REVIEW

A Review of Life Cycle Assessment (LCA) of Bioethanol from Lignocellulosic Biomass

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Abstract

Liquid biofuels are widely recognized alternatives to fossil fuel not only to combat the global warming potential, but also to reduce the reliance on fossil fuels to facilitate economic development. The production and use of lignocellulosic liquid biofuel have been emphasized because it is highly reproducible and does not compete with food. This study summarizes the LCA studies on lignocellulosic ethanol produced from various biomass resources focusing on energy balance, greenhouse gas (GHG) emission and other impact categories, and the production cost to discuss their potential environmental and socioeconomic impacts. Numerous efforts have been made to evaluate the life cycle of lignocellulosic ethanol with LCA methodologies and deals with feedstock, energy paths, conversion technologies, allocation methods, utilization of by-products etc. to determine the environmental impacts as well as the production cost. The environmental benefits are reported in most of the studies except for few examples. A wide variation was observed in the reported production cost of ethanol, which is dependent on the feedstock, conversion technologies, allocation methods and plant sizes. Onsite enzymes production/purchase appeared to be the main hotspot, demands a vigorous study to improve their productivity and reduce costs. Another promising alternative for compensating production costs seem to be the generation of valuable coproducts and integration of ethanol production processes (ethanol and energy). Reviewed literature indicates that despite the environmental benefits of ethanol produced from lignocellulosic biomass, its economic viability remains doubtful at present, even if highly optimistic assumptions are made for the cost calculation, especially in the case of enzyme. Hence, the biotechnological revolution is must for the sustainability of bioethanol, especially in the field of enzymes and microorganisms. Moreover, the adaptation of innovative technologies and renewable energy policy may help limit costs, but careful consideration of land use changes and soil quality is required to avoid any loss of productivity.

Discipline: Biofuel

Additional key words: GHG emissions, rice straw, enzymatic saccharification

Introduction

GHG emissions, which have dramatically proliferated due to tremendous energy use, have resulted in global warming, perhaps the most serious problem that humankind faces today. The growing concerns about climate change, rising costs of fossil fuel and the geopolitical uncertainty associated with uninterrupted energy supply have motivated individuals, organizations and na-

tions to seek clean and renewable substitutes. Liquid biofuels (bioethanol and biodiesel) are widely recognized alternatives to fossil fuel. It is known that renewable energy not only reduces the reliance on foreign oil and improves energy security, but also provides significant environmental benefits and enlarges rural economies^{69,115}. The first generation biofuels are produced from food or feed grains, thus compete with food or feed and contribute to higher food prices. Accordingly, the production of second generation biofuels from lignocellulosic biomass

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has been emphasized, because it does not compete with food or feed^{22,47,52,99,141}.

The life cycle GHG emissions of various forms of bioenergy and their ability to reduce GHG emissions vary widely, and are dependent on land use changes, choice of feedstock, agricultural practices, refining and conversion processes with differing socioeconomic and environmental impacts. It is thus essential to evaluate the environmental impact and the economic feasibility of lignocellulosic-based bioethanols. Environmental awareness influences the way in which legislative bodies such as governments will guide the future development of the lignocellulosic-based ethanol industry. Life cycle assessment (LCA) is a tool for evaluating the environmental effects of a product, process, or activity throughout its life cycle or lifetime, known as a 'from cradle to grave' analysis. Although several researchers have compiled LCA studies of lignocellulosic ethanol to discuss some of the key issues: energy pathways, system boundaries, functional units, allocation methods, utilization of coproducts etc.^{14,23,24,42,63,77,81,88,111,132}, some recent advances in LCAs of lignocellulosic bioethanol remain to be reported. Therefore, this study aims to compile recent LCA studies of lignocellulosic bioethanol, and discuss the energetic, environment and socioeconomic aspects of the bioethanol industry.

LCA Methodology

Although the concept of LCA evolved in the 1960s and there have been several efforts to develop its methodology since the 1970s, it has attracted considerable attention from those engaged in environmental science fields since the 1990s, which has seen the LCA concept promoted, sponsored and developed by various national and international organizations (SETAC: Society of Environmental Toxicology and Chemistry, USEPA: United States Environmental Protection Agency, ISO: International

Organization for Standardization, ILCAJ: Institute of Life Cycle Assessment, Japan etc.), and LCA practitioners. Consequently, consensus has been achieved on an overall LCA framework and a well-defined inventory methodology⁵⁹. The method has rapidly developed into an important tool for authorities, industries, and individuals in environmental sciences. The UNEP (United Nations Environment Programme)-SETAC initiative includes methods to evaluate the environmental impacts associated with water consumption and land use⁶⁵. A common methodological framework ("Version Zero") has also been developed by the Global Bioenergy Partnership (GBEP) Task Force on GHG Methodologies that could be applied to the LCA of bioenergy production and compared to the full lifecycle of its fossil fuel equivalent to improve the transparency and acceptance of the results⁴¹. The LCA methodology consists of four components: Goal definition and scoping, Inventory analysis, Impact assessment and Interpretation. Figure 1 shows the stages of an LCA⁶⁰. The purposes of an LCA can be: (1) comparison of alternative products, processes or services; (2) comparison of alternative life cycles for a certain product or service; (3) identification of parts of the life cycle where the greatest improvements can be made.

1. Goal definition and scoping

Goal definition and scoping defines the purpose of the study, the expected product of the study, system boundaries, functional unit (FU) and assumptions. The system boundary of a system is often illustrated by a general input and output flow diagram. All operations that contribute to the product life cycle, process, or activity fall within the system boundaries. The purpose of the FU is to provide a reference unit to which the inventory data are normalized and its definition depends on the environmental impact category and aims of the investigation. The functional unit is often based on the mass (kg) or volume (L) of the product under study, however the distance (km), land area (ha), energy (MJ) and economic values of products are also used.

2. Life cycle inventory (LCI) analysis

This phase includes all inputs and outputs from the processes. The inputs are energy (renewable and non-renewable), water, raw materials etc., while the outputs are products and co-products, and emissions (CO₂, CH₄, SO₂, NO_x, CO, etc.), water and soil (total suspended solids: TSS, biological oxygen demand: BOD, chemical oxygen demand: COD, adsorbable organically bound halogens: AOXs, etc.) and solid waste generation. Nowadays, many LCA databases exist and can normally be bought together with LCA software. Data from databases can also be

Fig. 1. Stages of an LCA⁶⁰

used for processes that are not product specific, such as general data on the production of electricity, coal or packaging, although site-specific data are required for product specific data.

3. Impact assessment

The life cycle impact assessment (LCIA) aims to understand and evaluate environmental impacts based on inventory analysis, within the framework of the goal and scope of the study. In this phase, the inventory results are assigned to different impact categories, based on the expected types of environmental impacts. Impact categories include global effects (global warming, ozone depletion etc.); regional effects (acidification, eutrophication, photo-oxidant formation etc.); local effects (nuisance, working conditions, effects of hazardous waste, effects of solid waste etc.); biodiversity, water and land use efficiency and impacts on human health (ISO 1025, 2006E⁶⁰).

4. Interpretation

The purpose of an LCA is to draw conclusions that can support a decision or provide a readily understandable LCA result. The inventory and impact assessment results are discussed collectively in the case of an LCIA, or the inventory only in the case of LCI analysis, and significant environmental issues identified for conclusions and recommendations consistent with the goal and scope of the study. This is a systematic technique to identify and quantify, check and evaluate information from LCI and LCIA results, and communicate them effectively. This assessment may include both quantitative and qualitative measures of improvement, such as changes in product, process, and activity design; raw material use, industrial processing, consumer use, and waste management. Cost and profit are the key indicators in decision-making on an investment, while costs are what producers or consumers understand best and an integral part of the decision-making process when identifying improvements of a product, process or activity, hence LCA results are also interpreted in the form of life cycle costing.

LCA studies on lignocellulosic bioethanol

The life cycle of lignocellulosic ethanol extensively evaluated by using LCA methodologies determines the economic and environmental impacts of different production processes. Figure 2 shows a schematic diagram of ethanol production process using the enzymatic hydrolysis method.

1. LCA of bioethanol produced from agri-residues

Agri-residues, known as the most abundant feed-

Fig. 2. Schematic diagram of the ethanol production process from lignocellulosic biomass using the enzymatic hydrolysis method

SSF: simultaneous saccharification and fermentation.

stocks for lignocellulosic ethanol, have gained increasing attention as a renewable energy source. LCA methodology has been extensively used to evaluate the life cycle of lignocellulosic ethanol. Several studies noted that lignocellulosic ethanol can improve energy security and contributes significantly to a reduction of GHG emissions^{37,44,81,115,130,132,137}. In contrast, the bioenergy system can release more GHG emissions than its fossil alternative when the energy used to feed the biomass conversion process comes from carbon-intensive fossil sources³⁸. The reduction in GHG emission is reported to be dependent on feedstocks, conversion technology, utilization of coproducts and allocation methods^{67,77,78,115,123}.

Biofuel production is reported to be beneficial in terms of the reduction of non-renewable energy consumption and the global warming impact if biomass from cropping systems is utilized. However, unless additional measures such as planting cover crops are taken, the utilization of biomass for biofuels would also tend to increase acidification and eutrophication, primarily because significant nitrogen and phosphorus related environmental burdens are released from the soil during cultivation⁷⁰. Ethanol produced from the lignocellulosic residues of banana fruit is also reported to be energetically feasible¹²⁹. Stolman¹¹⁹ noted that ethanol produced from grass clippings, corn stalks and other plants using future techniques is beneficial. The investigated bioenergy production processes from sugarcane bagasse revealed that the cogeneration option results in lower energy-related emissions (i.e. lower global warming, acidification and eutrophication potentials), whereas the fuel ethanol option is preferred in terms of resource conservation (since it is assumed to replace oil not coal), and also scores better in terms of human and eco-toxicity if assumed to replace lead-bearing oxygenates¹⁶.

The lignocellulosic (stover) ethanol pathway avoids

86–113% of GHG emissions if E85 is used in fuel flexible vehicles instead of gasoline^{30,108}. GHG emissions are reported to exceed 100%, which may be due to the carbon neutrality of biomass and the use of residues. Total fossil energy is also 102% lower, but emissions of CO, NO_x, and SO_x increase whereas hydrocarbon ozone precursors are reduced¹⁰⁸. It is worth noting that biomass combustions are assumed to be carbon neutral in all these studies. Conversely, emissions from stover ethanol are reported to be 65% lower for the near-term scenario (2010) due to the sharing of emissions with corn grains¹¹⁵. Emissions would be about 25–35% lower than the near-term scenario if the mid-term scenario (2020) is considered. The use of corn stover as a feedstock results in lower GHG emissions relative to conventional corn-grain ethanol^{70,132}, although this reduction is dependent on the allocation method used^{67,70,132}. The carbon intensity of stover-derived ethanol is reportedly 10–44% that of gasoline⁶⁷. The corn stover collection emits GHGs after corn harvesting unless equipment capable of performing a single-pass harvest becomes commercially available¹⁰⁸.

Abiotic resources and ozone layer depletion decrease when gasoline is replaced by stover ethanol fuels (E10 and E85), which is not relevant to the allocation method, however the remaining impacts are larger except the global warming potential (GWP). The GWP reduces when mass/energy allocation is applied, but increases in the case of economic allocation⁷⁷. System boundaries also cause considerable variation in LCA estimates since they not only vary according to start and end points (e.g. well to tank and well to wheel) but also over space and time in a way that can dramatically affect energy and GHG balances¹⁶. The GWP of the lignocellulosic ethanol plant is noted as significantly (twofold) worse than that of the gasoline refinery, but its improved eco-efficiencies make it superior in terms of abiotic and ozone layer depletion potentials⁷⁹. In contrast, GHG savings from ethanol and Ethyl Tertiary Butyl Ether (ETBE) blending are reported to be positive, even taking into consideration the modification of the refinery sector included²⁹.

Uihlein and Schebek¹²⁶ concluded that bio-based products and fuels from straw may also be associated with environmental disadvantages due to, e.g. land use or water eutrophication. The environmental impacts predominantly result from the provision of hydrochloric acid and, to a smaller extent, from the provision of process heat. The optional acid and heat recoveries yield environmental impacts that are approximately 41% lower than those of the fossil counterparts. LCA of lignocellulosic (straw, waste wood and other agricultural waste) ethanol produced by an enzymatic hydrolysis process shows that E10 improves the environmental performance in GHG

emissions if the energy required to generate the process steam is derived from biomass rather than fossil fuel for pretreatment of feedstock, but has inferior performances in terms of acidification, eutrophication, winter smog, summer smog, carcinogenic substances, heavy metals, ozone layer depletion and solid waste³⁸. The net energy ratios (output energy divided by input energy from fossil fuels) of ethanol production systems from high yield rice plants are also reported to be positive, where rice and its residues are used^{73,98}. Koga and Tajima⁷³ also noted that whole rice plant-based ethanol production systems improve energy efficiency and reduce GHG emission, because straw removals notably mitigate CH₄ emissions from the paddy field. The use of straw for energy (CHP: combined heat and power) in bioethanol production from wheat grains has significant benefits in terms of reduced global warming and the use of non-renewable energy, but the eutrophication and atmospheric acidification impact categories were slightly unfavorable in some cases³⁹. Cherubini and Ulgiati²⁵ noted that the use of agricultural residues in a biorefinery saves GHG (50%) and reduces demand for fossil fuels (80%), where the best management practices are employed. However, biomass harvest rates must be carefully established.

Although agri-residues are identified as abundant lignocellulosic biomass resources, there is debate regarding the actual amounts of residues which could be removed from arable soils with no loss of quality, as well as the potential tradeoffs in the overall energy chain compared to the use of fossil energy sources. Gabrielle and Gagnaire³⁹ noted that straw (wheat straw) removal had little influence on environmental emissions in the field, and incorporating it in soil resulted in sequestration of only 5–10% of its C in the long term. It is also noted that a certain portion of crop residues can be removed to produce bioethanol without degrading the soil quality, depending on the season, location, tillage and soil types^{87,94}. Selecting residues that contain relatively high levels of available cellulose and hemicellulose for removal or returning suitable crop residues that are rich in refractory compounds may increase the scope for removal of crop residues for ethanol production⁹⁴. Sheehan et al.¹⁰⁹ state that up to 60% of the stover can be collected and converted to fuel ethanol, however Blanco-Canqui and Lal¹³ suggested a stover removal rate as low as 25%, beyond which soil fertility and structural stability would be negatively affected. Graham et al.⁴⁶ noted that in current agricultural practice, only 28% of the stover is harvested, and the rest is left in the field for soil fertility. Although lignocellulosic biomass is reported to be the most promising feedstock considering its great availability and low cost, the large-scale commercial production of fuel bioethanol

from lignocellulosic materials has yet to be implemented^{4,62}, due to challenges and obstacles such as cost, technology and environmental issues needing to be overcome for the commercial production of lignocellulosic ethanol^{55,122}.

2. LCA of bioethanol from energy crops, woody biomass and forest residues

Presently, the contribution of energy crops as a proportion of total biomass energy is relatively small, but it is set to grow in the near future. The majority of LCA studies noted that bioenergy from energy crops reduces GWP and fossil energy consumption when the most common transportation biofuels are used to replace conventional diesel and gasoline^{14,26,50,102,110} in all but a few studies^{92,104}. In contrast, other environmental aspects such as acidification and eutrophication increase²⁶, and including land use change effects in GHG balances, biofuels substituting fossil fuels may lead to increased negative impacts¹⁰⁴. Pimentel and Patzek⁹² noted that ethanol production from switchgrass and woody biomass requires 50 and 57% more fossil energy than the ethanol fuel produced, respectively. In contrast, Cherubini et al.²³ believed these limitations could be partially overcome by developing second generation biofuels, produced from various lignocellulosic non-food crops and residues.

Estimated GHG emissions from cellulosic ethanol were 94% lower than those of gasoline, while genetic and agronomical improvement may further enhance the energy sustainability and biofuel yield of switchgrass¹⁰². Switchgrass fields are reported to be near-GHG neutral depending on the agricultural inputs (mainly N fertilization) and subsequent biomass yields. The use of ligneous biomass residue for energy at a cellulosic biorefinery is the main key to reducing GHG emissions rather than biofuels from annual crops, where processing energy is derived from fossil fuels³⁵. Spatari et al.¹¹⁵ noted that emissions from energy crop (switchgrass) ethanol were 57% in the case of the near-term scenario (2010) and lower for an E85-fueled automobile compared to gasoline, on a CO₂ equivalent per kilometer basis. Emissions could be 25-35% lower than those of the near-term scenario if the mid-term scenario (2020) were considered. Net energy gains per hectare of biofuels are affected by the crop yield, conversion rate, and energy inputs required to produce, deliver and process feedstock. The yearly net energy gain is reported to be greater in the case of field scale trials on marginal land than low-input switchgrass grown in small plots¹⁰².

González-García et al.⁴⁵ assessed the environmental performance of bioethanol produced from poplar biomass considering three ethanol applications (E10, E85 and E100), addressed the impact potentials per kilometer

driven by a mid-size passenger car, and compared it with gasoline. The authors noted that fuel ethanol derived from poplar biomass may help ease the exacerbation of global warming, and depletion of abiotic resources and the ozone layer by up to 62, 72 and 36%, respectively. Conversely, acidification and eutrophication would intensify. Tilman et al.¹²³ noted that biofuels derived from low-input high-diversity (LIHD) mixtures of native grassland perennials can provide more usable energy, greater GHG emission reductions, and less agri-chemical pollution than that of corn grain ethanol. LIHD biofuels are carbon negative because net ecosystem carbon dioxide sequestration (4.4 t/ha/year of carbon dioxide in soil and roots) is reported to be greater than the release during biofuel production (0.32 t/ha/year). LIHD biofuels can also be produced on agriculturally degraded lands and thus neither displace food production nor cause any loss of biodiversity via habitat destruction.

Switchgrass is reported to be effective at storing soil organic carbon (SOC), not just near the soil surface, but also at depths below 30 cm where carbon is less susceptible to mineralization and loss^{76,102,136}. Haney et al.⁵⁴ noted that perennial grass systems had higher SOC and water extractable organic C (WEOC) than the annual corn system. Among perennial grass systems, switchgrass had the lowest SOC and WEOC. Nitrogen leaching is reported to be less for switchgrass than corn, but greater than in alfalfa-corn cropping systems¹²⁸. Monti et al.⁸⁶ analyzed the energy crops (switchgrass, giant reed and cynara) production in terms of energy and hectares, and compared them with conventional wheat and maize rotation. This study concluded that on average, 50% lower environmental impacts can be achieved by substituting conventional rotation with perennial crops. The benefits are reportedly dependent on biomass yield and the preference to a specific energy crop strongly depends on weighting sets that may change considerably in terms of space and time.

It is also concluded that the economic and environmental aspects of high yield crop production systems are not necessarily conflicting, whereas under or over supply of nitrogen fertilizers leads to a decline in resource use efficiency^{18,19,51}. Pedersen et al.⁹⁰ reveal that in the USA, some long-term breeding of switchgrass has achieved large yields and may begin to contribute significantly to biofuel production. Genetically modified (GM) herbicide tolerant energy crops (sugar beet) are reported to be less harmful to the environment and human health than growing conventional crops, largely due to lower emissions from herbicide manufacture, transport and field operations¹⁰. These studies indicate that the social and environmental co-benefits, including carbon sequestration

opportunities, will be drivers of future energy cropping uptake, although they must also be ecologically sustainable, environmentally acceptable and economically competitive with fossil fuels¹¹⁰.

3. Land, water and other approaches in LCA of bioethanol

The global population continues to grow geometrically, exerting great pressure on arable land, water, energy and biological resources to provide an adequate food supply while maintaining the ecosystem. The availability of land on which to grow biofuel crops without affecting food production or GHG emissions from land conversion is limited, hence land use efficiency should be maximized to achieve climate change goals. Although lignocellulosic ethanol supply chains are considered feasible for making GHG savings relative to gasoline, an important caveat is that if lignocellulosic ethanol production uses feedstocks that cause indirect land-use change, or other resulting significant impacts, any benefit may be greatly offset^{113,114}. The effects of land use changes were noted as having a significant influence on the final GHG balance (about 50%)²⁵.

Several studies also noted that converting croplands or grasslands to produce energy crops may actually lead to an increase rather than fall in GHG emissions^{34,104}. Brandão et al.¹⁷ studied the different land use systems used for energy crops and noted that *Miscanthus* is the optimal choice in terms of GHG emissions and soil quality compared to oilseed rape, short-rotation coppice willow and forest residues, but performed worse in the categories of acidification and eutrophication, while oilseed rape showed the worst performance across all categories. Stephenson et al.¹¹⁷ reported that if willows are grown on idle arable land in the UK, or in Eastern Europe, and imported as wood chips into the UK to produce ethanol, this saves about 70–90% of GHG emissions compared to fossil-derived gasoline on an energy basis. In contrast, Searchinger et al.¹⁰⁴ estimated GHG emissions from land-use changes by using a global agricultural model and noted that corn-based ethanol, instead of achieving 20% savings, nearly doubles GHG emissions over 30 years and increases GHGs for 167 years. Biofuels from switchgrass also increase emissions by 50%, if grown on U.S. corn lands. It is also reported that the bioelectricity pathway outperforms the cellulosic ethanol across a range of feedstocks, conversion technologies, and vehicle classes; producing 81% more transportation kilometers and 108% more emission offsets per unit area of cropland than cellulosic ethanol¹²¹.

Based on the ecological footprint perspective, Stoeglehner et al.¹¹⁸ noted that biofuels will only be able

to contribute to a certain – maybe relatively limited – extent, to an overall sustainable energy supply that will vary widely between regions, and the sustainability of biofuel production depends on the amount of land available. It is reported that direct land use changes, the choice of calculation methods, utilization of coproducts and the technical design of production systems all impact on GHG balances and eutrophication for all biofuels¹⁵. The enhanced demand for biofuel crops under the EU Biofuel Directive has a strong impact on agriculture at a global and European level, while the incentive to increase production in the EU tends to increase land prices and farm income there and in other regions⁶.

The sustainability of biofuels depends on the selection of land on which feedstocks are grown. Several competing factors need to be balanced, such as changes in land use (clearing tropical forests or using peatlands for crop cultivation) to negate any of the intended future climate benefits, and impacts on biodiversity. In addition, developments in the agricultural sector for food and non-food crops will have important implications for water usage and its availability. The opportunity costs and rebound effects of land use changes must be addressed when considering any decision to assign land to biofuel feedstocks⁹¹. Although biomass residues have been identified as a potential feedstock for bioenergy, the global mature forest area will decrease by 24% between 1990 and 2100, due to both population growth and wood biomass demand in developing regions, and may even disappear by 2100 in some developing regions, such as Centrally Planned Asia, Middle East and North Africa, and South Asia¹³⁹.

The gross water consumption in the lignocellulosic ethanol production processes are reported to be 28–54 liters per liter of ethanol⁹⁵. The high water consumption results from the process water used in the $\text{Ca}(\text{OH})_2$ pretreatment, washing of solids prior to enzymatic hydrolysis. However, water consumption is reported to be only 0.3 L per liter of ethanol produced from agri-residues (corn stover or wheat straw), because the water requirement for crop production was attributed only to grains¹¹². Ethanol produced using a biochemical or thermochemical conversion process is expected to reduce GHG and air pollutant emissions, but involve similar or potentially greater water demands and solid waste streams than conventional ethanol biorefineries. Despite current expectations, significant uncertainty remains regarding how well next-generation biofuels will fare in terms of different environmental and sustainability factors when produced on a commercial scale in the U.S.¹³². Although ethanol production consumes huge amounts of water, its impact on water resources is seldom included. The land to man ra-

tio in developing countries is not as favorable as in developed countries, with far scarcer land resources creating serious problems in land resources management and possibly resulting in land degradation in such developing countries. The use of bioenergy also involves environmental challenges, for instance increased mono-cropping practices and greater fertilizer and pesticide use, which may jeopardize water and soil quality. Perhaps the main concern over land use change is the risk of large areas of natural forests and grasslands being converted to energy crop production, which would not only threaten biodiversity and ecosystems, but also result in a possible increase in GHG emissions.

Cost analysis

Costs are what producers, or consumers understand best and an integral part of the decision-making process when identifying potential improvements of a product, process, or activity, hence the use of life cycle costing (cost analysis of the entire life cycle) as a decision support tool in the bioethanol industry. The production cost of ethanol is dependent on both technical and economic parameters, such as the cost of feedstocks, choice of feedstocks, energy consumption, conversion technology and efficiency, and the value of coproducts^{1,2,4,5,31,82,135,137}. The production cost of lignocellulosic bioethanol is reported to be considerably higher than the market price of gasoline^{5,58,79,89,96,135}. Vadas et al.¹²⁸ noted that net energy production per hectare is greater for switchgrass than that of alfalfa-corn cropping systems, but may not return the potential income to farmers that alfalfa-corn could. The costs of cellulase and capital are the major expenses when producing lignocellulosic bioethanol⁹⁵, while industrial cellulase contributes about 40–55% of the enzymatic cellulosic ethanol production cost. The estimated costs of producing ethanol from lignocellulosic residues (verge grass, wheat milling residues and woody energy crop/willow) are 0.75–0.99 €/L. The authors noted that the cellulase cost (assumed 0.51 €/L) would have to be reduced at least tenfold and the capital cost by 30% to achieve ethanol production costs comparable to those of ethanol from starch crops. It is also noted that the production cost of cellulosic ethanol depends on feedstocks and their composition as well as plant capacity. The estimated production cost varies from about 0.38–0.48 US\$/L (plant size: feedstocks consumption is 2000 t/day)^{58,105}. For the same plant capacity the production cost of ethanol from corn stover is reported to be 0.71–0.87 US\$/L dependent on the assumed scenarios³¹. The production cost is noted to be 0.56–0.77 US\$/L depending on the feedstock and plant sizes⁴³. The simulated produc-

tion cost of ethanol is reported to be 0.94–1.20 US\$/L which depends on the ethanol yield⁷².

The economic viability, GHG emission and economic performance of lignocellulosic ethanol under extreme weather conditions are also reported to be dependent on the availability of feedstock (weather condition) and the use of single or multiple feedstocks⁷⁴. Wingren et al.⁹⁶ noted that the production cost is also dependent on enzymatic processes. The cost of ethanol produced from softwood based on simultaneous saccharification and fermentation (SSF), and separate hydrolysis and fermentation (SHF) are reported to be 0.57 and 0.63 US\$/L, respectively. The main reason for SSF being lower was the lower capital cost and the overall higher ethanol yield. Major economic improvements in both SSF and SHF could be achieved by boosting income from the solid fuel coproduct, reducing energy consumption and recycling process streams. A techno-economic evaluation of the spruce-to-ethanol process, based on SO₂-catalysed steam pretreatment followed by simultaneous saccharification and fermentation with various process configurations, achieved an ethanol cost of about 0.38–0.50 €/L. Anaerobic digestion of the stillage with biogas upgrading was a demonstrably favorable option in terms of both energy efficiency and ethanol production cost^{8,9} and the contribution of enzyme is reported to be 0.04–0.05 €/L⁹.

Ballerini et al.⁵ concluded that technical and economic optimization of the pretreatment step, the total substitution of lactose by pentose hydrolysate as the main carbon source for enzyme production, and the recycling

Fig. 3. Economics of ethanol production from wood in France⁵

FF: French Franc, odt: oven dry ton, bbl: barrel

of a fraction of the enzyme, the incorporation of pentose in ethanol fermentation, and the utilization of by-products all reduce the production cost of lignocellulosic ethanol. The authors also noted that since ethanol from biomass is tax-exempted, it could compete with gasoline assuming a crude oil price of around US\$50 (Fig. 3). In contrast, it is noted that with current technology, the production cost of cellulosic ethanol (0.75 \$/L) is almost double compared to the market price of oil (0.48 \$/L) and much of the optimism surrounding cellulosic ethanol has faded¹⁰⁷. The externality (environmental and health) cost of bioethanol is also reported to be dependent on the feedstock⁷⁵.

Hamelinck et al.⁵³ stated that the combined effect of higher hydrolysis-fermentation efficiency (to 68%), lower specific capital investments, increased scale (5 times) and lignocellulosic and woody biomass feedstock costs reduced to about 67% could slash ethanol production costs to 59–40% of the current level in 10–20 years or more. The production cost is reported to be slightly higher for wood-produced ethanol compared to that of switchgrass⁹². The production costs of bioethanol from energy crops vary widely due to the complex characteristics of the resource, their site specificity, national policies, labor costs and efficiency of the conversion technologies, but are expected to decline over time¹¹⁰ and it is noted to have clear socioeconomic benefits⁵⁰.

The coproduct revenue and utilization of the excess solid residue for heat and power production had a considerable effect on the process economics, and improved ethanol yield and reduced energy demand resulted in significant production cost reductions (0.41–0.50 €/L)^{100,101}. Sassner et al.¹⁰¹ also concluded that the utilization of pentose fractions for ethanol production helped achieve good process economy, especially in the case of *Salix* or corn stover. It is also noted that ethanol produced from softwood and sold as a low percentage blend with gasoline could ultimately be cost competitive with gasoline without requiring subsidy, but that production from straw would generally be less competitive¹¹⁴. Despite the environmental benefits of ethanol produced from coppice willow, its economic viability remains doubtful at present¹¹⁷. The author argued that the production cost could be reduced significantly if the willow were altered by breeding to improve its suitability for hydrolysis and fermentation. A techno-economic assessment of lignocellulosic ethanol also revealed that commercial success of pilot plants (0.3–67 MW) remains pending, although cost-competitive ethanol can be produced with efficient equipment, optimized operation, cost-effective syngas cleaning technology, inexpensive raw material with low pretreatment cost, high performance catalysts, off-gas

and methanol recycling, an optimal systematic configuration and heat integration, and a high value by-product with a plant capacity of 200 MW⁵⁶. The estimated cost of ethanol from wood varies between 0.50–0.76 US\$/L depending on the plant capacity^{3,40,97}.

The reported enzyme cost of lignocellulosic ethanol varied widely, with the on-site enzyme production/purchase cost reported to date perhaps the most contentious or dubious estimation/prophecy. In the USA, the costs associated with dedicated cellulase production are reported to be 0.1–0.5 US\$/gal ethanol^{1,2,80,135}. It is also predicted that in future, less cellulase will be necessary, due to increased specific enzyme activity: threefold in 2005 and tenfold in 2010¹³⁵. The present enzyme production cost is estimated as 265 \$/m³ (1 \$/gal), but with recent investments and continuous research efforts, this value may drop to 130 \$/m³ (0.5 \$/gal) by 2010^{20,103}. The most astonishing prediction seems to be of enzyme productivity: 600–2000 FPU/g glucose+Xylose between 2005 to 2010¹³⁵, which is subject to considerable doubt. Presently, the enzyme productivity achieved is reported to be 333 FPU/g glucose (NFRI, 2010; unpublished data). Conversely, the cost of cellulase is reported to be 0.51 €/L⁹⁵. The reported enzyme cost (production/purchase) is Canadian dollar (CAD) 12/million FPU (enzyme loading: 10 FPU/g cellulose)⁴⁸. These studies reported a wide variation of the cost of cellulase, hence the ethanol.

The key objective of an LCA study is to provide as complete a portrait as possible of energy consumption, environmental impacts, economic viability and their rebound effects and hence enable effective planning for a sustainable society. Reviewed LCA studies were ridiculous to compare due to deferring system boundaries, objectives and functional units. The main categories discussed in the LCA studies of lignocellulosic bioethanol were energy balance and GHG emissions, soil quality, land and water use, and production cost. Table 1 represents a brief summary of the LCA studies concerning energy balance and GHG emissions, soil quality and land and water use. Table 2 shows a summary of the reported cost of cellulase and the production cost of ethanol produced from cellulosic biomass by different authors.

Discussion

In recent years, the production of biofuels (bio-ethanol and bio-diesel) has been increasing rapidly, affecting virtually all aspects of field crop sectors, ranging from domestic demand and exports to price and the allocation of land area among crops. Adjustments in the agricultural sector are already underway, given the growing interest in renewable energy sources to reduce environmental

Table 1. A brief summary of some recent LCA studies of lignocellulosic bioethanol deal with energy balance/GHG emissions, land use change and soil quality

Reference	Methodology		Scope of the study			
	System boundary	Functional unit	Energy/GHG	Land use change	Soil quality	Toxicity
Blotnitz and Curran, 2007 ⁽⁴⁾	Cradle to wheel	ha, km	o	o	o	o
Cherubini et al., 2009 ⁽²³⁾	Cradle to wheel	ha, km	o			o
Cherubini and Ulgiati, 2010 ⁽²⁵⁾	Cradle to gate	year	o	o	o	o
Cherubini and Jungmeier, 2010 ⁽²⁶⁾	Cradle to wheel	year	o	o	o	o
Fleming et al., 2006 ⁽⁷⁾	Cradle to wheel	km	o	o	o	–
Gabrielle and Gagnaire, 2008 ⁽³⁹⁾	Cradle to gate	L	o	o	o	–
González-García et al., 2010 ⁽⁴⁵⁾	Cradle to wheel	km	o	–	–	o
Luo et al., 2009 ⁽⁷⁸⁾	Cradle to wheel	km	o	–	–	o
McLaughlin et al., 2002 ⁽⁸⁵⁾	Cradle to grave	mg -biomass	o	–	–	–
Monti et al., 2009 ⁽⁸⁶⁾	Cradle to gate	MJ, ha	o	o	o	o
Pimentel and Patzek, 2005 ⁽⁹²⁾	Cradle to gate	ha	o	–	–	–
Saga et al., 2010 ⁽⁹⁸⁾	Cradle to wheel	ha	o			
Schmer et al., 2008 ⁽⁰²⁾	Cradle to gate	ha, year	o	–	o	–
Searchinger et al., 2008 ⁽⁰⁴⁾	Cradle to gate	MJ	o	o	–	–
Searcy and Flynn, 2008 ⁽⁰⁶⁾	Cradle to gate	kWh, L	o	–	–	–
Sheehan et al., 2003 ⁽⁰⁸⁾	Cradle to grave	ha, km	o	o	o	
Spatari et al., 2005 ⁽¹⁵⁾	Cradle to wheel	km	o	o	–	–
Spatari et al., 2010 ⁽¹⁶⁾	Cradle to gate	L	o	o	o	–
Stephenson et al., 2010 ⁽¹⁷⁾	Cradle to gate	ton	o	o	–	–
Uhllein and Schebek, 2009 ⁽²⁶⁾	Cradle to gate	kg	o	o	o	o
Williams et al., 2009 ⁽³²⁾	Cradle to gate	ton, L	o	–	–	–
Wu et al., 2008 ⁽³⁶⁾	Cradle to wheel	km	o	o	o	–
Zah et al., 2007 ⁽⁴⁰⁾	Cradle to wheel	km	o	o	–	o

Note: considered (o), not considered (–)

Table 2. Summary of the reported cost of ethanol produced from different feedstock

Authors	Feedstock, feed rate, cost & yield		² Enzyme loading	Enzyme cost, \$/L	Cost of ethanol for different cases and years, \$/L						Remarks
	Rate, t/d	Cost, \$/t	L/t		1999	2000	2002	2005	2010	2012	
¹ Wooley et al., 1999 ⁽³⁵⁾	*CS, 2000	25.0	257.38–355.79	15–20 FPU	0.079	0.380	–	0.248	0.217	–	Enzyme cost need to be reduced ten fold, dollar value in 1997
McAlloon et al., 2000 ⁽⁸⁴⁾	*CS, 1050	35.0	272.52	–	0.050	0.396	–	–	–	–	Little information is available on enzyme production, dollar value in 1999
¹ Aden et al., 2002 ⁽¹⁾	*CS, 2000	30.0	272.52–339.51	12–17 FPU	0.026	–	0.346	–	0.283	–	Buying of enzymes, dollar value in 2000
¹ Aden et al., 2008 ⁽²⁾	*CS, 2000	60.0–46.0	257.38	–	0.085–0.026	–	1.110	0.666	–	0.351	Enzyme cost is assumed, dollar value in 2002
¹ Dutta et al., 2010 ⁽³¹⁾	*CS, 2000	60.1	–	30–40 mg protein	0.085	–	–	–	0.801	–	Enzyme cost is assumed, dollar value in 2007
¹ Eggeman et al., 2005 ⁽³²⁾	CS, 2000	35	–	15 FPU	0.039	–	–	0.262–0.441	–	–	Enzyme cost is assumed
Reith et al., 2002 ⁽⁹⁵⁾	¹ VG, 2000	20 [€]	152.49	–	0.510 [€]	–	0.920 [€]	–	–	–	Enzyme cost is assumed
Orikasa et al., 2009 ⁽⁸⁹⁾	*RS, 200	15000 [¥]	250.0	–	–	–	–	–	–	–	Enzyme cost is assumed
³ Barta et al., 2010 ^(8,9)	Spruce, 200000 ^a	68.15	254.0–270.0	10 FPU	0.058–0.073	–	–	–	0.548–0.722	–	Enzyme cost is assumed

CS: corn stover, RS: rice straw, VG: filter paper unit, ¹Plant life: 20 years, ²per g–cellulose, *dilute acid pretreatment, ¹Lime pretreatment, ³Plant life 15 years, €: cost in Euros, ^aAnnually

pollution and dependency on foreign oil, which might affect biodiversity, soil erosion and its quality. Hence, the interpretation should be based on agricultural intensity, socioeconomic aspects, land and water use and soil quality as well as the environmental and socioeconomic impacts of lignocellulosic ethanol.

The rate of decomposition of soil organic matter, both that originally in the soil and that added through crop residue mulch, is reported to be higher in the tropics than in temperate climates. Jenkinson and Ayanaba⁶¹ reported that 12 tons biomass/ha/year was insufficient to meet the ecosystem demand for maintenance or sequestration of the SOC pool in Ibadan, Nigeria. The amount of corn (*Zea mays* L.) stover needed to maintain SOC, which is responsible for favorable soil properties, was reported at 5.25–12.50 t/ha/year. Johnson⁶⁴ concluded that the minimum above-ground source carbon (MSC) requirement was 2.5 ± 1.0 ton/ha/year for moldboard plow sites and 1.8 ± 0.44 ton/ha/year for non-tilled and chisel plowed sites which is equivalent to 6.25 and 4.50 ton stover/ha/year, respectively. Above-ground biomass production is reported to be 5.46 and 10.00 ton/ha for paddy and corn, respectively^{64,89}. It is predicted that removal of corn stover from soil could decrease nitrogen-related emissions and also reduce the annual accumulation rates of SOC⁷¹. These estimates indicate that the need for stover to maintain SOC, and thus productivity, are a greater constraint to an environmentally sustainable cellulosic feedstock harvest than that needed to control water and wind erosion¹³¹. In contrast, Sheehan et al.¹⁰⁸ noted that SOC drops slightly in the early years of stover collection but remains stable over the 90-year time frame.

The various cultivation practices and ethanol production technologies have different impacts. GHG emissions in agriculture, for example, are largely determined by the emission of nitrous oxide or methane whereas the ethanol production process by CO₂ is a factor of electricity generation and fermentation. The production of the enzyme used for hydrolysis requires substantial fossil fuel or electricity for air compression, which also generates considerable CO₂ in the chain⁷⁸. The recalcitrance of lignocellulosic biomass still renders the proposed processes complex and costly, but there are grounds for optimism: the application of newly engineered enzyme systems and the construction of inhibitor-tolerant industrial yeast strains, combined with optimized process integration, promises significant improvements⁸³. The production of ethanol from lignocellulosic materials requires considerable research and development before reaching an economically viable stage. If crop residues are utilized in the ethanol industry, there should also be careful consideration to maintain soil organic carbon to avoid a

decline in productivity and soil.

The LCA results of lignocellulosic ethanol are more sensitive to the changes in parameters related to the biomass and ethanol yield. The cultivation practices, enzyme and ethanol production technologies are the main processes, which could significantly affect environmental impacts. Although the biofuel industry provides significant environmental benefits and enlarges rural economies^{68,91,115}, its negative impacts are also reported^{27,28}. Mandatory blending pushes up petrol prices as feedstock are not profitable to use in fuel production given current technologies⁶. The production of biofuels from lignocellulose is also limited by the amount of plant matter which can be sustainably produced and harvested¹²¹. The cultivation of bioenergy and biofuel crops affects biodiversity more directly, both positively and negatively. The changes to policy and land use should be addressed, not simply in terms of species abundance at field level, but also changes to landscape diversity, potential impacts on primary and secondary habitats and potential impacts on climate change³⁶. It is also reported that the production of biofuels is often not competitive with oil unless subsidized or benefiting from tax credits that offset those already provided to the alternatives¹³⁴.

A survey in the USA revealed that only 17% of Iowa's farmers currently have interest in harvesting their corn stover; though 37% are undecided. Farmers who anticipate the negative impacts of corn stover removal on environmental quality tended to be less interested in harvesting it¹²⁵. Farmer participation would be the main key in harvesting/supplying corn stover/crop residues for lignocellulosic ethanol in the near future. Jensen et al.⁶² investigated U.S. energy policy and revealed that it focused more on the producer stage of the lignocellulosic ethanol life cycle than the landowner or consumer stages, despite the need to reflect the requirements of land owners and consumers in future renewable energy policy to ensure steady feedstock supplies and the development of a strong lignocellulosic ethanol industry. Farrell et al.³⁵ suggested that the large-scale use of ethanol for fuel would certainly require cellulosic technology. Tilman et al.¹²⁴ noted that the biofuels policy in the USA has become increasingly polarized, and that political influence seems to be trumping science. Harnessing the best available science, continually updated information should be used to evaluate the extent to which various biofuels achieve their multiple objectives. The development of rigorous accounting rules is urged to assess the impacts of biofuels on the efficiency of the global food system, GHG emissions, soil fertility, water and air quality, and biodiversity³⁶ should be considered in the full life cycle of biofuels production, transformation, and combustion¹²⁴. The ISO series rec-

ommends using methods that reflect the physical relationship, e.g. the mass and energy content or using other relevant variables to allocate, such as the economic value of products, which is similar to the cost allocation methods in managerial accounting⁴⁹. The ecoinvent default allocation includes differentiated allocation factors based on physical-causal relationships, common physical parameters (mass or heating values), and/or the economic values of the valuable outputs of the multi-output process⁶⁶. However, Singh et al.¹¹¹ was against the use of allocation based on economic value. A common methodological framework was developed by the GBEP that could be applied to the LCA of bioenergy production and compared to the full life cycle of its fossil fuel equivalent to improve the transparency and acceptance of the results.

The GBEP is also promoting bioenergy activities, especially in developing countries. Lignocellulosic biomass comes from energy crops/grass, wood, agricultural residues and by-products, and forestry residues. Sukumaran et al.¹²⁰ concluded that despite the abundant biomass residues generated in India as agro-and forest residues, the only feasible feedstock among them would be the crop residues due to problems in terms of collection and logistics. The residues from major agricultural crops like rice, wheat and sugar cane are mostly consumed as fodder or used as raw material for paper industries, and less than 10% are available in surplus, which is also reliant on the weather due to the significant dependence of Indian agriculture on rainwater for irrigation. Biofuels provide about one-third of the total energy in developing countries, and up to 80% of energy in some sub-regions of Africa³³, while biomass often accounts for more than 90% of total rural energy supplies in developing countries, including Bangladesh and India¹¹, and biomass resources are utilized to an extreme and possibly dangerous extent⁶⁸. In Bangladesh, the household sector consumes 80% of total biomass energy and rural households use it almost exclusively for cooking. The combination of population growth with the decreasing per capita land area and growing energy needs puts great stress on the available biomass resources⁷, and requires judicious alteration of energy consumption patterns⁹³. Despite afforestation/reforestation initiatives, the Earth's forest cover is dwindling in various parts of the world (especially in developing countries), because supply is outpaced by ever-growing demand. Therefore, it is hard to imagine how biofuels can be produced from lignocellulosic biomass in developing countries, meet rural energy demand in the form of biomass and avoid deforestation. Concerns over sustainability and perceptions about the negative impacts of biofuels in particular are growing and prompting clos-

er scrutiny of policies designed to expand bioenergy use. A sustainable liquid biofuel program in developing countries may only be feasible if modern energy carriers replace the present inefficient biomass consumption in rural areas. Although there are several explanations for the contradictory results regarding the sustainability of biofuels, the socio-economic aspects concerning biofuel production and its rebound effects must be considered, especially in developing countries.

It is likely that biofuels from lignocellulosic biomass might be an alternative option to reduce GHG emissions and improve energy security in the developed countries where crop residues are known to be abundant, yet the main constraint is the ethanol production cost. The life cycle GHG emission of various forms of bioenergy and their ability to reduce GHG emissions vary widely, and are dependent on the land use changes, choice of feedstock, agricultural practices, refining and conversion process and finally the end use practices. GHG emissions may intensify still further if the forest land is cleared to make way for new energy crops¹²⁷. Commercial biofuel production may target higher-quality lands, due to better profit margins and relegate cereals and subsistence crops to low-quality land which will have knock-on effects on farm income, government payments and food prices. Hence, biofuel feedstock must be produced through biofuel plantations on agriculturally surplus/marginal soils or degraded/desertified soils which do not compete with those dedicated to food crop production, in order to ease pressure on land used for food and feed and avoid any potential conflicts with food production.

Although lignocellulosic biomass is known to be highly reproducible and does not compete with foods, substantial doubts remain concerning the economic and environmental performance of biofuels^{92,104,124,138}. The main bottleneck hampering the sustainability of lignocellulosic ethanol is its production cost. Wide variation was observed in both the reported cost of feedstock, enzymes and microorganisms, and fixed costs, which are dependent on the plant's life-span, yearly operating periods and regions, with the reported enzyme cost the most wide-ranging variable. It is also worth noting that most of those studies referred to the highly optimistic or futuristic enzyme cost reported by the NREL^{1,84,135}, which may or may not be achievable depending on whether a biotechnological revolution takes place in this sector. In an effort to reduce the enzyme production cost, research activities have also been undertaken to identify alternate carbon sources instead of commercial cellulose⁵⁷. It is also noted that the characteristics of electricity and biomass markets and fuel prices are crucial for the future of this sector¹². The reviewed literature indicates that the

biotechnological revolution is a must for the sustainability of bioethanol, especially in the fields of enzymes and microorganisms. In addition, strong renewable energy, an industrial policy to reduce fixed costs and an agriculture policy might be helpful in reducing feedstock costs, where agriculture is heavily government subsidized. Therefore, in-depth studies are required for each stage of the life cycle of bioethanol from lignocellulosic biomass, markets of bioenergy for any investment and commercial production.

Conclusion

This study revealed that environmental impacts and the production cost of bioethanol are dependent on feedstock, conversion technologies, system boundaries, allocation methods, the utilization of by-products and the end use characteristics. The reported hotspots reportedly vary among studies depending on the assumptions, system boundaries and impact categories employed. Although environmental benefits are reported in most studies, economic viability is doubtful with present technologies. Significant variation was observed in the reported production cost of bioethanol, especially enzyme cost. However, the biotechnological revolution is a must for the sustainability of this sector, especially in the field of enzymes and microorganisms. Extended national or international support may enable the prevailing hurdle in this sector to be overcome. In addition, the adaptation of innovative technologies and renewable energy policy (to reduce feedstock cost) may facilitate the production of economically viable bioethanol, especially from agri-residues where agriculture is heavily subsidized. If crop residues are employed in the bioethanol industry, there must be careful consideration of soil quality to avoid any productivity loss. Finally, the bioethanol industry must be vigorously evaluated using LCA methodologies for any investment and commercial production, and its sustainability.

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