) for Cu, Ni, and Zn and generalized by Dong et al. (2014) to all the other metals covered by WHAM, using water archetypes with different properties (pH, organic matter content (OM), etc.), which is integrated in the current version of USEtox.

Recent efforts were also made to improve terrestrial ecotoxicity. Owsianiak et al. (2013) proposed an approach based on empirical regression models to calculate the bioavailable fraction for nickel and copper. However, those empirical regressions are not supposed to be extrapolated outside their original scope (soil types) (Groenenberg et al. 2012). Plouffe et al. (2016) proposed a second approach using WHAM to integrate zinc speciation in FF and BF, using soil

Fig. 1 Metal fractions illustration (adapted from Plouffe et al. (2015a))



properties available in the Harmonized World Soil Database (HWSD) for all types of soils around the world (pH, cationic exchange capacity (CEC), OM, texture, and carbonate content). This database includes disaggregated data for 16,000 types of soils and is, to our knowledge, the most complete soil database (FAO/IIASA/ISRIC/ISSCAS/JRC 2012). The FF were determined with USEtox, using the soil-specific partition coefficient between soil particles and water (K_d) according to Zn speciation determined using WHAM (Plouffe et al. 2015a). Soil-specific BF_s were also based on WHAM results. The speciation predictions from WHAM were validated using measured field samples from the literature. WHAM gave better results than the empirical regression approach from Owsianiak et al. (2013) to predict the Zn soluble fraction in field samples (i.e., true solution metal plus colloids and dissolved complexes, as shown in Fig. 1), both in terms of rank of solubility between soils and in terms of absolute values. Such a validation was not possible for the truly dissolved fraction of Zn because of insufficient field data available.

With regard to the EFs, Gandhi et al. (2010) used the Biotic Ligand Model (BLM) to generate aquatic ecotoxicity EF. However, BLM are not available for all the metals. Dong et al. (2014) proposed the use of the free ion activity model (FIAM) instead of BLM, assuming that toxicity is only caused by the binding of free metal ions to critical sites in target organisms. For terrestrial ecotoxicity, the equilibrium partitioning method (EqP) is commonly applied in LCA when no terrestrial ecotoxicity data is available: it allows extrapolating terrestrial ecotoxicological data from the aquatic ecotoxicological data based on the substance partition between soil and water. The terrestrial biotic ligand models (TBLMs) were recommended by Owsianiak et al. (2013) and were considered the most robust way to obtain regionalized EF accounting for speciation, but TBLMs are not available for all the metals. Tromson et al. (2017) compared two methods to obtain terrestrial regionalized EF for metals: the TBLMs and the EqP with soil-specific K_d determined using WHAM (i.e., considering, as Dong et al. (2014) did for aquatic ecosystems, that toxicity is directly linked to the free metal ion). Their results showed a total absence of correlation between the EFs obtained using the EqP/WHAM approach and the TBLM. Hence, they recommended to keep using the generic EFs as default since no robust method is currently available that could be applied consistently to all the metals to regionalize the EFs.

Those recent developments in metal terrestrial ecotoxicity in LCA are limited to the direct metal emissions to soil. Still, a significant fraction of metal emissions is transferred to the soil from the atmosphere. For example, in Canada in 2014, over 138 tonnes of nickel, 346 tonnes of zinc, and 252 tonnes of copper are emitted to the air yearly, which is more than the direct emissions to soil (26, 303, and 209 tonnes/year, respectively, for Ni, Zn, and Cu) (INRP 2014). Atmospheric metal

emission dispersion was addressed at the continental scale (Liu et al. 2008; Sanderson et al. 2008), and at the local scale (Pizzol et al. 2012), but the study conducted by Roy et al. (2016) is, to our knowledge, the only one covering the entire world consistently. Source-receptor matrices (SRMs) were developed by Roy et al. (2016) from the Harvard/NASA GEOS-Chem global model (Yantosca et al. 2015). The GEOS-Chem model is a 3D model taking into account emissions, transport, deposition, and chemical transformation. The SRMs were simulated for meteorological data for 2005 and calibrated using the European Monitoring and Evaluation Program (EMEP) data. For each emission location (resolution of $2^\circ \times 2.5^\circ$), the SRMs provide the emitted fraction that falls on each of the receiving cells (resolution of $2^\circ \times 2.5^\circ$). However, no fate in soil is considered once the metal is deposited.

The present work aims to combine the atmospheric fate of metal and its speciation in soil in an LCA context. Zinc, nickel, and copper were chosen for this exploratory work because they are well documented, and they often appear as big contributors to ecotoxicity in LCA case studies. Therefore, this paper aims to create regionalized characterization factors to quantify the potential terrestrial ecotoxicity of metals (Zn, Ni, Cu) emitted to air in LCA. The influence of these new regionalized characterization factors on LCA results will also be analyzed through the case study of a kilowatt hour (kWh) of electricity produced in Québec—which terrestrial ecotoxicological impact is dominated by metal emissions (Plouffe et al. 2015b).

2 Methods

2.1 Fate factors from air to soil determination

Fate modeling was performed in two stages. First, the distribution of metals for all possible emissions worldwide: SRMs developed by Roy et al. 2016 provide the fraction of the metal deposited in each receiving $2^\circ \times 2.5^\circ$ cell for an emission in any of those cells at the global scale. Subsequently, this grid is overlapped using Arc GIS with a map of soil types from the Harmonized World Soil Database Version 1.21 in order to determine the distribution of the metal in the different soil types. To get the fraction of metal by type of soil, it is assumed that the metal is distributed uniformly in each cell of $2^\circ \times 2.5^\circ$. Then, the fraction considered to be transferred to one type of soil from the one-grid cell is proportional to the area fraction occupied by this soil type in the box (calculation details are available in the Electronic Supplementary Material Section 1). The surface of each soil in each cell is calculated using the Mollweide projection (in Arc GIS), since it is an equal area projection, and to be consistent with the IMPACT World+ project assumptions (Bulle

et al. 2017). This approach makes it possible to calculate the total amount of metal transferred from the atmosphere to each soil type (i.e., the atmospheric fate factors FF_{as} from air cell a to each soil type s) by summing the quantity of an emission in air cell a received in all the $2^\circ \times 2.5^\circ$ cells or fractions of cells intersecting soil type s from the HSWD map using Arc GIS. Secondly, the fate of the fraction within the soil (i.e., the fate factor of the metal directly emitted to soil—from soil to soil— FF_{ss} for each soil type s) (see Eq. (2)) is modeled using the Plouffe method for the three metals (Zn, Ni, and Cu) (Owsianiak et al. 2013).

$$FF_{ss} = \frac{\Delta C_{total,s} \cdot V \cdot \rho_b}{\Delta M_s} \quad (2)$$

with $\Delta C_{total,s}$ as the time-integrated incremental change in concentration of total metal in the soil s up to the infinite, V as the volume of soil, ρ_b as the soil density, and ΔM_s as the incremental change in the metal emission to the soil. All the values used for the calculation are summarized in the Excel spreadsheet in the Electronic Supplementary Material Section 2.

As recommended by Plouffe et al. (2016), the FF_{ss} is determined with USEtox, using partition coefficients between soil and water calculated using WHAM 0.7. Fate factors from soil to soil (FF_{ss}) were calculated for the 4513 types of soil using the soil properties (pH, OM, CEC, texture, and carbonate content) available in version 1.21 of the Harmonized World Soil Database. The background concentration of metals in the soil is taken into consideration according to the Kabata-Pendias and Mukherjee (2007) estimations (for more information, see the Electronic Supplementary Material Section 3). Partition coefficients based on soluble fraction were used, since the use of WHAM for soils was only validated for the soluble fraction of metal by Plouffe et al. (2015a). As we acknowledge that the true solution is the fraction that is supposed to best represent the bioavailable fraction (Diamond et al. 2010), we also did all the calculations for the true soluble fraction, which can be found in the Electronic Supplementary Material Section 4. We consider the topsoil properties (i.e., topsoil in the HSWD database (Harmonize soil database 2012)) to determine a site-specific K_d using WHAM 7.0. The fate modeling within the topsoil was performed using USEtox with this site-specific K_d . The speciation and fate of the metal in the deeper soil layers and in the groundwater are not determined once the metal is transferred from the topsoil: we assume that most target organisms of the terrestrial ecosystem are exposed mainly via the topsoil (following the approach published by Plouffe et al. 2015a, b).

Finally, the fate factors for atmospheric emission transferred to each type of soil (FF_{as}) were obtained by multiplying

the results of the first and the second steps (see Eq. (3)). Note that for one emission in one atmospheric cell ($2^\circ \times 2.5^\circ$), there are as many FF_{as} as receiving cells (obtained by crossing the 4513 types of soils with the atmospheric deposition grid of $2^\circ \times 2.5^\circ$). To better understand the spatial variability due to the atmospheric fate, aggregated FF_{as} will be presented as intermediary results (i.e., summing the FF_{as} of all the receiving cells for an emission cell) even if this is not what is done when calculating the CF_{as} in the next steps where the specific fate in each soil cell is considered, meaning the FF_{as} have to be kept disaggregated.

$$FF_{as} = AFF_{as} * FF_{ss} \quad (3)$$

2.2 Characterization factor calculation

Characterization factors for the terrestrial ecotoxicological impact of atmospheric emissions CF_{as} are calculated by multiplying the resulting $FF_{as,j}$ from the emission cell to the soil receiving cell j by the $BF_{s,j}$ specific to each of the receiving soil cell j and by a generic EF, and by summing the resulting impact across all the receiving soil cells (Eq. (4)).

$$CF_{as} = \sum_j FF_{as,j} * BF_{s,j} * EF \quad (4)$$

The BF_{s} are calculated using WHAM 0.7 and the soil properties from the HSWD based on the soluble fraction of metal in order to be consistent with the FF_{as} calculation. Here again, the background concentration of metals in soil is taken into consideration according to the Kabata-Pendias and Mukherjee (2007) estimations (see values in the Electronic Supplementary Material Section 3).

Following Tromson et al. (2017) recommendations, the EF are not regionalized: the generic aquatic EF from USEtox are used to derive terrestrial EF default values using the AMI method (Eq. (5)) (adapted from Payet 2004).

$$EF_{soil} = \frac{0.5}{\left((EF_{aquatic} * 0.5) * (k_d * \rho_s + f_w) \right)} \quad (5)$$

where K_d is the soil-water partitioning coefficient, ρ_s is the soil density, and f_w is the water fraction in soil. Those three values are taken directly from the generic soil of USEtox, and their values can be found in the Electronic Supplementary Material Section 2.

Regionalized “native resolution” CF_{as} were determined this way for each potential emission location in the world (i.e., each of the $2^\circ \times 2.5^\circ$ grid cells around the world). Aggregated CF_{as} at the country and global (global default) levels were also determined using a weighted average of native resolution CF_{as} weighted by the area of each cell within the country (or the world). This corresponds to an assumption of an equal probability of emission within a country, which is

a way, among others, to calculate aggregated CFs. An alternative proxy would have been to consider that metal emissions occur most probably in highly populated regions. However, it is probably totally wrong for metals since mining which generally occurs in areas with low population is one of the main contributors to the overall global emissions. Ideally, regionalized national inventories of pollutant release should be used as weighting factors to proceed to the aggregation (i.e., considering that the emission occurs more probably where more emissions are declared), but these inventories are not available for metal emissions at the global scale. An equal probability of emission was therefore considered as a proxy. Since the “native resolution” CF_{as} are made available in this paper under the form of shape files, any other relevant weighting set becoming available could later be used to re-aggregate the CFs if needed.

The uncertainty related to the spatial variability of the CF_{as} within each country and the world was determined using the min and the max values within the considered geographical region (it corresponds to the uncertainty related to not specifying more precisely the point of emission when using a country level or a global default CF_{as}). The detailed contribution to the overall spatial variability of the CF_{as} of the spatial variability of the different factors (FF_{as} , FF_{ss} , and BF_s) could not be analyzed in detail as for one point of emission a high number of receiving cells is implied with different values of FF_{as} , FF_{ss} , and BF_s . However, we did (1) an analysis of the spatial variability of the FF_{as} and the BF_s and (2) a partial analysis of the relative contribution of the spatial variability in the atmospheric fate compared to the spatial variability in the soil fate by doing a linear regression between the aggregated FF_{as} and the CF_{as} . By deduction, what is not correlated to the atmospheric fate may be correlated to the spatial variability in soil, whether it is due to the FF_{ss} or to the BF_s .

The characterization factor for an emission occurring in Montréal was compared when using respectively the generic USEtox CF_{as} (i.e., a CF_{as} obtained by multiplying the USEtox fate factor from air to soil with the generic EF from Eq. (5)), the global default CF_{as} , the country-level CF_{as} for Canada, and the native resolution-scale CF_{as} for Montreal, with the respective uncertainty on the impact score due to spatial variability when using a lower geographic resolution CF_{as} .

2.3 Illustrative example

The results were applied in an illustrative example to better assess the impact of the metals emitted to air and to soil in the LCA of the 1 kWh of electricity produced in Québec. This case study was selected because metals dominate the ecotoxicological impact, as previously shown by Plouffe et al. (2015b). The purpose of this illustrative example is to assess the feasibility and the influence of the integration of the developed regionalized CFs (i.e., of using the regionalized CF_{as}

developed as part of this project as well as regionalized CF_{ss} for direct metal emissions to soil) on the terrestrial ecotoxicity impact score. Only zinc, nickel, and copper are taken into account in this case study, acknowledging that this is a very partial assessment. However, the full results of the LCA are already available in Plouffe et al. (2015b). The impact score of the zinc, nickel, and copper emitted to the atmosphere during the production of 1 kWh in Québec is calculated (1) using the generic USEtox CF_{as} and CF_{ss} , (2) using the new characterization factors aggregated at the global level (i.e., global default CF_{as}), and (3) using the country-level CF_{as} . The native resolution-scale CF_{as} (i.e., at the native resolution of $2^\circ \times 2.5^\circ$) cannot be used because the life cycle inventory geographic information is only available at the country level (Wernet et al. 2016). Inventory data is taken from the ecoinvent database, using the “market for electricity, low voltage | electricity, low voltage | cut-off, U” process (Wernet et al. 2016). The LCA is realized using the openLCA software. The uncertainty related to spatial variability (i.e., due to using characterization factors at a lower resolution scale than the native one) is quantified by considering the min and the max CF_{as} value within the region.

3 Results and discussion

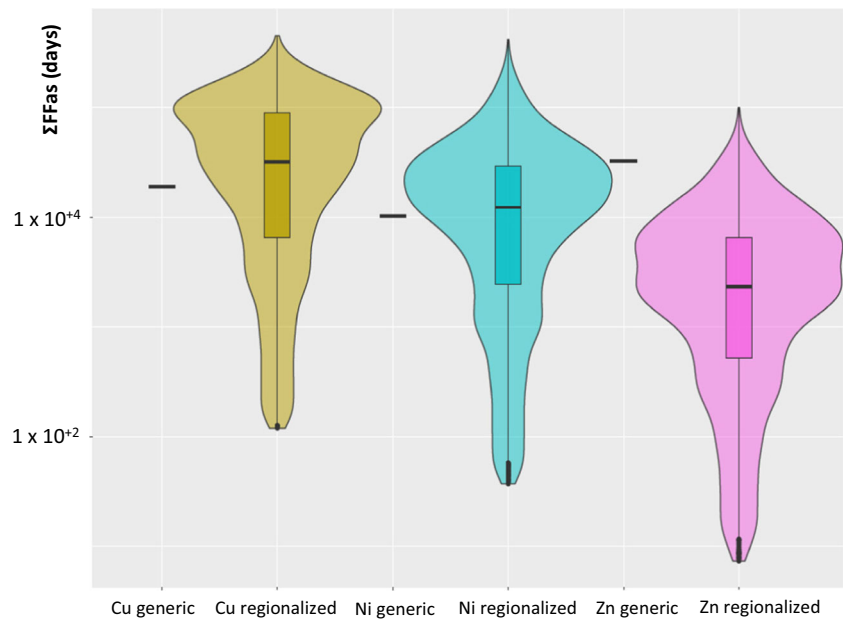
3.1 Fate factors from air to soil determination

The resulting FF_{as} for each of the $2^\circ \times 2.5^\circ$ emission cells (i.e., summing the FF_{as} of all the receiving cells for each emission cell) are illustrated in the violin graph in Fig. 2, showing the spatial variability and the frequency at which the FF_{as} are found around the world.

For the studied metals, there is a spatial variability of regionalized FF_{as} of around four orders of magnitude depending on their emission location. The FF_{as} values for the soluble fraction of zinc, copper, and nickel are distributed between 7.32 and 9.95E+04 days, 1.20E+02 and 4.51E+05 days, and 3.71E+01 and 4.17E+05 days, respectively, demonstrating the influence of the emission location on the atmospheric fate. The density distribution of the plots over the violin shows different frequencies of FF_{as} . The most probable value of FF_{as} is 2.31E+03 days for zinc, 4.17E+05 days for copper, and 1.23E+04 days for nickel. Another interesting result in Fig. 2 is that the difference is small between the generic value of FF_{as} from USEtox and the average value of the regionalized FF_{as} for Cu and Ni and that this difference is around one order of magnitude for Zn, which is within the uncertainty range of the USEtox fate factors.

Fate factors calculated for true solution and soluble fraction of zinc (Zn), copper (Cu), and nickel (Ni) are similar with a difference of less than two orders of magnitude, which is

Fig. 2 Violin graphs illustrating the aggregated atmospheric fate factor (ΣFF_{as}) spatial variability across all the $2^\circ \times 2.5^\circ$ atmospheric emission cells. This graph shows the values and frequency of occurrence of regionalized FF_{as} (Cu regionalized, Ni regionalized, and Zn regionalized) and gives also the USEtox generic CF_{as} (Cu generic, Ni generic, and Zn generic). The box plots show the min, max, 25th and 75th, and the mean



considered as the usual uncertainty in USEtox CF_{as} (Henderson et al. 2011; Huijbregts et al. 2010; Rosenbaum et al. 2008).

3.2 Characterization factor calculation

The resulting BF_{ss} for each type of soil are illustrated in the violin graph in Fig. 3, showing the spatial variability and the frequency at which the BF_{ss} are found around the world. Indeed, there is a spatial variability of regionalized BF_{ss} over eight orders of magnitude depending on the receiving soil. The most probable value of BF_{ss} is close to $1E-3$ for copper, nickel, and zinc.

The resulting CF_{as} for each of the $2^\circ \times 2.5^\circ$ cells are shown on the violin graph in Fig. 4, which illustrates the

spatial variability of the aggregated CF_{as} for the world. Figure 4 represents the CF_{as} values and the frequency at which the CF_{as} are found around the world. All results are presented for the metal soluble fraction (true solution results are available in the Electronic Supplementary Material Section 4). The regionalized characterization factors are systematically lower (between two and three orders of magnitude) than the generic CF_{as} from USEtox. This overestimation of the generic USEtox CF_{as} compared to the new CF_{as} can be explained by the integration of soil-specific BF_{ss} : the total metal in soil was considered available in the generic CF_{as} , whereas regionalized CF_{as} consider only the soluble fraction of the total metal as being available. The spatial variability between the lowest and highest regionalized CF_{as} are over three orders of magnitude (Fig. 4). These

Fig. 3 Violin graphs illustrating the spatial variability of BF_{ss} across the different soil types. This graph shows the values and frequency of occurrence of regionalized BF_{ss} calculated with WHAM 0.7 using the soil properties from the HSWD. The box plots show the min, max, 25th and 75th percentile, and the mean

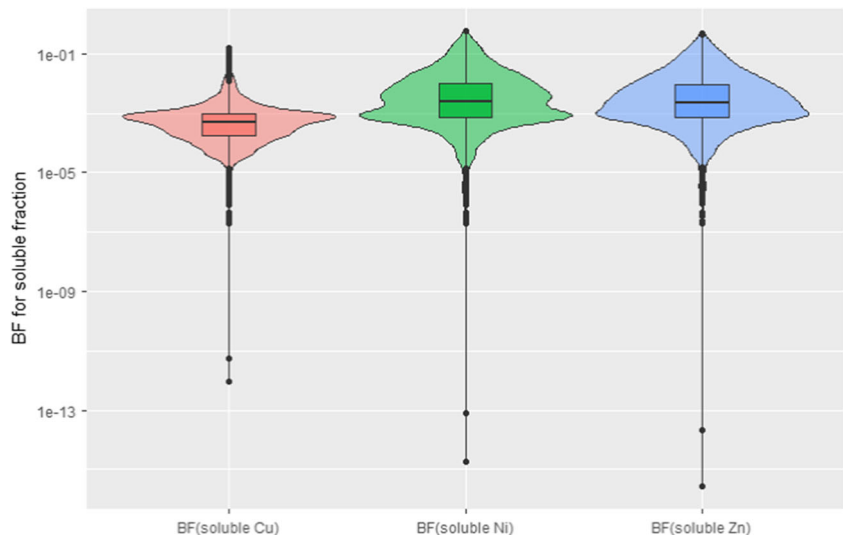
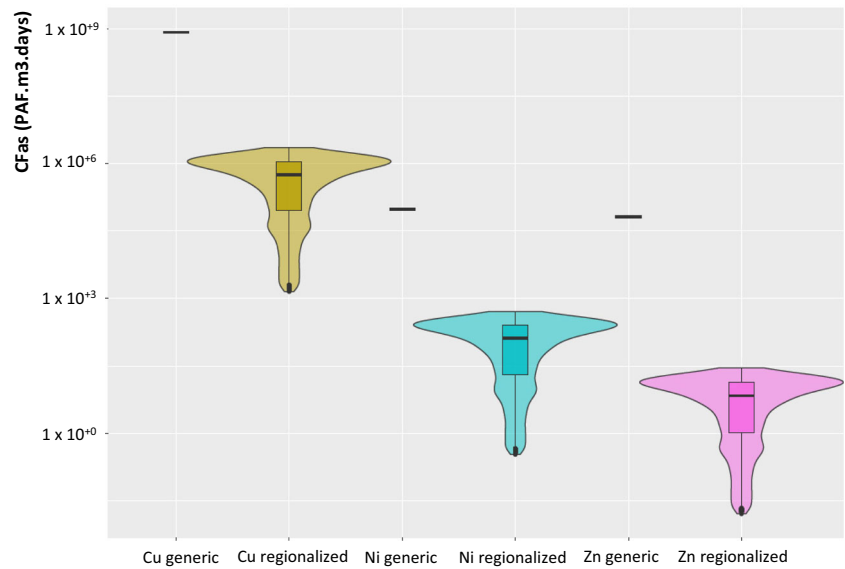


Fig. 4 Violin graphs showing the spatial variability and frequency of occurrence of CF_{as} across all the $2^\circ \times 2.5^\circ$ atmospheric emission cells (Cu regionalized, Ni regionalized, and Zn regionalized) and the generic USEtox CF_{as} (Cu generic, Ni generic, and Zn generic). The box plots show the min, max, 25th and 75th percentile, and the mean



results demonstrate that the location of the emission may have a significant influence on the impact scores in LCA. It means that the CF regionalization for metal atmospheric

emissions that we proposed, based on FF_{as} (sources_receptor method) and metal speciation in receiving soils, is needed to increase the robustness of LCA results.

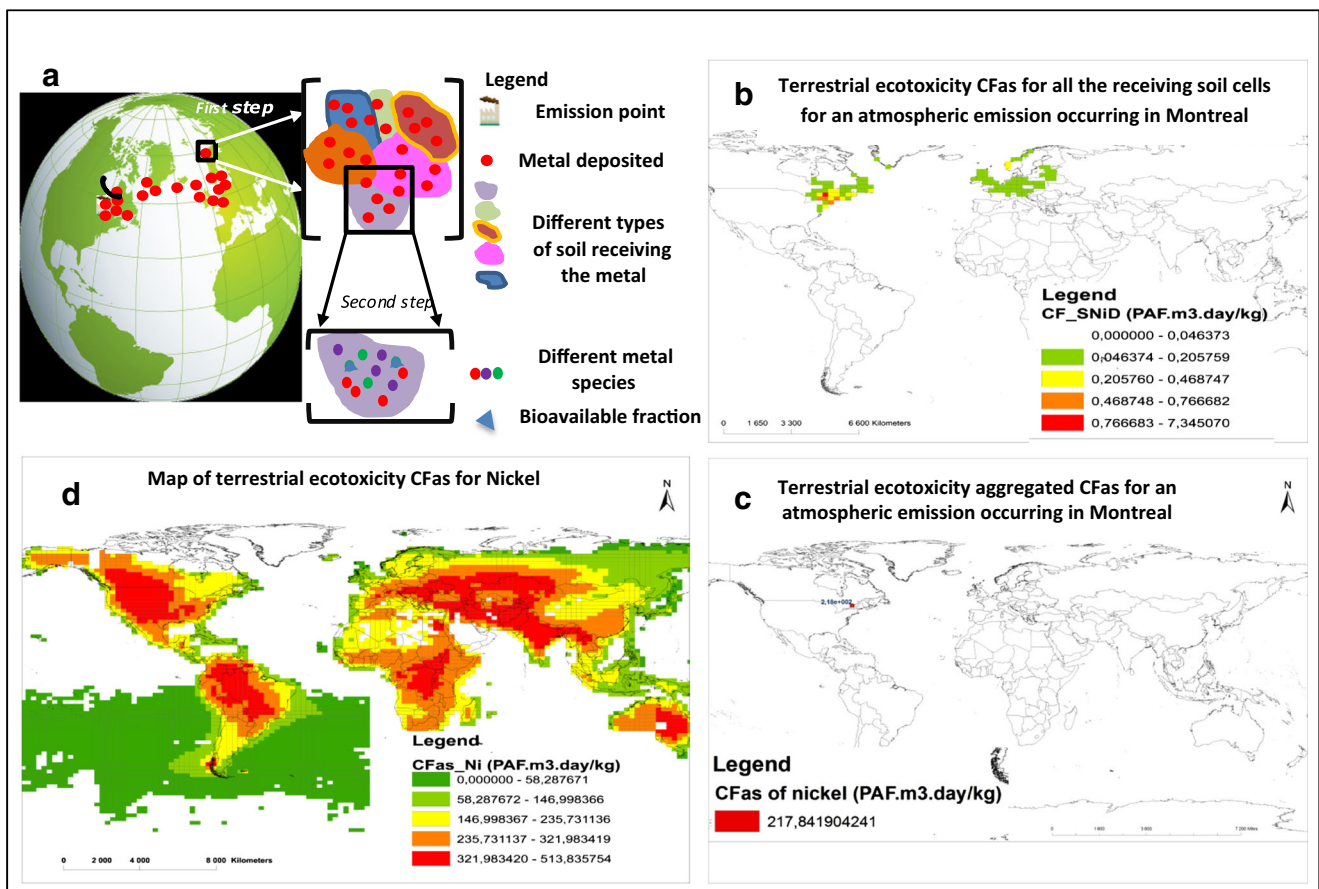
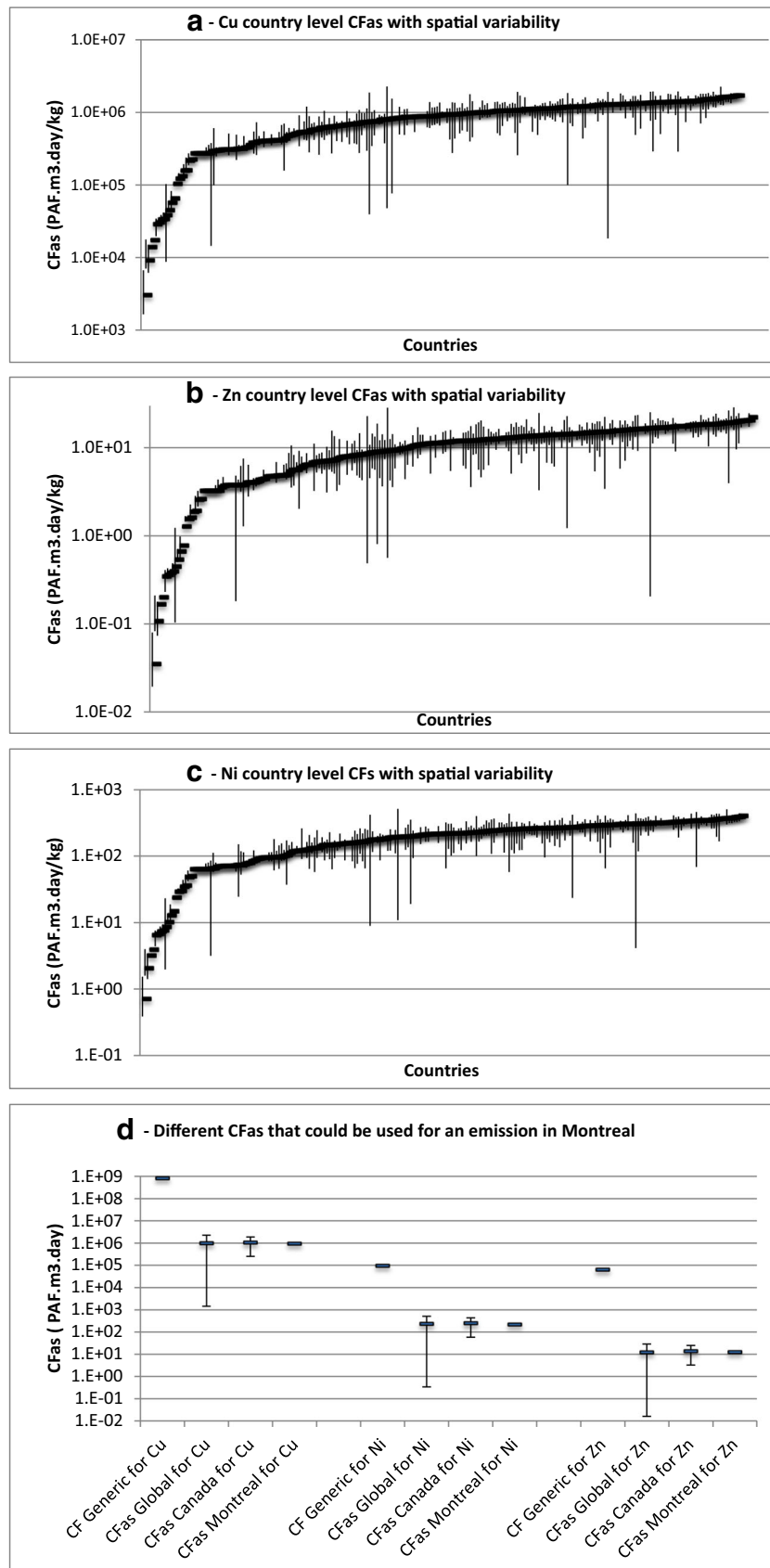


Fig. 5 Regionalized CF_{as} calculation steps. **a** Illustration of the different steps—the first step represents the distribution of an atmospheric emission over different soils and the second step the speciation in those receiving soils; **b** terrestrial ecotoxicity disaggregated CF_{as} for each of the

receiving soil cells for an emission in Montreal; **c** Montreal CF_{as} resulting from the aggregation of all the disaggregated CF_{as} from the map (**b**); **d** regionalized CF_{as} for all the different emission points around the world



◀ **Fig. 6** Terrestrial ecotoxicity CF_{as} at the country level for all the countries of the world for **a** Cu, **b** Zn, and **c** Ni with the corresponding spatial variability ranked by increasing value of country level CFs (error bars represent the min and the max CF_{as}), and **d** influence of the choice of the CF resolution scale on the CF_{as} value and uncertainty due to spatial variability in the case of an atmospheric emission occurring in Montreal

In Electronic Supplementary Material Section 5, a poor correlation is shown between FF_{as} and CF_{as} (R^2 respectively equals 0.30, 0.56, and 0.31 for Zn, Cu, and Ni), highlighting that both the atmospheric fate and the fate in soil contribute to the overall spatial variability of the CF_{as} . We acknowledge we did not use a regionalized EF (i.e., we did not account for the influence of speciation on the effect factor, which may reduce the observed difference between USEtox generic CFs and our regionalized CF_{as} when it will be possible to generate regionalized EFs).

Figure 5 gives an example of the results obtained at the different steps in CF_{as} calculation for Ni. These steps are summarized in Fig. 5a). The Fig. 5b map shows step 1 that calculates for each deposition point the fraction transferred to different type of soil (FF_{as} for an emission in Montréal to all the receiving soil cells). Step 2 calculates the soluble fraction in each type of soil (FF_{ss} for each receiving soil cell). The product of those steps is multiplied by a regionalized BF_{ss} specific to the receiving soil and by a generic EF to obtain the disaggregated CF_{as} for each receiving soil cell for an atmospheric emission occurring in Montréal. The map in Fig. 5c gives the value of the CF_{as} for an atmospheric emission in Montréal, i.e., the sum of the impact of all the cells of the world from Fig. 5b. The map on Fig. 5d is the resulting map for all emission points for which aggregated regional CF_{as} were calculated, the same way it was done for Montréal in Fig. 5b, c. Maps representing the CF_{as} calculated for each point of emission of Cu, Ni, and Zn in the world are available in the Electronic Supplementary Material Section 6 in PDF format and as shape files, which may, for example, be imported directly in openLCA in order to perform regionalized LCA studies (CF_{as} are available at the native resolution scale of $2^\circ \times 2.5^\circ$).

Because metals do not travel over long distances, emissions within continental zones are more likely to be deposited in soils and thus contribute to terrestrial ecotoxicity, which explains the increase in the CF_{as} closer to the center of the continents. The global default CF_{as} are $1.01E+06$ PAF m^3 day kg^{-1} for copper, $2.34E+02$ PAF m^3 day kg^{-1} for nickel, and $1.24E+01$ PAF m^3 day kg^{-1} for zinc.

In Fig. 6, the spatial variability of CF_{as} between countries and within each country is shown for Cu, Zn, and Ni. The spatial variability between the different country-level CF_{as} for all the countries of the world is around three orders of magnitude, but most of the countries are within the same order of magnitude. Moreover, the spatial variability within each country is, in most cases, less than one order of magnitude,

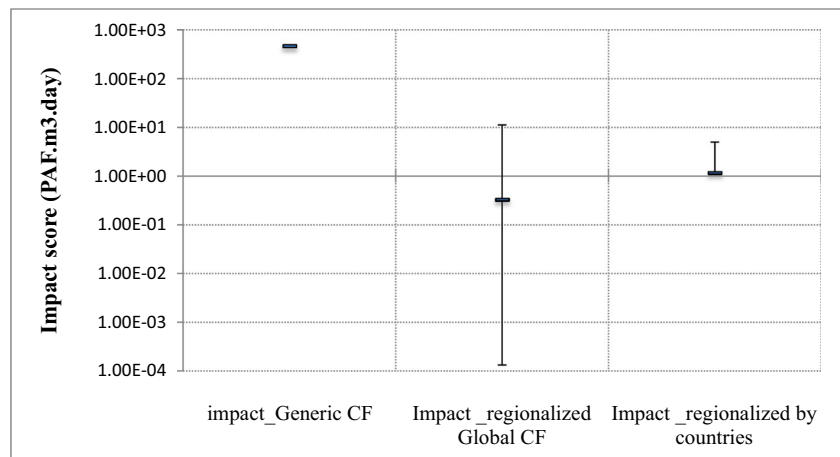
meaning that regionalizing at the country level may be enough most of the time. The countries with a higher spatial variability (i.e., for which it may be worth regionalizing further) are, in decreasing order, Chile, USA, Russia, Australia, India, and Canada. For most countries, it seems that the use of country-level CF_{as} (which corresponds to the geographical resolution of most LCA inventory databases) is a good compromise between the high uncertainty associated to spatial variability and the intensive data collection needed to regionalize at the native $2^\circ \times 2.5^\circ$ level. However, depending on the need in terms of uncertainty defined in the goal and scope of the study, additional spatialization efforts may be needed, following the approach recommended by Patouillard et al. (2016). As an example, the different CF_{as} that could be used for an emission occurring in Montreal, Canada, are compared in Fig. 6d (with their respective spatial variability corresponding to the uncertainty due to not specifying where precisely the emission occurs). The generic USEtox CF_{as} is higher than the new regionalized ones. The global default CF_{as} is at least three orders of magnitude lower than the generic one, with a spatial variability of around three orders of magnitude. The regionalized CF_{as} at the country level for Canada is quite close to the global default CF_{as} , but with smaller spatial variability (a little less than one order of magnitude). The detailed CFs at the native resolution scale and at the country level with the corresponding spatial variability are available in the Electronic Supplementary Material Section 1.

We have assumed that the metal was uniformly distributed over the different types of soils inside each $2^\circ \times 2.5^\circ$ receiving cell. This assumption may influence the result as in reality the deposition may not be homogeneous within the cell. However, most of the time, the receiving cell has one dominant soil type and even at the country level, the spatial variability of the CF remains in general small due to a low difference in soil properties (see Fig. 6), so we do not expect the corresponding uncertainty to be very important when compared to all the other sources of uncertainty in LCA modeling. Electronic Supplementary Material Section 7 illustrates the influence on the CF_{as} distribution to consider (1) a uniform deposition in the different soils of the $2^\circ \times 2.5^\circ$ cell versus (2) a 100% deposition in the dominant soil within the $2^\circ \times 2.5^\circ$ cell, which confirms the low influence of this assumption on the CFas value.

3.3 Illustrative example

The total impact score of the atmospheric emissions of Cu, Ni, and Zn for the case study is shown in Fig. 7 using different options for CF_{as} and CF_{ss} . Here again, when USEtox generic characterization factors are used, the impact score is three orders of magnitude higher than when using the global default or the country-level regionalized CF_{as} . Using the geographic information already available

Fig. 7 Terrestrial ecotoxicity impact score of the atmospheric and soil emissions of Cu, Ni, and Zn during the production of a kilowatt hour of electricity at Quebec using (1) USEtox generic CF_{as} and CF_{ss} , (2) global default regionalized CF_{as} and CF_{ss} ; (3) country-level CF_{as} and CF_{ss} (the two later with the corresponding spatial variability). Error bars represent terrestrial ecotoxicity impact scores calculated using the min and the max CF_{as} possible in the different countries and in the world



in the ecoinvent database, i.e., at the country level for emissions close to the foreground and at the global default level for nonregionally specified background emissions, we were able to significantly reduce the spatial variability to less than one order of magnitude—even if the two countries in which metal emissions were regionalized, i.e., Canada and the USA, are two of the countries identified as having a higher spatial variability—confirming the feasibility of regionalized LCA without very intensive data collection for the LCA practitioner.

4 Conclusions

This study shows the feasibility of integrating the atmospheric fate and metal speciation in soil in an integrated manner in order to characterize the terrestrial ecotoxicological impact of metals in LCA. The significant spatial variability of CF_{as} for an atmospheric emission is observed, which can influence the impact scores. This may, in turn, influence decision-making based on LCA studies. Therefore, we highly recommend to document the uncertainty related to spatial variability or to proceeding with the regionalization of the inventory when atmospheric metal emissions are present in a LCA study. The regionalized CF_{as} allow to calculate CF at different aggregation levels. When the exact emission locations are known, we propose a map to identify the site-specific characterization factors. When only the country of emission is known, shape files at native resolution can be directly imported into the openLCA software, which automatically calculates the aggregated CF_{as} at the country level (average weighted by surface area). LCA analysts can then choose a different aggregation level than the country level if needed. Unlike generic factors, aggregated regionalized factors come with an uncertainty value corresponding to their spatial variability (i.e., the uncertainty of not knowing precisely where an emission occurs). Since soluble fraction seems to be a good alternative as validated by Plouffe, we propose the use of

results which arise from soluble fraction. The EFs used to calculate the regionalized CF_{as} are generic EFs derived from the USEtox model, but we recommend integrating regionalized EF_{ss} as soon as it is available (for example, when TBLMs will be available for enough metals to be meaningfully used in LCA). Finally, this exploratory research was only conducted for Cu, Ni, and Zn, demonstrating the feasibility of our approach to integrate the atmospheric fate and the speciation of metals in soil. In terms of generalization of this approach, the model can be applied to any metal for which the SRMs and the WHAM model are both available. The SRMs were currently developed for metals with a very low volatility that are only transported, within the atmosphere, by sorbing with aerosol particles, it could be used for any non-volatile metal. The WHAM model is available for 19 of those metals, for which it is hence possible to generate FF_{as} using the approach proposed in the present paper.

Acknowledgements The authors would like to thank Pierre-Olivier Roy for his share of expertise and Pablo Tirado-Seco, Amir Yadghar, Hassana El-Zein, Clara Tromson, Geneviève Plouffe and Ivan Viveros Santos for their appreciated contribution. The International Life Cycle Chair (a research unit of the CIRAI) would like to thank its industrial partners for their financial support: Arcelor Mittal, Bombardier, Mouvement des caisses Desjardins, Hydro-Québec, LVMH, 477 Michelin, Nestlé, RECYC-QUÉBEC, RONA, SAQ, Solvay, Total, Umicore, and Veolia Environment. The research was supported by a grant from the Natural Sciences and Engineering Research Council of Canada.

References

- Bulle C et al (2017) IMPACT World+: a globally regionalized life cycle impact assessment method. Int J Life Cycle Assess (submitted)
- Diamond M et al (2010) The clearwater consensus: the estimation of metal hazard in fresh water. Int J Life Cycle Assess 15(2):143–147. <https://doi.org/10.1007/s11367-009-0140-2>
- Dong Y, Gandhi N, Hauschild MZ (2014) Development of comparative toxicity potentials of 14 cationic metals in freshwater. Chemosphere 112:26–33. <https://doi.org/10.1016/j.chemosphere.2014.03.046>

- Fairbrother A, Wenstel R, Sappington K, Wood W (2007) Framework for metals risk assessment. *Ecotoxicol Environ Saf* 68(2):145–227. <https://doi.org/10.1016/j.ecoenv.2007.03.015>
- Gandhi N, Diamond M, van de Meent D, Huijbregts MAJ, 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(13):5195–5201. <https://doi.org/10.1021/es903317a>
- Gandhi N, Diamond ML, Huijbregts MAJ, Guinée JB, Peijnenburg WJGM, van de Meent D (2011a) Implications of considering metal bioavailability in estimates of freshwater ecotoxicity: examination of two case studies. *Int J Life Cycle Assess* 16(8):774–787. <https://doi.org/10.1007/s11367-011-0317-3>
- Gandhi N, Huijbregts MAJ, van de Meent D, Peijnenburg WJGM, Guinée J, Diamond ML (2011b) Implications of geographic variability on comparative toxicity potentials of Cu, Ni and Zn in freshwaters of Canadian ecoregions. *Chemosphere* 8:268–277
- Groenenberg JE, Dijkstra JJ, Bonten LTC, de Vries W, Comans RNJ (2012) Evaluation of the performance and limitations of empirical partition-relations and process based multisurface models to predict trace element solubility in soils. *Environ Pollut* 166:98–107. <https://doi.org/10.1016/j.envpol.2012.03.011>
- Harmonized World Soil Database (version 1.2) (2012) Accessed 21–08-2016
- Hauschild M (2007) International consensus model for comparative assessment of chemicals. SETAC Europe Annual Meeting 2007
- Hauschild MZ, McKone TE, van de Meent D, Huijbregts M, Margni M, Rosenbaum RK, Jolliet O (2010) USEtox™ 1.01 - UNEP/SETAC model for the comparative assessment of chemicals released to air, water and soil and their toxic effects on the human population and ecosystems. UNEP/SETAC
- Haye S, Slaveykova IV, Payet J (2007) Terrestrial ecotoxicity and effect factors of metals in life cycle assessment. *Chemosphere* 68(8):1489–1496. <https://doi.org/10.1016/j.chemosphere.2007.03.019>
- Henderson AD, Hauschild MZ, van de Meent D, Huijbregts MAJ, 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(8):701–709. <https://doi.org/10.1007/s11367-011-0294-6>
- Huijbregts M, Hauschild M, Jolliet O, Margni M, McKone T, Rosenbaum RK, van de Meent D (2010) USEtox™ User manual. USEtox™ Team
- INRP (2014) Inventaire national des rejets de polluants. <http://ec.gc.ca/inrp-npri/donnees-data/index.cfm?do=query&lang=fr>. Accessed 02 Jan 2017
- ISO (2006) Management environnemental — Analyse du cycle de vie — Principes et cadre
- Jolliet O, Saadé M, Crettaz P (2005) Analyse du cycle de vie - Comprendre et réaliser un écobilan vol 23. Gérer l'Environnement, 1st edn. Presses Polytechniques et Universitaires Romandes, Lausanne
- Kabata-Pendias A, Mukherjee AB (2007) Trace elements from soil to human. Springer. <https://doi.org/10.1007/978-3-540-32714-1>
- Liu J, Mauzerall DL, Horowitz LW (2008) Source-receptor relationships between East Asian sulfur dioxide emissions and northern hemisphere sulfate concentrations. *Atmos Chem Phys Discuss* 8(2):5537–5561. <https://doi.org/10.5194/acpd-8-5537-2008>
- Nolan AL, Zhang H, McLaughlin MJ (2005) Prediction of zinc, cadmium, lead, and copper availability to wheat in contaminated soils using chemical speciation, diffusive gradients in thin films, extraction, and isotopic dilution techniques. *J Environ Qual* 34(2):496–507. <https://doi.org/10.2134/jeq2005.0496>
- Owsianiak M, Rosenbaum RK, Huijbregts MAJ, Hauschild MZ (2013) Addressing geographic variability in the comparative toxicity potential of copper and nickel in soils. *Environ Sci Technol* 47(7):3241–3250. <https://doi.org/10.1021/es3037324>
- Patouillard L, Bulle C, Margni M (2016) Ready-to-use and advanced methodologies to prioritise the regionalisation effort in LCA. *Mater Tech* 104(1):105. <https://doi.org/10.1051/mattech/2016002>
- Payet J (2004) Assessing Toxic Impacts on Aquatic Ecosystems in Life Cycle Assessment. École Polytechnique Fédérale de Lausanne
- Pizzol M, Bulle C, Thomsen M (2012) Indirect human exposure assessment of airborne lead deposited on soil via a simplified fate and speciation modelling approach. *Sci Total Environ* 421–422:203–209
- Plouffe G, Bulle C, Deschênes L (2015a) Assessing the variability of the bioavailable fraction of zinc at the global scale using geochemical modeling and soil archetypes. *Int J Life Cycle Assess* 20:1–14
- Plouffe G, Bulle C, Deschênes L (2015b) Case study: taking zinc speciation into account in terrestrial ecotoxicity considerably impacts life cycle assessment results. *J Clean Prod* 108(Part A):1002–1008. <https://doi.org/10.1016/j.jclepro.2015.06.050>
- Plouffe G, Bulle C, Deschênes L (2016) Characterization factors for zinc terrestrial ecotoxicity including speciation. *Int J Life Cycle Assess* 21(4):523–535. <https://doi.org/10.1007/s11367-016-1037-5>
- Rosenbaum RK, Bachmann TM, Gold LS, Huijbregts MAJ, 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(7):532–546. <https://doi.org/10.1007/s11367-008-0038-4>
- Roy P-O, Deschênes L, Cécile B (2016) Global scale atmospheric modeling of black carbon for estimating metal source-receptor relationships for life cycle assessment. *Int J Life Cycle Assess* (submitted)
- Sanderson M et al (2008) A multi-model study of the hemispheric transport and deposition of oxidised nitrogen. *Geophys Res Lett* 35. <https://doi.org/10.1029/2008GL035389>
- Strandesen M, Birkved M, Holm PE, Hauschild MZ (2007) Fate and distribution modelling of metals in life cycle impact assessment. *Ecol Model* 203(3-4):327–338. <https://doi.org/10.1016/j.ecolmodel.2006.12.013>
- Tromson C, Bulle C, Deschênes L (2017) Including the spatial variability of metal speciation in the effect factor in life cycle impact assessment : limits of the equilibrium partitioning method. *Sci Total Environ* 581-582:117–125. <https://doi.org/10.1016/j.scitotenv.2016.12.043>
- Wernet G, Bauer C, Steubing B, Reinhard J, Moreno-Ruiz E, Weidema B (2016) The ecoinvent database version 3 (part I): overview and methodology. *Int J Life Cycle Assess* 21(9):1218–1230. <https://doi.org/10.1007/s11367-016-1087-8>
- Yantosca B, Sager PL, Carouge C (2015) GEOS-Chem v10-01 online user's guide. <http://acmg.seas.harvard.edu/geos/doc/man/index.html>. Accessed 18–08 2016

Reproduced with permission of copyright owner. Further reproduction prohibited without permission.