



Review

Life Cycle Assessment of Bioethanol Production: A Review of Feedstock, Technology and Methodology

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Abstract: So far, a lot of efforts have been put in life cycle assessments (LCA) of bioethanol production. There are many works that have assessed bioethanol production in different points of view to illustrate the environmental impacts. This study reviewed former LCA studies on bioethanol produced from various biomass resources by considering the effect of methodological components, technical pathways and feedstock provision on the result of LCA studies. The review evaluated 48 papers published 2002–2021 with a focus on studies that included a complete set of environmental impact categories. However, due to lack of harmony among studies, comparing the LCA results was challenging but the review indicated that the final results of studies are influenced by LCA methodological components, such as system boundary, functional unit, etc. Around 80% of the reviewed papers show the reduction in global warming potential, while contrary results have been found about increasing acidification, eutrophication and photochemical oxidant formation impact categories because of the feedstock provision. Regarding technical aspects, results from the review revealed that most of the studies considered the pre-treatment as a crucial step in bioconversion processes. Despite several LCA studies of bioethanol production, there is still low attention given to uncertainty analysis in the publications.

Keywords: LCA; biofuel; methodological components; feedstock provision; technical aspects



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1. Introduction

Over the past few years, the policy context for renewable energy has changed considerably [1]. In this regard, international institutions help the countries to set the necessary targets towards a higher renewables' deployment. Renewable Energy Directive (RED) 2018/2001 of the European Parliament addressed energy generation from renewable sources in member countries until 2030 [2]. Further, organizations such as REN21: Renewables Now! International Renewable Energy Agency and International Energy Agency by providing up-to-date information support decision-makers from politics and industry for a sustainable renewable energy future.

Amongst the targets and strategy setting for renewable energies, the development of biofuels, including bioethanol, has also been considered. Bioethanol is the main renewable energy carrier after biodiesel in Europe [3] that can be produced from a variety of biomass feedstock, including, e.g., energy crops and lignocellulosic biomass. The United States is the world's largest producer of bioethanol. Brazil and the European Union are ranked in the second and third place, respectively [4]. The vast majority of U.S. bioethanol is produced from corn, while Brazil primarily uses sugarcane [4], and corn is the main crop for bioethanol production in Europe [5]. The substitution of fossil fuels started with ethanol produced from food crops containing starch or sugar as the first-generation bioethanol. However, the conversion of food crops into bioethanol raised concerns about food security

on a global scale [6,7]. Therefore, a second-generation of bioethanol based on non-food resources like lignocelluloses is under development. The following sections will present a summary of bioethanol production from various sources.

- Starch based bioethanol

Crops such as corn, wheat or barley are known as starch biomass with the purpose of producing ethanol. Moreover, other kind of starchy crops like cassava in tropical areas can be used for production of ethanol. In starchy materials polymers of glucose require to break down to fermentable sugars through a reaction with water and adding enzymes. Then enzymatic hydrolysis is followed by fermentation, distillation and dehydration to yield anhydrous ethanol.

- Sugar based bioethanol

Sugar-based feedstock such as sugarcane, sugar beets, sweet sorghum and molasses are potential raw materials used for ethanol production. They contain a large proportion of simple sugars that leads to cost reduction of conversion processes. Conversion starts by feedstock grinding to extract the sugar, then is followed by adding yeast for the fermentation process.

- Lignocellulose based bioethanol

Lignocellulosic materials, including agricultural and forestry residues, are abundant all over the world. They can be used to produce ethanol as second-generation bioethanol. Lignocellulose is a more complex substrate than sugar and starch-based materials. It is composed of three fractions, including cellulose, hemicellulose and lignin. Conversion of carbohydrate polymer to fermentable sugars is not easy since ethanol production from lignocellulosic biomass is a multi-stage conversion.

The most important reasons for bioethanol's widespread deployment are environmental challenges caused by fossil fuels and their depletion [6,8]. It is expected that switching to renewable sources like bioethanol reduces impacts on the environment. However, its production may have several impacts depending on feedstock type, local conditions, design and implementation of the respective conversion process [9]. Hence, to identify the environmental burdens of the bioethanol production system, a comprehensive evaluation is required. Life cycle assessment, defined according to ISO14040 and 14044 [9,10], is mainly used to identify and measure the environmental impact towards sustainability [11]. Several LCA studies have been carried out to determine savings in energy and emissions throughout the life cycle of bioethanol production.

The main aim of this paper is an overview of previous LCA studies for bioethanol production, followed by results on methodological and technological issues related to crop production based on the following sections. Moreover, the purpose of this review is to recognize bottlenecks and improve future research works by investigating the influence of different parameters on the final results of LCA.

2. Method

2.1. Approach of Review

In recent years, many LCA studies of bioethanol production were conducted. Each of them used different objectives, various feedstock, varying technological approaches, and different methodological components. Therefore, comparing their results and finding the best practice is a challenging task. In the first step of the overview, records were identified searching through Google Scholar, ResearchGate and SCOPUS.

The search approach was conducted on joining two key expressions: "life cycle assessment" and "bioethanol production". After removing the duplicated and unrelated items, the papers not meeting the inclusion/exclusion criteria were removed at this stage. Then, full-text assessment started to check eligibility. Papers were selected when they revealed LCA was carried out, feedstock provided from 1G and 2G agro-based substrates, a set of environmental impact categories was analyzed, as well as the biochemical processes

used in the biorefinery. Finally, eligible studies were included in a qualitative analysis of the effect of feedstock provision and conversion processes on LCA results, as well as methodological choices.

2.2. Data Description

The screening process classified 48 suitable studies out of 66 papers (as explained in Section 2.1) published between 2002 and 2021. The papers include 34 LCA studies on bioethanol from second-generation feedstock, 11 on bioethanol of first-generation feedstock. The remaining examined both types of resources.

Those studies that assessed only a single impact category or energy usage or, feedstock from non-agro residues or thermochemical conversion process are not included in this review. Bioethanol LCA studies have been conducted mainly by European countries since 2009. However, in recent years gradually the scope of the published studies has expanded to include Asia and South / North America. More than 50% of the papers assessed here compared the results with a reference case often being gasoline production, another feedstock type, or a different conversion process.

3. Results and Discussion

3.1. Technical Aspects

The biochemical conversion of biomass feedstock comprises the following main steps:

- Feedstock handling and pre-treatment;
- Saccharification to reach fermentable sugars;
- Fermentation process to convert sugars to ethanol;
- Ethanol recovery.

Figure 1 shows the scheme of production processes for 1G biomass conversion to ethanol. Although the mentioned processes are fundamental in bioconversion, each type of feedstock requires a special conversion method. All the above-mentioned processes require energy, water and chemicals. Therefore, conversion technologies are important sources of environmental impact during bioethanol production.

The pre-treatment stage in bioconversion plays a key role in influencing downstream processes [12]. Choosing a suitable pre-treatment method can change the following processes and LCA results. Shadbahr et al. [13] noted that modification of pre-treatment by increasing the concentration of sulfuric acid and decreasing residence time in the reactor improves environmental impacts. Moreover, the effect of the type of pre-treatment process on impact categories has been argued by Guo et al. [14]. Here, dilute acid and liquid hot water pre-treatment methods were examined. According to the results, the dilute acid method caused higher impacts on acidification potential, eutrophication potential, and ecotoxicity because of more chemical consumption whilst the latter increased global warming potential, abiotic depletion and ozone depletion potential due to more enzyme usage as an energy-intensive product.

In another study by Wang et al. [15], different types of pre-treatment methods, including dilute acid, steam explosion, liquid hot water and wet oxidation, are compared. The result reveals that dilute acid increased all impact categories of CML baseline 2000 method whereas steam explosion pre-treatment caused the lowest environmental burdens in all categories.

Ntihuga et al. [16] compared two scenarios of bioethanol production by continuous fermentation Blenke cascade system with and without co-product utilization. Results showed the scenario with yeast and CO₂ recycling, as well as double saccharification processes contributes to low impacts in all categories of CML 2010 method.

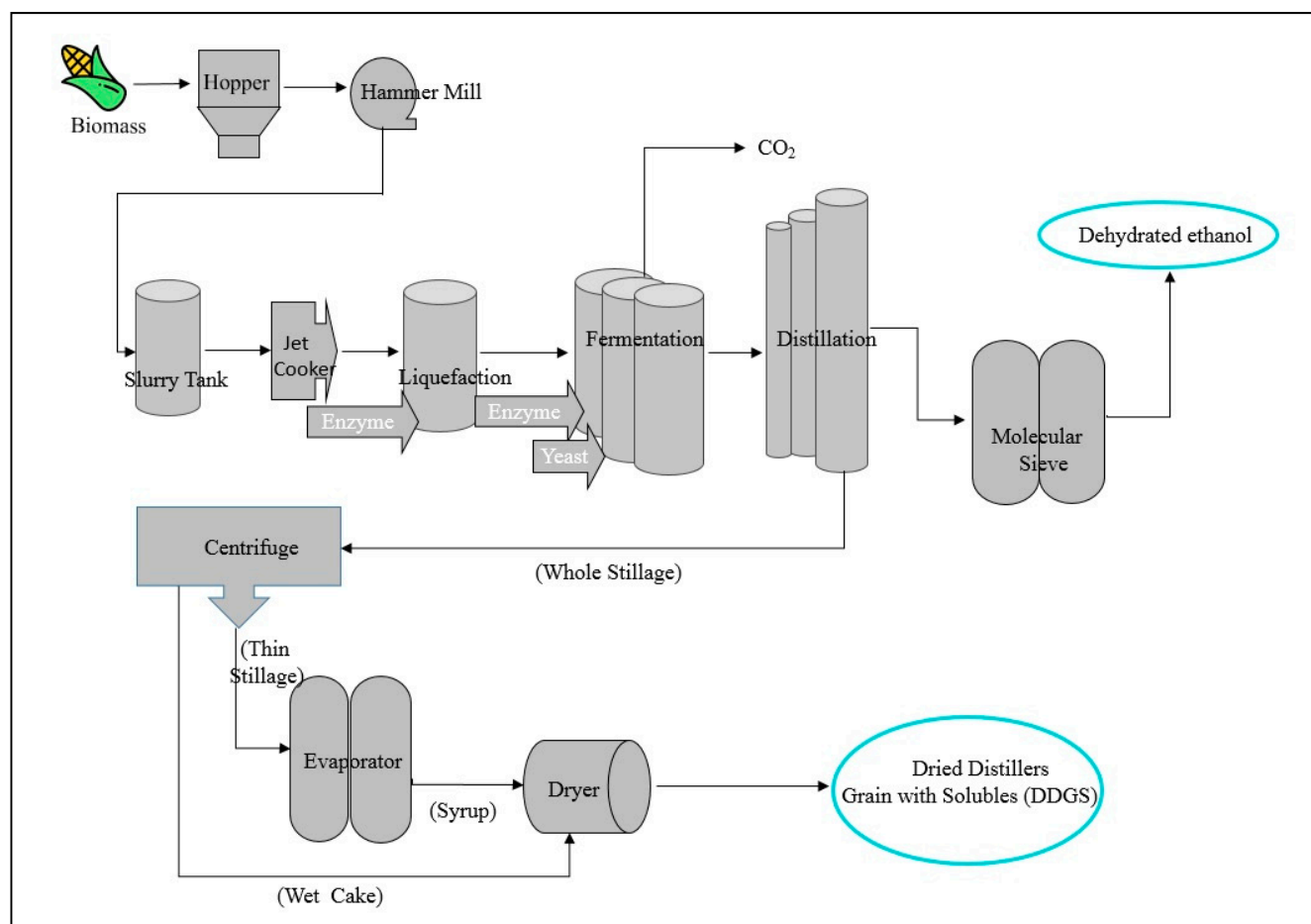


Figure 1. Schematic diagram of bioethanol production for first generation feedstock.

3.2. Feedstock Provision

The type of feedstock used for ethanol production determines the conversion processes or the agricultural measures to prepare the raw material, which is reflected in the life cycle assessment results. For instance, lignin-rich materials, such as wood, need an efficient pre-treatment process which results in higher energy consumption for the conversion of the biomass into bioethanol [13].

Most of the reviewed studies (46 papers) integrated the crop cultivation in their system boundaries. Figure 2 shows the type of biomass studied in the reviewed papers. These papers included all activities for feedstock preparation and transport. They investigated the effect of planting and fertilizing, residues utilization on LCA results, especially on the impact category land-use change. Besides others, it was found that using substrates from naturally occurring species would decrease LUC. Research conducted by Falano et al. [17] revealed that land use shifting from forest area to other species like *Miscanthus* increases GHG emissions from the soil whilst forest cultivation instead of grass leads to reverse results.

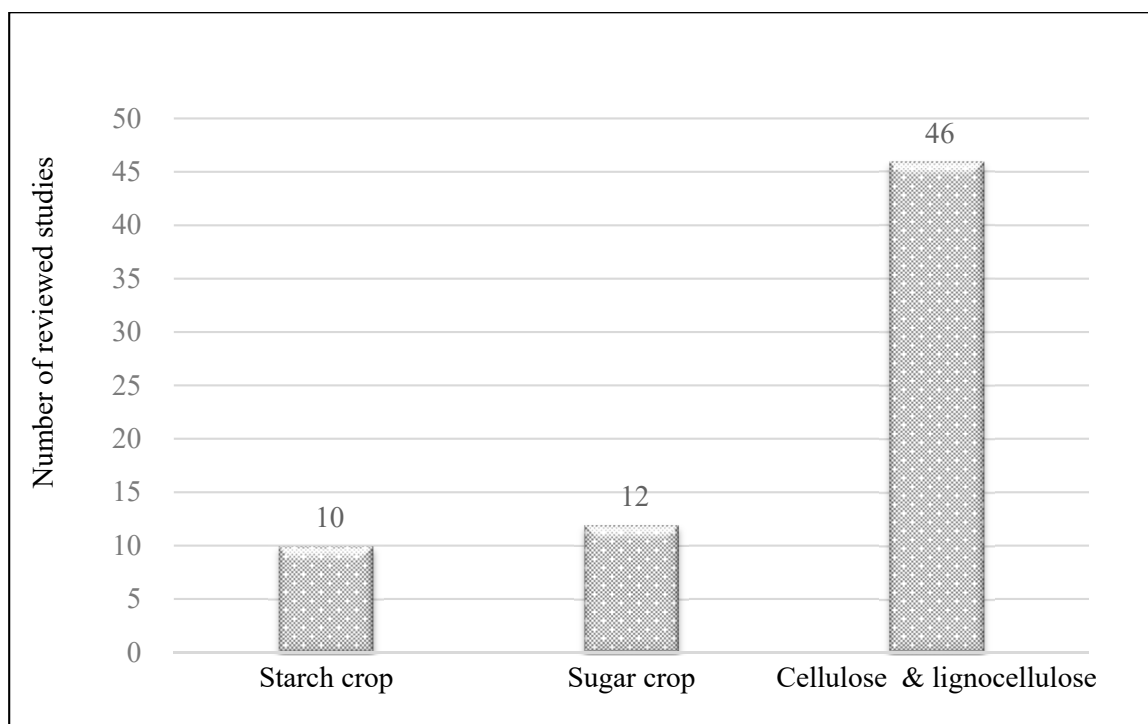


Figure 2. Type of biomass considered by reviewed papers (some of papers studied more than one feedstock).

Further, using alternative feedstock like residues, by-products or new species showed changes in environmental impacts [18]. One of the reviewed studies showed that residual materials have less contribution to land-use change in comparison to main crops due to joint production and reduced use of chemicals, water and fuel [19]. One of the papers that advanced practices like genetic improvements and developed breeding programs can influence the effectiveness of ethanol production [14]. The biomass type also determines the kind and number of agrochemicals that are used, which also influence the LCA results.

Among the LCA studies reviewed here, 21 included agrochemicals production in the system boundary. Their result showed that the use of chemicals, such as fertilizers and pesticides, increases acidification, eutrophication, land-use change and carcinogens.

3.3. LCA Methodology Components

3.3.1. System Boundary and Functional Unit

The system boundary is setting the frame for the assessed process. It also includes different variables, such as data availability, purpose of study, time, and budget. The boundary for the assessed bioethanol production systems ideally could be defined as cradle-grave covering all processes during a life span. Figure 3 shows a general overview of the processes and system boundaries in the reviewed papers.

The most widely used boundaries by the reviewed studies were cradle-gate (25 case studies), followed by cradle-grave (22 cases) and three cases of gate-to-gate type. Table 1 shows the studies that examined the LCA for bioethanol production.

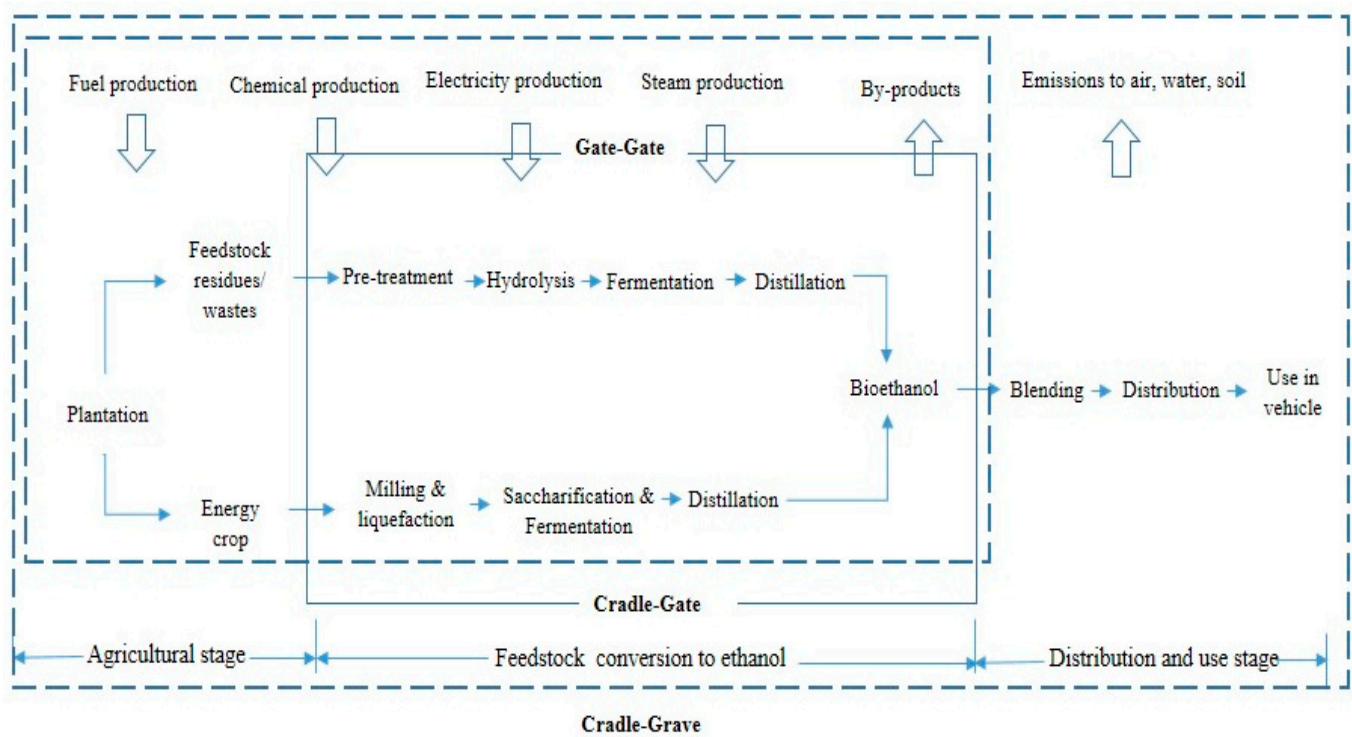


Figure 3. An overview of system boundaries and technological process of 1G and 2G bioethanol considered by reviewed papers.

Table 1. Summary of the reviewed publications. (ND = not detected).

Reference/Authors and Reference Number as Required by Energies	Biomass Source	Location	System Boundary	Functional Unit	Data Source/Type of Collected Data	Software	LCIA Method/Type	Allocation Method
[20]	Sweet potato	China	Cradle-gate	Ethanol product	Industry, GREET	ND	CML	ND
[21]	Sugarcane	Brazil	Cradle-gate	Ethanol product	Industry	SimaPro	Ecoindicator	Mass, energy
[16]	Wheat	Germany	Cradle-gate	Ethanol product	Experimental data, Lean database	GaBi	CML	ND
[22]	Corn grain	USA	Cradle-grave	Ethanol product	GREET, NREL, DEAM database	ND	TRACI, IPCC	System expansion
[23]	Sugarcane	Brazil	Cradle-grave	Caloric value	Ecoinvent, process simulation	SimaPro	TRACI, EDIP, IMPACT 2002, Ecological scarcity 2006, ReCiPe, CML, Ecoindicator	Energy
[24]	Wheat, sugar beet	Germany	Cradle-gate	Caloric value	Industry, ecoinvent	Umberto	ReCiPe, CED	Energy
[19]	Corn grain	Argentina	Cradle-gate	Caloric value	Industry, ecoinvent, NREL	SimaPro	ReCiPe, Ecoindicator	Economic
[25]	Sugarcane	Brazil	Cradle-grave	Driving distance	PestLCI software, industry, simapro databases	SimaPro	EDIP	Energy
[26]	Sugarcane	Brazil	Cradle-grave	Driving distance	Ecoinvent, EIPRO database,	CMLCA	CML	Economic
[27]	Sweet potato	China	Cradle-gate	Ethanol product	GREET, on-site survey	ND	CML	Economic
[28]	Sugar beet	Greece	Cradle-gate	Caloric value	Industry, ecoinvent	SimaPro	CML	ND
[14]	Poplar	Sweden, France, Italy, Spain, Slovakia	Cradle-grave	Driving distance	Aspen Plus	SimaPro	CML, Ecoindicator	System expansion, energy
[29]	Sweet sorghum	China	Cradle-gate	Ethanol product	GREET pilot plant	ND	CML	Energy, economic
[30]	Wheat straw	UK	Cradle-grave	Driving distance, ethanol product	Laboratory data, NREL	SimaPro	ReCiPe	Mass
[31]	Sweet sorghum	Mexico	Cradle-grave	Input biomass	Ecoinvent, Aspen Plus	SimaPro	CML	System expansion
[32]	Corn stover	Turkey	Cradle-grave	Driving distance	Ecoinvent, NREL	GaBi	IPCC, EDIP	ND
[33]	Willow	USA	Cradle-grave	Caloric value	GREET, Aspen Plus, NREL, ecoinvent	SimaPro	TRACI	Energy
[34]	Switchgrass	Austria	Gate-gate	Input biomass	Ecoinvent	SimaPro	CML, CED	ND
[13]	Woodchips	Canada	Gate-gate	Ethanol product	NREL, ecoinvent	SimaPro	Ecoindicator	ND
[35]	Corn stover, switchgrass	USA	Cradle-gate	Caloric value	GREET, ecoinvent, GaBi	ND	IPCC	ND
[36]	Corn stover, wheat straw	Austria	Cradle-gate	Input biomass	Ecoinvent, ETH-ESU 96	SimaPro	CML	ND
[37]	Poplar	Spain	Cradle-grave	Driving distance	NREL, Experimental data	ND	CML	ND

Table 1. Cont.

Reference/Authors and Reference Number as Required by Energies	Biomass Source	Location	System Boundary	Functional Unit	Data Source/Type of Collected Data	Software	LCIA Method/Type	Allocation Method
[7]	Corn stover	Netherlands	Cradle-grave	Driving distance	NREL, EIPRO database, ecoinvent	CMLCA	CML	Energy, mass, economic
[18]	Black locust, eucalyptus and poplar	Spain, Italy	Cradle-grave	Driving distance	NREL, on-site survey, ecoinvent	CMLCA	CML	ND
[8]	Alfalfa, Ethiopian mustard, flax shive, hemp hurds and poplar	Spain	Cradle-grave	Driving distance	NREL, ecoinvent	ND	CML	Mass
[15]	Wheat straw	UK	Cradle-grave	Driving distance	NREL, EMEP-EEA Guidebook, Aspen Plus, ecoinvent, IPCC guideline	SimaPro	CML	Economic, system expansion
[38]	Hemp hurds	Spain	Cradle-grave	Driving distance	NREL, ecoinvent	ND	CML	Mass, economic
[39]	Fiber sorghum	Italy	Cradle-grave	Input biomass	Experimental data, ecoinvent, IPCC guideline	SimaPro	ReCiPe	ND
[40]	<i>Arundo donax L.</i>	Italy	Cradle-grave	Driving distance	Experimental data, ecoinvent, IPCC guideline	SimaPro	ReCiPe	Energy, economic
[17]	Wheat straw, forest residue, poplar, and miscanthus	UK	Cradle-gate	Ethanol product	NREL, Aspen Plus, ecoinvent, GEMIS model	GaBi	CML	Economic, system expansion
[41]	Cassava	Thailand	Cradle-gate	Ethanol product	Industry, ecoinvent	SimaPro	CML	Energy
[42]	Switchgrass	Netherlands	Cradle-gate	Driving distance	NREL, ecoinvent	CMLCA	CML	Energy, economic
[43]	Flax shives	Spain	Cradle-grave	Ethanol product	NREL, on-site survey, ecoinvent	CMLCA	CML	Mass, economic
[44]	Alfalfa stems	Spain	Cradle-grave	Driving distance	NREL, ecoinvent	CMLCA	CML	Mass
[45]	Bagasse	South Africa	Cradle-gate	Input biomass	Industry, TEAM database	TEAM	CML	ND
[46]	<i>Brassica carinata</i>	Spain	Cradle-grave	Driving distance, ethanol product	NREL, on-site survey, ecoinvent, EIPRO database	CMLCA	CML	not used
[47]	Willow	Sweden	Cradle-gate	Input biomass	NREL, ecoinvent, on-site survey, Aspen Plus	SimaPro	CML	Economic
[48]	Eucalyptus	Spain	Cradle-grave	Driving distance	NREL, ecoinvent, on-site survey	CMLCA	CML	ND
[49]	Cassava straw, cassava root	China	Cradle-gate	Ethanol product	NREL, Aspen Plus	GaBi	CML	ND

Table 1. Cont.

Reference/Authors and Reference Number as Required by Energies	Biomass Source	Location	System Boundary	Functional Unit	Data Source/Type of Collected Data	Software	LCIA Method/Type	Allocation Method
[50]	Bagasse	India	Gate-gate	Input biomass	EPA, NREL, Aspen Plus, TEAM databases	TEAM	CML, IPCC	ND
[51]	Bagasse, Sugarcane	Brazil	Cradle-gate	Ethanol product	Industry, NREL, Aspen Plus	SimaPro	CML	Economic
[52]	Timber, recycled newsprint	USA	Cradle-gate	Ethanol product	Boustead model, NREL, Aspen Plus, USDA Forest Service database	EFRAT	Ecoindicator	ND
[53]	Hay, agricultural and forest wood waste	Canada	Cradle-grave	Driving distance	Ecoinvent	SimaPro	Ecoindicator	not used
[54]	Bagasse	South Africa	Cradle-gate	Caloric value	Industry, ecoinvent	SimaPro	IMPACT 2002	System expansion
[55]	Willow	UK	Cradle-grave	Ethanol product	NREL, GaBi databases, Aspen HYSYS	GaBi	EDIP	System expansion, energy
[56]	Maize grain, maize stover, sugarcane, sugar beet and wheat	USA, Brazil, France	Cradle-gate	Ethanol product	Ecoinvent	SimaPro	ReCiPe	Economic
[57]	Eastern redcedar, corn grain	USA	Cradle-gate	Caloric value	Ecoinvent, NREL, Aspen Plus SuperPro	SimaPro	IMPACT 2002, BEES+	System expansion
[58]	Sugarcane, bagasse	Brazil	Cradle-gate	Ethanol product	Designer, CanaSoft model, ecoinvent, NREL	GaBi	ILCD 2011, Ecoindicator	Economic

In cases of using bioethanol for FFVs blending refinery, distribution points and final bioethanol product have been included by a few studies [24,30,32,34]. However, an ideal cradle-grave system boundary for bioethanol (as fuel) should include manufacturing, maintenance and disposal of a vehicle, road construction, as well as upstream information, e.g., chemicals, but only a small portion of papers argued that.

Around 78% of the reviewed studies (mostly used second-generation feedstock) considered energy as a co-product of bioethanol production and heat/electricity production in their system boundary. Shadbahr et al. [13] directly investigated boundary changes on LCA results by shifting the system boundary from the pre-treatment stage to the whole bioethanol production plant. This study shows that after extending the system boundaries, the indicator values of the most environmental impact and damage categories improved. The credit given to on-site steam and electricity generation from residues was the main reason for improved impact indicator values.

Functional unit (FU) is another important component of LCA methodology defined as a reference flow to signify the output of processes within the LCA. The definition of the functional unit (FU) related to the purpose of the study influences the interpretation of the results. Moreover, selecting an appropriate functional unit can illustrate the environmental performance of the system boundary in different aspects. González-García et al. [43] analyzed impact categories for two different functional units comparing E10 fuel with E85. According to their findings, LCA results are changed by choosing functional units for similar system boundaries.

In general, there is no uniformity among functional units presented in the reviewed studies. Typical functional units selected in the reviewed LCA studies are related to biomass input, land area to produce the dedicated input or final output (i.e., final product or service of the process). About 85% of the reviewed studies used functional units relevant to outputs like mass/volume of ethanol product or km of transportation. Figure 4 shows the number of reviewed papers with different functional units.

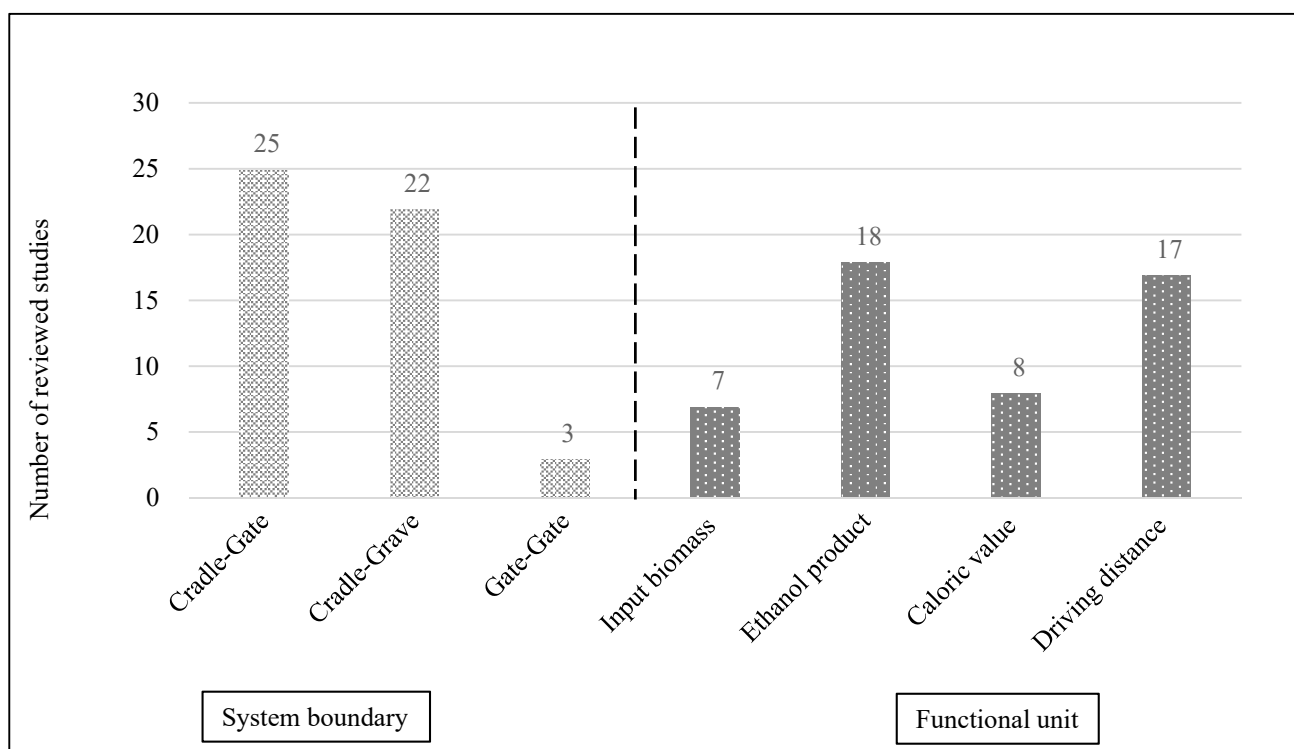


Figure 4. Number of reviewed papers in each selected functional unit and system boundary (some of studies chose more than one FU).

3.3.2. Life Cycle Inventory

Data Source

Different data sources can be used for providing the inventories of an LCA study. The reviewed papers obtained data from industry, expert interviews, estimation and LCA software databases. Every study tried to have completeness of data by adding different datasets. Further, most of the studies compensated missing real data by data sets from literature, software databases and simulations. Data gathering from reports, models and simulations are widely used in the reviewed LCA studies. Table 2 gives an overview of the employed data sources used for life cycle inventory preparation in the reviewed studies.

Table 2. Data sources used for life cycle inventory stage of the reviewed bioethanol LCAs.

Type of Collected Data	Data Source	Reference
Primary data for the agricultural activities	CanaSoft model	[58]
Emission data related to fossil fuel, combustion and auxiliary materials production	GREET model	[20,22,27,29,33,35]
Processes of bioethanol production	Aspen Plus, SuperPro Designer	[14,15,17,31,33,47,49–52,55,57,58]
Transportation, ethanol and electricity production, Agricultural data, facility and equipment design	NREL report	[7,8,13,15,17–19,22,30,32,33,37,38,42–44,46–52,55,57,58]
Emissions from capital goods production	EIPRO database	[7,26]
Emission from agricultural activities, fuel combustion and field operation	EMEP-EEA Guidebook	[15]
Emissions inventory	Boustead Model	[52]
Pesticide application emissions	PestLCI software	[25]

3.3.3. Software Use

There are different software tools available facilitating LCA compilation. As shown in Table 1 the reviewed studies used popular open-source or commercial software tools. SimaPro was the most used tool among all studies. Furthermore, some studies selected other software and tools such as TEAM or CMLCA to calculate environmental impacts. Moreover, in one paper authors connected inventory data with the Environmental Fate and Risk Assessment Tool (EFRAT) to assess eight impact categories [52]. Table 3 Shows the details of tools used by the reviewed studies.

Table 3. Life cycle assessment tools used in reviewed papers.

Software	Developer	Country
SimaPro	Pré-consultants	Netherlands
GaBi	PE International	Germany
Umberto	Ifu Institute	Germany
TEAM	Ecobilan-PricewaterhouseCoopers	France
CMLCA	Institute of Environmental Sciences (CML), Leiden University	Netherlands
EFRAT (Environmental Fate and Risk Assessment Tool)	Institute of Environmental Sciences (CML), Leiden University Department of Chemical Engineering and Department of Environmental Engineering at MichiganTech University	United States

3.3.4. By-Product Credit and Allocation

Most of the agricultural and industrial processes provide more than one output. Bio-ethanol production inevitably generates co-/by-products like animal feed, electricity, heat, etc. Hence, the breakdown of environmental loads between the main product and the

co-product is important. Choosing a suitable allocation method for sharing environmental loads among final outputs is still the most debated aspect in LCA methodology [59].

The allocation method based on the value of energy, mass and economy are very popular. Almost 56% of the reviewed studies introduced the allocation approaches in their work. The remaining followed the system expansion approach, neglect allocation procedure or did not mention in the paper. Since each study chose its allocation approach based on the purpose of the study, a comparison of the reviewed papers is hardly possible. 12 cases of reviewed papers took multiple approaches into consideration recognizing the influence of allocation methods on the outcomes of their work. Figure 5 presents an overview of allocation method selected by the reviewed papers.

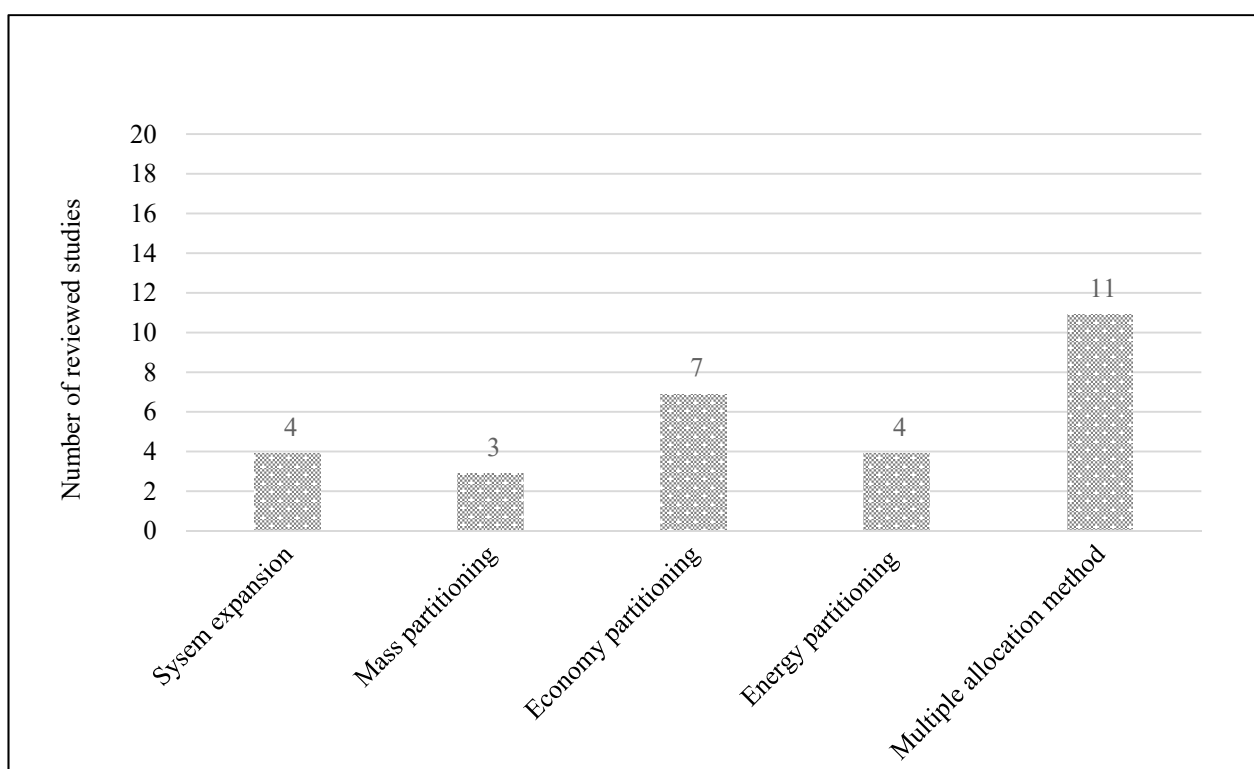


Figure 5. Different allocation methods in the reviewed papers.

3.3.5. Applied Assessment Methods and Impact Categories

Most of the LCAs of bioethanol are limited to the GWP as a single impact category. Here, only studies including a set of impact categories were considered. The review revealed that most studies preferred to omit the normalization and weighting step in the assessment stage. Only a few papers (18%) presented the results after normalization and two studies applied weighting step.

The impact assessment methods used in the papers comprised both midpoint (problem-oriented) and endpoint (damage-oriented) levels, as well as combined-methods in some cases. According to Figure 6, CML assessment method as a midpoint level method is most widely used, followed by EcoIndicator and ReCiPe.

Each applied assessment method covers a series of impact categories. Nevertheless, some of the studies just selected a few categories. Mostly acidification, eutrophication and ecotoxicity (air, water, terrestrial, marine or human) have been assessed. According to Figure 7, 100% of the reviewed studies used a climate-related impact category when investigated the life cycle impacts of bioethanol. More than 50% of the studies evaluate two further impacts related to air pollution (i.e., ozone depletion, photochemical ozone creation).

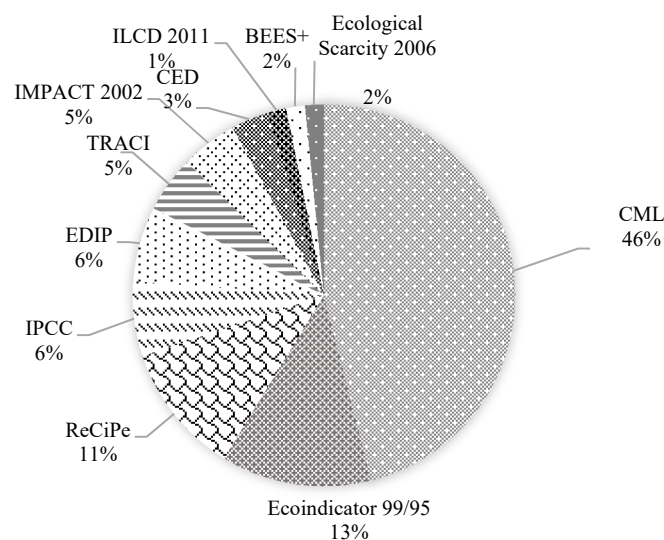


Figure 6. Share of most used assessment methods in previous LCA studies.

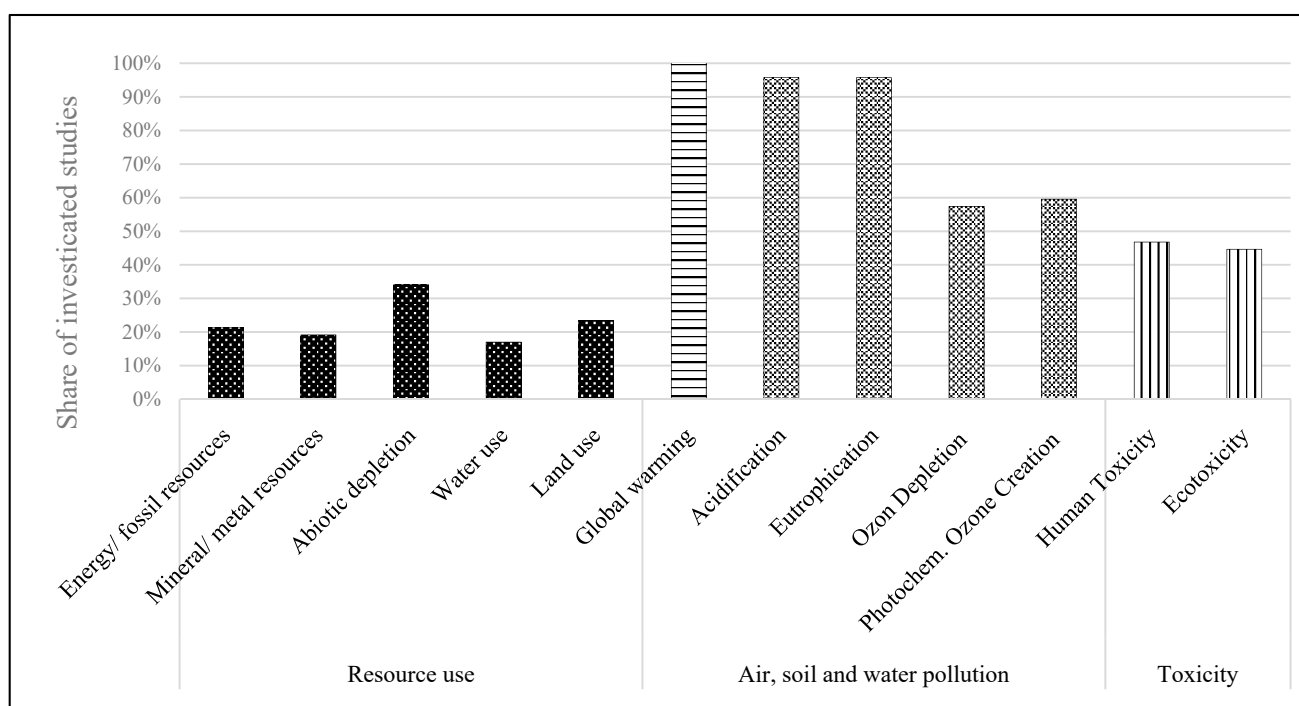


Figure 7. In each impact category, the proportion of studies that use the corresponding impact category is shown to the total number of all studies examined (in %).

Around 80% of the reviewed paper reported that bioethanol production despite the reduction in global warming potential increases acidification, eutrophication and photochemical oxidant formation impact categories. The assessment of the depletion of fossil energy resources is done by 21% of the studies through CED or the respective impact category within the ReCiPe. Additionally, the portion of studies that also examine an impact category related to abiotic resource use is low. However, 16 out of the 48 studies, considered impacts from abiotic resource use. Of these, 34% deploy the impact category Abiotic Depletion Potential from the Dutch CML method. All studies analyzing the abiotic degradation potential showed that bioethanol production from biomass offers advantages in this category compared to conventional fossil-based gasoline. 19% of the analyzed

studies also assessed the categories of mineral resource and metal depletion of ReCiPe and IMPACT 2002 methods.

Despite covering various categories by different assessment methods, some categories still receive poor attention. For instance, only a few studies consider the significant impact (directly/indirectly) of bioethanol production on land-use change and water consumption.

Approximately 17% of the studies investigate impact on water consumption, although this topic seems to be quite relevant for bio-based energy. This is probably mainly due to a lack of a methodological consensus for water use assessment in the context of LCA at the time most of the reviewed studies were published. There have been several attempts for water use assessment, but the first kind of approved method was just published in 2017 [60]. The studies reviewed here used ReCiPe for assessing the impact category water use.

Another very relevant impact category for the assessment of biofuels is land use. 23% of the reviewed studies assessed the impacts of land use. They mostly used impact categories are based on the Ecoindicator 99 and CML 2 Baseline 2000 methods. High impacts from land use were found when forests or grasslands are converted to biomass crops with low biofuel yield. Using waste materials or woody crops (e.g., from short rotation plantations) is more favorable in terms of land use impacts [17,57]. When comparing bio-based and fossil-based ethanol in terms of land use it would be advisable to prefer fossil-based fuels because there are always impacts from land use when switching to bio-based fuels [56].

Moreover, to pay more attention to land use impact category, Muñoz. et al. extended their research by adding novel midpoint impact categories on Biodiversity and Ecosystem Services [56]. The BES indicator comprises seven impact categories including biodiversity damage potential [61], climate regulation potential [62], biotic production potential [63], freshwater regulation potential, erosion regulation potential, water purification potential through physicochemical filtration and water purification potential through mechanical filtration [64] to consider relevant impacts to land occupation.

3.3.6. Uncertainties

Uncertainty analysis is included in the interpretation part of the LCA methodology to address the range of variation in the results. Most of the published papers have not included an uncertainty analysis. Among the reviewed LCA studies, 17% of them (8 cases) examined the uncertainties on the results. They analyzed uncertainty by a Monte Carlo function that was included in the LCA software. In these cases, all input/output data from the upstream processes to usages phase in transportation were included [39,57]. The reviewed papers indicated that uncertainties occur due to a lack of data or data inaccuracy in industrial processes, methodological elements selection (e.g., system boundary, allocation, impact categories and characterization models), as well as feedstock preparation.

According to the review, 13 studies used simulation software to prepare data inventories. In this regard, there is a risk of estimating the impacts of processes in the conceptual design, pilot and laboratory rather than commercial scale because it is not easy to examine the efficiency of the technologies in the early stages [65]. Not only the maturity of technology but also the construction of equipment and facilities could be a source of data scarcity. Amongst reviewed studies there was only one study, which included material used for plant construction [35]. The influence of technology in the result uncertainty was taken into account by Borrion et al. [30]. They found that the level of technology development affects the reliability of data and the results.

The methodological choices are another potential cause for uncertainties in LCA results. However, the recent LCA studies attempt to cover more impact categories but in most of them impact categories such as human toxicity, biodiversity, land and water use are still under poor attention. The data gap due to employing literature and similar projects to collect data is unavoidable, especially due to geographical or temporal differences in particular when normalization is considered in the study.

In our review, six of eight studies that included uncertainty analysis in their work, examined agricultural parameters to estimate the uncertainties. These studies showed that the gap of the data in the agricultural stage (e.g., land-use change, soil emissions, carbon cycle) contributes to uncertainties [34,36,56].

4. Conclusions

In recent years, a wide range of LCA studies has been conducted to measure the sustainability of bioethanol production. As was mentioned, the differences in methodological assumptions, technical and agricultural issues applied by authors hampered the comparison of LCA outcomes. In other words, it is not possible to compare the influence of each component of the methodology on the results with a high certainty, because various technical and agricultural-related parameters are also involved in the results of studies. However, in this paper selected studies were reviewed to identify bottlenecks in LCA analysis in terms of the three aspects mentioned above.

- Reviewed papers show that studies results depend greatly on LCA methodology choices. Selection of allocation method, the definition of assessment method, system boundary and functional unit besides choosing effective impact categories change the results.

However, there is an increasing trend of considering a set of impact categories in LCIA stage rather than only GHG impact. On the other hand, the share of studies that examine impact categories relevant to land and water use is not significant. Therefore, future studies should focus more on ecosystem quality to clarify the potential impacts of bioethanol production on land-use changes including water resources.

A handful of reviewed papers considered uncertainty analysis. Some of them studied conceptual models which may not precisely represent the industrial system that would be built in the future; hence, the differences between data obtained from the simulation and actual industrial data makes the uncertainties. Besides, many other variables caused by methodological assumptions and agricultural issues are involved in a wide range of uncertainties. Attention needs to be placed on uncertainty analysis in LCA studies. Furthermore, the studies can reduce the potential uncertainties by reconsidering the key elements of LCA methodology such as FU, system boundary, as well as data collection process.

- Although most authors added the feedstock production and processing into the system boundary, less than half of them considered agrochemicals' production. Given the important role of chemicals, such as fertilizers, pesticides in increasing acidification, eutrophication, land-use change and carcinogens, it seems necessary to include the production of those chemicals in LCA studies. Moreover, carrying out LCA analysis on the bioethanol from new generation of feedstock with less chemicals requirements or less agricultural practices would be useful to estimate and compare the relevant impacts with conventional feedstock.
- From the technical point of view, pre-treatment was the most considered process in the studies with a high potential for changing LCA results. As reviewed papers showed that the modified pre-treatment methods lead to environmental savings, because pre-treatment plays an important role in the next processes. However, advanced pre-treatment technologies with optimization of inputs and outputs in the bioethanol production can decrease the environmental impacts; the other processes of production also need to be considered. Furthermore, in most circumstances, studies neglected the production, transportation and use of the chemicals, nutrients, enzymes, which are the inputs for bioethanol production processes. This aspect needs to be investigated more by future studies.

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Acronyms

LCA	Life Cycle Assessment
FU	Functional Unit
FFVs	Flex Fuel Vehicles
1G	First generation
2G	Second generation
CED	Cumulative Energy Demand
CMLCA	Chain Management by Life Cycle Assessment
LUC	Land Use Change
GHG	Greenhouse Gases
E10	Mixture of 10% ethanol and 90% gasoline by volume
E85	Mixture of 85% ethanol and 15% gasoline by volume
ISO	International Standards Organization
BES	Biodiversity and Ecosystem Services
GWP	Global Warming Potential
IPCC	Intergovernmental Panel on Climate Change
EDIP	Environmental Design of Industrial Products
CML	Institute of Environmental Sciences of the Faculty of Science of Leiden University
BEES+	Building for Environmental and Economic Sustainability
ILCD	The International Reference Life Cycle Data System
TRACI	Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts
LCIA	Life Cycle Impact Assessment

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