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Global regionalized characterization factors for phosphorus and nitrogen impacts on freshwater fish biodiversity

Jinhui Zhou^{a,*}, José M. Mogollón^a, Peter M. van Bodegom^a, Arthur H.W. Beusen^{b,c}, Laura Scherer^a

^a Institute of Environmental Sciences (CML), Leiden University, Leiden, the Netherlands

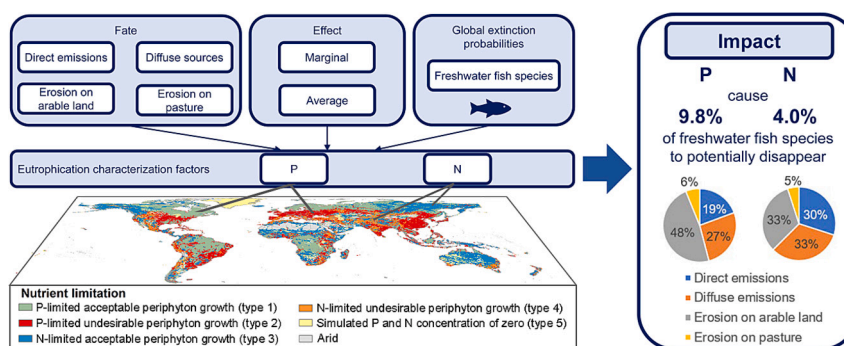
^b PBL Netherlands Environmental Assessment Agency, the Hague, the Netherlands

^c Department of Earth Sciences, Utrecht University, Utrecht, the Netherlands

HIGHLIGHTS

- Considering nutrient limitation in assessments of eutrophication impacts
- Evaluating global species loss of freshwater fish due to eutrophication
- Regionalizing characterization factors at half-degree resolution
- Highlighting country-level contributions of different nutrients to species loss
- Identifying global species loss of 13.8 % and erosion as the major contributor

GRAPHICAL ABSTRACT



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ABSTRACT

Inefficient global nutrient (i.e., phosphorus (P) and nitrogen (N)) management leads to an increase in nutrient delivery to freshwater and coastal ecosystems and induces eutrophication in these aquatic environments. This process threatens the various species inhabiting these ecosystems. In this study, we developed regionalized characterization factors (CFs) for freshwater eutrophication at 0.5×0.5 -degree resolution, considering different fates for direct emissions to freshwater, diffuse emissions, and increased erosion due to agricultural land use. The CFs were provided for global and regional species loss of freshwater fish. CFs for global species loss were quantified by integrating global extinction probabilities. Results showed that the CFs for P and N impacts on freshwater fish are higher in densely populated regions that encompass either large lakes or the headwaters of large rivers. Focusing on nutrient-limited areas increases country-level CFs in 51.9 % of the countries for P and 49.5 % of the countries for N compared to not considering nutrient limitation. This study highlights the relevance of considering freshwater eutrophication impacts via both P and N emissions and identifying the limiting nutrient when performing life cycle impact assessments.

* Corresponding author at: Institute of Environmental Sciences (CML), Leiden University, P.O. Box 9518, 2300 RA Leiden, the Netherlands.

E-mail address: j.zhou.12@cml.leidenuniv.nl (J. Zhou).

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1. Introduction

Humans have altered the phosphorus (P) and nitrogen (N) cycle by harvesting P and N from the geosphere and the atmosphere, respectively, and using them to boost agricultural production (Jenny et al., 2016). Mineral and synthetic fertilizers have contributed to a five-fold increase in global food production, boosting and supporting the 2.5-fold population growth during the past five decades. The long-term use of fertilizer and phosphate mining has doubled phosphorus (P) and tripled nitrogen (N) inputs to the global environment (Bouwman et al., 2009; FAO, 2023; Zhang et al., 2021). This has led to problems such as eutrophication (Jenny et al., 2016; Schindler and Vallentyne, 2008) and species toxicity (Kocour Kroupova et al., 2018). These issues negatively affect aquatic ecosystems and can also be harmful to human health (Bryan and van Grinsven, 2013; Vonlanthen et al., 2012). The rising global food demand and its concomitant fertilizer requirements indicate that eutrophication in freshwater systems is likely to continue increasing in the near future (Beusen et al., 2022; Mogollón et al., 2018a, 2018b; Nedelciu et al., 2020).

Life Cycle Assessment (LCA) is one approach used to evaluate the environmental impacts of eutrophication (Muralikrishna and Manickam, 2017). Ecosystem quality, represented by biodiversity, is one of the core areas of protection regarding which impacts are assessed. Characterization factors (CFs) link emissions to biodiversity loss (Payen et al., 2019), composed of multiple sub-factors following a cause-effect chain from midpoint factors (fate and exposure factors) to endpoint factors (effect and damage factors) (Rosenbaum and Hauschild, 2015).

In freshwater ecosystems, fish biodiversity is considered a good indicator of ecosystem health (Villéger et al., 2017; Whitfield and Elliott, 2002). This indicator has previously been used in LCA: CFs for freshwater fish biodiversity have been developed to assess the impacts of various environmental stressors, such as climate change (de Visser et al., 2023; Hanafiah et al., 2011), water consumption (Hanafiah et al., 2011; Pierrat et al., 2023), and hydropower (Turgeon et al., 2021). All these studies have been at a global scale and at different degrees of regionalization from biomes to the grid level. Due to the various environmental conditions (including the prevalence of nutrient limitations) and species compositions, impacts of P and N emissions on fish species vary spatially, which also requires regionalized CFs. Studies on global regionalization for P include LC-IMPACT (Azevedo et al., 2020), ReCiPe2016 (Huijbregts et al., 2017), and Jwaideh et al. (2022). However, the representativeness of the spatial variability of P impacts on freshwater species is insufficient since these studies only distinguish the relationships between species and P for four geographical zones globally. Other studies include fate factors (FFs) (Zhou et al., 2022) and effect factors (EFs) (Zhou et al., 2023) for N at half-degree resolution. However, none of these studies cover more than one nutrient concerning freshwater eutrophication.

In 2017, the United Nations Environment Programme (UNEP) launched the third phase of the Global Life Cycle Impact Assessment Method (GLAM) project as a part of the "Life Cycle Initiative" to standardize and review life cycle methods globally (Payen et al., 2019). GLAM advocates for the use of spatially explicit models with global coverage to develop regionalized CFs. Specifically for eutrophication, an additional recommendation from an earlier GLAM phase was to improve the modeling of physical and biogeochemical processes (Payen et al., 2019). Moreover, GLAM recommended the consideration of global as opposed to just local or regional species loss (Verones et al., 2019), for which global extinction probabilities (GEPs) have just recently been developed (Verones et al., 2022).

Our study aims to develop regionalized CFs for both P and N over the world (with a 0.5×0.5 -degree spatial resolution and yearly time step) while following the GLAM recommendations. Following the methods of Zhou et al. (2022, 2023), we coupled the local nutrient fate and concentrations from the Integrated Model to Assess the Global Environment – Global Nutrient Model (IMAGE-GNM) (Beusen et al., 2015) to 41 years

of freshwater fish species data (13,920 freshwater fish species) to estimate the impact at the local ecosystem level. Next, we employed GEPs (Verones et al., 2022) to extrapolate the regional species loss to global species loss. Finally, we integrated nutrient limitation information with the assessment of eutrophication impacts. Our study provides practical indicators for economic actors to estimate the P and N impacts on the global freshwater ecosystem and serves as a roadmap for impact assessment of eutrophication in other ecosystems.

2. Methods

2.1. Characterization factors

In LCA, CFs connect the life cycle inventory (e.g., emissions) to impacts (e.g., on the ecosystem). Ideally, CFs are composed of all the sub-factors in the cause-effect chain: fate, exposure, effect, and damage factors (Rosenbaum and Hauschild, 2015). In the case of eutrophication, FFs describe the fate of contaminants from emissions due to human activities eventually transported to the (aquatic) environment. In this study, we distinguished emission routes into direct emissions to freshwater, diffuse emissions excluding erosion, and increased erosion caused by agricultural land use and calculated FFs for P and N in 2010 by employing the approach of Zhou et al. (2022) (their FFs for N were developed for the year 2000). The same methodology was employed to assess the fate of P and N, considering that while residence times for various chemical forms may differ, our focus is on total phosphorus (TP) and total nitrogen (TN), mitigating the impact of the transformation time for different P and N forms. Exposure factors (XFs) translate the total concentration of a released substance to a concentration that is more closely linked to the effects on species. For marine eutrophication, as defined by Cosme et al. (2015), they translate the total concentration of a nutrient to the concentration of dissolved oxygen (Cosme et al., 2015). However, excessive nutrients also affect species in other ways: changes in the energy transfer in food webs (Wang et al., 2021), noxious toxins produced by harmful algae (Chorus and Welker, 2021), diminishing light penetration by algae blooms (Lehtiniemi et al., 2005), and decreases in cross-taxon congruence due to loss of balance between species (Wang et al., 2021). These phenomena highlight the importance of considering the comprehensive effects of nutrients on biodiversity loss along with hypoxia when assessing the impacts of nutrient emissions. Several studies include the overall effects by linking the nutrient concentration and the species richness (Azevedo et al., 2013; Zhou et al., 2023). Based on the species richness-nutrient relationships, several LCA studies (e.g., LC-IMPACT (Azevedo et al., 2020), ReCiPe2016 (Huijbregts et al., 2017), and Jwaideh et al. (2022)) have conceptualized and calculated CFs from fate to effect without considering exposure. We followed a similar approach to these studies.

Damage to biodiversity is often expressed through the potentially disappeared fraction (PDF) of species. In contrast to previous studies (Rosenbaum and Hauschild, 2015) that assessed impacts through the potentially affected fraction, we directly linked the nutrient concentration levels (kg/m^3) to PDF by regressing species sensitivity distributions (SSDs) for P following the method of Zhou et al. (2023) to calculate EFs for P ($\text{PDF}\cdot\text{m}^3/\text{kg P}$) and N ($\text{PDF}\cdot\text{m}^3/\text{kg N}$).

By linking fate and effect factors, we derived CFs for impacts on regional species loss (Eq. (1)). Additionally, we employed Global Extinction Probabilities (GEPs) (Verones et al., 2022) of freshwater fish for CFs that assess the global impacts of freshwater eutrophication (Eq. (2)). GEPs denote a scaling factor for potential regional species loss with respect to potential global extinctions (Verones et al., 2022). GEPs are available for 20 species groups across marine, terrestrial, and freshwater ecosystems. We selected the GEPs for freshwater fish to match the species group used in the EFs of the regional CFs, as recommended by Verones et al. (2022).

We estimated eight types of CFs for the combination of different emission routes and the marginal vs. average methods in assessing EFs

(i.e., marginal and average CFs for direct emissions to freshwater, diffuse emissions excluding erosion, erosion caused by land use transition from natural land to arable land, and that to pasture). Detailed descriptions of FFs, EFs, and GEPs can be found in Zhou et al. (2022, 2023) and Verones et al. (2022).

$$CF_{regional,e \rightarrow i} = \sum_j FF_{e \rightarrow i \rightarrow j} \times \frac{EF_j}{V_j} \quad (1)$$

$$CF_{global,e \rightarrow i} = \sum_j FF_{e \rightarrow i \rightarrow j} \times \frac{EF_j}{V_j} \times GEP_j \quad (2)$$

In Eq. (1) and Eq. (2), $CF_{regional,e \rightarrow i}$ and $CF_{global,e \rightarrow i}$ denote the cumulative CFs for regional species loss and global species loss in the source grid cell i (at a half-degree resolution) for an emission route e (erosion, diffuse sources excluding erosion, and direct emissions to freshwater). The subscript j indicates the downstream receptors of source cell i connected by advection. V_j is the water volume of the receptor j (m^3). The unit of CFs for erosion is $PDF \cdot year / (m^2 \cdot year)$, while that for diffuse sources excluding erosion and direct emissions to freshwater is $PDF \cdot year / kgX$, where X represents P or N.

For the additional erosion caused by agricultural land use, we subtracted the initial CFs of arable land/pasture and natural land to express the eutrophication impact of human activities ($CF_{erosion \rightarrow i, landuse}^*$, $PDF \cdot year / (m^2 \cdot year)$, Eq. (3)).

$$CF_{erosion \rightarrow i, landuse}^* = CF_{erosion \rightarrow i, landuse} - CF_{erosion \rightarrow i, natural\ land} \quad (3)$$

To link our CFs to the life cycle inventory, we suggest using molar mass-based conversion factors for P and N compounds (e.g., phosphate, ammonium, nitrate), although this is a simplification because the compounds can differ in solubility and bioavailability. Details can be found in Table 3 in Supplementary Material 3.

2.2. Model and data

SSDs were estimated by coupling the fish occurrence data to nutrient concentration levels within the same locations (i.e., in half-degree pixels) for each year (from 1970 to 2010). We compiled the occurrence data for freshwater fish species following the approaches of Barabossa et al. (2020, 2021) but included species recorded in both lotic and lentic habitats. We harmonized the species names based on FishBase (Boettiger et al., 2012) and Tedesco et al. (2017). In total, we acquired 13,920 freshwater fish species and 5,427,740 occurrence records from 1970 to 2010. Our dataset provides a comprehensive coverage of global freshwater fish species, as the cumulative discoveries to date amount to 18,642, including extinct and recently identified species (Fricke et al., 2023). Until 2010, 15,170 species were described (of which we covered 92%), with hundreds more added each year (Eschmeyer et al., 2010). Compared with a widely used dataset of global regionalized estimates of freshwater fish species (Abell et al., 2008), our identified species numbers in 75% of ecoregions (= 290/389 ecoregions) exceed their estimates, showing strong region-specific representativeness. A scarcity of occurrence records was found in East Asia and polar regions. The species number in each ecoregion and a comparison with Abell et al. (2008) can be found in Section S4 of Supplementary Material 1 and in Supplementary Material 2.

We used results from IMAGE-GNM (Beusen et al., 2022) from the year 2010 (the latest non-scenario year) to obtain estimates for nutrient loadings, emissions, and concentrations for TP and TN in order to derive FFs, EFs, and emission-weighting estimates (see Eq. (4) below). IMAGE-GNM was designed to be spatially explicit and dynamic, with a resolution of 0.5×0.5 degrees and a yearly time step (Beusen et al., 2015, 2016). This model uses a mechanistic approach to calculate nutrient transport processes based on the hydrological cycle. The hydrological patterns were simulated by incorporating the PCRaster GLOBAL Water Balance model (PCR-GLOBWB) (Sutanudjaja et al., 2018). For this

study, we applied the hydrological and nutrient estimates of IMAGE-GNM from 1970 to 2010 for generating SSDs for P (SSDs for N were derived from Zhou et al. (2023)).

2.3. Freshwater nutrient limitation

In a specific water body, eutrophication can be limited by P or N, or co-limited by both nutrients, depending on the ratio of P and N content and their concentrations (Francoeur et al., 1999). Although the N:P ratio that determines the limited nutrients for algal growth may vary depending on the organism and the study sites, remarkably consistent N:P ratios were found in total soil pools and the soil microbial biomass (Cleveland and Liptzin, 2007). Such a ratio is the Redfield N:P ratio and has been widely used for evaluating the limitation of nutrients in marine, soil, and freshwater systems (Cleveland and Liptzin, 2007; Jarvie et al., 2018; Rhee and Gotham, 1980; Rhee, 1978; They et al., 2017).

Following McDowell et al. (2020), we assess the locations affected by nutrient excess and undesirable periphyton growth, which are directly tied to hypoxic "dead zones" and the loss of higher trophic species, such as fish (Wurtsbaugh et al., 2019). Note that periphyton is a mixture of algae, cyanobacteria, heterotrophic microbes, and detritus that are attached to surfaces on river beds, and nutrient limitation can also affect the growth of other organisms, such as freshwater algae (McDowell et al., 2020). Still, periphyton growth and its relationship with the Redfield ratio are considered an appropriate indicator of the eutrophication potential. According to McDowell et al. (2020), the nutrient limitation and the periphyton growth state are determined by a Redfield N:P ratio of 7:1 (by mass) and concentration thresholds for P and N in non-arid zones. Non-arid zones were defined as regions where discharge is nonzero. McDowell et al. (2020) differentiated limitations and the periphyton growth state into four types. We added a fifth type of acceptable periphyton growth without defining the limiting nutrient when the simulated concentrations of both P and N are zero. Simulated concentrations of zero may imply very low concentrations that cannot be captured by the model process. This occurs, for example, in endorheic river basins or at the outflow of a lake with a long residence time of >30 years. The five types of limitations and the periphyton growth state include:

- 1) If the TN:TP ratio ≥ 7 and TP concentration < 0.046 mg P/L, or if TP concentration equals zero and TN is nonzero, the cell has acceptable periphyton growth and is P-limited (type 1).
- 2) If the TN:TP ratio ≥ 7 and TP concentration ≥ 0.046 mg P/L, the cell has undesirable periphyton growth and is P-limited (type 2).
- 3) If the TN:TP ratio < 7 and TN concentration < 0.800 mg N/L, or if TN concentration equals zero and TP is nonzero, the cell is considered to have acceptable periphyton growth and to be N-limited (type 3).
- 4) If the TN:TP ratio < 7 and TN concentration ≥ 0.800 mg N/L, the cell has undesirable periphyton growth and is N-limited (type 4).
- 5) If TN and TP concentrations are both simulated to be zero, the cell is assigned to have negligible periphyton growth (type 5).

By implementing this spatial information on nutrient limitation, more precise CFs can be derived to assess the impacts of freshwater eutrophication. That is, CFs for P can be applied where the cells are limited by P, and those for N can be used for regions limited by N.

2.4. Aggregation of CFs

To match the freshwater eutrophication CFs with life cycle inventory data, the grid-level CFs can be aggregated to any scale through Eq. (4) and Eq. (5). We provided CFs aggregated to the country level, the common level of inventory data, using two methods: (1) considering only the P/N-limited regions; (2) considering the entire region.

$$CF_{r,e} = \frac{1}{\sum_i E_{e \rightarrow i \in r}} \bullet \sum_i CF_{e \rightarrow i \in r} \bullet E_{e \rightarrow i \in r} \quad (4)$$

$CF_{r,e}$ (PDF·year·kg⁻¹) indicates the aggregation of nonzero CFs (i.e., we excluded CFs that are zero) for direct emissions to freshwater and diffuse emissions excluding erosion over a region r . In method 1, r only includes the P/N-limited regions within the country; in method 2, r indicates the whole country. $E_{e \rightarrow i \in r}$ (kg year⁻¹) is the emission-weighting data of the emission route e in the source cell i that belongs to the region r .

All nonzero $CF_{i \in r, erosion, landuse}^*$ were summed over a region r weighted by the area of the land use type ($A_{i \in r}$, m²) to provide $CF_{r, erosion, landuse}^*$.

$$CF_{r, erosion, landuse}^* = \frac{1}{\sum_i A_{i \in r}} \sum_i CF_{i \in r, erosion, landuse}^* \bullet A_{i \in r, landuse} \quad (5)$$

We also calculated the proportion of emissions (direct emissions and diffuse sources) and area (arable land and pasture) for nutrient-limited regions across all countries. It equals the ratio of emission or area between nutrient-limited regions and the whole country. If it remains unknown where in a country a nutrient is emitted and which nutrient is limiting, one could calculate the average of P and N impacts, weighted by such proportions. For ease of use, we also provide country-level CFs that already incorporate such weights.

2.5. Global impact of eutrophication on freshwater fish species

We calculated the global freshwater fish species loss due to eutrophication from global nutrient emissions and agricultural land use in 2010 to test the application of the developed CFs. The impacts were obtained from the product of inventory data (direct or diffuse emissions, or land occupation area and time) and the respective CFs:

$$I_{emission} = \sum CF_{i, emission} \times E_i \quad (6)$$

$$I_{erosion} = \sum CF_{i, erosion} \times A_i \times t \quad (7)$$

where $I_{emission}$ and $I_{erosion}$ (PDF·year) represent the total impact on freshwater fish species, taking the sum of the impact of each source cell i ; $CF_{i, emission}$ (PDF·year·kgX⁻¹, where X represents P or N) and $CF_{i, erosion}$ (PDF·year/(m²·year)) indicate the CF for P and N emissions and for erosion, respectively, choosing the average CFs for global rather than regional species loss; E_i (kgX) denotes the emissions, for which we considered here the global emissions over a year; A_i (m²) is the land use area occupied, for which we considered here the total agricultural land use area; and t (year) is the occupation time, for which we considered one year. All the P and N emissions to freshwater and land use areas were retrieved from IMAGE-GNM. The impact was aggregated over the world, considering the nutrient limitation.

3. Results

3.1. Regionalized characterization factors

The gridded CFs for global freshwater species loss show similar hotspots for P and N (Fig. 1, Fig. S1 and S2 in Supplementary Material 1). Results showcase only slight differences between using the average or marginal methods to assess effects. For the diffuse emissions and the direct emissions to freshwater, high CFs occur in regions that encompass either large lakes or the headwaters of rivers, especially if they are also densely populated. Most of these regions are in tropical and temperate zones. For erosion, high CFs belong to those areas with intensive agriculture and animal husbandry or with conditions favoring high erosion including steep slopes, vulnerable coastlines, and windy environments. Compared with pasture, more erosion per area occurs in arable land and thus leads to higher CFs. High values for all the emission routes are found in the Andes Mountains (upstream of the Amazon River), the

Sierra Madre do Sul - Sierra Madre Occidental (upstream of San Diego River and Marikina River), Great Salt Lake, Lake Tanganyika, and the Himalayas and Dangla Mountains (Yarlung Zangbo River and Mekong River Basin).

Low CFs for P and N are found in high latitudes, such as the north of Canada, northern Europe, the south of Argentina, the east of Russia, and the northeast of China. These regions are sparsely populated and thus less affected by human activities, or they are located on the coasts where most contaminants disperse from freshwater to offshore environments.

Compared with CFs for regional species loss (Fig. S3 and S4 in Supplementary Material 1), CFs for global species loss show similar spatial patterns in tropical and temperate zones. Nevertheless, for direct emissions, CFs for global species loss seldom have hotspots in polar regions, while CFs for regional species loss have hotspots in North Russia and North Canada. This difference results from considering or not considering GEP (Fig. S13 in Supplementary Material 1). In addition, CFs for global species loss show a larger variation between polar and temperate regions than CFs for regional species loss.

3.2. Nutrient limitation

Excluding the 7.8 % of global arid regions without freshwater discharge, the non-arid regions limited by P and N occupy 52.3 % and 36.5 %, respectively, while 3.4 % are covered by negligible P and N concentrations (Fig. 2). Among these, 23.5 % of the area limited by P and 11.7 % of the area limited by N is susceptible to undesirable rates of periphyton growth. Undesirable eutrophication (type 2 and type 4) ties up with densely populated regions, while acceptable nutrient freshwater concentrations (type 1 and type 3) are distributed in sparsely populated areas such as the polar region and arid zones. The US, China, and countries in North and West Europe, east South America, and Central Africa belong to P-limited regions with undesirable periphyton growth (type 2). These regions are affected by P contained in runoff, domestic and industrial wastewater from populated urban areas and sludge in arable land. Australia, the countries in North Africa and West Asia are more affected by N limitation (type 4), as they represent regions with historically relatively small proportions of arable land.

3.3. Aggregation of characterization factors

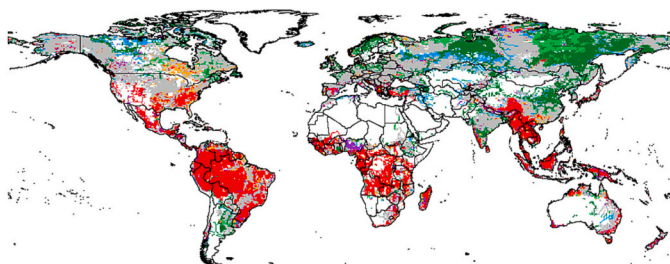
The influence of considering nutrient limitation on country-level CFs varies across countries for all emission routes, while the hotspots (e.g. Cameroon) and low-CF countries (e.g. Libya) (Figs. S5-S12 in Supplementary Material 1, data in "Country_CF_P.xlsx" and "Country_CF_N.xlsx" in Supplementary Material 3) maintain a similar pattern independent of nutrient limitation. Considering nutrient limitation leads to about half-half increase vs. decrease in country-level CFs. Take the average CFs for direct P emissions as an instance; the inclusion of nutrient limitation makes more countries have higher country-level CFs (56 countries higher vs. 52 countries lower than not considering nutrient limitation), whilst the opposite applies to direct N emissions (54 countries higher vs. 55 countries lower).

On a global scale, the inclusion of nutrient limitation leads to lower CFs for diffuse P emissions (by 2 % to 3 %) and higher CFs for other emission routes (by up to 30 %). Yet, considering nutrient limitation makes CFs for N higher (by 25 % to 62 %) (Fig. 3). The average CFs for all emission routes are higher than marginal CFs, independent of nutrient limitation.

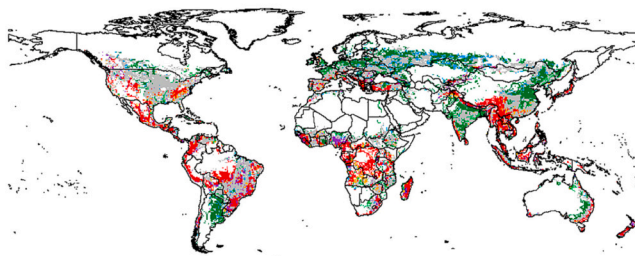
3.4. Global impact of eutrophication on freshwater fish species

Considering nutrient limitation in CFs, we calculated the impact of global P and N emissions as well as erosion enhanced by agricultural land use during the year 2010 on freshwater fish species richness as 0.138 PDF·year (Table 1). Among all the emission routes, erosion on arable land contributes the most to the impacts, while diffuse emissions

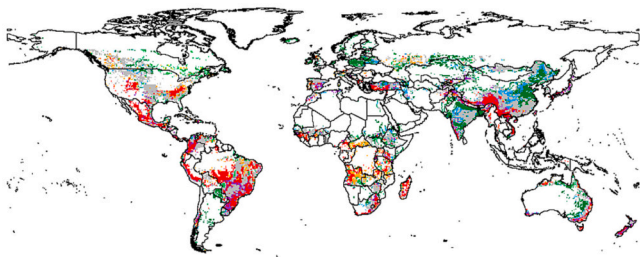
(a) Marginal CF for direct emissions



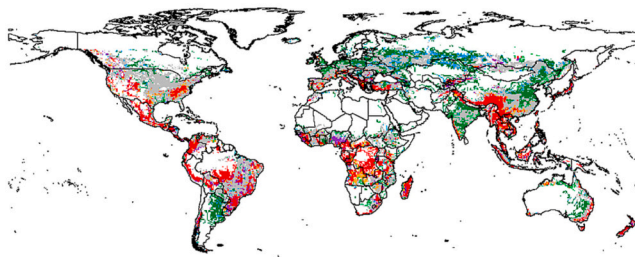
(e) Marginal CF for erosion on arable land



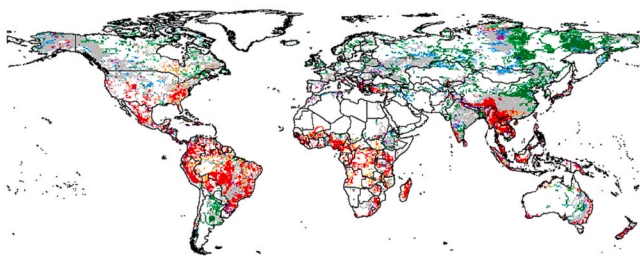
(b) Marginal CF for diffuse sources



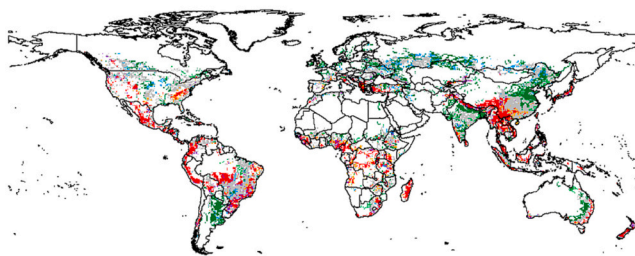
(f) Marginal CF for erosion on pasture



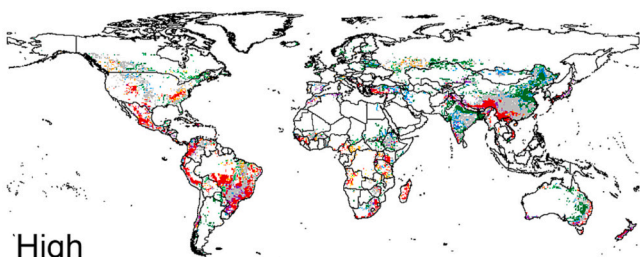
(c) Average CF for direct emissions



(g) Average CF for erosion on arable land



(d) Average CF for diffuse sources



(h) Average CF for erosion on pasture

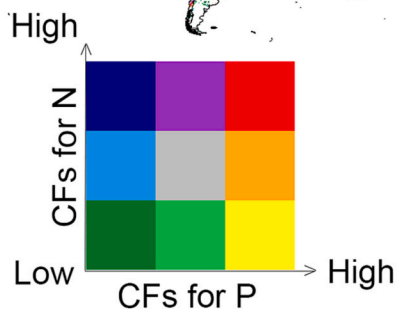
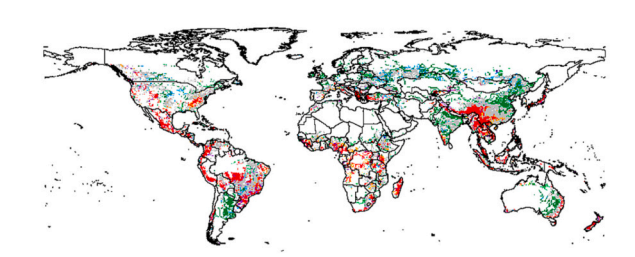


Fig. 1. Comparison of characterization factors (CFs) between P and N at a half-degree resolution. Low CFs and high CFs indicate CFs < first quartile (Q25) and CFs > third quartile (Q75), respectively, and medium CFs are between low and high CFs.

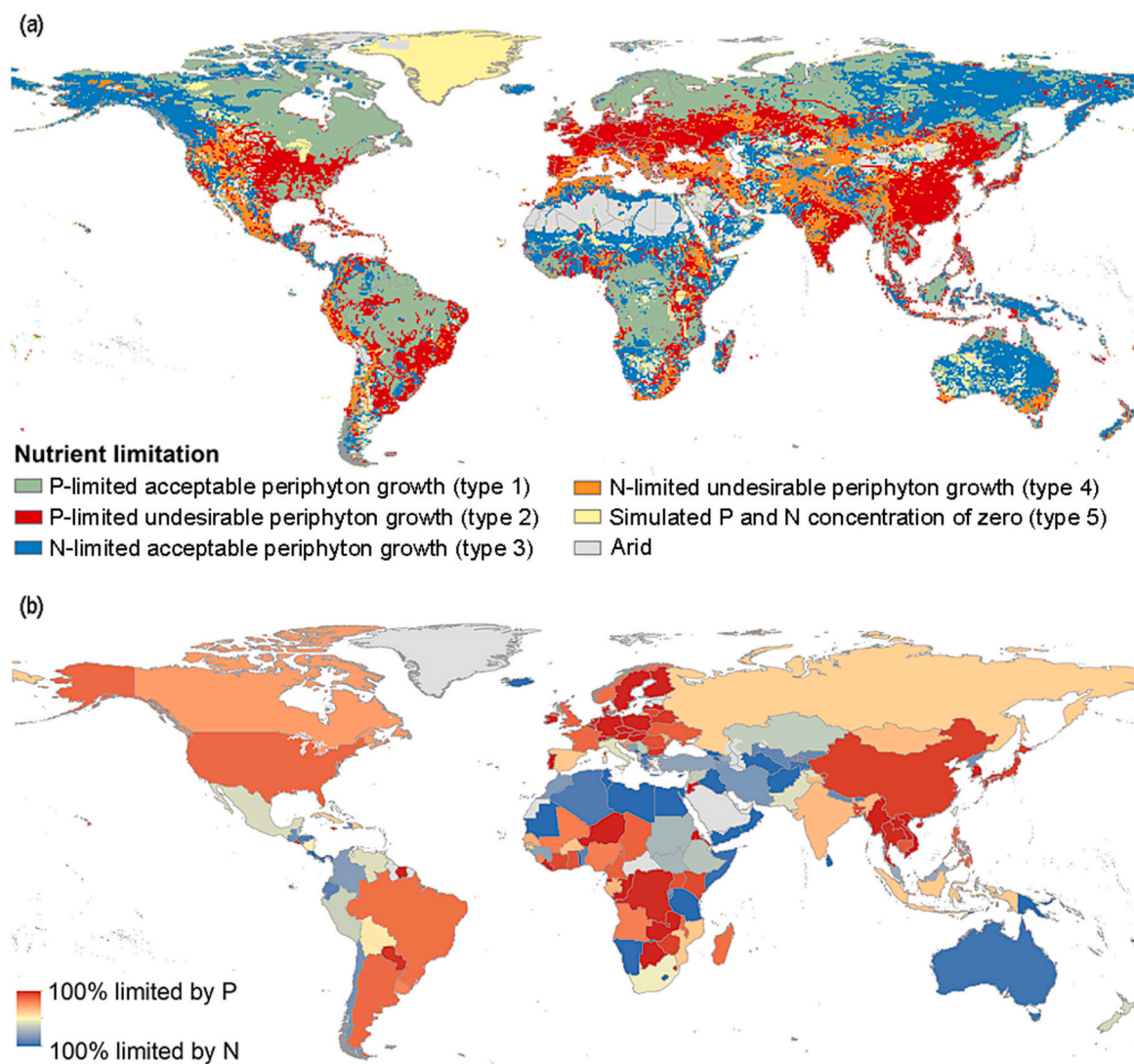


Fig. 2. Nutrient limitation for periphyton growth based on the Redfield ratio: (a) based on P and N concentrations and the type definition as described in Section 2.3 on the gridded scale, (b) based on the proportion of P–N limitation that is weighted by direct emissions in those countries where average CFs for this emission route are available (see Section 2.4). The proportion of P-limited and N-limited regions are provided in “Country_CF_P.xlsx” and “Country_CF_N.xlsx” in Supplementary Material 3.

are the second strongest contributor. Direct emissions rank third and still contribute considerably, while the erosion on pasture has the least influence. Regarding nutrients, P leads to more than double the species loss than N. The difference in impacts is particularly evident for erosion. In summary, P is the paramount nutrient for freshwater eutrophication (causing an impact of 0.098 PDF·year), but the impact of N should not be neglected (0.040 PDF·year).

4. Discussion

In this study, we used two methods for deriving EFs (average and marginal) and compiled the CFs for four emission routes at multiple spatial scales: global, country level, and half-degree grid level. These CFs can be used with emissions and land use areas from life cycle inventories to assess the nutrient-induced impacts on freshwater fish biodiversity. Our improvements include providing CFs that cover eutrophication more comprehensively than just hypoxia (Cosme and Hauschild, 2017) with a much finer resolution for both FFs and EFs than previous studies (Cosme et al., 2018; Payen et al., 2021; Verones et al., 2020). We note that our CFs for N also encompass the potential impact of N-induced toxicity (Kocour Kroupova et al., 2018), which is a different impact

category to eutrophication (Chislock et al., 2013; Dodds and Smith, 2016; Payen et al., 2019; Smith et al., 2006). However, N overloads have been deemed to predominantly affect the aquatic ecosystem through eutrophication (Chislock et al., 2013; Dodds and Smith, 2016; Wang et al., 2021), while direct toxicity contributes little to the influence because it only occurs at a very high concentration of certain forms of N, such as ammonia and nitrite (Jones et al., 2014; Kocour Kroupova et al., 2018; Thurston et al., 1981).

Compared with previous studies that only consider P-related freshwater eutrophication and N-related marine eutrophication in CFs, our study is the first to incorporate both P and N simultaneously and consider which of the two nutrients is limiting where. The information about nutrient limitation can guide users in the choice to assess the impacts of either P or N emissions. The CFs allow LCA practitioners to estimate the nutrient impact on freshwater fish species richness more accurately. This method can also serve as a prototype that may be adapted for eutrophication impact assessment related to marine or terrestrial ecosystems.

Based on the approach outlined above (considering nutrient limitation), the global impact of eutrophication on freshwater fish species is 0.138 PDF·year. The dominance of erosion as a contributor to the

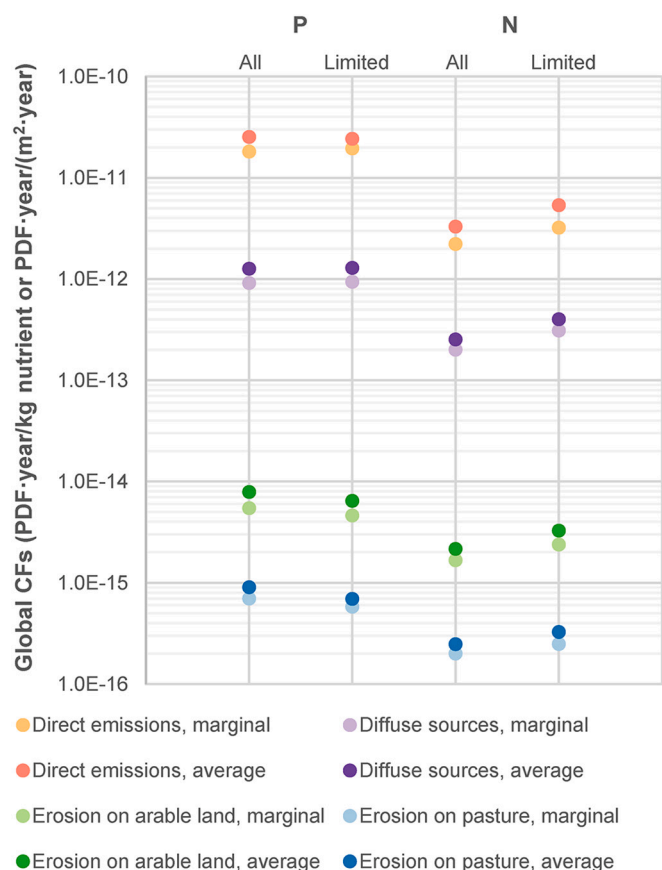


Fig. 3. Globally aggregated characterization factors (CFs) for P and N impacts on global species richness. The unit of CFs for direct and diffuse emissions is PDF-year/kg nutrient, where the nutrient represents P or N, while the unit of CFs for erosion is PDF-year/(m²-year). CFs consider either all regions (All) or only regions limited by the respective nutrient (Limited).

Table 1

Global species loss over the world considering the nutrient limitation. The unit is PDF-year.

	Impact of direct emissions	Impact of diffuse emissions	Impact of erosion on arable land	Impact of erosion on pasture	Sum
P	0.019	0.026	0.047	0.006	0.098
N	0.012	0.013	0.013	0.002	0.040

impacts of freshwater eutrophication is consistent with the findings of Scherer and Pfister (2015). Since the fate within the freshwater is relatively short (in the order of dozens of days) but the emissions and erosion are spread throughout the year, the exposure duration can be assumed to be roughly one year. This means that 13.8 % of the fish species potentially disappear due to freshwater eutrophication. This result approximates the 15.6 % (= 24.8 % × 63 %) of freshwater fish species threatened with extinction due to pollution, as estimated by Miranda et al. (2022). While water pollution is a broad category encompassing various substances, eutrophication stands out as the most widespread source (Boyd, 2019). Therefore, we deem it reasonable that our estimate is somewhat lower than theirs. This agreement shows the validity of using our CFs to reproduce the influence of freshwater eutrophication on the global ecosystem.

The model for nutrient transport and fate is crucial for determining CFs and can be a large source of uncertainty. For instance, 0.5 % of the 0.5 × 0.5 degree grid cells have a diffuse N loading from surface runoff that exceeds diffuse emissions, since IMAGE-GNM does not isolate the

influence of long-term retention of N in the soil surface from the short-term loads in surface water due to new emissions. The isolation of these processes might be possible by using a process-based mechanistic model such as the IMAGE-Dynamic Global Nutrient Model (DGNM) (Vilmin et al., 2020), which models water column and sediment dynamics, and the exchanges between them. A future version on a global scale can form a better basis for developing CFs (Vilmin et al., 2020).

A large amount of data and high spatial resolution allowed for distinguishing species richness-nutrient relationships across 425 ecoregions, which is substantially beyond the previous knowledge of only four geographical zones (Cosme et al., 2017; Cosme and Hauschild, 2017). This resolution could even be improved for the effect factors by considering the background concentration at different locations within the ecoregions. More regionalized environmental indicators can help to better assess impacts at local scales. Our study showcases the advantages of finer spatial resolutions, and we recommend the continuation of this practice when developing new CFs in the future. Next to spatial differentiation, the temporal dynamics in emissions from human activities should be considered in future studies (Potting and Hauschild, 2006; Seppala et al., 2001). Our study can be replicated to generate CFs across various years. Future CF research can delve into both annual and seasonal dynamics with enhanced temporal resolution, providing a more nuanced assessment of fate and effects. Analyzing the impact with consideration of temporal variations in CFs allows for a more accurate identification. Notably, FFs exhibit temporal variations between subsequent years, particularly in densely populated urban regions (Zhou et al., 2022). de Andrade et al. (2021) examined phosphorus FFs in a regional study, revealing minor monthly variations but advocating for consideration of seasonal changes due to differences in water availability. Additionally, EFs vary across years due to varying background concentrations (Zhou et al., 2023). It underscores the dynamic nature of nutrient fate and effect in freshwater systems, emphasizing the need for dynamic models to complement existing steady-state life cycle impact assessment models.

In conclusion, we developed regionalized CFs for freshwater eutrophication at a fine spatial resolution and proposed a method to consider nutrient limitation in CFs. This work provides life cycle impact indicators and a roadmap for considering nutrient limitation to finetune CFs for multiple emission routes to assess the eutrophication impacts on regional and global species richness for LCA practitioners. This roadmap and the consideration of comprehensive nutrient-species effects can be further used for developing regionalized CFs for eutrophication in other ecosystems.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.169108>.

CRedit authorship contribution statement

Jinhui Zhou: Conceptualization, Data curation, Formal analysis, Methodology, Visualization, Writing – original draft. **José M. Mogollón:** Conceptualization, Methodology, Supervision, Writing – review & editing. **Peter M. van Bodegom:** Conceptualization, Methodology, Supervision, Writing – review & editing. **Arthur H.W. Beusen:** Resources, Supervision, Writing – review & editing. **Laura Scherer:** Conceptualization, Formal analysis, Methodology, Supervision, Writing – review & editing.

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.

Data availability

The data that support the findings of this study are partly available in

the supplementary material of Barbarossa et al. (2020, 2021) and Zhou et al. (2022, 2023) and partly in the supplementary material of this article. Supplementary Material 1 shows maps of CFs for regional and global freshwater fish species loss at half-degree resolution, country-level CFs for regional and global species loss not considering and considering nutrient limitation, a comparison of EFs with EFs multiplied by GEPs, and an overview of the coverage of freshwater fish species used in this study. Supplementary Material 2 provides data on the coverage of freshwater fish species used in this study. Supplementary Material 3 is an archive with the gridded and country-level CFs and the gridded nutrient limitation map, as well as a readme file related to the data. Supplementary Material 3 can be accessed at Zenodo under <https://doi.org/10.5281/zenodo.10286168>.

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