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# Development of LCA characterization factors for terrestrial eutrophication at regional scale

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## Abstract

Available models for terrestrial eutrophication in Life Cycle Assessment (LCA) only allow estimation on a country scale, being this a problem, especially in large countries with high geographical, climatic and economical variability. This work aims to estimate characterization factors for terrestrial eutrophication at a regional scale, using Galicia (NW Spain) as a case study. Accumulated exceedance (AE) was used as the category indicator and real values of deposition, emissions and critical loads were required.

The results obtained confirm that AE is a suitable category indicator for estimation of terrestrial eutrophication. Being stable and suitable for LCA applications, the characterization factors obtained for variations in emissions inferior to 100 tons were selected as general characterization factors. Results were compared with available values for Spain and found to be lower, although further analysis is required in order to calculate the exact value of the uncertainties associated with each characterization factor.

**Keywords:** Accumulated exceedance; Life cycle impact assessment; Normalization; Site dependent characterization factors; Terrestrial eutrophication

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## 1. Introduction

Terrestrial eutrophication has been identified as the most important process causing vegetation damage in natural reserves in Europe [1]. It can be defined as the changes in species composition produced in the soil ecosystem due to high inputs of external nutrients [1, 2]. This generally leads to decreases in biodiversity due to competitive substitution [3]. As a result, nutrient imbalances appear and sensitivity of vegetation to disease, drought, frost and herbivores increases [4]. In the specific case of Galicia (NW Spain), terrestrial eutrophication caused by atmospheric deposition of nitrogen compounds has been reported as the main threat to sustainability of terrestrial ecosystems [5]. These authors reported that the threshold for nitrogen deposition ensuring protection of forest ecosystems have been exceeded in 40% of the region. Rodríguez and Macías [5] have also identified the Atlantic heathlands, which are protected natural areas in the Atlantic region, as the most sensitive Galician ecosystems to the effects of eutrophication. In this sense, Cuesta et al. [6] have also detected changes in the macrofauna associated with these heathlands due to external inputs of nitrogen to the system.

LCA has traditionally been considered a site and time independent tool and no consideration has been given to when and where emissions take place [7, 8]. Since the late 1990s, research has shown that location may have a strong influence on categories such as acidification, eutrophication (terrestrial and aquatic), ecotoxicity and smog (e.g.: [9-14]), especially when these categories are significant in the system under study. Spatial differentiation began at a continental level and developed afterwards at a country level. As impact characterization became more mature, the study of specific characterization factors has been reduced to a regional scale (especially in those countries that have large geographic, climatic and economic variability) being a focus of interest in different LCA impact categories [15-17]. So, Reap et al. [18] recommended as a research priority the development of spatially explicit models, capable of accounting for local environmental uniqueness, considering smaller spatial scales and allowing improved resolution around major pollution sources, deposition areas and sensitive zones. The significance of more detailed spatial differentiation seems to increase as the level of definition of the impact category is near the endpoint [9, 11, 13]. For example, Potting et al. [13] included the effect factor when calculating characterization factors, for 44 European regions (most coinciding with countries) and reported differences bigger than a factor of 1 000 fold for acidification and a factor of 500 fold for terrestrial eutrophication.

In the particular case of terrestrial eutrophication, several authors have proposed country-specific characterization factors based on different methodologies with diverse models (EcoSense, RAINS) and source-receptor matrices derived from a Lagrangian model developed by the European Monitoring Evaluation Programme (EMEP ) [9, 11, 13, 19]. By using EcoSense, Finnveden and Nilsson [20] calculated characterization factors for 4 regions in Sweden, but taking into account only the emissions of NOx but not of NHx, which are also important for terrestrial eutrophication. Aerial models allow the calculation of spatial-differentiated characterization factors in a easy and systematic way, but in the case of the RAINS model and source-receptor matrices derived from EMEP model, one of the major drawbacks is the use of information at a country scale, with, at the moment, no possibility to achieve a deeper and more specific spatial definition. This may turn out to be a problem, especially in geographically large countries (such as Spain) where characterization factors are likely to vary. Currently the only way to calculate these characterization factors is by measuring and calculating local emissions, deposition and critical loads.

The main objectives of this study are therefore:

1) To estimate characterization factors for terrestrial eutrophication as well as the normalization values due to NOx and NHx emissions at a regional scale, using Galicia as a case study.

2) To compare the results obtained with those available for Spain, in order to evaluate the influence of the calculation of characterization factors at a lower scale.

## 2. Experimental

### *2.1. The study area*

Galicia has a high diversity of geologic, edaphic and vegetation types [5]. Mean annual temperature ranges from 6ºC to 15ºC and the average precipitation varies from 700 mm/yr (southeastern areas) to 1 200 mm/yr (coastal area). The level of industrialization is low-medium, with some important industrial areas mainly located in the district of the largest cities. Agriculture and livestock rearing are two major activities that occupy 28% of the land area; these are widespread throughout the whole territory, and they constitute the most important source of nitrogen compounds emission to the atmosphere in this region. These specific climatic, biological and anthropogenic characteristics make Galicia quite different to the rest of Spain. For example, emissions are considerably higher than the average values in the rest of the country for NHx (26 g NHx/person in Galicia and 9 g NHx/person in Spain in 2001) and for NOx (46 g NOx/person in Galicia and 32 g NOx/person in Spain in 2001) [21-24]. This region is then an interesting place to test the possible differences in characterization factors, found in regions when compared with the whole country.

It can be argued if Galicia has the adequate extension to have its own characterization factors for terrestrial eutrophication. Establishing a fixed value of extension for the calculation of characterization factors for terrestrial eutrophication is complicated because it depends on the intrinsic characteristics of the area. It would have no sense to calculate separate characterization factors for homogeneous large areas (e.g. Siberia). Potting et al. [25] have suggested as a rule of thumb, based on European data, that the adequate resolution for site dependent factors for impact categories produced by airborne emissions could be about 25 000 km$^2$, which justifies the development of characterization factors for regions like Galicia (29 574 km$^2$). The development of characterization factors at lower levels would make an holistic methodology like LCA impracticable due to the amount of necessary data and for such specific analysis (e.g.: a river) another tools like Environmental Risk Assessment (ERA) should be applied.

The relationship between aerial emissions and deposition needs consideration in order to calculate the characterization factors for compounds causing terrestrial eutrophication in a region. In theory, emissions from Galicia can travel and affect other ecosystems outside the region, and, in the same way, emissions from other areas can deposit in Galicia and affect its ecosystems. However in this study, as in Gallego et al. [26], Galicia has been considered as a “closed box”, where the emissions produced are deposited in its own territory, and, therefore, the influence of the emissions from the neighboring regions has been disregarded. The reasons behind this assumption are:

1) NOx and NHx emissions produced in Galicia are mainly deposited in Galicia.

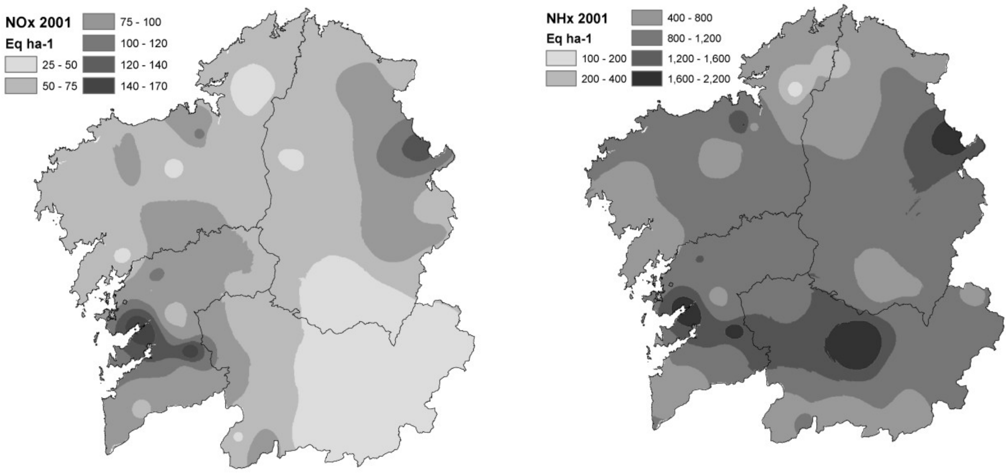

**Figure 1.** Deposition levels of NOx (A), and NHx (B) (adapted from [58]).

Gallego et al. [21, 22] established that mobile and industrial sources were responsible for 90% of NOx emissions in Galicia. The A9 highway (NW-SW) is the main high capacity road in Galicia and the main axis for intra-regional and interurban traffic, with more than 65,000 vehicles per day and almost 65% of the Galician population concentrated around it. Concerning industrial NOx emissions, the two provinces crossed by the A-9 accounted for 91.3% of the total NOx industrial emissions in 2001 [27]. These conditions lead to the high deposition levels observed in Western Galicia (Figure 1A). The high deposition located in the NE of the region (a sparsely populated area with little industrial presence) is surprising. The presence of high mountains, which can cause lower mobility of air and cause NOx to stagnate, and the proximity of the A6 highway, can be considered as reasonable explanations for this high deposition. In any case, there is no evidence that the majority of the NOx deposited in this area are from outside Galicia.

A large proportion of the NHx emitted locally is deposited in the immediate neighborhood of the source rather than being transported large distances [28-31]. This low mobility is also confirmed in Galicia: Gallego et al. [21, 22] identified dairy cows and broilers as the main sources of NHx in the region (38.8% and 41.1% of the total, respectively). Their production is concentrated in the North-East and South part of the region, correspondingly [32], which are the locations where more NHx is deposited (Figure 1B). The high depositions detected in the NE Galicia are also in agreement with the hypothesis of lower mobility of air due to the presence of high mountains explained for NOx.

2) Inputs of NOx and NHx from neighboring regions to Galicia can be disregarded.

This statement is based on the analysis of the levels of deposition and emission from NOx and NHx in Galicia and in the neighboring regions (<200 km of distance from the Galician frontier) reported by the Cooperative Program for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe [33]. It should be noted that the EMEP grids do not correspond with the boundaries of the Spanish regions, and therefore, the emissions and deposition have been proportionally distributed in the case of “frontier” grids. Additionally, the values calculated by EMEP are based on modeling of the data reported by only 16 monitoring stations in Spain and 5 in Portugal. Therefore, this data must be analyzed with caution when applied at a local scale. With these limitations in mind, Table 1 shows that the level of emissions and deposition in Galicia and neighboring regions is quite similar (the differences are less than 10%), which can be justified in two different ways:

- The net flux of emissions (entrance + outflow) is close to 0. That is, Galicia exports NOx and NHx to its surroundings at a similar rate than imports emissions from neighboring regions. Having this in mind, a variation of the emissions in Galicia (and therefore, a variation of the depositions in the neighboring regions) will produce in those regions a similar impact in terrestrial eutrophication as in Galicia, because the neighboring regions present very similar biogeographical conditions (type and distribution of vegetations, land uses, climate…) [34]. This means that, even if produced in other regions, the impact caused by Galician emissions will be

**Table 1.** Emission and deposition levels in Galicia and in the neighboring regions (from reported data by [33]).

| | Emissions | | | Depositions | | |
|---|---|---|---|---|---|---|
| | Galicia | Neighboring regions[a] | Variation[b] (%) | Galicia | Neighboring regions[a] | Variation[b] (%) |
| $NO_x$ (t/km$^2$) | 2.49 | 2.70 | 8.43 | 2.06 | 2.23 | 8.25 |
| $NH_x$ (t/km$^2$) | 0.49 | 0.48 | -2.04 | 0.41 | 0.40 | -2.44 |

[a] 200 km from the Galician border; [b] Variation in comparison with Galician values

similar and our characterization factors would still be valid.

• The dispersion pattern is similar both in the neighboring regions and in Galicia (the emissions are deposited near the production site) and thus, the external flux of NOx and NHx to Galicia can be ignored.

### *2.2. Methodology*

The critical load concept is a key element for determining the sensitivity of forest ecosystems to the deposition of atmospheric pollutants. For eutrophication, the critical load concept of nutrient nitrogen has been defined as "the highest deposition of nitrogen compounds below which harmful effects in ecosystems structure and function do not occur according to the present knowledge" [35]. Therefore, the critical load could be considered as the threshold value and, when exceeded, according to the present knowledge, harmful effects could occur to the ecosystem. Within LCA methodologies, three approaches are available to handle impact categories subjected to threshold values [36]:

• “Less is better”: Emissions are always considered harmful, independently of the previous situation of the ecosystem and therefore, ignoring the concept of critical load. One of the major drawbacks of this approach is that the impact produced by an increase in depositions for ecosystems exposed to levels around the critical load are considered equally important as ecosystems facing the same increase but having already been exposed at levels far above (and thus difficult to rescue) or far below (and thus hardly in danger) the critical load.

•“Only around threshold”: Characterization factors are based on how much the extension of ecosystems will exceed their critical load and become unprotected areas as a result of emissions; therefore, the category indicator will be the quantity of unprotected areas (UA) caused per amount of substance emitted (m$^2$/kg of emission). As a result, this approach gives priority to ecosystems exposed to levels around their critical load rather than ecosystems exposed to levels far below or far above. For terrestrial eutrophication and acidification, this option has been criticized for having unstable characterization factors and being strongly dependent on the variation percentage of the emissions, especially when they are small (as generally occurs in LCAs studies) [19, 37, 38]. In addition, this approach does not reflect the environmental benefits of the reduction of emissions when depositions are substantially higher than the critical load [19].

•“Only above threshold”: This method only considers those emissions that result in exposure increases at sites where critical loads are already exceeded, with the exposure increase level for the remaining scenarios valued at zero. This approach is more realistic than the first approach (“Less is better”), considered too protectionist, allowing to obtain stable factors with small reductions in the emissions, which is the principal drawback of the second option (“only around threshold”). One of the main criticisms here is that we can ignore an ecosystem exposed far below its critical load (and thus hardly in danger) and also ecosystems just below their critical load (and thus in danger of exceeding their critical load due to a small exposure increase). Another drawback is that this approach may characterize ecosystems as being equally vulnerable whether they are exposed far above or just above their critical loads. The methodology applied in this study (developed by [19]) is based on this third approach and the most important advantage of the indicator used in this study (accumulated exceedance, AE) is that the amount of depositions above the critical load can be taken into account thus avoiding this last disadvantage. AE methodology was provisionally selected by the European Platform on Life Cycle Assessment as the recommended model for midpoint evaluation of terrestrial eutrophication (while no recommendation was made at the endpoint level) [39].

Exceedance is defined as the amount of nitrogen deposition above the critical load. AE can be calculated with the following equation [19]:

$$AE = \sum_{i=1}^{n} A_i \cdot Ex_i \qquad (1)$$

where $A_i$ is the area of ecosystem i [ha], $Ex_i$ is the exceedance in ecosystem i [eq/ha/yr] and n is the number of ecosystems within the area of interest.

To calculate AE for a region j, the joint deposition of nitrogen compounds (NOx as $NO_2$ and NHx as $NH_3$) must be calculated and combined with the critical loads of the existing ecosystems. This joint deposition describes exactly what occurs in nature and from here forward will be called the “exact model”. However, Life Cycle Impact Assessment (LCIA) requires independent characterization factors (one for NOx and one for NHx) and so the exact model needs to be replaced by an “additive model” [19], which allows the calculation of regional characterization factors for terrestrial eutrophication based on AE (CAEp) for each pollutant P (= NOx, NHx) as:

$$CAE_P = \Delta AE^{X\_P}/\Delta E_{X\_P} = (AE - AE^{X\_P})/(EP - E_{X\_P}) \quad (2)$$

where AE is the total Accumulated Exceedance per year [keq/yr], $AE^{X-P}$ is the total AE after an X% annual variation (reduction or increase) of the emissions of pollutant P, EP is the annual emissions of pollutant P (t/yr) and $E^{X-P}$ is the emissions of P after the variation X.

The validity of the characterization factors must be tested with the help of the so-called characterization error [19], which describes the difference between the results calculated by the exact model and the results of the additive model. An error equal to zero indicates that pollutants NOx and NHx are independent of each other in the characterization equation, and therefore, the assumption of the non-interdependence between NOx and NHx outcomes is correct. In addition, the characterization error evaluates how the characterization model can describe situations of joint reduction or increase of emissions. This feature is required in LCA applications when at least two eutrophying compounds are considered. The absolute characterization error ($Er_{rab}$) is obtained using the following equation:

$$Err_{ab} = AE - AE^{X\_NOx,\ X\_NHx} - CAE_{NOx} \cdot \Delta E_{NOx} - CAE_{NHx} \cdot \Delta E_{NHx} \quad (3)$$

where $AE^{X\_NOx,X\_NHx}$ is the total AE in the region of interest after X emission variation of NOx and NHx in the reference year. The term AE – $AE^{X\_NOx,\ X\_NHx}$ represents the results of the exact model while the following terms stand for those from the additive model.

Combining equations 2 and 3:

$$Err_{ab} = AE^{X\_NOx} + AE^{X\_NHx} - AE^{X\_NOx,X\_NHx} - AE \quad (4)$$

Equation 4 gives absolute errors. Percentage or relative error (Errre) can be calculated by dividing the absolute error by the result of the exact model and multiplying the result by 100:

$$Err_{re} = (Err_{ab} \cdot 100)/(AE - A^{EX\_NOx,X\_NHx}) \quad (5)$$

### *2.3. Data collection*

Seppälä et al. [19] have estimated the deposition of NOx and NHx using source-receptor matrices derived from a Lagrangian model of long-range transport of air pollutants within a grid of 150 $km^2$ cells developed by EMEP. One of the major drawbacks of this approach is that only country-to-grid transfer matrices are available and therefore, only country-dependent characterization factors can be calculated with no possible further differentiation. The mentioned authors have established that it would be useful to calculate specific characterization factors for different regions inside large countries in Europe. To do so while avoiding the mentioned limitations, the deposition levels for NOx and NHx presented in this work were directly measured in the field (see section 2.3.1) and those values were later combined with critical loads of eutrophication calculated specifically for Galicia (section 2.3.2). Total emissions of NHx and NOx, which are also critical for calculation purposes (see equation 2), were estimated for 70 102 and 127 259 tons, respectively, by using the best available calculation methodologies and considering the main emission sources [21, 22]. All values are referenced to 2001, which is the most recent year for which all required data was available.

#### *2.3.1. Nitrogen deposition levels: Geostatistical analyses*

The deposition levels of nitrogen compounds were calculated using rainfall compositional data recorded by 23 monitoring sites from the Galician monitoring network of atmospheric pollutants and from the monitoring networks of the power stations of As Pontes and Meirama [5]. We used the monthly average concentrations of NOx and NHx (eq/L) in rainfall separately to create 12 maps by ordinary kriging over the whole territory. These maps were multiplied by the accumulated amount of precipitation ($L/m^2$) to account for the monthly deposition of these compounds in $eq/m^2$. Then, the monthly values were summed to obtain the annual deposition (Figure 1A for NOx and Figure 1B for NHx), as well as the total N deposition (Figure 2) as the sum of the single deposition values of NOx and NHx.

Because the monitoring stations only register wet deposition, dry deposition cannot be

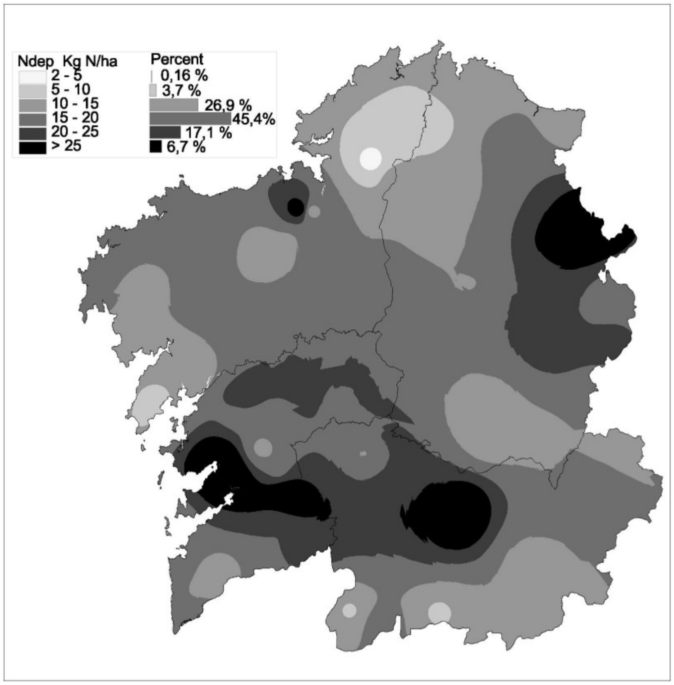

**Figure 2.** Depositions total N in terrestrial ecosystems in Galicia in 2001 (adapted from [5, 58]).

quantified. However, part of the emissions will deposit as dry deposition in terrestrial ecosystems and therefore an underestimation of characterization factors is likely to happen. In this sense it should be noted that precipitation levels in Galicia are very high (especially in the reference year 2001 where the precipitation was 34% higher than the average from the 2002-2007 period [40]) and well distributed throughout the year, and thus, the main deposition is wet deposition [5, 41]. Furthermore, estimation of dry deposition is a very complex process. It varies greatly depending on the complexity of the territory, the atmospheric conditions (wind circulation) and the type of ecosystems. There is often a high degree of uncertainty associated with dry deposition estimates [42]. This uncertainty is even higher for areas such as Galicia that have a rough landscape and are very windy. A very complex network would therefore be required for accurate estimation of dry deposition in the study area. As a first approximation, dry deposition of NHx and NOx in 2001 can be estimated in 9% and 14% of the total, respectively [33]. This data must be taken with care because it is a rough approximation based only in one monitoring station instead of a complex network. Considering the mentioned facts and following the recommendation of previous studies in Galicia [5, 41], dry deposition has not been considered.

### *2.3.2. Terrestrial ecosystem sensitivity to eutrophication*

The critical loads of eutrophication (Figure 3) were calculated using the Simple Mass Balance method [43]:

$$CL\ (N_{nut}) = N_i + N_u + N_{de} + N_{le(crit)} \qquad (6)$$

where CL($N_{nut}$) is the critical load of eutrophying N deposition and $N_i$, $N_u$, $N_{de}$ and $N_{le(crit)}$ are the amounts of N removed from the system by immobilization, uptake, denitrification and leaching processes, respectively. Specific critical loads have been calculated for all Galician forests, which occupy almost two thirds of the area in the region [32]. Detailed information on this method and its application to Galicia can be found in [5].

### *2.3.3. Exceedances of nitrogen deposition*

Exceedances of nitrogen deposition (Figure 4) were calculated as the difference between total N deposition values in 2001 and the critical loads of eutrophication, and expressed as equivalent units (eq), equating 1 eq to 1 mol of nitrogen. Although 40% of forest soils suffer from a risk of eutrophication in Galicia, these exceedances were generally moderate, less than 5 kg/ha (27.2%), and only 3.3% of the forests showed exceedances higher than 10 kg/ha.

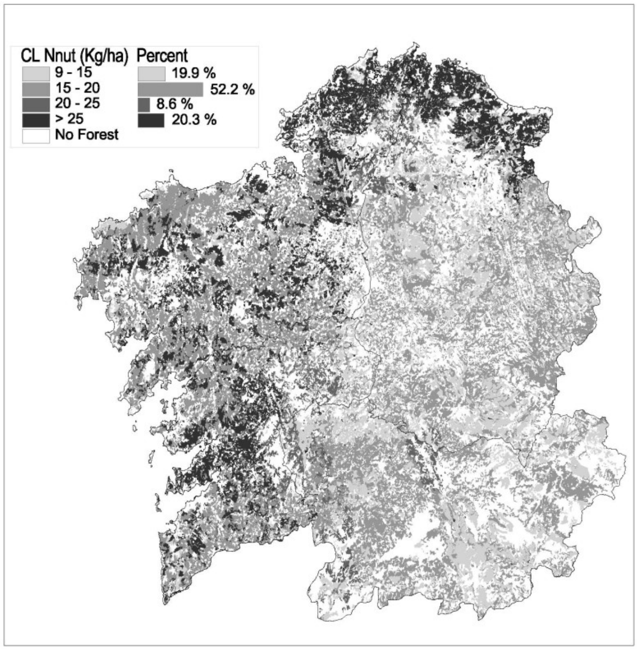

**Figure 3.** Critical loads of eutrophication in Galicia in 2001 (adapted from [5]).

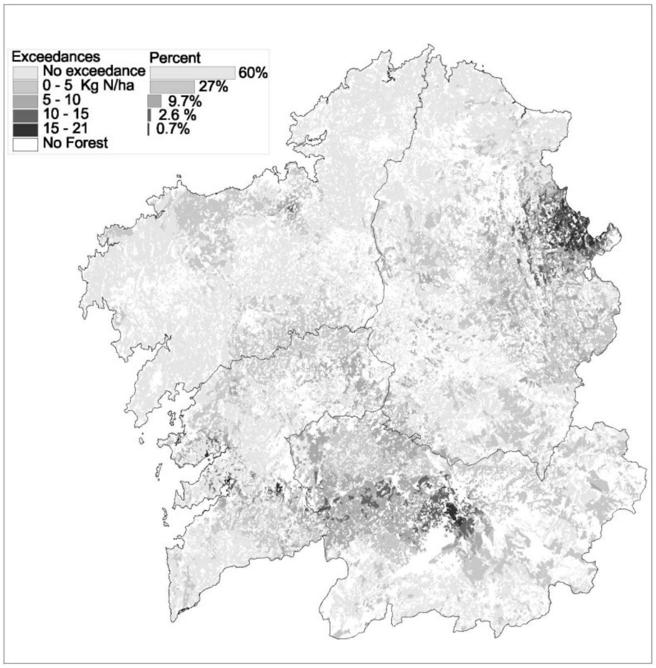

**Figure 4.** Exceedance of N in Galicia in 2001 (adapted from [5]).

## 3. Results

### *3.1. Characterization factors and normalization value*

The sum of all the exceedances of N (Figure 4) allowed us to calculate the total AE for Galicia in 2001 (20 124 keq/yr). The next step, while maintaining the same critical load values (Figure 3), was to calculate the variation in AE (in equation 2, $\text{AE-AE}^{\text{X\_ NOx}}$, $\text{AE-AE}^{\text{X\_ NHx}}$, respectively) with progressive variation (expressed as X) in NOx and NHx emissions ($\Delta E_{\text{X\_NOx}}$, $\Delta E_{\text{X\_NHx}}$, respectively). For these calculations, it was assumed that a percentage of reduction or an increase in the emissions meant the same percentage of reduction or increase in the depositions. These results allow the calculation, following equation 2, of the characterization factors for NOx and NHx (Table 2).

**Table 2.** Characterization factors and errors for $NO_x$ and $NH_x$ in Galicia with variation in the emissions.

| $NH_x$ (as $NH_3$) | | | | $NO_x$ (as $NO_2$) | | | | Characterization error | |
|---|---|---|---|---|---|---|---|---|---|
| Variation emission (%) | $\Delta E_{X\_NHx}$ (t) | $AE\text{-}AE^{X\_}_{NHx}$ (keq/yr) | $CAE_{NHx}$ (keq/t·yr) | Variation emission (%) | $\Delta E_{X\_NOx}$ (t) | $AE\text{-}AE^{X\_}_{NOx}$ (keq/yr) | $CAE_{NOx}$ (keq/t·yr) | Absolute (keq/yr) [a] | Relative (%)[a] |
| 0.014 | 1 | 1.96 | 1.96 | 0.008 | 1 | 0.074 | 0.074 | -0.01 | -0.05 |
| 0.14 | 100 | 196 | 1.96 | 0.08 | 100 | 7.4 | 0.074 | -0.19 | -0.09 |
| 0.25 | 175 | 342 | 1.95 | 0.25 | 318 | 23 | 0.074 | -0.35 | -0.10 |
| 0.50 | 351 | 680 | 1.94 | 0.50 | 636 | 47 | 0.074 | -1.20 | -0.17 |
| 0.75 | 526 | 1 015 | 1.93 | 0.75 | 954 | 70 | 0.074 | -2.67 | -0.25 |
| 1 | 701 | 1 345 | 1.92 | 1 | 1 273 | 94 | 0.074 | -4.68 | -0.33 |
| 2 | 1 402 | 2 624 | 1.87 | 2 | 2 545 | 187 | 0.074 | -19.16 | -0.69 |
| 3 | 2 103 | 3 838 | 1.82 | 3 | 3 818 | 280 | 0.073 | -42.59 | -1.05 |
| 4 | 2 804 | 4 990 | 1.78 | 4 | 5 090 | 372 | 0.073 | -73.20 | -1.38 |
| 5 | 3 505 | 6 085 | 1.74 | 5 | 6 363 | 465 | 0.073 | -11.39 | -1.75 |
| 6 | 4 206 | 7 122 | 1.69 | 6 | 7 636 | 556 | 0.073 | -160.77 | -2.14 |
| 7 | 4 907 | 8 100 | 1.65 | 7 | 8 908 | 648 | 0.073 | -216.83 | -2.54 |
| 8 | 5 608 | 9 020 | 1.61 | 8 | 10 181 | 739 | 0.073 | -273.50 | -2.88 |
| 9 | 6 309 | 9,892 | 1.57 | 9 | 11 453 | 830 | 0.072 | -335.72 | -3.23 |
| 10 | 7 010 | 10 713 | 1.53 | 10 | 12 726 | 920 | 0.072 | -401.15 | -3.57 |
| 20 | 14 020 | 16 317 | 1.17 | 20 | 25 452 | 1 366 | 0.071 | -1 288 | -7.63 |
| 30 | 21 031 | 19 146 | 0.91 | 30 | 38 178 | 2 229 | 0.069 | -2 321 | -11.92 |
| 40 | 28 041 | 20 115 | 0.72 | 40 | 50 904 | 3 054 | 0.068 | -3 447 | -17.13 |
| 50 | 35 051 | 20 124 | 0.57 | 50 | 63 630 | 4 227 | 0.066 | -4 227 | -21.00 |

[a] Negative (positive) values mean that the additive model produces larger (smaller) category indicator results compared to the exact model.

According to [44], one of the advantages of the AE is that it changes smoothly when deposition varies, and this can be observed in Table 2, where characterization factors are very similar when they are calculated on the basis of emission reduction below 2% for NHx, (expressed as $NH_3$) and below 20% for NOx (expressed as $NO_2$) (Table 2). At these percentages of reduction the characterization factors are quite stable, whereas, in the case of other category indicators used in LCA such as unprotected areas [16], characterization factors are strongly dependent on the reduction percentage, especially when they are small [19, 37, 38].

Normalization is an optional step in LCIA which consists of calculating the magnitude of the category indicator results relative to reference information [45]. This reference value for Galicia in 2001 is the total amount of accumulated exceedance (20 124 keq/yr).

### *3.2. Uncertainty*

The procedure described entails several assumptions/choices that will affect the results of the characterization factors and their associated uncertainties. The huge amount of variables handled and the ignorance of their confidence intervals make very difficult the calculation of quantitative uncertainty values. Instead, a qualitative analysis is presented, highlighting those parameters or choices that we consider can produce more uncertainty and possible solutions to reduce it.

To diminish this uncertainty associated to critical loads it would be necessary to:

•Increase our knowledge of the spatial distribution estimates of biomass productivity of forest stands [5, 41] and their uptake rates.

•Obtain, with empirical methods, specific values of N denitrification ($N_{de}$) and N critical leaching ($N_{le(crit)}$) for Galicia. The used data was obtained from published reviews relating to different climatic zones in Europe [46, 47] and this fact can make these values slightly different to those corresponding to Galician forest soils.

In order to decrease the uncertainty in the deposition rate it is necessary to increase the monitoring stations. These new stations must be well located, trying to measure the deposition in areas that are currently not well covered. Old and new stations must be equipped to measure dry depositions because this data will improve the reliability of the characterization factors.

For the total emissions of NOx and NHx, as established by [21, 22], the NOx emissions produced by agricultural, fishery and forest vehicles and machinery and by maritime and railway traffic are the main focus of uncertainty due to the lack of specific data for Galicia and, for NHx, agricultural emissions (especially from cattle, broilers and urea) because of the absence of more specific emission factors.

To assume Galicia to be a “closed box” can be considered also as an important source of uncertainty. The justification of this hypothesis was already described (section 2.1), but it must be proved empirically. In order to apply this methodology to other regions, each case must be analyzed individually to determine which amount of the emissions are deposited in that same region and which are deposited in other regions.

## 4. Discussion

The results corresponding to a variation in emissions of less than 100 tons were chosen as the final characterization factors: 0.074 keq/t•yr for NOx and 1.96 keq/t•yr for NHx. Although the decision seems quite arbitrary, this value is based on the fact that below this level, which is suitable for LCA applications, the characterization factors are maintained stable. When dealing with LCA studies, the main matter usually implies marginal changes in emissions compared with the total regional emissions, as the assessment concerns the emissions of functional units of products (expressed typically as kg of emission per functional unit, FU) [19, 48]: e.g., an LCA of a small wastewater plant where FU = 1 p.e. /yr implies the emission of 0.2 kg of NHx/FU [49]. But, following the same argument proposed by [19] the characterization factor for terrestrial eutrophication should not only describe the impact produced by the FU, but should also describe the impact from the whole production. Emissions per functional unit used in LCA are derived from the emissions in a period of time (normally one year or one batch of production) that really represents the effects of the whole production, and our characterization factors must describe the effects caused by the associated total emission range. Going back to the example and assuming a treatment capacity of 10 000 p.e., the whole emission of NHx will be 2 t and therefore, our characterization factors must describe the impact of the whole production (2 t) and not only that amount produced by the functional unit (0.2 kg NHx/p.e•yr). The level of emissions described in the example is normal in LCA applications in Galicia. Therefore, the characterization factors should usually describe eutrophying emissions representing some hundreds of kilograms or a few tons. Actually, to the best of our knowledge, none of the LCA of products developed in

**Table 3.** Characterization factors and errors for $NO_x$ and $NH_x$ for Spain [19] and Galicia (1 and 100 tons emission variation).

| | Characterization Factors (keq/t·yr) | | Characterization Errors (1 ton) | | Characterization Errors (100 tons) | |
|---|---|---|---|---|---|---|
| | $NO_x$ (as $NO_2$) | $NH_x$ (as $NH_3$) | Absolute (keq/yr)[a] | Relative (%)[a] | Absolute (keq/yr)[a] | Relative (%)[a] |
| Spain | 0.877 | 3.431 | 0.03 | 0.00 | -0.03[a] | 0.00[a] |
| Galicia | 0.074 | 1.96 | -0.05 | -0.01 | -0.19[a] | -0.09[a] |

[a] Negative (positive) values mean that the additive model produces larger (smaller) category indicator results compared to the exact model.

Galicia so far (considering the effects of the whole production amount) reaches values even near 100 tons of emissions of NOx or NHx (e.g.[49–52]). Therefore, characterization factors considering variations in emissions lower than 100 tons are suitable for LCA applications. In rare cases, when annual emissions are in kilotons, the characterization factors change negligibly. For example, as mentioned above, with variations of 2% in the emissions of NHx (1 402 tons) and 20% for NOx (25 452 tons) the characterization factors are still quite constant (differences lower than 5%). For extreme cases in Galicia, where the variation in emissions is even larger, LCA practitioners can derive characterization factors using extrapolation of the results from Table 2.

Comparison of our results with values from the literature are not easy because, as far as we know, only [19] have obtained characterization factors for terrestrial eutrophication using AE as a category indicator. Table 3 compares both sets of data where the following aspects need to be taken into account:

•Due to the absence of specific characterization factors for Galicia in [19], values for the whole of Spain were used.

•The calculation of the category indicator (accumulated exceedance) has been done in a different way. We used real measured data for wet depositions while [19] obtained their values (including also dry depositions) from source-receptor matrices derived from a Lagrangian model of long-range transport of air pollutants. Moreover, we calculated the specific critical loads for Galicia (section 2.2.) while [19] used the data compiled in the work under the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CRLTAP) [53]. Emissions, on the other hand, were calculated in both cases following the EMEP/CORINAIR Emission Inventory Guidebook [54].

•A factor of scale must also be considered as our study is focused on a small region (Galicia) while [19] used data obtained for all of Europe as their area of interest.

•The factor of time is probably less important due to different years being used in both studies: 2001 here and 2002 in [19].

•Even if the best available data is used, the level of uncertainty is not calculated in any study.

As observed in Table 3, NOx and NHx characterization factors for Galicia are lower than the ones calculated for the entire country (Spain). The differences between characterization factors (twelve-fold for NOx and two-fold for NHx) are in the range observed for European countries in other studies [13, 19]. The differences between NOx and NHx Galician characterization factors can be explained on the basis that terrestrial eutrophication in Galicia is mainly produced by NHx emissions instead of NOx [5]. The environmental significance of these results is that using the Spanish factors in Galicia would result in an overestimation of environmental damages or benefits, depending on whether emissions are increasing or decreasing. This conclusion must be taken cautiously because these results only show a tendency and for the exact quantification of this overestimation a detailed comparison of both studies is required in order to calculate the exact value of the uncertainties associated with each characterization factor. However, this is not possible with the currently available data. Characterization errors are insignificant in both studies, indicating that in both cases, with less than 100 tons of emissions reduction, the results of the additive model are similar to the ones obtained with the exact model. It must be clarified that these errors don't measure the accuracy of the data (emissions, critical load…). Therefore, at this level of variation of emissions, the emissions of NOx and NHx can be considered practically independent and the additive characterization model describes well situations of joint reduction or increase of those emissions.

The importance of spatial differentiation in LCA and its influence in real case studies have already been reported by other authors (e.g. [55-57]). The practical applicability in LCA of the results here presented is limited as they represent only one region and are based on several important assumptions/choices. So, these characterization factors can be only applied to LCA analysis of products with production processes mainly located in Galicia (e.g. [49-52]); and even there, its use will not necessary lead to different conclusions. In fact, site-dependent characterization factors are probably not necessary for each region with the extension of Galicia as happens in the particular case described by [20]. There, the small difference between regions in Sweden and the absence of a characterization factor for NHx could be the reason why the alternatives proposed for the Swedish two regions studied obtain the same ranking using site-independent or site-dependent characterization factors. However, Finnveden and Nilsson [20] considered that the use of spatial differentiated characterization factors was "still policy relevant, since it is often suggested in the public discussions that different policies are needed in different regions". In this sense, the Galician Government has authority in environmental issues and therefore specific data for this region is of interest for the development of its specific environmental policies.

Having said this, the main objective of this study was not the direct application of the characterization factors, but the development of regional characterization factors. In this sense, and always taking into account the specific conditions of Galicia and its weaknesses (section 3.2 Uncertainties), this study can be used as a base for the future development of characterization factors in other regions. As far as more similar studies are available, the practicability of this first attempt will increase. Related with this, one of the main drawbacks for the future development of these specific characterization factors is the data required. This gap will hopefully be filled, at least at a European level, in the near future with the development of European Directives such as the EU Water Framework Directive.

## 5. Conclusions

According to the state-of-the-art characterization factors for terrestrial eutrophication, the present development of the models used for their calculation only allows differentiation at a country-scale basis. In this sense, this paper presents the calculation of terrestrial eutrophication characterization factors for NOx and NHx at a regional level, using Galicia (NW Spain) as a case study. The procedure uses measured and estimated data of depositions and emissions of pollutants and critical loads of the natural ecosystems. Even if the best available data is used, the results must be considered as a first approximation, because several assumptions/choices, like assuming Galicia as a "closed box", have been made. Having this caution in mind, the Galician characterization values obtained are different that those obtained by Seppälä et al. [19] for a higher scale (Spain), but a more detailed comparison of both studies is required in order to calculate the exact value of the uncertainties associated with each characterization factor.

Seppälä et al. [19] have established that accumulated exceedance was a good category indicator for terrestrial eutrophication in LCA as it produced stable characterization factors with low variation for 39 countries, which is typical for LCIA applications. We confirm this finding with our work because with variation in emissions below 100 tons, the characterization factors for Galicia are also constant and characterization errors negligible.

## 6. Recommendations and Perspectives

The high amount of uncertainty reported in this study is a major concern in our calculations and therefore future studies should focus on the reduction of this uncertainty. In the case of emissions, the use of scientifically contrasted and widely used guidebooks (e.g., [54]) and continuous attempts to increase the quality of the emissions inputs in the context of integrated assessment work, encourages some optimism for obtaining realistic values. Regarding critical loads, the establishment of more specific values for different ecosystems and the specific identification of the limits of these ecosystems will allow the improvement of the characterization factors. In the case of depositions, more monitoring stations would allow more specific results. By improving the accuracy of all these variables, updated and more reliable regional characterization factors should be established.

A thorough comparison of the AE and hazard index methodologies for terrestrial eutrophication would be interesting as both apply the "only above threshold" approach [9, 19]. This comparison should be done using the same input data and integrated assessment model, while critically discussing the advantages and disadvantages of each methodology.

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