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Improving bioenergy sustainability evaluations by using Soil Nitrogen Balance coupled with Life Cycle Assessment: A case study for electricity generated from rye biomass.

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Abstract

The use of Life Cycle Assessment (LCA) as an environmental tool to evaluate the sustainability of different bioenergy pathways has become a common practice since the European Renewable Energy Directive was published in 2009. In the evaluation of bioenergy produced out from dedicated energy crops, nitrogen fertilizer production and use are commonly identified as the most important contributors to fossil energy consumption and to several environmental impacts categories including Global Warming Potential. In considering the impacts produced by the nitrogen fertilization of energy crops and in addition to the effects of fertilization schemes on the biomass yield, more attention should be paid to the changes in soil nitrogen to know if fertilization doses and application schemes are sufficient enough to maintain soil nitrogen stocks and ensure that soil quality is preserved for future years. To this aim, in this work soil nitrogen balance is used as an indicator to estimate the evolution of soil nitrogen stocks and complement LCA calculations. In this paper, the effects of three nitrogen top fertilization doses (null, 30 and 80 kg N/(ha·y)) used for rye cultivation are compared when ry is grown as a dedicated energy crop for electricity generation under the Spanish province of Soria conditions. A LCA was carried out using experimental crop testing results and a centralised (25MWe) straw power plant data in combination with soil nitrogen balance obtained in each of the experimental crop trials. After that, the LCA results were compared with those obtained when electricity is generated from natural gas in Spanish power plants. According to the average calculations, each additional kg N/(ha·y) applied in top fertilization produces a reduction of 0.18% on GHG savings with respect to natural gas electricity, as well as a worsening in the energy balance of

0.00084 TJ fossil energy per TJ of electricity generated but reduces soil nitrogen deficit in 0.43 kg N/(ha·y). For top fertilization doses of 80 kg N/(ha·y) the average GHG savings with respect to natural gas were 63.7% and the average non-renewable energy consumption was 6,4 times less for the bioenergy system than for natural gas. Fossil energy accounted for more than 95% of total non-renewable energy in this calculation. This work evidences that determinate biomass growing conditions associated to high GHG savings and improved energy balances may cause detrimental effects for soil fertility due to considerable associated negative soil nitrogen balances. This finding suggests the convenience to include the soil nitrogen balance as a complementary indicator for bioenergy LCA calculations.

Keywords: bioenergy; sustainability; life cycle assessment (LCA); soil nitrogen balance; fertilizers; energy crops.

Nomenclature

Variables

Symbols	Description	Units
E	electrical energy generated.	MJe/(ha·y)
RY	rye whole plant yield at 0% humidity.	kg crop/(ha·y)
H	humidity percentage of the biomass.	kg water/kg crop
SL	bales storage loses.	dimensionless
$NHV_{CP,H}$	net heating value of rye at constant pressure and at H% humidity.	MJ/kg
η	conversion efficiency of the biomass power plant.	MJe/MJ crop
$NHV_{CP,0}$	net heating value of rye at constant pressure and at 0% humidity.	MJ/kg
AM	amount of machinery.	kg/(ha·y)
W	weight of the machinery.	kg
OR	operating rate.	h/(ha·y)
LT	lifetime of the machinery.	h
N_2O	emissions of N_2O to the air.	kg N_2O/(ha·y)
N_{tot}	total nitrogen input from fertilizers.	kg N/(ha·y)
N_{cr}	nitrogen contained in crop residues.	kg N/(ha·y)
$N\text{-}NH_3$	losses of N in form of NH_3.	kg $N\text{-}NH_3$/(ha·y)
$N\text{-}NO_3^-$	losses of N in form of NO_3^-.	kg $N\text{-}NO_3^-$/(ha·y)
P	precipitation plus irrigation if it exits.	mm/y
c	clay percentage of the soil.	%
L	root depth of the crop.	m
S	total nitrogen supply.	kg N/(ha·y)
N_{org}	nitrogen content of soil organic matter.	kg N/ha
U	nitrogen uptake by the crop.	kg N/(ha·y)
N_Er	nitrogen losses by soil erosion that reach surface water..	kg N/(ha·y)
S_{er}	quantity of soil eroded.	kg soil/(ha·y)
N_{es}	nitrogen content in top soil.	kg N/kg soil
P_{ro}	phosphorus emitted through run-off to rivers.	kg P/(ha·y)
P_{rol}	quantity of P lost through run-off for a land use category.	kg P/(ha·y)
F_{ro}	correction factor for fertilization with phosphorus.	dimensionless
P_2O_{5min}	quantity of P_2O_{5min} contained in mineral fertilizers.	kg P_2O_5/(ha·y)
P_{er}	phosphorus emitted through erosion to rivers.	kg P/(ha·y)
P_{es}	phosphorus content in top soil.	kg P/kg soil
$M_{leach\ i}$	agricultural drainage related emission of the heavy metal i.	mg metal/(ha·y)
$m_{leach\ i}$	average amount of leaching of heavy metal i.	mg metal/(ha·y)
A_i	allocation factor heavy metal i.	dimensionless
$M_{agro\ i}$	input of heavy metal i from agricultural production.	mg metal/(ha·y)
$M_{deposition\ i}$	input of heavy metal from atmospheric deposition.	mg metal/(ha·y)
$M_{erosion\ i}$	heavy metal i emissions to surface water through erosion.	mg metal/(ha·y)
$C_{tot\ i}$	heavy metal i content of soil.	mg metal/ kg soil
$M_{soil\ i}$	change in the content of metal i in the soil due to the agricultural system.	mg metal/(ha·y)
$Inputs_i$	total input of heavy metal i to the agricultural soil.	mg metal/(ha·y)

$Outputs_i$	total output of heavy metal i from agricultural soil.	mg metal/(ha·y)
SNB	soil nitrogen balance.	kg N/(ha·y)
N_Fert	nitrogen provided by fertilizers.	kg N/(ha·y)
N_Seed	nitrogen provided by sowing seed.	kg N/(ha·y)
N_AtDep	nitrogen provided by atmospheric deposition.	kg N/(ha·y)
N_FrLiv	nitrogen fixed by the effect of soil free living organisms.	kg N/(ha·y)
N_BioFix	nitrogen symbiotic biological fixation of legumes.	kg N/(ha·y)
N_HarvEx	nitrogen exported by the harvest.	kg N/(ha·y)
$N\_NO_3^-$	nitrogen losses by leaching in the form of NO_3^-.	kg N/(ha·y)
N_NH_3	nitrogen losses by volatilization in the form of ammonia.	kg N/(ha·y)
$N\_N_2O_{cr+Fert}$	nitrogen loses due to N coming from fertilizers and crop residues an emitted as N_2O.	kg N/(ha·y)
N_NO_x	nitrogen loses due nitrogen oxides released during the denitrification.	kg N/(ha·y)

Parameters

Symbols	Description	Units
EF_1	IPCC factor 1, express the fraction of N from inputs that is emitted as N in form of N_2O.	kg N-N_2O/kg N inputs
EF_4	IPCC factor 4, express the fraction of N in form of NH_3 that is converted in N in form of N_2O.	kg N-N_2O/kg N-NH_3
EF_5	IPCC factor 5, express the fraction of N in form of NO_3^- leached and emitted as N in form of N_2O.	kg N-N_2O/kg NO_3^-
F_{rn}	enrichment factor for nitrogen.	dimensionless
F_{erw}	fraction of eroded soil that reaches the rivers.	dimensionless
F_{rp}	enrichment factor for phosphorus.	dimensionless
F_{rh}	enrichment factor for heavy metals.	dimensionless

1. Introduction

The sustainable production of domestic biomass is a key issue to support the development of Bioeconomy and the Set Plan (Strategic Energy Research Plan), the two basic pillars for implementation of a low carbon economy in the EU objectives [1]. In this context, and in view of the limited residual biomass availability, the sustainability of the biomass value chains [2] from dedicated crops grown in the EU territory for the production of energy and bioproducts is being intensively studied in order to determine the real potential of this source of biomass [3] to supply the foreseen growing demand of those industries.

LCA [4] is the environmental tool designed by the Renewables Energy Directive (RED) in 2009 [5] to evaluate the sustainability of transportation biofuels value chains. The binding sustainability criteria established from 2018 onwards by the RED is the achievement of a percentage of greenhouse gases (GHG) emissions savings of at least 60 % with respect to the fossil energy of reference. The same sustainability criteria has been proposed as a strong recommendation for biomass devoted to electricity, heating and cooling [6]. Since RED was put in force, potential GHG savings of different pathways to produce bioenergy for transportation [7-10] and to generate electricity and heat [11-14] are being evaluated intensively [15,16]. Most of previous assessments point out the crucial importance of fertilizers and their associated emissions for energy balances, GHG emissions and other environmental impacts caused by energy crops value chains. As a result, many works deal with the role of fertilizers in bioenergy sustainability assessments [17-23]. In these studies very scarce attention is paid to the effects that the crops biomass production and its exportation may have on the soil nitrogen balance. However, this parameter [17,24] can be an important sustainability indicator due to its relation to soil fertility and the fertilization requirements of further crops.

Nitrogen balance is commonly used in the analysis of agronomic systems [25]. Nevertheless, the authors have not found any reference in which it is linked to bioenergy LCAs. There are some references that analyse the nitrogen use efficiency of different bioenergy options [20,26,27] or the effect on the global warming potential of using different nitrogen fertilizers [19], but none of them tries to clarify whether the use of minor fertilizations doses in energy crops results in a deficit of nitrogen in the soil or not. Consequently, there is a compromise to be assessed between these possible soil nitrogen deficits, and the expected lower GHG emissions and improved energy balances obtained when lower fertilization doses are applied to bioenergy crops.

The objective of this study is to assess the energy balance and environmental impacts associated to the production of electricity from dedicated rye biomass grown under different nitrogen fertilization doses, utilizing the soil nitrogen balance in conjunction with current LCA to provide a better and more holistic approach to the environmental sustainability analysis [28]. The biomass production conditions are referred to those of Soria province, situated in an extensive cereal production area in central-northern Spain (Castilla y León Region). In this Region, and due to present grain market prizes and the pedo-climatic conditions of the Region, the profitability of the production of grain for food and feed uses is small and alternative uses for the biomass cropped need to be explored in order to increase the farmers’ income. Therefore, the aerial part of rye crop produced in the considered area has been selected in this study due to the satisfactory sustainability results as a fuel for electricity production obtained in previous study [12].

The systems analysed consider data concerning biomass production, as well as soil and biomass characterization obtained in an experimental parcel. Relevant data of straw bales transportation and transformation into electricity in a 25 MWe biomass power plant located in northern Spain have been taken into account as a reference to evaluate the impacts of rye biomass conversion. The results of the LCAs were compared to those of electricity produced from natural gas under Spanish conditions. The percentage of GHG savings with respect to natural gas were calculated to evaluate the accomplishment of EU sustainability criteria for biomass electricity [6] and were confronted with associated soil nitrogen balances. Primary energy consumptions and other environmental impacts such as eutrophication, acidification, among others, were also evaluated due to its importance to bioenergy sustainability assessments. As fossil energy is the most important source of non-renewable energy, fossil primary energy consumptions were confronted as well with associated soil nitrogen balances in other to establish possible relations.

2. Experimental design: plots, soils and biomass yields and characterization

In order to obtain the field data for the LCAs inventories and nitrogen balances, different fertilization experimental tests were carried out on a plot of a total surface of 8500 m^2 where rye was established as winter cereal during two consecutive campaigns, in the period 2011-2012. This plot was located in the province of Soria. The fertilization trials were carried out in strips of 800-900 m^2 using commercial machinery and typical management farmers' techniques for all fieldworks except for top fertilization where two additional doses of calcium ammonium nitrate 27% N in addition to the typical one used by farmers (80 kg N/(ha·year)) were utilized, namely: low dose (30 kg N/(ha·year)) and null (0 kg N/(ha·year)). Three trials were performed for every fertilization dose and each one was established in a portion of the cited parcel with soil of different characteristics (S1, S2 and S3). All the details about the experimental design and the plot pedo-climatic conditions are shown in **Table 1**.

Table 1
Experimental design summary

1. Location	Soria		
Coordinates	41º 36’ 40.0” N 2º 28’ 55.6” W		
Altitude	1035 m		
2. Climate	Continental Mediterranean with cold winters		
3. Genotype			
Species (variety)	Secale Cereale (Petkus)		
4. Plots			
Quantity / years / type / size	9 / 2 / Strips / 0.04-0.05 ha		
5. Experimental period	Year 1 (Y1)	Year 2 (Y2)	
Duration	09/2010 to 06/2011	09/2011 to 06/2012	
Average Temperature	10.3 ºC	11.0 ºC	
Total rainfall	447 mm	293 mm	
6. Crop management practices	Y1	Y2	
Seeding dose (kg/(ha·y))	Rye (120)	Rye (120)	
Base Fertilization (kg /(ha·y))	NPK 8-24-8 (300)	NPK 8-24-8 (300)	
Top Fertilization (kg N /(ha·y))	Calcium Ammonium Nitrate 27% (0/30/80)	Calcium Ammonium Nitrate 27% (0/30/80)	
Herbicides (kg/(ha·y))	Dicamba (0.125) 2,4-D (0.370)	No treatments were applied	
7. Soil type	Soil 1 (S1)	Soil 1 (S2)	Soil 3 (S3)
Texture	Sandy	Sandy	Sandy loam
Clay (%) / Sand (%) / Silt (%)	8/84/8	4/84/12	12/76/12
pH	7.19	6.85	7.13
Organic matter (%)	0.54	0.84	1.3
Nitrogen (%)	0.050	0.070	0.100
Total Phosphorus (%)	0.0058	0.0103	0.0187

When rye was harvested the biomass productivity was measured for each trial and biomass sampling and characterization were performed too. The proportions between the harvested biomass, the stubble and the roots were also measured, and the differences between their respective carbon and nitrogen contents were determined. It was found that stubble and roots masses were 13 % and 9.1 % of the harvest yield, respectively. There were no remarkable differences between harvest and stubble compositions, and the carbon content (%) in aerial biomass (harvested biomass + stubble) was approximately the same as for roots, while nitrogen content was on average 53% higher for roots than for shoots. The Table 2 shows a detailed summary about biomass characterization and productivity results of the rye trials, also including information about biomass yield and the heating value of the biomass referred to dry biomass and to 12% humidity content, which is the reference value for the water content of the biomass bales which feed the biomass power plant (see later Section 3.3.1). A detailed biomass assessment of the combustion quality properties of this biomass has been reported elsewhere in the literature [29].

Table 2
Trials productivity and biomass composition

Trial	**Year**	**Top Fertilization Dose (kg/ha)**	**Soil**	**Harvest yield (kg,db[a]/ha)**	**N in aerial biomass (%)**	**N in roots (%)**	**C in aerial biomass & roots (%)**	**NHV$_{cp,0}$[b] (MJ/kg, db[a])**	**NHV$_{cp,12}$[c] (MJ/kg, wb[d])**
1	Y1	0	S1	7092	0.57	0.87	44.9	16.68	14.39
2	Y1	0	S2	10142	0.90	1.38	45.2	16.85	14.53
3	Y1	0	S3	9001	1.15	1.76	44.4	16.57	14.29
4	Y1	30	S1	8182	0.82	1.26	44.6	16.57	14.29
5	Y1	30	S2	10442	1.01	1.55	45.0	16.79	14.48
6	Y1	30	S3	10792	0.86	1.32	45.6	16.93	14.61
7	Y1	80	S1	10548	1.00	1.53	45.6	16.97	14.64
8	Y1	80	S2	11815	0.83	1.27	45.4	16.88	14.56
9	Y1	80	S3	13200	0.87	1.33	45.3	16.85	14.53
10	Y2	0	S1	4416	0.68	1.04	45.6	16.82	14.51
11	Y2	0	S2	5028	0.81	1.24	46.0	16.94	14.61
12	Y2	0	S3	5875	0.81	1.24	45.5	16.81	14.50
13	Y2	30	S1	6349	0.73	1.12	45.7	16.84	14.53
14	Y2	30	S2	6024	0.80	1.23	45.9	16.93	14.61
15	Y2	30	S3	6091	0.91	1.39	45.8	16.87	14.55
16	Y2	80	S1	7577	1.29	1.98	46.0	16.92	14.60
17	Y2	80	S2	8298	1.24	1.90	45.2	16.96	14.63
18	Y2	80	S3	6452	1.07	1.64	45.7	16.88	14.56

[a] Dry basis
[b] Net heating value at constant pressure in dry basis
[c] Net heating value at constant pressure and 12 % moisture content
[d] Wet basis

3 Life cycle assessment methodology

3.1. Goal, scope and evaluation of data sources and tools

LCA was selected as the environmental assessment tool to determine the energetic and environmental performance of rye to be used to generate electrical energy and to compare it with a fossil reference energy system.

LCA involves a systematic set of procedures for compiling and examining the inputs and outputs of materials and energy and the associated environmental impacts directly attributable to the functioning of a product or service system throughout its life cycle [30]. This environmental management tool is regulated by ISO 14040 [30] and ISO 14044 [31] standards, and according to them, LCAs should follow four steps: (1) goal and definition, (2) inventory analysis, (3) impact assessment and (4) interpretation.

All the information concerning the biomass production system has been obtained from real plots (see **Table 1**), whilst data on biomass conversion have been obtained from an existing 25 MWe located in Northern Spain.

The generation of electricity from natural gas has been chosen as the reference system with the purpose of making comparisons for being one of the cleanest fossil energy sources for electricity generation. Average data of efficiency and emissions of Spanish natural gas power plants were taken as a reference [32].

Simapro 8.1.0.60 software tool and Ecoinvent 3.1 [33] European database have been used to conduct the LCAs in this study. The recycled version of Ecoinvent 3.1 database in Simapro has been selected for being the alternative which more closely resembles Ecoinvent 2.

3.2. Functional unit

The functional unit chosen for this biomass system is the generation of 1 TJ of electrical energy (TJe) from rye cultivated in Soria and burned in a 25 MWe Spanish straw power plant. The generation of 1 TJe from natural gas in Spanish conditions was selected as the functional unit for the reference fossil system.

3.3 Systems description

The characteristics and burdens of the bioenergy system and the natural gas system are described in this section. Natural gas Spanish electricity was chosen as the reference system for comparisons because it is the cleanest fossil energy source available.

3.3.1 Bioenergy system

The bioenergy system is composed of three subsystems: rye production (agricultural system), biomass power plant and transport that are going to be explained in detail. **Fig. 1** summarises the processes included in the bioenergy system.

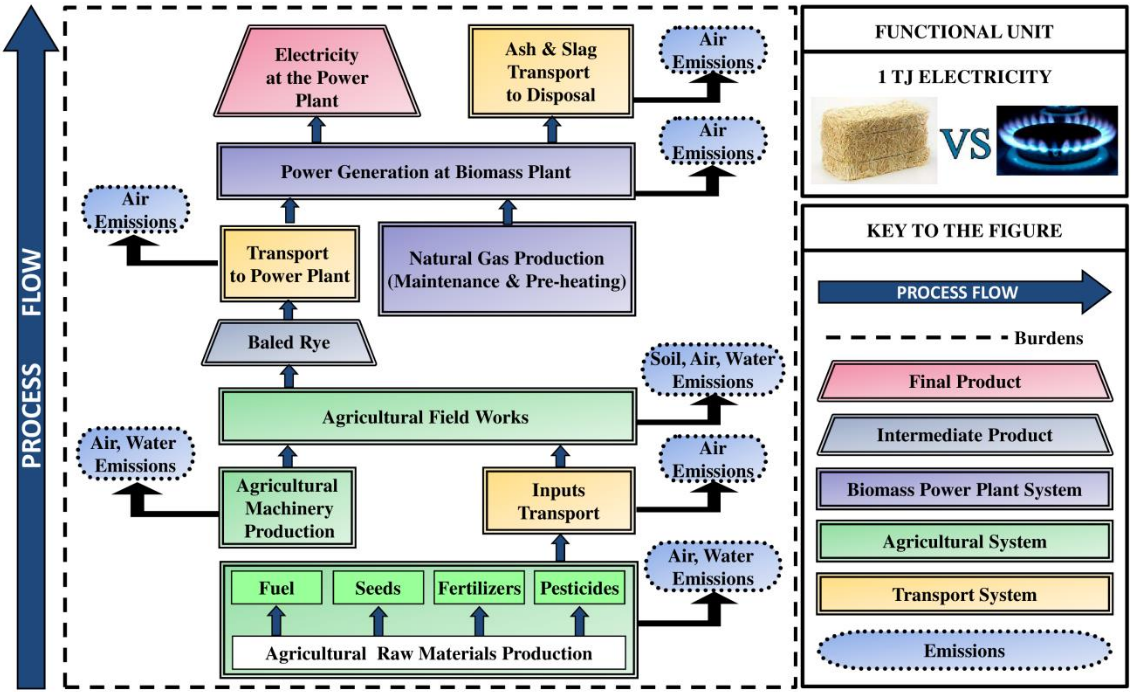

Fig. 1 Bioenergy system burdens and phases included in the analysis.

(1) Rye biomass production subsystem: This phase was defined by the crop management practices followed, the machinery used as well as the agricultural raw materials consumed (seeds, fertilizers, and pesticides). The **Table 3** shows a detailed summary of the field operations performed for rye cultivation. The fuel consumption of some field operations depends on plot productivity. The maximum, minimum and the average consumption (in parentheses) are shown in **Table 3**.

Table 3
Operations performed for rye cultivation.

Operation	Tractor		Implement			
	Weight (kg)	Power (kW)	Type	Weight (kg)	Operating rate (h/ha)	Fuel consumption (L/ha)
Primary tillage	5470	103	Plough	1390	1.00	20
Secondary tillage	5470	103	Harrow	400	0.66	10
Base fertilization	3914	66	Spreader	110	0.20	4
Sowing	5470	103	Seeder	830	0.60	8
Herbicide treatment	3914	66	Boom sprayer	230	0.50	4
Top fertilization[a]	3914	66	Spreader	110	0.20	4
Rolling	3914	66	Roller	1000	0.40	8
Mowing-Swathing[b]	3914	66	Mower	150	1.89-1.16 (1.49)[c]	15.77-9.68 (12.42)[c]
Baling[b]	9000	144	Large square baler	1700	1.33-0.82 (1.05)[c]	26.51-16.43 (20.96)[c]
Automatic bale loading	5470	103	Atomatic bale loader trailer	2500	0.48	10.9
Bale loading to lorry	5470	103	Forklift	1870	0.40	4

[a] Top fertilization was not performed for trials with 0 kg of nitrogen.
[b] Field work fuel consumption and operating rates depend on the harvest yield.
[c] Maximum-Minimum (Average) values.

(2) Biomass power plant subsystem.

This system was modelled using real data supplied by officers of an existing 25 MWe Spanish straw power. This plant is considered representative of other existing biomass power plants in Spain. The plant consumes biomass at average humidity of 12 % and produces electricity with an average efficiency of 29 %. The biomass power plant consumes small amounts of natural gas in start-up and pre-heating and generates ashes and slags as residues of straw burning. Natural gas average consumption and ash and slag average generation per kg from straw burned are shown in **Table 4**. Aerial emissions are submitted online to regional authorities and its average values are presented in **Table 4** as well**.** Fossil carbon dioxide emissions of natural gas burning are taken into account. The emissions of carbon dioxide from straw combustion have not been accounted for because CO_2 was previously fixed from the air by the crop no more than one year before being burned.

Table 4
Biomass power plant consumptions, residues and emissions

Items	Type	Amount	Units
Natural gas	Consumption	0.0389	MJ/kg dry biomass
Slags	Residue	93.72	g/kg dry biomass
Ashes	Residue	9.38	g/kg dry biomass
Carbon Dioxide from natural gas combustion	Emission	2.16	g/kg dry biomass
Nitrogen oxides	Emission	1.85	g/kg dry biomass
Carbon monoxide	Emission	1.05	g/kg dry biomass
Sulphur dioxide	Emission	0.36	g/kg dry biomass
Particulate matter	Emission	0.27	g/kg dry biomass

(3) Transport subsystem.

A summary of the elements considered in the transport system is shown in **Table 5**. The information shown includes the materials to be transported, origin and destination points, distances and means of transport. Transport distances and transport means of agricultural inputs until regional storehouses are considered as in Ecoinvent [34]. A distance of 10 km was assumed as a good estimate of the average transport distance for agricultural inputs from the storehouse to farmers´ plots. The average transport distances and transport means for rye bales, ashes and slags were provided by officers in charge of the power plant.

Table 5
Transport system characteristics

Material	From	To	Distance	Vehicle
Seed	Field	Processing center	30 km	Lorry 16-32t
	Processing center	Regional storehouse	100 km	Lorry 16-32t
	Regional storehouse	Demostration plot	10 km	Tractor and Trailer
Fertilizers and pesticides	Manufacturer	Regional storehouse	600 km 100 km	Train Lorry >16t
	Regional storehouse	Demostration plot	10 km	Tractor and Trailer
Rye bales	Demostration plot	Biomass plant	60 km	Lorry 16-32t
Ash and slag	Biomass power plant	Disposal site	37 km	Lorry 16-32t

The output of the whole bioenergy system is the electrical energy generated. This energy output was calculated as shown in Eq. **(A.1)**.

3.3.2 Natural Gas System

The natural gas system represents the generation of electricity in Spanish power plants fuelled with natural gas. The system takes into account the exportation of natural gas to Spain from main exporter countries (Algeria 73% and Norway 27%), including the gas field operations for the extraction, losses, emissions, and purification. The long distance transport to Spain as well as the inland delivery to the power plant is considered, including the energy consumption, losses and emissions. Average inputs needed for Spanish natural gas power plants, as well as transformation efficiencies and emissions are inventoried [32].

3.4. Life cycle inventory structure

The inventories used to consider natural gas consumption of the biomass power plant and the transportation of agricultural inputs, biomass and power plant residues are taken from Ecoinvent v3.1. The methods used for the inventory analysis of the agricultural system mainly follow those proposed in the life cycle inventories of agricultural production systems [34] and they are updated following a new method for field emissions [35].

3.4.1 Fertilizer production

The inventories for fertilizer production include the consumption and transport of raw materials and intermediate products as well as the energy consumption and the emissions generated in the production processes [34].

3.4.2 Pesticide production

The inventory data of the emissions, energy and substance consumption in the production of the herbicides sprayed is retrieved from Ecoinvent [36]. The active matter substances contents (see **Table 1**) are taken from the commercial herbicide formulations used.

3.4.3 Seed production

The seed production system was modelled as described in Sastre et al [12] for non-hybrid rye. The management techniques, machinery and input consumptions considered were similar to the ones described in this study for typical fertilization doses. The seed production yield was 5.500 kg/ha. The transports for the seed production system are shown in **Table 5**. The energy consumption for drying, cleaning, seed dressing, and bag filling of the cereal seed in the processing plant has been estimated in 0.0328 kWh/kg [37].

3.4.4 Diesel and motor oil consumption and combustion emissions of agricultural machinery

The diesel consumption of agricultural machinery for the trials is shown by field operation performed in **Table 3**. According to the Spanish platform of agricultural machinery [38], the consumption of motor oil for tractors is 1 % of the diesel consumption. The inventories for the extraction, the transportation of crude oil, its transformation into diesel, and its distribution are taken from Ecoinvent [39]. The agricultural machinery exhaust emissions are also taken into account [40].

3.4.5 Agricultural machinery manufacture

The inventories for agricultural machinery manufacture are specific to the different types of machinery (tractors, harvesters, tillage implements or other implements) [34].

The amount of machinery consumed for carrying out a specific agricultural operation was calculated as shown in Eq. **(A.3)**.

3.4.6 Field and fertilizer nitrogen derived emissions

There are several nitrogen derived emissions that affect GWP among other impact categories.Nitrous oxide (N_2O), nitrogen oxides (NO_x), ammonia (NH_3) emissions to the air, nitrate (NO_3^-) leaching to ground water and the nitrogen emission from eroded particles that reach the surface water were accounted in this study. This section describes the methods used for inventorying each type of emission.

The calculation of the nitrous oxide emissions (N_2O) is shown in Eq. **(A.4)** and follows the methodology proposed by the RSB [41] and Nemecek et Kägi [35].

During the denitrification process nitrogen oxides (NO_x) are produced in soil. These emissions were estimated in a 21 % of nitrous oxide (N_2O) emissions (see Eq. **(A.4)**). They are produced in a parallel process and should not be discounted from N_2O emissions.

The ammonia emissions (NH_3) due to the application of mineral fertilizers are calculated by a constant emissions factor for each group of fertiliser. A 2 % of the nitrogen content of calcium ammonium nitrate and 4 % of the nitrogen in multinutrient fertilizer are emitted in the form of ammonia.

The calculation of nitrate emissions follows the methodology proposed by the RSB [41] which is also proposed by Nemecek [35]. This calculation is shown in Eq. **(A.5).**

In order to be consistent with the calculation of phosphorus water emissions (see **3.4.7**), the emissions of nitrogen to surface water due to soil erosion were accounted for as well. This calculation is shown in Eq. **(A.6).**

3.4.7 Emissions of phosphorus to the water

Phosphorus is an essential nutrient for crops that must be supplied to them in sufficient quantities. However, a portion of this phosphorus is not used by the crops and reaches the water generating environmental impacts. Three different types of phosphorus emissions to water are distinguish in this study: leaching of phosphates to ground water, run–off of phosphates to surface water and eroded soil particles to surface water.

Phosphate leaching to ground water was calculated as an average quantity of phosphates leached for a land use category. An average emissions value of 0.07 kg P/(ha·y) was considered appropriate for arable land in this study.

Run–off of phosphorus to surface water was calculated as an average corrected by phosphorus fertilization as shown in Eq. **(A.7)**.

Emissions of phosphorus from eroded soil particles that reach surface water was calculated as shown in Eq. **(A.8)**.

3.4.8 Emissions of heavy metals to agricultural soil, surface water and ground water

The following seven heavy metal are selected to inventory their emissions due to damage they cause to agricultural ecosystems: Cadmium (Cd), Chromium (Cr), Copper (Cu), Lead (Pb), Mercury (Hg), Nickel (Ni) and Zinc (Zn). The model estimates the emissions of heavy metal due to drainage to the ground and the surface water, erosion of soil particles that reach surface water and the balance of inputs and outputs of heavy metals in the agricultural soil. Drainage emissions of heavy metal emissions are calculated with constant leaching rates as shown in Eq. **(A.9)**.

The emissions of heavy metals to the surface water due to erosion are calculated in a similar manner to phosphorus. This calculation is shown in Eq. **(A.11)**.

The balance between inputs and outputs of heavy metals provides the changes of each heavy metal in the soil due to the cultivation of rye as shown in Eq. **(A.12)**.

3.4.9 Emissions of pesticides to agricultural soil

All active matters of pesticides applied for crop protection are assumed to end up as emissions to the soil. The amount of active matter of the applied pesticides is simultaneously considered as input and as output in the form of emissions to agricultural soil.

3.5. Life cycle impact assessment

In the Life Cycle Impact Assessment (LCIA) phase of an LCA the inputs and outputs of elementary flows that have been collected and reported in the inventory are translated into impact indicator results [42]. LCIA includes mandatory and optional steps. Mandatory steps of classification and characterization have been performed and optional steps of normalization and weighing have been avoided.

3.5.1. Environmental impact assessment methods

The impact assessment method chosen to evaluate the GWP was the 2013 version of the IPCC [43] for 100 years' time horizon. This method calculates the cumulative radiative forcing caused by a unit mass emission of a GHG, integrated over a 100 year time horizon, as compared with the cumulative radiative forcing due to emission of a unit mass of carbon dioxide (CO_2) over the same time horizon. The CML method [44] has been also chosen to evaluate the effects of the systems on other important impacts categories. The version used was the baseline version 4.2 released by de CML in April 2013. This method included the following categories: abiotic depletion (ADep), abiotic depletion based on fossil fuels (ADep FF), global warming potential 100 years IPCC 2007 (GWP 100y), ozone layer depletion (OLDP), human toxicity (HuTx), fresh water ecotoxicity (FWAEtx), marine aquatic ecotoxicity (MAEtx), terrestrial ecotoxicity (TeEtx), photochemical oxidation (PhoChOx), acidification (Ac) and eutrophication (Eutro).

3.5.2. Energy assessment method

Cumulative Energy Requirement Analysis (CERA) [45] was the method chosen to assess the energy consumed to generate electricity from rye biomass and from natural gas. This method aims to calculate the energy use throughout the life cycle of a good or service. It included the following primary energy types: non-renewable fossil (NR Fossil), non-renewable nuclear (NR Nuclear), non-renewable biomass (NR Biomass), renewable biomass (R Biomass), renewable solar, wind, geothermic, etc. (R Others) and renewable hydraulic (R Water).

4 Soil nitrogen balance methodology

To perform the soil nitrogen balance methodology several references have been taken into account [17,24] and several adjustments have been performed to make it coherent with LCA methodology, in particular with inventories for field and fertilizer nitrogen derived emissions (see point **3.4.6.).**

The methodology for soil nitrogen balance is summarized in **Fig. 2**. The balance is calculated by accounting for all the different outputs and inputs of the system as shown in Eq. **(A.13)**.

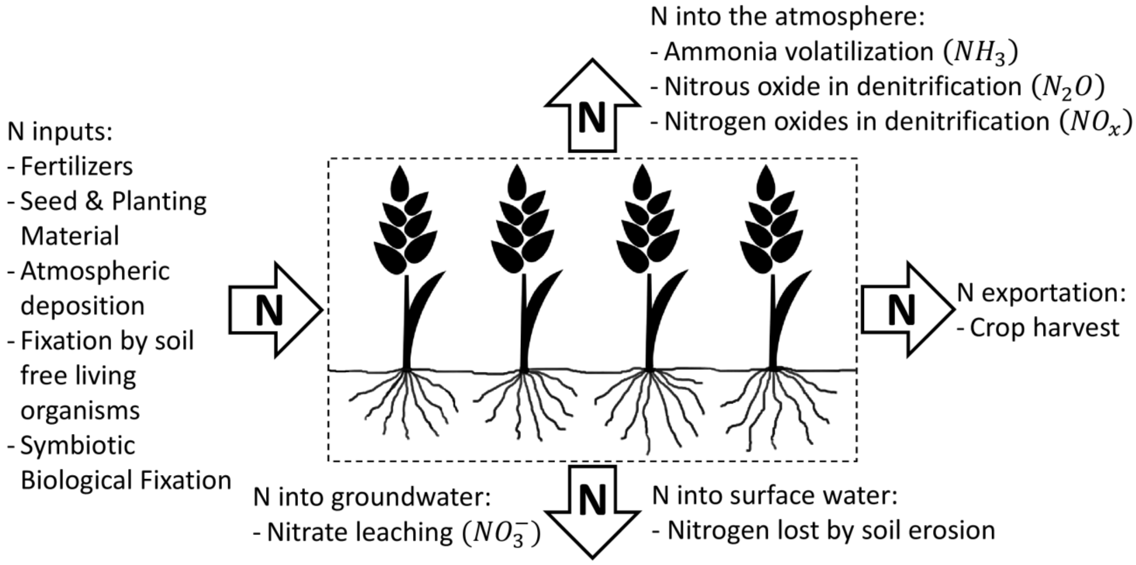

Fig. 2. Inputs and outputs considered in the soil nitrogen balance.

5 Results

5.1 Soil Nitrogen Balance

The results of the soil nitrogen balance are shown in **Table 6.** It can be seen that soil nitrogen balances were negative for most of the trials. The nitrogen deficits were in general higher for null and low nitrogen doses and for the first cropping year compared to the second one. Nitrogen inputs were mainly due to N provided by fertilizers while outputs were dominated by N exported with the harvest although N lost in form of nitrates had also a remarkable importance.

Table 6
Soil nitrogen balance results

Trial	Code [a]	Soil Nitrogen Balance (kg N/(ha·y))	Inputs (kg N/(ha·y)) [b]				Outputs (kg N/(ha·y)) [b]					
			N_Fert	N_Seed	N_AtDep	N_FrLiv	N_HarvEx	N_NO$_{-3}$	N_Er	N_NH$_3$	N_N$_2$O$_{Cr+Fert}$	N_NO$_x$
1	Y1_0_S1	-32.27	24	1.91	7	3	40.42	24.29	1.98	0.96	0.35	0.18
2	Y1_0_S2	-72.87	24	1.91	7	3	91.28	13.07	2.78	0.96	0.49	0.20
3	Y1_0_S3	-93.10	24	1.91	7	3	103.51	19.83	3.97	0.96	0.52	0.22
4	Y1_30_S1	-29.54	54	1.91	7	3	67.09	23.80	1.98	1.56	0.72	0.30
5	Y1_30_S2	-61.98	54	1.91	7	3	105.46	16.95	2.78	1.56	0.82	0.32
6	Y1_30_S3	-57.89	54	1.91	7	3	92.81	24.34	3.97	1.56	0.79	0.33
7	Y1_80_S1	-20.11	104	1.91	7	3	105.48	24.17	1.98	2.56	1.32	0.51
8	Y1_80_S2	-24.79	104	1.91	7	3	98.06	35.47	2.78	2.56	1.30	0.53
9	Y1_80_S3	-34.03	104	1.91	7	3	114.84	26.70	3.97	2.56	1.35	0.52
10	Y2_0_S1	-22.16	24	1.91	7	3	30.03	24.61	1.98	0.96	0.32	0.17
11	Y2_0_S2	-37.91	24	1.91	7	3	40.73	28.81	2.78	0.96	0.35	0.19
12	Y2_0_S3	-42.28	24	1.91	7	3	47.59	25.11	3.97	0.96	0.37	0.19
13	Y2_30_S1	-10.54	54	1.91	7	3	46.35	25.61	1.98	1.56	0.66	0.29
14	Y2_30_S2	-20.66	54	1.91	7	3	48.19	33.06	2.78	1.56	0.67	0.31
15	Y2_30_S3	-22.53	54	1.91	7	3	55.43	26.49	3.97	1.56	0.69	0.30
16	Y2_80_S1	-12.36	104	1.91	7	3	97.74	24.19	1.98	2.56	1.30	0.50
17	Y2_80_S2	-23.55	104	1.91	7	3	102.90	29.38	2.78	2.56	1.32	0.52
18	Y2_80_S3	9.87	104	1.91	7	3	69.04	28.76	3.97	2.56	1.23	0.48

[a] The code for each trial is composed by the year, the top fertilization dose expressed as kg N/(ha·y) and the soil type reference.
[b] The meaning of the abbreviations used for the inputs and outputs of the soil nitrogen balance are shown in section 4.

5.2 Global Warming Potential

The results of rye electricity for the GHG savings with respect to natural gas electricity using the IPCC 2013 method for the GWP 100 years' time horizon are presented in **Fig. 3 to 5**. In **Fig. 3**, where GHG savings are related to rye whole plant yield, it can be seen that trials tended to follow three separated curves, each one for a fertilization dose. These curves suggest a positive correlation between yield and GHG savings and they appear to be very similar among them, but with a displacement of the amount of savings obtained for the same crop yield. These savings are much higher on average when null (77.7 %) and low (71.6 %) nitrogen were applied in top fertilization than when typical fertilization doses of 80 kg N/ha (63.7 %) were used. **Fig. 3** also reflects the effect of the different environmental conditions of each agricultural year in crop yield, obtaining lower GHG savings for all the trials with the same nitrogen top fertilization dose in the second year than the corresponding trials for the first year. The EU sustainability criteria of 60% GHG savings was accomplished for 16 out of 18 trials.

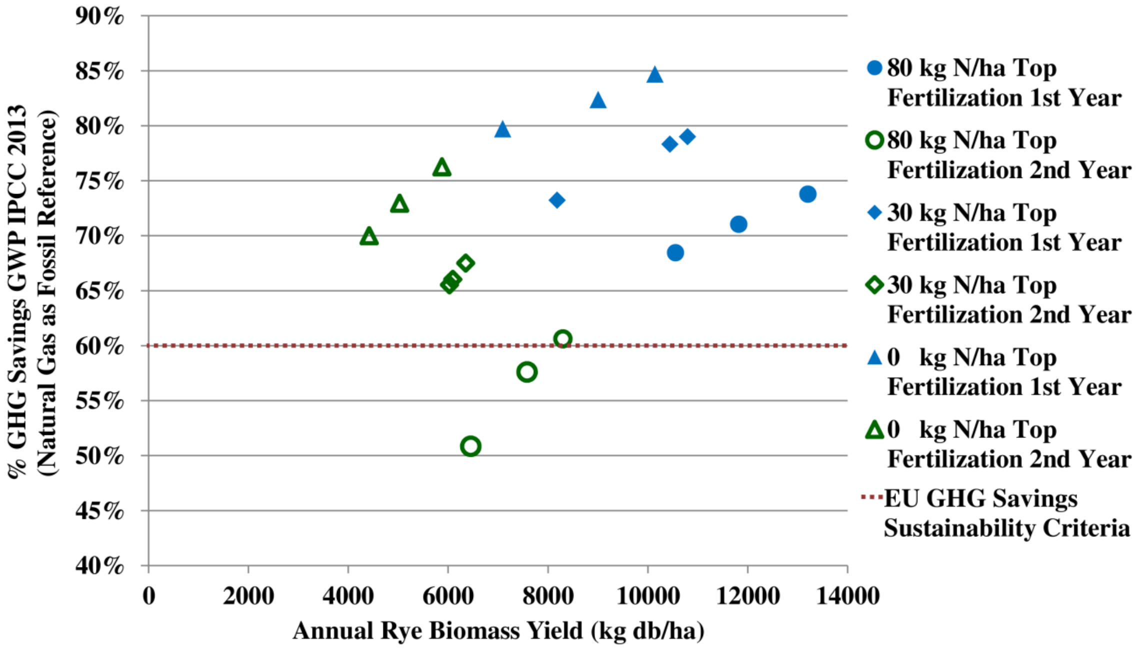

Fig. 3. Relationship between the GHG savings with respect to natural gas calculated with the GWP IPCC 2013 100 years method and the aerial rye biomass yield.

In **Fig. 4** GHG savings are related to soil nitrogen balance. It can be observed that there is a quite clear positive correlation between GHG savings and soil nitrogen deficits. However, in this figure, trials do not align forming clear curves depending on the fertilization doses as clearly as they do in **Fig. 3.** The main reason for this is the importance of the nitrogen exported by the harvest in soil nitrogen. The results of the N balance vary considerably within the same group of trials due to the differences in the nitrogen contents of aerial biomass (see **Table 2**). The nitrogen deficits were on average higher for the null (-50 kg N/ha) and low (-34 kg N/ha) nitrogen top fertilization doses than for the typical ones (-17 kg N/ha), therefore showing and inverse correlation between the nitrogen top fertilization dose and the nitrogen deficit in soil. It is remarkable that typical fertilization doses produced non-negligible nitrogen deficits. It can be also observed that first year trials generated in general more nitrogen deficits (-47 kg N/ha) than the ones evaluated during the second year (-20 kg N/ha), consequently producing more GHG savings. The reduction of 1 kg/ha of the soil nitrogen deficit implied a reduction of the GHG savings of 0.31 % according to the case study conditions and the authors calculations.

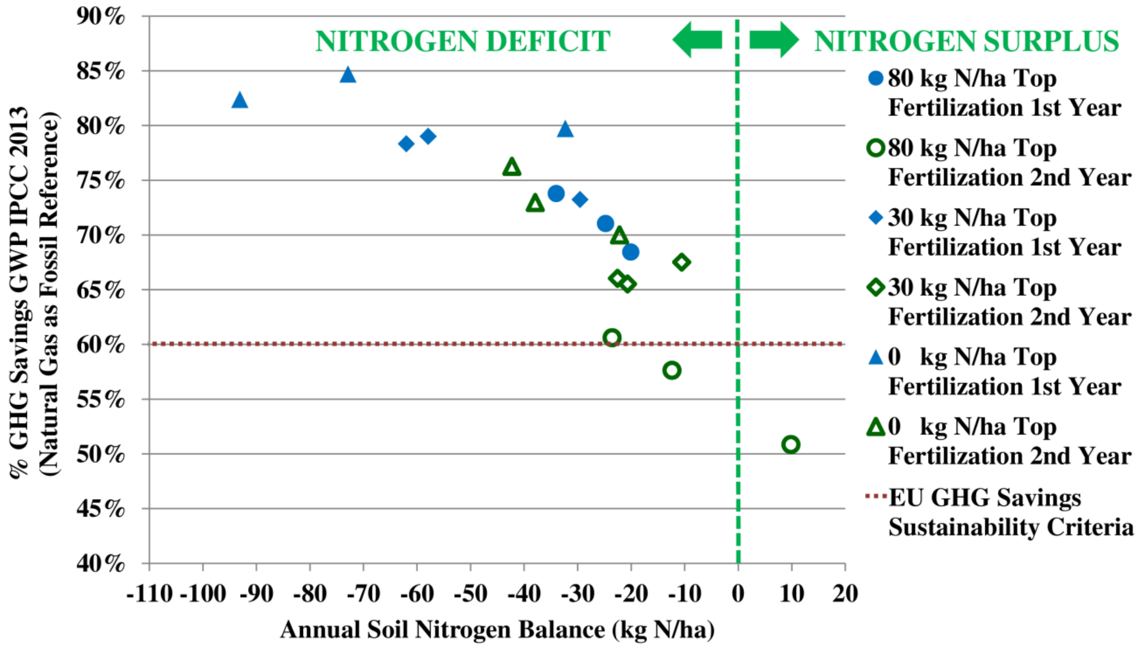

Fig. 4. Relationship between the GHG savings with respect to natural gas calculated with the GWP IPCC 2013 100 years method and the soil nitrogen balance.

Fig. 5 relates GHG savings to the total fertilization efficiency. This figure shows that trials formed a quite clear curve that appears to have a horizontal asymptote. According to this curve, there is a positive correlation between GHG savings and total fertilization efficiency. Average total fertilization efficiency was higher for the first agricultural year because the meteorological conditions were better, allowing the crop to grow more but also to extract more nitrogen coming from fertilizers as well as from soil stocks. Total fertilization efficiency was obviously higher for null and low fertilization doses because rye was able to grow extracting the majority of the N it needed from the soil instead of from fertilizers.

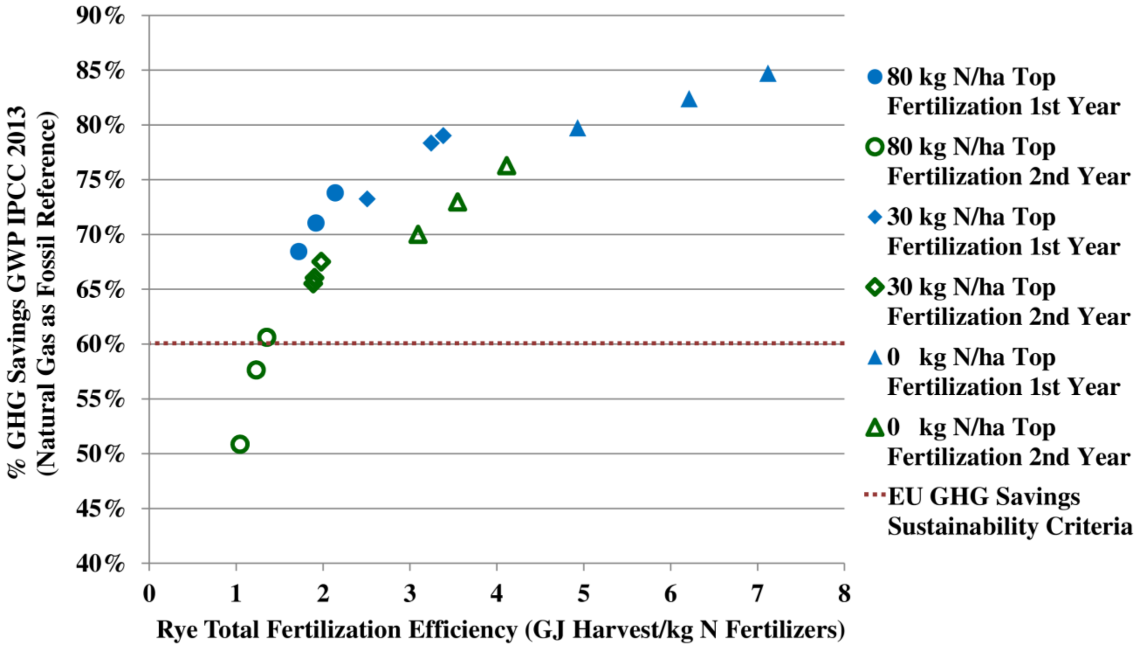

Fig. 5. Relationship between the GHG savings with respect to natural gas calculated with the GWP IPCC 2013 100 years method and the total fertilization efficiency.

5.3 Cumulative Energy

The results of rye electricity for the cumulative energy including the different types of primary energy are presented in **Fig 6 to 10.**

Fig. 6 shows the relation between the electricity generated from rye biomass per fossil energy consumed and the aerial rye biomass yield. It can be observed that trials lined following three clear tendencies according to the top fertilization dose. These alignments indicate a clear positive correlation between electrical energy generated per unit of fossil energy consumed and the rye biomass yield. As in it happened for GHG savings (**Fig. 3**), rye yield also has a positive correlation with respect to the electricity generation per fossil energy consumption. The results for the first year trials were better than the ones for the second year, regardless of the top fertilization dose. The electricity generated was between 2.6 and 3.8 times higher than the fossil energy consumed by the first year trials, while for the second year ones this ratio was between 1.7 and 2.6.

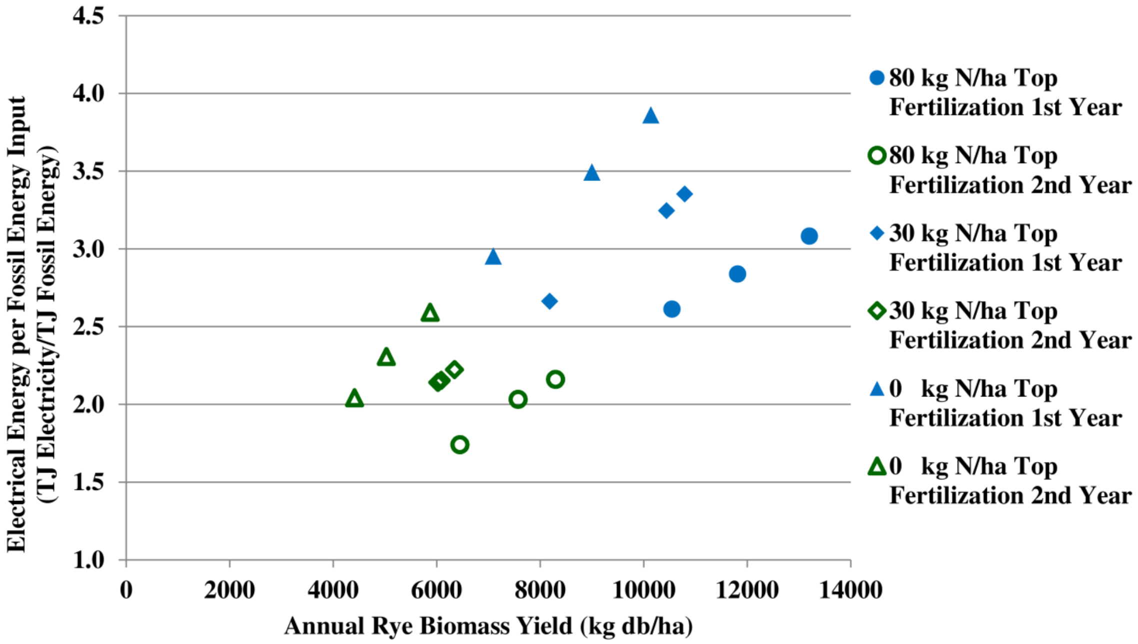

Fig. 6. Relationship between the electrical energy generated per fossil energy consumed and the aerial rye biomass yield.

In **Fig. 7** the relation between the electricity generated from rye per fossil energy consumed and the soil nitrogen balance can be observed. It can be seen in this figure that, there is a positive correlation between the energy ratio and the nitrogen deficit. Results for the three top fertilization doses of the first year revealed a higher efficiency in the use of fossil energy, but, in turn, they produced worse soil nitrogen balances with a wide range of variability. The range for the soil nitrogen balance for the first year trials went from -93 to -20 kg N/ha, while for the second year this range was narrower, from -42 to 10 kg N/ha. The figure shows that only one trial achieved a positive soil nitrogen balance. The reduction of 1 kg/ha of the soil nitrogen deficit implied a worsening of the energy balance of 0.021 TJ electricity/TJ fossil according to the case study conditions and the authors calculations.

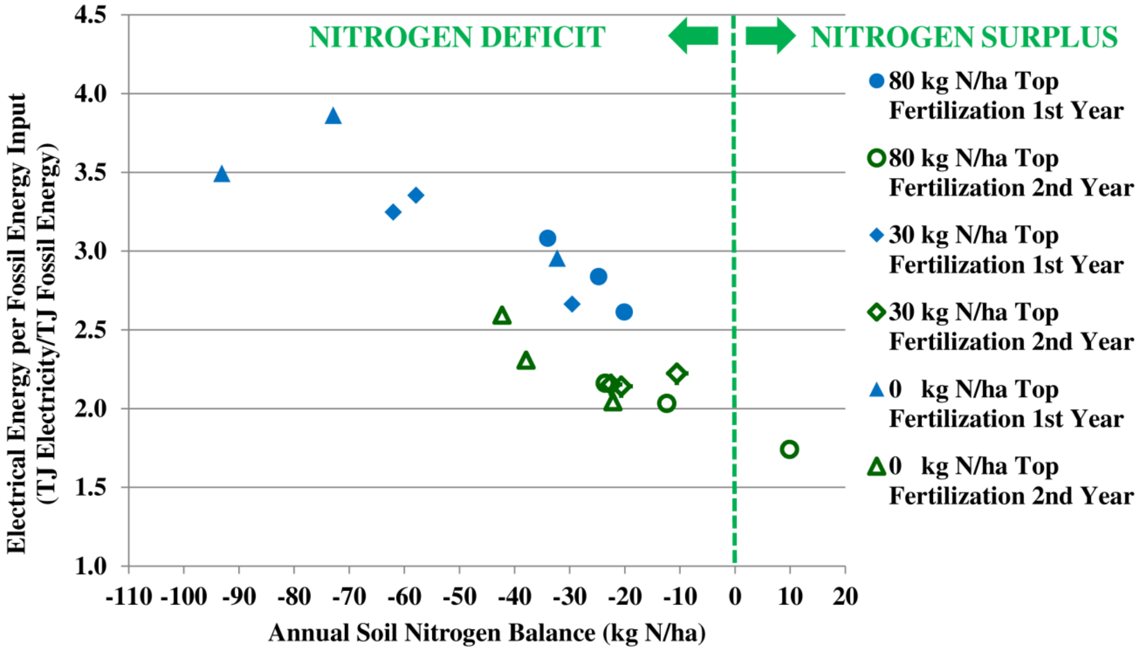

Fig. 7. Relationship between the electrical energy generated per fossil energy consumed and the soil nitrogen balance.

Fig. 8 shows the relation between the electricity generated from rye biomass per fossil energy consumed and the total fertilization efficiency. Trials lined up following three clear tendencies according the top fertilization doses as in **Fig. 3 and 6**. These tendency lines evidence a clear positive correlation between the total fertilization efficiency and the electrical energy generated per unit of fossil energy consumed. The range of variation of total fertilization efficiency was narrower for typical (1.0 to 2.1 GJ/kg N) and low (1.9 to 3.4 GJ/kg N) top fertilization doses than for null top fertilization (3.1 to 7.1 GJ/kg N). Total fertilization efficiency for the same fertilization dose was lower for the second year trials when compared to the first year ones.

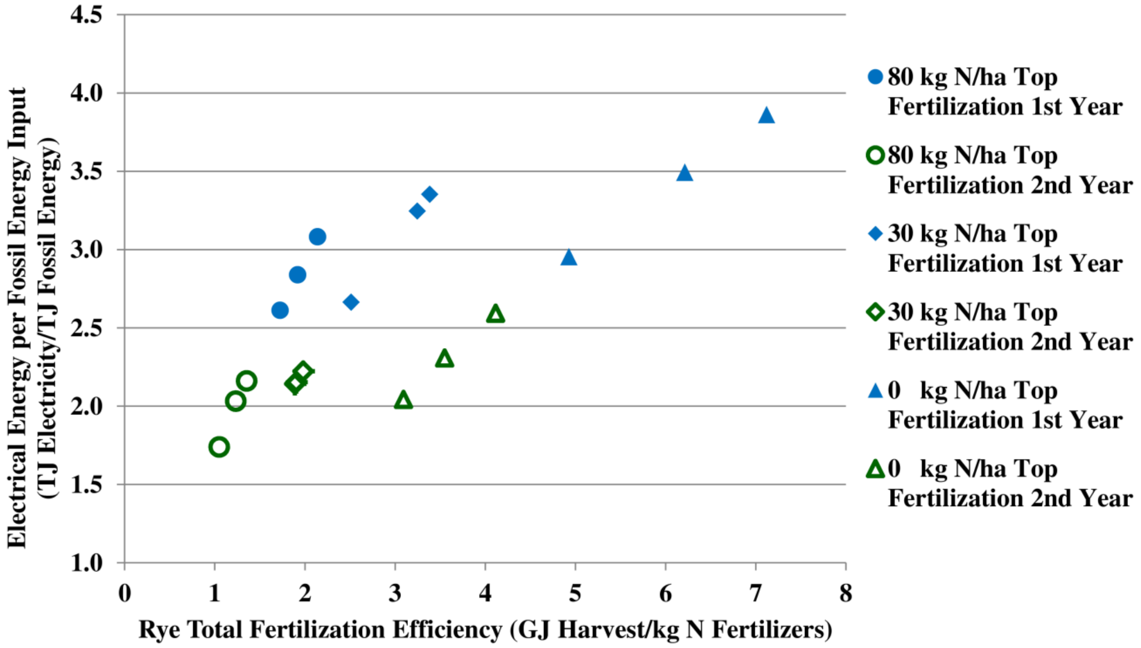

Fig. 8. Relationship between the electrical energy generated from rye biomass per fossil energy consumed and the total fertilization efficiency.

Fig. 9 shows the primary energy consumed per electrical energy generated differentiating between non-renewable and renewable energy sources. The total primary energy was higher for the three scenarios of rye electricity with 4.06, 4.03 and 4.01 TJ primary energy/TJe for typical, low and null top fertilization doses, respectively, compared to natural gas, with 2.89 TJ primary energy/TJe. This higher content of primary energy for rye electricity scenario is mainly because of the accounting of the biomass energy content of rye bales. However, non-renewable energy consumption, mostly consisting in fossil energy, was more than six times higher for natural gas electricity than for the three rye electricity scenarios. The three scenarios of rye fertilization were very similar in terms of renewable energy because the rye net heating value did not change much over the years and among top fertilization doses. The same occurred for non-renewable energy, but in this case because the lower yields obtained for null and low fertilization doses were compensated by the lower amount of N fertilizers consumed, which use high amounts of energy in their production, mostly coming from non-renewable energy sources. The average non-renewable energy consumption for natural gas was 6.4 times higher than the consumption for the rye higher top fertilization dose of 80 kg/ha.

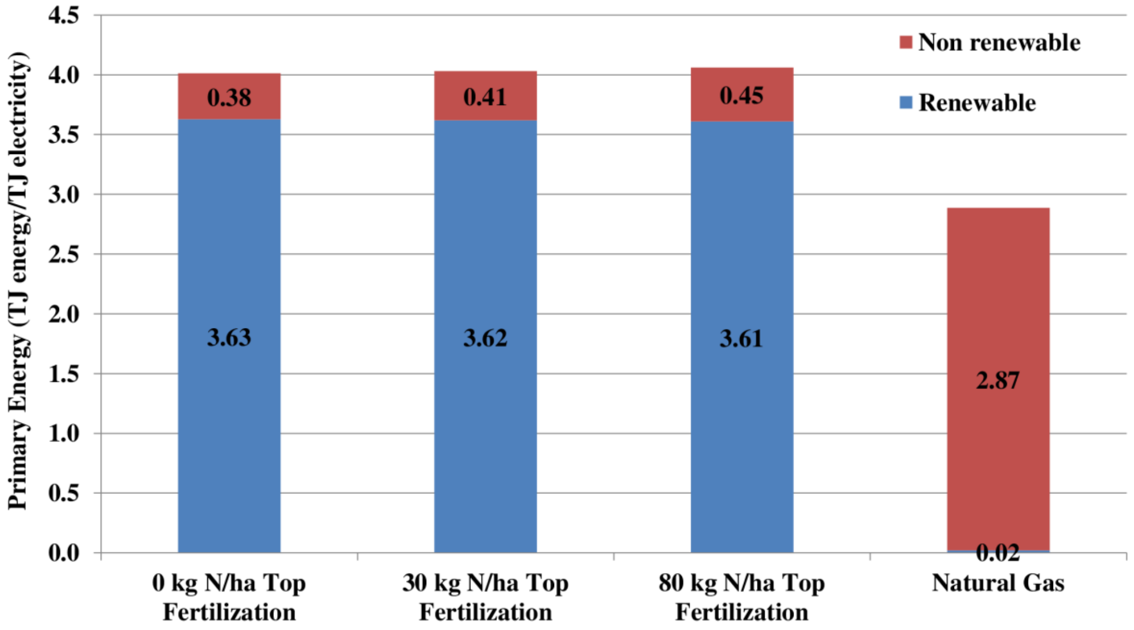

Fig. 9. Average total primary energy consumed in the generation of 1 TJe from rye biomass for the three different doses of nitrogen top fertilization and from natural gas.

In **Fig. 10** the consumption by phases of the different types of primary energy needed for the generation of 1 TJe from the three different scenarios of rye can be observed. Fertilizer production and transport was the most important phase for six out of nine categories in the figure. Power plant operation was the most important phase for renewable biomass energy consumption because bales are burned during this phase and the biomass energy of rye was accounted for here. As renewable biomass energy is the most consumed type of energy, power plant operation was also the most important phase for total renewable energy and for total primary energy. Field works were the second most important phase for five energy sources including fossil energy. As it could be expected, the share that fertilizers production and transport took grew with the top fertilization dose in all the energy sources analysed.

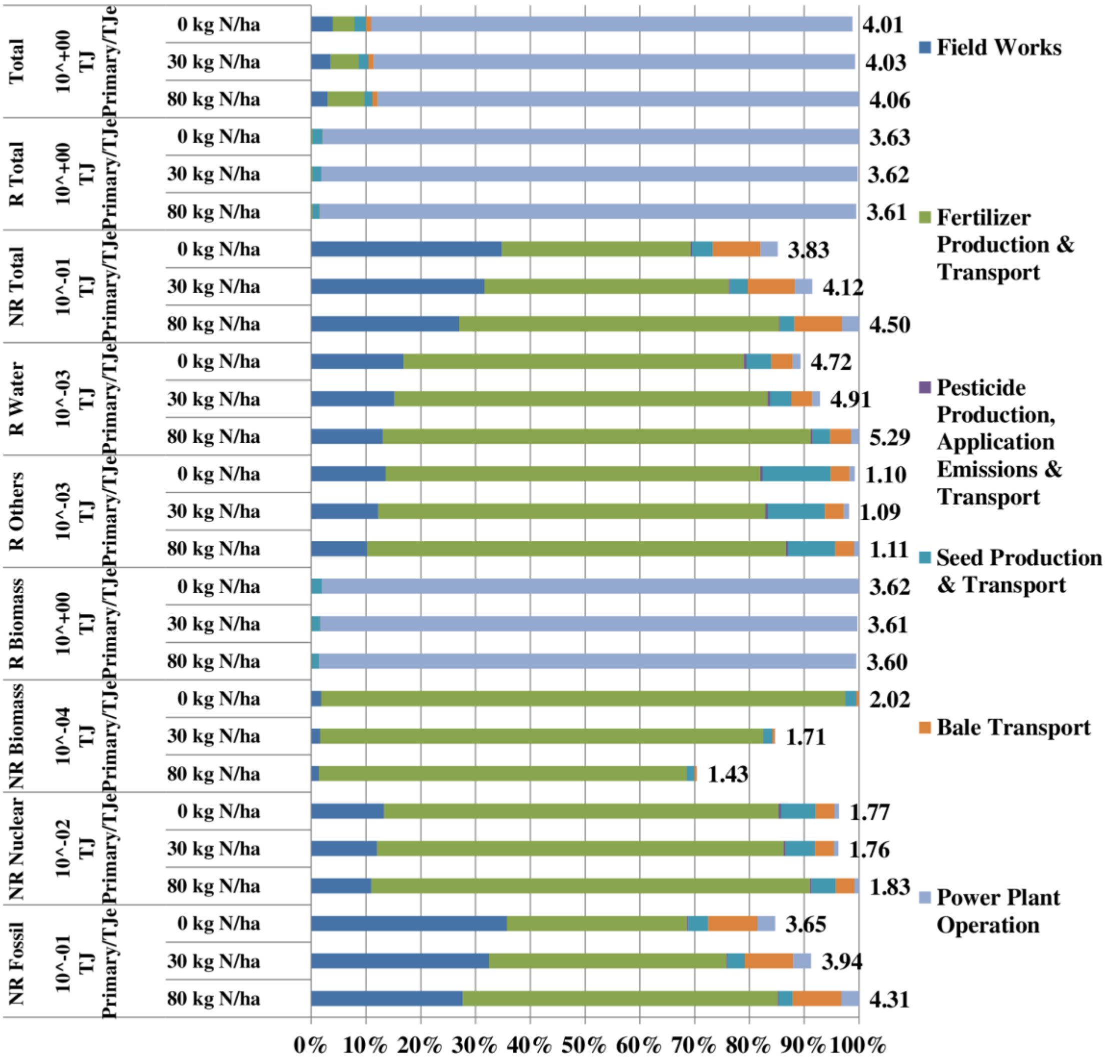

Fig. 10. Average primary energy types consumed by phases in the generation of 1 TJe from rye biomass for the three different doses of nitrogen top fertilization.

3.3 Impact Categories of CML method.

Fig. 11 compares the three rye biomass systems for the production of electricity with respect to natural gas for the eleven impact categories of the CML method. Rye biomass was a better option only for four out of the eleven categories. However, the global warming potential and the abiotic depletion due to fossil fuels consumption are found among these categories, and both of them are usually considered within the most important ones. Among the three fertilization scenarios considered for the generation of electricity from of rye, the one corresponding to typical fertilization doses (80 kg N/(ha·y)) showed the higher impacts for eight out of the cited eleven impact categories studied.

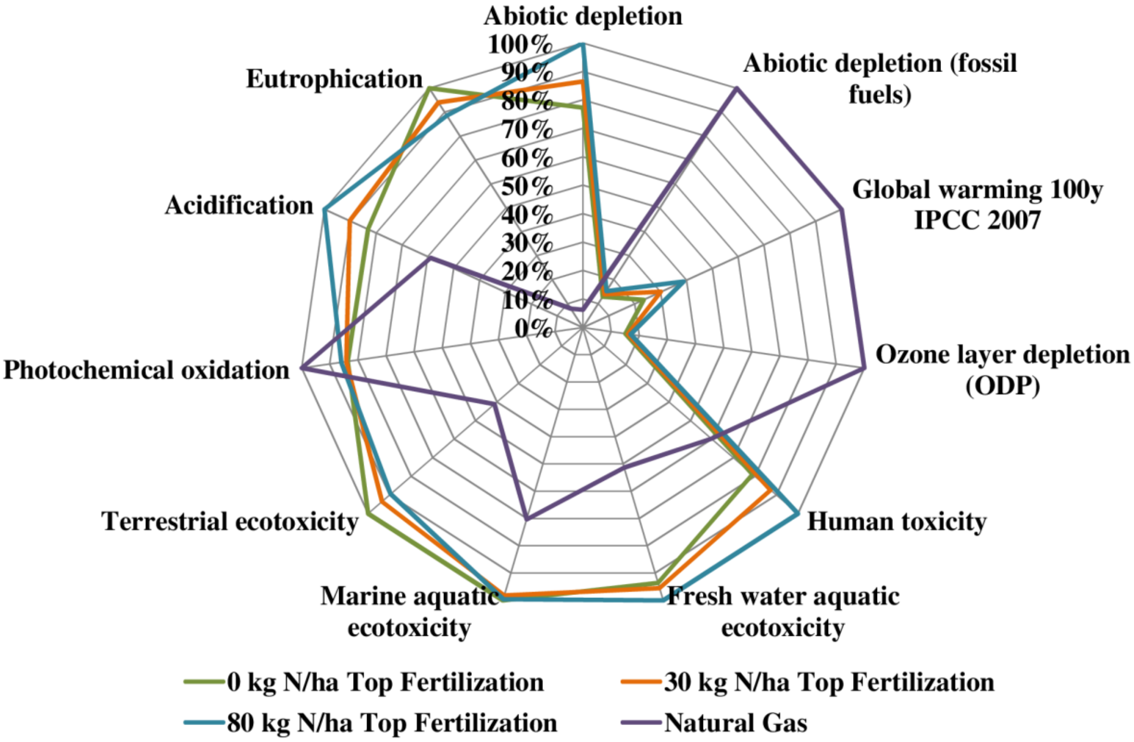

Fig. 11. Comparison of the CML method impact categories between the LCA of the three fertilization schemes used to grow rye and generate electricity and the LCA of natural gas also to generate electricity.

In **Fig. 12** it can be observed the impacts by phases incurred for the CML method categories in the generation of 1 TJe from the three different scenarios of rye top fertilization. Fertilizer productions in conjunction with field and fertilizer emissions were the most important phases for all the impact categories except for acidification on which power plant operation had a higher influence due to the aerial SO_2 emissions. The impacts within the same impact category grew with the nitrogen top fertilization dose in nine out of eleven impact categories. For eutrophication, the bigger impacts were mainly due to the use and emissions derived from the phosphorus fertilizers that were applied at the same rate for all scenarios. This circumstance in conjunction with the lower yields for null and low top fertilization doses resulted in higher impacts for these scenarios. For marine aquatic ecotoxicity, results were very similar among the three fertilization scenarios considered, and they were mainly influenced by the production of fertilizers.

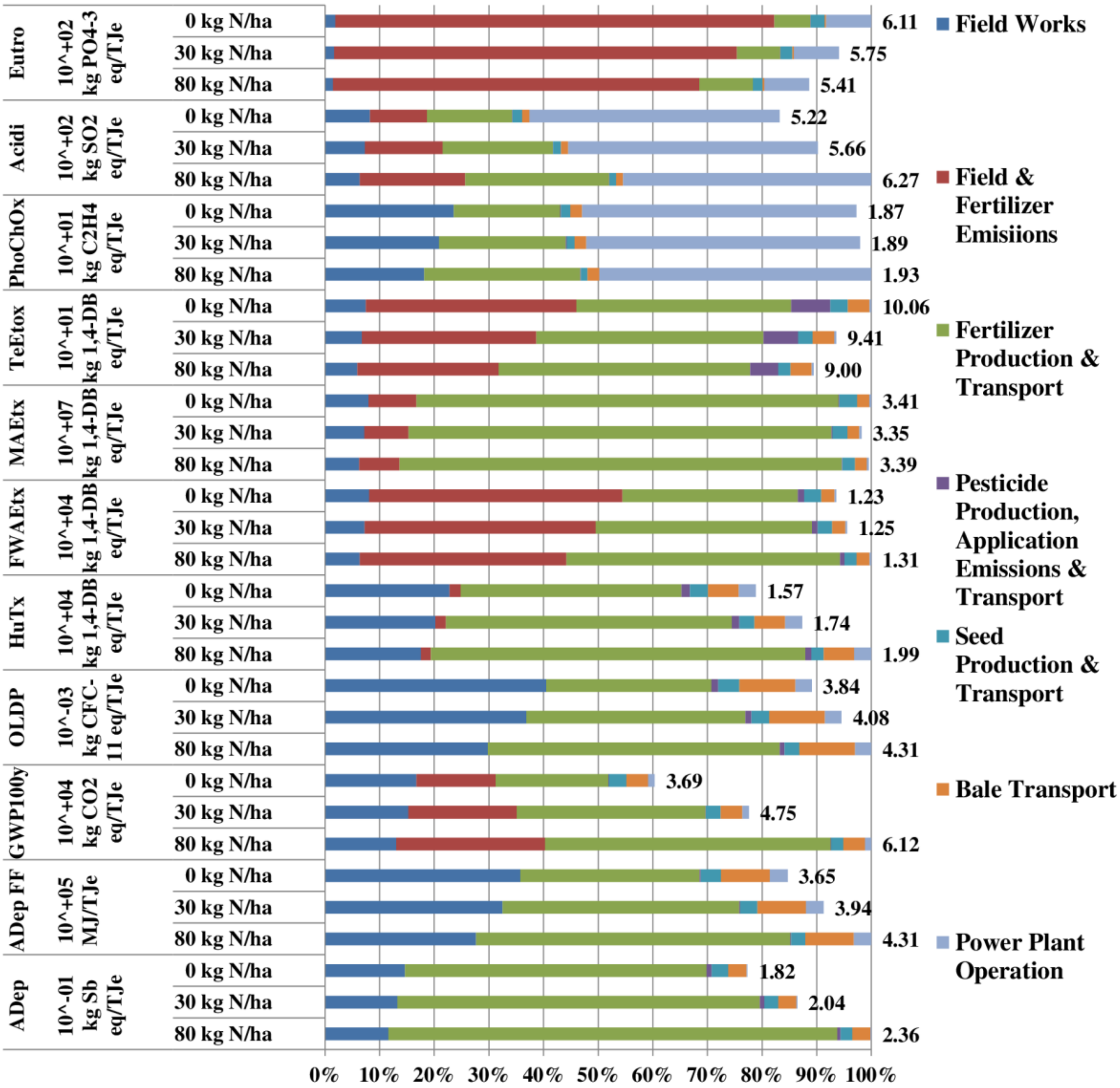

Fig. 12. Average results by phases to generate 1 TJe from rye biomass for the three different doses of nitrogen top fertilization using the CML method.

6. Discussion

From the results described, new and relevant relationships between the amounts of nitrogen provided in top fertilization with respect to GHG savings, fossil energy balances and soil nitrogen balances in the production of electricity from the aerial biomass of rye compared to natural gas are obtained. Comparing between trials it was discovered that, on average, each additional kg of N applied in top fertilization produced a reduction of 0.18% of GHG savings (see **Fig 3**), as well as a worsening in the energy balance (see **Fig 6**) of 0.00084 TJ fossil energy/TJe. On the contrary, each additional kg of N was able to reduce the soil nitrogen deficit in 0.43 Kg of N (see **Fig. 4 and 7**). The results shown in **Fig. 4** also revealed that even for the higher top fertilization dose of 80 kg N/(ha·y) significant deficits in soil nitrogen stocks were observed in most of the trials (see **Table 6**). The compensation of soil nitrogen deficits in the tests of the **Fig. 4** using previous relationships would produce a reduction in GHG savings to values between 52.7 % and 60.1 %. These new results imply that only 1 out 18 trials now will accomplish the 60 % GHG savings EU sustainability criteria worsening previous LCA results where 16 out 18 did (see **Fig. 5**). Considering the described results, from the point of view of the authors, the soil nitrogen deficit must be accounted and compensated when evaluating the sustainability of a particular crop. Increasing the nitrogen fertilization above typical doses to compensate soil nitrogen deficits, could worsen the energy balance, endanger the achievement of the EU GHG savings sustainability criteria and increase the impacts in other important impact categories as for example eutrophication.

A number of alternatives can be utilized in order to improve soil nitrogen stocks while reducing the needs for and increased use of chemical nitrogen fertilizers. Among these can be cited the use of soil nitrogen stock improvement techniques like the introduction of legumes in the crop production systems, the no-tillage farming, the optimization of top fertilization, and the reduction of specific nitrogen soil extractions by the optimisation of the crop collection time and/or the use of low nitrogen content species. The introduction of legumes in crop rotations could improve soil nitrogen stocks from 80 to 300 kg N/ha per year [46]. The use of no tillage farming has evidenced an increase of soil organic matter when compared to conventional farming [47], thus improving the content of organic nitrogen in soil. The optimization of top fertilization aims to improve the nitrogen use efficiency which can be achieved by optimising the nitrogen application to better fit the time windows when the crops require more nitrogen and considering the possibility of splitting top fertilization in two applications [48]. Other possible ways are the optimization of the processes implicated in the most important phases of the LCA. The optimization of farming operations [47] and proper sizing of machinery [49] could improve energy balances and increase GHG savings. No tillage farming is a good option to optimize the use of machinery that has other positive effects for the soil that were previously discussed. The use of ammonium sulphate for top fertilization instead of calcium ammonium nitrate could be tested due to its lower associated impacts on GWP. However, biomass yield should be maintained and possible soil acidification effects should be controlled. The reduction of transport distances of biomass is other optimization alternative of the value chains that could be achieved by incentivizing local farmers next to the power plant to grow bioenergy crops and sell them to the industry.

Another finding of this work is the large differences among the cropping years in regard to the environmental impacts produced by the crop value chain studied. In the case of GHG savings, this indicator showed an average value considering all trials of 76.7 % the first year while for the second crop year it was only 65.3 %. These differences can be mostly due to the different biomass annual yields associated to the annually variable climatic conditions, and particularly the rainfall conditions (see **Table 1**).

These results strongly support the need to carry out the LCA evaluations for an adequate number of years when the inter-annual variability of biomass yield is high, in order to achieve more confident LCAs results on a determinate crop surface.

To be coherent with the proposal for using a more holistic approach to environmental bioenergy sustainability evaluation, apart from GWP, other impacts included in the CML method have been measured. It has been discovered that regardless of the top fertilization scenario, biomass rye electricity produced considerable higher impacts than natural gas on eutrophication, acidification, abiotic depletion, human toxicity and ecotoxicities (see **Fig. 11**). Although most of the alternatives previously proposed to improve GHG savings and nitrogen balances will reduce as well these impacts, they could not be enough to achieve better results than natural gas for these categories. Therefore, the negative results obtained for these categories should be opposed to the positive ones obtained for GWP, fossil energy consumption, ozone layer depletion and photochemical oxidation. The results of the primary energy evaluation have evidenced that independently of the top fertilization scenario the consumption of non-renewable energy was lower for rye biomass electricity than for natural gas, whereas renewable energy and total primary energy were always higher due to the accounting of the renewable biomass energy contended in the rye biomass consumed.

Total fertilization efficiency or its inverse nitrogen intensity (kg N/GJ) [20] are commonly used to compare among performances of bioenergy crops or scenarios for a specific crop. However, these indicators tend to lead to better results for experiences with low nitrogen fertilization doses (see **Fig. 5 and 8**) that can produce considerable soil nitrogen deficits (see **Table 6**) and compromise future soil fertility.

The use of soil nitrogen balance has evidenced that some bioenergy experiences with high GHG savings may be causing problems for soil fertility sustainability due to considerable negative nitrogen balances. Therefore, the inclusion of soil nitrogen balance as a complementary indicator for bioenergy LCAs and the establishment of a maximum soil nitrogen deficit within the sustainability criteria would be recommendable.

7 Conclusions

The results derived from this study evidenced that even typical fertilization doses for dedicated bioenergy crops could produce nitrogen deficits in soil stocks. Using null and low fertilization doses resulted in more GHG savings with respect to natural gas and better results for the majority of the impacts assessed. However, they produced a significant raise in nitrogen deficits, compromising the soil sustainability and future crop fertility.

Raising the nitrogen fertilization above typical doses aiming to compensate soil nitrogen deficits could compromise the achievement of the EU sustainability criteria of 60 % GHG savings.

Besides, it could generate excessive impacts in other relevant impact categories, such as eutrophication.

To overcome this problem and be able to achieve EU sustainability criteria and sustainable soil nitrogen balances the following recommendations are made:

The use of soil nitrogen improvement techniques like crop rotation with legumes, no-tillage farming and optimization of top fertilization practices.

The reduction of crop specific nitrogen soil extractions by the optimisation of the crop collection time and/or the use of low nitrogen content species.

The optimization of field works and biomass transport as well as the use of the most energy efficient and less emitting fertilizer among the different options available for the crop.

Even though having all the data of the soil and the biomass characterization at your disposal may not be the most common case, it is recommendable to perform at least an estimative soil nitrogen balance using the data from bibliography or databases that best fit the particular case study. The authors believe that the use of soil nitrogen balance as a sustainability indicator in conjunction with LCA, can help to provide a more holistic approach to the sustainability evaluation of bioenergy systems by bringing into question some experiences that accomplish the EU 60% GHG savings sustainability criteria but generate excessive soil nitrogen deficits, therefore requiring improvement. The authors think that the establishment of maximum sustainable soil nitrogen deficit rates within the bioenergy sustainability criteria could be a good strategy to improve the reliability of the LCA results.

Acknowledgements

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Appendix A

A.1 Introduction

This appendix contains all the equations needed for the life cycle inventory calculations. They are provided separated from the main body of the article in order to make it more readable.

A.2 Energy output calculation

The energy output of the system was obtained through Eq. **(A.1)**:

$$E \ = RY/(1-H)\cdot(1-SL)\cdot NHV_{CP,H}\cdot\eta \qquad \text{(A.1)}$$

where E (MJe/(ha·y)) is the electrical energy generated, RY (kg crop/(ha·y)) is the rye whole plant yield at 0% humidity (See **Table 2**), H (kg water/kg crop) is the water content of rye in a per unit basis, SL are the bales storage losses in a per unit basis, $NHV_{CP,H}$ (MJ/kg) is the net heating value of rye at constant pressure at water content H (see Eq. **(A.2)**) and η (MJe/MJ crop) is the conversion efficiency of rye into electricity in a per unit basis.The humidity considered for rye bales was 12%. A value of storage loses of 1% was considered appropriate for this study. The efficiency was 29 % as this is the average efficiency of the 25 MWe real biomass plant of this considered for this study.

The net heating value of rye at constant pressure at water content *H* was calculated as shown in Eq. **(A.2)**:

$$NHV_{CP,H} = NHV_{CP,0} \cdot (1 - H) - 2.444 \cdot H \qquad \text{(A.2)}$$

where $NHV_{CP,H}$ (MJ/kg) and *H*(kg water/kg crop) are described previously in Eq **(A.1)** and $NHV_{CP,0}$ is the is the net heating value of rye at constant pressure at 0% water content obtained from **Table 2.**

A.3 Agricultural machinery manufacture calculation

The amount of machinery consumed for carrying out a specific agricultural operation was calculated as shown in Eq. (A.3):

$$AM = W \cdot OR/LT \qquad \text{(A.3)}$$

where *AM* (kg/(ha·y)) is the amount of machinery consumed for carrying out a specific agricultural operation, *W* (kg) is the weight of the machinery used to carry out the operation (see **Table 3**), *OR* (h/(ha·y)) is the operating rate (see **Table 3**) and *LT* (h) is the lifetime of the machinery which is obtained from the Spanish platform for the knowledge of agricultural machinery [38].

A.4 Field and fertilizer nitrogen derived emissions calculation

The calculation of the nitrous emissions (N_2O) is shown in Eq. **(A.4):**

$$N_2O = 44/28 \cdot (\mathrm{EF}_1 \cdot (N_{tot} + N_{cr}) + \mathrm{EF}_4 \cdot N\_NH_3 + \mathrm{EF}_5 \cdot N\_NO_3{}^{-}) \qquad \text{(A.4)}$$

where N_2O (kg N_2O/(ha·y)) are the emissions of N_2O to the air, 44/28 is conversion factor of N-N_2O in N_2O, EF_1 (kg N-N_2O/kg N inputs) is the factor expressing the fraction of nitrogen inputs that is converted into nitrogen in the form of N_2O, N_{tot} (kg N/(ha·y)) is the total nitrogen input from fertilizers (base + top fertilization), N_{cr} (kg N/(ha·y)) is the nitrogen contained in the crop residues (stubble + roots), EF_4 (kg N-N_2O/kg N-NH_3) is the factor expressing the fraction of N-NH_3 that is converted in N-N_2O, *N-NH*$_3$ (kg N-NH_3/(ha·y)) are the losses of nitrogen in the form of ammonia (See **3.4.6)**, EF_5 (kg N-N_2O/kg NO_3^-) is the factor expressing the fraction of N-NO_3^- leached that is converted in N-N_2O and *N-NO*$_3^-$ (kg N-NO_3^-/(ha·y)) is the nitrogen leaching to ground water in the form of nitrate (See Eq. **(A.5)**).Average values of 0.01 has been considered for EF_1 and EF_4 and of 0.0075 for EF_5.

The calculation of nitrate emissions (NO_3^-) is shown in Eq. **(A.5):**

$$N\_NO_3^- = 21.37 + P/(c \cdot L) \cdot (0.0037 \cdot S + 0.0000601 \cdot N_{org} - 0.00362 \cdot U) \qquad \text{(A.5)}$$

where $N\_NO_3^-$ (kg N-NO_3^-/(ha·y)) is the nitrogen leaching to ground water in the form of nitrates, P (mm/y) is the precipitation (see **Table 1)**, c (%) is the clay content of the soil (see **Table 1**), L (m) is the root depth of the crop. S (kg N/(ha·y)) is the nitrogen supply through fertilizers (see **Table 1)**, N_{org} (kg N/ha) is the nitrogen in soil organic matter and U (kg N/(ha·y)) is the nitrogen uptake by the crop. An average of 1.32 m has been considered for rye root depth. The organic nitrogen is estimated in an 85% of total nitrogen (see **Table 1).** The total nitrogen uptake by the crop is estimated as the sum of the nitrogen content of the harvest, the stubble and the roots (see **Table 2**).

The emissions of nitrogen to surface water due to soil erosion are shown in Eq. **(A.6):**

$$N\_Er = S_{er} \cdot N_{es} \cdot \mathrm{F_{rn}} \cdot \mathrm{F_{erwn}} \qquad \text{(A.6)}$$

where N_Er (kg N/(ha·y)) is the quantity of nitrogen emitted through erosion to rivers, S_{er} (kg soil/(ha·y)) is the quantity of soil eroded [50], N_{es} (kg N/kg soil) is the nitrogen content in top soil (See **Table 1**), $\mathrm{F_{rn}}$ is the enrichment factor for nitrogen and $\mathrm{F_{erw}}$ is the fraction of eroded soil that reaches the river. An average value of 1.86 was used for the nitrogen soil enrichment factor and an average value of 0.2 was used for the factor that represents the fraction of eroded soil that reaches the river.

A.5 Emissions of phosphorus to the water calculation

Run–off of phosphorus to surface water was calculated as an average corrected by phosphorus fertilization as shown in Eq. **(A.7)**:

$$P_{ro} = P_{rol} \cdot F_{ro} = P_{rol} \cdot (1 + 0.2/80 \cdot P_2O_{5min}) \qquad \text{(A.7)}$$

where P_{ro} (kg P/(ha·y)) is the quantity of phosphorus emitted through run-off to rivers, P_{rol} (kg P/(ha·y)) is the average quantity of P lost through run-off for a land use category, F_{ro} is the correction factor for fertilization with phosphorus, P_2O_{5min} (kg P_2O_5/(ha·y)) is the quantity of P_2O_5 contained in mineral fertilizers. A emissions value of 0.175 kg P/(ha·y) was considered for P_{rol} for being an average value appropriate for open arable land.

Emissions of phosphorus from soil particles that reach surface water were calculated as shown in Eq. **(A.8)**:

$$P_{er} = S_{er} \cdot P_{es} \cdot \mathrm{F_{rp}} \cdot \mathrm{F_{erw}} \qquad \text{(A.8)}$$

where P_{er} (kg P/(ha·y)) is the quantity of phosphorus emitted through erosion to rivers, S_{er} (kg soil/(ha·y)) is the quantity of soil eroded [50], P_{es} (kg P/kg soil) is the phosphorus content in top soil (See **Table 1**), $\mathrm{F_{rp}}$ is the enrichment factor for phosphorus and $\mathrm{F_{erw}}$ is the fraction of eroded soil that reaches the river. An average value of 1.86 was used for the phosphorus soil enrichment factor and an average value of 0.2 was used for the factor that represents the fraction of eroded soil that reaches the river.

A.6 Emissions of heavy metals to agricultural soil, surface water and ground water calculation

Drainage emissions of heavy metal were calculated using constant leaching rates as shown in Eq. **(A.9)**:

$$M_{leach\,i} = m_{leach\,i} \cdot A_i \tag{A.9}$$

where $M_{leach\,i}$ (mg metal/(ha·y)) are the agricultural drainage related emission of the heavy metal i, $m_{leach\,i}$ (mg metal/(ha·y)) is the average amount of leaching of heavy metal i (Cd = 50, Cu = 3600, Zn = 33000, Pb = 600, Ni = n.a, Cr = 21200 and Hg =1.3), A_i is the allocation factor for the share of agricultural inputs in the total inputs for heavy metal i. The allocation factor measures the proportion of the total input of each heavy metal that is attributable to the agricultural system studied discounting the effects of the atmospheric deposition of heavy metals. This calculation is shown in Eq **(A.10)**:

$$A_i = M_{agro\,i} \,/\, (M_{agro\,i} + M_{deposition\,i}) \tag{A.10}$$

where $M_{agro\,i}$ (mg metal/(ha·y)) is the total input of heavy metal i from agricultural production (fertiliser + seeds + pesticides) [35] and $M_{deposition\,i}$ (mg metal/(ha·y)) is the total input of heavy metal from atmospheric deposition (Cd = 700, Cu = 2400, Zn = 90400, Pb = 18700, Ni = 5475, Cr = 3650 and Hg = 50).

The emissions of heavy metals to the surface water due to erosion are calculated in Eq. **(A.11)** in a similar way to phosphorus:

$$M_{erosion\,i} = C_{tot\,i} \cdot S_{er} \cdot \mathrm{F_{rh}} \cdot \mathrm{F_{erw}} \cdot A_i \tag{A.11}$$

where $M_{erosion\,i}$ (mg metal/(ha·y)) are the agricultural related heavy metal emissions to surface water through erosion, $C_{tot\,i}$ (mg metal/ kg soil) is the heavy metal i content of soil, S_{er} (kg soil/(ha·y)) is the quantity of soil eroded, $\mathrm{F_{rh}}$ is the enrichment factor for heavy metals, $\mathrm{F_{erw}}$ is the fraction of eroded soil that reaches the river and A_i is the allocation factor for the share of agricultural inputs in the total inputs for heavy metal i (see Eq.**(A.10)**).An average for Spanish soils (mg/kg) was used for the heavy metals content of soil (Cd = 0.15, Cu = 15.6, Zn = 47.15, Pb = 19.13, Ni = 14.23, Cr = 19.67 and Hg = 0.0551) [51]. For the watershed of the region, the average erosion is 0.82 mm/ha·y [50]. Considering 1300 kg/m3 as soil density, 10,660 kg soil/ha are lost annually. The value for the enrichment factor was 1.86. The value for the fraction of eroded soil that reaches the river was 0.2.

The balance between inputs and outputs of heavy metals provides the changes of each heavy metal in the soil due to the cultivation of rye as shown in **Eq (A.12)**:

$$M_{soil\,i} = \left(\sum Inputs_i - \sum Outputs_i\right) \tag{A.12}$$

where $M_{soil\,i}$(mg metal/(ha·y) is the change in the content of metal i in the soil due to the agricultural system, $Inputs_i$ (mg metal/(ha·y)) is the total input of heavy metal i to the agricultural soil (fertilizers, pesticides, seed) [35], $Outputs_i$ (mg metal/(ha·y)) is the total output of heavy metal i from agricultural soil (exported biomass, leaching and erosion). The heavy metal contents of rye (mg/kg rye) are taken from the Phyllis 2 database [52] (Cd = 0.1, Cu = 1.8 and Pb = 0.6). The atmospheric deposition of the heavy metal is not attributable to the rye energy system and therefore, it should not be accounted for, resulting $\Sigma \text{Inputs}_i = M_{\text{agro } i}$ (see Eq. **(A.10)**).

A.7 Soil nitrogen balance calculation

The soil nitrogen balance is calculated as shown in Eq. **(A.13)**:

$$SNB = (N\_Fert + N\_Seed + N\_AtDep + N\_HarvEx + N\_BioFix) - (N\_HarvEx + N\_NO_3^{-} + N\_Er + N\_NH_3 + N\_N_2O_{Cr+Fert} + N\_NO_x) \qquad \text{(A.13)}$$

where *SNB* (kg N/(ha·y)) is the soil nitrogen balance, *N_Fert* (kg N/(ha·y)) is the nitrogen provided by fertilizers (See **Table 1**), *N_Seed* (kg N/(ha·y)) is the nitrogen provided by sowing seed, *N_AtDep* (kg N/(ha·y)) is the nitrogen provided by atmospheric deposition, *N_FrLiv* (kg N/(ha·y)) is the nitrogen fixed by the effect of soil free living organisms, *N_BioFix* (kg N/(ha·y)) is the nitrogen symbiotic biological fixation of legumes [46], *N_HarvEx* (kg N/(ha·y)) is the nitrogen exported by the harvest (see **Table 1**), $N\_NO_3^-$ (kg N/(ha·y)) are the nitrogen losses by leaching in the form of NO_3^- (see Eq. **(A.5)**), *N_Er* (kg N/(ha·y)) are the nitrogen losses by soil erosion that reach surface water (see Eq. **(A.6)**), N_NH_3 (kg N/(ha·y)) are the nitrogen losses by volatilization in the form of ammonia (see **3.4.6**), $N\_N_2O_{Cr+Fert}$ (kg N/(ha·y)) are the nitrogen loses due to nitrogen coming from fertilizers and crop residues an released in the form of nitrous oxide (see Eq. **(A.4)**) and N_NO_x (kg N/(ha·y)) are the nitrogen loses due nitrogen oxides released during the denitrification process (see **3.4.6**). The nitrogen provide by seed is calculated by multiplying the sowing dose by the nitrogen content of the seeds. The nitrogen content of rye seeds is obtained by dividing the protein content [53] by the nitrogen to protein factor [54]. Within the range of possible values [55], 7 kg N/(ha·y) has been considered a good estimation for the atmospheric deposition. A value 3 kg N/(ha·y) has been considered a good estimation for the fixation of free living organisms [55]. No symbiotic biological fixation has been accounted for rye.

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