



Review



Harmonizing methods to account for soil nitrous oxide emissions in Life Cycle Assessment of agricultural systems

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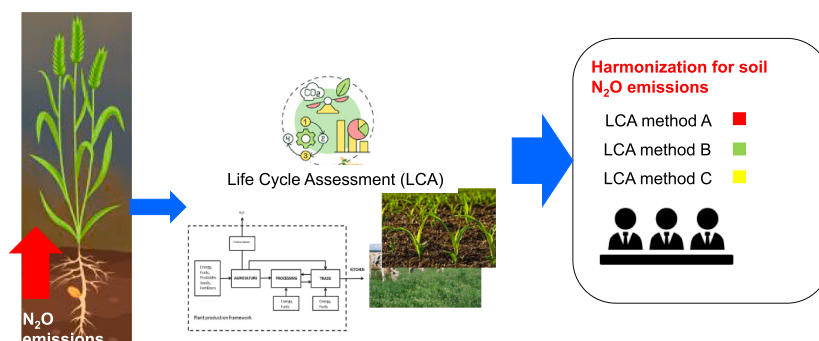
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HIGHLIGHTS

- Methods used in life cycle assessment (LCA) of agricultural systems to account for soil N₂O need to be improved.
- A method harmonization was carried out and recommendations for soil N₂O emissions were identified in agricultural LCA.
- The results showed that a high level of accuracy corresponded to a low level of applicability and vice versa.
- DNDC, DAYCENT and direct measurements scores high on accuracy to assess soil N₂O emissions in LCA of agricultural systems.
- Alternative to DNDC and measurements are IPCC Tier 1 or 2 methodologies to assess soil N₂O emissions in agricultural LCA.

GRAPHICAL ABSTRACT



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ARTICLE INFO

Editor: Mark van Wijk

Original content: [Criteria for LCA assessment of soil N₂O emissions in agricultural systems](#) (Original data)

Keywords:

LCA
Cropping systems
Livestock systems
Soil N₂O emissions
Methods
Harmonization

ABSTRACT

CONTEXT: Worldwide greenhouse gas emissions (GHG) reached 59 Gt of CO₂eq in 2019 and agricultural soils are the primary source of N₂O emissions. Life cycle assessments (LCA) have been successful in assessing GHG from agricultural systems. However, no review and harmonization attempt has been focused on soil N₂O emissions, despite the need to improve LCA methodologies for assessing GHG in agricultural LCA.

OBJECTIVE: We therefore undertook a review and harmonization of existing methods to account for soil N₂O emissions in LCA of agricultural systems and products: i) to compare current methods used in LCA; ii) to identify advantages and iii) disadvantages of each method in LCA; iv) to suggest recommendations for LCA of agricultural systems; v) to identify research needs and potential methodological developments to account for soil N₂O emissions in the LCA of agricultural systems. In this paper, we consider as soil N₂O emissions, those originated from soils in relation to fertilisers (organic and manufactured), crop residues, land use/land management change, grassland management, manure and slurry applications and from grazing animals.

METHODS: The approach adopted was based on two anonymous expert surveys and a series of expert workshops ($n = 21$) to define general and specific criteria to review LCA methods for GHG emissions used in LCA of agricultural systems. A broad list of keywords and search criteria was used as the research involved GHG assessment in agricultural LCA. Reviewed papers and methodology were then assessed by LCA and soil N₂O emission experts ($n = 14$).

RESULTS AND DISCUSSION: >25,000 scientific papers and reports were identified, 1175 were screened, 263 included in the final review and 31 scientific papers were related to soil N₂O emissions. The results showed that a high level of accuracy corresponded to a low level of applicability and vice versa, following the assessment framework developed in this work through participatory approaches.

SIGNIFICANCE: The choice of LCA methods, critical for high quality LCA of agricultural systems, should be based on the assessment objectives, data availability and expertise of the LCA practitioner. However, it is preferable to use DNDC model after calibration and validation or direct field measurements, considering system effects. When necessary data are lacking, IPCC tier 2 methodology where available should be used, otherwise 2019 IPCC Tier 1 methodology. This LCA method development should be synchronous with improvements of quantification methods and the assessment of a wider range of agricultural management practices and systems.

1. Introduction

Worldwide greenhouse gas (GHG) emissions reached 59 Gt of CO₂eq in 2019, while N₂O represents 4% of the total global emissions. However, following the Intergovernmental Panel for Climate Change 6th assessment report, nitrous oxide has a global warming potential with a 100 year horizon 273 times larger than carbon dioxide. Agriculture, forestry and land use sector contributed 22% of the total global GHG emissions (IPCC, 2022). Because of the large amounts of GHG emissions, there is an increasing demand for GHG emission reduction for every sector of the economy, including agriculture (IPCC, 2022).

Agricultural soils are the primary source of anthropogenic nitrous oxide (N₂O) emissions (Wang et al., 2018). Soil emissions due to synthetic fertilizer applications to soils accounted for 0.75% of the total global GHG emissions in 2019 (IPCC, 2022). N₂O emissions from manure management contributed 5% to global greenhouse gas emissions within the livestock production chains, while feed production accounted for 9.8% in 2015 (FAO, 2023).

Soil N₂O emissions are by-products of microbial processes transforming nitrate to nitrogen gas under microaerobic and anaerobic conditions (denitrification) or ammonium to nitrate under aerobic conditions (nitrification) (Oertel et al., 2016; Ussiri and Lal, 2013). These emissions are part of the N nitrogen cycle together with other pollutants (e.g. Ammonia, nitrate) which can cause other impacts such as acidification and eutrophication (Brady and Weil, 2002).

Nitrous oxide emissions are largely affected by the soil moisture and soil oxygen availability making these emissions highly variable throughout the season (Bastos et al., 2021; Dorich et al., 2020; Olesen et al., 2023). Indeed, a key parameter is the water filled pore space (WFPS), WFPS value above 60% creates favourable conditions for soil N₂O emissions through denitrification (Laville et al., 2011), but optimum N₂O production may occur at about 80% WFPS (Butterbach-Bahl et al., 2013). Thus, climate and soil types affect soil N₂O emissions (Butterbach-Bahl et al., 2013; Dorich et al., 2020; Loubet et al., 2011).

Further, N fertilizer use, N content and C/N ratio of manure or slurry, and the C/N ratio of crop residues also influence soil N₂O emissions (Dorich et al., 2020; Kimming et al., 2011; Sagar, 2010; Tuomisto et al.,

2012; Ussiri et al., 2009). Soil N₂O emissions are often characterized by peak emission events after fertilizer or manure applications, freezing-thaw periods and ploughing of grass, where most of the emissions occur during the growing season (Dorich et al., 2020; Giltrap et al., 2020; Olesen et al., 2023; Taki et al., 2019). Otherwise, the background N₂O emissions are generally low in concentration which makes field monitoring difficult and costly (Goglio et al., 2013; Laville et al., 2011; Olesen et al., 2023).

Accounting for fluxes of N₂O in LCA of agro-ecosystems is important for evaluating which management practices may enhance or mitigate climate change effects for different crop-livestock systems (Grossi et al., 2019; Sykes et al., 2019). Soil N₂O emissions from soils are evaluated mostly with regards to land management and land management changes (e.g. tillage, fertilizer application), and land use changes (from and to grassland/ cropland/ forest), following intergovernmental panel for climate change (IPCC) classification (McConkey et al., 2019; Ogle et al., 2019a, 2019b).

Life Cycle assessment (LCA) is an assessment method commonly used to assess crop, livestock systems and products due to its ability to identify environmental hotspots and trade-offs across different types of pollution (Cederberg et al., 2013), use of resources (e.g. energy and materials), biodiversity and human health impacts (Huijbregts et al., 2017; van der Werf et al., 2020; Zampori and Pant, 2019). LCA has also been widely used to assess climate change impacts of agricultural products and production systems (Grossi et al., 2019; Poore and Nemecek, 2018). This includes the assessment of different types of fertilizer, tillage practices and residues management within cropping systems (Goglio et al., 2014; Nemecek et al., 2015; Zaher et al., 2013). Other LCA research assessed the influence of the method used to estimate N₂O emissions on the overall LCA results of agricultural systems (Cabot et al., 2023; Goglio et al., 2018; Sinisterra-Solis et al., 2020).

Recently, a combined approach has been proposed for assessing livestock products and systems taking into account crop-livestock interaction (Ershadi et al., 2020; Marton et al., 2016; Parajuli et al., 2018). Considering the importance of mitigating GHG emissions there is an increasing need to assess complex livestock systems under current and future climate (Godfray et al., 2018; Willett et al., 2019).

Furthermore, improved LCA methodologies are required to better capture systems effects, crop-livestock interactions and circular economy (Costa et al., 2020; Grossi et al., 2019; Van Zanten et al., 2018).

Several harmonization attempts were focused mostly on sectors other than agriculture (Segura-Salazar et al., 2019; Siegert et al., 2019; UNEP, 2023a, 2023b), while others specifically focused on wines (Jourdain et al., 2020), food waste advocating for a better integration between LCA and soil science (Morris et al., 2017) or generally on livestock systems (FAO, 2020). No harmonization attempt exists for soil N₂O emissions in the LCA of agricultural systems, including crop-livestock interaction (FAO, 2020). Within this study, we therefore undertook a review and harmonization of existing methods to account for soil N₂O emissions in Life Cycle Assessment in agricultural systems, including excreted N on pasture, and products: i) to compare current methods used in LCA; ii) to identify advantages and iii) disadvantages of each method in LCA; iv) to suggest recommendations for LCA of crop-livestock systems; v) to identify research needs and potential methodological developments to account for soil N₂O emissions in the LCA of agricultural systems. In this paper, we consider soil N₂O emissions as those originated from soils in relation to fertilisers (organic and manufactured), crop residues, land use/land management change, grassland management, manure and slurry applications and from grazing animals. All the manure management emissions related to manure handling, storage and animal housing are out of scope of the present research as they do not originate from soil. This paper is part of a broader research project (PATHWAYS) aiming at assessing pathways to sustainability for livestock and food systems integrating crop-livestock interactions. In particular, the research presented in this paper is part of an effort to harmonize LCA methods related to GHG emissions in LCA of crop-livestock systems and soil N₂O emissions were investigated together with soil C, manure emissions and enteric fermentation. However, this paper will only present and discuss the outcomes limited to soil N₂O emissions.

2. Methodology

2.1. Search criteria

A systematic literature search was conducted using Scopus, Google Scholar and the Web of science search engines. The systematic literature search and review had a broader scope, which was to identify methodologies to assess soil C sequestration, soil N₂O emissions and enteric fermentation; rather than just soil N₂O emissions in the LCA of agricultural systems, as described by Goglio et al. (2023a). Thus, search terms and search term combinations employed are described below in Table 1, including all papers published between 2012 and 2022. These were selected as considered relevant for LCA of crop-livestock and agricultural systems and for soil N₂O emissions.

2.2. Screening and review procedures

The collected sources were screened against the following criteria: i) Peer-reviewed publications in a scientific journal, published by the European Commission, FAO or other international organizations; ii) English language publication; iii) Method is related to and applicable for LCA; iv) Method is related to agricultural systems or their components; v) Method is applicable for agricultural systems. A systematic review of the existing literature, based on the methodology described above, was conducted to provide a comprehensive assessment on how LCA methodologies include livestock GHG emissions in relation to soil N₂O from both cropland and grassland within crop-livestock systems. These include cropping systems receiving manure, sludge, slurry, grazed systems and all the related crop and grassland management practices (e.g. tillage, fertilizer management, residue management, weed control, irrigation). To achieve this, a review protocol was developed (Fig. 1), describing the search and screening process including an iterative

Table 1
Combinations of search terms for the subgroup “GHG Emission Issues”.

Database	Combination	Search strings ^a
Scopus & Web of Science	1	("LCA" OR "Life Cycle Assessment" OR "life cycle analysis") AND ("enteric fermentation")
	2	("LCA" OR "Life Cycle Assessment" OR "life cycle analysis") AND ("soil*") AND ("emissions" OR "nitrous oxide" OR "N2O" OR "carbon dioxide" OR "CO2" OR "carbon sequestration" OR "GHG" OR "greenhouse gas*" OR "C dynamics" OR "soil") AND ("carbon") AND ("livestock")
	3	("Life Cycle Assessment" OR "life cycle analysis") AND ("wheat" OR "maize" OR "grass" OR "barley" OR "oat" OR "soy*" OR "faba beans" OR "alfalfa" OR "clover" OR "sorghum" OR "Rye" OR "Ley") AND ("soil emissions" OR "soil carbon" OR "soil nitrogen" OR "soil organic matter" OR "nitrous oxide") AND ("feed" OR "fodder" OR "farming system" OR "farm")
	4	("Life Cycle Assessment" OR "life cycle analysis") AND ("livestock" OR "dairy" OR "cattle" OR "sheep" OR "pig*" OR "poultry" OR "goat*" OR "milk" OR "egg*" OR "chicken*" OR "cow*" OR "husbandry") AND ("emissions") NOT ("waste" OR "biofuel" OR "bioenergy")
	5	("LCA" OR "Life Cycle Assessment" OR "life cycle analysis") AND ("manure" OR "slurry") AND ("handling" OR "storage" OR "treatment" OR "emissions")
	6	("LCA" OR "Life Cycle Assessment" OR "life cycle analysis") AND ("emissions") AND ("livestock*" OR "dairy" OR "sheep" OR "pig" OR "poultry" OR "goat" OR "milk" OR "egg*" OR "Chicken" OR "cow" NOT "waste" OR "biofuel" OR "bioenergy")
Google Scholar	7	"LCA" "enteric fermentation" OR "enteric emissions"
	8	"LCA" "manure application" OR "manure emissions"
	10	"LCA" "crop soil emissions"
	11	"LCA" "livestock"
	18	"LCA" "wheat soil emissions"

^a Last access in March 2022.

process of article selection based on restrictive criteria.

First ("identification step"), the literature search was performed, according to the queries defined in Table 1, in Scopus, Web of Science and Google Scholar databases. Searches led to a total of 29,151 papers. When the Google search engine was used in the search, the selection of papers was stopped at page 15 of the search results (Each Google Scholar page contained approximately 10 items). Papers with research which was not fully relevant to the crop-livestock sector such as rice, plastic, biofuel, and bioenergy were excluded. Energy papers related to biogas without any relation to feed, and soil emissions were also excluded as were papers with insects, fish or feed production without any focus on livestock.

The second step involved the review of abstracts and titles, article accessibility, language, region and removal of duplicate papers. The "screening" was accomplished by using restrictive criteria ("refine results") excluding appearances before 2012 and papers which were not accessible (1175 papers). Further selection was performed based on the content of the abstract and by excluding off-topic material. Finally, 621 papers were selected as "Eligible" for full-text reading.

After the full-texts were read, the final step was to exclude papers which were not directly used in LCA application or did not focus on the key topic of "GHG emissions". This resulted in a 263 papers included in the qualitative analysis related to soil C, soil N₂O emissions, manure emissions and enteric fermentation. Of these, 31 papers dealt with soil N₂O emissions in LCA of agriculture systems and 16 were identified as describing key methods "Method identification". Direct measurements

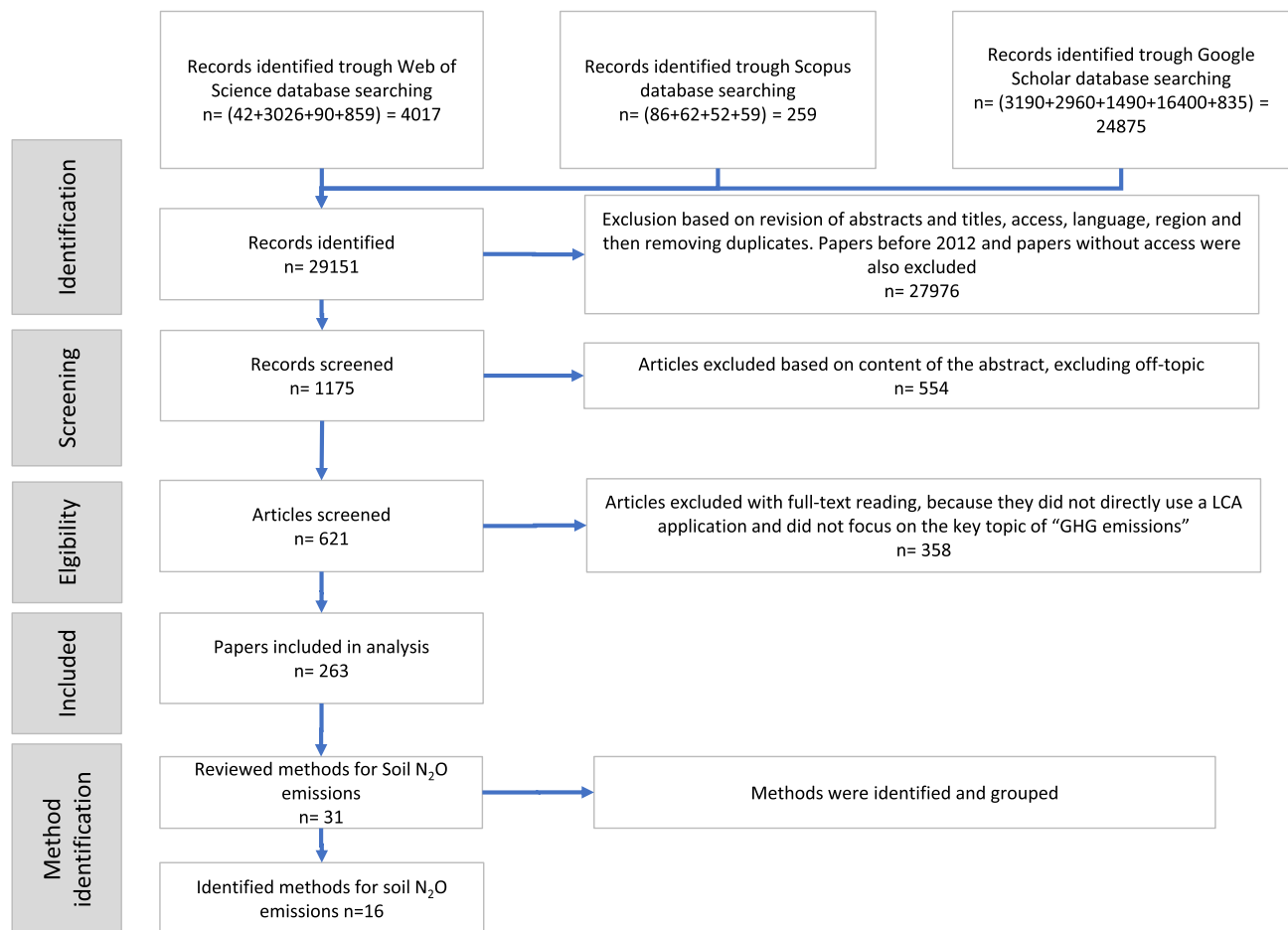


Fig. 1. Methodological steps of the literature search process.

methods have been added in this assessment even if they are not part of a LCA of crop-livestock systems, as they have been reported in major publications related to greenhouse gas emissions, such as the IPCC (De Klein et al., 2006; Hergoualc'h et al., 2019), or used in LCA of cropping systems (Goglio et al., 2018; Zaher et al., 2013).

2.3. General and specific criteria for method assessment

2.3.1. General criteria

The papers included in this review were then reviewed using both general and specific criteria to assess the LCA methods for crop-livestock systems and products. General criteria used in the harmonization of LCA methods for crop-livestock systems for GHG emissions were selected using a participatory approach based on a modified DELPHI method, extensively described by Goglio et al. (2023a). Briefly, the selection of key topics was carried out through an anonymous survey which allowed us to screen the various topics and provide a priority list on the basis of a preliminary literature review.

General criteria to assess for the LCA methods across the key identified topics were identified. This process began with a review of frameworks used to assess LCA methods. It was undertaken together with articles and publications from literature including the Food and Agriculture Organization (FAO) Livestock Environmental Assessment Programme (LEAP) reports and the Product Environmental Footprint Category Rules (PEFCR) general guidelines (FAO, 2018; Zampori and Pant, 2019), considering only publicly available sources. Next, an anonymous survey of LCA experts was carried out using Google survey. The general criteria selected through the survey were then further partially reformulated to ensure better consistency and coherence across

the key topics selected. Goglio et al. (2023b) describes the general criteria defined for the harmonization of LCA methods for agricultural systems.

2.3.2. Specific criteria identification

Following the definition of the general criteria, specific evaluation criteria were defined for each specific topic in several workshops ($n = 4$). In this paper, only the identification of criteria for soil C and soil N_2O emissions were extensively described. Soil C criteria were here presented as soil C and soil N_2O emissions are closely related (Olesen et al., 2023; Saggarr, 2010). However, LCA methods related to soil C in agricultural systems are going to be part of a separate paper. Further information on the specific criteria can be found in Goglio et al. (2023a).

The specific criteria selected for "Soil C dynamics & Soil N_2O emissions" are reported in Goglio et al. (2023b) together with their scale: adaptability to different soil types, adaptability for different land uses, and adaptability to different climates. With adaptability to different soil types, we defined the degree at which a LCA method can be applied to different soil types, e.g. peat soils, sandy mineral soils and other type of mineral soils. Instead, with adaptability to land uses, we define the level at which the LCA method can be applied to different land uses (e.g., grassland, cropland); while with the adaptability to different climates, we define the level at which a LCA method can be used in different climatic conditions (e.g., Temperate, Continental, Boreal). Finally, the accuracy was defined as the ability of the LCA methods to capture daily changes and the long-term dynamics of the soil N_2O and CO_2 emissions. With regards to this accuracy definition, it is assumed that the LCA practitioner has sufficient expertise to adopt the methodology and that observations have been carried out with a protocol.

2.4. Assessment of the LCA methods for soil N₂O emissions

The assessment of the LCA methods was carried out by providing a scoring for the general and specific criteria. The results of the assessment were discussed among the group of experts ($n = 14$) in a series of workshops ($n = 22$), then they were further reviewed by other experts external to the PATHWAYS project. The identified experts had expertise in LCA of agricultural systems and soil N₂O emission quantification. All the discussions were conducted as a community of peers among experts (Macombe et al., 2018), in line with the harmonization approach for LCA of livestock systems and products (Goglio et al., 2023a). Targeted and structured discussions were organised to solve eventual disagreement in the scoring of the LCA methods, as previously carried out (Goglio et al., 2023a; Macdiarmid et al., 2016).

3. Results

3.1. Quantitative results

Throughout the systematic review, only 31 LCA methods which assessed soil N₂O emissions in relation to agricultural systems (0.1%) were included in the final review. These LCA methods satisfied most of the general criteria adopted in this research (Fig. 2): average score > 2.4, across transparency and reproducibility, completeness, fairness and acceptance, robustness criteria (with a scale of 1–4). For these criteria, >94% of the LCA methods scored 2 or higher. In contrast, the LCA methods assessed here resulted in low applicability (on average 1.7) with 78% of the LCA methods reviewed in this study scoring 2 or lower with a scale ranging from 1 to 4 (Fig. 2). Four methods scored 3 for applicability: Brentrup et al., 2000, IPCC Tier 12,006, IPCC Tier 12,019 methodology and Sozanska et al. (2002) (see section 3.2. for details) (Brentrup et al., 2000; De Klein et al., 2006; Hergoualc'h et al., 2019; Sozanska et al., 2002).

Two of the specific criteria were satisfactorily fulfilled (>2.4 on average with a 1–3 scale): adaptability to soil types and land uses. For this set of criteria, all the methods achieved a score of 2 or higher. However, on average, the LCA methods reviewed scored poorly for adapting to different climates and had reasonably low accuracy (<2.2 with a 1–3 scale) (Fig. 2). Only four methods scored 3 for adaptability to different climates (IPCC Tier 1 (2006) and IPCC Tier 1 (2019), DNDC and direct measurements, for details see section 3.2) (De Klein et al., 2006; Li et al., 1996); while 78% scored 2 or less with a range of 1–3. For accuracy, as defined in Goglio et al. (2023b), only direct measurements scored 4, DNDC and DAYCENT scored 3; while most of the methods

(83%) scored 2 or lower with a 1–3 scale (Fig. 2).

3.2. Description and scoring of key identified methodologies

In this section, a brief description of each identified LCA methodology is presented. The different methods are discussed following a tiered approach as proposed by the IPCC. Three tiers have been proposed following the FAO LEAP framework (FAO, 2020): Simple empirical models and emission factors (Tier 1); Basic process or complex empirical models (Tier 2); Complex process-based models and direct measurements (Tier 3) (FAO, 2020). Direct observations generally fall under the scope of Tier 3 methods, while simple emission factors specific to large geographical areas are Tier 1. The scoring of each method is presented in Table 2.

3.2.1. Simple empirical models and emission factors (Tier 1)

Brentrup – This method relies on Bouwman, 1995 (Bouwman, 1995), and simply multiplies the total N applied by 1.25% to estimate N₂O emissions for both mineral and organic sources, without distinguishing the source type (Brentrup et al., 2000). This is in contrast with the current IPCC guidelines which uses other values for soil N₂O emissions from both mineral and organic sources (Hergoualc'h et al., 2019). The Brentrup method scored on average 2.6 with the lowest value for the accuracy criterion (1, Table 2).

EMEP/EEA – This method (Amon et al., 2019) was primarily developed for use by national inventory compilers. The authors state that due to its empirical nature, and lack of consideration for site specific soil conditions its use in modelling situations may not be appropriate. It estimates emissions due to manure application and grazing distinguishing between manure types from different livestock categories (Amon et al., 2019). The method scored 2.2 on average among the criteria with lowest values for accuracy, similar to the Brentrup method (1, Table 2).

GLEAM – The updated guidelines (FAO, 2022) for version 3.0 of the GLEAM model provide further guidance on the GLEAM model structure. The equations for N₂O are the same or adapted from the IPCC 2006 or 2019 equations. Thus, they provide a differentiation between crops (based on N biomass content and biological N fixation factors), fertilizer type (ie. manure vs synthetic fertilizer) soil and climatic factors affecting soil N₂O emissions (FAO, 2022). Manure application factors vary slightly from IPCC. Further indirect emissions from leaching losses are estimated based on a nitrogen balance method, different from the IPCC methodology (De Klein et al., 2006; FAO, 2022; Hergoualc'h et al., 2019). GLEAM scored 2.2 and again had the lowest score for the

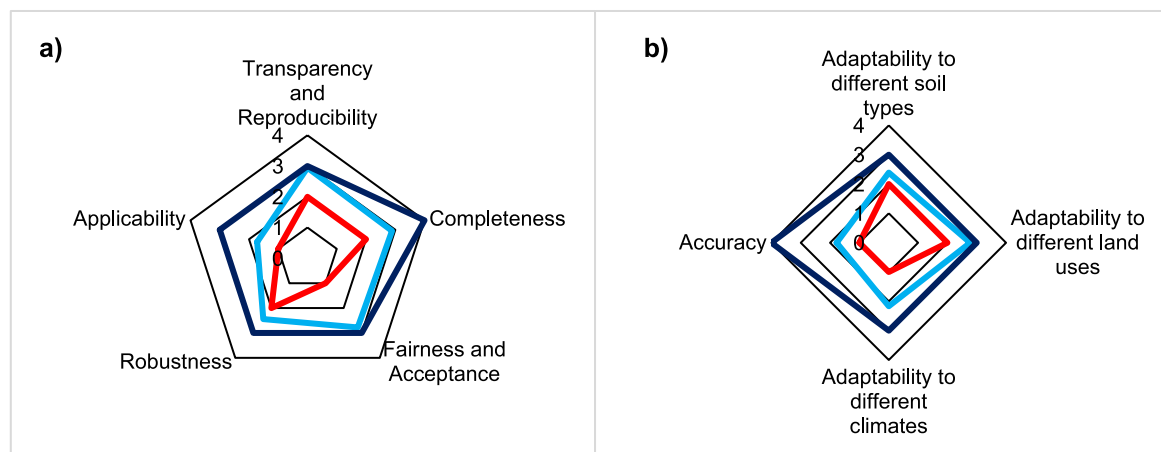


Fig. 2. Results from the scoring of the five generic criteria (a) and four specific criteria (b) for LCA methods used to assess soil N₂O emissions. Dark blue colour indicates the maximum value obtained, red colour the minimum value and light blue colour the average value. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 2

Details of the described methods, including the general criteria and specific criteria scoring. Trans.: Transparency; Rep.: Reproducibility; Com.: Completeness; Fair.: Fairness; Accept.: Acceptance; Robust.: Robustness; App.: Applicability; Adapt.: Adaptability; Acc.: Accuracy.

Group	LCA publication ^a	Method name	Method publication ^{b, c and d}	General criteria					Specific criteria				Mean Score
				Trans. and Rep.	Com.	Fair. and Accept.	Robust.	App.	Adapt. to different soil types	Adapt. to different land uses	Adapt. to different climates	Acc.	
Simple empirical models and emission factors (Tier 1)	Schmidt Rivera et al. (2017)	Brentrup 2000	Brentrup et al. (2000)	3	3	3	3	3	2	3	2	1	2.6
	Berton et al. (2016)	EEA 2013 (2019 reviewed)	Amon et al. (2019)	3	3	3	2	2	2	2	2	1	2.2
	MacLeod et al. (2018)	GLEAM model. IPCC 2006 tier 2 combined with LCA analysis	FAO (2017)	3	3	3	2	2	2	2	2	1	2.2
	Cederberg et al. (2013)	IPCC (2006) Tier 1	Lasco et al. (2006)	3	3	3	2	3	3	3	3	1	2.7
	Jeswani et al. (2018)	IPCC (2006) Tier 2	De Klein et al. (2006)	3	3	3	2	2	2	3	2	1	2.3
	González-Quintero et al. (2021)	IPCC 2006 (2019 refinement) Tier 1	Hergoualc'h et al. (2019)	3	3	3	2	3	3	3	3	1	2.7
	Bonesmo et al. (2013)	Bonesmo et al., 2012	Bonesmo et al. (2012)	3	2	2	2	2	2	3	2	2	2.2
	Alemu et al. (2017)	Holos	Little et al. (2008)	3	3	3	2	1	2	2	2	2	2.2
Basic process or complex empirical models (Tier 2)	Avadí (2020)	INDIGO-N combined with IPCC Tier 1 emission factors	Bockstaller et al. (2022)	3	3	3	2	1	2	2	2	2	2.2
	de Vries et al. (2015)	INITIATOR	de Vries et al. (2003)	3	2	2	3	1	3	3	1	2	2.2
	Bonesmo et al. (2013)	Sozanska et al. (2002)	Sozanska et al. (2002)	2	2	1	2	3	2	3	2	2	2.1
	Carauta et al. (2021)	CANDY	Franko et al. (1995)	3	4	3	2	2	2	3	2	1	2.4
	Cederberg et al. (2013)	CERES-EGC	Goglio et al. (2013)	3	3	3	3	1	3	3	2	2	2.6
Complex process based models and direct measurements (Tier 3)	Cederberg et al. (2013)	DAYCENT	Del Grosso et al. (2005)	3	3	3	3	1	3	3	2	3	2.7
	Grossi et al. (2021)	DNDC	Li et al. (1994)	3	3	3	3	1	3	3	3	3	2.8
	Rotz (2018)	ECOSYS	Metivier et al. (2009)	3	3	3	3	1	2	3	2	2	2.4
	^c	Direct observations	^{c and d}	3	3	3	3	1	3	3	3	4	2.9

^a Publications where the method has been used in the LCA of agricultural systems.

^b Key publication where the method has been extensively described.

^c The LCA method assessment was based on a LCA of cropping systems using direct observations (Goglio et al., 2018).

^d Several research studies discussed direct N₂O observations techniques (Glenn et al., 2012; Pattey et al., 2007; Rochette et al., 2018; Rochette and Eriksen-Hamel, 2008; Venterea et al., 2020).

accuracy (1, Table 2).

IPCC (2006) Tier 1 – The Tier 1 method utilizes simple emission factors to estimate direct and indirect N₂O emissions, with little differentiation between sources of N (including crop types) or climatic factors (De Klein et al., 2006). With regards to fertilizer management, it distinguishes between mineral and organic fertilizer. IPCC Tier 1 (2006) method scored 2.7 with the lowest value for the accuracy criterion (1, Table 2). IPCC Tier 1 (2019) adopted a broader differentiation between N sources and climatic factors than the IPCC Tier 1 (2006) (De Klein et al., 2006; Hergoualc'h et al., 2019). As shown in Table 2, both IPCC Tier 1 (2019) and IPCC Tier 1 (2006) had the same scores for all the assessed criteria with the smallest value for accuracy (1).

IPCC (2006, 2019) Tier 2 – The Tier 2 method goes beyond Tier 1 through additional differentiation of emissions from synthetic nitrogen types, climatic conditions, and can include country or regional specific Emissions Factors (EFs) (De Klein et al., 2006; Hergoualc'h et al., 2019; Liang et al., 2020). IPCC Tier 2 had an average score of 2.3, considering both versions together (2006, 2019) and low accuracy values (1, Table 2) (De Klein et al., 2006; Hergoualc'h et al., 2019). Among the methods using the IPCC Tier 2 framework, there is also the most recent version of the Swiss Agricultural LCA (SALCA) method (Nemecek et al., 2023). Regional framework utilising emission factors were also proposed by Cayuela et al. (2017) for Mediterranean conditions.

3.2.2. Basic process or complex empirical models (Tier 2)

Bonesmo – The Bonesmo et al., 2012's paper utilised IPCC 2006 as the basis for N₂O estimation (Bonesmo et al., 2013), but further refined this into a seasonal (quarterly) estimation, utilising Sozanska et al. (2002)'s data; distinguishing only pasture manure from all other soil N inputs (e.g. organic, synthetic fertilizer, residues). This improved the estimation as it took into account the effects of soil water and temperature on direct N₂O emissions. Indirect N₂O emissions due to nitrate leaching were estimated by multiplying 30% by the total N inputs in kg ha⁻¹ and the emission factor derived from IPCC 2006 (De Klein et al., 2006). The Bonesmo method averaged a 2.2 score with the largest value for Transparency and Adaptability to different land uses (3, Table 2).

Holos – Holos is a Canadian model (Little et al., 2008) which has been further enhanced beyond its original scope to use country and regional specific EFs based upon Rochette et al. (2018) and Liang et al. (2020). The model allows for differentiation across crops, soil types, texture and climate by using regional annual precipitation to potential evapotranspiration ratios, allowing for improved emission estimation for Canada. With regards to fertilizer management, it distinguishes between organic and mineral fertilizer (Liang et al., 2020; Rochette et al., 2018). The model had on average a 2.2 score with the lowest applicability value (1, Table 2). Thus, the application of the current Holos version is limited outside Canadian conditions.

INDIGO-N – The Indigo-N v3 model (Bockstaller et al., 2022) provided a new semi-mechanistic approach to estimate nitrate leaching as a source of indirect N₂O losses. While this novel approach tried to account for other losses and agronomic interventions, the direct N₂O emissions, ammonia volatilisation and EFs continued to rely on IPCC Tier 1 or 2 data or equations. These distinguish between different crops (e.g. legumes vs cereals) and fertilizer type (e.g. mineral vs organic). INDIGO N scored as Holos (2.2) with lowest value for applicability (1, Table 2).

INITIATOR – The model INITIATOR (Integrated NITrogen terrestrial systems was partitioned to surface water Impact Assessment Tool On a Regional scale) estimates N₂O emissions through a series of empirical equations estimating nitrification and denitrification (de Vries et al., 2003), and was utilised to assess the impacts of Dutch agriculture on N₂O emissions. INITIATOR distinguish between organic, mineral fertilizer and biological N fixation (de Vries et al., 2003). INITIATOR averaged 2.2 across the assessment criteria and the lowest values were obtained for applicability and adaptability to different climates (1, Table 2).

Sozanska et al. (2002) – This method uses a single regression

equation developed for soils in the UK (Sozanska et al., 2002). Whilst this equation was useful as a tool for use with Geographical Information System (GIS) data, the authors acknowledge the uncertainty is high due to the application of short term measured data to arrive at annual estimates. Further, Sozanska et al. (2002)'s method pools all the type of N input together in the calculation. Sozanska et al. (2002)'s method resulted in average score of 2.1 with lowest scores for Fairness and Acceptance (1, Table 2).

3.2.3. Complex process-based models and direct measurements (Tier 3)

CANDY – Utilising the CANDY (CARbon Nitrogen DYnamics) model, a daily time step processing of agricultural soils in 10 cm increments down to 2 m is used for estimating C and N dynamics. These were linked to water and crop sub-models. Two nitrogen forms (nitrate and ammonium) are considered, and processed as nitrogen inputs, conversions and losses, through e.g. nitrification of ammonium to nitrate and denitrification leading to gaseous losses. These are estimated through equations, and related to the crop model for uptake, soil temperature and water (Franko et al., 1995). CANDY averaged 2.4 across the assessment criteria with the highest score for completeness (4, Table 2) and the lowest for accuracy (1, Table 2).

CERES-EGC – The model comprises components to simulate the cycles of water, carbon and nitrogen in agro-ecosystems (Lehuger et al., 2009), and was itself adapted from the semi-empirical NOE model (Goglio et al., 2013). Operating on a daily time step, the agro-ecological model simulates crop development and soil interactions with nitrogen, carbon and water. The N₂O emissions are estimated from 15 parameters, 4 of which require site-specific measurements and the remaining 11 are derived from literature reviews. CERES-EGC had a mean score of 2.6 and had the lowest score for applicability (1, Table 2).

DAYCENT – DayCent was developed as a daily time step version of the CENTURY model (Parton et al., 1994). Daycent is a full agroecological system model, simulating fluxes of C and N between the atmosphere, soil and plant system using a series of empirical equations (Del Grosso et al., 2005; Rotz, 2018). In contrast to e.g. IPCC assumptions of all emissions occurring within the same year of application, DAYCENT includes carry over effects of nitrogen between years and crops. Due to the complex nature of the model, input data goes far beyond simple empirically based calculations, as per other Tier 3 models. DAYCENT had an 2.7 average score and the lowest value for applicability (1, Table 2).

DNDC – DNDC is a mechanistic agroecosystem model (Li et al., 1994), which has been widely used to examine the potential impacts of agricultural management, climate and soils on N₂O emissions, crop yields and other N and C gases. The model includes detailed processes for estimating decomposition, nitrification, denitrification, urea hydrolysis, fermentation and methanogenesis. The model has been shown to perform well in comparison to specific field trials but requires extensive parameterisation to operate under varying soil or climatic conditions (Ehrhardt et al., 2018). DNDC had 2.8 average score and a low applicability value (1, Table 2). Similar to many complex process-based models, DNDC requires extensive expertise and a comprehensive user manual (Gillespy et al., 2014; Goglio et al., 2018).

ECOSYS – The ECOSYS model allows ecosystem behaviour to be represented in a fully integrated manner under user-defined conditions of soil, climate and management (Wegegedara et al., 2020a, 2020b). Of particular relevance to soil N₂O, the soil organic matter microbial populations are represented through five complexes to characterize soil dynamics under varying conditions at an hourly timestep (Metivier et al., 2009). ECOSYS resulted in an average value of 2.4 and the lowest value for applicability (1, Table 2).

Direct measurements – Direct measurements have been employed at this stage only for LCA of cropping systems (Goglio et al., 2018), as they are a challenge to be carried out (Laville et al., 2011; Olesen et al., 2023). Direct measurements methods for soil N₂O emissions include chamber, eddy covariance and flux gradient measurements (Glenn et al.,

2012; Pattey et al., 2007; Rochette and Eriksen-Hamel, 2008). These methods averaged 2.9 scores despite a low applicability (1) (Table 2).

4. Discussion

4.1. Identified key methodological issues

The LCA methods assessed in the present review of soil N₂O emissions were transparent and easy to reproduce, complete, robust, fair and accepted. However, a large proportion have low applicability (50%) and accuracy (39%), whilst the majority of the methods (78%) had low adaptability to different climates. The five methods with very high applicability (3) were Brentrup et al. (2000); Sozanska et al. (2002); IPCC Tier 1 methodology 2006 and 2019 (De Klein et al., 2006; Hergoualc'h et al., 2019) and direct measurements. Brentrup et al. (2000), IPCC Tier 1 (2006) and IPCC Tier 1 (2019) were probably the more general methods which could be applied for every condition, soil climate, soil type and soil management, though they do not include the effects of nitrification inhibitors, slow release fertilizer, timing of fertilizer or manure applications, and type of spreading and distribution for manure and slurry (De Klein et al., 2006; Hergoualc'h et al., 2019). On the other hand, Sozanska et al., 2002's method was generally more accurate, however it required very specific data such as water filled spore space (WFPS) measured directly from the field (Bastos et al., 2021; Rochette et al., 2018; Venterea et al., 2011), which is often not available to the LCA practitioner. However, together with WFPS, during the season, different conditions have to be verified for the soil N₂O emissions to occur such as high nitrate availability and high temperature. While assessing accuracy, these aspects were taken into account as these soil parameters are subject to daily changes which affect emissions (Bastos et al., 2021; Sagggar, 2010).

The LCA based on DNDC, DAYCENT or direct measurements scored 3 in accuracy, as DNDC and DAYCENT accounts for soil moisture and temperature, soil C and N dynamics and crop N uptake at a daily time step (Brilli et al., 2017; Del Grosso et al., 2020; Ehrhardt et al., 2018; Giltrap et al., 2020; Li et al., 1996). Instead, while the direct measurements performed very well for all the criteria except applicability, their use in a LCA is limited by the difficulties in carrying out the monitoring both from a technical and financial stand point (Dorich et al., 2020; Giltrap et al., 2020; Laville et al., 2011; Olesen et al., 2023), which make these data hardly available to the LCA practitioner. Further, it may be challenging to allocate the soil N₂O emissions related to a particular crop management since the impacts can carry over to the period when the following crop is grown as discussed previously for crop residues (Goglio et al., 2017; Olesen et al., 2023). As for LCA method for soil C in agricultural LCA (Goglio et al., 2015), a compromise also has to be found between accuracy and applicability of the LCA method for soil N₂O. This compromise is dependent on data availability, LCA practitioner expertise in coherence with the LCA objectives (Goglio et al., 2015). In some cases, simpler methods for estimating N₂O emissions may not include some field management practices (e.g. impacts of urease and nitrification inhibitors, split fertilizer application, or N credit from legumes).

Most of the methods assessed fit into two categories: IPCC Tier 1 methodology and subsequent updates or agroecosystem models, such as DNDC and DAYCENT (Del Grosso et al., 2005; Gilhespy et al., 2014; Goglio et al., 2018). Different from soil C, empirical or regression models are currently not available, except those proposed by Sozanska et al. (2002), however this latter method was developed for the Atlantic climate and depends on data which were rarely available to the common LCA practitioner. Other methods were developed to estimate soil N₂O emissions based on parameters such as rainfall, soil characteristics and N management in Canadian conditions (Rochette et al., 2018), which could be used in the LCA of livestock systems. This type of data is more commonly used and collected in agricultural LCA (Goglio et al., 2018; Styles et al., 2014).

For large scale site-dependent assessment, either attributional,

consequential or anticipatory LCA, using the IPCC Tier 1 methodology (using the 2019 updated and disaggregated by climate type values) is a sensible compromise between the accuracy and the applicability of the LCA method. For countries where IPCC Tier 2 emission factors are available, the latter methodology should be preferred as it is more accurate in capturing local conditions (Cayuela et al., 2017; Hergoualc'h et al., 2019), however it might be challenging to collect data with enough quality to use IPCC emission factors in both cropping, grassland, agricultural and livestock systems. Indeed for the latter, a higher level of system complexity is achieved as feed (e.g. cropping systems) and fodder (e.g. grassland systems) producing systems need to be assessed (Rotz, 2018).

Improving soil N₂O emissions quantification is important as N₂O impacts global warming, but can also contribute and affect other impact categories in combination with other important emissions such as ammonia and NO_x. These impact categories can include biodiversity loss, stratospheric ozone-depletion, eutrophication (which is related to water quality degradation) and acidification (which is affected by air pollution). Indeed, the effects on climate change (i.e. global warming) can alter indirectly several of the ecosystem services provided by the cropping and grassland systems, including water availability (Brady and Weil, 2002; Hergoualc'h et al., 2019; Pörtner et al., 2022)."

4.2. Research need, future studies

Soil N₂O emissions derived from both soil tillage management and fertilizer management including manure, sludge or slurry spreading are often dependent on the interaction between soil characteristics, rainfall and temperature (Bastos et al., 2021; Sagggar, 2010). While the pattern of soil N₂O emissions related to the mineral and organic fertilizer application is rather well known (Dorich et al., 2020; Giltrap et al., 2020; Taki et al., 2019), the interaction with residues from legume crops is less clear (Chirinda et al., 2010; Olesen et al., 2023). The latter together with grassland and cover crop management are particularly important in livestock systems (Parajuli et al., 2018).

Within the LCA context, there is a general need to ensure that crop and grassland management issues are considered and accurately accounted for in the LCA of agricultural systems, as previously discussed for organic agriculture (van der Werf et al., 2020). This is in view of pollution shifts and trade-off across impact categories, related to the N biogeochemical cycle (Brady and Weil, 2002; Styles et al., 2015; Zhou et al., 2023). With regards to soil N₂O emissions, only the DNDC was able to fully capture soil N₂O drivers, crop management, soil and climate characteristics (Brilli et al., 2017; Del Grosso et al., 2020; Li et al., 1996). However its applicability is low due to a large data requirement and a need for modeller expertise, as previously discussed for soil C (Giltrap et al., 2020).

Emissions from crop residues can happen during the growing season of the following crop, when high biomass is degraded and high water content is available (Olesen et al., 2023). This can cause allocation issues among crops in agricultural LCA, thus a system approach might be necessary, as previously discussed (Goglio et al., 2017; Sieverding et al., 2020). However, even with a system approach in agricultural LCA, the environmental impacts from a specific crop within a specific cropping system should be allocated, if the latter is used as feed in a livestock system (Rotz, 2018).

Therefore, soil N₂O emission methods need to be developed to capture crop management effects on soil N₂O emissions, similar to DNDC, without limitations from data requirements. An option is the method by Sozanska et al. (2002), even though it did not capture many aspects of crop management such as tillage, residue management, type of fertilizer, rainfall patterns (Bastos et al., 2021; Sagggar, 2010). Alternative methods could be based on statistical methods used for gap-filling (e.g., random forest or neural networks), which use a series of covariates factors to estimate soil N₂O emissions (Dorich et al., 2020). On the other hand, regression models, similar to those developed in Rochette et al. (2018),

which capture more aspects of crop and grassland management, such as crop type, some fertilizer management, soil and climate characteristics, should be developed for European conditions. These could be a compromise between accuracy of the model and applicability of the methods for LCA of agricultural systems.

Further, soil N₂O emission play an important contribution to the overall global GHG budget (0.75%) (IPCC, 2022). Thus, efforts should be made to improve the estimates by increasing the available data across Europe and by comparing agroecosystem model performance of the key models identified here (ie. DNDC and DAYCENT) to better improve the overall GHG estimates. Previously a metanalysis was carried out in Canadian conditions (Liang et al., 2020) and a similar analysis could be performed in Europe by assessing all scientific evidence related to the impact on soil N₂O emissions due to crop/grassland management practices, soil and climate conditions (Liang et al., 2020; Rochette et al., 2018). This research would contribute to the overall improvement of the IPCC GHG emission calculations (Hergoualc'h et al., 2019).

4.3. LCA recommendations

Our review of soil N₂O emissions leads to the recommendation that it is preferable to use the DNDC model after calibration and validation or use of direct field measurements, taking in consideration system effects (Goglio et al., 2017). However, when the necessary data to run the DNDC model or field observations are lacking, the use of IPCC tier 2 methodology (2019) with disaggregated EFs should be prioritized where available, otherwise IPCC Tier 1 methodology following the 2019 guidelines should be used (Hergoualc'h et al., 2019). When using 2019 IPCC Tier 2 or IPCC Tier 1 methodology to assess soil N₂O emissions, the methodological limitations should be made clear by the LCA practitioner (Hergoualc'h et al., 2019). Independently from the methodological choice carried out, it is key to provide arguments for this choice and describe its potential limitations, in agreement with the ISO standards (ISO, 2006a, 2006b, 2013).

Especially for large site-dependent or site-generic studies (Potting and Hauschild, 2006), a preliminary assessment could still be carried out using simpler methods such as IPCC Tier 1 (2019) (Hergoualc'h et al., 2019), as data might not be available for the LCA practitioner. This should be complemented with a clear description of limitations of the methodology as suggested by the ISO standards (ISO, 2006b, 2006a, 2013) and discussed in the present research. Further, conclusions about these LCAs should be taken with caution as they poorly reflect local conditions and the effect of crop and grassland management. Indeed, local conditions are key in soil N₂O emissions as these are subject to a large spatial variability (Del Grosso et al., 2020).

This harmonization of LCA methods has been carried out with a participatory approach involving several experts ($n = 14$) which have been involved at different stage of the process, following the criteria previously drawn (Goglio et al., 2023a). This approach allowed for the development of scoring criteria to assess LCA methods through workshops and targeted discussion, as previously discussed for social LCA (Macombe et al., 2018). This harmonization approach allowed for the discussion of state of the art practices and the identification of future development priorities and future needs in a coherent manner for several topics including soil C, manure emissions, enteric fermentation, biodiversity, animal welfare, nutrition aspects and circular economy (Goglio et al., 2023a).

5. Conclusion

In this research, an attempt to harmonize LCA methods for soil N₂O emissions in agricultural systems was carried out by comparing methods, showing their limitations and making recommendation on their use. It was observed that a high level of accuracy corresponded to a low level of applicability and vice versa. Thus, the choice of the methodology in relation to the LCA objectives is particularly critical to enable

high quality LCA assessments.

Following the analysis of the available literature, a series of recommendations was proposed. A general recommendation for soil N₂O from agricultural systems is that the choice of LCA methods should be based on the LCA objectives, data availability and expertise of the LCA practitioner. For all soil N₂O assessments, more complex methods are available but have greater data requirements. IPCC Tier 1 methodology has been employed in most of the assessments analysed here. Independently of the method used, method limitations should be discussed in the LCA of agricultural systems in view of the assessment objectives, data requirements and expertise available. Further, within the IPCC, there is a urgent need to develop higher Tier methods to improve the overall assessment of soil N₂O emissions. This could be achieved to a broader testing and comparison of field observations with the models identified here to improve the IPCC methodology. This research should be combined with a metanalysis of all the drivers affecting soil N₂O emissions in cropland/grassland systems.

Future development of LCA methodology is necessary to improve LCA of agricultural systems. For soil N₂O emission, effort should be placed towards developing a basic process model (i.e. soil N₂O regression models) which optimises applicability and accuracy. The LCA method development related to soil N₂O emissions must be synchronous with improvements of quantification methods and the assessment of different agricultural management.

Funding sources

This research has been developed within the PATHWAYS project, funded by the European Union's Horizon 2020 Research and Innovation Programme European Union through Horizon 2020 Research and Innovation Programme under grant agreement No 101000395.

CRedit authorship contribution statement

Pietro Goglio: Writing – review & editing, Writing – original draft, Visualization, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation. **Simon Moakes:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation. **Marie Trydeman Knudsen:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization. **Klara Van Mierlo:** Methodology, Investigation, Formal analysis, Data curation. **Nina Adams:** Methodology, Investigation, Formal analysis, Data curation. **Fossey Maxime:** Methodology, Investigation, Formal analysis, Data curation. **Alberto Maresca:** Writing – review & editing, Methodology. **Manuel Romero-Huelva:** Methodology, Investigation, Formal analysis, Data curation. **Muhammad Ahmed Waqas:** Methodology, Investigation, Formal analysis, Data curation. **Laurence G. Smith:** Writing – review & editing, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization. **Giampiero Grossi:** Writing – review & editing, Supervision. **Ward Smith:** Writing – review & editing, Supervision. **Camillo De Camillis:** Writing – review & editing, Supervision. **Thomas Nemecek:** Writing – review & editing, Supervision. **Francesco Tei:** Writing – review & editing, Supervision. **Frank Willem Oudshoorn:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Pietro Goglio reports financial support was provided by European Commission Horizon2020 programme. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data related to the LCA harmonization will be made available upon request.

Criteria for LCA assessment of soil N₂O emissions in agricultural systems (Original data) (Zenodo)

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