



A life cycle assessment approach for nitrogen footprint quantification: the reactive nitrogen indicator

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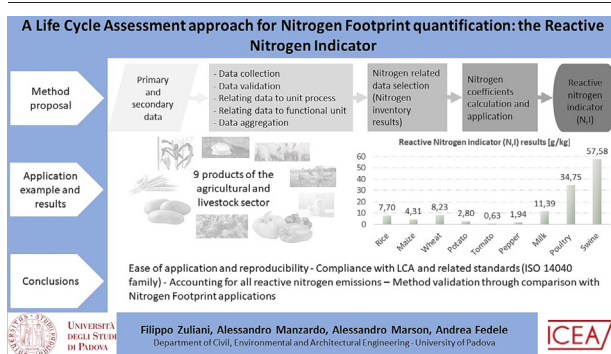
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HIGHLIGHTS

- Method for reactive nitrogen emissions and nitrogen footprint accounting
- Calculate the nitrogen footprint with a LCA approach
- Nitrogen footprint of products from the agricultural and livestock sectors
- Reactive nitrogen emissions and nitrogen footprint from inventory analysis

GRAPHICAL ABSTRACT



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ABSTRACT

Among the environmental issues that have recently catalyzed the attention of the scientific world, we must undoubtedly include the perturbation in the biogeochemical flows of nitrogen and phosphorus, which have been identified as one of the major risks on a global scale, also considering its social implications, since the use of macronutrients is essential to guarantee the food needs of the world population.

In this context, there is a growing interest in the evaluation of the environmental impact related to this issue, particularly with regard to the effects of changes in the nitrogen cycle and the methods for quantifying them. In the latter field, several researches have recently been developed focusing on the indicator known as the nitrogen footprint, associated with the environmental releases of reactive nitrogen.

This study proposes an innovative method to quantify the reactive nitrogen emissions of a product system through the reactive nitrogen indicator; the method is designed using as a reference the requirements of the international standards ISO 14040 and ISO 14044, in order to be aligned with the operating procedures of the life cycle assessment technique, thus differing from the previous approaches to calculate the nitrogen footprint.

As part of the study, the proposed method is applied to calculate the reactive nitrogen emissions of a set of agricultural and livestock supply chain products, using secondary inventory data from an internationally recognized database.

A validation of the method was also carried out by comparing references in the literature regarding the nitrogen footprint accounting for the same products, generally obtaining a good level of agreement.

The proposed method, due to its reproducibility, ease of application and completeness, can therefore be usefully applied to any product system for the calculation of reactive nitrogen emissions, thanks to an innovative approach that meets the requirements of life cycle assessment.

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1. Introduction

The role of Nitrogen as an essential element for life on earth and a fundamental constituent of many compounds found in living cells within plants, animals and humans is known. However, the abundant supply of gaseous dinitrogen (N_2) in the atmosphere is in a chemical form that plants and animals cannot use directly; only a few special microorganisms can convert atmospheric nitrogen into reactive forms that plants and animals can use (Galloway et al., 2004). Reactive Nitrogen (N_r), commonly defined as all nitrogen forms apart from N_2 , is therefore an indispensable nutrient for agricultural production and human alimentation (Bodirsky et al., 2014). Taking this aspect into account, it should be emphasized that “food insecurity at the global level has been increasing steadily over the past 8 years - since FAO first started collecting data in 2014. In 2020, the year the COVID-19 pandemic spread across the globe, it rose nearly as much as in the previous five years combined. Updated projections of the number of undernourished people suggest that nearly 670 million people will still be undernourished in 2030” (FAO, 2022). Strategies linked to the achievement of the goal of “zero hunger” are also strongly present in the context of the sustainable development goals which, among other things, provide, by 2030, to “duplicate the agricultural productivity” and to “ensure sustainable food production systems and implement resilient agricultural practices that increase productivity and production, that help maintain ecosystems, that strengthen capacity for adaptation to climate change, extreme weather, drought, flooding and other disasters and that progressively improve land and soil quality” (United Nations, 2022). And precisely the provision of N_r of anthropic origin, through mineral fertilisers, has certainly contributed and will continue to contribute to the increased production of agricultural products needed to feed the increasing global population (Erisman et al., 2008) and hence to food security.

Regarding sustainability in the food production, on the other hand, it has been highlighted that the continuous increase in production of N_r “has led to a progressive alteration of the natural cycle of nitrogen, exceeding the limit defined for the planetary boundary, fixed as a safe operating space for human societies to develop and thrive” (Steffen et al., 2015). Besides the benefits of the use of nitrogen compounds, nitrogen, in its various chemical forms, plays indeed a major role in several environmental issues such as “acidification and eutrophication of soil, groundwater and surface waters, decreasing ecosystem vitality and biodiversity, also causing groundwater pollution through nitrate leaching. Nitrogen compounds give also a contribution to carbon sequestration, global climate change, and formation of ozone, oxidants and aerosols, potentially posing a threat to human health and affecting visibility. Each of the emissions takes part in the cycling of nitrogen causing a number of different effects with its consequent linkages” (Erisman et al., 2011).

There are several studies that describe and quantify the environmental effects of human perturbations on nitrogen cycle; an in-depth analysis of this topic is the one proposed by Galloway et al. (2004), in which reference is also made to papers that addressed the nitrogen cycle on a global and regional scale, to the major phases and components of the nitrogen cycle, and to the relationship to public policies.

Recently, with reference to the quantification of impacts related to the nitrogen cycle, different calculation and communication methods have also been proposed that fall within the scope of the nitrogen footprint (NF) tool. In this context, the most recognized definition of the nitrogen footprint as “the total amount of reactive nitrogen released to the environment as a result of an entity’s resource consumption, expressed in total units of N_r ” (Leach et al., 2012). This definition is included within a general project named N-PRINT that also presents a first accounting tool for the nitrogen footprint, the nitrogen-calculator (N-Calculator), with the aim of helping consumers to understand and possibly reduce their personal nitrogen footprint.

Subsequently to this first approach, various applications have been proposed and implemented using different analysis and calculation methods, according to specific objectives. All tools are characterized by a life cycle approach with different degrees of application, by considering various

phases of the life cycle to calculate the NF of the analyzed entities. Furthermore, current applications are exclusively in the agricultural and livestock sectors with reference to food products.

Early works in this topic refer to the tools proposed by Leach et al. (2012), mentioned above, by carrying out an analysis of reactive nitrogen released for the production of food, for the use of energy during the production phase and for the consumption of food (Mungcharoen and Suwanmanee, 2021; Pierer et al., 2014). This approach is based, for the most relevant part of NF calculation, on the concept of virtual nitrogen (Burke et al., 2009; Galloway et al., 2007), understood as any nitrogen that was used in the food production process and is not in the food product that is consumed. Virtual nitrogen is used to estimate the N_r lost to the environment along the production, processing, and consumption stages of the life cycle. The release of N_r is thus obtained not by an analysis of the actual emissions, but by difference with respect to the nitrogen content of the final product. This methodology can therefore be applied only to food products for which the virtual nitrogen coefficients are available and does not allow highlighting the actual flows of nitrogen-containing substances in all phases of the product life cycle.

Other and more recent applications, developed mainly in the Chinese area (Chen et al., 2020; Xu et al., 2020a, 2020b), use specific emissions factors - contained, for example in the IKE eBalance database (IKE Environmental Technology CO., 2006) - to obtain N_r losses related to farm inputs and eutrophication potential factors based on internationally adopted characterization models to obtain N_r losses in the food crop production processes. These applications are also related to food products, but the NF calculation is based on actual emissions of nitrogen-containing substances, with an approach much closer to the principles of LCA (ISO, 2020a, 2020b). However, it should be noted that the database and the eutrophication potential factors used in this approach take into account only compounds that are responsible for most of the environmental impacts related to the perturbation of the nitrogen cycle (mainly NH_3 volatilization, N_2O emissions, leaching of NO_3 and NH_4^+).

With reference to the different approaches developed to date and presented above, as well as pointed out by Pelletier and Leip (2014), although research has usually been based on LCA, “consistency and comparability of studies are hampered by the current lack of common methodologies”. To address these potential weaknesses, the authors developed a two-step method (consisting of classification and characterization) for systematic inventorying and aggregation of nitrogen mobilization flows in product systems and emissions in life cycle assessment. In particular, they proposed an approach, which they refer to as “characterization”, based on N-equivalent factors for nitrogen-containing compounds, obtained by calculating the ratio of N mass to total molecular mass for each compound of interest. The approach proposed by the authors is certainly valid, even if it is not adequately highlighted its compliance with the principles and requirements of the LCA envisaged by international standards (ISO, 2020a, 2020b). In fact, in the study, the inventory analysis phase is not addressed from a methodological point of view; furthermore, the characterization phase and those identified as characterization factors could be improved with greater alignment with the provisions of the reference standards. The proposed characterization factors, indeed, should be considered a method for aggregating the results of the inventory analysis, rather than impact-oriented factors derived from a characterization model, based upon an identifiable environmental mechanism, as required by the LCA standards (ISO, 2020b). Finally, the authors apply the method to the aggregate inventory of a set of products and not to a specific product system, making it difficult to compare the results with previous specific NF calculation applications, also basing the calculation on a limited number of substances.

Considering the importance and diffusion of the LCA technique and the issues presented regarding nitrogen flows and related environmental impacts, it is considered useful to propose a method to quantify the overall perturbation of the nitrogen cycle attributable to a product system in accordance with the approach used by researchers who make use of the LCA technique and related tools (software and databases) and based on the requirements of the LCA standards ISO 14040 and ISO 14044 and. This article

presents such a method that, with the aim of overcoming the potential limitations of existing approaches, can be applied, using the concept of reactive nitrogen, to any product system starting from the results of a life cycle inventory analysis; the method takes into account all the emissions of nitrogen-containing substances, in order to calculate the reactive nitrogen indicator (N_rI), which, to all intents and purposes, corresponds to what is currently referred to as nitrogen footprint.

The proposed method is described in detail, presenting the underlying mathematical model and then applied to nine products from the agricultural and livestock sectors using secondary data from a specific database in order to demonstrate its efficacy and consistency; the results obtained in terms of N_rI are furthermore compared with the NF values reported in some scientific papers for the same products.

It must also be emphasized that, in this study, the wording “reactive nitrogen indicator” (N_rI) is used rather than “nitrogen footprint” (NF), while referring to the same metric. In fact, given the definition of footprint contained in international standards, “metric(s) used to report life cycle assessment results addressing an area of concern” (ISO, 2017), the debate on which metrics can actually be considered as footprints is still open (see, for example, Matuščík and Kočí, 2020 or Ridoutt et al., 2016). The current definition and framework for NF makes this an indicator that could be defined “at inventory level” as it provides an aggregate quantification of reactive nitrogen emissions, but does not include an assessment of the actual environmental impacts related to the nitrogen cascade, which, as mentioned, can be multiple and heterogeneous (e.g., eutrophication, global warming, acidification, etc.); an impact-oriented NF metric should, moreover, consider the issue of spatial variability of N impacts (see, for instance, De Vries et al., 2013), a factor that is not addressed in any of the NF proposed tools. This specific topic, however, must be considered outside the scope of the present study.

2. Material and methods

2.1. Method framework and supporting tools

The method proposed for the quantification of reactive nitrogen emissions of a product system, based on the calculation of the reactive nitrogen indicator (N_rI) using a LCA approach, is shown, in its general framework, in Fig. 1.

The method is based on the provisions of international standards for life cycle assessment (ISO, 2020a, 2020b) and can be applied as a stand-alone tool, considering only nitrogen streams, or, more reasonably, it can be one of the outputs of a LCA analysis as in the applications carried out in the present study.

According to this last approach, the first phase of the method consists precisely in the complete inventory analysis of the considered product system, for the details of which reference can be made to the requirements of the above-mentioned international standards. Once the results of inventory analysis have been obtained, the selection of nitrogen inventory data must be made: from an operational point of view, this step consists in a selection of the results of the full inventory, in order to identify the streams of nitrogen-containing substances attributed to the system, thus obtaining the Nitrogen inventory results. Subsequently, the nitrogen coefficients are calculated for each of the nitrogen-containing substances detected as a result of the nitrogen inventory. Finally, the reactive nitrogen indicator is obtained by applying the nitrogen coefficients to the streams of

nitrogen-containing substances resulting from the nitrogen inventory. The method and calculation formulas are described in detail in the following paragraphs.

The SIMA PRO software (PRé Sustainability, 2022) and the ecoinvent database (Wernet et al., 2016) were used for the application part of this work. Although different databases could have been used as a source for secondary data, based on the analysis carried out, the ecoinvent database proved to be most complete and updated among those available within the SIMA PRO software. In particular, the ecoinvent v3 database (converted ecoinvent 3.6, data compiled in December 2019) was used with system model “cut-off by classification”.

2.2. Nitrogen coefficients determination

Starting from the standard inventory results of the datasets contained in the ecoinvent database, which include >1500 input and output streams, nitrogen-containing substances and compounds were selected, identifying approximately 290 streams. The ecoinvent database specifies, for each stream, the output compartment (“air, water, soil”) and input compartment (“raw”). As far as nitrogen is concerned, streams related to the input compartment (raw) are not exhaustive with respect to the total molecular nitrogen absorbed by the product system. This probably happens because nitrogen is not considered a “scarce resource” and therefore, from an LCA perspective, the potential impacts attributable to its depletion are not accounted for.

Consistent with the purposes of the proposed indicator, in this approach only the output flows were considered, ignoring the flows identified as ‘raw’ for nitrogen and for any other nitrogen-containing items (e.g.: granite, gravel, sodium nitrate); however, the input flows are useful and must be taken into account to carry out mass balances in the inventory analysis phase. For this purpose, particular attention must be paid to the mass balance of the processes that absorb molecular nitrogen from the air (e.g., production of ammonia or combustion processes) for which the incoming flows are not specified in the standard databases.

For each of the identified nitrogen-containing substances, the nitrogen coefficient (N_c) was calculated according to the following formula (excluding the exceptions specified below):

$$\text{nitrogen coefficient}(N_{c_i}) = \frac{\text{molar mass of N contained in the substance/compound } i}{\text{molar mass of the substance/compound } i} \quad (1)$$

Table 1 shows, by way of example, the nitrogen coefficients calculated for some of the nitrogen containing substances; for molecular formula and molecular mass reference was made to the public web database “Pubchem” (National Center for Biotechnology Information, 2022). The complete list of calculated nitrogen coefficients is available as Supplementary material (S1).

In order to calculate the nitrogen coefficients, some assumptions were made regarding database streams, as specified below:

- the emissions of “nitrogen oxides” were considered as nitrogen dioxide, estimated to be substance most likely present among the various forms of nitrogen oxides;
- for the determination of nitrogen content, for the “nitrogen organic bound” stream, the standard nitrogen-to-protein conversion factor of 6,25 was used;

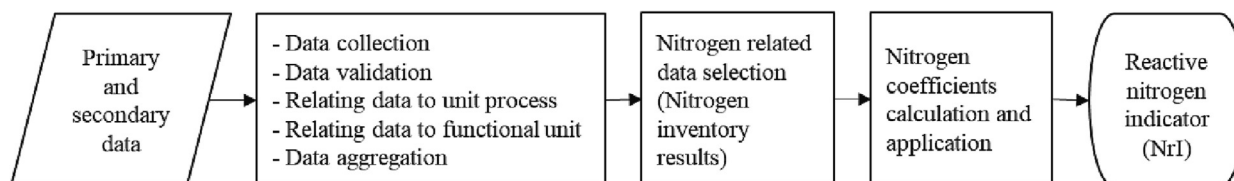


Fig. 1. Method framework.

Table 1
Nitrogen coefficients (non-exhaustive list).

Substance/compound	Molecular formula	Compartment	Molar mass [g/mol]	Nitrogen atoms	Nitrogen content [g/mol]	Nitrogen coefficient
Ammonia	NH ₃	Air, water, soil	17,031	1	14,007	0,822
Ammonium, ion	NH ₄ ⁺	Air, water, soil	18,039	1	14,007	0,776
Dinitrogen monoxide	N ₂ O	Air, water, soil	44,013	2	28,014	0,636
Nitrate	NO ₃	Air, water, soil	62,005	1	14,007	0,226
Nitric oxide	NO	Air	30,006	1	14,007	0,467
Nitrite	NO ₂ ⁻	Air, water	46,006	1	14,007	0,304
Nitrogen dioxide	NO ₂	Air, water, soil	46,005	1	14,007	0,304
Nitrogen fluoride	F ₃ N	Air	71,002	1	14,007	0,197
Nitrogen, organic bound	n.a.	Water	n.a.	n.a.	n.a.	0,160
Particulates, <2,5 μm	n.a.	Air	n.a.	n.a.	n.a.	0,087
Particulates, >2,5 μm, and <10 μm	n.a.	Air	n.a.	n.a.	n.a.	0,028
Urea	CH ₄ N ₂ O	Air, water, soil	60,056	2	28,014	0,466

- database includes different types of particulate matter emissions (Particulates, <2,5 μm; Particulates, >2,5 μm, and <10 μm; Particulates, >10 μm) which, as is known, may contain nitrogen compounds that can be traced, for their chemical composition, to ammonium ion (NH₄⁺) and nitrate ion (NO₃⁻). To estimate the nitrogen content of particulate, reference was made to the results reported by Sillanpää et al. (2006) and Masri et al. (2015) regarding the chemical composition of particulate matter in urban sites in Europe and U.S.; starting from these researches, an average value of the nitrogen coefficient was calculated equal to 0,028 for “Particulates, >2,5 μm, and <10 μm” and equal to 0,087 “for Particulates, <2,5 μm”. No estimates were made regarding the output “Particulates, >10 μm” as no reliable studies were found in the literature.
- the elementary flow “Nitrogen” was considered an emission of molecular nitrogen (N₂), for instance, as a result of denitrification processes; a coefficient of 0 was assigned to this flow, according to Pelletier and Leip (2014), who state that “denitrification, through a series of intermediate gaseous nitrogen oxide products, returns reactive nitrogen to its most thermodynamically stable form, nitrogen gas (N₂). Therefore, N₂ must be assigned a characterization factor of 0 in impact assessments that quantify contributions to perturbation of the nitrogen cycle”.

The approach adopted is that proposed by Pelletier and Leip (2014); however, in this work it is preferred not to use the term “characterization factor” because the coefficients obtained are not functional to calculate of an effective impact indicator result for a defined impact category, but rather they are used to aggregate the inventory results into a single indicator: exactly the reactive nitrogen, as specified below.

2.3. Calculation of reactive nitrogen indicator

Starting from the nitrogen coefficients, obtained with (1), the reactive nitrogen indicator, defined as the total reactive nitrogen released by a product system in the different environmental compartments, is calculated according to the following:

$$\text{reactive nitrogen indicator } (N_r I) = \sum_{i=1}^n \sum_{j=1}^m s_{i,j} * N_{c_i} \left[\frac{g}{\text{functional unit}} \right], \quad (2)$$

where:

- N_rI is the reactive nitrogen indicator calculated for the product system;
- i is the identification index of the specific substance/compound;
- j is the identification index of the substance compartment of emission;
- n is the total number of nitrogen-containing substances and compounds for the product system;
- m is the total number of the compartments of emission (for this study m = 3; the considered compartments are: air, water and soil).
- s_{i,j} is the emission of the substance i in the compartment j as resulting from inventory analysis [g/functional unit];
- N_c_i is the nitrogen coefficient for the substance/compound i;

As already mentioned, the reactive nitrogen indicator, which, according to the current meaning, accounts for the nitrogen footprint, provides an aggregate quantification of the reactive nitrogen emissions, without specifying the type of related impacts.

The calculation formula is made explicit in order to clearly highlight the different emission compartments and to make it easily applicable to the standard outputs of the inventory analyzes of LCA studies. A particular application of (2) also allows, considering index j fixed, to quantify the reactive nitrogen emissions of the system produced for each primary emission compartment (air, water, soil).

2.4. Application of the method and comparison with the results of the literature

The proposed method was applied to a set of 9 products from the agricultural and livestock food sector (rice, maize, wheat, potato, tomato, pepper, milk, poultry, swine). The products were selected from the ecoinvent database, using the following datasets:

- Rice: 1 kg Rice, non-basmati {CN} | rice production, non-basmati. The dataset is representative of Chinese production.
- Maize: 1 kg Maize grain {RoW} | production. The dataset is representative of average production outside the United States, South Africa, Brazil, Canada, and India.
- Wheat: 1 kg Wheat grain {RoW} | wheat production. The dataset is representative of average production outside the United States, South Africa, Brazil, Canada, India, Australia, Germany, France, and Spain.
- Potato: 1 kg Potato {RoW} | production. The dataset is representative of average production outside the United States, Canada, Ukraine, Russia, China, and India.
- Tomato: 1 kg Tomato, fresh grade {RoW} | tomato production, fresh grade, in heated greenhouse. The dataset is representative of average production outside Netherlands.
- Pepper: 1 kg Bell pepper {GLO} | bell pepper production, in heated greenhouse. The dataset is representative of a global average production.
- Milk: 1 kg Cow milk {RoW} | milk production, from cow. The dataset is representative of average production outside Canada.
- Poultry: 1 kg Chicken for slaughtering, live weight {GLO} | chicken production. The dataset is representative of a global average production.
- Swine: 1 kg Swine for slaughtering, live weight {RoW} | swine production. The dataset is representative of average production outside Canada.

The choice of products was made considering also the results of scientific works in which the NF was calculated for the same products; the results obtained in terms of N_rI are then compared with the NF values reported in scientific papers, with the aim of evaluating the actual applicability of the proposed method and its validity to calculate an aggregate indicator of the reactive nitrogen emissions of a product system. For the analysis, production datasets were selected that are representative of a cradle to gate approach (i.e., no product distribution scenarios are included). The dataset inventory results were used to calculate the N_rI as described above, using

a simple algorithm to speed up the procedure. Specifically, the algorithm, through database or electronic spreadsheet formulas, extracts from the inventory analysis the flows of nitrogen-containing substances (Nitrogen inventory results), calculates the reactive nitrogen flow using the Nitrogen coefficients and finally the reactive nitrogen of the product system as the sum of the nitrogen flows.

3. Results

Fig. 2 shows the results of the calculation of the reactive nitrogen indicator (N_rI) for 9 products of the agricultural and livestock sector, as a result of the application of the proposed method.

As expected and already highlighted in several studies, the indicator shows higher values for livestock products than for agricultural sector products. The absolute lowest values of the indicator are also found for products for which the datasets refer to the production “in heated greenhouse” (tomato and pepper).

Fig. 3 shows, for each product, the contribution to the N_rI of the main streams.

Regarding the results of the N_rI calculation for the nine products, considered as a whole, the streams that give the greatest contribution are “nitrate” in the compartment water, “ammonia”, “nitrogen oxides” and “dinitrogen monoxide” in the compartment air. Specifically, for all the products analyzed, with the exception of tomato and pepper (production in heated greenhouse), “nitrate” in the compartment water is the stream that contributes the most to N_rI with percentages ranging from 42,92 % (swine) to 74,33 % (potato) and an average percentage of 49,97 % for the nine products analyzed. For these same products, the second stream by contribution is “ammonia” in the compartment air with percentages ranging from 12,00 % (potato) to 42,84 % (swine), and an average percentage of 25,36 % for the nine products analyzed. The ammonia output stream was, proportionally, much more significant for products in the livestock sector (with an average percentage of contribution to N_rI equal to 40,10 % and with values ranging from 34,96 % of milk to 42,84 % of swine) compared to what was found for agricultural sector products (for which an average percentage of contribution to N_rI equal to 17,99 % was calculated, with values ranging from 4,85 % of pepper to 28,46 % of rice). The preponderant contribution of streams nitrate and ammonia and the percentage values obtained with reference to N_rI are in line with what is reported in the literature results with reference to NF (see, for example Xu et al., 2020b). Regarding dinitrogen monoxide in the compartment

air, which has important implications for environmental impact in terms of global warming, an average percentage contribution to N_rI equal to 5,86 % was detected, with values ranging from 2,60 % (rice) to 10,15 % (tomato). Is worth highlighting that the stream of “nitrogen oxides” in compartment air, which is not taken into account as a contribution to the NF in the previous studies pertaining to the emissions in the production phase, is the most significant stream for greenhouse-grown products, with a contribution to N_rI equal to 57,21 % for tomato and 44,42 % for pepper. Furthermore, the design and application has also allowed to bring out the contribution of other streams such as “particulates” in the compartment air (with emissions of the average order of magnitude of 10^{-1} g of reactive nitrogen per kg of product and an average contribution to N_rI for the products analyzed products of approximately 1,4 %) or, albeit to a lesser extent, cyanide in the compartment air and atrazine in the compartment soil (with emissions of the average order of magnitude of 10^{-2} g of reactive nitrogen per kg of product). It is emphasized that these emissions, like others that are not mentioned here for the sake of brevity, are not considered in previous studies relating to NF, although they could make a non-negligible contribution in some specific applications.

As mentioned above, the proposed method also makes it easy to calculate the amount of reactive nitrogen emission for each primary environmental compartment (air, water, and soil), thus providing a rough indication of the nature of the potential impacts, caused by emissions of reactive nitrogen, for the product system. An example of such an additional analysis for 4 of the products studied in this study is shown in Fig. 4.

It can be seen that almost all of the reactive nitrogen emissions for the considered products have, as their first destination, the air and water compartments with a different prevalence of one of the two; emissions in the soil compartment are, instead, much lower and almost absent for greenhouse-grown products.

4. Discussion

Table 2 shows the comparison of the results of the calculation of N_rI for the 9 products of the agricultural and livestock sector with some results found in the literature regarding the nitrogen footprint accounting (NF). The geographic area of reference is indicated for dataset and literature results: correspondence in this regard was sought where possible (e.g. for rice); otherwise, database global geographic areas were used (see § 2.4). For the results of the literature, the source and an indication of the boundaries (stages of the life cycle and reference geographical area) of the system

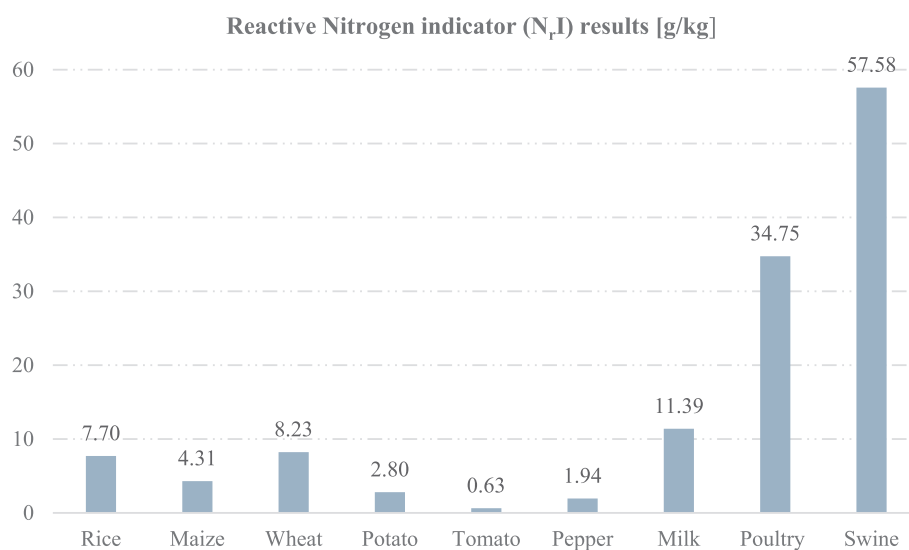


Fig. 2. Results of the reactive nitrogen indicator for 9 products of the agricultural and livestock sector.

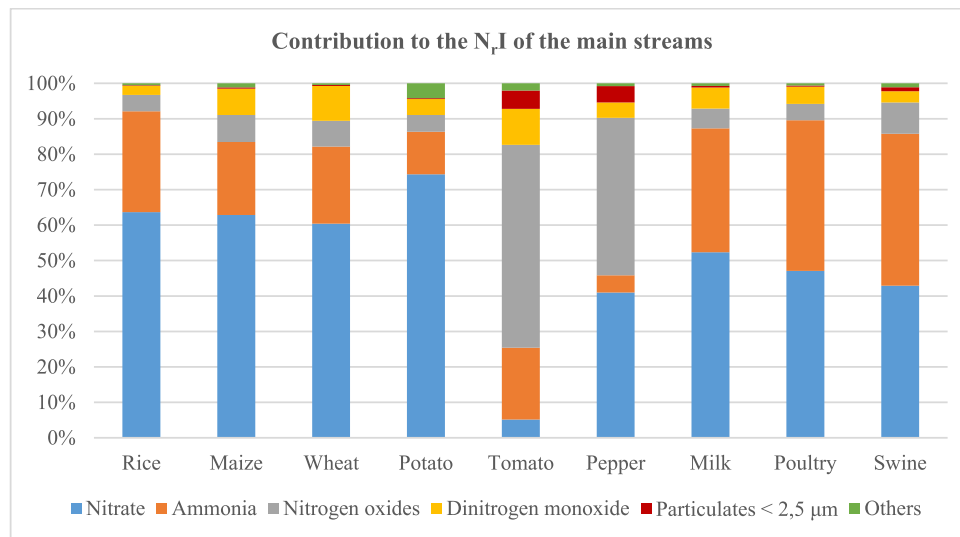


Fig. 3. Contribution to the N_I of the main streams for 9 products of the agricultural and livestock sector.

to which the calculation of NF is applied are shown. With regard to the literature data, the following should also be noted (for more details, refer to the specific sources indicated):

- the reported results of the work of [Leip et al. \(2014\)](#) are obtained by analyzing the graphs contained in the paper, since the exact values of the results are not disclosed: they are therefore affected by a greater degree of uncertainty than the others data. The data range refers to the lower and upper quartiles and takes into account the maximum and minimum values of the results obtained using two different impact assessment models (CAPRI and MITERRA);
- the reported results of [Mungcharoen and Suwanmanee \(2021\)](#) include two values of which the higher takes into account the effect of international trade;
- with respect to [Xu et al. \(2020b\)](#) data, the lower and higher values of the results obtained from the experiments (three water management practices combined with three nitrogen management practices) are reported;
- regarding the data from [Xu et al. \(2020a\)](#), the average values of the experiments on two modes of management are reported: the farmers' practice (higher value) and the reduced inputs of water and nitrogen fertilizer (lower value); furthermore, for maize cultivation, only "summer maize" results were considered, ignoring "spring maize";
- [Wang et al. \(2020b\)](#) reported data are the lowest and the highest values of the results obtained for four experimental treatments ("farmers' practice", "soil remediation", "soil remediation and crop planting density optimization", "integrated soil-crop system management") for two reference years (2017 and 2018);

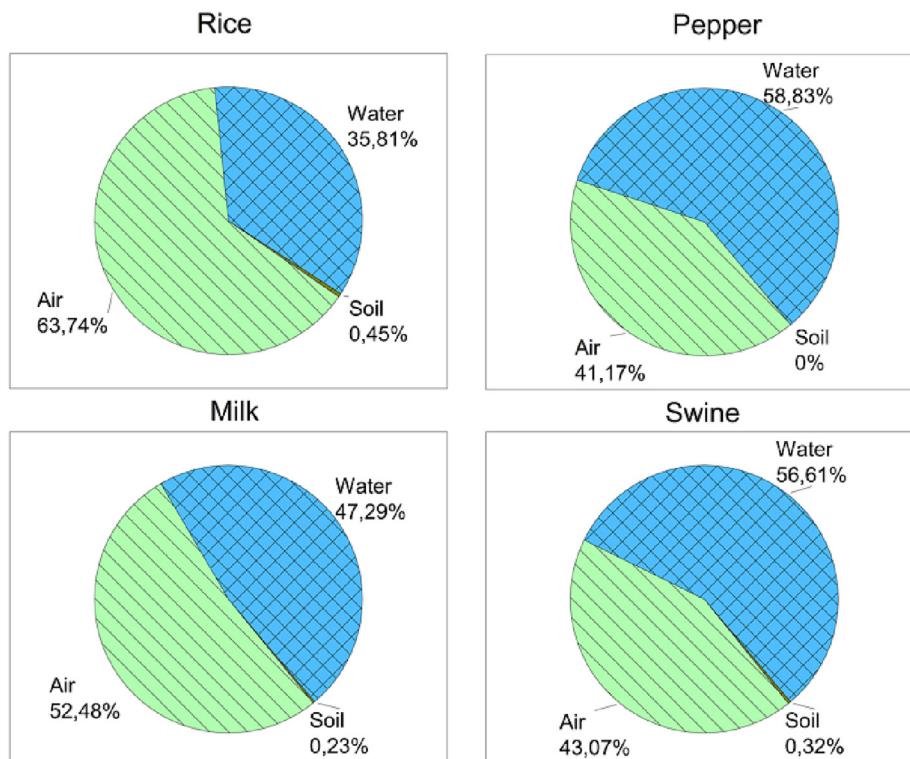


Fig. 4. Primary environmental compartment of emission of reactive nitrogen (example for 4 products).

Table 2
Comparison of reactive nitrogen indicator results with literature results for nitrogen footprint.

Product	N _r I [g/kg]	Database geographic area	NF (literature) [g/kg]	System boundaries (literature)	Literature source
Rice	7,70	China	11,4/22,1 11,6 1,3 ± 0,1/6,1 ± 1,3	Farm-to-fork (Thailand) Cradle-to-farm-gate (China) Cradle-to-farm-gate (China)	(Mungcharoen and Suwanmanee, 2021) (Chen et al., 2020) (Xu et al., 2020b)
Maize	4,31	Rest of the world	3,27/11,65	Cradle-to-farm-gate (China)	(Xu et al., 2020a)
Wheat	8,23	Rest of the world	2,88/5,81	Cradle-to-farm-gate (China)	(Xu et al., 2020a)
Potato	2,80	Rest of the world	3,2 0,3–5	Farm-to-fork (Austria) Farm-gate (European Union)	(Pierer et al., 2014) (Leip et al., 2014)
Tomato	0,63	Rest of the world	0,42 ± 0,05/1,18 ± 0,06	Cradle-to-farm-gate (China)	(Wang et al., 2020b)
Pepper	1,94	Global	1,2 ± 0,06/2,1 ± 0,05	Cradle-to-farm-gate (China)	(Wang et al., 2020a)
Milk	11,39	Rest of the world	33,4/34,3 3,2	Farm-to-fork (Thailand) Farm-to-fork (Austria)	(Mungcharoen and Suwanmanee, 2021) (Pierer et al., 2014)
Poultry	34,75	Global	256,9/803,1 67 50–170	Farm-to-fork (Thailand) Farm-to-fork (Austria) Farm-gate (European Union)	(Mungcharoen and Suwanmanee, 2021) (Pierer et al., 2014) (Leip et al., 2014)
Swine	57,58	Rest of the world	49,3/49,7 60–200	Farm-to-fork (Thailand) Farm-gate (European Union)	(Mungcharoen and Suwanmanee, 2021) (Leip et al., 2014)

- as regards Wang et al. (2020a) data, the mean of lower and the mean of higher values for three tested treatments (“farmers’ current practice”, “soil remediation”, “integrated soil-crop system management”) are reported.

It can be noted that the results, in terms of N_rI, are in most cases comparable with those published and analyzed relating to NF, with greater convergence for some agricultural products such as rice, maize, potato, tomato and pepper. A very high correspondence can also be found if we consider the ratio of the results (in terms of N_rI and NF) for potato, cereals and swine using as a reference the studies in which NF is calculated for different products (Leip et al., 2014; Pierer et al., 2014). The differences found, which in some cases are even high (for example regarding milk and poultry), can be attributed to several elements, the main ones being the approach used for the analysis – mainly for the absence of standard methods for NF calculation as already highlighted – the considered system boundaries and the allocation choices (important for example in the case of milk) which in some studies are not disclosed. Regarding the system boundaries, in the scientific works analyzed, the NF calculation approach includes, in some cases, also the food consumption stage (e.g., Pierer et al., 2014 or Mungcharoen and Suwanmanee, 2021) thus generating, for NF, greater final results that are more evident in the case of products from the livestock sector – for the higher protein content which determines higher nitrogen emissions in the final stages of the life cycle. Minor differences in results are in general found with studies that refer to a more LCA-compliant approach with cradle-to-farm-gate system boundaries and actual emissions calculation for the production phase (Wang et al., 2020a, 2020b; Xu et al., 2020a, 2020b). Additionally, the geographical context of reference, as mentioned above, is a relevant factor for the final results; by example and limiting the analysis to what concerns N_rI calculation, it is observed that, for the item “rice, non-basmati”, different results (with deviations >70 %) are obtained using datasets of different geographical contexts (7,70 g/kg for the “China” dataset, 13,12 g/kg for the “U.S.” dataset and 12,90 g/kg for the “rest of the world” dataset). Moreover, the methodological approach that considers the contribution of almost 300 output streams – using the proposed method and database for N_rI – could have a not negligible impact on the final results as regards the comparison with NF calculation applications. This last consideration is particularly valid for studies that use the LCA approach and account analytically for the releases of N_r in the production phase (Chen et al., 2020; Wang et al., 2020a, 2020b; Xu et al., 2020a, 2020b). In fact, in these examples, only the contribution of volatilization of NH₃, emissions of N₂O, leaching of NO₃⁻ and NH₄⁺ is considered for the production phase, while the contribution of other emissions, such as nitrogen oxides, is ignored; based on application carried out in this study, however, nitrogen oxides emissions are relevant for greenhouse-grown products, as seen, and, in any case, not negligible also for the other

products analyzed (with percentage contribution to N_rI ranging from 4,58 % for rice to 8,82 % for swine).

As seen, the proposed method produces results similar to those of the NF indicator, but, unlike the approaches currently used, it is perfectly aligned with the LCA technique and with the related standards requirements.

In particular, compared to the tools proposed by Leach et al. (2012) and the studies that can be traced back to them, the method is applicable to any product system, not only to food products, since it considers the actual emissions of the analyzed entity, rather than using virtual nitrogen coefficients. Compared to other applications that use specific emission and eutrophication potential factors, the proposed method is designed to take into account the contribution of any nitrogen-containing emissions, not only some of them, allowing a more complete and accurate assessment of reactive nitrogen flows. Compared finally to the methodological proposal of Pelletier and Leip (2014), which has the same approach, the presented method provides a perfectly aligned with LCA and inventory-oriented scheme, including a detailed description of all the phases and explanation of calculation formulas that allow replication of the procedure. In this regard, a punctual comparison is not possible as the formulas underlying the method, although intuitive, are not explicit in the published research. Furthermore, the applications presented in this study refer to inventories of specific product systems, rather than to a generic set of consumer products, thus allowing for a test of the method very close to real operating conditions such as those found in LCA studies. Finally, the streams considered in the inventory analysis are very numerous (the contribution of nearly 300 nitrogen-containing substances was calculated), thus allowing to obtain more accurate results, also accounting for the contribution of particulate emissions (not considered in the approach of Pelletier and Leip, 2014), which, as seen, is in some cases significant.

The proposed method can also be easily integrated with additional analyzes, such as those presented and related to the primary compartment of emission of reactive nitrogen emissions, thus providing information on potential environmental impacts and indications useful for identifying significant issues.

As regards the proposed method, it is based on the LCA technique and therefore affected by the limits of the latter relating to the inventory analysis phase (Islam et al., 2016) or to the handling of multifunctionality (Moretti et al., 2020). The N_rI indicator can be applied without preclusion to inventories developed with different end-of-life or co-product allocation approaches. For the presentation of the results in this study, reference was made to the ecoinvent databases with a cut-off approach, as required by various sectoral rules (e.g., International EPD System, 2022). For the sake of completeness, the results obtained with the end-of-life allocation approach “at point of substitution” (APOS) are given in the Supplementary information (S2). The variation of the results obtained with the “APOS” approach compared to the “cut-off” approach is limited and between

0,12 % for swine and –8,18 % for chicken. For a more detailed discussion of the differences in the approaches, see Wernet et al. (2016).

With regard to the data used and the results obtained in this study, they can be traced back to the contents of the selected database and, therefore they are conditioned by the methodological choices inherent in them, with reference, for example, to the system boundaries and allocation choices. In particular, to calculate the inventory of agricultural and livestock products, the ecoinvent database uses the methodological approaches for life cycle inventory of agricultural products contained in the World Food LCA Database (Nemecek et al., 2019) to which reference is made for details. These guidelines provide for a cradle-to-gate approach for both crop and animal production considering, as output flows, direct emissions from field and farm, indirect emission from inputs (e.g., energy carriers, fertilisers, pesticides), waste and wastewaters as well as products and co-products. Regarding direct emissions from crop and animal production, limiting the discussion to the substances with the greatest contribution to reactive nitrogen, for ammonia, the emission factors for mineral fertilisers are taken from the EMEP guidelines (European Environment Agency (EEA), 2016) for crop production, while the emission factors for animal housing are taken from the IPCC 2006 guidelines (Eggleston et al., 2006); for nitrate leaching, the SALCA-NO₃ model by Richner et al. (2014) is applied for Europe, while, for non-European countries, the SQCB-NO₃ model is used (Faist Emmenegger et al., 2009); for nitrogen oxides, the emission factor for the application of mineral and organic fertilizer is taken from European Environment Agency (EEA) (2016), while the emission factor for manure storage is based on the volatilization fraction given by IPCC (Eggleston et al., 2006).

It is noted that the proposed method was applied using predefined datasets of a specific database; however, as explained, it is designed to be applied to any product system. Obviously, in the case of output streams different from those analyzed, it may be necessary to calculate and apply the relative nitrogen coefficients to obtain the final results.

It should be further underlined that the use of secondary data in this study is functional to the comparison with the published studies in order to verify if the results obtained with the proposed method are in line. In fact, the contents of the databases represent average data and obviously differ from the results of an approach based on primary data. In the case of a specific application, the primary data must be privileged, in line with the requirements of the ISO 14040 and ISO 14044 standards.

Concerning the calculation of the nitrogen coefficients, the method is subject to the limits relating to the assumptions made for some database streams (see § 2.2) regarding which the following is highlighted:

- for “Particulates, <2,5 µm” and “Particulates, >2,5 µm, and <10 µm” streams a deepening could be performed to estimate the nitrogen content more accurately, considering that the results of the literature show variable compositions based on the reference geographic area. A specific study should also be performed to evaluate the possible nitrogen content in the stream “Particulates, >10 µm”, even if it is supposed a non-relevant nitrogen content that does not affect the validity of the final results of the applications. It is noted that, for particulate emission, an average coefficient calculated from literature data is proposed, thus introducing an uncertainty factor into the method. For completeness, it is specified that the contribution of particulate emissions to the total emissions, calculate by mass, for each of the product systems analyzed is <0,15 %, varying from 0,02 % for “Particulates, >2,5 µm, and <10 µm” (tomato) to 0,11 % for “Particulates, <2,5 µm” (swine);
- a methodological consideration should be made, in a specific study, regarding the molecular nitrogen released as a result of denitrification processes. In fact, a negative value could be assigned to this stream, in order to enhance the positive contribution of the processes, natural or anthropic, that reconvert reactive nitrogen into molecular nitrogen, thus avoiding potential environmental impacts. It must be emphasized that, in the applications carried out, the flows referred to as “nitrogen” are quite low and their contribution does not significantly affect the results;

- the assumptions relating to the streams “nitrogen organic bound” and “nitrogen oxides” are considered consistent with the scientific evidence and not significant with respect to the final results, even if an in-depth and contextual analysis of the actual composition of the emissions generically referred to as nitrogen oxides could be performed in the inventory analysis phase for more accurate applications.

Regarding the additional analyzes carried out to highlight the primary destination compartments of the reactive nitrogen emissions, it should be noted that the results obtained must only be considered as indicative of the potential impacts resulting from reactive nitrogen emissions. Furthermore, in this regard, the method only allows identifying the output compartment of the flows from the inventory analysis and does not provide information on the actual final destinations, due to the extreme mobility of reactive nitrogen in the various compartments presented in different studies (Galloway et al., 2004).

An uncertainty analysis was not performed for the study as it was not included in the objectives: specifically, for the secondary data used, the uncertainty factors included in the ecoinvent database can be used. The proposed method for the calculation of reactive nitrogen is based on molecular mass ratios and therefore does not introduce further contributions to the uncertainty, except for the assumptions illustrated in § 2.2.

Finally, as regards the part of the research relating to the comparison between the results of N_rI and NF, no considerations were made relating to the reference context and time periods, both as regards the validity of the databases and as regards the publications analyzed: only the differences relating to the geographical context and the system boundaries have been made explicit. In particular, it should be noted that the results of the NF reported literature are related to the specific studies conducted and therefore conditioned by the reference context analyzed in terms of, for example, the cultivation and breeding techniques, the technologies used, the reference time periods, the meteorological conditions, etc., while the databases are representative of average conditions of different contexts.

5. Conclusions

With reference to the quantification of reactive nitrogen flows in the environment that refers to an entity, a method is proposed to calculate and express, in aggregate form, the reactive nitrogen emissions of a product system in its life cycle using the reactive nitrogen indicator (N_rI). The method is developed in accordance with the LCA technique and the reference standards of (ISO 14040 and ISO 14044), is suitable to be supported by databases and software commonly applied in the LCA field, and produces results corresponding to those of the metric known as nitrogen footprint.

The proposed method has some specific characteristics that make it a suitable tool to overcome the limitations of current approaches to calculating the nitrogen footprint, since it is perfectly aligned with the LCA technique, is designed to account for all reactive nitrogen emissions, including, for example, the potentially significant contribution of particulate emissions, and is easy to replicate.

The method was successfully and efficiently applied to nine products in the food and livestock sectors, also thanks to the use of standard calculation procedures. The results obtained were also compared with some results found in the literature and related to the calculation of the nitrogen footprint, obtaining, in general, a good level of agreement; the differences found, in some cases even significant, are justifiable in view of the different approaches underlying each study and application and the methodological choices of the database.

Finally, in our opinion, the question of the real environmental impact attributable to the nitrogen cycle remains open. In fact, both in the case of the proposed method and in the case of NF as currently defined, these are evaluated at the inventory level, without expressing a specific quantification for the impact categories involved.

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CRedit authorship contribution statement

Filippo Zuliani: Conceptualization, Methodology, Investigation, Formal analysis, Software, Supervision, Validation, Data curation, Writing-Original draft preparation.

Alessandro Manzardo: Validation, Data curation, Validation, Writing - Review & Editing.

Alessandro Marson: Software, Data curation, Writing - Review & Editing.

Andrea Fedele: Writing - Review & Editing.

Data availability

The data used are included in international databases mentioned in the manuscript.

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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