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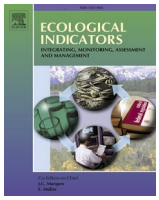

Original Articles

Novel Endpoint Characterization Factors for Life Cycle Impact Assessment of Terrestrial Acidification

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ABSTRACT

Human activities involving combustion and agricultural practices, among others, lead to the release of acidifying compounds such as nitrogen oxides (NO_x), sulfur oxides (SO_x), and ammonia (NH_3). These substances are the main drivers of human-induced terrestrial acidification, a geochemical process resulting mainly in the decline of soil pH, causing ecosystem damage and biodiversity loss. A relevant tool to quantify impacts of human activities is Life Cycle Assessment where characterization factors are used to estimate the potential environmental impacts per unit of emission. These are derived from models of environmental processes occurring along the stressor's impact pathway, connecting an emission to its potential environmental damage.

Here, new ecosystem quality characterization factors for terrestrial acidification were developed, assessing the potential global loss of vascular plant species. The final values combine four elements: existing fate factors, updated soil response factors, recently revised effect factors, and the Global Extinction Probability. The latter allows to convert the local decline in species richness into a global species loss.

The regionalized marginal characterization factors provided represent the aggregated global biodiversity impact in all the world's ecoregions due to an acidifying emission (of NO_x, NH_x, or SO_x) from a specific country. The values cover five orders of magnitude (from 10^{-16} to 10^{-11} $PDF_{global}.yr.kg^{-1}_{emitted}$), and the comparison to currently implemented values has helped both validate the calculation pathway and confirm the need for updated factors. Following current harmonization recommendations, terrestrial acidification impacts can now be compared to those from other stressors estimated in global Potential Disappeared Fraction of species.

1. Introduction

Terrestrial acidification refers to the decrease in base cations (weak acid positively charged ions in the soil) or the accumulation of hydrogen ions (H^+) within terrestrial systems (Norton and Veselý, 2003). Human-induced terrestrial acidification, arising mainly from activities like fossil fuel combustion and agriculture, is caused by the release of acidifying compounds such as nitrogen oxides (NO_x), sulfur oxides (SO_x), and ammonia (NH_3) into the atmosphere (Bouwman et al., 2002; Psenner, 1994; van Zelm et al., 2015). Subsequent chemical reactions in the air produce acidifying dissociation products that may be deposited onto terrestrial systems. The distribution of these deposits depends upon atmospheric conditions (such as temperature, precipitation, and wind), chemical interactions, and topography (van Zelm et al., 2015), and thus causes varied responses around the globe. Soil characteristics render some areas more sensitive due to unreactive geology or base-poor soil (van Zelm et al., 2015).

Terrestrial acidification leads to a decline in soil pH and leaching of essential base cations (Ca^{2+}, Mg^{2+}, K^+), affecting nutrient regulation, reducing phosphorous and magnesium content in plant tissues, decreasing biomass production, hindering germination and regeneration, and fostering competitive exclusion by acid-tolerant species (van Zelm et al., 2015). Ultimately, this phenomenon results in ecosystem damage and biodiversity loss, particularly among vascular plants (which often lie at the bottom of the whole food web).

Biodiversity loss is one of the three dimensions of the ongoing triple planetary crisis, in addition to climate change and pollution (UNFCCC, 2022). Several environmental management tools exist to give decision support across all three dimensions of this crisis; Life Cycle Assessment (LCA) is one of them (Hauschild et al., 2018; Hellweg et al., 2023; ISO, 2006). LCA assesses the environmental impacts of products or processes over their entire life cycles, covering multiple impacts simultaneously. It

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is a comparative tool that gives relative impact values of two or more systems with the same function (e.g., transporting 1 L of liquid, comparing a glass and plastic bottle) (Hauschild et al., 2018; Hellweg et al., 2023). Although not site-specific, it allows to contrast the environmental performances of various products/processes, across their different life phases and for a defined set of impact categories (Hauschild et al., 2018; Hellweg et al., 2023).

Carrying out an LCA requires iterating over four steps (Hauschild et al., 2018). First, the practitioner defines the goal and scope of the study: which questions are to be answered, which impacts should be covered, what temporal and geographical boundaries should be set (Hauschild et al., 2018). Then, the Life Cycle Inventory (LCI) requires collecting quantitative data on all the elementary flows (extracted resources and emissions) occurring during the entire life cycle of the product or process in question (Hauschild et al., 2018). In the third – Life Cyle Impact Assessment (LCIA) – phase, characterization factors (CFs) are used to translate the emission and resource extraction flows collected in the LCI phase (e.g. emitted amounts of NO_x, SO_x, NH_3) into environmental impacts. According to the ISO-norm 14044, practitioners are free to assess the impacts of different stressors (e.g., climate change, land use, terrestrial acidification) either at midpoint or damage level. While midpoint indicators are usually known to be "located on the impact pathway at an intermediate position between the LCI results and the ultimate environmental damage" (Jolliet et al., 2004), damage-level indicators model the whole impact pathway, all the way up to the final damage caused in one of three Areas of Protection (AoP; ecosystem quality, human health, and resource extraction) (Hauschild et al., 2018). For biodiversity loss, belonging to the Area of Protection "Ecosystem Quality", the damage-level indicator recommended by the Life Cycle Initiative hosted by the UN Environment Programme is the "Potentially Disappeared Fraction of Species" (PDF) (Verones et al., 2017). It accounts for the decline in species richness due to a certain stressor. In contrast, one current midpoint indicator for terrestrial acidification quantifies the acidification potential in kg SO_2-equivalents, which is not linearly related to biodiversity loss due to different regional ecosystem sensitivities. Therefore, any LCA aiming to specifically assess biodiversity loss requires going beyond the use of midpoint indicators and applying damage-level indicators (Hellweg et al., 2023). Indeed, while midpoints assess the environmental mechanism, only damage level indicates species losses. Nevertheless, damage-level characterization factors are not currently available and updated for all impact categories. Moreover, whenever possible and relevant in the LCIA phase, the development of spatially differentiated characterization factors (CFs) with global coverage is preferable over spatially generic models (Mutel et al., 2019). Finally, CFs should, if possible, be calculated for different taxonomic groups (e.g., plants, invertebrates) and should quantify both regional losses (potentially reversible, local extinctions) and global losses (irreversible extinctions) (Verones et al., 2017). This is why, by developing a damage-level indicator for terrestrial acidification, this study contributes to the completion of LCIA. It is one step further in the direction of giving LCA its full power: assessing and enabling the comparison of environmental impacts from different stressors along the entire value chain of a product or process (Hauschild et al., 2018; Hellweg et al., 2023).

In the case of terrestrial acidification, state-of-the-art, spatial characterization factors (CFs) are composed of three elements:

- The fate factor (FF) considers the origin and transport of a pollutant from its source to the destination environment. Depending on the emission location, the local atmospheric conditions, and weather characteristics, the amount of substance deposited, and the regions of deposition vary (Roy et al., 2012b).
- The soil response factor (RF) characterizes alterations in soil properties within a specific area at a receptor site resulting from the considered pollutant deposition. In the case of terrestrial acidification, soil pH is a common indicator of soil property changes (Roy et al., 2012a).
- The effect factor (EF) quantifies the reduction in species richness of local biodiversity (at the receptor site) caused by the change in soil characteristics. Vascular plants are the only species group currently taken into consideration (Roy et al., 2014).

The relevance of terrestrial acidification is also recognized by the GLAM (Global Guidance for Life Cycle Impact Assessment Indicators and Methods) initiative (Frischknecht and Jolliet, 2019, 2016). GLAM is a project of the Life Cycle Initiative hosted by the UN Environment Programme. It has been running since 2013 and aims to provide guidance on the best currently available life cycle impact assessment indicators through processes of global consensus building. In phases one and two, GLAM has given recommendations on mostly midpoint indicators related to greenhouse gas emissions, fine particulate matter, land use, toxicity, water use, and soil quality, as well as freshwater eutrophication, terrestrial acidification, and cross-cutting issues (Frischknecht and Jolliet, 2019, 2016). In the third phase, the UN initiative promotes the development of damage-level characterization factors to foster the comparison of impacts across environmental stressors (Frischknecht and Jolliet, 2019, 2016).

In the current, third phase of the project, the aim is to provide operational models for relevant impact pathways for human health, resources, ecosystem quality, and ecosystem services. Within ecosystem quality, terrestrial acidification was included as a relevant impact pathway to be updated. The foundational work of Roy et al. (2014) has been used in many LCIA methods (e.g., ReCiPe, LC-IMPACT, TRACI). This study therefore revises the CFs for terrestrial acidification at endpoint level. This work includes the following improvements:

First, an update on the response factors. We apply new input datasets, as the weather and deposition that affect soil geochemical processes have changed. While the 2005-measurement-based atmospheric calculations used in Roy et al. (2012b) can still be considered a proxy for current atmospheric reactions influencing the atmospheric fate of emitted pollutants, the evolution of climate conditions in the last 20 years is likely to cause more significant changes in the soil response aspect. Moreover, the acid deposition data used by Roy et al. (2012a) are representative of the time period from 1961 to 1990, according to the National Climatic Data Center of the National Oceanic and Atmospheric Administration (Roy et al., 2012a), and thus need to be updated. It is also worth noting that what we have coined here as "Response Factors" were originally named "Sensitivity Factors" in the previous studies (Roy et al., 2014; Roy et al., 2012a). Due to the potential confusion between the latter term and that of "sensitivity analysis" (which can be performed on each component of the CFs), we have given a more appropriate name to these soil response factors.

Second, the effect factors implemented in the current state-of-the-art method rely on the work conducted by Roy et al. (2014) and Azevedo et al. (2013), quantifying the relative loss of species richness among the vascular plant group. In that work, only 0.6 % of the world's accepted vascular plant species (2,409 out of 383,671 registered species) are covered by the method (Roy et al., 2014). Moreover, the study at the biome scale does not encompass ecosystem-level damage and lacks spatial refinement regarding acidification's impact on biodiversity. Gade et al. (2021) developed new EFs covering 189,185 vascular plant species (49 % coverage) and ecoregion damage, addressing the previously mentioned concerns. The present study thus uses these updated effect factors.

Finally, the current approach only provides regional CFs by accounting for local species loss in the different biomes (Roy et al., 2014) but does not provide CFs for global losses. As a last step, the Global Extinction Probability (GEP) from Verones et al. (2022) converts local biodiversity losses into global losses.

This study provides novel CFs for terrestrial acidification, tackling the improvements mentioned above. The main contribution lies in the

calculation of a new response factor, which is then combined with the existing fate factors from Roy et al. (2012b) and the new effect factors from Gade et al. (2021). These three factors allow to calculate new CFs quantifying the regional impacts on vascular plant richness. Thanks to the GEP application in line with the harmonization goal of GLAM, global terrestrial acidification impacts are now compatible with those from other impact categories also assessing impacts in global Potential Disappeared Fraction of species.

2. Materials and Methods

Fig. 1 illustrates terrestrial acidification's impact pathway. The model takes into consideration anthropogenic emissions of NO_x, SO_x, and NH_3 released into the atmosphere that are subsequently deposited onto terrestrial systems and affect local biodiversity.

The methodology employed to calculate CFs for terrestrial acidification builds on the LCIA framework described by Jolliet et al. (2004) and used in Roy et al. (2014). The spatially explicit CFs quantify alterations in the global occurrence of vascular plant species, expressed as a potential disappeared fraction of species (PDF) per emission rate of acidifying substance (kg of pollutant emitted per year). As presented in Eq. (1), $CF_{s,p}$ expresses the characterization factor of a source location s emitting a marginal amount of a pollutant p (NO_x, SO_x, or NH_3) into the atmosphere. It quantifies the biodiversity impacts that a marginal additional emission of a pollutant p from the source location s has everywhere in the world. Indeed, the approach accounts for terrestrial biodiversity impacts on all pollutant-receiving areas r (adding up to a total of N_r regions around the world). The Global Extinction Probability (GEP) factor (Verones et al., 2022) is integrated to transform the potential regional, reversible species loss in receiving environments into a potential irreversible species loss at the global scale. The final CFs expressed in $PDF_{global}.yr.kg_{emitted}^{-1}$ provide a global coverage of impacts. They are calculated at grid cell level (2° × 2.5°) and are further aggregated to the country level, as described in Supporting Information 3.1 and 3.2.

$$CF_{s,p} = \frac{1}{N_r} \times \sum_r \left(FF_{s,r,p} \times RF_{r,p} \times EF_r \times GEP_{vascPlants,r} \right) \quad (1)$$

As explained in the subsections below, each of the CF's fate, soil response, effect, and GEP components are derived as matrices with a number of rows corresponding to the number of emission locations and a number of columns matching the number of deposition-receiving environments. Applying the Hadamard product across all four terms yields a matrix with spatially distributed impacts across receiving locations. The row average of this matrix results in a column vector reflecting the total impact per emitting location (Supporting Information 3.2).

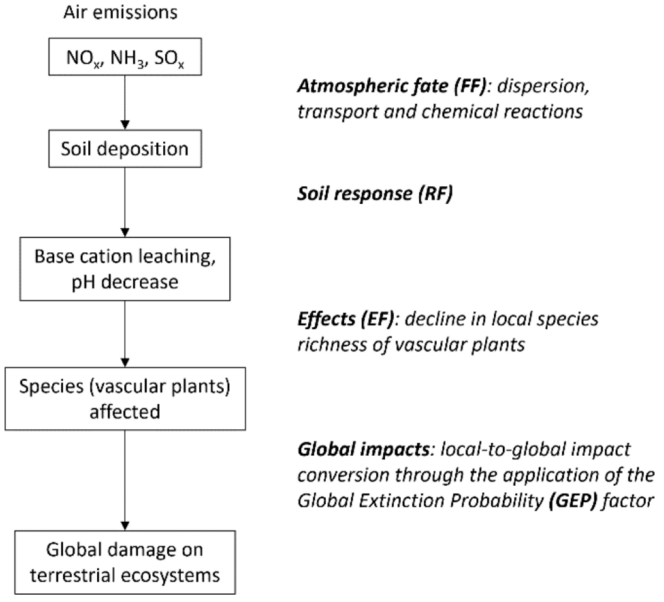

Fig. 1. Cause-effect pathway for terrestrial acidification (adapted from van Zelm et al. (2015)). The endpoint is damage to terrestrial ecosystems, expressed as a global potentially disappeared fraction of species (PDF).

2.1. Fate Factors

The fate factors ($FF_{s,r,p}$) consider the fate and transport of a pollutant (p) from an emission source (s) to a receiving environment (r) in terms of substance deposition rate (kg of nitrogen or sulfur deposited per year) per substance emission rate (kg of nitrogen or sulfur emitted per year). For example, $FF_{NewYork,Paris,NH3}$ tracks the mass fraction of NH_3 (pollutant p) getting deposited in Paris (receiving environment r) due to an emission in New York (emission source s). These factors were calculated using the GEOS-Chem atmospheric chemistry model, incorporating meteorological inputs from NASA (Roy et al., 2012b). As described in the study by Roy et al. (2012b), the 2005 reference year of GEOS-Chem undergoes perturbations through marginal additional emissions of NO_x, NH_3, and SO_x. The resultant fractional deposition values are placed in source-receptor matrices (SRMs). In these pollutant-specific matrices (e.g., NH_3), each row represents a source pixel location (e.g., New York), and each column denotes a receiving pixel location (e.g., Paris) (Roy et al., 2012b). The resulting FFs are given in three SRMs (one for each substance considered: NO_x, SO_x, NH_3) at a grid cell resolution of 2° × 2.5° (Roy et al., 2012b). Each cell of the pollutant-p SRM gives the fate factor corresponding to the substance being emitted in the source grid cell *cellS* and deposited in the receiving grid cell *cellR*, according to Eq. (2) (Roy et al., 2012b):

$$FF_{cellS,cellR,p} = \frac{dDeposition_{cellR,p}}{dEmission_{cellS,p}} \quad (2)$$

The matrix calculation defined in Eq. (1) requires a spatial transformation step, such that the receiving spatial units r of the $FF_{s,r,p}$ are ecoregions (rather than 2° × 2.5° grid cells) which is consistent with the spatial resolution of the RFs, EFs and GEPs (Supporting Information 3). Ecoregions define terrestrial areas with shared climates, hydrology, underlying geology, as well as flora and fauna (Dinerstein et al., 2017; Olson et al., 2001). The spatial transformation involves the area-based reassignment of information from one set of spatial objects to another. A spatial overlay helps identify the fractions of each target spatial unit (ecoregions) in each deposition cell. These fractions become the coefficients of a spatial transformation matrix (STM), such that a square FF source-receptor matrix, multiplied by the STM (*STMrec*), yields a fate matrix linking emission in 2°× 2.5° cells (matrix rows) to deposition in ecoregions (matrix columns).

In addition, on the emission side, to aggregate from the native scale of the CFs (the 2°× 2.5° emission units) to spatial units useful in applied LCA work (in this case, countries), sector-based emissions of NO_x, NH_3, and SO_2 gave weights to a second, likelihood-based spatial transformation (Supporting Information 2 and 3). In this case, the spatial transformation matrix (*STMem*) gives higher weight to areas with higher emissions, such that a country's CF reflects the emission conditions in that country. The approach is described in detail elsewhere (Henderson et al., 2021), and Supporting Information 4 provides a case study demonstrating it in Australia.

As a result, the adapted pollutant-p-specific FF matrix linking N and S emissions from source countries s to N and S deposition in receiving ecoregions r is calculated according to Eq. (3):

$$\left[FF_p\right]_{s,r} = \left[STMem\right]_{s,cellS} \times \left[FF_p\right]_{cellS,cellR} \times \left[STMrec\right]_{cellR,r} \quad (3)$$

2.2. Soil Response Factors

Pollutants reaching the receiving location subsequently affect the

local soil environment. Chemical reactions regulating the nitrogen cycle (nitrification, assimilation, ammonification, denitrification), nitrogen immobilization associated with organic matter decomposition, and plant respiration are affected by these acidifying inputs (Wallman et al., 2005). Depending on the characteristics of the soil, the buffering capacity of the soil solution may not neutralize the excessive acid deposition (Blaser et al., 1999; Dangles et al., 2004). As a result, the local pH (i.e. hydrogen ion concentration) may decrease (Roy et al., 2012a). As presented in Eq. (4) and based on the study by Roy et al. (2012a) the response factor ($RF_{r,p}$) quantifies the change in molarity of H^+ ions in the receiving ecoregion r due to a marginal increase of the acidifying pollutant p.

$$RF_{r,p} = \frac{d[H^+]_r}{dDeposition_{p,r}} \approx \frac{\Delta[H^+]_r}{\Delta Deposition_{p,r}} \approx \frac{10^{-pH_{new}} - 10^{-pH_{ref}}}{0.1 \times Deposition_{ref_{p,r}}} \quad (4)$$

As commonly used in LCIA studies dealing with models of complex nonlinear environmental processes, the marginal change approach is adopted here. It assumes that "an additional amount of a certain stressor introduces very small changes on top of a ceteris paribus background situation" (Huijbregts et al., 2011), which is indeed the case for terrestrial acidification's response factors. We also relied on the experiments, tests, and validations of various studies calculating acidification CFs to choose a 10 % marginal addition relative to the pollutants' current deposition rates (Roy et al., 2012a; Seppälä et al., 2006; van Zelm et al., 2007).

The RF of ecoregion r is thus calculated through the hydrogen ion molarity difference between a reference state and a 10 % increase of pollutant p deposition with respect to the reference state. The calculation is done at the ecoregion level given the shared climates, hydrology, and geology of the ecoregions. For each ecoregion and each of the three ionized main contributors to terrestrial acidification (NO_3^-, NH_4^+, SO_4^{2-}), the RF values are derived from the outputs of soil chemistry model PROFILE, run with and without marginal additions to pollutant deposition (Akselsson et al., 2006; Sverdrup et al., 2020; Warfvinge and Sverdrup, 1992).

PROFILE is a steady-state geochemical soil model originally developed for assessing critical loads of acidifying substances. PROFILE estimates the pH of soil layers across the 1-meter soil column based on input data covering local climate conditions (precipitation and temperature), soil layer characteristics (mineralogy, particle size distribution, etc.), and deposition (acidifying substances, dust, and salts). PROFILE calculates weathering rates and models geochemical processes related to nutrients (nitrification, immobilization, denitrification, nutrient uptake). With spatially differentiated inputs, the model can be used to calculate spatially differentiated pH responses. In this work, we start from the approach described by Roy et al. (2012a), updating input datasets where possible and modifying the calculation structure as described below.

Mineralogy is clearly a critical parameter input parameter to a geochemical weathering model such as PROFILE (Casetou-Gustafson et al., 2019). In the current modeling context, an effective model is one that can provide a relative response to acidifying inputs and differentiating responses between spatial units based on underlying geophysical parameters. The global model is not intended to predict pH responses at specific sites, and a site-specific, locally calibrated model will predict pH responses more accurately. Therefore, since we have used a harmonized, global database of soil mineralogy, we expect that any systematic errors in input data will be reflected in the model, but that these errors should have a restricted influence on the relative differences that the model is intended to highlight.

Table 2.1 summarizes the parameters and data sources used in the PROFILE model. The complete set of input data used in this study is available in Supplementary Information 1 and 2. ISRIC-WISE data (v1.2) (Batjes, 2012; ISRIC, 2012) provides a global description of soil properties at a 5×5 arcminute resolution and a 20 cm depth resolution across five soil layers. Within each of the five layers, multiple soil orders may be present, each with different properties. Based on the fraction of each soil order observed, we calculate representative averages of soil properties and mineral content for each layer at each site, as described in Supporting Information 1.

With inputs of varying resolutions, modeling at the highest resolution possible (e.g., the soil grid cells of five arc minutes, which are approximately 10 km at mid-latitude) can lead to a false sense of accuracy. In this case, ecoregions provide naturally meaningful modeling units, as characteristics relevant to the PROFILE model are typically shared across the ecoregions. Therefore, we have calculated area-weighted averages of the input parameters by ecoregion, and calculated soil response factors at the ecoregion scale.

Running the model twice for each ecoregion (once with reference state conditions and once with increased pollutant deposition) allows to derive the respective hydrogen ion molarities and thus the corresponding soil response factor. As shown in Eq. (4), the updated RFs are expressed in $mol_{H+}.L^{-1}.kg_{deposited}{}^{-1}.yr$.

The last step remaining to make the resulting pollutant p-specific RF vector applicable in Eq. (1) consists of transforming it into a matrix having its number of rows matching the number of emission source location s. This process is done through the application of the RF vector as it is to all emission locations as detailed in Eq. (5).

$$\left[RF_p\right]_{s,r} = [1]_{s,1} \times \left[RF_p\right]_{1,r} \quad (5)$$

2.3. Effect Factors

The effect factors (EFs) represent the decline in vascular plant species richness resulting from a marginal lowering of the soil pH (PDF per unit molar concentration of H^+ ions). As an update from previous work (Azevedo et al., 2013), Gade et al. (2021) had access to a larger inventory of terrestrial plants. Thanks to the use of popular science helping register species occurrence points in the global research network GBIF (Global Biodiversity Information Facility), they managed to cover 49 % of the world's reported vascular plant species (Gade et al., 2021). Although this represents a significant increase in species coverage compared to the previous study from Azevedo et al. (2013), they acknowledge that some areas in the world are under-sampled when it comes to GBIF species occurrences (Gade et al., 2021). Nonetheless, this inventory allowed them to derive ecoregion-specific species sensitivity

Table 2.1
Summary of data inputs to PROFILE.

Group	Parameter	Description	Resolution	Reference
Atmospheric deposition	Acidifying substances, wet and dry deposition	World Meteorological Organization produced a global assessment of precipitation chemistry and deposition	1 × 1°	(Vet et al., 2014)
Atmospheric deposition	Dust deposition	DustCOMM total annual deposition, PM_{10}.	1.9 × 2.5° (lat, lon)	(Kok et al., 2021) Data: (Kok and Adebiyi, 2021)
Weather characteristics	Precipitation, temperature	WorldClim v2.1 historical climate data	10′ (arcmin) (1/6°)	(Fick and Hijmans, 2017)
Soil	Multiple; see Supplementary Information	ISRIC-WISE global harmonized soil dataset	5 × 5′ (arcmin)	(Batjes, 2012; ISRIC, 2012)

distribution (SSD) curves, which then enabled them to determine the local Potentially Not Occurring Fraction of species (PNOF) (Gade et al., 2021). In order to better represent the ecoregion soil acidity characteristics, the new method developed computed area-weighted EFs accounting for the local probability of receiving acidifying substances and for the probability of species being impacted at each pH unit (Gade et al., 2021). Eq. (6) from Gade et al. (2021) estimates the EF of ecoregion r where $A_{k,r}$ is the area that pH unit k covers within the ecoregion r, and $A_{tot,r}$ is the sum of $A_{k,r}$ for all pH units k considered. Finally, following the guidelines outlined in the Global Guidance on Environmental Life Cycle Impact Assessment Indicators – Volume 2 (Frischknecht and Jolliet, 2019) and for consistency purposes, the PNOF values were transformed into PDF using a conversion factor of 1.

$$EF_r = \frac{1}{A_{tot,r}} \times \sum_k \left(A_{k,r} \times \frac{dPNOF_{k,r}}{d[H^+]_r} \right) \quad (6)$$

In this work, due to the same mathematical constraints explained in the previous subsection (2.2), a transformation step was required to transform the EF vector into a dimension-matching matrix (Eq. (7)).

$$[EF]_{s,r} = [1]_{s,1} \times [EF]_{1,r} \quad (7)$$

2.4. Global Extinction Probability

The Global Extinction Probability (GEP) (Verones et al., 2022) is a dimensionless scaling factor, ranging between 0 and 1 and used to convert regional PDF values (representing a reversible impact) into global PDF values (irreversible impact). The GEP accounts for species

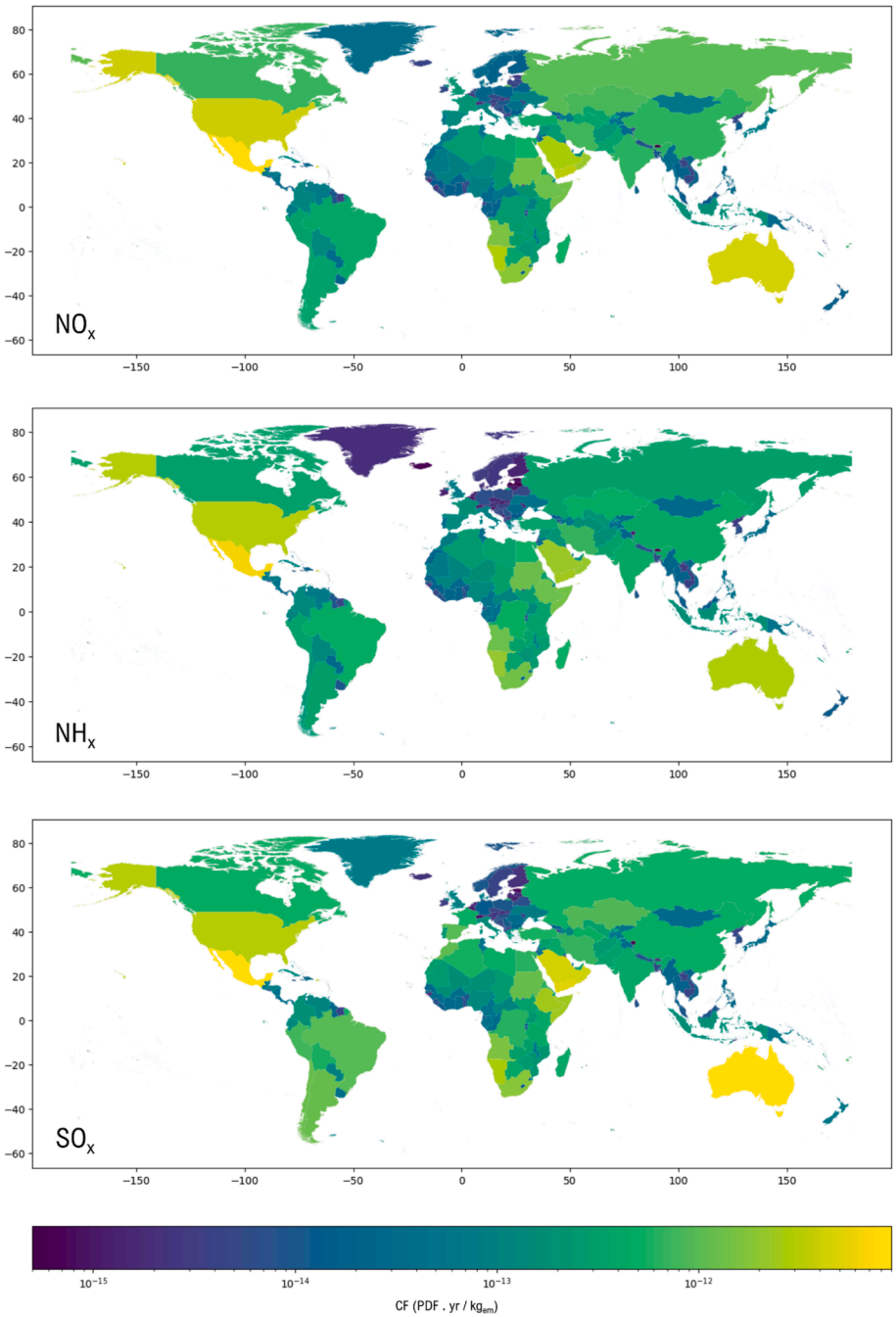

Fig. 2. Endpoint country Characterization Factors for Terrestrial Acidification for NO_x, NH_x, and SO_x.

threat level, range area, and occurrence certainty. It is available at several scales; when applied, the raw GEP values are adjusted such that the sum of all GEP values equals one. Here the GEP for vascular plants is used at the ecoregion level, since it is the native scale of the effect factors.

Verones et al. (2022) highlight that it is important to first aggregate the regional CFs to the required resolution and then apply the corresponding GEP. It ensures that all GEP values still add up to one. This recommendation is straightforward when the receiving environment on which the pollutant is deposited is the same as its emission location. In our case, multiplying the regional country CFs with the corresponding country GEP would be in line with the recommendation. However, terrestrial acidification involves the transportation of the emitted pollutant. By following the recommendation, one would thus assume that the GEP in the emitting country and in the receiving ecosystem are the same. However, this is not the case. Therefore, the GEP had to be applied on the receiving side (ecoregion level), where the impact actually takes place. This consideration, occurring before the averaging aggregation to the final country CFs, forced the transformation of the ecoregion GEP row-vector into a matrix matching the FF matrix size (Eq. (8)). In order to stay within the probabilistic framework described by Verones et al. (2022) and keep the sum of all GEP values equal to 1, the modified GEP matrix's elements were all divided by the number of countries ($N_{countries} = 205$), assuming an equal share to each emission location (see 3.3).

$$[GEP_{vascPlants}]_{s,r} = \frac{1}{N_{countries}} \times [1]_{s,1} \times [GEP_{vascPlants}]_{1,r} \quad (8)$$

3. Results and Discussion

3.1. New damage-level terrestrial acidification CFs

Combining the fate, soil response, effect factors and the GEP together as defined in Eq. (1), results in the characterization factors for NO_x (top map), NH_x (middle map) and SO_x (bottom map) displayed in Fig. 2.

The distribution of CFs is similar across the pollutants, reflecting similar transport for each pollutant, and thus similar receiving ecoregions. However, they differ significantly from the individual contributing factor (RF, EF) maps detailed in the following subsections. Indeed, unlike many LCIA impact categories, terrestrial acidification implies transportation of emitted substances. The impacts of an emission released in a certain country do not solely occur in this specific country. According to the transportation and deposition patterns given by the fate factors, the acidifying substance is distributed and can cause harm in ecoregions a great distance away from the emission location. Therefore, a selected country's CF must be interpreted as the overall and aggregated impact that an emission from this specific country has in the whole world.

Supporting Information 4 displays the building of the CF in more detail, using Australia as an example. Here it is clearly shown that a country CF for terrestrial acidification is composed of two main elements. First, on the emission side, the fate factor translates where emissions from different parts of the country eventually deposit (Fig. S3a and b). The second element expressed in $PDF_{global}.yr.kg_{deposited}^{-1}$ reflects the actual impact that the deposited acidifying substance has on biodiversity (Fig. S3c). It combines the other three terms of the CF: the local soil response (RF), the local effect (EF), and the corresponding GEP. In summary, to calculate the final country CF, the impact values from the second element are "weighted" by the fate mass fractions which indicate how much of the pollutant released ends up in each of the receiving ecoregions. This procedure can explain why some countries may be covered by quite acidification-sensitive ecoregions while showing a relatively low CF (e.g.: Laos, Cambodia) and vice-versa (e.g.: Russia). In the case of Australia, however, the emissions from the main sources of acidifying pollutants (ore extraction mines) mostly stay within the country and deposit in the acidification-sensitive ecoregions in the state of Western Australia. Both elements of the final CF are thus on the high side of their respective scales, driving the Australian CF value up.

Terrestrial acidification is already accounted for in various LCIA methodologies based on the work of Roy et al. (2014). One of them, the recent LC-IMPACT methodology, implements Ecosystem Quality CFs that are "regionalized with a global coverage" and "include aspects of species extinction vulnerabilities" (Verones et al., 2020). With such characteristics, the existing LC-IMPACT terrestrial acidification CFs offer a reference point to qualitatively validate the CFs calculated in the present study.

Fig. 3 shows the values for each country of the comparative ratio between the novel characterization factors calculated in this study and the ones implemented in LC-IMPACT. The statistics (Supporting Information 5) reveal that about 38 % of the new CFs are within the same order of magnitude as the previous ones (grey color), whereas 22 % lie between one and five orders of magnitude lower (blue colors), and 40 % between one and four orders of magnitude higher than the existing factors (red colors). This quantitative analysis leads to two main conclusions. On the one hand, the rather even ratio distribution between lower, similar, and higher values indicates an overall similar global average value to the existing method. Such observation thus helps validate the updated calculation pathway. On the other hand, the country-specific discrepancies in ratio values confirm the need and benefit of updated factors in LCIA methodologies.

The following subsections explore and evaluate the different updated components of the CFs in order to discuss their contributions to the final values.

3.2. Update and contribution of soil RFs

The calculations run by the geochemistry model PROFILE resulted in substance-specific soil response factors at ecoregion scale (Fig. 4). The values range from 4.5×10^{-17} to 5.3×10^{-9} $mol_{H+}.yr.L^{-1}.kg^{-1}_{deposited}$ with 80 to 92 % of them between 4.5×10^{-17} and 8.0×10^{-10} $mol_{H+}.yr.L^{-1}.kg^{-1}_{deposited}$. The patterns of distribution are similar across the three substances tested. The highest RFs are found along the US-Canada Pacific coast all the way up to Alaska, in Australia (in particular in the southwestern part of the country), along the southwestern coast of the Arabic Peninsula, and around the Taklimakan Desert in Central Asia. On the contrary, the Taklimakan Desert as well as the Sahara Desert, and the center of the Arabic Peninsula stand out as the ecoregions with the lowest RFs. A low RF typically reflects a high Acid Neutralizing Capacity (ANC) of the corresponding ecoregion's soil. Comparing the RF values with Roy et al. (2012a) "soil sensitivity factors" could have helped validate the modeling and calculation approach. However, the differences in units ($mol_{H+}.yr.ha^2.L^{-1}.kg^{-1}_{deposited}$ instead of $mol_{H+}.yr.L^{-1}.kg^{-1}_{deposited}$ in the present study) and resolution (99 515 grid cells worldwide in contrast to our 767 ecoregions) made this testing impossible.

As indicated in Fig. 4, the grey ecoregions represent the receiving locations where PROFILE has calculated a 0-pH difference between the reference deposition state and after a 10 % increase in pollutant deposition. This trend can only be observed for nitrogen-based acidifying substances and for a total of 110 ecoregions distributed in northwestern Canada, southern Argentina, southern Australia, the most southern point of Africa, eastern Siberia, and southern Russia. Such a value would translate into soils that have very high ANCs. However, these results are contradictory to the rest of the values found for the sulfur-based pollutant which shows quite high RFs for these regions. This reflects an area of potential improvement in the modeling, as PROFILE can account for the nitrification and/or denitrification processes of the nitrogen cycle in different ways. Here, we have accounted for nitrification and denitrification of ammonia compounds by adjusting deposition in keeping with Bouwman et al. (2002), rather than allowing PROFILE to model these processes. While our modeling of the N cycles is consistent with Roy et al. (2012a), further investigation should be carried out to

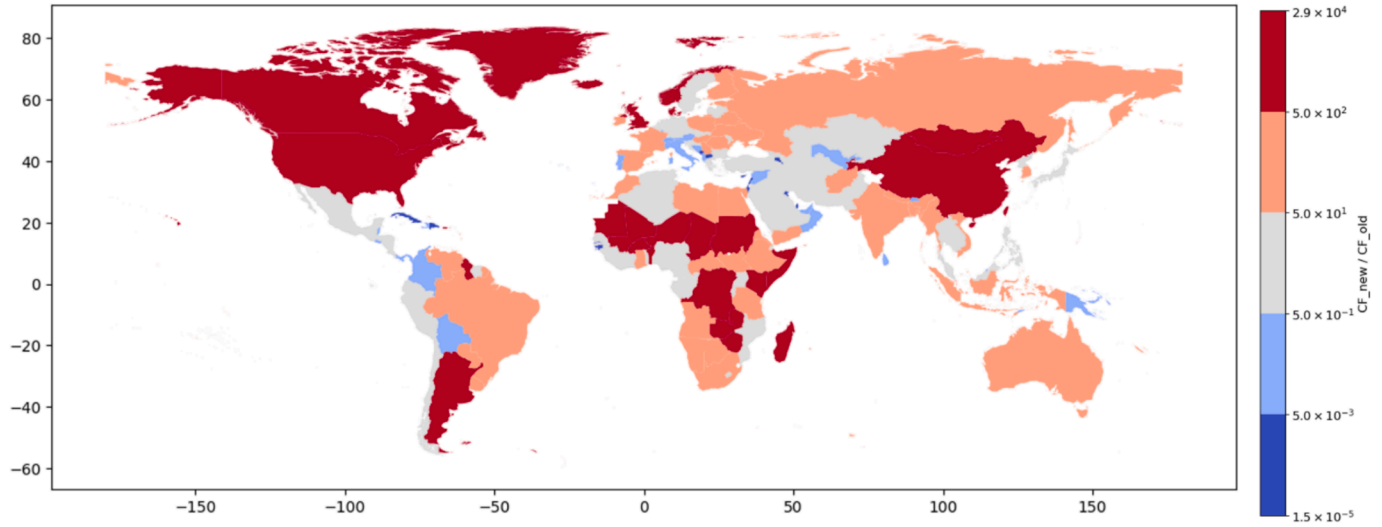

Fig. 3. Ratio of the updated ("new") Terrestrial Acidification CFs over the existing ("old") CFs from LC-IMPACT. Red areas have a higher CF in the new version, blue ones are smaller. Grey areas experience little to no change. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

investigate the accuracy of the soil response factors with respect to nitrogen substances.

3.3. Application of the GEP

This study integrates the GEP into the regionalized calculation of global species loss in order to account for the endemism and vulnerability of species in the deposition area of the acidifying substance. If the GEP were to be applied on the emission side, as suggested by the original GEP framework (Verones et al., 2022), it would then be assumed that both GEPs in the receiving and emitting locations are the same. However, the ratio comparison of CF values derived from the application of the GEP on the emission (denominator) and receiving (numerator) sides shows this is not the case (Supporting Information 6). The ratios are (almost) all inferior to one and the difference between the two approaches can lead to up to a five-order-of-magnitude difference. One element of explanation for these observed discrepancies is the size of the emitting country. Indeed, a very small emitting country tends to have a very small GEP as it is covered by a much lower number of pixels with GEP values than substance-receiving ecoregions (Verones et al., 2022). This thus drives up the ratio value on the map. An additional contributor to discrepancies is the fate factor. The larger the transport distance, the larger the potential for a difference in GEP values.

This analysis confirms the need to apply the GEP on the receiving side (ecoregion level), where the impact is actually happening. To do so, we made a value choice and assumed an equal share of the GEP to each emission location (Eq. (8)). This value choice preserves the probabilistic framework of the GEP (i.e., the sum of all GEP values equaling one).

With this conceptual choice, a better comparison of global impacts with other impact categories is possible. However, some questions were raised when making value choices to adapt the GEP framework to such transportation-based impact categories (Eq. (8)). Because the aggregation of the final characterization factors to country level uses emission weighting, we have made the decision to apply the GEP normalization equally across countries. This reflects a value decision to treat all emitting countries equally with respect to the ecoregions that their emissions may affect. Nevertheless, in the aim of harmonizing CF calculation methods, this concern highlights the need for clear guidance on how to apply the GEP to regional CFs when accounting for differences in emission and receiving sides (e.g. plastic entanglement regional CFs (Høiberg et al., 2024)). Indeed, without full scientific consensus, different approaches may be chosen and the interpretations based on the comparison of results across impact categories may differ.

3.4. Implications in LCA and outlook

The novel damage-level characterization factors presented in this study are important for a better quantification of terrestrial acidification impacts on biodiversity. The method has been discussed and presented several times to a global, expert audience within the GLAM project of the life cycle initiative and is the consensus-based outcome of that exercise. As shown in section 3.1, it has the potential to change the practitioner's conclusions and interpretation of impacts. Compared to the currently implemented values in LC-IMPACT (Verones et al., 2020), some countries are now responsible for higher or lower vascular plant species loss than they were previously.

In order to avoid any misuse and misinterpretation of an impact assessment of terrestrial acidification, the practitioner should be aware of the following points.

First, the presented CFs should not be applied to offshore activities. Due to the grid-cell-to-country spatial transformation applied on the emission side, the adjusted fate factors only account for land-based emissions and omit any acidifying pollutants being released offshore. We recommend that non-terrestrial activities be assigned the global average.

Second, an impact assessment with the presented CFs may lead to an overestimation of the relative contribution of terrestrial acidification to biodiversity loss as compared to the other drivers. Indeed, the model considers only vascular plants as a proxy for the whole terrestrial realm when quantifying the impacts of terrestrial acidification on ecosystem quality. The focus on vascular plants is a function of the available effect factor data, which has rightfully focused on the species group most affected by acidification. However, most other impact categories, such as land use, eutrophication, or climate change, include various taxonomic groups in the quantification of their impacts on terrestrial species (Verones et al., 2020).

Nevertheless, thanks to the Global Extinction Probability, terrestrial acidification can be included in damage-level LCIA methods and be cross-compared with other environmental stressors assessing the potential global loss of terrestrial biodiversity.

Since terrestrial acidification is not the only impact category involving pollutant transportation between the release point and the impact location (similar to photochemical ozone formation or marine eutrophication), this study is the first to tackle the issue of GEP inclusion

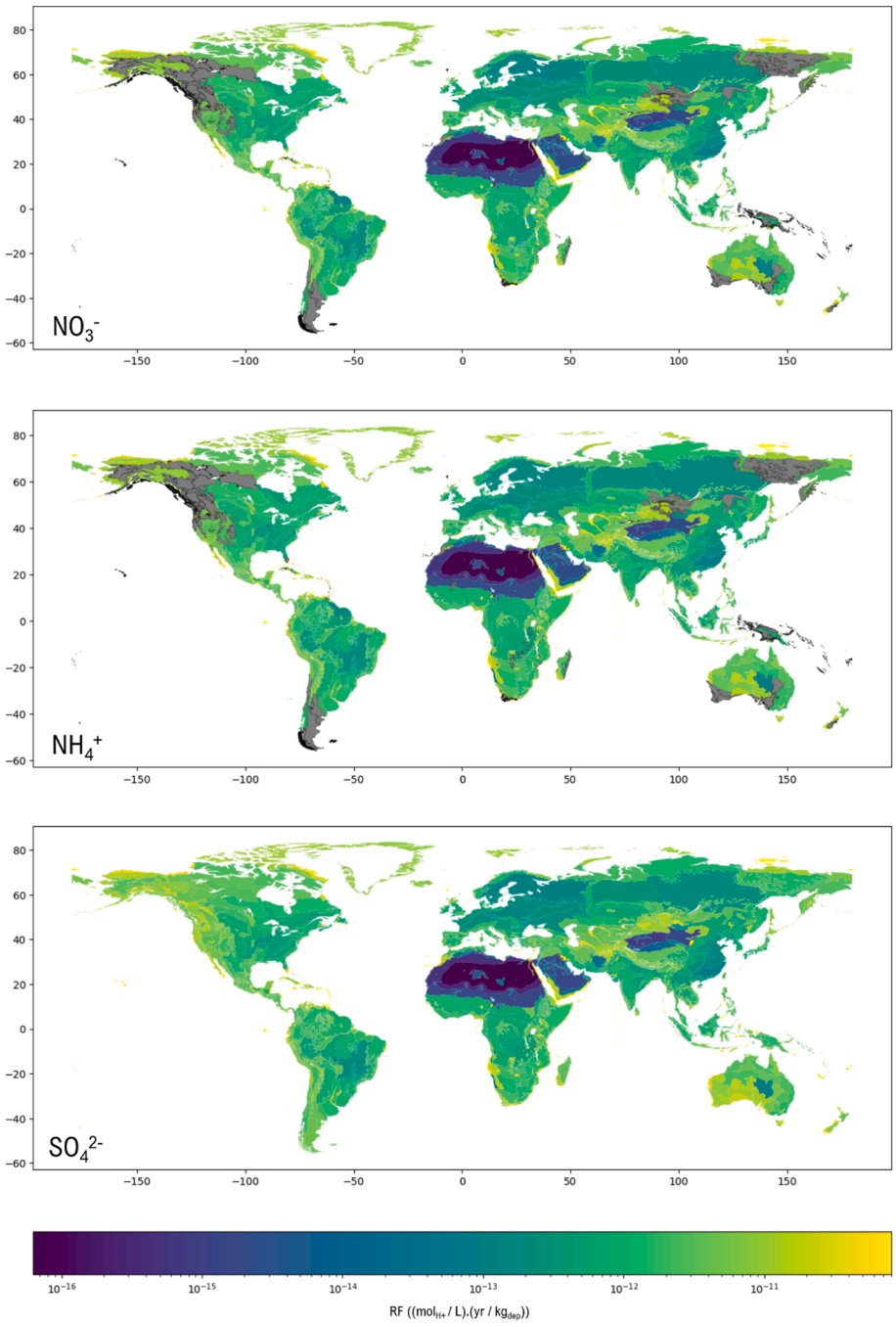

Fig. 4. Marginal RFs obtained for the three acidifying substances tested. The grey areas represent ecoregions where RFs = 0 $mol_{H+}.L^{-1}.yr.kg^{-1}_{deposited}$.

with airborne emissions. Therefore, reaching a scientific consensus and methodological harmonization on the GEP application would be beneficial for the LCIA community (specifically when the impact does not happen in the same region as the impact driver). This research also enables future work to evaluate changes in the effect factor with respect to species group coverage. Exploring the more indirect relationships between acidification and other species groups will help grasp the impact pathway in its entirety. Furthermore, this study calls for an update of the fate component. Not only would it benefit from updated atmospheric modeling and weather data, but there is also a need to take offshore emissions into consideration. As the ocean is becoming increasingly exploited due, in part, to trade and offshore drilling, proper accounting of these emissions reaching terrestrial systems is an important area for future research. Finally, this work will also enable the investigation of potential future terrestrial acidification impacts on biodiversity based on different climate scenarios.

4. Conclusion

Together with the other impact categories in ecosystem quality and the other Areas of Protection, this updated model will be an important step forward in assessing damage-level impacts from human activities within the framework of LCIA. It provides cutting-edge characterization factors combining spatially transformed country-to-ecoregion fate factors (adapted from Roy et al. (2012b)), updated soil response factors at ecoregion level, and recently revised ecoregion effect factors which account for the potential regional loss of vascular plant species (Gade et al., 2021). In addition, the integration of the Global Extinction

Probability allows to convert the local decline in species richness into a global species loss. With this fourth component, terrestrial acidification impacts are now comparable to those from other drivers of biodiversity loss estimated in global Potential Disappeared Fraction of species. However, it is important to keep in mind that Life Cycle Assessment is a relative tool and cannot grasp all the dimensions of environmental management and sustainability on its own. Nevertheless, its combination with complementary methodologies such as Quantitative Risk Assessment will bring valuable support to decision-makers.

CRediT authorship contribution statement

Marion N. Lebrun: Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Formal analysis, Data curation, Conceptualization. **Martin Dorber:** Writing – review & editing, Methodology, Formal analysis. **Francesca Verones:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Andrew D. Henderson:** Writing – review & editing, Supervision, Software, Methodology, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecolind.2025.113241.

Data availability

The final country characterization factors and the intermediate soil response factors at the ecoregion level are available on Zenodo: https://doi.org/10.5281/zenodo.13833176.

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