

Available online at www.sciencedirect.com





Energy 31 (2006) 3362-3380

www.elsevier.com/locate/energy

Renewability and life-cycle energy efficiency of bioethanol and bio-ethyl tertiary butyl ether (bioETBE): Assessing the implications of allocation

João Malça^a, Fausto Freire^{b,*}

^aDept. of Mechanical Engineering, ISEC, Coimbra Polytechnic Institute, 3030-199 Coimbra, Portugal ^bDept. of Mechanical Engineering, Faculty of Sciences and Technology, University of Coimbra, Pólo II, 3030-199 Coimbra, Portugal

Abstract

Biofuels are expected to play an increasingly important role in the transportation market, as we search for ways to reduce fossil fuels depletion and emissions. However, the extent to which biofuel can displace petroleum-based fuels depends on the efficiency with which it can be produced. To demonstrate that biofuel has a positive energy balance—i.e. more energy is contained in the fuel than is used in the production—a life-cycle approach must be employed. This paper presents a Life-Cycle Energy Analysis of bioethanol (from sugar beet or wheat) and bioETBE systems in France. Physical and economic data was collected. A systemic description was implemented and the energy used throughout was calculated. A novel indicator aiming at characterizing the renewability of (bio)energy sources is proposed—the energy renewability efficiency (ERenEf). ERenEf measures the fraction of final fuel energy obtained from renewable sources. Inventory results-calculated using four different allocation approaches and ignoring co-product credits-are analyzed in order to understand the effect of allocation in the energy efficiency and renewability results. Sensitivity analysis shows that allocation has a major influence in the results. This research concludes that bioethanol produced in France is clearly favorable in terms of primary energy. A maximum ERenEf value of 48% was obtained for wheat-based ethanol (mass allocation), meaning that 48% of the biofuel energy content is indeed renewable energy. Fossil energy savings when gasoline is displaced by bioethanol, bioETBE or E5 are calculated. In particular, pure bioethanol may save up to 0.70 MJ, depending on whether wheat or sugar beet is used and on the allocation procedure adopted. © 2006 Published by Elsevier Ltd.

Keywords: BioETBE; Bioethanol; Energy efficiency; Energy policy; Life cycle assessment; Renewability

1. Introduction

The transport sector is almost exclusively dependent on petroleum-based fuels and emerging attention has been given to the potential use of biomass as the basis for production of an alternative (and renewable) motor vehicle fuel—the *biofuel* [1]. More recently, the global warming problem has been increasingly a focus of

*Corresponding author. Tel.: + 351 239 790700/39; fax: + 351 239 790701.

E-mail address: fausto.freire@dem.uc.pt (F. Freire).

^{0360-5442/} $\$ - see front matter $\$ 2006 Published by Elsevier Ltd. doi:10.1016/j.energy.2006.03.013

Nomenclature

BioETBE ETBE produced from bioethanol BioMTBE MTBE produced from biomethanol cumulative energy demand, MJ CED DDGS distiller's dried grains with solubles gasoline blended with 5% (by volume) bioethanol E5 E85 bioethanol blended with 15% (by volume) gasoline $E_{\rm in, prim}$ total accumulated inputs, in primary energy terms, MJ/kg $E_{\rm in,\ fossil,\ prim}$ total accumulated fossil inputs, in primary energy terms, MJ/kg $E_{\rm in, ren, prim}$ total accumulated non-fossil (renewable) inputs, in primary energy terms, MJ/kg ERenEf energy renewability efficiency, % energy requirement, MJ/MJ $E_{\rm req}$ European Union EU ETBE ethyl tertiary butyl ether ETBE(15) gasoline blended with 15% (by volume) bioETBE FEC fuel energy content, MJ/kg FER fossil energy ratio, MJ/MJ gross energy requirement, MJ/kg GER GS green syrup ISO International Organization for Standardization life cycle assessment LCA LCEA life cycle energy analysis LCEE life cycle energy efficiency, MJ/MJ life cycle inventory LCI lower heating value, MJ/kg LHV MTBE methyl tertiary butyl ether NEV net energy value, MJ Reid vapor pressure RVP SI spark ignition tonne of oil equivalent TOE

attention and greater use of biofuels, which have been able to compete with (and displace) petroleum-based fuels in the transportation market place, could help to comply with the Kyoto Protocol [1].

Biofuels originate from plant oils, sugar beets, cereals, organic waste and the processing of biomass. Biological feedstocks that contain appreciable amounts of sugar—or materials that can be converted into sugar, such as starch or cellulose—can be fermented to produce alcohol (bioethanol) to be used in gasoline engines. Bioethanol feedstocks can be classified in: (i) sugar feedstocks (e.g. sugar beet and sugar cane); (ii) starch feedstocks (e.g. wheat, corn and barley) and (iii) cellulosic feedstocks (e.g. trees, forestry processing residues and grasses). Bioethanol can also be used as a feedstock to produce Ethyl Tertiary Butyl Ether (bioETBE), through the chemical reaction of bioethanol with isobutylene—a by-product of the petroleum refining process. Plant oils (colza, soybean, sunflower, palm, etc.), animal fats, or even recycled cooking greases can be converted into a diesel substitute (biodiesel).

Bioethanol and biodiesel have been known as motor fuels for many decades. For example, in the beginning of the XX century, Henry Ford designed the Model T in the expectation that bioethanol produced by American farmers would be used as its primary fuel. Analogously, the concept of using vegetal oil as an engine fuel dates back to the end of the XIX century, when Rudolf Diesel developed the first engine to run on peanut oil.

The European Commission White Paper [2] calls for dependence on oil (currently 98%) in the transport sector to be reduced by using alternative fuels such as biofuels [1]. In addition, due to the increasing mobility of people and things, the transport sector accounts for more than 30% of final energy consumption in the European

Union (EU) and is expanding. Therefore, an increasing use of biofuels for transport is emerging as an important policy strategy to substitute petroleum-based fuels. However, the extent to which biofuel can displace fossil fuels depends on the efficiency with which it can be produced. In fact, all processing technologies, including biofuel options, involve (directly and/or indirectly) the use of fossil fuels in their production and/or operation. Therefore, in practice the actual benefits of biofuels displacing their fossil fuel equivalents depend crucially on biofuels' energy balances, which indicate the relative magnitude of fossil fuels input relative to subsequent fossil fuel savings resulting from their use as alternatives to conventional fossil fuels. To demonstrate that biofuel has a positive energy balance—i.e. more energy is contained in than is used in the production—a life cycle approach must be employed, allowing quantification of the renewability of biofuel delivered to consumers.

Based on "net energy analysis" studies, which were first published in the 1970s (e.g. [3,4]) and following the thrust for a more holistic approach to system analysis, there has recently been a substantial development of life-cycle methodologies to assess the energetic and environmental performance of product systems from "cradle-to-grave", namely life cycle energy analysis (LCEA) and environmental life cycle assessment (LCA). These methodologies have been receiving increasing attention, first by researchers and product manufacturers and more recently among policy-makers. Nonetheless, a methodological allocation problem arises in a life cycle study involving only one of several products from the same process: how are the resource consumption and emissions associated with this process partitioned and distributed over these co-products? This has been one of the most controversial issues in the development of the LCA methodology, as it may significantly influence or even determine the results of the assessment [5]. Biofuel technologies have diverse main products and co(by)-products. Thus, suitable allocation procedures need to be established and applied to partition the primary energy inputs between the different biofuel co-products [6]. However, this effect has often been underestimated in biofuel LCEA's and LCA's studies. In addition, it is important to recognize that there is no single allocation procedure that is appropriate for all biofuel processes [7]. According to the ISO 14041 [8], whenever several alternative allocation procedures seem applicable, a sensitivity analysis should be conducted.

This paper discusses bioethanol and bioETBE potential to displace the use of gasoline in spark ignition (SI) engines by reviewing important advantages and technical aspects. Production trends in Europe are analyzed with focus on the current European environmental policy context, which strongly promotes biofuels, but asks for studies addressing the life-cycle perspective of biofuels and demonstrating they are energy efficient and have the potential of becoming competitive and cost-efficient [1]. This has motivated the development of a life cycle approach to calculate the renewability and energy efficiency of biofuel energy systems, including a sensitivity analysis assessing the implications of allocation in the results. In practical terms, an important goal of this paper is to demonstrate the methodology by presenting an LCEA applied to bioethanol and bioETBE chains in France. The energy use throughout the alternative biofuel life cycles is analyzed and the overall energy efficiencies are calculated aiming at characterizing the renewability of French biofuels, which are able to compete with (and displace) petroleum-based fuels in the market place. Different allocation methods are explored in order to understand their effect on the calculation of overall energy efficiency and renewability of bioethanol chains (sugar beet and wheat) and its derivative bioETBE.

This paper is organized in 6 sections, including this introduction. Section 2 describes bioethanol and bioETBE technical aspects and analyzes current production trends in Europe, with particular emphasis on the French situation. Section 3 gives an overall view of the methodology, proposes a novel indicator—the *energy renewability efficiency*—and discusses the implications of multifunctionality. Section 4 demonstrates the implementation of the modeling approach and analyzes the inventory results obtained using different allocation approaches. Section 5 presents the main results and discusses the implications of allocation for the renewability and energy efficiency of bioethanol and bioETBE, produced from sugar beet and wheat. Section 6 concludes.

2. Bioethanol and bioETBE: technical issues and production in the European context

Bioethanol has been increasingly used in SI engines due to the following three main features: It was originally used as a gasoline extender, displacing gasoline derived from imported crude oil, in particular when oil prices boosted after the oil shocks of 1973 and 1979. Secondly, as a result of the phasing out of leaded fuel,

bioethanol became popular as a high-quality octane enhancer. Due to its better anti-knock characteristics, bioethanol provides a valuable additive to mid-to-low-octane gasoline, replacing benzene and other toxic chemicals often used by gasoline refiners as octane enhancers. Thirdly, owing to environmental concerns, bioethanol is used as an emission reducing oxygenate (oxygen-rich compound). In fact, adding bioethanol to gasoline increases the oxygen content of the fuel, improving the combustion of gasoline and reducing the exhaust emissions normally attributed to imperfect combustion in motor vehicles, such as carbon monoxide and unburned hydrocarbons [9,10].

However, blending bioethanol with gasoline raises fuel volatility, which increases the amount of ozonecausing evaporative hydrocarbon emissions. These emissions can be reduced by reformulating gasoline at the refining stage to have a lower vapor pressure, despite increasing refining costs. Another issue is that ethanol exhibits a 41% lower volumetric energy content (MJ/l) than gasoline (see Table A1, Appendix A). This and other operational problems that face the alcohol, namely storage and shipping challenges to avoid water contamination, can be overcome by converting bioethanol into its derivative bioETBE prior to blending with gasoline [11,12]. ETBE is produced in the petroleum refinery through the chemical reaction of ethanol with (fossil) isobutylene, in the presence of heat and a catalyst. BioETBE offers the same benefits as bioethanol e.g. reduced air pollution, increased fuel octane, reduced oil imports—without the technical and logistic difficulties shown by the alcohol [13,14]. BioETBE also contributes, although partially, to the share of renewable fuels in the transportation sector, as the percentage of bioETBE obtained from bioethanol amounts to 47% by mass [1,14].

It should be emphasized that ETBE is superior to MTBE (Methyl Tertiary Butyl Ether), another fuel oxygenate extensively used worldwide. MTBE is produced by reacting methanol and isobutylene. Like ETBE, MTBE enhances fuel's octane rating and improves air quality by reducing tailpipe emissions of carbon monoxide and ozone precursors [15,16]. However, due to its high solubility in water and high resistance to biodegradation, MTBE transfers readily to groundwater and causes contamination problems when fuel spills and leaks occur [16]. Thus, in spite of its relatively recent usage, MTBE has become one of the most frequently detected groundwater pollutants [16–18] and has been partially replaced by bioETBE in France since 1990 [19]. Another advantage of ETBE over MTBE is its lower Reid vapor pressure (RVP), allowing blenders to use a higher RVP base gasoline in final fuel formulation [20,21]. In addition it should be noted that currently nearly all methanol is produced from natural gas, although it could be produced from wood, coal, or any carbonaceous material that can be converted to synthesis gas (carbon monoxide and hydrogen) [22]. However, even if renewable sources were used to produce methanol, bioETBE would remain more renewable energy efficient, since the percentage of MTBE obtained from methanol only amounts to 36% as compared to the 47% of bioETBE from bioethanol [1].

Bioethanol can be used as a fuel for SI engines both in its pure form or blended with gasoline in several proportions. In Brazil, bioethanol is used as neat ethanol in 100% alcohol-fuelled passenger cars or is blended with gasoline in proportions of usually about 22% [23]. In several states of the USA, a small amount of bioethanol (10% by volume) is added to gasoline, known as gasohol or E10. Blends having higher concentrations of bioethanol in gasoline are also used, e.g. in flexible-fuel vehicles that can operate on blends of up to 85% bioethanol—E85 [24]. In Europe, Sweden uses bioethanol (i) blended directly with gasoline up to 5% by volume E5; (ii) in the form of E85 in modified light-duty vehicles and (iii) as a diesel replacement in trucks and buses, with ignition improvement additives. Unlike Sweden, in other European countries—e.g. France and Spain—bioethanol is mainly converted to bioETBE, which is used in SI engines in proportions of up to 15% by volume [14].

The main world regions responsible for the production of liquid biofuels are Brazil, the USA and Europe. Bioethanol is by far the most produced biofuel, with more than 18 million tonnes in 2003, essentially due to Brazil—9.9 million tonnes—and the USA—8.4 million tonnes. The EU biofuel production represented approximately 1.75 million tonnes in 2003 [25].

The European Commission has declared its intention to promote biofuels in different proposals and directives establishing minimum biofuel content in transportation fuels on the basis of an agreed schedule [1] and allowing Member States to apply a partial or total tax exemption for biofuels when used in their pure state or in mixtures [26]. In particular, the European Commission Green Paper [27] sets the objective of 20% substitution of conventional fuels by alternative fuels in the road transport sector by the year 2020.

Consequently, there has been a significant increase in biofuel production and use in Europe. For example, in 2004 an annual growth of 26% was observed [28]. However, only eight Member States make any real contribution to the total European biofuel production. Germany has the leading position and France is second, with a production of more than 400 thousand tonnes in 2004 representing 0.8% of the total national liquid fuel consumption [28,29].

Bioethanol production in France is based on two alternative chains (sugar beet or wheat), which can generally be described by the following three main stages [30,31]: (i) the agricultural sector; (ii) the biomass transformation industry; and (iii) the petroleum industry, responsible for the refining and mixing processes necessary to obtain the final combustible. In France, bioethanol has not been usually blended directly into gasoline. However, this is changing because direct incorporation of bioethanol in gasoline (up to 5% by volume, E5) has benefited from a tax reduction of $37 \in$ per hectoliter of ethanol since 2004 [25,29]. Currently, bioethanol is chemically combined with isobutylene to produce bioETBE, which is then mixed with gasoline for up to 15% by volume at the oil refinery. Eventually, the final fuel is delivered to the fuel energy transportation market replacing its fossil fuel equivalent.

3. Methodology: life cycle energy analysis

3.1. Life cycle inventory and energy renewability efficiency

LCEA is based on the standardized LCA methodology [32], limited to assess the energy aspects and, in this study, with a particular focus on energy efficiency indicators aiming at characterizing the renewability of bioethanol product systems. An LCA study offers a clear and comprehensive picture of the flows of energy and materials through a system and gives a holistic and objective basis for comparison. LCA is based on systems analysis, treating the product process chain as a sequence of sub-systems that exchange inputs and outputs. The results of an LCA quantify the potential environmental impacts of a product system over the life cycle, help to identify opportunities for improvement and indicate more sustainable options where a comparison is made. The LCA methodology consists of four major steps. The first component of an LCA is the definition of the *goal and scope* of the analysis. This includes the definition of a reference unit, to which all the inputs and outputs are related. This is called the *functional unit*, which provides a clear, full and definitive description of the product or service being investigated, enabling subsequent results to be interpreted correctly and compared with other results in a meaningful manner. In this study, in particular, the functional unit should enable the comparison of the energy used throughout the alternative biofuel product systems.

The second component of an LCA is the inventory analysis, also Life Cycle Inventory (LCI), which is based primarily on systems analysis treating the process chain as a sequence of sub-systems that exchange inputs and outputs. Hence, in LCI the product system (or product systems if there is more than one alternative) is defined, which includes setting the system boundaries (between economy and environment, and with other product systems), designing the flow diagrams with unit processes, collecting the data for each of these processes, performing allocation steps for multifunctional processes and completing the final calculations [33]. Its main result is an inventory table, in which the material and energy flows associated with the functional unit are compiled and quantified [32].

The "*end point*" defined for this life cycle study is the final (bio)fuel product, quantified by the energy content (MJ/kg). The various bioethanol LCI results are, subsequently, compared on the basis of 1 MJ of biofuel energy content. In LCA, the selection of the final product as an "end point" is often designated by the "cradle-to-gate" approach, instead of the more metaphoric "cradle-to-grave". The "gate" can be seen here as the fuel pumping station where (bio)fuel is delivered to vehicles. The choice of the "cradle-to-gate" approach is appropriate because it enables LCI results and biofuels' energy efficiencies to be analyzed in a variety of different ways, namely concerning allocation, enabling optimization or comparison with fossil fuels displaced. In fact, delivered energy as an end point avoids the complexities of adding further assumptions, in particular concerning vehicle performance factors, as it would be if, for example, "kilometers traveled" were adopted as the reference (end point). In some studies, kilogram (or liter) of biofuel have often been used as a reference, e.g. [34–37]. However, this is definitely not an adequate basis for comparison of the function provided by different (bio)fuels.

The functional unit chosen for this investigation is 1 MJ of fuel energy content, measured in terms of the lower heating value (LHV, heat of combustion excluding the latent heat in combustion products, i.e. the specific enthalpy of vaporization of water). This is consistent with the goal and scope of the study, which is to calculate the life cycle energy efficiency of bioethanol and bioETBE chains and compare these values with their fossil fuel equivalents, aiming at assessing the renewability of alternative biofuel product systems or, inversely, fossil fuel resource depletion.

Energy resource depletion must be quantified in terms of primary energy—energy embodied in natural resources (e.g. coal, crude oil, uranium or biomass) that has not undergone any anthropogenic conversion or transformation. As such, primary energy values are an indicator of energy resource availability and implicitly take into account the *energy quality*. Primary energy is the sum of the final energy with all the transformation losses, with fuel primary energy values being greater than their final energy values. In fact, consumers buy final energy, but what is really consumed is primary energy, which represents the cumulative energy content of all resources (fossil and non-fossil) extracted from the environment. In the case of fuels (or electricity), energy inputs required during the extraction, transportation and production processes measured in terms of primary energy ($E_{in, prim}$, MJ/kg), do not include the final fuel energy, i.e. the *fuel energy content* (FEC, MJ/kg). Even though, the energy requirement of fossil fuels should also include the FEC, in which case the result is referred to as the *gross energy requirement* (GER, MJ/kg) [7]

$$GER = E_{in,fossil,prim} + FEC.$$
 (1)

In (bio)energy analysis studies it is essential to distinguish between fossil ($E_{in,fossil,prim}$) and non-fossil ($E_{in,ren,prim}$) energy inputs, because we are concerned about the renewable nature of biofuels. Therefore, the essential comparison that needs to be made is between the fossil primary energy input to biofuel' life cycle ($E_{in,fossil,prim}$) and the fossil primary energy requirements throughout the life cycle of non-renewable fuels, including the fossil fuel energy content, i.e. the GER.

The life cycle inventory results provide an opportunity to quantify the total energy demand and, therefore, the overall energy efficiency. Quantifying the overall energy efficiency of a biofuel is helpful to determine how much (fossil) energy must be expended to convert the energy available in the raw materials (biological cultures) to 1 MJ of available energy in the transportation fuel. The more fossil energy required to make the biofuel, the less we can say that this biofuel is "renewable". Thus, the renewable nature of a fuel can vary across the spectrum of "completely renewable" (i.e. no fossil energy input) to non-renewable (i.e. fossil energy inputs as much or more than the energy output of the fuel) [38].

Within the energy analysis and LCA literature there is lack of consensus concerning the definition (and designation) of energy efficiency indicators to be used in a life-cycle perspective and, in particular, to characterize the energy requirements of renewable energy systems. In fact, various indicators have been used, often with the same meaning but different definition, or inversely, e.g. energy efficiency [39]; overall energy efficiency [4,40]; overall energy balance [41]; cumulative energy demand [42]; gross energy requirement and net energy requirement [43].

In particular, Sheehan et al. [38] have used the *life cycle energy efficiency* (LCEE), defined as the ratio between the biofuel energy content and the biofuel GER

$$LCEE = \frac{FEC}{(E_{in,fossil,prim} + FEC)}.$$
(2)

The LCEE can be seen as a measure of the fraction of the GER (primary energy required throughout the biofuel life cycle plus the biofuel energy content), which actually ends up in the fuel product. The same authors have also adopted the *fossil energy ratio* (FER), defined as

$$FER = \frac{FEC}{E_{in,fossil,prim}}.$$
(3)

According to this definition, if the fossil energy ratio is less than 1 the fuel is non-renewable, as more energy is required to make the fuel than the energy available in the final fuel product. Biofuel with FER greater than 1 can be considered as (partially) renewable. In theory, a total renewable fuel would have no fossil energy requirement and, thus, its fossil energy ratio would be infinite. Other authors have also used the FER

indicator, but under a different designation, for example "energy efficiency" [39], whereas others have used the "energy requirement" (E_{req}), defined as the "primary energy input per delivered energy output" [6,7,44,45]. This indicator is also used in Refs. [35,41], but under the designation of "net energy" and "overall energy balance", respectively. It should be noted that E_{req} is the inverse of FER.

The "*net energy value*" (NEV), defined as the biofuel FEC minus the fossil energy required to produce the biofuel (E_{prim})

$$NEV = FEC - E_{in, fossil, prim}$$
⁽⁴⁾

is proposed in Refs. [34,36]. In this case, negative *net energy values* indicate that (bio)fuel is non-renewable, while positive values indicate the fuel is renewable to a certain extent. In this study the energy requirement E_{req} is used to identify the relative contributions to the total primary energy input from different stages of the production chains and to evaluate the implications of the allocation method chosen for the energy efficiency of bioethanol.

In addition, a novel indicator is proposed—the *energy renewability efficiency*, aiming at characterizing the renewability of an energy source. The energy renewability efficiency (ERenEf)—to our knowledge, not previously proposed in the literature—measures the fraction of final fuel energy obtained from renewable sources. It can be defined as

$$\text{ERenEf} = \frac{(\text{FEC} - E_{\text{in,fossil,prim}})}{\text{FEC}}.$$
(5)

A biofuel may be considered renewable if ERenEf assumes values between 0% and 100%. In case there were no inputs of non-renewable energy, the biofuel would be completely renewable with an ERenEf of 100%. If the ERenEf is lower than zero, then the biofuel should be characterized as non-renewable since the nonrenewable energy required to grow and convert biomass into biofuel would be greater than the energy present in the biofuel final product. In this case, the biofuel is, indeed, not a fossil energy substitute and increasing its production does little to displace oil imports or increase the security of energy supply. By definition, nonrenewable energy sources have negative values of ERenEf, with increasing negative values as life cycle energy efficiency decreases. For example, gasoline (the fossil fuel displaced by bioethanol) shows an ERenEf value of -22.0%, meaning that the total primary energy required to produce gasoline is 22.0% greater than its final energy content [46].

3.2. Multifunctionality and allocation

Most industrial and agricultural processes are multifunctional. In particular, many of the feedstocks for biofuels are either co-produced with other products or are by(sub)-products of other production processes. Biofuel production systems generate large quantities of co(by)-products and, thus, LCA practitioners are faced with the problem that the product system under study provides more functions than that which is investigated in the functional unit of interest. This leads to the following central question: how should the resource consumption and energy used be distributed over the various co(by)-products? An appropriate procedure is thus required to partition the relevant inputs and outputs to the functional unit under study.

Options for dealing with co-production include: (i) sub-dividing the process into two or more sub-processes; (ii) expanding the product system to take into account potential effects of providing a new use for the coproducts on systems currently using the co-products—known as system boundary expansion—and (iii) allocating inputs and outputs between product streams based on causal relationships [8]. The international standard on LCA [8] states that allocation¹ should be avoided where possible by sub-division or system boundary expansion. Where this is not possible allocation should be undertaken using causal relationships, based on economic or physical properties of the co-products. However, Ekvall and Finnveden [47] have analyzed a large number of LCA studies where subdivision or system expansion was applied and found no case study where an allocation problem is completely eliminated through sub-division. In general terms,

¹The meaning of allocation in LCA is often used misleading. According to the ISO 14041, sub-division and system boundary expansion are not formally part of the allocation procedure.

3369

system expansion requires that there is an alternative way of generating the exported functions and that data can be obtained for this alternative production [47]. Many co-products are competing with other co-products, so expanding the system boundary would only result in an increasingly complex system [6,48]. In particular, many of the co-products of biofuel technologies have no separate main means of production. Hence, a simple substitute cannot be identified and, consequently, it is necessary to use an allocation procedure.

According to the ISO 14041 [8], allocation should reflect the physical relationships between the environmental burdens and the functions, i.e. how the burdens are changed by quantitative changes in the functions delivered by the product system. Thus, allocation can be based on physical properties of the products, such as mass, volume, energy, because data on the properties are generally available and easily interpreted. Where such physical causal relationships cannot be used as the basis for allocation, the allocation should reflect other relationships between the environmental burdens and the functions. In some cases, this allocation may coincide with allocation based on physical, causal relationships. The choice and justification of allocation procedures is a major issue for life cycle assessment [31], especially since they can have a significant influence on subsequent results [6,7,35,36,49,50].

In many biofuel studies mass of co-products has been chosen as a basis for allocation [39,51,52]. Other studies have used the energy content, e.g. [53,54]. However, the main reason for using mass seems to arise because both main and co-products can be weighed, and the use of energy content would only be relevant if both main and co-products were actually burned as fuels. Allocation can also be based on the exergy content [55,56]. Another method is based on the replacement value of co-products, in which energy credits can be assumed equal to the energy required to produce a substitute for the co-products [36,54]. Allocation based on the relative economic value (market price) of main and co-products has been used by Elsayed et al. [6], Spirinckx and Ceuterick [49] and Guinée et al. [57]. However, there are concerns about the effect of price variation on the calculation of allocation parameters. In most studies no discussion is provided regarding the selection of the allocation procedure and, in general, no complete justification can be found concerning the reason to choose one and not a different allocation procedure. In fact, it is important to recognize that there is no single allocation procedure which is appropriate for all biofuel processes [7]. Therefore, whenever several alternative allocation procedures seem applicable, a sensitivity analysis should be conducted [8]. Additionally, a sensitivity analysis to allocation rules is especially useful in reducing uncertainty due to choices in LCA [58]. Another important source of uncertainty in a LCA study is parameter uncertainty due to data inaccuracy, lack of data or unrepresentative data. In some cases, the uncertainty is inherent to data, namely for forecasted market data or data subjected to uncontrollable phenomena (e.g. climatic variation) [59]. Data uncertainty and variability of the most relevant parameters are analyzed and reported in Section 4.

A key aspect of performing an error analysis is to identify the most important sources of errors, in order to focus the efforts to areas where large improvements can be gained. According to Ref. [58], methodological choices (e.g. system boundaries, allocation methods) tend to have large influence, which may well override many other types of uncertainty. This type of uncertainty cannot be eliminated, but is rather easily illustrated by identifying the relevant alternatives and performing sensitivity analysis. Next section analyzes the energy use throughout the life cycle in order to assess the effect of the allocation approach chosen. The inventory results were calculated using four different allocation approaches based on: (i) output weight; (ii) energy content, (iii) economic value and (iv) replacement value of co-products, which are compared with results obtained ignoring co-product credits.

4. Life cycle modeling and inventory results

4.1. Goal, scope and main assumptions

This section presents the modeling of the LCI for bioethanol (from sugar beet or wheat) and bioETBE chains in France. Sector production data along with life cycle data from commercial databases have been combined to build the LCI model. Relevant assumptions are outlined and a systemic description of the bioethanol production schemes is implemented, including agricultural, transportation and industrial transformation stages. The results are summarized in terms of indicators of fossil fuel depletion, particularly analyzing biofuel energy requirement E_{req} values.

Energy analysis of biofuel systems frequently shows varying results due to different assumptions on critical variables that have a decisive impact on the energy balance, e.g. biomass yields and conversion technologies, fertilizer application rates, co-product evaluation, number of energy inputs included in the calculations [36,37]. Below, the main assumptions are outlined. A "cradle-to-gate" approach is adopted since the combustion of biofuels does not modify the energy balances, provided that no additional energy source is necessary for the combustion. To convert energy inputs into primary energy terms, the following efficiencies were considered: 91% for natural gas; 83% for oil; 94% for coal and 33% for electricity [52]. Primary energy inputs associated with fertilizers and energy content of diesel fuel used in farm and transportation activities are based on data from [52]. Agricultural production data has been collected for sugar beet [52] and wheat [60]. Agricultural inputs (fertilizers, agrochemicals and use of machinery) depend on the individual farm practices and agricultural yields are related to differences in natural conditions such as climate and soil conditions. Thus, a large source of variation $(\pm 50\%)$ can be expected for agricultural data [59,61]. The energy associated with transportation activities is estimated by using the following assumptions [51,52]:

- fertilizers: 200 km by rail and 100 km by road;
- biomass: 100 km by road, from farms to biofuel production plants;
- bioethanol: 200 km by rail, from distilleries to the refinery;
- bioETBE: 100 km by road, from the refinery to local distribution depots.

Road and rail transportation energy inputs were obtained using the French model [51]. Uncertainty associated with transport distances and energy intensity (MJ/ton km) is estimated as $\pm 30\%$. The energy embodied in the materials used to construct biofuel plants, transportation equipment and farm machinery ("capital energy") was not considered, since it becomes negligible when distributed over the throughput achieved in the lifetime of those equipments [36,40]. Data on industrial processes is site specific and the inherent uncertainty is typically low (<10%). (Bio)fuels' properties used in this paper are reported in Table A1 (Appendix A).

Parameters used for allocation are presented in Table 1. Energy content and replacement credits of coproducts were estimated using data from [41,62]. However, because pulps from sugar beet may have quite distinct alternative uses [62], the adoption of the replacement method gives considerably different energy credits. Thus, an average credit for pulps was calculated based on the alternative potential uses. Economic parameters were obtained from average market prices of products and co-products [63]. Market prices present high variability, since they are largely influenced by external factors such us oil price, dollar value and feedstock market fluctuations. From Table 1, it is apparent that there is no linear relation between allocation column data.

4.2. Bioethanol and bioETBE life cycles

Figs. 1 and 2 illustrate two alternative ways of producing bioethanol, namely from sugar beet and wheat, respectively. The flow charts simplify the actual chain of processes for the sake of clarity and the arrows show

Allocation	Mass (kg/kg bioethanol)			Energy (LHV)	Economic (Market	Replacement credits of co-	
procedure	S. beet #A	S. beet #B	Wheat	- (WJ/Kg)	value) (e/ton)	products (wij/kg co-product)	
Bioethanol	1	1	1	26.8	115	_	
Sugar	_	7.92	_	15.5	147		
Pulps	0.60	3.06	_	15.6	37	1.4	
DDGS	—	—	1.35	15.0	107	2.1	

Table 1 Data used for allocation

the direction of the logistics flow. The agricultural production of sugar beet and wheat cultivation includes several steps, namely: soil preparation, plowing, weeding, fertilization, sowing and harvesting.

The production of ethanol from sugar beet (Fig. 1) comprises two steps: (i) green juice and green syrup (GS) are produced at the sugarhouse, by subjecting biomass to a sequence of processes, namely washing and diffusion to obtain green juice and afterward purification, evaporation and crystallization to obtain GS from green juice, with sugar as a co-product; (ii) ethanol is produced both from green juice and GS at the distillery through the following processes: fermentation using yeast, followed by distillation to increase ethanol concentration and, finally, dehydration to obtain anhydrous ethanol. Sugar beet pulp is the most important by-product of the sugar beet conversion process and can be used in the following alternative ways: (i) added to an anaerobic digester, producing biogas; (ii) dried and sold for animal feed instead of equivalent products from cereals fermentation (low-protein animal feed); (iii) dried and burnt for process heat or (iv) converted into more ethanol by simultaneous saccharification and fermentation [62]. The purification step also produces foams that are used as organic fertilizer. Vinasses, another co-product from ethanol distillation of green syrup, are concentrated and spreaded on agricultural land. Details concerning the technological description and the mass and energy balances of these steps can be found in great detail in Ref. [52]. The technological processes involved to obtain ethanol from sugar beet are not self-dedicated to the production of ethanol. Instead, the whole chain is shared by the alcohol and sugar industries. According to current industrial and commercial practices in France, 50% of the bioethanol produced comes from green juice and the other half comes from GS [52]. With this share, 3.96 kg of sugar are obtained for each kg of bioethanol produced (Table 2). The commercial feasibility of producing ethanol from sugar beet involves a comparison of alternative revenue streams from sugar beet with ethanol or sugar product forms, i.e. the trade-off between relative sugar and alcohol returns is a key factor driving the allocation of sugar beet between sugar and bioethanol [64]. In contrast to *joint production*, in which the relative output volume of the co-products is fixed [56], the share of sugar and ethanol extracted from sugar beet is independently variable (combined production, illustrated in



Fig. 1. Flow chart illustrating the bioethanol production chain from sugar beet.



Fig. 2. Flow chart illustrating the bioethanol production chain from wheat.

	Ethanol (sugar b	eet)		Ethanol (wheat)
	#A	50%A/50%B	#B	
Agricultural production				
Land (ha)	-0.172	-0.521	-0.870	-0.469
N fertilizer (kg)	-21.1	-63.8	-106.5	-99.2
P_2O_5 fert. (kg)	-12.0	-36.5	-60.9	-15.9
K ₂ O fert. (kg)	-30.1	-91.2	-152.3	-15.7
Diesel (1)	-27.7	-83.9	-140.1	-53.9
Leaves (t)	0.75	2.27	3.79	_
Straw (t)	_		—	3.05
Biomass (t)	11.58	35.06	58.55	3.58
1st industrial conversion stage				
Natural Gas (MJ)	-6127.8	-17933.6	-29378.2	-18702.7
Oil (MJ)	-4860.2	-13377.5	-21604.4	_
Coal (MJ)	-4009.6	-9429.7	-14609.7	_
Electricity (kWh)	_	-653.3	-1306.7	-1602.8
Sugar (t)	—	3.96	7.92	_
Pulps (t)	0.60	1.83	3.06	_
DDGS (t)	—	_	—	1.35
Bioethanol (t)	1	1	1	1

 Table 2

 Agricultural and industrial data for the production of 1 tonne of bioethanol

#A: Ethanol production from sugar beet; #B: Sugar production, with sub-production of ethanol; t = tonne; l = liter.

Fig. 1 by dashed lines), which offers the sugar beet transformation industry the opportunity to broaden its revenue base and to assure continued financial viability by pursuing the ethanol and/or sugar options. For combined production, allocation can be avoided simply by modeling directly the consequences of a change in the output of the co-product of interest (that which is used in the product system under study) [5]. This approach is used in the LCI of bioethanol produced from green juice (route a in Fig. 1).

The production route of ethanol from wheat (Fig. 2) includes a sequence of mechanical and chemical processes, which can be divided in two main stages. Firstly, feedstock processing, including grinding of grains, liquefaction and saccharification, where enzymes are introduced to break down the starch in the wheat into fermentable sugar. Secondly, fermentation of sugar juice using yeast to produce ethanol at 10–15% concentration, distillation of this solution to recover the ethanol at higher concentrations (95%) and dehydration to obtain anhydrous ethanol used as fuel. The leftover residue from the fermentation process (Distiller's Dried Grains with Solubles (DDGS)) is the wheat equivalent of pulps from sugar beet but with higher protein content and can be sold as high-protein animal feed.

Bioethanol can be used as motor fuel in pure form or blended with gasoline. It can also be used to produce bioETBE. The production of bioETBE (Fig. 3) takes place at the petroleum refinery by reacting bioethanol with isobutylene, 47% and 53% by mass, respectively. Like bioethanol, bioETBE is an oxygenate, used as an additive to gasoline, resulting in a cleaner burning fuel as it increases the level of oxygen available during combustion, reducing, thus, the emissions of carbon monoxide and improving air quality [13]. In addition, bioETBE overcomes some of the technical and logistic drawbacks of bioethanol, as detailed in Section 2.

Allocation was avoided by extension of system limits to include additional functions related to the following co-products: leaves from sugar beet cultivation; foams from purification of green juice in the sugarhouse and vinasses from distillation of bioethanol, after fermentation of green juice and green syrup in the distillery. Sub-division was used by splitting ethanol production chain (from sugar beet) into case #A (route a, Fig. 1), where only ethanol is produced, and case #B (route b, Fig. 1), where sugar is the major product; however, allocation has to be performed for case #B, where ethanol is jointly produced with sugar.



Fig. 3. Flow chart illustrating bioethanol use: (i) as a single fuel, (ii) blended with gasoline and (iii) blended with gasoline, after conversion in its derivative bioETBE.

Table 3 BioETBE production data

	BioETBE production	
2nd industrial conversion stage	Bioethanol (t)	-0.47
	Isobutylene (t)	-0.53
	Electricity (kWh)	-14
	Natural Gas (MJ)	-127
	Oil (MJ) ^a	-27550
	Coal (MJ)	-162
	BioETBE (t)	1

^aOil energy includes isobutylene feedstock energy.

Allocation had to be applied in eight individual processes: (i) sugar beet cultivation, green juice production and purification, evaporation and crystallization of green juice and (ii) wheat cultivation, milling and malting. The substitution approach, in which each by-product generates energy credits equals to those associated with producing a substitute for that co-product, is used assuming that: (i) sugar beet pulps can be used in four different ways as described in this section, resulting in an average energy credit and (ii) DDGS left after fermentation and distillation in the wheat-to-ethanol process replaces soy bean meal as a high-protein animal feed. The equivalent quantity of soy bean meal is calculated on the basis of the protein content [62].

The use of resources (energy and materials) and the output of co-products for the production of 1 ton of bioethanol are listed in Table 2. The production process is separated into two stages, the production of the agricultural energy feedstock and the conversion of the feedstock into bioethanol. Three distinct situations are shown for ethanol production based on sugar beet: (i) case #A; (ii) case #B and (iii) case "50%A/50%B", representing the current commercial practice in France. Production data for bioETBE is illustrated in Table 3 [52]. Input and output coefficients are given negative and positive signs, respectively.

4.3. Allocation and implications for inventory results

Table 4 reports the mass allocated "cradle-to-gate" fossil energy use (1 MJ of bioethanol as functional unit) and identifies the relevant contributions from the different stages. Bioethanol production is by far the most energy intensive stage. Agriculture activities are less important in terms of primary energy requirement and the transportation stages hardly contribute to the energy balances. Thus, uncertainties associated with these processes become of minor importance. Fossil energy input during biomass production mainly results from the energy content of fertilizers and from the use of agricultural machinery. Case #B of sugar beet based ethanol exhibits the worst results due to the higher energy requirements associated with processing of green syrup as compared to green juice processing.

Table 4	
Bioethanol "cradle-to-gate" primary energy requirement (E_{req}) using mass allocation	

Stage	Ethanol (sugar	Ethanol (wheat)			
	#A	50%A, 50%B E _{req} (MJ/MJ)	#B		
Biomass production	0.05664	0.04043	0.03830	0.12296	
Biomass transport	0.00864	0.00617	0.00584	0.00147	
Bioethanol production	0.56070	0.73279	0.92923	0.38915	
Bioethanol transport	0.00348	0.00348	0.00348	0.00348	
TOTAL	0.630	0.783	0.977	0.517	

Table 5 BioETBE "cradle-to-gate" primary energy requirement (E_{req}) using mass allocation

	BioETBE production							
	Sugar beet (#A) 50%A, 50%B Sugar beet (#B)							
	Ereq (MJ fossil/MJ BioETBE)							
Process energy	0.0839							
Isobutylene	0.6873							
Bioethanol	0.2194	0.2728	0.3405	0.1805				
TOTAL	0.991	1.044	1.112	0.952				

Table 6 Allocation approach: implications for bioethanol primary energy requirement (E_{req})

Allocation procedure	Ethanol (sugar beet)							Ethanol (wheat)	
	#A		50%A/50%B		#B				
				$E_{\rm req}~({\rm MJ}/{\rm MJ})$					
Without co-product credits	0.702	(100%)	1.860	(100%)	3.041	(100%)	1.210	(100%)	
Mass	0.630	(89.7%)	0.783	(42.1%)	0.977	(32.1%)	0.517	(42.7%)	
Energy	0.652	(92.9%)	0.875	(47.0%)	1.116	(36.7%)	0.691	(57.1%)	
Market Value	0.671	(95.6%)	0.770	(41.4%)	0.947	(31.1%)	0.540	(44.6%)	
Replacement	0.671	(95.6%)	0.721	(38.8%)	0.848	(27.9%)	1.104	(91.2%)	

Table 5 summarizes energy requirements associated with the conversion of bioethanol into bioETBE. Similar to Table 4, case #B exhibits the highest energy input. The energy associated with isobutylene has the greatest impact on the life-cycle energy of bioETBE (approximately 70%). Refinery processes and, in particular, isobutylene production are well-known processes with relatively low uncertainty.

Implications for biofuel life-cycle primary energy requirements are presented in Tables 6 and 7 for each of the four allocation procedures used. For comparative purposes, results obtained ignoring co-product credits are also presented. A minimum energy requirement of 0.517 MJ/MJ was obtained for wheat based ethanol, using mass allocation. The percentage of energy use assigned to bioethanol is shown inside brackets—42.7% of the total energy requirements are assigned to bioethanol if mass allocation is used. For case #A of sugar

J. Malça, F. Freire / Energy 31 (2006) 3362-3380

Allocation procedure	BioETBE (sugar beet)							BioETBE (wheat)	
	#A		50%A/50%B		#B				
				$E_{\rm req}~({\rm MJ/MJ})$					
Without co-product credits	1.016	(100%)	1.420	(100%)	1.831	(100%)	1.194	(100%)	
Mass	0.991	(97.5%)	1.044	(73.5%)	1.112	(60.7%)	0.952	(79.7%)	
Energy	0.999	(98.3%)	1.076	(75.8%)	1.160	(63.3%)	1.012	(84.8%)	
Market Value	1.005	(98.9%)	1.040	(73.2%)	1.101	(60.1%)	0.960	(80.4%)	
Replacement	1.005	(98.9%)	1.023	(72.0%)	1.067	(58.3%)	1.157	(96.9%)	

Table 7 Allocation approach: implications for BioETBE primary energy requirement (E_{req})

beet based ethanol at least 89.7% of the energy used to produce the biofuel is assigned to bioethanol, since the contribution of pulps (the other co-product) is not relevant. The same does not apply for case #B, where sugar is the major output and, thus, allocation results in about a 30–40% energy assigned to bioethanol. Cases #A and #B, as well as case "50%#A/50%#B", are not as sensitive to allocation as ethanol from wheat, in which variations in E_{req} due to allocation are significant. Table 6 also shows that energy replacement values for wheat based ethanol result in less energy credits than the other methods, i.e. it is the least favorable allocation approach for bioethanol from an energy use standpoint. Similar conclusions can be drawn from Table 7.

5. Results and discussion

Fig. 4 illustrates the energy renewability efficiency of bioethanol compared with gasoline. ERenEf results for each alternative bioethanol chain are presented for the five procedures adopted. The impact of parameter uncertainty has also been estimated and found to be considerably less important than the effect of allocation, since high uncertainty of agricultural and transportation data is associated with less important processes. As shown before (Table 6), ethanol from wheat is much more sensitive to the allocation procedure chosen than sugar beet based ethanol, with ERenEf values ranging from -10% to 48%.

Ethanol produced from wheat exhibits the maximum energy renewability efficiency. In particular, a maximum ERenEf value of 48% is achieved using mass allocation, meaning that approximately 50% of the bioethanol energy content is indeed renewable energy. However, when replacement is used as the allocation procedure, wheat based ethanol shows a negative ERenEf value, which is due to low energy credits from co-products substitution.

Ethanol from sugar beet (case #A) is clearly renewable, even before adding co-product energy credits, with ERenEf values between 30% and 37% regardless of the allocation approach chosen. A maximum ERenEf of 37% is achieved using mass allocation. However, as detailed in Section 4.2, it should be noted that sugar beet based ethanol is jointly produced with sugar and higher shares of sugar production imply lower bioethanol ERenEf due to high energy inputs associated with processing of sugar. Thus, case #B (where sugar is the major product) presents considerably low ERenEf (between -12% and 15%). Furthermore, since case #B and case "50%#A,50%#B" are mainly dedicated to sugar production, ERenEf values without co-product credits are meaningless (omitted from Fig. 4).

All chains exhibit higher ERenEf values than gasoline (ERenEf = -22%), which clearly indicates that considerable reductions in fossil fuel depletion would be achieved by replacing gasoline with bioethanol. In particular, the higher energy savings are achieved for wheat based ethanol.

Fig. 5 presents the energy renewability efficiency of bioETBE, which shows lower ERenEf values than bioethanol due to high energy inputs associated with the synthesis process of isobutylene for ETBE production. Analogously to Fig. 4, wheat based production is highly sensitive to allocation, with ERenEf



Fig. 4. Energy renewability efficiency (ERenEf) values: bioethanol and gasoline (Sb#A or Sb#B-sugar beet, case #A or case #B).



Fig. 5. Energy renewability efficiency (ERenEf) values: bioETBE and gasoline (Sb#A or Sb#B-sugar beet, case #A or case #B).

values between -19% and 5%, while sugar beet (case #A) based production is nearly independent of the allocation procedure used (ERenEf varies less than 3%).

Finally, Table 8 compares energy requirement E_{req} and fossil energy savings when gasoline is displaced by E5 and ETBE(15)—blends typically used in the European context—together with results for pure bioethanol and bioETBE. It should be noticed that although E5 and ETBE(15) require more primary energy than their FEC, they represent actual fossil energy savings when compared to gasoline. Pure bioethanol displacing 1 MJ of gasoline may save up to 0.70 MJ, depending on whether wheat or sugar beet is used and on the allocation procedure adopted.

Based on the values presented in Table 8, the impact of producing bioethanol in France can be estimated in terms of total amount of fossil fuel saved, to which corresponds a reduction in CO₂ emissions. In France (see discussion in Section 2), almost the entire production of bioethanol has been converted to bioETBE before displacing gasoline in the transportation market. For example, in 2004 about 170 600 ton of bioETBE [28] were produced. Assuming that bioETBE saves on average (from sugar beet and wheat, Table 8) 0.25 MJ of fossil fuel energy per MJ of bioETBE replacing gasoline (up to a maximum of 15% by volume), the total amount of fossil fuel saved can be estimated as 1.54×10^9 MJ (1.706×10^8 kg $\times 36.1$ MJ/kg $\times 0.25$ MJ/MJ). Thus, from our calculations about 36775 ton of oil equivalent (TOE) were saved for France in 2004 (1 TOE = 41.868 GJ).

	Bioethand	ol	BioETBE		E5 ^a		ETBE(15) ^b		Gasoline
	S.beet	Wh	S.beet	Wh	S.beet	Wh	S.beet	Wh	
<i>E</i> _{req} (MJ/MJ) FES ^c (MJ/MJ)	0.63 0.59	0.52 0.70	0.99 0.23	0.95 0.27	1.20 0.02	1.20 0.02	1.19 0.03	1.19 0.03	1.22

Table 8 Primary energy requirement (E_{req}) and fossil energy savings (FES) (mass allocation)

S.beet: Sugar beet; Wh: Wheat.

^aGasoline blended with 5% bioethanol by volume.

^bGasoline blended with 15% bioETBE by volume, which corresponds indirectly to 6.6% bioethanol by volume.

^cFossil energy savings are calculated as FES(i) = E_{req} (gasoline)- E_{req} (i).

6. Conclusions

This paper has demonstrated how a life cycle energy approach can be used to assess the renewability and energy efficiency of biofuels. A novel indicator aiming at characterizing the renewability of (bio)energy sources is proposed—the *energy renewability efficiency* (ERenEf)—which measures the fraction of final fuel energy obtained from renewable sources. ERenEf values were calculated for bioethanol (from sugar beet or wheat) and its derivative bioETBE and were compared to gasoline. Implications for fossil fuel depletion associated with the various stages of biofuel production have been addressed by estimating primary energy inputs and calculating the accumulated energy requirement. LCI results together with E_{req} and ERenEf values are presented using four different allocation approaches (and ignoring co-product credits) in order to understand the effect of allocation in the overall energy efficiency and renewability of biofuel production.

Results demonstrate that the LCEA of wheat based ethanol is highly sensitive to the allocation method used. In fact, ERenEf values for bioethanol (wheat) can vary more than 50%, ranging between -10% (replacement method) and 48% (mass allocation), with 31% for energy allocation and 46% for allocation based on market values. However, the energy renewability efficiency of ethanol from sugar beet (case #A) varies only between 33% and 37%, being less sensitive to allocation because in this case co-production is not much relevant. ERenEf values calculated ignoring co-product credits can be very low, emphasizing the importance of performing allocation. Nevertheless, this study demonstrates that bioethanol produced in France, whether from sugar beet or wheat, is clearly favorable in primary energy terms. BioETBE shows lower ERenEf values than bioethanol, requiring on average as much primary energy as its FEC. E5 and ETBE(15) require more primary energy than their FEC due to the major share of fossil energy in these blends.

Regardless of the kind of biofuel (pure or blended) assessed in this research, actual fossil energy savings are achieved in comparison to gasoline, even though to different extents depending on the fraction of bioethanol in the final commercial blend. Energy savings per MJ of FEC range from 0.02 MJ (E5) to 0.70 MJ (bioethanol). It can be concluded that the use of bioethanol and bioETBE as liquid transportation fuels (pure or blended) reduces fossil fuels depletion and increases the security of energy supply. This conclusion is in line with Directive 2003/30/EC recommendations to increase the share of biofuels in the transportation sector [1] and with Directive 2003/96/EC that allows EU Member States to apply an exemption or reduced rate of excise duty to all biofuels sold as from 2004 [26].

Furthermore, it must be emphasized that optimum use of co-products, such as protein-rich residues for animal feed (e.g. pulps and DDGS) and wheat straw as an energy source, is needed to improve the energy efficiency of bioethanol production. In some cases, however, overestimating the benefits of these co-products is speculative, since the practicality and economic viability of their massive use is still subjected to uncertainty. Although the inventory analysis presented in this paper is focused only on bioethanol and bioETBE, the methodology presented and, in particular, the assessment of the implications of the allocation approach used might be applied to other biofuels. In addition, the inventory analysis performed in this research can be used as a starting point for a complete LCI, since it lays out a formal methodology for conducting an LCA for biofuels. In a complete LCA, a detailed list of environmental impacts should be adopted with the aim of addressing the sustainability of bioethanol chains, which is beyond the scope of this paper.

\cdot , , , , , , , , , , , , , , , , , , ,										
	Ethanol	ETBE	E5 ^a	ETBE(15) ^b	Gasoline					
Specific volume (l/kg)	1.259	1.342	1.329	1.335	1.333					
LHV (MJ/kg)	26.8	36.1	41.7	41.6	42.5					

Table A1 Properties of fuels (pure and blended)

^aGasoline blended with 5% bioethanol by volume.

^bGasoline blended with 15% ETBE by volume.

Acknowledgements

We are grateful for critical inputs from Dr. Stelios Rozakis, Institut National de la Recherche Agronomique (INRA). The first author (J. Malça) is also grateful for financial support from INRA, France, during his research visit to INRA, Thyverval-Grignon in 2002. The authors would like to thank the editor and two anonymous reviewers for important suggestions during the revision of the paper. The assistance of Dr. Deolinda Simões in reviewing the written English of the final manuscript is gratefully acknowledged.

Appendix A

(See Table A1).

References

- EPC (European Parliament and Council). Directive on the promotion of the use of biofuels or other renewable fuels for transport, 2003/30/EC, 2003. http://europa.eu.int
- [2] EC (European Commission). European transport policy for 2010: time to decide. White Paper, 2001. http://europa.eu.int
- [3] Boustead I. The milk bottle. Milton Keynes: Open University Press; 1972.
- [4] Boustead I, Hancock G. Handbook of industrial energy analysis. New York: Ellis Horwood Ltd., Wiley; 1979.
- [5] Weidema B. Avoiding co-product allocation in life-cycle assessment. J Ind Ecol 2000;4:11–33.
- [6] Elsayed M, Matthews R, Mortimer N. Carbon and energy balances for a range of biofuels options. Final Report, Resources Research Unit, Sheffield Hallam University, UK, 2003.
- [7] Mortimer N, Cormack P, Elsayed M, Horne R. Evaluation of the comparative energy, global warming and social costs and benefits of biodiesel. Report, Resource Research Unit, Sheffield Hallam University, UK, 2003.
- [8] ISO 14041. Environmental management—LCA—Goal and scope definition and inventory analysis. ISO, 1998.
- [9] Al-Hasan M. Effect of ethanol-unleaded gasoline blends on engine performance and exhaust emission. Energy Convers Manage 2003;44:1547–61.
- [10] Hsieh WD, Chen RH, Wu TL, Lin TH. Engine performance and pollutant emission of an SI engine using ethanol-gasoline blended fuels. Atmos Environ 2002;36:403–10.
- [11] Bensaïd B. Engines/motor fuels: possible long-term trends. IFP (Institut Français du Pétrole) Panorama 2004 Conference—Energy, Oil, Natural Gas, the Automobile and the Environment, Paris, January 2004.
- [12] His S. Biofuels in Europe. IFP (Institut Français du Pétrole) Panorama 2004 Conference—Energy, Oil, Natural Gas, the Automobile and the Environment, Paris, January 2004.
- [13] Vinuesa JF, Mirabel Ph, Ponche JL. Air quality effects of using reformulated and oxygenated gasoline fuel blends: application to the Strasbourg area (F). Atmos Environ 2003;37:1757–74.
- [14] EBN (European Bioenergy Networks). Liquid Biofuels Network-Activity Report, France, 2003.
- [15] Toran L, Lipka C, Baehr A, Reilly T, Baker R. Seasonal and daily variations in concentrations of methyl tertiary-butyl ether (MTBE) at Cranberry Lake, New Jersey. Water Res 2003;37:3756–66.
- [16] Kharoune M, Pauss A, Lebeault J. Aerobic biodegradation of an oxygenates mixture: ETBE, MTBE and TAME in an upflow fixedbed reactor. Water Res 2001;35(7):1665–74.
- [17] Schmidt T, Schirmer M, Weib H, Haderlein S. Microbial degradation of methyl tert-butyl ether and tert-butyl alcohol in the subsurface. J Contaminant Hydrol 2004;70:173–203.
- [18] Klinger J, Stieler C, Sacher F, Brauch HJ. MTBE (methyl tertiary-butyl ether) in groundwaters: monitoring results from Germany. J Environ Monit 2002;4(2):276–9.

- [19] Conseil B, Fombarlet C, Chatin L. ETBE: industrial experience of Elf Antar France (L'ETBE: l'expérience industrielle d'Elf Antar France). Pétrole Tech 1995;393:55–8 (in French).
- [20] Schmidt T. Analysis of methyl tert-butyl ether (MTBE) and tert-butyl alcohol (TBA) in ground and surface water. Trends Anal Chem 2003;22(10):776–84.
- [21] Ahmed I. Greenhouse gas emissions from ethanol and MTBE—a comparison. Final Report, Institute for Local Self-Reliance, Washington DC, 1994.
- [22] MacLean H, Lave L. Evaluating automobile fuel/propulsion system technologies. Prog Energy Combust Sci 2003;29:1-69.
- [23] Calle F, Cortez L. Towards ProAlcool II-a review of the Brazilian bioethanol programme. Biomass Bioenergy 1998;14(2):115-24.
- [24] Puppán D. Environmental evaluation of biofuels. Periodica Polyt Ser Soc Man Sci 2002;10(1):95–116.
- [25] EurObserv'ER. The EurObserv'ER biofuels barometer survey, June 2004.
- [26] EPC (European Parliament and Council). Directive restructuring the Community framework for the taxation of energy products and electricity, 2003/96/EC, 2003. http://europa.eu.int
- [27] EC (European Commission). Towards a European Strategy for the Security of Energy Supply. Green Paper, COM(2000) 769 final, 2000. http://europa.eu.int
- [28] EurObserv'ER. The EurObserv'ER biofuels barometer survey, June 2005.
- [29] Direction Générale de l'Énergie et des Matières Premières. The oil industry in 2004 (L'industrie pétrolière en 2004). Paris, 2005 (in French).
- [30] Freire F, Malça J, Rozakis S. Integrated economic and environmental life cycle optimization: an application to biofuel production in France. In: Henggeler Antunes C, Figueira J, Clímaco J, editors. Multiple criteria decision aiding. 2004. p. 247–272 (based on the papers presented at the 56th Meeting of the European Working Group Decision Multiple Criteria Decision Aiding, Coimbra, Portugal, 3–5/10/2002).
- [31] Freire F, Malça J, Rozakis S. Biofuel production systems in France: integrated economic and environmental life cycle optimization. In: Proceedings of the II International Conference on Mechanical Engineering, COMEC 2002, Santa Clara, Cuba, 2002.
- [32] ISO 14040. Environmental management—life cycle assessment—principles and framework. ISO; 1997.
- [33] Guinée J, Gorrée M, Heijungs R, Huppes G, Kleijn R, de Koning A, et al. Handbook on life cycle assessment. Operational guide to the ISO standards. Berlin: Springer; 2002.
- [34] Shapouri H, Duffield J, Graboski M. Estimating the net energy balance of corn ethanol. Report no. 721. US Department of Agriculture, 1995.
- [35] Kim S, Dale B. Allocation procedure in ethanol production system from corn grain. I-System expansion. Int J LCA 2002;7(4):237-43.
- [36] Shapouri H, Duffield J, Wang M. The net energy balance of corn ethanol: an update. Report no. 813. US Department of Agriculture, 2002.
- [37] Henke J, Klepper G, Schmitz N. Tax exemption for biofuels in Germany: is bio-ethanol really an option for climate policy? 2003 International Energy Workshop, Laxenburg, Austria, 2003.
- [38] Sheehan J, Camobreco V, Duffield J, Graboski M, Shapouri H. Life cycle inventory of biodiesel and petroleum diesel for use in an urban bus. Final Report, National Renewable Energy Laboratory, Golden, CO, 1998.
- [39] Ecobilan. Energy and greenhouse gas balances of biofuels' production chain in France. Report, ADEME, France, 2002.
- [40] Boustead I. Eco-Profiles of the European plastics industry. Methodology, Report. Association of Plastics Manufacturers in Europe, Brussels, 2003.
- [41] Armstrong A, Baro J, Dartoy J, Groves A, Nikkonen J, Rickeard D, et al. Energy and greenhouse gas balance of biofuels for Europe—an update. Report no. 2/02. CONCAWE, Brussels, 2002.
- [42] Wagner H, Pick E. Energy yield ratio and cumulative energy demand for wind energy converters. Energy 2004;29(12–15):2289–95.
- [43] Wilting H. An energy perspective on economic activities. PhD thesis, Groningen, 1996.
- [44] Elsayed M, Mortimer N. Carbon and energy modelling of biomass systems: conversion plant and data updates. Final Report, Sheffield Hallam University, UK, August 2001.
- [45] Malça J, Rozakis S, Freire F. Life cycle energy analysis of biofuels: an application to bioethanol production in France (Análise Energética do Ciclo de Vida de Biocombustíveis: Aplicação à Fileira do Bioetanol em França). 3rd Polytechnic Engineering Meeting, Coimbra, November, 2003 (in Portuguese).
- [46] Hekkert M, Hendriks F, Faaij A, Neelis M. Natural gas as an alternative to crude oil in automotive fuel chains well-to-wheel analysis and transition strategy development. Energy Policy 2005;33:579–94.
- [47] Ekvall T, Finnveden G. Allocation in ISO 14041-a critical review. J Cleaner Prod 2001;9:191-5.
- [48] Beer T, Grant T, Morgan G, Lapszewicz J, Anyon P, Edwards J, et al. Comparison of transport fuels. Final Report (EV45A/2/F3C) to the Australian Greenhouse Office. Australia: CSIRO Division of Atmospheric Research, 2001.
- [49] Spirinckx C, Ceuterick D. Comparative life-cycle assessment of diesel and biodiesel. Int J LCA 1996(3):127-32.
- [50] Wang M, Hanjie L, Molburg J. Allocation of energy use in petroleum refineries to petroleum products: implications for life-cycle energy use and emission inventory of petroleum transportation fuels. Int J LCA 2004;9(1):34–44.
- [51] Epelly O. Ecobilan du diester & Elements d'analyse de la filière gazole. Rapport Final Onidol, Tome II. Janvier, 1993 (in French).
- [52] Poitrat E, Leviel B, Vergé C, Gosse G. Total costs and benefits of biomass in selected regions of the European Union (Biocosts). Report to the European Commission on the Case Study "ETBE from sugar beet under French conditions", 1998.
- [53] Culshaw F, Butler C. A review of the potential of biodiesel as a transport fuel. Report R71, Energy Technology Support Unit, Harwell, UK, 1992.
- [54] Gover M, Collins S, Hitchcook G, Moon D, Wilkins G. Alternative road transport fuels—a preliminary life cycle study for the UK. Report R92, Energy Technology Support Unit, Harwell, UK, 1996.

- [55] Dewulf J, Van Langenhove H, Mulder J, van den Berg M, van der Kooi H, de Swaan Arons J. Illustrations towards quantifying the sustainability of technology. Green Chem 2000;2:108–14.
- [56] Frischknecht R. Allocation in life cycle inventory analysis for joint production. Int J LCA 2000;5(2):85-95.
- [57] Guinée J, Heijungs R, Huppes G. Economic allocation: examples and derived decision tree. Int J LCA 2004;9(1):23-33.
- [58] Björklund A. Survey of approaches to improve reliability in LCA. Int J LCA 2002;7(2):64-72.
- [59] Weidema B, Fress N, Petersen E, Ølgaard H. Reducing uncertainty in LCI: developing a data collection strategy. Environmental Project No. 862, Denmark, 2003.
- [60] Herbert V. Analyse Technico-Économique de la Production d'Éthanol Carburant de Blé, Mémoires et Thèses no 13. INRA, France, 1995 (in French).
- [61] Ferret R, Mendoza G, Castilla M. The influence of agricultural data uncertainty in the comparative life cycle assessment of biodegradable hydraulic lubricants. The International Environmental Modelling and Software Society Conference, Complexity and Integrated Resources Management, University of Osnabrück, Germany, June 2004.
- [62] Edwards R, Griesemann JC, Larivé JF, Mahieu V. Well-to-wheels analysis of future automotive fuels and powertrains in the European context. Report v. 1b. January 2004.
- [63] Rozakis S, Sourie JC, Vanderpooten D. Integrated micro-economic modelling and multicriteria methodology to support public decision-making: the case of liquid bio-fuels in France. Biomass Bioenergy 2001;20:385–98.
- [64] Jolly L. Agriculture: a source of green energy. Symposium Agriculture, Mauritius, October 2003.