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Abstract

N biogeochemical flows and associated N losses exceed currently planetary boundaries and represent a major threat for sustainability. Measuring N losses is a resource-intensive endeavour, and not suitable for *ex-ante* assessments, thus modelling is a common approach for estimating N losses associated with agricultural scenarios (systems, practices, situations). The aim of this study is to review some of the N models commonly used for estimating direct field emissions of agricultural systems, and to assess their suitability to agricultural systems featuring organo-mineral or organic fertilisation, non-arable crops, or happening under tropical and sub-tropical conditions.

Simple N models were chosen based on their frequent use in LCA, following a literature review, including ecoinvent v3, Indigo-N v1/v2, AGRIBALYSE v1.2/v1.3, and the Mineral fertiliser equivalents (MFE) calculator. Model sets were contrasted, among them and with the dynamic crop model STICS, regarding their consideration of the biophysical processes determining N losses to the environment from agriculture, namely plant uptake, nitrification, denitrification, NH_3 volatilisation, NO_3 leaching, erosion and run-off, and N_2O emission to air; using four reference agricultural datasets. Models' consideration of management drivers such as crop rotations and the allocation of fertilisers and emissions among crops in a crop rotation, over-fertilisation and fertilisation technique, were also contrasted, as well as their management of the mineralisation of soil organic matter and organic fertilisers, and of drainage regimes. We highlighted the reasons for the differing model outputs.

Among these models, Indigo-N is the most data intensive, and ecoinvent the least. For the four agricultural datasets, the ecoinvent model predicted significantly lower values for NH_3 than the AGRIBALYSE and the STICS models. For N_2O, no significant differences were found among models. For NO_3, the ecoinvent and AGRIBALYSE models predicted significantly higher emissions than the STICS model, regardless of the fertilisation regime. For both emissions, values of Indigo-N were close to those of the STICS model. By analysing the reasons for such differences, and the underlying factors considered by models, a list of recommendations was produced regarding more accurate ways to model N losses (by including, for instance, the main drivers regulating emissions).

Keywords: agriculture, fertilisation, field emissions, nitrogen, organic, tropical

1 Introduction

1.1 Nitrogen modelling in agricultural LCA

Nitrogen is the main limiting factor for terrestrial and aquatic primary production, yet anthropogenic activities have altered the natural N cycle by massively increasing the flow of reactive nitrogen in the biosphere. This biogeochemical flow, as well as the global phosphorus flow and damage to genetic biodiversity, are considered to have exceeded the planetary boundaries (Steffen et al., 2015), with agriculture as a major contributor to the excess (Campbell et al., 2017). The production and use of agricultural fertilisers, together with symbiotic fixations due to human activities, represent an important part of those inputs to the environment. Those are sources of losses to the different environmental compartments, causing a series of impacts, the so called "nitrogen cascade" (Fowler et al., 2013; Galloway et al., 2003). This includes global (e.g. climate change) and local impacts (e.g. aquatic eutrophication, soil degradation). Understanding, quantifying and modelling these losses is thus an increasingly relevant research topic (Gao and Guo, 2014; Oenema et al., 2012; Yang et al., 2017). N losses, conditioned by both pedoclimatic conditions and agricultural strategies (e.g. rotations, fertilisation), predominantly take the form of ammonia (NH_3) volatilisation, nitrate (NO_3) leaching, nitrification-driven nitric oxide (NOx) emission to air and denitrification-driven nitrous oxide (NOx and N_2O) emissions to air (EMEP/EEA, 2016). Fertilisation strategies play a key role in N efficiency in agriculture, through unbalanced amounts exceeding crop requirements, time lag between fertilisation and crop uptake, and lack of emission mitigation management for some fertilising strategies, are leading yet manageable drivers of N losses (Padilla et al., 2018). Management of crop cover through rotation, catch crops, or intercropping to insure sufficient N uptake during drainage periods (e.g. winter in Europe, rainy seasons in the tropics) is another major driver (Abdalla et al., 2019).

Life Cycle Assessment (LCA) is widely used to estimate the environmental impacts of agricultural activities. Such assessment is based on life cycle inventories (i.e. resource consumption and emissions associated with a production system) (ISO, 2006), which include direct field emissions associated with fertilisation. Mineralisation, drainage, plant uptake, nitrification and denitrification, and volatilisation should be considered to estimate all N losses in agricultural LCA. Consideration of symbiotic fixation would be a plus, but seldom included. The most common approaches/concepts used to model these mechanisms are listed in the Supplementary Material (Table S1).

Measuring N losses is a resource-intensive endeavour, and not suitable for *ex-ante* assessments, thus modelling is a common approach for estimating N losses associated with agricultural scenarios (systems, practices, situations). Researchers in agricultural subjects use different types of models for estimating N losses according to their scientific questions, their level of familiarity with available models and agricultural systems studied, and their resource constraints (e.g. time, data). For instance, the modelling continuum relevant in LCA context in France is presented in Fig. 1. For example Brilli et al. (2017) reviewed very complex models, the International Soil Modeling Consortium website (https://soil-modeling.org/resources-links/model-portal) described a wide variety of agroecosystem models, and Jones et al. (2017) delivered a synthesis of agricultural systems modelling and modelling comparison/improvement initiatives.

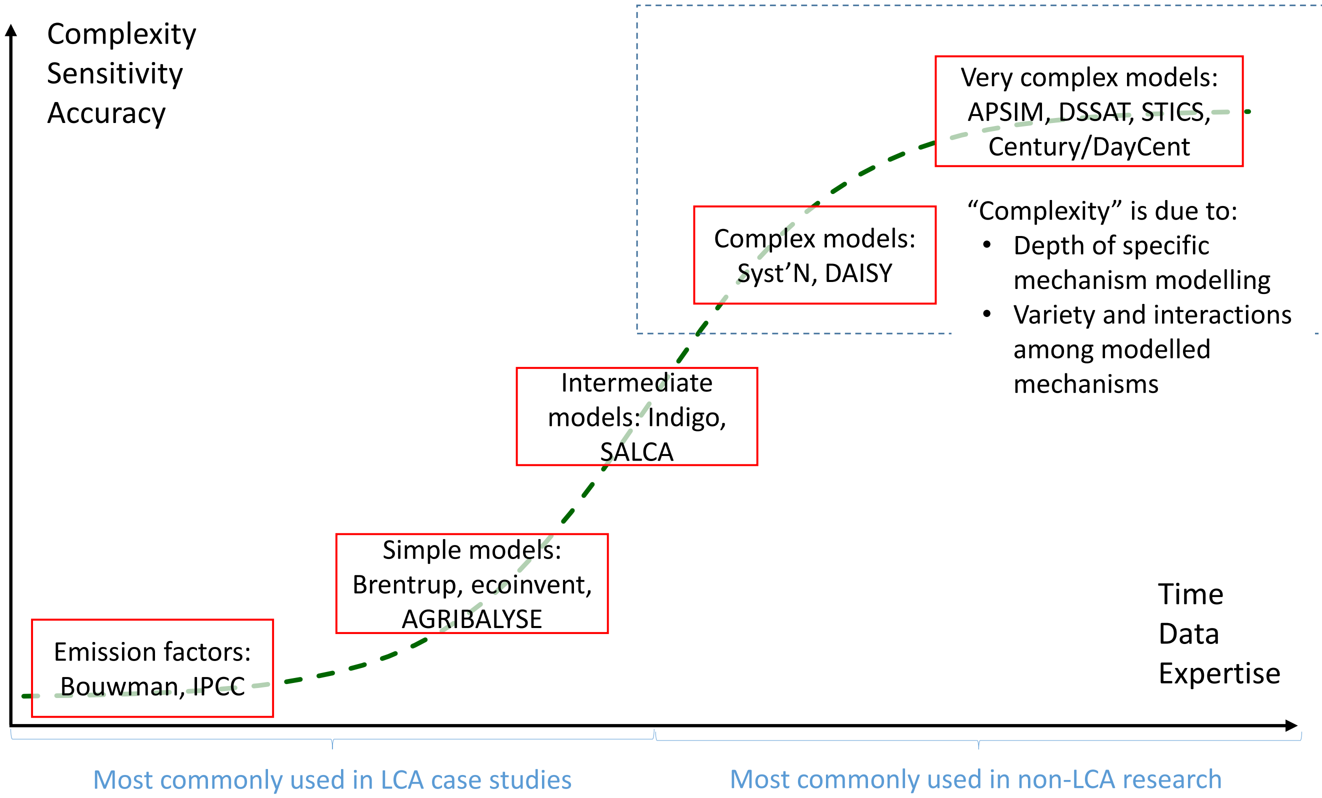

Fig. 1. Modelling continuum for estimation of N emissions in the French LCA context

At both ends of the modelling continuum are "simple" models —i.e. empirical equations with or without parameters, usually based on regressions on emissions datasets (Brentrup et al., 2000; Koch and Salou, 2016; Nemecek and Schnetzer, 2012)— and "complex" simulation models —i.e. functional or mechanistic and dynamic biogeochemical/crop models (Addiscott and Wagenet, 1985; Brilli et al., 2017; Manzoni and Porporato, 2009)—. Another key dichotomy used to classify models is their mechanistic (generic) or functional (basic parameters for default conditions, adjusted by factors to other conditions) nature, where a trend towards the latter has been observed in the last decades (Cannavo et al., 2008). Other authors suggest that a mechanistic representation of biophysical processes should lead to a reduced number of analytical generalizable models, as opposite to a large number of situation-specific complex models (Manzoni and Porporato, 2009). It has also been noted that the mathematical features of models across the modelling continuum are more linked with models' fields of application than to their intended spatial and temporal scales of application (Cannavo et al., 2008; Manzoni and Porporato, 2009). A discussion on N models classification criteria, as well as on keywords associated with N models definition and classification, is presented in the Supplementary Material.

Output from mainly simple models requiring few and available input variables can be used to calculate environmental indicators (Buczko and Kuchenbuch, 2010). Such models are designed as "operational" in Bockstaller et al. (2015). Among them, pre-calculated emission factors (EF) for the different N emissions are used, especially by environmental researchers, but these factors are often generic and may not accurately represent the studied situation. EF are usually derived from simple, generic empiric models such as those proposed by the IPCC Guideline for National Greenhouse Gas Inventories (IPCC, 2006) and its 2019 update (Hergoualc'h et al., 2019). Nitrogen balances, among the most used nitrogen indicators (Bockstaller et al., 2015; Rasmussen et al., 2017) whose use is also recommended by the FAO and the OECD, are also suitable to predict

N field emissions, yet they have been suggested to be poor predictors of nitrate leaching risk, unless considered at multi-annual temporal scales (Bockstaller et al., 2009). N-balances are computed at different levels of aggregation (i.e. from the farm to the continent), based on empiric equations often calibrated for specific conditions (Roy et al., 2003).

When N direct field emissions are the focus of research, LCA practitioners tend to use complex models (*e.g.* soil-plant dynamic models) that provide detailed information and help interpret LCA results. Yet, such models require relatively high time, data and knowledge, and thus are not widely used for agricultural LCA, but instead, LCA practitioners typically use the most accessible models, in terms of data demand and ease of use, such as those included in pre-defined model sets associated with databases like ecoinvent (Frischknecht et al., 2005), the World Food LCA Database (Nemecek et al., 2014), the Agri-footprint database (Blonk Agri-footprint BV, 2014) and the French agricultural LCI database AGRIBALYSE (Colomb et al., 2015; Koch and Salou, 2016). More often than not, LCA practitioners use default pre-calculated emission factors, such as those provided in Albanito et al. (2017), Bouwman and van der Hoek (1997), Bouwman (1996), IFA/FAO (2001) and IPCC (Hergoualc'h et al., 2019; IPCC, 2006). This strategy is in principle aligned with the nature of LCA, which aims at **estimating** impacts, to be analysed in a comparative fashion (Bernstad and la Cour Jansen, 2012; Heijungs, 2021; Prado, 2018).

On the face of this situation, the aim of this study is to review simple N models used for estimating direct field emissions of agricultural systems, and to assess their suitability to agricultural systems featuring contrasting agricultural situations: organo-mineral or organic fertilisation, non-arable crops (e.g. perennials, vegetable gardening, associated crops), or happening under tropical and sub-tropical conditions. To achieve it we selected a set of models representing a broad gradient of complexity and approaches. Models were described and their outputs associated with a set of example cropping systems compared to provide some information about their relative sensitivity, although it is not possible to conclude on their predictive quality in the absence of a sound set of measured emission data (Bockstaller et al., 2008; Buczko et al., 2010). Comparing outputs of various N models has been recommended in Bockstaller et al. (2008) as a suitable model comparison strategy. Ideally, the models' predictions should be compared to a dataset of field measurements of gaseous N emissions and nitrate leaching, but such datasets are still rather rare.

On the base of said comparison, in contrast with key factors determining N emissions from agriculture, recommendations were offered on the minimum requirements a model set would have to fulfil to accurately represent N emissions from contrasting agricultural situations, in a context of LCA applications.

1.2 General limitations of simple N emission models in agriculture

Few models across the modelling continuum are able to model N dynamics across agricultural situations (Cannavo et al., 2008). In the LCA context, N direct emission models commonly used, for instance those simple models used by popular LCI databases such as ecoinvent v3.5 and AGRIBALYSE v1.3 (and earlier versions), are predominantly representative of conventional fertilisation of field crops by synthetic fertilisers. These models are not well adapted to the *modus operandi* of organic fertilisers, or to agricultural systems other than field crops. Moreover, these models often disregard the fertilisation efficiency, that is to say, the effect on emission intensity due to fertiliser inputs beyond the plant needs or after their peak absorption period, as well as the position of a crop of interest within a crop rotation.

Various aspects challenge modelling of direct emissions from organic fertilisation in LCA. For instance, the content and quality of nutrients in organic fertilisers is often unknown or very variable, especially the less industrialised ones, such as digestates, composts, separated solid and liquid phases (of slurries, sludge and digestates), and animal effluents. Moreover, organic fertilisers contain both organic and mineralised N, where the organic fraction experience varying rates of mineralisation according with management and pedoclimatic conditions. Several approaches have been developed to model N mineralisation of added organic matter (i.e. agricultural residues, organic fertilisers) and soil organic matter (Benbi and Richter, 2002; Clivot et al., 2017; Kwiatkowska-Malina, 2018; Manzoni and Porporato, 2009), including mineralisation kinetic curves (Doublet et al., 2011; Morvan et al., 2006; Parnaudeau et al., 2006); yet simple N models often include pre-calculated mineralisation factors representative of specific agricultural situations. Simple models and emission factors for direct field emissions predominantly focus on conventional mineral fertilisation of field crops (Meier et al., 2015). Furthermore, most LCA-oriented models focus on single crops rather than on crop rotations, which consequently disregards the abovementioned delayed N (and C) dynamics of organic fertilisation and crop residues left on the field.

The specificities of perennial crops are not captured by the most commonly used simple models such as those used by ecoinvent —i.e. SALCA-N (Richner et al., 2014)—, nor by emission factors such as the popular ones proposed by Bouwman and colleagues (Bouwman et al., 2002a, 2002b, 2002c; Stehfest and Bouwman, 2006). These specificities include deep root system expansion, relatively high yields and low nutrient requirements, and much longer rotation times, when compared with arable crops (Bessou et al., 2013; Cerutti et al., 2014). A similar challenge applies to vegetable gardening, featuring much shorter rotation times, and associated crops in the same field, where interactions among crops with different N absorption behaviours are not easy to estimate (Perrin et al., 2014). Associated crops are seldom modelled in LCA, and their direct emissions are complex to estimate, as crops are associated due to reinforcing mechanisms (including N absorption) which are difficult to represent with simple models (Bessou et al., 2013).

Simple models and emission factors for direct field emissions are predominantly based on temperate weather conditions. Only the IPCC (IPCC, 2006) and Bouwman and van der Hoek (1997) provide emission factors for tropical and sub-tropical conditions, and for conventional field crops (Bessou et al., 2013). The draining regimes, as well as other pedoclimatic conditions affecting these emissions, are different across agro-climatic zones (van Wart et al., 2013). It has been suggested that IPCC-based results are flawed for N_2O emissions in tropical environments (van Lent et al., 2015).

The practice of LCA in developing countries faces additional challenges than in developed ones (Basset-Mens et al., 2018), including the paucity of background inventory data (Perrin et al., 2014), as well as the lack of reliable statistics and adapted direct emission models. Most developing countries feature tropical and sub-tropical conditions.

2 Material and methods

We performed a literature review to select (section 3.1) and investigate the known general limitations of simple N models (section **Erreur ! Source du renvoi introuvable.**), to frame the specific limitations identified during our data-based comparison of selected models (section 3.2), enabling us to provide N modelling recommendations in an LCA context (section 0).

2.1 Criteria for model selection

We established criteria for selecting simple N models, as well as a strategy for an objective and comprehensive comparison.

Simple N models to be tested were chosen based on their applicability in LCA, following a literature review. For instance, French researchers apply LCA to many different agricultural systems, including organic, gardening, perennial, and tropical ones, and have produced several methodological proposals and case studies regarding direct field emissions estimation, be it specific equations or combinations and adaptations of existing models (e.g. Bockstaller and Girardin 2010; Bellon-Maurel et al. 2015; Koch and Salou 2016; Brockmann et al. 2018). We privileged models and model sets used in the European and French research environment (both in European and non-European contexts), as it is one of the more prolific communities in agricultural LCA, as represented for instance in the international LCA Food conferences (https://www6.inra.fr/lcafoodconferencearchives/).

We consider emission factors to be, by definition, less representative of a particular agricultural situation (an agricultural system under given pedoclimatic conditions) than the outcomes of a simple model or model set that captures the main determinants of emissions, and whose inputs include parameters that can be calibrated to the given situation or to a similar one. Pre-calculated emission factors, notably those proposed by Bouwman and colleagues (Bouwman and van der Hoek, 1997; Bouwman, 1996; Bouwman et al., 2002c, 2002a, 2002b; L. Bouwman et al., 2013; Lex Bouwman et al., 2013; Stehfest and Bouwman, 2006), are widely used in agricultural LCA. Nonetheless, as suggested in Goglio et al. (2015), simple models should be preferred to fixed emission factors for soil organic C modelling in agricultural LCA, because they allow for a better adaptation to specific conditions. Therefore, generic emission factors were excluded from this review, except for comparison purposes.

2.2 Model (outputs) comparison strategy

Selected models were fed with agricultural and pedo-climatic data from four reference agricultural datasets (see section 2.3), and ran to obtain predictions of N emissions. For model-estimated parameters such as plant N uptake, we always retained the agricultural datasets data. Results of simple models were compared with outputs from the complex dynamic model STICS (Brisson et al., 2003), and the resulting differences analysed. STICS was retained for a direct comparison of simple models with a complex simulation model, which takes into consideration more parameters and mechanisms of emissions than simple models. Moreover, simulation models are better equipped to represent dynamic of emissions, and also the cumulative effect of repeated inputs of organic matter, a key mechanism associated with organic fertilisation (Constantin et al., 2012, 2010). The level of predictive error associated with STICS has been computed for a wide range of systems and pedo-climatic conditions, and determined to be, in decreasing order of relative importance, more prevalent for nitrate leaching, plant biomass, N uptake, and soil water (Coucheney et al., 2015). All retained simple models were implemented in Excel, and fed with the experimental datasets. In the case of multi-annual datasets, we retained average annual values to feed the simple models, while presented STICS results consist of the mean of annual outputs.

Disaggregation of mineral- and organic-fertilised subsystems was possible for all agricultural datasets. A few measurements were available for nitrate losses in the Senegal and Reunion Island sites, which were used as reference points to assess the quality of model predictions, beyond their sensitivity. Measurements were made with lysimetric plates at 40 and 100 cm, respectively.

A 3-way ANOVA and post-hoc Tukey's tests (Piepho, 2018) were firstly performed to assess the effect of three factors and their interactions on N emissions across models: the type of N emission considered (*Emission*), the study site (*Site*) and the fertilisation regime (*Ferti*).

Outputs from selected models were then compared, per specific emission, across agricultural datasets (study sites) after normalisation because the emissions simulated by the models were not in the same scales for the different sites and were therefore normalised by the average method (**Erreur ! Source du renvoi introuvable.**) to enable the comparison between models across sites.

$$x' = \frac{x - \text{mean}\ (x)}{\max(x) - \min\ (x)} \qquad \textbf{Eq. 1}$$

where *x'* is the normalised emission, *x* is the output model, mean (x) is the average value of the different models, min (*x*) and max (*x*) are the minimum and maximum values of the different models in each site.

A second 3-way ANOVA and corresponding post-hoc Tukey's tests were then performed to conduct pairwise comparison across models. The three factors tested were the model itself (*Model*), *Emission* and *Ferti*. The normality of the residues was checked prior to the statistical analyses and p-values were corrected with the Benjamini-Hochberg procedure (https://stat.ethz.ch/R-manual/R-devel/library/stats/html/p.adjust.html) to reduce the false discovery rate. The significance threshold was fixed to 5%. Data were processed using the R software (R Core Team, 2020).

2.3 Agricultural datasets for model comparison

We used reference agricultural datasets to test the models, and highlighted the reasons for their differing results. Datasets used for model comparison include one for field crops in France, two for market vegetable gardening in Benin and Senegal, and one for sugarcane in Reunion Island. Such variety permits to capture differing agricultural systems under very contrasting pedo-climatic conditions: temperate, tropical wet (continental and islander) and tropical dry. All datasets feature data for mineral and organic fertilisation. The main pedoclimatic conditions of all four sites are synthesised in Table 1. Common total and mineral N contents for organic fertilisers, as detailed in (Galland et al., 2020), were retained across sites to reduce parameter uncertainty.

Table 1. Pedoclimatic conditions in the sites where the agricultural activities represented by the reference datasets take place

Key features	Feucherolles, France	South Benin[a]	Sangalkam, Senegal	Reunion Island
Soil texture	Silty	Sandy	Sandy	Clayey
Soil type (FAO/IIASA, 2009)	Luvisol	Arenosol	Arenosol	Nitisol (Ferralsol)
Total topsoil C (%)	1.10	0.70	0.64	1.86
Total topsoil N (%)	0.11	0.05	0.06	0.16
Topsoil clay fraction (%)	16.12	13.00	9.12	43.30
Topsoil pH	7.34	6.02	6.61	6.10
N in Soil Organic Matter (kg N/ha)	4 997	1948	2 689	6 720
Global agro-ecological zone (IIASA/FAO, 2012)	Temperate oceanic forest	Tropical rainforest	Tropical shrubland	Tropical mountain system
Average annual precipitation	583	1101	424	2 665
Average annual temperature (°C)	10.7	25	26.5	25

[a] Average of 12 sites (Perrin, 2013)

2.3.1 Temperate field crop: maize in central France

The first dataset used for comparisons comes from the long-term field experiment QualiAgro (https://www6.inra.fr/qualiagro/), corresponding to a field trial located on the Plateau des Alluets le Roi, Feucherolles, about 20 km west of Paris, France. QualiAgro is part of the SOERE-PRO network (System of Observations, Experiments and Environmental Research on Organic Residual Waste, https://si-pro.fr/).

The trial consists of a maize-wheat rotation in the period 1998-2013, fertilised with the mineral fertiliser Urea Ammonium Nitrate solution (aka "Solution 390", a liquid mixture of urea and ammonia nitrate, featuring 30% N in the form of 25% N-NO_3, 25% N-NH_4 and 50% N-NH_3) at two mineral fertilisation rates: minimal and optimal; and amended with four different organic products (cattle manure, compost of organic waste, compost of sludge and green waste, and compost of green waste). The experimental setup is described in Cambier et al. (2014) and Bourdat-Deschamps et al. (2017), and both annual fertiliser inputs (for the optimal mineral fertiliser rates) and resulting crop yields depicted in Table 2. Plant uptake was estimated between 150 and 188 kg N/ha (according with the fertilisation scenario, which includes a control mineral fertiliser-only scenario).

Table 2. Fertiliser treatments for the central France maize-wheat dataset (1998-2013)

Crop	year	Average of 4 organo-mineral treatments			Mineral treatment	
		Organic fertilisers (kg N/ha)	Solution 390 (kg mineral N/ha)	Yield (kg/ha)	Solution 390 (kg mineral N/ha)	Yield (kg/ha)
wheat	1998-1999	-	-	-	-	-
maize	1999-2000	294	79	11 274	79	7 608
wheat	2000-2001	-	102	7 991	51	7 902
maize	2001-2002	335	68	11 497	68	11 076
wheat	2002-2003	-	124	7 196	62	6 501
maize	2003-2004	352	50.8	11 700	50.8	9 647

wheat	2004-2005	-	122	8 642	61.5	7 614
maize	2005-2006	312	51.5	8 234	51.5	6 152
wheat	2006-2007	-	121.1	7 341	60.3	5 698
barley	2007-2007	326	82.3	9 760	82.3	7 426
maize	2008-2009	330	-	8 395	108	8 453
wheat	2009-2010	-	173.5	8 189	110	7 702
maize	2010-2011	315	12.5	6 722	136	6 965
wheat	2011-2012	-	199	6 254	99	5 339
maize	2012-2013	287	-	8 210	110	8 457
Annual average		170	79	8 094	101.8	7 103
No irrigation; fertilisers spread by broadcaster, with soil incorporation; rooting depth: 1.8 m						

2.3.2 Tropical wet garden crop: tomato in south Benin

The second dataset represents off-season (i.e. grown during the dry season, irrigated, featuring low yields) field tomato production in south Benin during 2011-2012, as described in Perrin (2013), who used STICS to estimate N emissions from 12 different systems (Table 3). In average, these tomato systems received 448.7 kg N/ha, 337.4 kg N/ha of which from poultry manure, resulting in a yield of 5 092 kg FM/ha. Plant uptake was estimated at 200 kg N/ha.

For this dataset, as a STICS-based comparison device, emission factors computed with STICS as presented in Perrin (2013) were retained —expressed as a function of total N inputs—: N_2O = 0.6%, NO_3 = 10% (range 0 to 52%) and NH_3 = 10% (range 0 to 37%). NO_x emissions are not originally computed by Indigo-N v2.70 or STICS, but Perrin (2013) estimated an emission factor based on total N inputs (for the specific conditions of her study, to complement her STICS results).

Table 3. Fertiliser treatments of the south Benin off-season tomato dataset (2011-2012)

Fertiliser (kg N/ha)	Average of 8 organo-mineral treatments	Average of 4 mineral treatments	Weighted average of all treatments
Urea (46% N)	27.8	65.3	40.3
NPK (16-16-16)	87.6	38.0	71.1
Dried poultry droppings (0.5% N)	506.1	0	337.4
Total	621.5	103.3	448.7
Yields (t FM/ha)	4.2	6.8	5.1
Irrigation: 500 mm/ha in average; fertilisers spread by hand, without soil incorporation; rooting depth: 0.5 m; FM = fresh mass			

2.3.3 Tropical dry garden crop: market vegetables in north-west Senegal

The third dataset includes historical data (2016-2018) from the experimental site set up in 2016 by the Laboratoire Mixte International Intensification Ecologique des Sols Cultivés en Afrique de l'Ouest (LMI IE SOL), in Sangalkam, near Dakar, Senegal, in the context of the SOERE-PRO network. The area, neighbouring the *Niayes* coastal strip, is semi-arid.

The experimental design features a total randomised set-up, with 16 m² plots and three replicates per fertilisation treatment (Table 4). Three organic fertilisers are studied, at two applied doses representing 100 and 200% of the recommended dose of mineral fertilisers: poultry litter (210 kg N/ha), sewage sludge (122 kg

N/ha), and agricultural digestate (103 kg N/ha). The crops consist of rotations of lettuce-carrot-tomato. The fertilisation and other agricultural practices are considered as representative of peri-urban market vegetable gardening in the greater Dakar area. The cumulative plant uptake by this rotation was estimated at 350 kg N/ha, but 593 kg N/ha were added per year (considering only the mean of all treatments furnishing 100% of fertiliser needs of the rotation), 197 kg N/ha of which were furnished by mineral fertilisers.

Table 4. Fertiliser treatments of the Sangalkam market garden vegetables dataset (2017-2018)

Fertiliser (kg N/ha)	Treatment 1 (organo-mineral)	Treatment 2 (organo-mineral)	Treatment 3 (organo-mineral)	Weighted average of all treatments
Urea (46% N)	233.7	153.3	45.5	197.0
Limed sewage sludge (1% N)	262.6			152.0
Digestate of cattle manure (0.5% N)		506.0		172.4
Dried poultry droppings (0.5% N)			157.1	71.3
Total	496.3	659.3	202.6	592.7
Annual yields of the rotation (t FM/ha)	21.3	12.4	18.5	18.7
Irrigation: 1305 mm/ha in average; fertilisers spread by hand, with soil incorporation; rooting depth: 0.5 m; FM = fresh mass				

2.3.4 Tropical wet field crop: sugarcane in Reunion Island

The fourth dataset includes data (2017-2018) from the experimental site set up in 2014 by the Recycling and risk research unit of CIRAD in La Mare, near Saint-Denis in Reunion Island, France (20°54'12.2"S, 55°31'46.6"E). The experimental trial took place in a highly monitored site belonging to the SOERE-PRO network, designed to investigate the long-term impact of organic fertilisation on the different compartments of the sugarcane agroecosystem. The trial was planted in March 2014 with one sugarcane variety (R579) and a 1.5 m row-spacing. The trial was irrigated throughout the crop cycle (29 mm/week) except for the last two months before harvest. The trial consisted of six treatments, each with a different fertiliser, which were repeated in 5 blocks. Each plot made up of six sugarcane rows of 28 m, constituting a total plot area of 250 m^2. The data used in the present study were obtained from three distinct fertilisation treatments (Table 5) according with the dominating source of nutrients: urea, sewage sludge and swine slurry (the last two complemented with urea applications). In average, 152.2 kg N/ha were furnished, 54.7 kg N/ha of which by mineral fertilisers, to satisfy a plant uptake of 150 kg N/ha, resulting in a yield of 36 t/ha.

Table 5. Fertiliser treatments of the Reunion Island sugarcane dataset (2017-2018)

Fertiliser (kg N/ha)	Treatment 1 (mineral)	Treatment 2 (organo-mineral)	Treatment 3 (organo-mineral)	Weighted average of all treatments
Urea (46% N)	71.8	47.6	44.6	54.7
Limed sewage sludge (1% N)		24.0		8.0
Swine slurry (0.4% N)			268.5	89.5
Total	71.8	71.6	313.1	152.2
Yields (t FM/ha)	91.0	110.0	94.0	99.3
No irrigation; fertilisers spread by broadcaster, with soil incorporation; rooting depth: 1.0 m; FM = fresh mass				

3 Results and discussion

3.1 Selected simple models and their features

Several simple models are contrasted in Table 6: ecoinvent v3 (an international model widely used in LCA), World Food LCA database v3 (a European model, heavily based on ecoinvent), Indigo-N v1/v2 and AGRIBALYSE v1.2/v1.3 (French models used in French LCA research and case studies), Calculateur AzoteViti and Mineral fertiliser equivalents (MFE) calculator (recent French research models), and FAO N-balances (international models with tropical calibration, used in FAO case studies). These models, among others, are used by LCA practitioners to complete their agricultural life cycle inventories. Model sets were contrasted regarding their consideration of the biophysical processes determining N losses to the environment from agriculture, namely plant uptake, NH_3 volatilisation, NO_3 leaching, N transfer by erosion and run-off, N_2O emissions by nitrification ($NH_4 \rightarrow NO_3$) or denitrification ($NO_3 \rightarrow N_2$). Models' consideration of management drivers such as rotation over-fertilisation and fertilisation technique were also contrasted, as well as regarding their management of the mineralisation of soil organic matter (SOM) and organic matter provided by fertilisers, drainage regimes, and the allocation of fertilisers (and thus of emissions, mainly by leaching) among crops in a crop rotation.

Table 6. Direct emission model sets used in France

	International model sets		French model sets		French research models		Other international approaches	
Features	ecoinvent v3	World Food LCA database v3	Indigo-N v1/v2	AGRIBALYSE v1.2/v1.3	Calculateur AzoteViti	Mineral fertiliser equivalents (MFE) calculator	FAO N-balances (plot/farm scale only)	Pre-calculated emission factors
Source	Nemecek and Schnetzer (2012)	Nemecek et al. (2015)	Bockstaller and Girardin (2010)	Koch and Salou (2015, 2016)	Bellon-Maurel et al. (2015)	Brockmann et al. (2018)	Roy et al. (2003)	Various (e.g. Bouwman et al., 2002c)
Geographical validity	Switzerland, Europe, Global (SQCB)	Global (main food-exporting countries)	France	France, a few tropical	France	Denmark, France, Germany, Netherlands, Poland	Calibrated for Africa, but global applicability	Variable, but mainly global
Crops covered	Field crops	Field crops, grasslands	Field crops, grasslands	Field crops, grasslands, vegetables, rice, fruits	Grape vines	Crop-independent	Field crops	Field crops, other crop types
Types of fertilisers	Mineral, manure, sugarcane vinasse	Mineral	Mineral, certain organic	Mineral, certain organic	Mineral, most organic	Mineral, most organic	Mineral, most organic	Mainly mineral
Timescale	Annual	Annual	Roughly annual [i]	Roughly annual [i]	Annual	Annual and long-term	Annual	Annual
Physical scale	Plot, farm (AGRAMMON)	Plot	Plot, farm	Plot	Plot	Plot	Plot, farm	Any
N uptake by plants (plant requirements)	Pre-calculated factors based on a combination of STICS (Brisson et al., 2003) and factors from Flisch et al. (2009)	See ecoinvent v3	N uptake coefficients per crop type and sowing date, based on plant needs	N uptake coefficients per crop type (for SQCB only)	Computed from the N needs for grape production, from literature	Not considered	NUTMON model [h]: millet, sorghum, maize, rice, wheat, and other crops	Not considered
N mineralisation of added organic matter	Not explicitly considered	Fixed factors with corrections (EMEP/EEA 2013 model). See NH_3	Mineralisation factors for harvest residues, minimum value depending on	Research mineralisation kinetic curves used only for allocation of fertilisation	AZOBIL equation (Machet et al., 1990) modified by a monthly soil moisture curve	Plant available nitrogen (PAN) mineralisation factors (WEF, 2005)	Fixed factors of various origins (literature, NUTMON model)	Implicit

			soil and its increase due to over-fertilisation					
Soil mineral N	Not explicitly considered	Not explicitly considered	Minimum value depending on soil and increase due to over-fertilisation	Not explicitly considered	Not explicitly considered	Not explicitly considered	Not explicitly considered	Not explicitly considered
N mineralisation of soil organic matter (SOM)	Fixed factors with correction factors (SALCA-NO3 and SQCB-NO3 models). See NO_3	See ecoinvent v3	Mineralisation equation (Taureau et al., 1996) [a]	Leaching models retained do not require mineralisation	Implicit in the N balance approach used (Kücke and Kleeberg, 1997)	Deliberately set to 0	Same as previous	Implicit
NH_3 volatilisation model	AGRAMMON model tier 3 [b] (https://www.agrammon.ch/) for emissions from leaf surface, mineral fertilisers, manure and vinasse. Emission factors for manure management (Menzi et al., 1997).	EMEP/EEA 2013 tier 2 [c] (EMEP/EEA, 2013)	Volatilisation coefficient from literature, per type of fertiliser, the limestone content, the time of year, and the soil tillage	EMEP/EEA 2009 tier 2 (EMEP/EEA, 2009) for organic fertilisers EMEP/CORINAIR 2006 tier 2 (EMEP/CORINAIR, 2006) for mineral fertilisers	Volatilisation coefficients from literature: NH_4 assumed to follow an exponential decay with half-life of 12 h. Affected by rain.	Same as ecoinvent 3 for organic fertilisers EMEP/EEA 2013 tier 2 for mineral fertilisers	All N gaseous emissions are considered together, as N Empiric equations from the NUTMON model	Emission factors for chemical fertilisers, for developed and developing countries (Bouwman and van der Hoek, 1997)
N_2O emission model	IPCC 2006 tier 1 [g] (De Klein et al., 2006; IPCC, 2006), for direct (mineral and organic fertilisers, and crop residues) and indirect emissions (from NO_3 leached)	See ecoinvent v3	Empiric equation (Bouwman, 1996) with denitrification computed from IPCC 1997: 1.25%	IPCC 2006 tier 1	empiric equation (Bouwman, 1996) with denitrification computed from IPCC 2006: 1%	IPCC 2006 tier 1	See NH3	Factors and empiric equation for N_2O and NO (Bouwman et al., 2002a, 2002c; IFA/FAO, 2001) Emission factors for tropical and sub-tropical

								N_2O, per continent, country, crop type and fertiliser type (Albanito et al., 2017)
NOx emission model	Fixed factor for NOx emissions from N_2O (from a personal communication)	Fixed factors for mineral and organic fertilisers from EMEP/EEA 2013	excluded	EMEP/EEA 2009 tier 1	excluded	EMEP/EEA 2013 tier 2	See NH3	See N_2O
NO_3 leaching model	For Europe: SALCA-NO3 [d] (Richner et al., 2014). For other countries: SQCB-NO3 [e] (Faist Emmenegger et al., 2009), an adaptation of the de Willigen (2000) model (Roy et al., 2003). Drainage not considered by either model.	See ecoinvent v3	Empiric equation explicitly including drainage (Burns, 1976) modified (Laurent and Castillon, 1987), for post-fertilisation, using N absorption curves. COMIFER [a,f] (COMIFER, 2001) for winter drainage, based on N-balances.	ARVALIS method (Tailleur et al., 2012) for field crops. DEAC (Cariolle, 2002) for grassland. SQCB-NO3 for perennials and vegetables. IPCC 2006 tier 1 (De Klein et al., 2006) for tropical. Only DEAC and ARVALIS method include drainage.	Empiric equation (Burns, 1975). Calculated from N budget after deducting NH_3 and N_2O emissions. Monthly timestep.	Empiric equations from NUTMON model (Roy et al., 2003), distinguishing background nitrate emissions from SOM N. Drainage not considered.	Empiric equations from the NUTMON model.	NO_3 leaching factors are often computed with dynamic models (e.g. Groenendijk et al. 2005; Kasper et al. 2019) or measurements fitted to regression models (e.g. Vázquez et al. 2005; Bruun et al. 2006). Some models include drainage.
Nitrification (NH_4 --> NO_3)	Not considered	Not considered	Not explicitly considered	Not explicitly considered	Not considered	Not explicitly considered	Not explicitly considered	Not explicitly considered
Denitrification (NO_3 --> N_2O)	Implicitly considered (NOx), not considered (SALCA-NO3)	See ecoinvent v3	1.25% of remaining N after volatilisation (IPCC 1997)	Not explicitly considered	1% of remaining N after volatilisation (IPCC 2006) (De Klein et al., 2006)	Not explicitly considered	Not explicitly considered	Not explicitly considered

Mineral fertiliser equivalents	not considered	not considered	MFE coefficients per crop type	not considered	not considered	Calculated with the PAN formula	not considered	N/A
Consideration of over/under fertilisation	not considered, SQCB-NO3 seems to be calibrated for adequate fertilisation	not considered	Calculation of an increase in soil mineral N after harvest due to over-fertilisation following Machet et al. (1997)	not considered	not considered	not considered	not considered	Assumes adequate fertilisation
Required input data	Target crop, N inputs	Target crop, N inputs	Target crop, next crop, specific dates, recommended and actual N inputs	Target crop, N inputs, French region	Monthly climate data, soil data, N inputs	Fertiliser application data	Detailed farm operation data	N/A

[a] Current version of the method: COMIFER (2013). [b] AGRAMMON parameters: total ammonia nitrogen (TAN) in fertilisers, emission rates, various corrections factors related with management. [c] EMEP/EEA 2013 tier 2 parameters: TAN in fertilisers, amount of mineral fertilisers, emission factors per soil pH, correction factors (application method, application time and season). [d] SALCA-NO3 parameters: N mineralisation from the SOM per month, N uptake by vegetation (if any) per month, N input from the spreading of fertiliser, soil depth. Assumes a priori a soil with 15% clay and 2% humus, but modifiable. [e] SQCB-NO3 parameters: precipitation and irrigation, clay content, rooting depth, N in fertilisers, N in organic matter, N uptake by plants. [f] COMIFER N-balances consider residues, residues from over-fertilisation, mineralisation of crop residues and humus, mineral and organic N. [g] IPCC 2006 tier 1 parameters: total N in fertilisers, N in crop residues, N from mineralisation of SOM, NH_3 losses, NO_2 losses, NO_3 losses. [h] The NUTMON (Nutrient Monitoring for Tropical Farming Systems) model is not available online, as it has been replaced by the MonQI (Monitoring for Quality Improvement) model (https://www.monqi.org/). [i] i.e. current and next crops.

3.1.1 International models

The ecoinvent database v3 (Nemecek and Schnetzer, 2012) retains the AGRAMMON model (https://www.agrammon.ch/), calibrated for Swiss conditions, for NH_3 volatilisation of both mineral and organic fertilisers. For NO_3 leaching, ecoinvent v3 retains the SQCB-NO3 model (Faist Emmenegger et al., 2009), which is based on a widely used (including by FAO) regression model by de Willigen (2000), which in turn is based on NUTMON data (Roy et al., 2003). The Nutrient Monitoring for Tropical Farming Systems (NUTMON) model, was calibrated for tropical conditions. It is currently obsolete and has been replaced and extended by the Monitoring for Quality Improvement (MonQI) model (https://www.monqi.org/). The models used in ecoinvent v3 are claimed to have global applicability, but the analysis of certain modelling elements suggests it is an exaggerated claim. For instance, very few added organic matter used as fertiliser are represented (manure, sewage sludge and sugarcane vinasse only). Moreover, the model set does not explicitly compute mineral fertiliser equivalents for these organic fertilisers, nor does it consider over-fertilisation. A similar statement can be made on the nitrate leaching model proposed in Brentrup et al. (2000).

The World Food LCA database v3 (Nemecek et al., 2015) uses the same approach as ecoinvent v3 for N emissions, except that it retains the EMEP/EEA (2013) tier 2 model (EMEP/EEA, 2013) for NH_3 volatilisation. This guideline/model set proposes volatilisation factors for mineral fertilisers and manure only. No results were computed for this model, as the modelling principles are virtually identical to ecoinvent's. The most recent 3.5 version of the database maintains the 3.0 version model selection (Nemecek et al., 2020), but updating EMEP/EEA tier 2 to the 2016 version (EMEP/EEA, 2016).

The Agri-footprint database model set was not retained because it systematically and exclusively uses IPCC (De Klein et al., 2006; IPCC, 2006) simple models.

The FAO N-balance approach (Roy et al., 2003), at the plot and farm scales, are tailored to tropical conditions, as they heavily rely on the NUTMON model. All gaseous emissions are considered aggregated, and fixed N mineralisation factors considered. No results were computed for this model.

3.1.2 French models

AGRIBALYSE v1.3 (Koch and Salou, 2016) proposes a combination of models, some of which are tailored to French conditions. The overall modelling strategy is coherent and comprehensive, yet outdated models were retained for NH_3 volatilisation —EMEP/EEA 2009 (EMEP/EEA, 2009) and EMEP/CORINAIR 2006 (EMEP/CORINAIR, 2006)—, while various models are used for NO_3 leaching, according to the type of crop, including semi-qualitative ones, namely SQCB-NO3, factors from IPCC 2006 (De Klein et al., 2006, Table 11.3), and the method described in Tailleur et al. (2012).

Indigo-N v2, a subset of the Indigo environmental assessment method (Bockstaller et al., 2008; Bockstaller and Girardin, 2010), consists of a combination of simple models for each different N emission, around an annual mass balance of nutrients allocated to a crop location. The model relies on mineralisation factors, correction factors for management, and empiric equations. It is calibrated to field crops and prairies under temperate conditions. It is the only model accounting for the effects of over- and under-fertilisation on nutrient emissions. The main originality of this model is the calculation of NO_3 losses. Without being a complex soil-plant dynamic model, Indigo-N addresses effects of climatic and soil conditions, fertilisation in regard with the plant's needs or some management practices (e.g. soil management between crops, the following crop). Here the 2.70 version of Indigo-N was implemented in Excel for the comparison.

The approach and associated Excel tool “Calculateur AzoteViti” (Bellon-Maurel et al., 2015), tailored to viticulture, implements models and a modelling approach similar to those in Indigo-N. Consequently, no separate results are presented for this model.

The approach and associated Excel tool “Mineral fertiliser equivalent (MFE) calculator” (Brockmann et al., 2018), is tailored to temperate conditions (a list of European countries is parameterised). Its models to estimate N mineralisation of added organic matter, and NH_3 volatilisation (a combination of EMEP/EEA 2013 tier 2 and AGRAMMON), are flexible as to represent contrasting agricultural situations. Nonetheless its use of the obsolete NUTMON model for NO_3 leaching, as well as its reliance on country-specific data to compute emission factors, limit its suitability to represent contrasting agricultural situations. Moreover, the outputs from this model are not directly comparable with those of Indigo-N and other models, because it considers organic fertilisation only. Moreover, it considers nitrate losses after fertilisation but not as a result of the whole crop cycle. The model was retained nonetheless because it represents a useful approach for organic agriculture.

3.1.3 Model data requirements

The retained model sets (ecoinvent, AGRIBALYSE, MFE and Indigo-N) have different input data requirements, further detailed and contrasted with those of STICS in Table 7. . MFE does not require any data beyond the fractioning of N in the fertiliser and basic knowledge of the fertilisation mechanism, as it is based on emission and operational correction factors. Indigo-N features similar data requirements (at a larger time resolution) than complex models such as STICS, whose simulations are based on very detailed data files for soil, crop, and (daily) weather. The basic data for non-expert use are quite similar. Pedo-transfer functions are available in STICS to estimate parameters that are less often measured (or not measurable). What changes a lot is the interface that makes Indigo easy to use, and the level of expertise to properly interpret the results.

Table 7. Data requirements of selected N emission model sets used in France, compared with those of a complex dynamic system (STICS)

Data	ecoinvent v3	Indigo-N v1/v2	AGRIBALYSE v1.2/v1.3	STICS v8.5 *
Weather data	• Annual rain and irrigation (mm)	Data provided for France, but needing adaptation for other geographies: • Mean annual temperature (°C) • Drainage after winter runoff (January-March) (mm) • Drainage after spring runoff (April-June) (mm) • Winter drainage (mm) • Inter-annual frequency of drainage after winter runoff (fraction ≤1) • Inter-annual frequency of spring winter drainage (fraction ≤ 1) • Excess mineralisation during drainage period (%)	• Duration of draining period (days) • Drained surface (%)	• Irrigation (yes/no) • Detailed daily weather data (temperature, rain, etc)
Soil data	• Rooting depth (m) • Clay content (%)	• Texture (list provided) • Clay content (%) • Soil depth class (list provided) • Soil organic matter content (%) • Soil pebble content (%) • Soil limestone content (%) • Soil status as hydromorphic and humiferous (yes/no)	• Texture (list provided) • Rooting depth (cm) • Soil pebble content (%) • Soil organic matter content (%)	• Texture (list provided) • Soil pebble content (%) • Soil organic matter content (%) • pH • Soil capacity • C/N • Soil density by horizon • Permanent wilting point • Etc…
Land preparation		• Type of soil labour (list provided) • Frequency of organic matter inputs (list provided) • Frequency of burial of crop residues (list provided) • Reversal of previous-year prairies (yes/no) • Reversal of previous-year fallows (yes/no)	• Frequency of organic matter inputs (yes/no) • Season of organic fertilisation (list provided)	• Type of soil labour (list provided) • Dates of all soil labour • Active/inert fractions of SOC, which correspond to all Indigo-N parameters in this category

Crop	• N uptake by crop (kg/ha)	• Crop (list provided) • Previous crop (list provided) • Sowing date • Harvest date • Expected yield (kg/ha) • Recommended N inputs (alternative calculation is provided if unknown) • Fate of crop residues (list provided) • Date of residues burial • Irrigation (yes/no) • Irrigation mode (list provided)	• Crop (list provided) • Sowing date • Harvest date • Expected yield (kg/ha) • Co-product yield (kg/ha)	• Crop (list provided) • Sowing date • Harvest method • Harvest date •
Intermediate and next crop		• Intermediate crop (list provided) • Sowing date of intermediate crop • Next crop (list provided) • Sowing date of next crop	• Intermediate crop (list provided) • Sowing date of intermediate crop • Next crop (list provided) • Sowing date of next crop	• The rotation definition informs whether there is an intermediate crop
Fertilisation	• Amount and N content of fertilisers (kg) • TAN content of organic fertilisers (%)	• Fertiliser (list provided) • Quantity (kg, t, m3) • Date of input • Localised input (yes/no) • Burial of input within 24 h (yes/no)	• Fertiliser (list provided) • Quantity (kg, t, m3)	• Fertiliser (list provided) • Quantity (kg, t, m3) • % of N-NH_4 • % of dry matter • % of C • Date of input and associated soil labour
Background data provided	• Correction factors for application of slurry and manure • Coefficient of NH_3 volatilisation (mineral fertilisers) • FAO eco-zones and their assigned carbon content and annual precipitation • USDA soil orders and their assigned clay contents • Crops and their rooting depth as assumed for calculations • Crops and their nitrogen uptake as assumed for	• Minimal mineral N in soil, per soil type • Soil useful reserve, per soil texture and soil depth class • Number of days after which a crop reaches 50% of N uptake, per crop • N absorption values until the onset of winter, per crop • Proportion of N mineralised from crop residues • N content of fertilisers • Percentage of mineralisable N in organic fertilisers • Coefficient of organic fertiliser equivalence, per organic fertiliser	• NPK content of fertilisers • TAN of organic fertilisers • Coefficient of NH_3 volatilisation (mineral fertilisers) and TAN-based coefficient of NH_3 volatilisation (organic fertilisers) • Default factors for estimation of N added to soils from crop residues • Coefficient of N allocation from organic fertilisers to crops, per fertiliser and season • NPK content in exported crops • ARVALIS data for estimation of	• Detailed soil, crop, and weather data files (mainly for field crops)

	calculations • NOx emission coefficient from N_2O	• Coefficient of NH_3 volatilisation, affected by burial and chalk content of soil	leaching (see Table 1) • Default N_2O emission factors from managed soils

* Required input data for STICS represents minimal user-modified inputs, assuming the vast majority of data needs is fulfilled from provided data files for soils, crops, weather, etc.
TAN: total ammonia nitrogen

3.2 Comparison of simple model outputs and specific model limitations

As shown in Fig. 2 and Fig. 3, the N outputs estimated with the different models were related to the type of emission considered and influenced both by the study site and the fertilisation regime. Predicted N emissions are indeed significantly higher in wet study sites, regardless of the type of emissions considered, as supported by the lack of interaction effect between the factors *Site* and *Emission* (see p-values and further details in the Supplementary Material, Table S2). The fertilisation regime significantly influenced the model outputs by doubling the N emissions in the two wettest sites (Benin and Reunion Island) when supplied with organic fertilisers as compared to mineral fertilisation.

As expected, the 3-way ANOVA showed no significant effect of fertilisation regime or emission type as a result of the normalisation procedure. There were no interaction effect of *Model* and fertilisation regime, showing that the ability of models to predict N emissions were not significantly affected by the fertilisation regime. A significant interaction effect between *Emission* and *Model* was however found, indicating that the effect of the model depended on the type of N emission considered (see p-values and further details in the Supplementary Material, Table S3).

Table 8. Predicted values across models for all treatments together (different lowercase letters indicate significant difference at $p < 0.05$ on normalised outputs)

N flow	ecoinvent	AGRIBALYSE	MFE	Indigo-N	STICS
NH_3	11^{a}	38^{b}	28^{ab}	27^{ab}	25^{b}
N_2O	3.1^{a}	3.1^{a}	3.1^{a}	3.2^{a}	3.9^{a}
NO_3	54^{b}	43^{b}	51^{ab}	17^{ab}	12^{a}

For NH_3, the ecoinvent model predicted significantly lower values than the AGRIBALYSE and the STICS models (Table 8). The ecoinvent outputs were systematically at the lowest level while the AGRIBALYSE NH_3 outputs were particularly high in situations of organic fertilisation and in the Senegal site. The NH_3 estimations for these two models highly relied on emission coefficients from EMEP/EEA, IPCC and Bouwman and colleagues publications (see Table 6), which are intended to have a global validity, but which fail to accurately represent emissions in contrasted tropical wet and dry climates. The ecoinvent methodology, in particular, deploys the AGRAMMON model for volatilisation. It can also be noted that AGRIBALYSE applies a volatilisation factor to total ammonia nitrogen (TAN), while ecoinvent applies it to total N. MFE, Indigo-N and STICS models were closer despite punctual divergences although they do not use at all the same calculation.

For N_2O emissions there were no significant differences across models although the outputs of STICS appeared slightly higher than the other models (Table 8), especially in situations with organic fertilisation (Figure 2). Gaseous emissions are predicted across retained models via linear regressions that include parameters such as total N and total ammonia nitrogen (TAN) inputs, as well as emission factors from EMEP/EEA, IPCC and Bouwman and colleagues publications (see Table 6). Ecoinvent and AGRIBALYSE used the same calculation method and other methods used emissions factors which seem to be close to those of ecoinvent and AGRIBALYSE. Denitrification and N_2O emissions are calculated by the STICS model according to the NOE model (Hénault et al., 2005) that considers edaphic parameters such as temperature, water field pore space, soil pH and mineral N availability.

For NOx emissions, AGRIBALYSE always showed higher value than while MFE yielded results between those from AGRIBALYSE and ecoinvent for both French sites (QualiAgro and Reunion Island), and similar results to AGRIBALYSE for African situations (Fig. 2c).

Regarding NO_3, the ecoinvent and AGRIBALYSE models predicted significantly higher emissions than STICS, regardless of the fertilisation regime. NO_3 emissions were rather high for ecoinvent, AGRIBALYSE and MFE, especially in tropical conditions for organic fertilisation (Fig. 3), and lower for Indigo-N and STICS models which are in line with the two available measured values. Thus, ecoinvent, AGRIBALYSE (for tropical conditions) and MFE seem to overestimate NO_3 leaching. They take into account mainly precipitation, irrigation, rooting depth, soil texture and fertiliser inputs, but overlook other factors like evapotranspiration in the calculation of drainage. For instance, the nitrate model used by ecoinvent for non-European contexts and AGRIBALYSE for French vegetables (SQCB-NO3) consists of a regression equation calculating NO3 leaching in function of precipitation + irrigation, rooting depth, clay content, N in soil organic matter, fertiliser amount and crop uptake. In cases where N inputs are below plant requirements, SQCB may yield negative results, and when such inputs are beyond plant needs, predicted leaching soars. The reason for such behaviour is that the model consists of a linear regression calibrated to specific conditions, whose validity is not global, as it seems to exclude situations where crops requirements were not exactly met. High level of precipitation is commonly observed in tropical wet conditions, potentially leading to high leaching output in emission models that not take into consideration evapotranspiration. Indigo-N includes potential evapotranspiration in leaching predictions. STICS computes actual evapotranspiration by taking into account the climatic conditions, the soil water status and the crop physiological state (Constantin et al., 2015; Coucheney et al., 2015). Neither are tackled by the first group of models others factors such as N adsorption and immobilisation, which reduce available NO_3 in the soil, what can explain overestimation of NO_3 leaching. Furthermore, estimating the N fraction of organic fertiliser available for the crop and likely to be lost remained complicated for these models under tropical conditions (heat, wind, moisture) in which the mineral fraction can be rapidly lost through volatilisation and where, conversely, the mineralisation of the organic fraction can be greatly accelerated as compared to temperate conditions (Wetselaar and Ganry, 1982).

All studied models are sensitive to N inputs, but the ecoinvent and Indigo-N models for nitrate leaching are highly sensitive to drainage. A variation of plus or minus 10% produced a higher variation of NO_3 leaching in tropical wet contexts (Fig. 4; see more details in the Supplementary Material, Table S4 and Table S5). Thus, drivers of drainage, namely soil organic carbon and clay content, rooting depth, and water inputs through precipitation and irrigation, should be carefully considered.

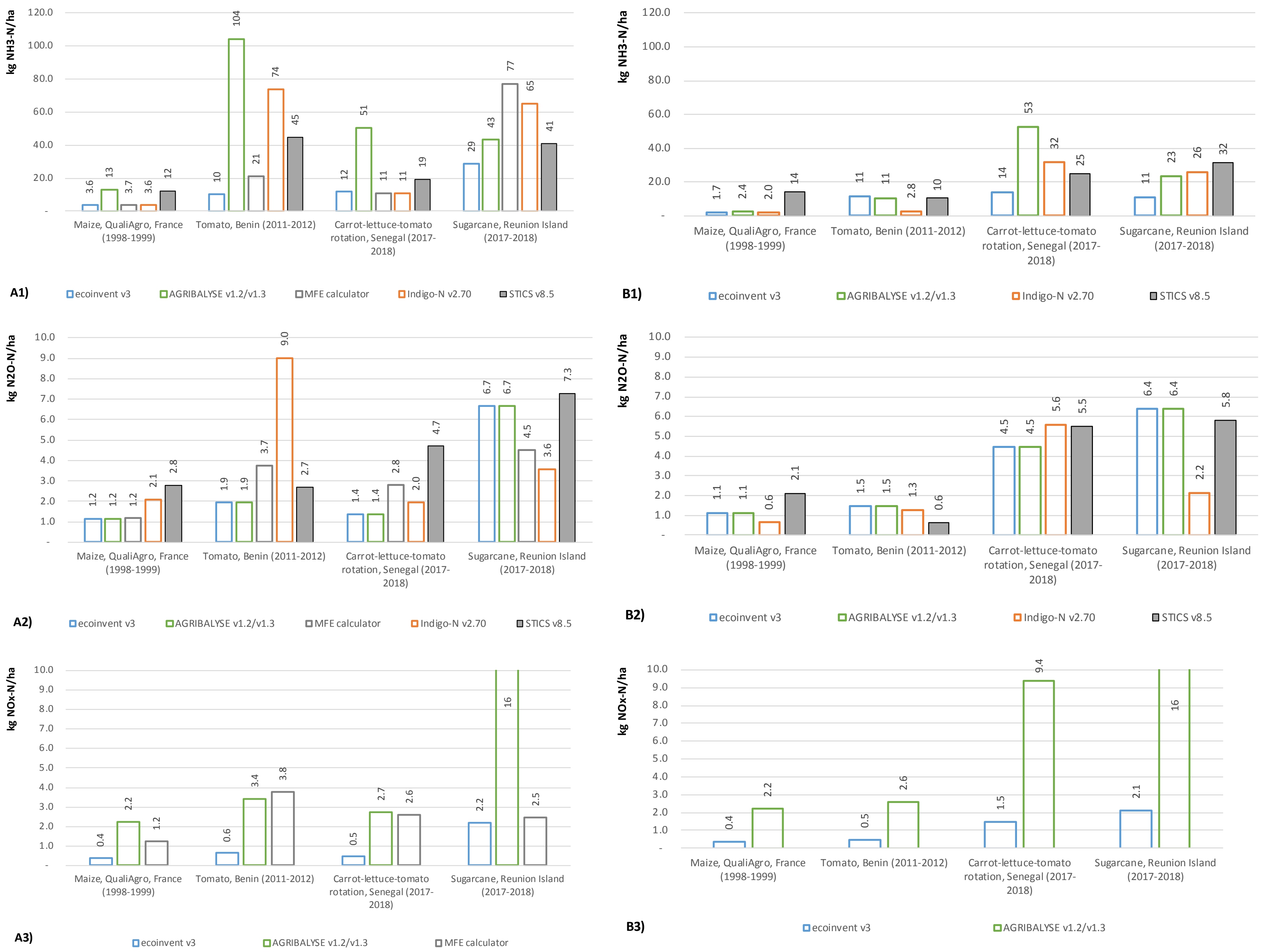

Fig. 2. Estimation of N gaseous direct field emissions across sites and models: A) fertilisation treatments dominated by organic inputs, B) mineral fertilisation treatments equivalent to organic ones; 1) ammonia, 2) nitrous oxide, 3) nitrogen oxide (NO + NO_2)

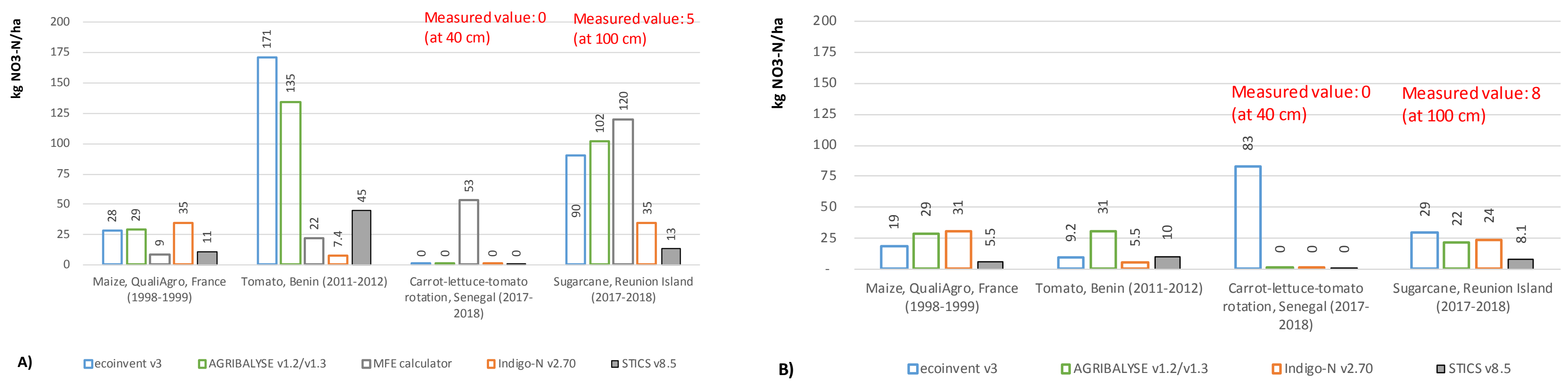

Fig. 3. Estimation of nitrate direct field emissions across sites and models: A) fertilisation treatments dominated by organic inputs, B) mineral fertilisation treatments equivalent to organic ones. Reference values based on averaged lysimetric measurements.

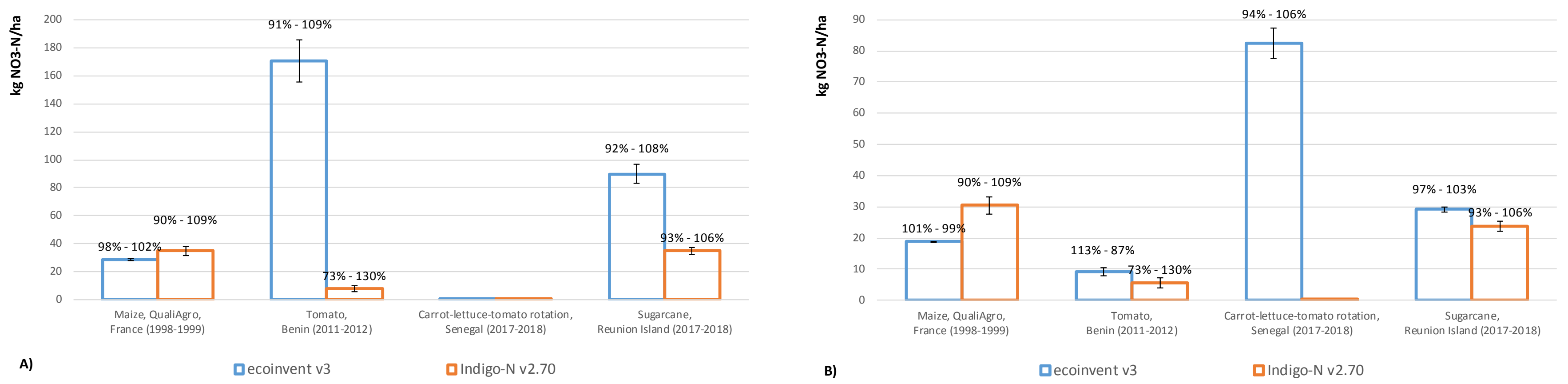

Fig. 4. Sensitivity of ecoinvent and Indigo-N models to a 10% change in precipitation, irrigation and drainage parameters affecting NO_3 leaching predictions for A) fertilisation treatments dominated by organic inputs, B) mineral fertilisation treatments equivalent to organic ones

3.3 Recommendations for N modelling under various agricultural situations

Based on the models we compared, and additional knowledge we have from other models and approaches, we discuss here the principles that should guide future models, in such a way that these future models would be better adapted to organic fertilisation, non-field crops (vegetable, perennial crops and grasslands), and varied pedo-climatic conditions.

A guiding principle of these recommendations was that a balance is sought between simplicity (e.g. data requirements) and comprehensiveness (e.g. consideration of key determinants —mechanisms, drivers— of emissions) (Bockstaller et al., 2015). The ideal N model for LCA should be as simple as possible and as complex as necessary.

3.3.1 Allocation of N inputs among crops in a rotation and long terms effects of organic matter inputs

All N inputs, be it organic fertilisers or crop residues returned to soil, should be allocated among the successive crops in a rotation.

The consideration of crop rotation in LCA has been amply discussed in the literature (e.g. van Zeijts et al. 1999; Goglio et al. 2017), and specific approaches have been implemented in French agricultural LCA databases (Koch and Salou, 2016; Wilfart et al., 2016). It is a consensual conclusion that added nutrients and their associated environmental impacts should be transferred from the crop where they occur to other crops in the rotation, but the basis for such allocation are not always agreed upon. For instance, fertiliser inputs and their direct emissions could be allocated evenly among all crops in the rotation, or weighted by some criteria such as individual crop requirements. Nevertheless, organic inputs may increase the soil organic matter content and thus increase the N mineralisation rate, which is the case for organic fertilisers (Noirot-Cosson et al., 2016; Obriot et al., 2016) as well as for crop or catch crop residues (Constantin et al., 2012, 2010; Tribouillois et al., 2016). Furthermore, the mineralisation of input organic N does not meet the crop period and can last more than one crop period. Such dynamics can only be handled by a dynamic model such as STICS. Thus, a compromise has to be found between this need of modelling dynamics and simplicity of representation of process and data parsimony.

3.3.2 Mineralisation of N in added organic matter (organic fertiliser and crop residues)

The estimation of added organic matter mineralisation in form of organic fertilizer or from incorporation of crop residue is relevant for computing over-fertilisation and related emissions. The N supply by organic fertiliser is often expressed as mineral fertiliser equivalents (MFE) (e.g. Brockmann et al., 2018). MFE is a measure of the capability of organic fertilisers to substitute mineral ones, based on their content of mineralised and rapidly mineralisable N. The model used in Brockmann et al. (2018), based on the Plant Available Nitrogen calculation (WEF, 2005), estimates MFE from mineralisation rates (k_{min}) affecting added organic matter, the mineral N content of all fertilisers (as N-NH_4 and N-NO_3), and the N emissions (NO_3 and NH_3 losses) from all organic fertilisers. Used k_{min} were obtained, as pre-calculated factors, from literature (Sullivan, 2008; WEF, 2005). Brockmann et al. (2018) calculated “first year” and “long term” MFE, based respectively on short- and long-term N mineralisation rates. An alternative approach to MFE is for instance the coefficient of equivalence of effective mineral N in fertilisers (KeqN), which represents the ratio between the amount of N provided by a synthetic mineral fertiliser and the total amount of N provided by an organic source which allows the same N absorption by the crop (COMIFER, 2013).

For contrasting agricultural situations, we suggest the use of mineralisation kinetic curves based on moisture- and temperature-normalised days to determine k_{min}, instead of pre-calculated mineralisation factors. We assumed that this approach, created for temperate conditions, is also valid for tropical ones, as supported by the study of Sierra et al. (2010) for carbon mineralisation under maize and banana. These curves would inform the availability of mineral N to crops, and thus the risk of emissions, once faced with time-specific N needs of the crops (e.g. N absorption curves associated with plant development) and drainage events (e.g. associated with precipitation and irrigation). The use of normalised time allows analysing the role of soil properties on mineralisation, in isolation from climatic factors (confounding factors). Mineralisation kinetic curves for various organic inputs to agriculture occurring under contrasting agricultural situations are available in the literature, for instance, for the vast majority of organic residual fertilisers used in France (Bouthier et al., 2009; Houot et al., 2015), including animal effluents (Morvan et al., 2006) and ago-industrial wastewaters (Parnaudeau et al., 2006); for European catch crop residues (Justes et al., 2009), aboveground crop residues and green manures (Machet et al., 2017), root residues and green manures (Chaves et al., 2004), agricultural composts (Amlinger et al., 2003), and a large variety of plant materials (Jensen et al., 2005); as well as for African leguminous cover crops (Baijukya et al., 2006) and Brazilian root, stem and leaf residues (Abiven et al., 2005).

The Nicolardot et al. (2001) model (later included in STICS as a "decomposition sub-model"), is a dynamic mineralisation model based on the C:N ratio of crop residues and requiring fitting of initial parameters. Once integrated into STICS, there is no more fitting to be done, as it retains the default settings set in Nicolardot et al. (2001) complemented with settings from Justes et al. (2009).

3.3.3 Mineralisation of N in soil organic matter

We propose the empiric equations proposed in Clivot et al. (2017), which are based on 65 field experiments in France, where mineralisation is predicted from soil parameters. Among the various models proposed, the "soil-history-biological" model explains 77% of the computed potential net N mineralisation rate variance. Under this model, the N mineralisation rate of SOM is determined, in descending order of importance, by soil organic N, soil C:N ratio, edaphic factors (clay and $CaCO_3$ content, pH), the effect of returning crop residues to soil, and the activity of soil microorganisms.

This equation should be tested and re-parameterised under tropical conditions to ensure its validity. Tropical soils regularly have very acidic pH values that strongly influence the results obtained with the Clivot equation. An important challenge also concerns the validity of the functional relationship established in temperate conditions between mineralisation and soil clay content stabilising SOM. Clay mineralogy (Motavalli et al., 1995) as well as a higher degree of humification of SOM in tropical soils (Grisi et al., 1998) are supposed to modify the relation.

3.3.4 Consideration of over/under fertilisation

The excess of fertilisation is one of the major component of nitrate leaching. In the Indigo-N model, two hypotheses justify changes in N emissions due to over- and under-fertilisation: that N inputs beyond the optimal dose required by crops entails increased N leaching; and that inputs below the crop requirements do not prevent N leaching in a linear manner. This is due to the minimum amount of mineral nitrogen at harvest, available for instance in COMIFER (2013). The model thus calculates an increase of leachable N consisting of either zero (under under-fertilisation) or 50% (Machet et al., 1997) of the difference between total inputs (minus losses by volatilisation and leaching) and the theoretical optimal dose (COMIFER, 2013). This simplified way to cope with more complex relations between over-fertilisation and increase of soil mineral nitrogen at harvest (ten Berge, 2002) seems to be an acceptable compromise.

3.3.5 NH_3 volatilisation

As volatilisation happens rapidly after fertiliser application (Sommer et al., 2004), it should be deducted from the computation of the other emission pathways.

The EMEP/EEA (2016) tier 2 model (EMEP/EEA 2016, Chapter 3.D - Crop production and agricultural soils) uses emission factors, for mineral fertilisers, for various pedoclimatic conditions (i.e. discriminated by temperature and pH). Tier 1 proposes emission factors for a few organic matter commonly added as fertilisers/amendments, namely sewage sludge and animal effluents. In principle, tier 2 is deemed suitable for contrasting agricultural situations, but correction factors should be applied to better account for agricultural management and climatic conditions. Correction factors are proposed, for instance, for soil preparation and irrigation (Bockstaller and Girardin, 2010), and for spreading technology, incorporation into soil, and seasonality (Brockmann et al., 2018; Nemecek and Schnetzer, 2012).

The AGRAMMON model (https://www.agrammon.ch/) tier 3 proposes emission factors for organic residues added as fertilisers: animal effluents and sugarcane vinasse (Nemecek and Schnetzer, 2012), as well as compost, digestate and sewage sludge (Brockmann et al., 2018). These factors are expressed as emission rates of the available mineral N (TAN) in the added organic matter. The model includes correction factors for spreading technology, incorporation into soil, and seasonality.

Both models are complementary, and a combination of them with correction factors would be suitable to estimate NH_3 volatilisation for contrasting agricultural situations. Both models lack specific factors for industrial organic fertilisers (e.g. manure composts enriched with N-rich materials such as pressed cake and rendered animal products), which should be found in other sources.

3.3.6 N_2O emissions

The widely used IPCC (2006) tier 1 (De Klein et al., 2006, Chapter 11 - N2O emissions from managed soils, and CO2 emissions from lime and urea application) features emission factors for direct N_2O emissions for various soils and added organic matter, based on literature. It proposes as well emission factors for indirect N_2O emissions due to volatilised and re-deposited N (NH_3, NOx), as well as to N lost to leaching and runoff. As various pedoclimatic conditions, crops and added organic matter are considered by specific emission factors, tier 1 is deemed suitable for contrasting agricultural situations, but correction factors should be applied to better account for agricultural management and climatic conditions. Correction factors are proposed, for instance, for soil type, incorporation into soil, and irrigation (Bockstaller and Girardin, 2010).

No model includes denitrification N2 losses as an N-balance element, although it may represent a non-negligible amount of nitrogen (Mathieu et al., 2006; Saggar et al., 2013). The empirical equation in Le Gall et al. (2014) is one possible solution to calculate it in simple way.

3.3.7 NOx emissions

The EMEP/EEA (2016) tier 1 model (Chapter 3.D - Crop production and agricultural soils) uses emission factors for various added organic matter derived from Stehfest and Bouwman (2006). These emission factors are based on a dataset of observations from global agricultural systems, representing 10 climate classes, multiple soil types; organic, organo-mineral and mineral fertilisation; and a variety of field and non-field crops and perennials. In principle, tier 2 is deemed suitable for contrasting agricultural situations. AGRIBALYSE, for instance, retained the EMEP/EEA approach, while ecoinvent uses a single fixed emission factor.

3.3.8 NO_3 leaching

As shown for AGRIBALYSE in Table 6, no single approach seems suitable to represent contrasting agricultural situations, mainly because most models are rather simple models adapted to specific situations. Observed discrepancies among models outputs with the few measured values for the tropical site of the study were observed for the simplest models, namely ecoinvent, AGRIBALYSE and MFE. This shows the limits of approaches based on an emission coefficient function depending on climatic conditions, be it a dry/wet differentiation (AGRIBALYSE), a single regression equation (ecoinvent), or even a more elaborated approach using correction factors (MFE). The approach implemented in the Indigo-N model deserves more attention since it considers processes, though in a simplified way. It computes post-fertilisation leaching and post-harvest leaching (associated with draining events, which under temperate conditions correspond to the winter period). For post-fertilisation leaching, it combines Burns leaching coefficients with a correction factor that associates the timing of fertilisation with that of maximum N uptake by crops, according with plant uptake curves for different crops (from literature). For post-harvest leaching, it combines Burns leaching coefficients with post-harvest N-balances, which take into consideration mineralisation of added organic matter (crop residues, organic fertilisers), mineralisation of SOM, intermediate crops, mineral N inputs, and increased N losses due to over-fertilisation. This last part was inspired from the COMIFER approach behind the AGRIBALYSE model for temperate situations (COMIFER, 2013; Taureau et al., 1996).

Such a formalism, despite being originally designed for field crops in temperate climate only, allows a great flexibility for representing different drainage regimes (e.g. winter rains in temperate climates, rainy seasons in tropical climates) and agricultural systems featuring different cycle lengths (e.g. vegetable cycles of <2 months vs. fruit tree cycles of several years). An adaptation and enhancement of this approach, suitable for contrasting agricultural conditions, would compute leaching coefficients associated with drainage regimes and soil characteristics, along the duration of a crop or crop rotation. Mineralisation of organic nitrogen from input and its cumulative effects should be better represented without demanding additional data.

4 Conclusion

A set of operational models across the N modelling continuum used in LCA to assess impacts due to nitrogen losses were compared each other and to a complex model integrating a lot of processes like STICS. The theoretical analysis and their implementation on four very contrasted sites, temperate and tropical showed several shortcomings of such models. Although the comparison was limited to four sites with two fertiliser regimes, the contrasted situations and especially the implementation under tropical conditions, made possible to highlight important discrepancies among models highlighting their limitations. For nitrate leaching, especially, models based on simplification excluding major drivers, e.g. using emission coefficients or regression equations failed (and in general, fail) to yield sound results. This can be explained by their limitations, including the poor integration of organic fertiliser and crop residues, the lack of consideration of some processes like N_2 emissions, fertiliser surplus, etc. We provide recommendations for building a model that more accurately represents the mode of action of organic fertilisers and considers the pedo-climatic conditions prevalent beyond temperate conditions. The approach of the Indigo-N model, designed for temperate conditions, could be a solid basis for the perspective when developing a model which would implement the recommendations' derived from our model analysis and comparison. Lastly, we compared here four LCA models and an agri-environmental model. One step further would be to integrate into the comparison more simple models such as those listed the review by Buczko and Kuchenbuch (2010).

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Figure captions

Fig. 1. Modelling continuum for estimation of N emissions in the French LCA context

Fig. 2. Estimation of N gaseous direct field emissions across sites and models: A) fertilisation treatments dominated by organic inputs, B) mineral fertilisation treatments equivalent to organic ones; 1) ammonia, 2) nitrous oxide, 3) nitrogen oxide ($NO + NO_2$)

Fig. 3. Estimation of nitrate direct field emissions across sites and models: A) fertilisation treatments dominated by organic inputs, B) mineral fertilisation treatments equivalent to organic ones. Reference values based on averaged lysimetric measurements

Fig. 4. Sensitivity of ecoinvent and Indigo-N models to a 10% change in precipitation, irrigation and drainage parameters affecting NO_3 leaching predictions for A) fertilisation treatments dominated by organic inputs, B) mineral fertilisation treatments equivalent to organic ones