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Comparison of biofuel life-cycle GHG emissions assessment tools: The case studies of ethanol produced from sugarcane, corn, and wheat



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ABSTRACT

The use of alternative fuels, particularly bio-based fuels, has been an important strategy to achieve greenhouse gas (GHG) emission reductions compared to petroleum-based fuels. However, discrepancies between results obtained by using different attributional life-cycle assessment (LCA) tools have challenged the credibility of the individual assessments, and as result, the progress towards or compliance with GHG mitigation targets. The objective of this study was to identify the main differences and commonalities in methodological structures, calculation procedures, and assumptions for the major commercial biofuel, ethanol, across three public LCA tools, BioGrace (EU), GHGenius (Canada), and GREET (U.S.), and a research-oriented fourth, the Virtual Sugarcane Biorefinery (VSB), a Brazilian platform for sugarcane ethanol assessments.

The calculated emissions across models ranged from 16 to 45 for sugarcane, 43–62 for corn, and 45–68 g $CO_2eq MJ^{-1}$ for wheat ethanol. Harmonizing the three public models with VSB assumptions for sugarcane ethanol produced in Brazil, the range was reduced to 16–17 g $CO_2eq MJ^{-1}$ for sugarcane ethanol. Agricultural production (e.g., N₂O emissions from fertilizers; energy and fuel use; straw field-burning; and limestone application) and ethanol shipping were found to be the major causes for variations for differences calculated for sugarcane ethanol. Similarly, harmonizing BioGrace and GHGenius calculations using GREET assumptions for U.S. corn ethanol generated nearly identical results (models varied within a 3% range). The coproduct treatment method was found to be the most influential parameter in the variations calculated for both corn and wheat ethanol. The application of the tools as part of GHG emissions accounting requirements is often defined via regulations and differences and/or conflicting assumptions set-forth in these models lead to most differences observed. Our study provides recommendations for promoting transparency in LCA calculations and assumptions across the tools used in research and development or for regulatory tools regarding biofuels.

1. Background

The use of bio-based alternative fuels has been considered an important strategy to achieve reductions in greenhouse gas (GHG) emissions from the transportation sector dominated by petroleum fuels, promote rural economy and improve energy security. Government agencies around the world have developed initiatives and policies to encourage the production and use of biofuels, which could contribute to multiple sustainability goals. Increasingly the growing variety of biofuels, including high energy density hydrocarbon fuels, makes the issue of reducing GHG emissions of increased importance [1-6].

To measure the progress towards one of the primary goals of biofuel policies, i.e., GHG emission reductions, several models have been developed to quantify life-cycle GHG emissions of biofuels and their reference fuels (typically petroleum gasoline, diesel, and jet fuels). Some models were designed to comply with regulatory requirements, whereas others were adopted and/or modified from existing research and development tools investigating multiple pathways for fuels coupled to vehicle life-cycle systems.

This study examined three publicly available life-cycle GHG emissions models for transportation fuels: GHGenius [7,8] used in some Canadian provinces to determine the carbon intensity of fuels under the Low Carbon Fuel Standards (LCFS); GREET (Greenhouse Gases, Regulated Emissions, and Energy use in Transportation Model) [9], used by the U.S. Environmental Protection Agency (EPA) along with other models for the rulemaking of the Renewable Fuel Standard 2 (RFS2).

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Abbreviations			in Transportation, by Argonne National Laboratory, ANL, U.S.		
CARB	California Air Resources Board	GWP	global warming potential – 100-year		
CCLUB	Carbon Calculator for Land Use Change from Biofuels	IPCC	Intergovernmental Panel on Climate Change		
	Production, sub-model of GREET	ISO	specific standard from the International Organization of		
CH_4	methane		Standards		
CNPEM	Brazilian Center for Research in Energy and Materials	JEC	Joint European Commission		
CO_2	carbon dioxide	km	kilometer		
CO ₂ eq	carbon dioxide equivalent	LCA	life-cycle assessment (GHGenius); life-cycle analysis		
CONCAV	VE Conservation of Clean Air and Water in Europe		(GREET)		
CRC	Coordinating Research Council, Inc.	LCFS	Low Carbon Fuel Standard		
CTBE	Brazilian Bioethanol Science and Technology Laboratory	MJ	megajoule		
EPA	U.S. Environmental Protection Agency	N_2O	nitrous oxide		
EU	European Union	RFS2	U.S. Renewable Fuel Standard 2		
EU-CAR	European Council for Automotive Research and	RSB	Roundtable on Sustainable Biomaterials		
	Development	UK-RTFC	OUnited Kingdom Renewable Transportation Fuels		
EU-RED	European Union Renewable - Energy Directive		Obligation		
FQD	Fuel Quality Directive, EU	VSB	Virtual Sugarcane Biorefinery, CBTE, CNPEM, Brazil		
GHG	greenhouse gas	WTT	well-to-tank		
GREET	Greenhouse Gases, Regulated Emissions, and Energy Use	WTW	well-to-wheel		

Derived GREET versions are used by the California Air Resources Board (CARB) LCFS (U.S.); and BioGrace (BIOfuel GReenhouse gas emissions Alignment of Calculations in Europe) [10], simplified for use in compliance with the European Union Renewable Energy Directive (EU-RED) and the Fuel Quality Directive (FQD), with harmonized data for EU nations.

Because the LCA models were designed for different purposes, significant variations have been observed in GHG emissions estimates, especially for biofuels [11-22]. Many factors, including assumptions, input data, treatment of coproducts, level of details built in the calculation structure, and specific attributes of the Life-cycle Assessment (LCA) approach utilized could influence the estimates of carbon intensity per functional unit of biofuel used in the transportation sector. In addition, much of the literature does not identify the specific model version used nor the level of maturity of the technology, making meaningful comparisons of results difficult, and, at times misleading. The inconsistency among these GHG modeling tools and their use has led to a great variability in GHG emissions results for the same biofuel pathway such as ethanol from sugar or starch crops, biodiesel from soybean or rapeseed which are commercial and bagasse or corn stover to ethanol which are in demonstration or initial commercialization phase. Macedo et al. [6] pointed out technical challenges in the evaluation of biofuels, including the need for reliable data (i.e. both at agronomic and conversion stages) and for a higher level of scientific consensus on a series of aspects, such as the treatment of coproducts, land use change, and reference systems [11,12,15,16].

One of the most debated pathways is ethanol produced from corn grown in the U.S.: some studies concluded that its GHG emissions would be nearly twice as high as those from gasoline [16,17], mainly driven by the (direct and indirect) land use changes induced by diverting corn to produce ethanol. Other studies found that corn ethanol offers advantageous GHG reductions of up to 40% in comparison with gasoline in scenarios which maintain the reference land use as continuing to be productive or improved agricultural land [11,18-20]. In

the case of Brazilian sugarcane ethanol, Cavalett et al. [21], Seabra et al. [22], Wang et al. [13] reported GHG emissions savings between 67% and 86% relative to the baseline gasoline. Khatiwada et al. [14] assessed GHG emissions for Brazilian sugarcane ethanol under North American (i.e., EPA's RSF2 and California Air Resources Board CARB LCFS) and European Directives and regulatory schemes (i.e., EU-RED and United Kingdom Renewable Transportation Fuels Obligation (UK-RTFO)). The authors found that GHG emissions savings at that time could vary as much as 30% within the same method (e.g., the EPA RFS2 approach can range between 61% and 91%) and more than 40% using the methods required by different regulations (e.g., CARB LCFS approach 31% and EU-RED approach showed 72%). Agricultural practices (especially soil carbon and nitrogen dynamics), coproduct credits from surplus electricity, and uncertainties around economic modeling approaches for indirect land use change were the major drivers of methodological divergences.

The discrepancy in GHG emissions estimates from the application of different LCA models noted in the previous analyses affected the acceptance of LCA results and their use in the biofuel policy context and the reported progress towards meeting the established GHG emissions reduction targets. A few studies on the comparison of LCA models used for regulatory purposes investigated methodological differences. Hennecke et al. [23] compared BioGrace versus the RSB (Roundtable on Sustainable Biomaterials) version approved under the EU-RED. Chum and Warner [24] performed a harmonization study for dry-mill corn ethanol production based on natural gas energy source using GHGenius and BioGrace models with GREET 2015 data as model inputs. Harmonized model results agreed well with each other within 2.5%. O'Connor [25] and Unnasch et al. [26] evaluated a broader range of LCA models for the Coordinating Research Council, Inc. (CRC) of petroleum and automotive equipment.

In this paper, in addition to the three publicly available LCA models (i.e. GREET, BioGrace and GHGenius) the Virtual Sugarcane Biorefinery (VSB) [27], developed by the Brazilian Bioethanol Science and

> Fig. 1. Schematic representation of the main methodological steps for the comparison of LCA models.



Technology Laboratory (CTBE) served as reference for the sugarcane ethanol production in Brazil and its parameters served to harmonize the three public models. The primary objective of this study was to quantify the main differences and identify commonalities in methodological structures, calculation procedures, and assumptions, to understand how and why the models generate different GHG emissions results. In addition, this comparative analysis provides recommendations for more consistent and transparent LCA methodologies and best practices reporting results of GHG emissions for biofuel pathways used globally.

2. Methods

The general aspects of the LCA methodology are provided in section 2.1 since it is the common methodological framework to all of the models considered in our study. Fig. 1 depicts the methodological steps used to compare LCA models. Step (1) of the analysis compares the main characteristics, parameters, and assumptions of the models. Step (2) extracts the life-cycle inventories from the models and converts to common units (e.g., mass and energy per tonne of feedstock, kilometers (km) of transportation). This step generates preliminary numerical comparison among models which use their own inputs and categories in different units, thus making direct comparison more difficult. Step (3) standardizes activity categories (e.g., fertilizer manufacture, diesel use, and natural gas use) associated with the life cycle of ethanol production and use. The categories were standardized across the models when the values were extracted from each model for each category. For instance, GHGenius presents a category named "land use changes, cultivation", which aggregates a series of items such as limestone application, N fertilizer use, and field-burning of sugarcane straw with manual harvest, among others. This definition of land use changes associated with cultivation used in GHGenius is different than that in the other models. The standardization process, in this case, was not straightforward; the intricate calculation mechanisms of the models had to be analyzed for each specific case in the models. Step (4) is the comparison of the results calculated for the GHG emissions of ethanol followed using the default calculation mechanisms of each LCA model. This part of the analysis was intrinsically linked to the other steps, given that the differences observed in the calculated results were discussed based on information obtained in the previous steps. In Step (5), a harmonization procedure utilizing modified steps to select parameters and assumptions within the models was performed using one of the models as "default" for harmonizing the other methods parameters and assumptions.

2.1. Life cycle assessment

The LCA methodology is often used to assess the environmental impacts associated with a product, process or activity, by the identification and quantification of energy and materials flows used, as well as waste and emissions released. The approach has been utilized as a standard to estimate life-cycle GHG emissions of biofuels. The general framework for conducting the assessment can be found in the International Organization for Standardization (ISO) documents [28,29]. The general procedure includes the following steps: defining scope, system boundaries, functional unit, and reference systems; determining mass and energy flows; and treating coproducts; and assigning impacts to energy and material flows.

Fig. 2 shows a schematic overview of the typical life cycle (well-towheel or WTW) of ethanol produced from biomass. The scope of the LCA approach usually takes into consideration the agricultural (including cropping, harvesting, and other relevant operations), transport, industrial (conversion of feedstock to biofuel), transport of the fuel through distribution, and fuel use stages. Inputs and outputs from each stage may vary depending on the type of feedstock considered, any preprocessing conducted, type of conversion plant, the location of these different activities, the considered system boundaries, and the LCA assumptions utilized.

2.2. Considered LCA models

In this section, the LCA models utilized for the comparison carried out in this paper are briefly described. Detailed assumptions and calculation mechanisms based on selected characteristics summarized in Table 1, including key drivers of differences among modeling (e.g., geography, LCA approach and treatment of coproducts, and upstream life-cycle data). These aspects are further discussed in section 3.

2.2.1. GREET

The GREET model [9] was developed in 1996 by the Argonne National Laboratory (ANL) for the U.S. Department of Energy and sponsored by the U.S. Department of Energy's Office of Energy Efficiency and Renewable Energy. It was originally designed to be a simple-to-use tool through which researchers could evaluate fuel-cycle energy and emission impacts of various transportation technologies. Since its inception, the model continues to be updated and expanded by the ANL. The most recent versions are the GREET1 2017 for fuel-cycle analysis, GREET2 2017 for vehicle-cycle analysis, and a graphic interface named GREET.net 2017. The model includes more than 100 fuel pathways such as petroleum fuels, natural gas fuels, biofuels, hydrogen, and electricity produced from various energy feedstock sources. Three classes of vehicles and various technologies are addressed. GREET version 2016 was used in this study.

In 2007, GREET version 1.8b was modified by Lifecycle Associates for CARB to quantify GHG emissions for implementing LCFS in California, where it is used for regulatory purposes and updated periodically.

2.2.2. GHGenius

GHGenius [7,8] is a model developed for Natural Resources Canada by $(S\&T)^2$ Consultants based on the 1998 version of Lifecycle Emissions Model by Delucchi [30]. The model calculates energy and emissions associated with conventional and alternative fuel production starting with the input from the past years (beginning with 1995) and uses projections into the future (2050). This study made use of GHGenius version 4.03 of 2013, which is capable of modeling multiple geographic



Fig. 2. Schematic overview of the life cycle of ethanol fuel and the main inputs and emissions from each stage.

Table 1

Selected characteristics of the LCA models investigated in this study.

Parameters	GREET	GHGenius	BioGrace	VSB
Model version	2016	4.03a	4d	2015
Developed for regulatory use?	No	No	Yes	No
Type of LCA	Attributional	Attributional	Attributional	Attributional
Functional unit	Service (km, mile)	Service (km)	Energy (MJ)	Service (km)
	Energy (Btu, MJ)	Energy (MJ)		Energy (MJ)
Life cycle data	Internal calculation	Internal calculation	JRC database	Ecoinvent v2.2
Default treatment of coproducts	Substitution (corn and wheat)	Substitution	Energy	Economic
	Energy (sugarcane)			
Heating value	LHV or HHV	HHV or LHV	LHV	LHV
Domestic and international land-use change	CCLUB model	C stocks	C stocks	-
Geography	U.S.	Canada, U.S., Mexico, India	Europe	Brazil
Gasoline baseline (g CO ₂ eq per MJ)	90.2	95.0	83.8	87.5
Impact categories	GHG,	GHG,	GHG	GHG,
	Energy,	Energy,		Energy,
	Water use,	Cost effectiveness		Ozone depletion and others
	Air pollutants			

regions, including not only country-level analyses for Canada, U.S., Mexico, and India, but also accounting for regional specificities. The public model version 5.0a is currently used as the basis for regulations; it was released in 2018, after this study was completed.

2.2.3. BioGrace

BioGrace [10] is a spreadsheet model used for the calculation of biofuel GHG emissions originated from the European cooperative harmonization effort, in which country model owners from Germany, the Netherlands, Spain, and the United Kingdom developed the calculator to implement the EU-RED and the FQD. The current model manager is the Institute for Energy and Environmental Research (IFEU) from Germany (BioGrace 4d version was used).

The calculation is based on a database with default values (EU averages) of 22 commercial feedstock/biofuels pathways elaborated by a joint group of experts, including the Joint European Commission (JEC) from the Joint Research Center (JRC), the European Council for Automotive Research and Development (EUCAR), and the Conservation of Clean Air and Water in Europe (CONCAWE). The complete and updated database and their models are not public, although results from multiple fuels are published periodically. BioGrace 4d uses a set of default values from 2011 that is accessible to users [31]. The model also allows users to input their own data as an approved voluntary scheme to demonstrate compliance with the EU-RED.

2.2.4. Virtual Sugarcane Biorefinery

The VSB was developed by the Sustainability Division of the CTBE from the Brazilian Center for Research in Energy and Materials. The tool evaluates sugarcane biorefinery configurations, combining chemical engineering computer simulation software (e.g., Aspen Plus), mathematical modeling, and sustainability assessment methods with the objective of identifying and evaluating technical parameters and sustainability impacts related to novel and existing biorefinery configurations. Sustainability impacts calculated in this tool include environmental (e.g., climate change, human toxicity, ecotoxicity, particulates, acidification, eutrophication, energy, land, and water use), economic (e.g. internal rate of return, net present value, and levelized production cost) and social (e.g. number of jobs, occupational accidents, wages, educational level, and gender distribution) aspects [27,32–47].

2.3. Approach for comparison of the LCA models

All four models were used to estimate GHG emissions impacts for sugarcane ethanol produced in Brazil. GREET and GHGenius were utilized for assessing the impact of corn ethanol produced in the U.S., whereas BioGrace calculated the impact of corn ethanol produced in the EU. GHGenius and BioGrace were used to assess wheat ethanol production, the first with data from the U.S. and the latter from the EU. For the sake of consistency, modeling was performed for the same year, when possible. For GREET and GHGenius, the year was set to 2015; VSB already utilizes average values for sugarcane ethanol in 2015 as default. The BioGrace 4d version, however, does not support changes to the default model year (2011).

2.3.1. Harmonization procedure

The objective of the harmonization procedure was to assess to what extent a difference in specific steps would affect the results and how close final figures obtained would be in relation to each other, after models are harmonized having one of the models as "default".

In summary, the modification steps described for sugarcane ethanol in Brazil and corn ethanol in the U.S. accomplished the following: (1) applying consistent approaches for coproducts treatment (i.e., economic allocation for sugarcane ethanol and substitution for corn ethanol); (2) removing overseas transportation (for sugarcane ethanol only), equaling and harmonizing ground transportation distances and GHG emissions from ethanol used in vehicles; (3) harmonizing nitrogen, limestone (for both ethanol cases), and field-straw burning GHG emissions (for sugarcane ethanol only); (4) harmonizing energy use in agricultural operations; and (5) harmonizing GHG emissions from the ethanol production processes (industrial conversion stage) and yields to those of VSB for sugarcane ethanol in Brazil and of GREET for corn ethanol in the U.S.

3. Results and discussion

3.1. General comparison of the models

Table 1 introduced some of the key drivers of differences among modeling results are discussed. Although GREET and GHGenius are used by policymakers for regulatory compliance in the U.S. and Canada, respectively, BioGrace is the only model developed to assess compliance of biofuels in the EU. Modified versions of GREET are used for compliance in the CARB LCFS.

The LCA scope is consistent mostly among GREET, GHGenius, and VSB. BioGrace, however, it is a well-to-tank (WTT) calculation following the EU-RED legislation specified values of various parameters. The model includes a threshold value or cut-off criterion of 0.1 g CO₂eq per MJ of fuel for inclusion of component inputs or processes in the accounting of the total GHG emissions of the biofuel pathway. The JEC model is capable of modeling both WTT and tank-to-wheel emissions, making use of specific values for the chosen propulsion system [31]. GREET and GHGenius have coupling choices of WTT with several types of vehicle cycle modeling tools to obtain overall WTW results in public

models. VSB also accounts for the vehicle emissions and provides multiple functional units such as CO_2eq emissions in terms of km traveled and include the exhaust emissions from the use of fuel in vehicles, as well as the emissions from the whole supply chain.

All four models provide life-cycle GHG emissions inventories; however, only VSB applies a structured life cycle impact assessment method and assesses other local and global environmental impacts. Life-cycle GHG emissions associated with gasoline (in g CO2eq per MJ of fuel) are used as the fossil fuel reference system to estimate the GHG emissions reductions obtained by biofuels. The fossil fuel reference emission between the models can differ as much as 12% across models (95.0 g CO2eq per MJ for GHGenius and 83.8 g CO2eq per MJ for BioGrace). The 12% difference includes producing, transporting and refining the various crude oils and alternative sources (tar sands or shale oil) used to produce the fossil fuel in the countries or states that are using the fossil and renewable fuels. Differences in the fossil fuels reference systems are expected between models, because they are designed to model different oil refineries with different technologies, petroleum quality and mix of products in the different regions of the world.

Climate impacts are typically expressed in terms of global warming potentials using a time horizon of 100 years (GWP_{100}), although the GWP values are updated over time with improvements from climate science. BioGrace utilizes GWP_{100} from the IPCC 2001 report [48] as default [10]. It provides, however, the option of switching the factors to those from the IPCC 2007 report [49]. GREET and VSB use GWP_{100} from the IPCC 2013 report [50] as default. GREET allows users to select values among all five IPCC reports [48–52]. GHGenius uses factors from the IPCC 2007 [49] report as default; however enabling the use of factors from IPCC 1995 and 2001 reports [48,52]. For the sake of comparison, characterization factors for all models were harmonized to use GWP_{100} values in the IPCC 2007 report [49].

3.1.1. LCA approach and treatment of coproducts

All four models use an attributional LCA approach as default, meaning that the focus of the models is on describing the environmentally relevant physical flows (e.g., materials and energy) to and from the environment for the biofuel production at defined system boundaries [53–57]. This differs from a consequential LCA approach, which aims to describe how environmentally relevant flows will change in response to possible substitution of products' decisions [54].

The choice of the procedure used to treat coproducts is one of the most controversial topics in LCA. The issue arises when a system produces more than one valuable output, as in a multi-functional system. The concern is associated with biofuels production systems because other useful products are often coproduced with the fuel of interest. The ISO 14040 and 14044 documents recommend avoiding allocation whenever possible either through subdivision of processes or by expanding the system boundaries to include the functions associated with the coproducts generated. This includes displacement or substitution methods that are intrinsically more complex as their implementation requires significant amount of market data in laborious levels of detail [56]. For instance, in the case of corn ethanol, a major coproduct is dried distillers' grains with soluble materials (DDGS), a protein rich animal feed, for beef, dairy, swine, and poultry. DGS (wet form) is also sold to closely located beef and dairy animals farms. DDGS substitutes corn, soy meal, and urea at specific displacement ratios determined, which is one example of the expansion of the boundaries. The aggregate amounts of the various displaced conventional products would have generated GHG emissions that are offset by the displacement ratios and amounts of the corresponding nonfuel product output. The emissions displaced are the credits for production of fuel products from the total system GHG emissions [56]. This example also includes another level of expansion of the system given by the amount of methane generation credit for avoided methane in dairy production. Current dry mills (70%) also extract corn oil (non-edible) for the production of biodiesel

and thus the boundaries are further increased with this additional energy coproduct. In summary, multi-functionality here is treated by subtracting from the product system the impacts from reference products (in the example corn, soy meal and urea) that may be displaced by the coproducts in the market. The substitution method can be considered a step into the consequential LCA approach.

When it is not possible to avoid allocation, the main recommendation by ISO is to use methods that reflect the physical properties or other relevant variables [55]. The most recent ISO document on this matter is the Sustainability Criteria for Bioenergy [58], which provides further guidance on the treatment of coproducts. Documentation of the selected procedures for coproducts treatment is needed and justified, including sensitivity analysis to illustrate the consequences of using alternative procedures.

The models investigated in this study made use of different coproduct treatment approaches as default: the VSB considers economic allocation while BioGrace makes use of energy for partitioning as per regulation, although JEC [31] suggests substitution as the most appropriate approach. GHGenius utilizes the substitution method, whereas GREET allows the user to calculate multiple approaches depending on the biofuel pathway. For instance, the impacts of sugarcane ethanol and surplus electricity produced in Brazil are allocated according to energy as default, whereas the impacts of corn ethanol in the U.S. can be calculated by substitution or other allocation methods.

3.1.2. Upstream life-cycle data

The life-cycle stages of a process or product include extraction, manufacture, logistics, and use; a product used as an input to a process carries the impact load from the previous stages. To account for these impacts in the assessment, GREET and GHGenius use internal calculation mechanisms based on sector inventories: power, transportation, industrial, and resource extraction. BioGrace and VSB utilize the JRC-EUCAR-CONCAWE database v4.a [31] and the Ecoinvent database v2.2 [59], respectively.

Main relevant items in terms of GHG impacts include fertilizers, diesel, and natural gas, among others, accounting for their manufacture and use. Table 3 presents the upstream life-cycle data for selected inputs. The impacts can vary significantly across the models. For instance, regarding diesel, the impact can vary as much as 30% with 116.4 g CO_2eq per MJ estimated by GHGenius (with 75% of the total impact associated with the combustion stage) and 81.6 g CO_2eq per MJ by the VSB (with 55% of the total impact associated with the combustion stage); for the nitrogen fertilizer manufacture, variation can be as much as 43% with 5.88 g CO_2eq per kg of nitrogen for BioGrace and 3.35 g CO_2eq per kg of nitrogen for the VSB.

3.1.3. Nitrous oxide (N_2O) emissions

The contribution of N_2O is an important variable in the calculation of climate impacts, magnified by its high characterization factor that can be as much as 300 times greater than that of CO_2 depending on the IPCC GWP method chosen. N_2O emissions on biofuel pathways come mainly from nitrogen fertilizer application and organic matter decomposition [60], depending on soil type, climate, crop, tillage method, and fertilizer and agricultural residues application rates.

LCA studies accounting for N₂O emissions often utilize the default emissions factors published by IPCC, which present estimates from several sources [61]: direct soil emissions of N as N2O, at 1% of synthetic N fertilizer application and 2% for manure; volatilization of N as NH₃ at a rate of 10% of total N in the case of synthetic N application or 20% of total N in the case of manure application, with 1% of the N in the NH₃ converted to N₂O; and runoff and leaching to groundwater as nitrate at 30% of total N applied, with 0.75% of it converted to N₂O. The total resulting effect is that 1.325% of N in synthetic fertilizer is emitted as N₂O (due to direct emissions of NH₃ and nitrate to N₂O).

Table 2 presents the emissions factors for direct and indirect N₂O

Table 2

Emissions factors for direct and indirect N_2O emission from fertilizers and agricultural residues.

0				
Emission factors ^a	GREET	GHGenius	BioGrace	VSB
Direct N ₂ O emissions ^b			1.00%	
Sugarcane	0.895%	1.25%	-	1.00%
Corn	0.900%	1.25%	-	-
Wheat	-	1.00%	-	-
Indirect N ₂ O emissions ^c				
Volatilization of N as NH ₃	10%	10%	10%	30%
N in NH3 converted to N2O	1.00%	1.00%	1.00%	1.00%
Runoff/leaching as nitrate	30%	30%	30%	5%
Nitrate converted to N ₂ O	0.75%	0.75%	0.75%	0.75%
Total N ₂ O emitted				
Sugarcane	1.220%	1.575%	1.325%	1.460%
Corn	1.225%	1.575%		-
Wheat	-	1.325%		-

^a GHG models utilize as basis default Tier 1 emission factors published by IPCC, which estimates emissions from several sources [66]: volatilization of N as NH₃, at a rate of 10% of total N in the case of synthetic N application (ranging from 3% to 30%) or 20% of total N in the case of manure application; direct soil emissions of N₂O, at 1% in case of synthetic N and 2% in case of manure; runoff and leaching to groundwater as nitrate at a rate of 30% of total N applied (ranging from 10% to 80%) with 0.75% of it converted to N₂O; default resulting effect is that 1.325% of N in synthetic fertilizer is emitted as N in N₂O.

 $^{\rm b}$ BioGrace and the VSB utilize the default IPCC values for the direct $\rm N_2O$ emissions, whereas GHGenius and GREET consider differentiated values for the crops.

^c BioGrace, GHGenius, and GREET models utilize the default IPPC values for the indirect N₂O emissions, whereas the VSB considers specificities of the soil used for sugarcane production in Brazil with a higher value for volatilization of N as NH₃ (30%) and lower for runoff/leaching as nitrate (5%), according to recommendations from experts.

Table 3

Upstream life cycle data for selected inputs.

Input	GREET ^a	GHGenius ^a	BioGrace ^a	VSB ^c			
kg CO ₂ eq per kg of nutrient (manufacture)							
Nitrogen (N)	4.48	3.51	5.88	3.35			
Phosphate (P ₂ O ₅)	1.51	0.73	1.01	2.16			
Potassium (K ₂ O)	0.66	0.47	0.58	0.55			
g CO ₂ eq per kg of input (manufacture $+$ use)							
Limestone (CaO)	236.0	790.0	129.5	131.6			
g CO ₂ eq per MJ of fuel (production $+$ combustion)							
Diesel	90.2	116.4	87.6	81.6			
Coal	96.0	103.7	111.3	-			
Natural gas	66.7	83.1	67.6	-			

^b The European life cycle database v4.a.

^a Internal calculation.

^c Ecoinvent database v2.2.

emissions from fertilizers and agricultural residues assumed by each model. Although the GHG models investigated in this study make use of the IPCC 2006 method as a basis to account for field emissions, small differences in the assumptions may lead to significant variability in the obtained results.

For instance, regarding direct N_2O emissions associated with the use of N-fertilizer, BioGrace and the VSB use the default IPCC values for the direct N_2O emissions (equivalent to 1%), whereas the others consider different values for the crops (GHGenius uses 1% for wheat and 1.25% for corn and sugarcane, whereas GREET assumes 0.895% for Brazilian sugarcane and 0.9% for corn).

3.1.4. Land use changes

GREET allows for the inclusion of GHG emissions associated with direct land use change (LUC) due to corn crop management and soil carbon data from the CENTURY model, Carbon Online Estimator (Tier 3) for the U.S. and induced land use change based on its Carbon Calculator for Land Use Change from Biofuels Production (CCLUB) module [62]. The CCLUB model combines land conversion (area and type) data from Purdue University's Global Trade Analysis Project model and the CENTURY model for estimates of U.S. soil carbon and Winrock International for international land use [63–66]. BioGrace and GHGenius also present the option of calculating land use changes based on the carbon stocks change method for various biomass feedstocks as an additional feature of the models. The assumptions and treatment of the LUC issue obviously varies across considered GHG emission models. however we did not assess the influence of land use change parameters and assumptions in GHG emissions associated with ethanol production in this paper. It is because our study focus on the attributional value chain GHG emissions of ethanol fuel pathways. Several issues on LUC modeling regarding biofuel pathways have been recently addressed in the scientific community (e.g. Refs. [67-73]). LUC results are especially dependent on the modeling assumptions and normally gain robustness when site-specific information is taken into account. Therefore, the analysis of LUC parameters and assumptions present in the LCA models deserves specific further analysis.

3.2. Life-cycle inventories

Tables 4–6 present the main agricultural and industrial inputs and products logistics and transportation parameters for ethanol from sugarcane, corn, and wheat, respectively, considered in each LCA model. Detailed lists of all the agricultural and industrial inputs and yields used for ethanol produced from the three types of biomass can be found in Tables 1S–4S of the supplementary material.

3.2.1. Sugarcane ethanol production in Brazil

The differences observed for most of the agricultural and industrial inputs reflect the sources of data selected: GHGenius uses average values from three studies [22,74,75] with the exception of diesel used in agricultural machinery. GREET uses data documented in Wang et al. [19] including the same references as GHGenius, and BioGrace derives data from Macedo et al. [75] in 2008, whereas the data for VSB are based on recent agriculture and industrial sectorial data, recommendations from experts, literature, and process simulations [27].

A key driver, the application of limestone, triggers CO₂ emissions due to its degradation: GHGenius assumes an application rate of more than double the values used by the other models. GHGenius uses the value reported by Macedo et al. [75] in the estimate, which considers an application rate of 1900 kg CaO per ha. In comparison, Seabra et al. [22] and Macedo et al. [74] reported a field application rate of 450 kg and 366 kg CaO per ha, respectively, in 2011 and 2008, from agronomic best practices at these times, and the 2011 publication projected rates to 2020.

Another important parameter is the proportion of mechanized harvested sugarcane versus the manual harvesting of field-burned straw, an old practice of the ethanol and sugar industry. However, since 2002, a series of state and federal laws have gradually reduced sugarcane field burning practices in face of concerns with climate change and health effects [76]. The models investigated in this study give the user a choice of the percentage of manual sugarcane harvesting. Changing the default values can be tricky and complex, given that models are not designed for non-LCA designers as users. Original default values of the models were maintained for this comparative assessment prior to harmonization. GREET assumes a time series for straw burning with values ranging from 95% of manual harvesting in 1995 to 14% in 2015. The value for 2015, which was used in this study, is close to the default value of VSB tool of 18.4%. BioGrace, on the other hand, considers that 100% of the straw is field-burned as default. GHGenius assumes no field burning as default; this value, however, may be manually set by the user.

All four models assume that sugarcane ethanol is produced in Brazil. VSB uses domestic transportation by truck from the producer through

Table 4

Main agricultural, industrial and transportation parameters for sugarcane ethanol.

Parameters	Unit	GREET	GHGenius	BioGrace ^a	VSB
Per tonne of sugarcane					
N fertilizer	kg	0.80	1.08	0.91	1.23
P ₂ O ₅ fertilizer	kg	0.30	0.58	0.41	0.14
K ₂ O fertilizer	kg	1.00	1.47	1.08	1.31
Limestone	kg	5.20	11.54	5.34	5.00
Diesel for machinery	L	1.1	2.9	0.8	1.9
Sugarcane transportation	km	19.3	20.0	20.0	27.3
Default straw burning	%	14%	0%	100%	18.4%
Per liter of ethanol					
Sulfuric acid	g	-	7.40	16.06	4.94
Lime	g	10.85	11.00	17.97	7.48
Cyclohexane	g	-	-	1.06	0.71
Ethanol transportation in Brazil	km	692 (truck 100%)	400 (truck 100%) additional 63 (rail 2%) and 137 (barge 34%)	700 (truck 100%)	345 (truck 100%)
overseas from Brazil	km	11,934 (ship 100%)	12,558 (ship 100%)	10,186 (ship 100%)	-
in destination country	km	208 (truck 100%)	-	150 (truck 100%)	-

^a A factor of +40% is applied to industrial inputs for BioGrace to encourage voluntary data disclosure contribution from the private sector.

Table 5

Primary agricultural, industrial and transportation parameters for corn ethanol in the U.S.

	Unit	GREET ^a	GHGenius	BioGrace ^b
Per tonne of corn				
N fertilizer	kg	16.7	17.2	13.3
P ₂ O ₅ fertilizer	kg	5.7	5.0	8.9
K ₂ O fertilizer	kg	6.0	6.9	6.6
Limestone	kg	45.3	-	412.0
Diesel (machinery operation)	L	4.2	4.8	26.3
Corn transportation	km	80.5	100	50
Per L of ethanol				
Electricity	MJ	0.7	0.9	-8.0
Natural gas	MJ	6.1	7.9	27.1
Coal	MJ	0.5	1.8	-
Ethanol transportation	km	837 (barge	121 (truck	150 (truck
		13.2%)	100%)	100%)
		1287 (rail	additional 802	
		78.9%)	(rail 22%)	
		128 (truck		
		7.9%)		
		additional 48		
		(truck 100%)		

^a GREET considers three types of corn mills existent in the U.S. for the production of ethanol: dry mill without corn oil extraction (17.72%); dry mill with corn oil extraction (70.88%); and wet mill (11.40%) based on Wang et al. [70].

^b A factor of +40% is applied to industrial inputs for BioGrace to encourage voluntary data disclosure contribution from the private sector; electricity produced with required steam is accounted as a credit to the product system.

the distribution system to tank whereas the other models include not only Brazilian ground distribution but also overseas shipping from Brazil to the destination country.

3.2.2. Corn ethanol

The GREET model uses as default weighted-average values of the three types of corn mills: dry mills without corn oil extraction (17.7%); dry mills with corn oil extraction (70.9%); and wet mills (11.4%) [18]. Therefore, the values presented in Table 5 for the industrial stage in the GREET model are, in fact, of the amounts of inputs and outputs used by the different types of existing mills. Detailed industrial inputs for ethanol produced from the three types of mills can be found in Table 5S of the supplementary material. In terms of agricultural inputs, GREET makes use of the study by Wang et al. [77] as a basis to project values for 2014. The same values are used in the following years. Other configurations can be selected.

By using GHGenius, it was possible to model an average U.S. dry-

Table 6

Primary agricultural, industrial and transportation parameters for wheat ethanol.

	Unit	GHGenius	BioGrace ^a
Inputs per tonne of wheat			
N fertilizer	kg	18.0	21.0
P ₂ O ₅ fertilizer	kg	10.3	4.2
K ₂ O fertilizer	kg	0.8	3.1
Diesel (machinery operation)	L	8.5	19.8
Wheat transportation	km	100	50
Inputs per L of ethanol			
Electricity	MJ	10.8	-5.6
Natural gas	MJ	13.4	20.2
Ethanol transportation	km	121 (truck 100%) additional 802 (rail 22%)	150 (truck 100%)

^a A factor of +40% is applied to industrial inputs for BioGrace to encourage voluntary data disclosure contribution from the private sector. Industrial inputs considering a configuration with steam production from a natural gas CHP system.

mill corn ethanol facility in 2015 by selecting the U.S. as the modeling location. In GHGenius, the base year for which default data for corn agricultural production and the amount of inputs was originally developed is 1994. The model adjusts these inputs for 2015 according to trends from 1964 to 2011 [78].

BioGrace models an average EU dry-mill corn ethanol facility powered by a natural gas combined heat and power (CHP) system using the default 2008/2009 dataset derived from Neeft [10]. Table 5 compares the primary model parameters used in the three models for corn and ethanol production and transportation.

3.2.3. Wheat ethanol

Only GHGenius and BioGrace include a wheat ethanol pathway. The GHGenius model assumes an average-wheat ethanol facility using natural gas CHP for steam and power production, with on-site electricity generation. Similar to the corn ethanol pathway, the base year for wheat agricultural production and the amount of inputs used in the U.S. is 1994 extrapolated to 2015. BioGrace models five types of wheat ethanol plants: four with on-site electricity generation (unspecified process fuel, steam from lignite CHP, steam from straw CHP, and steam from natural gas CHP) and one with electricity consumption from the grid (steam from natural gas boiler). For comparative reasons, steam from CHP was selected for evaluation in this study using the default 2008/2009 dataset based on Neeft [10]. Data for wheat and ethanol production is summarized in Table 6. The different locations in part explain the variations in model inputs, such as fertilizer application

rates and electricity usage.

3.3. Comparison of calculated GHG emissions

Differences in net life-cycle GHG emissions impacts across the models are shown in Fig. 3. Impacts values for the various categories are found in Table 6S of the supplementary material.

Brazilian sugarcane ethanol estimated life-cycle GHG emissions vary as much as 65% across models, with GHGenius presenting the highest value at 45.1 g CO₂eq per MJ of ethanol compared to 16-24 g CO₂eq per MJ obtained with the other models. For GHGenius, the main differences observed in comparison to the other models result from data on fuel and energy use, limestone, and fertilizer GHG emissions at the agricultural stage (farming), as well as low energy efficiency bagasse combustion at the industrial stage. GHG emissions from the use of diesel are responsible for 7.8 g CO2eq per MJ of ethanol according to GHGenius, while GREET, BioGrace, and VSB estimate 4.3, 1.3, and 2.2 g CO2eq per MJ of ethanol, respectively. As expected, the GHGenius model results for limestone application are 4.9 g CO₂eq per MJ of ethanol, which is 12 times compared to BioGrace (0.4 g CO₂eq per MJ), with GREET (1.3 g CO₂eq per MJ) and VSB (0.9 g CO₂eq per MJ) having intermediate values. Fertilizer and soil N2O emissions are estimated at 10.8 g CO₂eq per MJ of ethanol by GHGenius in comparison to 1.7 by GREET, 3.3 by BioGrace, and 7.9 g by VSB (in g CO2eq per MJ). GHGenius also includes GHG emissions from sugarcane bagasse combustion (5.1 g CO_2eq per MJ of ethanol), whereas the other models consider carbon emissions from bagasse combustion as biogenic and, therefore, do not include them in the life-cycle results; GREET calculates them.

The amount of diesel used in machinery operation (in L per tonne of

sugarcane) was estimated as 2.9 by GHGenius, which was higher than the 1.1 by GREET or 0.8 by BioGrace, and 1.9 by VSB, as shown in Table 4. In addition to the amount used, GHGenius estimated a higher life-cycle GHG emissions factor for diesel (production and combustion in agricultural machinery) when compared to GREET, BioGrace, and VSB: 116.4 in GHGenius, 90.2 in GREET, 87.6 in BioGrace, and 81.6 in VSB, all in g CO₂eq per MJ of diesel fuel, as shown in Table 3. In the case of limestone, a similar explanation is valid. The discrepancy observed for the impact calculated by GHGenius in comparison to the other models may be attributable to two main factors, such as the higher application rate of lime (Table 4) and the much larger upstream life-cycle impact value associated with limestone (Table 3).

Ethanol shipping is also a very important category for sugarcane ethanol; values calculated through GREET, BioGrace, and VSB would be similar with a maximum variation of 15%: (in g CO_2eq per MJ of ethanol) 16.8 for GREET, 18.9 for BioGrace, and 16.1 for VSB.

The influence of the approach for the treatment of coproducts utilized by the models when evaluating sugarcane ethanol is minor, as shown in Table 7S of the supplementary material. Excess electricity generated in the production of sugarcane ethanol corresponds to 5% of the total impact in GREET (energy allocation), 0% in BioGrace (energy allocation), 9% in GHGenius (substitution method), and 4% in VSB (economic allocation). Results from the impact of excess electricity production in this paper are specifically linked to the default assumptions considered in the investigated models. Khatiwada et al. [14] stated that, if the system expansion method (substitution method) were adopted, the credits associated with electricity generated could even offset all the GHG emissions resulting from ethanol production in Brazil. However, the calculation of such credits depends on the selection of several factors and assumptions (e.g. average electricity



Fig. 3. Greenhouse gas emissions impacts of ethanol produced from sugarcane, corn, and wheat in g CO₂eq per MJ of ethanol calculated by GREET, GHGenius, BioGrace, and VSB models.

generation by biorefineries, type of biomass used as feedstock (sugarcane bagasse and/or straw), electricity mix considered, and GHG emissions related to the sources of electricity considered), which are associated with uncertainties and variabilities, and whether it substitutes base or marginal electricity production. In addition to the electricity generated as a coproduct, the majority of sugarcane biorefineries in Brazil coproduce ethanol and sugar [32] – considering this fact could also lead to variability in the partition of the GHG emissions impacts obtained. The VSB model can account for variations in the portfolio of the products of a sugarcane biorefinery, as well as for the use of additional feedstocks for electricity production (in field mechanized sugarcane harvest) [39].

For corn ethanol, life-cycle GHG emissions obtained vary as much as 30% across models, with BioGrace estimating 43.4 (specific process), GHGenius 61.9 (average) and GREET 57.7 (country average) g CO₂eq per MJ of ethanol, respectively. The main differences observed in this case are due to the treatment of the coproducts utilized. The default method used by BioGrace (energy) led to a 50% partitioning of GHG emissions between ethanol and its coproducts, whereas the substitution method used by GREET and GHGenius provides a credit of 12.8 and 16.7 g CO₂eq per MJ, respectively, for non-energy products to ethanol (Fig. 3 and Table 6S). GHGenius estimates GHG emissions from fertilizers at 20.2 g CO₂eq per MJ of ethanol in comparison to 16.2 g by GREET and 4.3 g by BioGrace. The use of off-site electricity, coal, and other inputs required for corn ethanol production resulted in a total of 18.1 g CO₂eq per MJ of ethanol by GREET, 15.9 g by GHGenius, and 0 g by BioGrace (electricity from a natural gas steam plant generates corn

ethanol power needs without crediting excess electricity produced). Table 5 shows the differences in fertilizers, limestone, and diesel inputs at the agricultural stage, as well as the demand for electricity and fuels in the industrial stage between BioGrace and the other two models.

The same observations made for corn are valid for wheat ethanol: BioGrace presented the lowest GHG emissions impacts due to the treatment of the coproducts utilized. Additionally, the discrepancy presented in terms of the inputs considered by BioGrace and GHGenius, in this case, may be explained by the fact that BioGrace contemplates the production of wheat in the EU, whereas GHGenius considers wheat produced in the U.S.

3.4. Harmonization

The procedure to harmonize parameters and assumptions generated similar impact values calculated by the models (Fig. 4). Note that all following numbers are presented in g CO_2eq per MJ of ethanol. Upon harmonization, default impacts for sugarcane ethanol (24.0 by GREET, 23.0 by BioGrace, and 43.9 by GHGenius) changed to values close to 16.1 calculated with VSB assumptions (17.5 by GREET, 17.3 by BioGrace, and 17.2 by GHGenius) for ethanol produced in Brazil, representing a maximum variation of 8%. In terms of corn ethanol, considering the average of dry and wet corn milling facilities, the results obtained with default parameters (43.4 by BioGrace and 61.9 by GHGenius) changed to values close to 57.7 calculated by GREET (57.0 by BioGrace and 56.2 by GHGenius), achieving a maximum variation of 3%.



Fig. 4. Steps toward harmonization of public GHG emissions models for ethanol from sugarcane (using VSB as a basis) in Brazil (top) and corn ethanol (using GREET as a basis) in the U.S. (bottom).

The results obtained in this work are in line with the previous harmonization study by Chum and Warner [24]. In that study, impacts for corn ethanol produced in dry milling facilities (43.4 by BioGrace and 62.8 by GHGenius) changed to 54.1 by BioGrace and 53.7 by GHGenius within a maximum variation of results of 2.5% in comparison with the value of 52.5 by GREET.

3.5. Methodology comments

Although few LCA models and feedstocks were investigated to produce ethanol, the conclusions from this study are broadly applicable to LCA work carried out in a regulatory/policy context. The feedstocks selected for ethanol currently provide more than 90% of the ethanol produced and used globally [79].

Results of the harmonization emphasize that to understand the comparisons of LCA results from commercial biofuels one must recognize: the geographic and temporal context for feedstock agronomic production and of the performance of the industrial sector as technologies used for biofuel production improved over time (as it is the case for the two major feedstocks addressed here, as also shown by Chum et al. [80,81]; the product slate of the biorefinery evolves; the approach for modeling land use and coproduct credits; broad differences (e.g., uses natural gas instead of coal) or gaps (e.g., no pesticide or adoption of different country agricultural production data for lack of specific country data) in inventories; the scope and geographic context of the supply chain for chemicals, infrastructure, and biofuel transport and use (can be different than the production).

Some models have been continually improved (e.g., GREET), including relevant commercial pathways, while others lag, for instance, from 2013 GHGenius (used in the study) was updated in 2018. Default data in BioGrace is out of date and may not reflect the current commercial practices. However, BioGrace has a different purpose as it encourage users to provide their own data.

Regarding sugarcane ethanol production in Brazil, for instance, the amount of surplus electricity generated as a coproduct in a number of biorefineries could be included in the calculation; however, it is not an option in BioGrace as the EU regulation does not consider this aspect. This is an example of the tool following a directive that is not a research tool. This distinction could be clearer for researchers in the model documentation.

Significant progress has been accomplished with the GREET.net improved graphic interface that makes navigation easier for these complex models with intricately linked spreadsheets. Continual updating is needed to decrease inconsistencies in comparing web and spreadsheet results (e.g., field-burned straw). User-friendly interfaces for non-LCA experts would allow users to choose among options (treatment of coproducts, transportation distances, type of vehicles, etc.) to interpret results. Results obtained by the models presented by life-cycle stages, for instance, would make visualization of the influence of specific inputs on results more informative. This is just an example of presentation of results to facilitate the comparison across models/conditions to identify most critical inputs to GHG impacts.

4. Conclusions

Our work shows that LCA tools calculate similar results when harmonized to the maximum possible extent. The results of the harmonization procedure appear encouraging in the context of policies dealing with GHG emissions reduction targets, given that relatively few modification steps of identified parameters and assumptions were used to generate similar GHG emissions results for ethanol. On the other hand, the same fact shows that the models are highly sensitive to these assumptions and methodological choices, which may be interpreted as a risk factor in the policy arena.

The elaboration and application of the life-cycle GHG emissions assessment tools ultimately align with the biofuels regulations or directives defined by governments individually or regionally, which vary and may have different and/or conflicting requirements. Meanwhile, different results obtained by using such models suggest that modeling tools should provide transparent data sources and assumptions used in LCA calculations, to facilitate the understanding of results in terms of geographical locations of production of biomass and biofuels consumption. Regional directive requirements have harmonized data for specific regions (EU); states have set specific tools for their use (California); or Canadian provinces similarly have set up requirements as they implement and measure progress against stated objectives. Requirements change as more scientific information emerges that justifies changes. BioGrace is a public tool specifically designed for assessment of commercial bioenergy systems in the European Union. The other three tools used started and continue to be used as research tools, able to assess technology improvements (or feedstock supply improvements), or to compare biofuel pathways in scenario analyses.

Review papers discussing biofuels GHG emission reductions must consider data sources and clarify the models utilized, because their results depend on the purpose of the LCA, the boundaries of the systems investigated, the stage of technology development, the source of biomass production data (from small plots, field trial or commercial production), the locations across the supply chain of biomass, and the use of resulting fuels.

5. Recommendations

This study recommends strengthened efforts to strive for transparency about the structures and underlying assumptions of LCA models, their calculation mechanisms, and in the reporting of results. To this end, around the world efforts should be redoubled within stakeholder industries, the LCA/GHG modeler community, governmental entities, and multi-lateral governmental and non-governmental organizations (e.g., CRC, Inc.) to work together to increase transparency of models as well as to reach consensus on key areas such as treatment of coproducts. Similarly, efforts to increase the understanding of climate effects on emissions must continue to improve these tools (and many organizations are working towards this end). This study mainly expresses the perspective of the LCA model user community, oppose to the view of a model developer.

It would facilitate comparisons if the structure of the models and their presentation followed consolidated instructions for LCA studies (or well defined), providing clear definitions of study scope and major functional unit metrics being assessed (e.g., GHG emissions as CO_2eq per MJ and/or per km) as well as explicit detail about the life-cycle dataset and inventories used. This would also facilitate the understanding of the main assumptions of the models. Similarly, built-in conversion of results from the individual model units to some common functional unit to facilitate mutual understanding of results across models and regions, not just by modelers but by the entire user and stakeholder communities.

Life-cycle inventories should be continually updated and improved to incorporate the most data on biomass feedstock productivity, taking into account location-specific agricultural practices as well as the vintage of industrial technologies being used. Research communities, the other hand, should provide clear indication on the technology stage of development (and thus level of confidence/uncertainty in data used), and biomass feedstock development stage (for studies examining prospective future scenarios).

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Authors' contributions

All six authors contributed equally to the conception and design of this study, interpretation of the results, and editing of the text.

Conflicts of interest

The authors declare no competing financial interests.

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Appendix A. Supplementary data

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References

- [1] De Jong S, Antonissen K, Hoefnagels R, Lonza L, Wang M, Faaij A, et al. Life-cycle analysis of greenhouse gas emissions from renewable jet fuel production. Biotechnol Biofuels 2017;10:1–18. https://doi.org/10.1186/s13068-017-0739-7.
- [2] IEA. IEA bioenergy annual report 2016 2016 n.d https://www.ieabioenergy.com/ publications/iea-bioenergy-annual-report-2016/, Accessed date: 1 May 2017.
- [3] EPA US. Lifecycle analysis of greenhouse gas emissions under the renewable fuel standard. 2016https://www.epa.gov/renewable-fuel-standard-program/lifecycleanalysis-greenhouse-gas-emissions-under-renewable-fuel, Accessed date: 20 May 2017.
- [4] Hickox WH, Werner B, Gaffney P. Air Resour Board 2014:1–6.
- [5] European Commission. Renewable energy directive. 2017https://ec.europa.eu/ energy/en/topics/renewable-energy/renewable-energy-directive, Accessed date: 20 May 2017.
- [6] Macedo IC, Nassar AM, Cowiec AL, Seabra JEA, Marelli L, Otto M, et al. Greenhouse

gas emissions from bioenergy. In: Souza GM, Victoria RL, Joly CA, Verdade L, editors. Bioenergy sustain. Bridg. Gaps. Sao Paulo; 2015. p. 583–616.

- [7] (S&T)2 consultants inc. GHGenius model 4.03 volume 1: model background and structure. 2013. p. 445.
- [8] (S&T)2 consultants inc. GHGenius model 4.03 volume 2: data and data sources. 2013. p. 575.
- [9] ANL. GREET model. 2017https://greet.es.anl.gov/, Accessed date: 20 September 2018.
- [10] EU. Biograce. 2015http://www.biograce.net/content/ghgcalculationtools/ recognisedtool/, Accessed date: 20 April 2017.
- [11] Canter CE, Dunn JB, Han J, Wang Z, Wang M. Policy implications of allocation methods in the life cycle analysis of integrated corn and corn stover ethanol production. Bioenergy Res 2016;9:77–87. https://doi.org/10.1007/s12155-015-9664-4.
- [12] Cherubini F, Strømman AH, Ulgiati S. Influence of allocation methods on the environmental performance of biorefinery products - a case study. Resour Conserv Recycl 2011;55:1070–7. https://doi.org/10.1016/j.resconrec.2011.06.001.
- [13] Wang L, Quiceno R, Price C, Malpas R, Woods J. Economic and GHG emissions analyses for sugarcane ethanol in Brazil: looking forward. Renew Sustain Energy Rev 2014;40:571–82. https://doi.org/10.1016/j.rser.2014.07.212.
- [14] Khatiwada D, Seabra J, Silveira S, Walter A. Accounting greenhouse gas emissions in the lifecycle of Brazilian sugarcane bioethanol: methodological references in European and American regulations. Energy Policy 2012;47:384–97. https://doi. org/10.1016/j.enpol.2012.05.005.
- [15] Luo L, van der Voet E, Huppes G. Life cycle assessment and life cycle costing of bioethanol from sugarcane in Brazil. Renew Sustain Energy Rev 2009;13:1613–9. https://doi.org/10.1016/j.rser.2008.09.024.
- [16] Koponen K, Soimakallio S, Kline KL, Cowie A, Brandão M. Quantifying the climate effects of bioenergy – choice of reference system. Renew Sustain Energy Rev 2018;81:2271–80. https://doi.org/10.1016/j.rser.2017.05.292.
- [17] Hill J, Polasky S, Nelson E, Tilman D, Huo H, Ludwig L, et al. Climate change and health costs of air emissions from biofuels and gasoline. Proc Natl Acad Sci U S A 2009;106:2077–82. https://doi.org/10.1073/pnas.0812835106.
- [18] Flugge MJ, Lewandrowski J, Rosenfeld J, Boland C, Hendrickson T, Jaglo K, et al. A life-cycle analysis of the greenhouse gas emissions of corn-based ethanol. 2017. p. 185. Rep Prep by ICF under USDA Contract No AG-3142-D-16-0243.
- [19] Wang M, Han J, Dunn JB, Cai H, Elgowainy A. Well-to-wheels energy use and greenhouse gas emissions of ethanol from corn, sugarcane and cellulosic biomass for US use. Environ Res Lett 2012;7. https://doi.org/10.1088/1748-9326/7/4/ 045905.
- [20] Wang MQ, Han J, Haq Z, Tyner WE, Wu M, Elgowainy A. Energy and greenhouse gas emission effects of corn and cellulosic ethanol with technology improvements and land use changes. Biomass Bioenergy 2011;35:1885–96. https://doi.org/10. 1016/j.biombioe.2011.01.028.
- [21] Cavalett O, Chagas MF, Seabra JEA, Bonomi A. Comparative LCA of ethanol versus gasoline in Brazil using different LCIA methods. Int J Life Cycle Assess 2013;18:647–58. https://doi.org/10.1007/s11367-012-0465-0.
- [22] Seabra JEA, Macedo IC, Chum HL, Faroni CE, Sarto CA. Life cycle assessment of Brazilian sugarcane products: GHG emissions and energy use. Biofuels, Bioprod Biorefining 2011;5:519–32. https://doi.org/10.1002/bbb.289.
- [23] Hennecke AM, Faist M, Reinhardt J, Junquera V, Neeft J, Fehrenbach H. Biofuel greenhouse gas calculations under the European renewable energy directive – a comparison of the BioGrace tool vs. the tool of the roundtable on sustainable biofuels. Appl Energy 2013;102:55–62. https://doi.org/10.1016/j.apenergy.2012.04. 020.
- [24] Chum H, Warner E. Tools for greenhouse gas (GHG) assessment for biofuels: a comparison. IEA bioenergy conf. 2015http://ieabioenergy2015.org/fileadmin/ veranstaltungen/2015/IEA_Bioenergy_Conference/S08-4_Chum.pdf, Accessed date: 20 May 2017.
- [25] (S&T)2 Consultants I, Coordinating Research Council I. Transportation fuel life cycle assessment: validation and uncertainty of well-to-wheel GHG estimates. 2013. p. 293.
- [26] Unnasch S, Riffel B, Sanchez S, Waterland L. Review of transportation fuel life cycle analysis. 2011.
- [27] Bonomi A, Cavalett O, Pereira da Cunha M, Lima MAP. Virtual biorefinery. Cham: Springer International Publishing; 2016. https://doi.org/10.1007/978-3-319-26045-7
- [28] International Organization for Standardization ISO 14040. Environmental management – life-cycle assessment – principles and framework. 2006https://www.iso. org/standard/37456.html.
- [29] Standardization IO for ISO 14044. Environmental management life-cycle assessment – requirements and guidelines. 2006https://www.iso.org/standard/38498. html.
- [30] Delucchi M. A lifecycle emissions model (LEM): lifecycle emissions from transportation fuels, motor vehicles, transportation modes, and electricity. 2003https:// escholarship.org/uc/item/9vr8s1bb, Accessed date: 15 May 2017.
- [31] Joint European Commission. Well-to-wheels analysis of future automotive fuels and powertrains in the european context. 2014http://iet.jrc.ec.europa.eu/about-jec/, Accessed date: 15 April 2017.
- [32] Cavalett O, Junqueira TL, Dias MOS, Jesus CDF, Mantelatto PE, Cunha MP, et al. Environmental and economic assessment of sugarcane first generation biorefineries in Brazil. Clean Technol Environ Policy 2012;14:399–410. https://doi.org/10. 1007/s10098-011-0424-7.
- [33] Pereira LG, Chagas MF, Dias MOS, Cavalett O, Bonomi A. Life cycle assessment of butanol production in sugarcane biorefineries in Brazil. J Clean Prod 2015;96:557–68. https://doi.org/10.1016/j.jclepro.2014.01.059.

- [34] Pereira LG, Dias MOS, Junqueira TL, Chagas MF, Cavalett O, Filho RM, et al. Production of butanol and other high valued chemicals using ethanol as feedstock integrated to a first and second generation sugarcane distillery. Chem Eng Trans 2014;37:805–10. https://doi.org/10.3303/CET1437135.
- [35] Pereira LG, Dias MOS, Junqueira TL, Pavanello LG, Chagas MF, Cavalett O, et al. Butanol production in a sugarcane biorefinery using ethanol as feedstock. Part II: integration to a second generation sugarcane distillery. Chem Eng Res Des 2014;92:1452–62. https://doi.org/10.1016/j.cherd.2014.04.032.
- [36] Pereira LG, Dias MOS, MacLean HL, Bonomi A. Investigation of uncertainties associated with the production of n-butanol through ethanol catalysis in sugarcane biorefineries. Bioresour Technol 2015;190:242–50. https://doi.org/10.1016/j. biortech.2015.04.095.
- [37] Pereira LG, Dias MOS, Mariano AP, Maciel Filho R, Bonomi A. Economic and environmental assessment of n-butanol production in an integrated first and second generation sugarcane biorefinery: fermentative versus catalytic routes. Appl Energy 2015;160:120–31. https://doi.org/10.1016/j.apenergy.2015.09.063.
- [38] Moraes BS, Junqueira TL, Pavanello LG, Cavalett O, Mantelatto PE, Bonomi A, et al. Anaerobic digestion of vinasse from sugarcane biorefineries in Brazil from energy, environmental, and economic perspectives: profit or expense? Appl Energy 2014;113:825–35. https://doi.org/10.1016/j.apenergy.2013.07.018.
- [39] Cardoso T de F, Cavalett O, Chagas MF, Morais ER de, Carvalho JLN, Franco HCJ, et al. Technical and economic assessment of trash recovery in the sugarcane bioenergy production system. Sci Agric 2013;70:353–60. https://doi.org/10.1590/ S0103-90162013000500010.
- [40] Cavalett O, Chagas MF, Junqueira TL, Watanabe MDB, Bonomi A. Environmental impacts of technology learning curve for cellulosic ethanol in Brazil. Ind Crops Prod 2017;106:31–9. https://doi.org/10.1016/j.indcrop.2016.11.025.
- [41] Junqueira TL, Chagas MF, Gouveia VLR, Rezende MCAF, Watanabe MDB, Jesus CDF, et al. Techno-economic analysis and climate change impacts of sugarcane biorefineries considering different time horizons. Biotechnol Biofuels 2017;10:50. https://doi.org/10.1186/s13068-017-0722-3.
- [42] Souza A, Watanabe MDB, Cavalett O, Ugaya CML, Bonomi A. Social life cycle assessment of first and second-generation ethanol production technologies in Brazil. Int J Life Cycle Assess 2018;23:617–28. https://doi.org/10.1007/s11367-016-1112-y.
- [43] Watanabe MDB, Chagas MF, Cavalett O, Guilhoto JJM, Griffin WM, Cunha MP, et al. Hybrid input-output life cycle assessment of first- and second-generation ethanol production technologies in Brazil. J Ind Ecol 2016;20:764–74. https://doi. org/10.1111/jiec.12325.
- [44] Dias MOS, Junqueira TL, Cavalett O, Cunha MP, Jesus CDF, Rossell CEV, et al. Integrated versus stand-alone second generation ethanol production from sugarcane bagasse and trash. Bioresour Technol 2012;103:152–61. https://doi.org/10.1016/j. biortech.2011.09.120.
- [45] Mariano AP, Dias MOS, Junqueira TL, Cunha MP, Bonomi A, Filho RM. Butanol production in a first-generation Brazilian sugarcane biorefinery: technical aspects and economics of greenfield projects. Bioresour Technol 2013;135:316–23. https:// doi.org/10.1016/j.biortech.2012.09.109.
- [46] Mariano AP, Dias MOS, Junqueira TL, Cunha MP, Bonomi A, Filho RM. Utilization of pentoses from sugarcane biomass: techno-economics of biogas vs. butanol production. Bioresour Technol 2013;142:390–9. https://doi.org/10.1016/j.biortech. 2013.05.052.
- [47] Dias MOS, Pereira LG, Junqueira TL, Pavanello LG, Chagas MF, Cavalett O, et al. Butanol production in a sugarcane biorefinery using ethanol as feedstock. Part I: integration to a first generation sugarcane distillery. Chem Eng Res Des 2014;92:1441–51. https://doi.org/10.1016/j.cherd.2014.04.030.
- [48] IPCC. Climate change 2001: the scientific basis. Cambridge University Press; 2001.
- [49] IPCC. Climate change 2007: the physical science basis. Fourth assessment report (AR4) vol. 996. 2007http://www.ipcc.ch/publications_and_data/ar4/syr/en/ contents.html, Accessed date: 20 September 2018.
- [50] IPCC. Climate change 2013: the physical science basis. New York: Cambridge University Press; 2013.
- [51] IPCC. The IPCC scientific assessment. New York: Cambridge University Press; 1990.
- [52] IPCC. Climate change 1995: the science of climate change. New York: Cambridge University Press; 1995.
- [53] Finnveden G, Hauschild MZ, Ekvall T, Guinée J, Heijungs R, Hellweg S, et al. Recent developments in life cycle assessment. J Environ Manag 2009;91:1–21. https://doi. org/10.1016/j.jenvman.2009.06.018.
- [54] Ekvall T, Weidema BP. System boundaries and input data in consequential life cycle inventory analysis. Int J Life Cycle Assess 2004;9:161–71. https://doi.org/10.1007/ BF02994190.
- [55] Guinée JB, Gorrée M, Heijungs R, Huppes G, Kleijn R KA de. Handbook on life-cycle assessment. Operational guide to the ISO standards. Dordrecht: Kluwer Academic Publishers; 2002.
- [56] Heijungs R, Guinée JB. Allocation and 'what-if' scenarios in life cycle assessment of waste management systems. Waste Manag 2007;27:997–1005. https://doi.org/10. 1016/j.wasman.2007.02.013.

- [57] Wang M, Huo H, Arora S. Methods of dealing with co-products of biofuels in lifecycle analysis and consequent results within the U.S. context. Energy Policy 2011;39:5726–36. https://doi.org/10.1016/j.enpol.2010.03.052.
- [58] International Organization for Standardization ISO 13065. Sustainability Criteria for bioenergy. 2015https://www.iso.org/standard/52528.html, Accessed date: 24 May 2017.
- [59] Econvent. Econvent. 2010http://www.econvent.org/database/older-versions/ econvent-version-2/reports-on-econvent-2/reports-on-econvent-2.html, v2.2, Accessed date: 23 January 2017.
- [60] Stehfest E, Bouwman L. N2O and NO emission from agricultural fields and soils under natural vegetation: summarizing available measurement data and modeling of global annual emissions. Nutrient Cycl Agroecosyst 2006;74:207–28. https://doi. org/10.1007/s10705-006-9000-7.
- [61] IPCC. N2O emissions from managed soils, and CO2 emissions from lime and urea application. Guidel Natl Greenh Gas Invent 2006:54.
- [62] ANL. Carbon calculator for land use change from biofuels production. CCLUB); 2015https://greet.es.anl.gov/publication-cclub-manual, Accessed date: 5 May 2017.
- [63] Taheripour F, Tyner WEWM. Global land use change due to the U.S. cellulosic biofuels program simulated with the GTAP model. 2011https://greet.es.anl.gov/ files/luc_ethanol, Accessed date: 5 May 2017.
- [64] van Deusen PCHL. COLE web applications suite. 2013http://www.ncasi2.org/ COLE/, Accessed date: 3 May 2017.
- [65] Harris NL, Grimland SBS. Land use change and emission factors: updates since proposed RFS rule. 2009.
- [66] Kwon H-Y, Hudson RJM. Quantifying management-driven changes in organic matter turnover in an agricultural soil: an inverse modeling approach using historical data and a surrogate CENTURY-type model. Soil Biol Biochem 2010;42:2241–53. https://doi.org/10.1016/j.soilbio.2010.08.025.
- [67] El Akkari M, Réchauchère O, Bispo A, Gabrielle B, Makowski D. A meta-analysis of the greenhouse gas abatement of bioenergy factoring in land use changes. Sci Rep 2018. https://doi.org/10.1038/s41598-018-26712-x.
- [68] DeCicco JM, Liu DY, Heo J, Krishnan R, Kurthen A, Wang L. Carbon balance effects of U.S. biofuel production and use. Clim Change 2016. https://doi.org/10.1007/ s10584-016-1764-4.
- [69] Hudiburg TW, Wang W, Khanna M, Long SP, Dwivedi P, Parton WJ, et al. Impacts of a 32-billion-gallon bioenergy landscape on land and fossil fuel use in the US. Nat Energy 2016. https://doi.org/10.1038/nenergy.2015.5.
- [70] Jain AK, Verburg PH, Popp A, Tabeau A, Prestele R, Rounsevell MDA, et al. Hotspots of uncertainty in land-use and land-cover change projections: a globalscale model comparison. Glob Chang Biol 2016. https://doi.org/10.1111/gcb. 13337.
- [71] Dunn JB, Qin Z, Wander MM, Mueller S, Kwon H. Soil carbon sequestration and land use change associated with biofuel production: empirical evidence. GCB Bioenergy 2014. https://doi.org/10.1111/gcbb.12237.
- [72] Maia SMF, Davies CA, Bernoux M, Cerri CC, Cerri CEP, Galdos MV, et al. Payback time for soil carbon and sugar-cane ethanol. Nat Clim Change 2014. https://doi. org/10.1038/nclimate2239.
- [73] Fargione J, Hill J, Tilman D, Polasky S, Hawthorne P. Land clearing and the biofuel carbon debt. Science 2008;80:1235–8. https://doi.org/10.1126/science.1152747. 319.
- [74] Macedo IC, Leal MRLV, da Silva J. Greenhouse gases emissions in the production and use of ethanol from sugarcane in Brazil. 2004 (in Portuguese). www.unica.com. br/download.php?idSecao = 17&id = 11168105, Accessed date: 17 September 2017.
- [75] Macedo IC, Seabra JEA, Silva JEAR. Green house gases emissions in the production and use of ethanol from sugarcane in Brazil: the 2005/2006 averages and a prediction for 2020. Biomass Bioenergy 2008;32:582–95. https://doi.org/10.1016/j. biombioe.2007.12.006.
- [76] UDOP. Legislation for sugarcane straw burning (in Portuguese) n.d. http://www.udop.com.br/index.php?item=legislacao1&op=40 (accessed April 1, 2017).
 [77] Wang M, Wu M, Huo H. Life-cycle energy and greenhouse gas emission impacts of
- [77] Wang M, Wu M, Huo H. Life-cycle energy and greenhouse gas emission impacts of different corn ethanol plant types. Environ Res Lett 2007;2:024001https://doi.org/ 10.1088/1748-9326/2/2/024001.
- [78] U.S. Department of Agriculture (USDA). Economic research service (ERS): corn and other feedgrains. 2011https://www.ers.usda.gov/topics/crops/corn/, Accessed date: 12 December 2017.
- [79] U.S. Department of Energy. Maps and data global ethanol production. 2016https://www.afdc.energy.gov/data/10331, Accessed date: 17 September 2018.
- [80] Chum HL, Zhang Y, Hill J, Tiffany DG, Morey RV, Goss Eng A, et al. Understanding the evolution of environmental and energy performance of the US corn ethanol industry: evaluation of selected metrics. Biofuels, Bioprod Biorefining 2014;8:224–40. https://doi.org/10.1002/bbb.1449.
- [81] Chum HL, Warner E, Seabra JEA, Macedo IC. A comparison of commercial ethanol production systems from Brazilian sugarcane and US corn. Biofuels, Bioprod Biorefining 2014;8:205–23. https://doi.org/10.1002/bbb.1448.