Life cycle environmental sustainability of lignocellulosic ethanol produced in integrated thermo-chemical biorefineries

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Abstract: There is a growing interest in producing biofuels and bio-chemicals from lignocellulosic feedstocks in integrated biorefineries. However, the sustainability implications of integrated biorefineries are still poorly understood. Using a life cycle approach, this paper examines environmental impacts of second-generation ethanol produced in thermo-chemical refineries together with chemicals and energy. Four feedstocks are considered: wheat straw, poplar, Miscanthus and forest residue. The results suggest that the production of ethanol from these feedstocks offers significant savings in eight out of 12 environmental impacts when the system is credited for the avoided impacts from producing the co-products from fossil resources. Ethanol from forest residue is the best and wheat straw the worst option for most impacts. Land use change has a significant effect on the global warming potential (GWP) of ethanol. For example, conversion of forest to grow Miscanthus increases the GWP from –234 to 6685 g CO₂ eq./l of ethanol. The effect is opposite when grassland is converted to grow poplar: the GWP is reduced by two times because of carbon sequestration by poplar. The thermo-chemical route for producing ethanol from poplar and forest residue is more sustainable for most impacts than the bio-chemical conversion with the same feedstocks. Although ethanol saves up to 83% of GHG emissions per MJ of fuel compared to petrol, the savings are much smaller (~3%) for current ethanol blends of 5%. Therefore, unless a much higher proportion of ethanol was used, the contribution of second-generation ethanol to climate change mitigation would be small.

Supporting information may be found in the online version of this article.

Keywords: biofuels; ethanol; environmental impacts; integrated biorefineries; life cycle assessment (LCA); lignocellulosic feedstocks
Introduction

Concerns over the increasing costs of fossil fuels, climate change and energy security are some of the drivers for a growing interest in biofuels, with many countries setting mandatory targets to increase their use in transport. As a result, world ethanol production from biomass has increased five-fold between 2000 and 2012, from 17 billion to more than 83 billion litres, while biodiesel production expanded from less than 1 billion to 22.5 billion litres during this period. Globally, the majority of the ethanol is produced from first-generation feedstocks, representing over 80% of liquid biofuels by energy content, with the USA and Brazil being the main producers.

However, large-scale production of biofuels from first-generation feedstocks competes with food production for arable land and water. There are also concerns over greenhouse gas (GHG) emissions associated with the land-use changes (LUCs) caused by the production of feedstocks for biofuels. These issues could be avoided by using second-generation (lignocellulosic) feedstocks instead, including energy crops (e.g. poplar, Miscanthus) and wastes (e.g. agricultural, forestry and municipal waste). It is envisaged that these feedstocks will be used in integrated biorefineries to maximise their utilisation by co-producing biofuels, energy and chemicals. The concept of an integrated biorefinery is similar to a conventional oil refinery, but instead of using fossil feedstocks, fuels and the co-products are produced from biomass.

In general, an integrated biorefinery can use thermo-chemical or bio-chemical processes, or a combination of both. Each technological route has its advantages and disadvantages but they all have to overcome a range of technological issues before they can become a commercial reality. In addition to these, integrated biorefineries also face a number of sustainability challenges – environmental, economic and social – which must be evaluated carefully on a life cycle basis to avoid shifting the issues along supply chains. The life cycle approach is also required by various legislative acts related to biofuels, including the EU Renewable Energy Directive and the US Energy Independence and Security Act.

A number of studies have examined life cycle environmental impacts of lignocellulosic ethanol produced in different production processes and from various feedstocks. For the latter, most have focused on two feedstocks – corn stover and switchgrass – largely in the USA. A small number of studies have assessed environmental impacts of biofuels from other lignocellulosic feedstocks, such as Miscanthus, wheat straw, poplar, willow and forest residues, in Europe and other regions. In terms of production processes, the majority have examined the production of ethanol via the National Renewable Energy Laboratory (NREL) bio-chemical process which utilises dilute acid pretreatment followed by enzymatic hydrolysis, while some studies have also assessed the bio-chemical route but with different pre-treatment options, such as sulphuric acid-catalysed steam explosion, uncatalysed steam explosion and ammonia fiber expansion. By comparison, there are only few life cycle assessment (LCA) studies of thermo-chemical processing of lignocellulosic feedstocks. For example, Mu et al. compared the thermo-chemical and bio-chemical routes producing ethanol from four feedstocks (wood chips, corn stover, waste paper and wheat straw) but considered only three environmental impacts: GHG emissions, water consumption and fossil energy use. Cherubini and Ulgiati also compared the two routes, considering a broader range of impacts, but examined two feedstocks only: corn stover and wheat straw. Wang et al. focused on the thermo-chemical route but considered only GHG emissions.

In this paper we go beyond the previous studies of thermo-chemical biorefineries to assess the life cycle environmental sustainability of thermo-chemical integrated biorefineries by considering a range of environmental impacts and several lignocellulosic feedstocks, based on their availability around the world. The effects of potential LUC on the GHG emissions from energy crops used for the production of ethanol are also discussed. Furthermore, we also compare the results of this study with ethanol produced by bio-chemical conversion as well as with ethanol produced from fossil feedstocks. Finally, we discuss the potential of ethanol produced in thermo-chemical biorefineries to contribute to a reduction of GHG emissions using currently prevalent ethanol/petrol blends.

The following section gives an overview of lignocellulosic feedstocks and their global availability, followed by a brief description of conversion technologies envisaged for use in integrated thermo-chemical refineries.

Lignocellulosic feedstocks and thermo-chemical conversion technologies

Feedstocks

The main sources of lignocellulosic biomass include residues from agriculture and forestry as well as energy crops such as perennial grasses and different wood species. The availability of different types of residues varies by country and by region. The main agricultural residues available

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in Asia, Europe and North America are corn stover and wheat straw. Around 5.1 billion dry tonnes per year of agricultural residues, such as corn stover, wheat straw and rice husks, are produced globally. Since they are inexpensive, renewable and widely available in many countries, they have a significant potential for biofuel production in the long term. However, excessive removal of crop residues from agricultural fields could lead to soil erosion and decrease in crop yields.

It is expected that availability of agricultural and other residues will increase substantially in the future. For instance, in the USA, there is an estimated potential for annual production of 1.3 billion tonnes of agricultural and forestry waste without the need for new land; this would be sufficient to meet more than one third of the current demand for transportation fuels. In the UK, around 27 million tonnes of second-generation feedstocks could be available per year, of which more than 80% are agricultural and forestry residues. Energy crops, such as Miscanthus and poplar, could provide an additional 10% of the feedstock. Currently, the UK has the largest cultivation of energy crops in Europe. Their production is also growing in other European countries, including Finland, Germany, Italy, Spain and Sweden as well as in North America.

Thermo-chemical conversion technologies

The thermo-chemical route converts biomass to fuels, chemicals and energy using pyrolysis, gasification or liquefaction. Pyrolysis is the thermal decomposition of materials in the absence of oxygen. It converts the organic portion of the feedstock to char and volatile gases containing non-condensable vapours (syngas) and condensable tars which form bio-oil. The bio-oil is a low-viscosity combustible product which can be upgraded to an alternative fuel or used as a feedstock for chemicals. However, because of low volatility and other changes in its chemical composition, the bio-oil undergoes a phase separation over time, hence causing storage problems. The char can be used as soil improver or for process heat. Pyrolysis has been applied to different types of feedstock, including grass, woody biomass, straw, bagasse and municipal solid waste. The yield and composition of pyrolysis products depend on the composition of the feedstock, the pyrolysis technique used and the operating conditions, including temperature, residence time and heating rate. Gasification requires pre-treatment, such as drying, screening or grinding, to increase the surface area for further downstream processing. In this conversion, the biomass is decomposed into syngas, char and ash at temperatures of 600–1000°C. To prevent combustion, the conversion process is carried out in the presence of a controlled amount of oxygen, steam or air. The syngas is then cleaned up to remove any impurities, such as sulphur and unreacted carbon, using carbon beds or amine. For the production of chemicals and transportation fuels, the purified syngas is reacted with steam in the water-gas shift reaction to balance the H₂/CO ratio. The syngas is then passed through a catalyst in the Fisher-Tropsch process, producing liquids such as methanol, ammonia and mixed alcohols, which can be converted into ethanol and other fuels or chemicals. Any unreacted syngas is normally burned to generate electricity.

Liquefaction is a direct conversion of biomass to bio-oil in a catalytic reaction at high pressure (5–10 MPa) and temperature (250–325°C). Liquefaction of biomass can be direct and indirect. The former involves rapid pyrolysis to produce bio-oil and/or condensable organic vapours. Indirect conversion is not considered a thermo-chemical process but rather a chemical upgrade, such as that carried out in the Fisher Tropsch process. Indirect liquefaction can be carried out using either an alkali, acidic, or glycerine medium to produce bio-oil.

By comparison, the bio-chemical route includes anaerobic digestion, chemical and biological conversion. Anaerobic digestion is a biological process carried out in the absence of oxygen in which anaerobic bacteria are used to produce biogas from organic matter. The solid and liquid residues (digestate) can be used as fertiliser. Chemical conversion involves use of different reactions and chemicals to convert biomass into fuels. For example, transesterification is the most-widely used method for producing biodiesel. Biological conversion of biomass involves feedstock pre-treatment, its conversion to sugars, fermentation and processing of the fermentation products to produce ethanol.

Methods

There are no commercial thermo-chemical biorefineries in operation at present so most studies are based on conceptual designs. This study is based on the Aspen Plus model developed by the NREL with gasification used as a conversion technology. Since the NREL design considers only poplar as a feedstock, the Aspen model has been modified in this work to consider the following four feedstocks: wheat straw and forest residue as wastes and poplar and Miscanthus as energy crops. These were chosen based on the global feedstock availability and future trends, as...
discussed earlier. Each feedstock is considered separately rather than their blend so that four Aspen models have been created. The outputs from the models have then been used to estimate the environmental impacts of ethanol production from different feedstocks. The impacts have been estimated using LCA, following the ISO 14040 and 14044 LCA guidelines. The LCA software GaBi v.4.4 has been used for life cycle modelling and the impacts have been calculated applying the CML 2 method. The following 12 environmental impacts are considered: global warming, abiotic depletion of elements and fossil resources, acidification, eutrophication, freshwater, marine and terrestrial ecotoxicity, human toxicity, ozone layer depletion, photochemical smog and land use.

Further details on the system, data and assumptions are provided below.

**Goal and scope of the study**

The main goals of this study are to:

- assess the life cycle environmental sustainability of ethanol produced in an integrated thermo-chemical refinery using different second-generation feedstocks;
- examine the effects of potential LUC on the GHG emissions from energy crops used for the production of ethanol;
- compare the environmental sustainability of ethanol from the thermo-chemical route with that produced by bio-chemical conversion;
- compare the environmental impacts of second-generation ethanol with that produced from first-generation feedstocks as well as with ethanol produced from fossil resources; and
- compare ethanol from the thermo-chemical process with petrol and to assess its potential to reduce GHG emissions when blended with petrol.

As indicated in Fig. 1, the scope of the study is from ‘cradle to gate’, with the latter representing the refinery gate. The following stages are considered: feedstock cultivation (where relevant); feedstock collection and transportation to the refinery; and production of ethanol and its co-products, here assumed to be butanol and propanol.

The use of the products as well as their distribution are excluded from the system boundaries. However, the use of ethanol in vehicles is considered later in the paper when ethanol is compared to petrol, where the system boundary is from ‘cradle to grave’ for both. The impacts from construction and decommissioning of the refinery are excluded from the study since the impacts of infrastructure per unit of product are negligible over the (long) lifetime of industrial plants.

The study is based on the unit of analysis (functional unit) defined as the production of 1 litre of ethanol. To enable comparisons of ethanol from different feedstocks, the amount of ethanol produced in the refinery is assumed to be equal for each feedstock. Instead, the amounts of the feedstocks and the output of the co-products are varied, depending on the composition of the feedstock (see Table S1 in Supporting Information). The data for the refinery are summarised in Table 1. The refinery is assumed to be based in the UK. Further details on the system are provided below.

**System description**

After cultivation and harvesting (if relevant), the feedstock is transported to the refinery, where it is first dried and then gasified. The resulting syngas is cleaned up and conditioned and then sent to the alcohol synthesis unit to produce mixed alcohols. These are then separated to recover ethanol, propanol and butanol. These process steps...
would result from the biomass potassium reacting with the silicate compounds in olivine. Ash and sand particles from the syngas exiting the gasifier and flue gases from char combustor are removed in cyclone separators.

**Syngas clean-up:** In this process, firstly the tars in syngas are reformed to additional CO and H₂ in a tar reformer unit. The water-gas shift reaction also occurs in the reformer. The syngas is reacted with the tar reforming catalyst in an entrained flow reactor. The tar reformer operates at 890°C and the energy needed is provided from the heat generated by the hot catalyst and combustion of additional syngas and unreacted gases from the alcohol synthesis reactor in the regenerator. The hot reformed syngas is then cooled through a heat exchange with other process streams and scrubbed with water to remove impurities, such as particulates, ammonia, halides and recalcitrant tars. The cooled and quenched syngas is then compressed using the centrifugal compressor. The hydrogen sulphide and carbon dioxide are removed from the syngas by an acid gas removal unit. Sulphur is also removed to avoid contamination of the molybdenite catalyst used in the alcohol synthesis process. After heat recovery, the remaining low-quality heat in the flue gas from the catalyst regenerator is used for feedstock drying.

**Gasification:** A circulating fluidised bed gasifier is used to convert the dried biomass to syngas and char with the aid of steam which acts as a fluidising medium and a reaction agent. Heat is supplied indirectly by circulating hot olivine ‘sand’ between the gasifier and the char combustor, where the temperatures are maintained at 890°C and 995°C, respectively. The hot flue gases from the char combustor are used to reheat the olivine. A small amount of magnesium oxide must be added to the fresh olivine to avoid the formation of glass-like bed agglomerations that would result from the biomass potassium reacting with the silicate compounds in olivine. Ash and sand particles from the syngas exiting the gasifier and flue gases from char combustor are removed in cyclone separators.

**Alcohol synthesis:** In this stage, the syngas is further compressed and heated to about 300°C. The heated syngas is then passed across a bed of molybdenite catalyst for conversion into alcohol. The product gas containing the alcohols is cooled and separated from the unconverted syngas.
Alcohol separation: The crude mixed alcohol is degassed and dried using a molecular sieve. The resulting product is distilled in a distillation column to separate ethanol from other alcohols, i.e. propanol, butanol and methanol. Ethanol, propanol and butanol are then sent to storage, while any methanol and water mixture is recycled back to the alcohol synthesis unit.

Energy use: The system is self-sufficient in terms of energy. Electricity is generated by using 28% of the unconverted syngas. Steam is produced by recovering heat from the hot process streams throughout the plant. All process units in the plant are fed with steam indirectly except for the gasifier where steam is injected directly from the low-pressure turbine exhaust.

Waste management: Solid waste generated by the refinery consists of spent catalysts, sand and ash, all of which are assumed to be landfilled. Although ash could be used as soil improver or as an aggregate in the construction industry, at this stage it is not known if and how much of that would actually be used, so that only disposal by landfill is considered.

Data sources

As mentioned earlier, the refinery data are based on the conceptual design developed by NREL and modelled in Aspen Plus but adapted to enable consideration of different feedstocks. All background LCA data are from the Ecoinvent database, except for poplar and Miscanthus which are from GEMIS as these were not available in Ecoinvent. The LCA data for transport are also from Ecoinvent, assuming that all the materials used in the system are transported to the refinery by a 40 t truck over the distance of 100 km.

Allocation of environmental impacts

The refinery produces several outputs so that it is necessary to allocate the impacts between them. Following the ISO 14040/44 guidelines, the system expansion or the ‘avoided burden’ approach has been used, with the system being credited for producing the co-products in alternative production systems. For these purposes, propanol is assumed to be produced from propene and butanol from propylene and their impacts have been subtracted from the total impacts from the system. The LCA data for the system credits have been sourced from the Ecoinvent database.

As the choice of allocation method can affect the results, we have also applied economic allocation. For this, the impacts have been allocated proportionally between ethanol and its co-products based on their respective market prices in 2012. The market price assumed for ethanol is £808/t, for propanol £1345/t and butanol £1323/t.

Results and discussion

In this section, the environmental impacts estimated using system expansion to credit the system for the co-products are discussed first. This is followed by the discussion of the results for economic allocation.

System expansion

As shown in Fig. 2, ethanol from forest residue is the best option for eight out of 12 impacts considered; poplar has the lowest impacts for three categories and wheat straw for one. Ethanol from wheat straw is the worst option for all impacts except for land use, which is highest for Miscanthus. These results are discussed in more detail below, first for the global warming potential and then for the other impacts.

Global warming potential (GWP)

As per standard LCA practice, biogenic CO₂ emissions are excluded from the GWP as they are part of the natural carbon cycle. Similarly, biogenic carbon storage in the products is not considered as this carbon will be released during the use of ethanol in vehicles (considered later in the paper). The results discussed below also exclude the GHG emissions from LUC associated with the cultivation of energy crops; these are discussed separately in the section on LUC.

The results in Fig. 2 suggest that the GWP of ethanol is negative for all four feedstocks, indicating a saving in GHG emissions because of the credits for the co-products, especially for propanol. Ethanol from forest residue is the best option with the GWP of −325 g CO₂ eq./l. The greatest contributor to the impact from forest residue is its transport to the refinery (58% before system credits). This is due to its high moisture content so that a greater mass has to be transported per litre of ethanol produced compared to the other feedstocks (Table S1 in Supporting Information). Wheat straw is the worst alternative but it still saves 32 g CO₂ eq./l. The lower GHG savings for ethanol from wheat straw are due to the high contribution of the cultivation stage which is responsible for more than 90% of the GWP for wheat straw (before the system credits).

Other environmental impacts

Similar to the GWP, the abiotic depletion potential (ADP, fossil), ozone layer depletion potential (ODP) and photochemical oxidants creation potential (POCP) are...
Production contributes considerably to several environmental impacts (before system credits), including depletion of fossil resources (84%), EP (78%), HTP (94%), ODP (75%), TETP (>99%) and land use (>99%). Except for the TETP and land use, most of these impacts are associated with the fertilisers, pesticides and fuels used during cultivation. The agricultural stage is also the major contributor to the impacts associated with Miscanthus and poplar, including depletion of fossil resources (>66%), HTP (>54%), ODP (>61%), TETP (>87%) and land use (>99%). Most of the FAETP and MAETP for all four feedstocks are from the disposal of ash from the gasifier and char combustor. Note that herbaceous biomass (wheat straw and Miscanthus) has higher ash content compared to woody biomass (poplar and forest residues; Table S1) so that the quantity of ash produced from the former is 3–6 times higher than from...
the latter. Air emissions from the char combustor and syngas clean-up are responsible for most of the acidification and photochemical smog across all the feedstocks.

**Economic allocation**

Applying economic allocation based on the quantities and market prices of ethanol and the co-products results in 81% of impacts being allocated to ethanol, 17% to propanol and the rest to butanol. The environmental impacts in Fig. 3 show a similar trend as for the system expansion, with ethanol from forest residue being overall the best option, followed closely by poplar and Miscanthus. Wheat straw is the worst option for all impacts except for land use for which it is the best alternative. Therefore, while the absolute values for the impacts are different for the two allocation methods, the ranking of the options is the same for most of the impacts.

**Land use change (LUC)**

As mentioned in the introduction, GHG emissions from LUC can be high. In this section we consider how direct LUC would affect the GWP of ethanol. Note that consideration of indirect LUC is outside the scope of this paper. As LUC is relevant only to energy crops, only Miscanthus and poplar are considered. The following options are explored for different current land use, using the UK as an example:

i) Miscanthus:
- conversion from forest land to perennials with 20 t CO₂ eq./ha yr.⁵⁶ and
- conversion of grassland to perennials with 6.7 t CO₂ eq./ha yr.⁵⁶

ii) Poplar:
- conversion of current forest land to poplar forest assuming GHG emissions of 2.5 t CO₂ eq./ha yr.⁵⁷ and
- conversion from grassland to forest land with -1.5 t CO₂ eq./ha yr (the negative value is due to the GHG emissions from grassland clearing being lower than the CO₂ sequestration by poplar forest). ⁵⁷

The results indicate that the effect of LUC on the GWP of ethanol from Miscanthus is much more pronounced than

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**Figure 3.** Life cycle environmental impacts of 1 litre of ethanol produced in the thermo-chemical refinery (economic allocation) [System boundary: cradle to gate. The values shown against each bar indicate the total impact based on allocation among the ethanol and refinery co-products in proportion to their economic value. Some impacts have been scaled to fit. To obtain the original values of the impacts, multiply the values shown on the graph by the factor shown on the x-axis for relevant impacts. For impacts nomenclature, see Figure 2.]
for poplar (Fig. S1 in Supporting Information). Conversion of forest land to cultivate Miscanthus increases the impact from -234 to 6885 g CO₂ eq./l and to 2279 g for the conversion of grassland. If instead an existing forest is converted to a poplar forest, the GWP of ethanol decreases from -231 to 740 g CO₂ eq./l. However, if grassland is converted to a poplar forest, the GWP is actually reduced to -486 g CO₂ eq./l. This is due to the amount of carbon sequestered by the poplar forest being greater than the emissions released during the grassland conversion.

This analysis shows that the GWP is very sensitive to the LUC and should therefore be considered carefully, particularly as the GWP savings from the co-product credits in some cases are not high enough to compensate for the emissions from the LUC. The impact from the LUC would be even higher if the system produced only ethanol as there would be no credits for the co-products to compensate for the GHG emissions from LUC.

Comparison with ethanol from bio-chemical refinery

This section compares the environmental impacts of ethanol produced in a thermo-chemical biorefinery with that produced in a bio-chemical plant. The data for the latter are based on Falano et al., who considered the same feedstocks as in this study. Given that ethanol from forest residue and poplar has the lowest impacts for most impact categories for both processing routes, these two feedstocks are chosen for comparison here. The life cycle inventory data for the bio-chemical refinery for poplar and forest residue are summarised in Table S3 in Supporting Information.

In addition to ethanol, the plant produces acetic and lactic acids as well as electricity which is sold to the grid. As shown in Fig. 4 for system expansion, the thermo-chemical refinery is environmentally more sustainable for both feedstocks for seven out of 12 impacts than the bio-chemical: GWP, ADP (elements), FAETP, HTP, POCP, TETP and land use. This is mainly because the thermo-chemical refinery requires a lower amount of feedstock and chemicals per litre of ethanol than the bio-chemical plant. However, the thermo-chemical process has higher impacts than the bio-chemical option for the AP, EP, MAETP and ODP for both feedstocks. This is due to various reasons, including the emissions from the gasifier and differences in the credits for the different co-products. For the ADP (fossil), the best option is ethanol from poplar produced in the bio-chemical refinery.

Figure 4. Comparison of ethanol from thermo-chemical and bio-chemical refineries (system expansion) [System boundary: cradle to gate. Some impacts have been scaled to fit. To obtain the original values of the impacts, multiply the values shown on the graph by the factor shown on the x-axis for relevant impacts. For impacts nomenclature, see Figure 2.]
The comparison based on the economic allocation (Fig. 5) indicates a similar trend as for the system expansion except that here the thermo-chemical conversion is better for eight impacts: GWP, ADP (fossil), FAETP, HTP, MAETP, ODP, TETP and land use. The bio-chemical option has lower ADP (elements), AP, EP and POCP. This is mainly due to the higher NOx and SOx emissions from the gasification process in the thermo-chemical refinery.

Therefore, the results of this comparison suggest that on balance, assuming all the impacts are of equal importance, producing ethanol from poplar and forest residue via the thermo-chemical route is environmentally more sustainable than via the bio-chemical.

Comparison with ethanol from fossil resources

In this section, the environmental impacts of ethanol produced in an integrated thermo-chemical biorefinery are compared with the impacts of ethanol produced in a conventional refinery from ethylene, which in turn is made from fossil resources. The LCA data for the fossil-based ethanol are from the Ecoinvent database.53

The results in Fig. 6 suggest that, in comparison to ethanol from poplar, Miscanthus and forest residue, ethanol derived from fossil resources has higher impacts for most categories, except for acidification, freshwater ecotoxicity and land use. On the other hand, compared to ethanol from wheat straw, it is a better option for six out of 12 impacts: acidification, eutrophication, freshwater, human and terrestrial toxicity as well as land use.

Thus, although ethanol from biomass is more sustainable for some impacts, including depletion of fossil resources and the GWP, some of its other impacts, such as toxicity and land use, are higher than for fossil ethanol. These results highlight the importance of considering a wider range of impacts rather than GHG emissions alone to provide a more balanced picture and avoid solving the problem of climate change at the expense of other environmental impacts.

Comparison with ethanol from first-generation feedstocks

One of the main reasons for the drive towards using second- instead of first-generation feedstocks for biofuel production is the competition of the latter with food production. However, it is unclear how these feedstocks compare for the environmental impacts, particularly if second-generation ethanol is produced in an integrated
As indicated in Fig. 7, ethanol from poplar and forest residue has lower impacts than ethanol from wheat and refinery. To find out, we compare here second-generation ethanol considered in this study with that produced from two first-generation feedstocks used for ethanol production in the UK: wheat and sugarbeet. The LCA data for the latter have been sourced from CCaLC\textsuperscript{58} and Foteinis \textit{et al.}\textsuperscript{39} respectively.

As indicated in Fig. 7, ethanol from poplar and forest residue has lower impacts than ethanol from wheat and
sugarbeet across all the categories considered. The only exception is eutrophication for which ethanol from sugarbeet has a lower impact than ethanol from any of the second-generation feedstocks considered here. Ethanol from sugarbeet also has lower acidification, freshwater and terrestrial ecotoxicity than ethanol from wheat straw. Similarly, compared to Miscanthus, ethanol from sugarbeet also has lower freshwater ecotoxicity. Furthermore, ethanol from wheat has lower depletion of elements as well as freshwater and terrestrial ecotoxicity than ethanol from wheat straw.

Therefore, on balance, it could be concluded that second-generation ethanol from integrated thermo-chemical refineries is environmentally more sustainable than from first-generation biofuels. Combined with the fact that they would not compete with food production systems, provided that energy crops are grown on marginal land, would favour them over first-generation fuels. Cultivating energy crops on marginal land would also avoid GHG emissions from LUC. However, the crop yields could be lower owing to poor soil quality and higher inputs of fertilisers and irrigation water may be required, thus increasing the impacts.

Comparison with petrol

Finally, ethanol from the second-generation feedstocks considered in this paper is compared to petrol. To take into account the impacts from the use of the fuels, the comparison is carried out from cradle to grave. This is particularly important for the GWP because of the emissions of fossil CO_2 from petrol. Note that for ethanol the biogenic CO_2 emitted during its use is not taken into account as that is equivalent to the amount of CO_2 sequestered from the atmosphere by the feedstocks during their growth.

Since ethanol and petrol have a different energy content, their impacts are compared per MJ of energy. The impacts of ethanol per litre estimated above have been converted to be expressed per MJ assuming a lower heating value (LHV) of 21.5 MJ/l (the LHV of petrol is assumed at 32.85 MJ/l53). The LCA data for petrol have been sourced from Ecoinvent,53 taking into account all life cycle stages from the extraction and processing of crude oil to the use of petrol in vehicles. The LCA data for the use of ethanol in vehicles are also from Ecoinvent and they have been added to the cradle-to-gate impacts estimated in this study.

The results in Fig. 8 show that ethanol saves significant GHG emissions in comparison to petrol, ranging from 83% for wheat straw to 96% for forest residue ethanol. However, these savings are estimated on the basis of a 100% replacement of petrol by ethanol, which is currently not the case in almost any country in the world (except in e.g. Brazil). In the UK, for example, the law allows only up to 5% (vol.) ethanol to be added to unleaded petrol60; most other countries in Europe use 5% blends.

If we consider the 5% blend of ethanol with petrol (known as E5), the saving in GHG emissions would be much lower, ranging between 2.6 and 3.2% per MJ of fuel (Fig. 9). To put this in context, the GHG emissions from transport in the UK were 68 million tonnes of CO_2 eq. in 2010, contributing 12% to the total national GHG emissions in that year.61 If 5% of ethanol considered here were added to all the petrol used annually in the UK, a total
reduction in the GHG emissions would be on average 0.35% per year. Given that the UK has an overall target of 80% reduction of GHG emissions by 2050 on 1990 levels, this would represent a very small saving overall. As the above results show, the savings of the magnitude required would only be achievable if pure ethanol was used in vehicles. Similar is true for the other environmental impacts (Fig. 9) for which the savings relative to pure petrol range from 0.1 to 7%. The exception is human toxicity which would be reduced by between 31 and 39%, depending on the type of feedstock.

However, some of the impacts would be higher for the wheat-straw ethanol blend than for pure petrol: terrestrial ecotoxicity is three times higher while the depletion of elements, acidification, eutrophication and freshwater ecotoxicity are 3–26% higher. The depletion of elements associated with the other feedstocks is also higher for the ethanol blend than for petrol (9–24%). Furthermore, freshwater ecotoxicity is 8% higher for the blend with ethanol from Miscanthus than for pure petrol.

In summary, the reductions in GHG emissions and some other impacts are small for the 5% blends of ethanol and petrol currently used in the UK, Europe and some other countries such as Canada, India and Mexico. If, on the other hand, a higher proportion of ethanol were to be added to petrol, some other impacts would increase for some of the feedstocks, particularly acidification, eutrophication and freshwater and terrestrial ecotoxicity.

Conclusions

In this study, life cycle environmental impacts of ethanol produced from lignocellulosic feedstocks via a thermo-chemical route have been investigated. The following lignocellulosic feedstocks have been considered: wheat straw, forest residue, poplar and Miscanthus. In addition to ethanol, the refinery is assumed to produce propanol and butanol. Two approaches have been considered for allocating the impacts among the co-products: system expansion and economic allocation. The results suggest that, when the system is credited for the avoided impacts for the co-products, eight out of 12 impacts are negative, including depletion of abiotic resources and the global warming potential. Ethanol from forest residue is the best feedstock option for eight impacts. Ethanol from poplar has the lowest freshwater and marine ecotoxicity as well as photochemical oxidants creation potential while the fuel from wheat straw has the lowest land requirement.

If the impacts are allocated based on the economic value of the co-products, ethanol from forest residue is the best option for most impacts, followed closely by poplar and Miscanthus. Using wheat straw to produce ethanol is the worst option for all impacts except for land use. Although the absolute values for the impacts are quite different between the two allocation methods, the ranking of ethanol from different feedstock options remains the same across the impact categories.
The effect of land use change on the global warming potential is significant. Conversion of forest land to cultivate Miscanthus increases the impact from -234 g CO₂ eq./l to 6685 g and to 2279 g for converting grassland. If an existing forest is converted to a poplar forest, the GWP decreases from -231 to 740 g CO₂ eq./l ethanol. However, converting grassland to poplar forest reduces the GWP to -486 g CO₂ eq. because of the carbon sequestration by the forest. Thus, land use change is a critical factor for energy crops and should be considered carefully to avoid the unintended consequences of biofuel production.

The results also suggest that, based on the assumptions made in the study, ethanol produced from poplar and forest residue via the thermo-chemical route is more environmentally sustainable than ethanol from the same feedstocks produced by the bio-chemical route for the majority of impact categories, including the global warming potential. This is because of the better ethanol yields and lower use of chemicals in the thermo-chemical process.

Ethanol from fossil-derived ethylene has higher impacts than ethanol from poplar, Miscanthus and forest residue; the exceptions are acidification, freshwater ecotoxicity and land use. However, it is a better option than ethanol from wheat straw for six out of 12 impacts: acidification, eutrophication, freshwater and terrestrial ecotoxicity, human toxicity and land use. Therefore, while ethanol from second-generation feedstocks is a better option for some impacts, it is not necessarily environmentally more sustainable than fossil-derived ethanol for all the impacts.

In comparison to first-generation, second-generation ethanol from the integrated thermo-chemical refinery is more sustainable for most impacts. The exception is eutrophication for which ethanol from sugarbeet is a better option than second-generation ethanol. Ethanol from both sugarbeet and wheat has lower freshwater and terrestrial ecotoxicity than ethanol from wheat straw.

Potentially significant savings in GHG emissions could be achieved by using ethanol compared to petrol, ranging from 83% per MJ for wheat straw to 96% for forest residue, assuming that ethanol fully replaces petrol. However, taking into account that today most countries use 5% ethanol blends, this saving reduces to around 3% per MJ of fuel. For the UK, this would mean a total saving of only 0.35% per year if 5% ethanol was added to all petrol consumed today. Similar is true for the other impacts except for human toxicity, which could be reduced by up to 39%. Therefore, unless the proportion of ethanol mixed with petrol increased substantially to the point of replacing it, the potential of second-generation ethanol from integrated biorefineries to contribute towards climate change mitigation is very small.

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