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Solving the multifunctionality dilemma in biorefineries with a novel hybrid mass–energy allocation method

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Abstract

Processing biomass into multifunctional products can contribute to food, feed, and energy security while also mitigating climate change. However, biorefinery products nevertheless impact the environment, and this influence needs to be properly assessed to minimize the burden. Life cycle assessment (LCA) is often used to calculate environmental footprints of products, but distributing the burdens among the different biorefinery products is a challenge. A particular complexity arises when the outputs are a combination of energy carrying no mass, and mass carrying no energy, where neither an allocation based on mass nor on energy would be appropriate. A novel hybrid mass–energy (HMEN) allocation scheme for dealing with multifunctionality problems in biorefineries was developed and applied to five biorefinery concepts. The results were compared to results of other allocation methods in LCA. The reductions in energy use and GHG emissions from using the biorefinery's biofuels were also quantified. HMEN fairly distributed impacts among biorefinery products and did not change the order of the products in terms of the level of the pollution caused. The allocation factors for HMEN fell between mass and economic allocation factors and were comparable to energy allocation factors. Where the mass or the energy allocation failed to attribute burdens, HMEN addressed this shortcoming by assigning impacts to non-mass or to nonenergy products. Under the partitioning methods and regardless of the feedstock used, bioethanol reduced GHG by 72–98% relative to gasoline. The GHG savings were 196% under the substitution method, but no GHG savings occurred for sugar beet bioethanol under the surplus method. Bioethanol from cellulosic crops had lower energy use and GHG emissions than from sugar beet, regardless of the allocation method used. HMEN solves multifunctional problems in biorefineries and can be applied to other complex refinery systems. LCA practitioners are encouraged to further test this method in other case studies.

Keywords: allocation, biochemical, biofuels, biomaterial, biorefinery, GHG emissions, HMEN method, life cycle assessment

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Introduction

To cope with population growth and the rapid depletion of fossil resources, the EU and the United States have proclaimed their interest in strengthening green growth in the bioeconomy (EC, 2012, The White House, 2012). More than 30 countries have expressed their intentions to increase their reliance on biological resources (Bosch *et al.*, 2015), so the share of biomaterials and bioenergy is expected to increase in the coming decades. The development of biorefineries is crucial for achieving the transition to a bioeconomy. Biorefineries convert biomass into food, energy, chemicals, and

materials (Sacramento-Rivero *et al.*, 2016). They can contribute to sustainable resources use and so conserve finite resources, while mitigating climate change and other impacts. However, environmental impacts occur during the production and conversion phases of biomass into energy and bio-based materials (Creutzig *et al.*, 2015; Gerssen-Gondelach *et al.*, 2016), and these impacts should be reflected in the environmental assessment of individual biorefinery products.

To support environmental claims about biorefineries, a life cycle assessment (LCA) is used (Cherubini & Jungmeier, 2010; Nuss & Gardner, 2013; Pereira *et al.*, 2015; Silalertruksa *et al.*, 2017), but as only one product is often of interest for the LCA, environmental loads associated with a biorefinery system are often split among all biorefinery products using appropriate

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allocation methods. Allocation in multifunctional processes has been extensively discussed in the literature (Azapagic & Clift, 1999), and several methods for solving these problems have been proposed (Jungmeier *et al.*, 2002). The LCAs for petroleum refineries and for bioenergy and biorefinery systems are, in some cases, sensitive to allocation methods (Wang *et al.*, 2004; Börjesson, 2009; Gnansounou *et al.*, 2009; Luo *et al.*, 2009; Cherubini *et al.*, 2011). The choice of an adequate method is still a contentious issue, and an arbitrary choice can lead to incorrect LCA results (Reap *et al.*, 2008) and thus poor decision-making (Weidema, 2000).

Studies on petroleum refineries and bioenergy systems have so far used mass (Gabrielle & Gagnaire, 2008), energy (Huo *et al.*, 2009), and exergy (Cherubini *et al.*, 2008) as parameters to allocate resource use and GHG emissions to the products and coproducts of these systems. Mass, energy, and exergy allocations are often considered to be based on physical parameters because they use measurement units such as weight or energy content as their basis. But the direct application of these methods to biorefineries is challenging because of the complexity of biorefining processes, the large number of end products, and the diversity of their functions. Mass allocation is unsuitable for nonmass products like electricity, while, without further assumptions, the energy allocation would not work for nonenergy products (e.g., fertilizers) that do not have a heating value (Singh *et al.*, 2010). The exergy allocation is complex as a result of difficulties in establishing exergy values for some substances (Cherubini *et al.*, 2011).

The economic allocation (Spirinckx & Ceuterick, 1996; Guinée *et al.*, 2004) and the linear programming method (Azapagic & Clift, 1999; Babusiaux, 2003; Pierru, 2007; Hirshfeld & Kolb, 2012; Elgowainy *et al.*, 2014; Balakrishnan *et al.*, 2015) have also been used to split burdens among petroleum refinery, bioenergy, or biorefinery products. The economic allocation considers the financial incentives, which are the main drivers of production and associated impacts. But the method cannot be applied to systems where coproducts do not yet have a market or where market prices fluctuate (Wang *et al.*, 2011). The linear programming method models the physical and technical relationships between the inputs and outputs and environmental burdens of the system. It follows similar logics of economic allocation (Bredeson *et al.*, 2010) and provides detailed information on the operations within the biorefinery (Balakrishnan *et al.*, 2015). But the approach does not work if there is a fixed ratio between products and coproducts because the functional outputs cannot be varied independently (Azapagic & Clift, 1999). Linear programming is also data intensive, and the data needed may not be easily accessible.

Other studies avoid allocation either via system subdivision (Furuholt, 1995; Wang *et al.*, 2004; Bredeson *et al.*, 2010), substitution (Kim & Dale, 2002; Eriksson *et al.*, 2007), or using the system expansion approach (Njakou Djomo *et al.*, 2015). The subdivision method disaggregates processes into subprocesses within a given system and splits off those that are relevant to the functional output. It captures the differences in environmental loads of producing individual products at the next sublevel (Wang *et al.*, 2004). But biorefinery processes are very integrated and cannot be meaningfully split into subprocesses. The use of the substitution method requires the identification of the main product and the coproducts. The main product is then allocated the entire burden, but also credited with the impacts that the coproducts can avoid by replacing other products on the market. However, in the case of the biorefinery, the identification of the main product is not obvious because the overall idea of biorefining is to utilize synergies in the production with the purpose of obtaining multiple products (Parajuli *et al.*, 2015).

The system expansion method broadens the boundary to the point where allocation is not needed and where the compared systems cover the same functional unit (Ahlgren *et al.*, 2015). However, system expansion is difficult to apply when aggregating different functions. Neither does it show impacts of individual products, as its results refer to a group of functions rather than a single product or function. Finally, a small number of studies have used the surplus method (Fu *et al.*, 2003; Pimentel & Patzek, 2005) to overcome allocation problems with bioenergy systems. This latest method identifies and assigns all burdens to the main product. In this method, coproducts are burden-free and thus considered as waste products. This, however, seems to be a simplification of the reality for the reason that some biorefinery coproducts have well-established markets (e.g., protein and lignin) and cannot thus be regarded as wastes (Maes *et al.*, 2015).

As none of the methods above are without drawbacks when applied to biorefineries, new methods for solving multifunctionality problems in biorefineries could help to settle the debate. In response to this situation, a number of researchers proposed a framework to deal with allocation in biorefining (Cherubini *et al.*, 2011; Sandin *et al.*, 2015). Although appealing, their approaches may be criticized in that they fail to reduce the number of options that LCA practitioners are faced with and thus increase the risk of controversies. For many authors, mass- and energy-based allocation methods are very attractive as they are simple and based on clear and defensible rationales (Pelletier *et al.*, 2015). They provide an engineering perspective to allocation within a given refinery (Wang *et al.*, 2004). However, they are not

directly applicable to biorefinery systems for the reasons mentioned above. Therefore, a new method based on physical parameters and which is an improvement over the current mass and energy allocation is needed. Here, we present a new hybrid mass–energy ‘HMEN’ allocation method and the results from its application to different biorefinery concepts. To test the robustness of this new approach, its results were compared to results of other allocation methods in LCA. Finally, gasoline and natural gas were used to illustrate the effects of replacing conventional fossil fuels with bioethanol and biogas.

Materials and methods

Biorefinery models

Different biorefinery concepts exist today, with different pathways for biomass conversion and different final products. Simple biorefinery concepts use current available technologies to convert, via a platform, a single biomass feedstock into two or three marketable products. Complex biorefinery concepts use novel technologies to convert, via a number of platforms, a variety of biomass feedstocks into several marketable products (IEA, 2009). Both energy (e.g., biofuels) and nonenergy products (e.g., feed, biomaterial, and/or biochemical) from simple

and complex biorefineries were evaluated from cradle-to-gate in this study (Fig. 1).

Allocation framework

To overcome the limitations associated with allocation based solely on energy content or mass, a new hybrid mass–energy (HMEN) allocation method was developed (Fig. 1). This new allocation method captures well the differences in energy efficiency between biorefinery systems. A dispatch factor for energy $\alpha = \frac{\sum \eta_i}{\eta_0}$ was derived from the following relationship: $\eta_0 = \sum \eta_i + \sum \eta_j$, where η_0 is the overall energy efficiency of the biorefinery, $\sum \eta_i$ is the efficiency of the energy stream, and $\sum \eta_j$ the efficiency of the material stream. As this relation equals to $\frac{\sum \eta_i}{\eta_0} + \frac{\sum \eta_j}{\eta_0} = 1$ when its two sides are divided by η_0 , it thus follows that $1 - \alpha$ represents the dispatch factor for the material stream (Table 1). Based on the abovementioned dispatch factors, the classical equations for distributing burdens between energy and nonenergy products can be modified as shown in Eqns (1) and (2).

$$\Phi_i = \alpha \frac{\theta_i}{\sum_i^n \theta_i} \quad (1)$$

$$\Phi_j = (1 - \alpha) \frac{\lambda_j}{\sum_j^m \lambda_j} \quad (2)$$

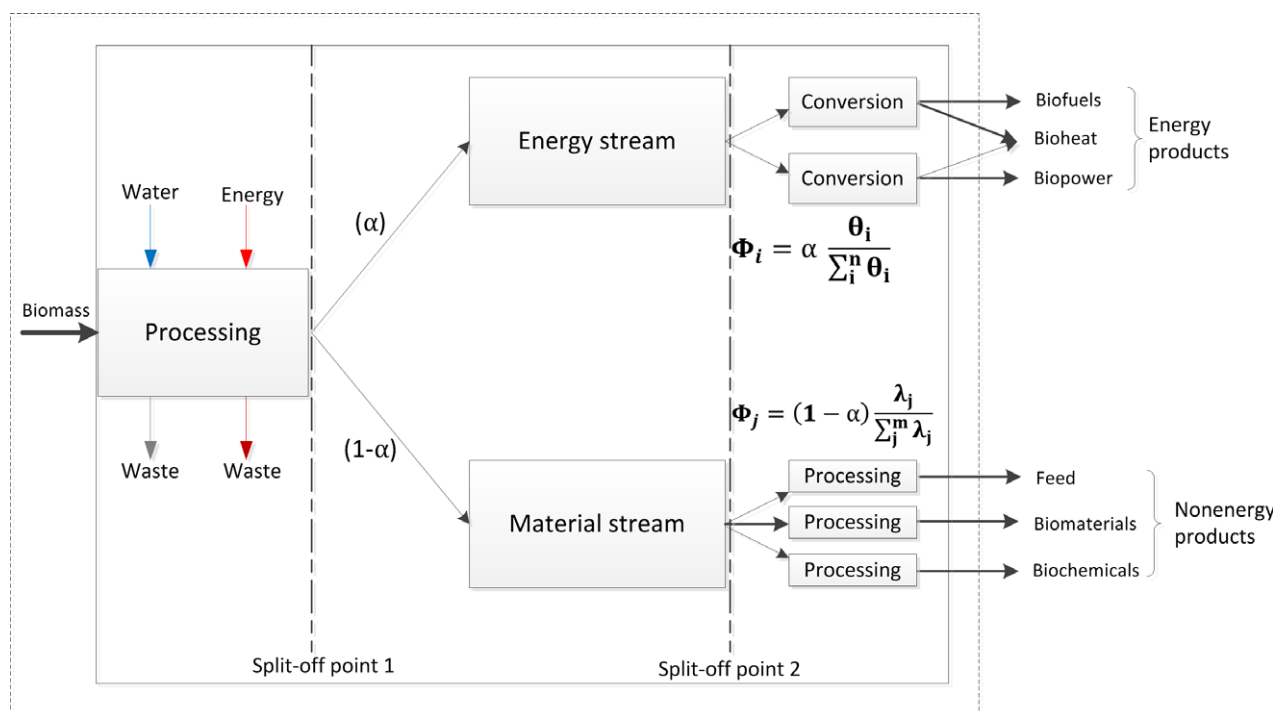


Fig. 1 System boundary and schematic representation of the hybrid mass–energy (HMEN) method. At split-off point 1, a dispatch factor (α) which divides a biorefinery into energy and material streams is computed. At the split-off point 2, the dispatch factor is combined with mass and energy allocation method to derive the allocation coefficients. The dotted line represents the system boundary.

Table 1 Key figures for the processing and conversion of the different feedstocks in the selected biorefinery plants

Biorefinery 1 (BioR ₁)			Biorefinery 2 (BioR ₂)			Biorefinery 3 (BioR ₃)			Biorefinery 4 (BioR ₄)			Biorefinery (BioR ₅)		
Component	Mass (t h ⁻¹)	Energy (GJ h ⁻¹)	Component	Mass (t h ⁻¹)	Energy (GJ h ⁻¹)	Component	Mass (t h ⁻¹)	Energy (GJ h ⁻¹)	Component	Mass (t h ⁻¹)	Energy (GJ h ⁻¹)	Component	Mass (t h ⁻¹)	Energy (GJ h ⁻¹)
<i>Inputs</i>			<i>Inputs</i>			<i>Inputs</i>			<i>Inputs</i>			<i>Inputs</i>		
Straw	3.44	57.6	Straw	44.1	635.0	Rapeseed	2.5	92.4	Sugar beet	103.5	1760	Green grass	0.29	5.2
Enzymes	0.034	0	Manure	71.3	42.81	Water	1.1	0.0	Catch crop & leaves	64.3	1093	Silage grass	0.17	3.0
			Biowaste	43.5	27.13	Steam	0.2	0.36	Lime (CaO)	7.5	0	Manure	0.24	4.6
			MSW	21.4	223.6	Electricity	0	0.45	Carbon dioxide	11.5	0	Electricity	0	0.09
			Enzyme	2.14	0				Steam	15.6	35.2	Heat	0.14	0.32
<i>Outputs</i>			<i>Outputs</i>			<i>Outputs</i>			<i>Outputs</i>			<i>Outputs</i>		
2G Bioethanol	0.573	16.0	2G bioethanol	8.58	231.7	1G Bio-oil	1.0	37	1G bioethanol	14.5	388	Biogas	0.17	3.11
Lignin pellets	0.47	10.1	Biogas	1.69	30.39	Protein products*	0.61	9.6	Biogas	45.0	799	Fiber*	0.09	1.46
C5 molasses*	0.965	15.0	Biomethane	1.68	72.2	Soapstock*	0.02	0.4	Sugar*	43.1	646	Coagulated Protein*	0.07	1.32
			Lignin pellets	4.18	92.04	Fiber*	0.76	12.0	Protein products*	12.9	193	Digestate*	0.30	6.6
			Digestate*	3.65	80.2	Ash [†]	0.03	0.0	Foam earth* (CaCO ₃)	8.5	0			
			Heat	111.5	251.9				Carbon dioxide [†]	15.0	0			
			Electricity	0	35.81									
Performance	%		Performance	%		Performance	%		Performance	%		Performance	%	
Overall efficiency	71.3		Overall efficiency	85.5		Overall efficiency	63.4		Overall efficiency	70.0		Overall efficiency	95.0	
Efficiency, energy stream	45.3		Efficiency, energy stream	77.0		Efficiency, energy stream	40.0		Efficiency, energy stream	41.0		Efficiency, energy stream	24.0	
Dispatch factor, energy (α)	64.0		Dispatch factor, energy (α)	90.0		Dispatch factor, energy (α)	63.0		Dispatch factor, energy (α)	59.0		Dispatch factor, energy (α)	25.0	

The overall energy efficiency is calculated as the total energy output of biorefinery products (i.e., energy and nonenergy products) divided by the total energy input.

The efficiency of the energy stream is computed as the sum of energy output of the energy products divided by the total energy input.

The dispatch factor for energy (α) is calculated by dividing the efficiency of the energy stream by the overall energy efficiency.

Note that for the BioR₁ and BioR₂, part of the biomass input is used to produce the energy needed for the process, so no external energy is required.

*Represents the nonenergy products which can be materials or chemicals.

[†]Represents waste (i.e., products with no economic value).

In Eqns (1) and (2) above, θ_i denotes the energy content of a given energy product i from a multifunctional process, and λ_j is the mass of a given nonenergy product j from a multifunctional process, while $\sum_i^n \theta_i$ and $\sum_j^m \lambda_j$ represent the total energy content and the total mass of the generated energy and nonenergy products, respectively. The indices n and m represent the number of energy and nonenergy products, respectively. If $\alpha = 0$, the biorefinery produces only material or chemical products; if $\alpha = 1$, the biorefinery generates only an energy product, and if $0 < \alpha < 1$, the biorefinery system yields both energy and material/chemical products. Note that Eqns (1) and (2) are reduced to a classical energy or mass allocation method, respectively, when $\alpha = 1$ or $\alpha = 0$.

Finally, the share of environmental burdens of a bioenergy/biofuel product from the biorefinery can be calculated as indicated in Eqn (3). Similarly, the environmental burdens assigned to a biomaterial/biochemical product from the biorefinery can be computed using Eqn (4):

$$E_i = \Phi_i E_T = \left(\alpha \frac{\theta_i}{\sum_i^n \theta_i} \right) \cdot E_T \quad (3)$$

$$E_j = \Phi_j E_T = \left((1 - \alpha) \frac{\lambda_j}{\sum_j^m \lambda_j} \right) \cdot E_T \quad (4)$$

where E_i is the share of environmental burdens for a given energy product, E_j represents the share of environmental impacts for a given biomaterial/biochemical product from the biorefinery, and E_T is the total environmental impact of the biorefinery system. Note that the sum of all allocated burdens should equal the total environmental impact generated by the biorefinery (i.e., 100% rule).

Case study selection

The HMEN method was used to estimate and compare the share of energy use and GHG emissions of biorefinery products. Five biorefinery plants were selected, based on the criteria that (i) they generate both energy and nonenergy products, and (ii) there are data available on the overall efficiency of the system. The selected biorefinery plants differed in terms of feedstock used and product outputs (Table 1). The first biorefinery plant (BioR₁) converts straw into bioethanol, lignin pellets, and molasses (Larsen *et al.*, 2012). The second plant (BioR₂) transforms a mixture of straw, manure, industrial waste and biowaste to bioethanol, biogas, biomethane, heat, power, lignin, and digestate (MEC I/S, 2015). The third plant (BioR₃) processes oilseed rape grains into bio-oil, meal cake, and soapstock (Schneider & Finkbeiner, 2013), while the fourth plant (BioR₄) converts sugar beet and catch crop leaves into bioethanol, biogas, sugar, protein products, and foam earth (CaCO₃) (ECN, 2010). The fifth plant (BioR₅) uses grass, grass silage, and manure to produce biogas, fiber, protein products, and fertilizers (ECN, 2010).

Because the impacts from biomass production can be larger than those from conversion (Jungmeier *et al.*, 2002), the study also covers all relevant agricultural operations as well as upstream production of inputs to these operations. Biorefinery inputs such as enzymes, electricity, and heat were included

where relevant in the calculation (Table 1). However, changes in soil carbon stock due to land use changes were excluded from the analysis, as were the storage and end use of biorefinery products. As manure, industrial waste, and biowaste are wastes, only the energy use and GHG emissions related to their transport to the biorefinery plants were considered. A transport distance of 50 km was assumed for all biomass feedstock. Data on energy use and GHG emissions during the production and transport of biomass were derived from the Ecoinvent database (Ecoinvent, 2014), while the data on electricity and heat used for enzyme production were derived from Dunn *et al.* (2012).

Comparison with other allocation methods

The robustness of the HMEN method was assessed by comparing its results to those of the mass, energy, economic allocation, surplus method, system expansion, and the substitution method. For the substitution approach, it was assumed that biofuels represent the main product of the biorefinery in each case study. The economic allocation was based on market prices, while the energy allocation was based on the lower heating values of the different biorefinery products (Table S1).

Comparison of biorefinery biofuels and conventional fuels

The energy use and GHG emissions of the different biofuels (i.e., bioethanol, biogas, biomethane) were compared to those of conventional fuels. To this end, it was assumed that bioethanol replaces gasoline, while biogas or biomethane replaces natural gas. It was further assumed that biofuels are carbon neutral because the CO₂ emitted during the combustion of biofuels corresponds to the CO₂ uptake by the feedstock during their growth. Gasoline production uses 52 GJ t⁻¹ and emits about 624 kg CO₂ t⁻¹ (Ecoinvent, 2014), whereas its combustion releases an additional ~3341 kg CO₂ t⁻¹ (Cherubini & Jungmeier, 2010). Natural gas production consumes 50 GJ t⁻¹ and emits 340 kg CO₂ t⁻¹ (Ecoinvent, 2014), while about 2805 kg CO₂ t⁻¹ is emitted during natural gas combustion. Assuming an energy content of 43 GJ t⁻¹ for gasoline and 27 GJ t⁻¹ for bioethanol, 1 t bioethanol can thus replace 0.62 t gasoline. Similarly, 1 t biogas can displace 0.35 t natural gas if assuming an energy content of 18 and 52 GJ t⁻¹ for biogas (@ 55% CH₄) and natural gas (@ 92% CH₄), respectively. The energy content of biomethane (@ 96% CH₄) is 43 GJ t⁻¹, so 1 t biomethane can displace 0.83 t natural gas (Table S1).

Results

The allocation coefficients, the energy use, and GHG emissions computed using the HMEN and other allocation procedures are shown in Table 2. Under the mass allocation approach, liquid biofuels (e.g., bioethanol) received a lower allocation factor because of their lower weights relative to the other biorefinery products (Table 2). However, under the economic allocation, liquid biofuels were assigned high allocation coefficients

because of the high ratio of price relative to the ratio of differences in mass between liquid biofuels and other cogenerated products. For example, the ratio of the price differences between bioethanol and lignin pellets was 10 times higher than the mass ratio (i.e., 0.33) between these two biorefinery products. The energy allocation and the HMEN method gave similar weights to all energy products, but their allocation factors for nonenergy products differed when these products represented a significant share of the coproducts (Table 2). Importantly, where mass and energy allocation failed to distribute burdens to certain products because of their zero mass value or energy content, the HMEN approach addressed this shortcoming by assigning actual burdens to these products (Table 2). This shows how problematic LCA results can be when mass or energy allocation is applied to biorefineries. Indeed, there is a disproportionate advantage for products without energy content (e.g., foam earth) over products with energy content when the energy allocation was used. The same was true for products with mass over products without mass (e.g., electricity) when the mass allocation was adopted (Table 2). Although the allocation factors differed between the HMEN approach and the other partitioning methods (i.e., mass or economic allocation), the estimates of energy use and GHG emissions per product were in most cases within the same order of magnitude (Table 2). HMEN also had the same prioritization of biorefinery products as the energy allocation in all cases. Its ranking of products was also similar to that of the mass allocation method in nearly all cases, but differed from the economic allocation in some cases. The similarity observed between the HMEN method and the energy allocation in this study reflects the underlying conceptual linkage between the two approaches. In fact, dispatch factors were computed using the energy efficiency, which is, in turn, based on the total energy outputs and energy inputs of the biorefinery systems.

The substitution method credited the main products (i.e., biofuels) with the impacts generated by the alternative goods displaced by their coproducts. No allocation factors were computed here, but because the total avoided burdens exceeded in some cases the overall impacts of the biorefineries, negative estimates for energy use and GHG emissions were obtained (Table 2). This contrasted with the partitioning methods, which all computed a positive total physical energy use and GHG emissions. Such results suggest that when the substitution approach is used, some biorefinery products may become a net sink for energy and/or GHG emissions even before a comparison of these bio-based products with their conventional counterparts.

Figure 2 shows the comparison of the different allocation methods used in this study. Estimates of other allocation schemes were compared to those of the surplus method set as a reference. For simplicity, only biorefineries producing bioethanol were selected (i.e., BioR₁, BioR₂, and BioR₄). The comparison between the mass, energy, economic, and HMEN method showed only small differences among them. Indeed, all estimates of energy use (Fig. 2b) and GHG emissions (Fig. 2a) were within the same order of magnitude. Under the substitution approach, it was clear that avoided impacts by some of the coproducts more than compensated for the overall environmental impacts of some biorefinery products. Figure 2 also shows that the substitution method is not only sensitive to the choice of the conventional good displaced by the coproducts, but also to the number and type of coproducts generated. In fact, large energy and GHG credits were given to the main product when the biorefinery system generated many energy products as in the BioR₂ case (Fig. 2).

The variability in specific energy use and GHG emissions of biorefinery products is presented in Fig. 3. These estimates were obtained by dividing the energy use and GHG emissions in Table 2 by the amount of final products of each biorefinery plant in Table 1. Given that substitution provided only results for the main product, the choice of the main product was varied in each case study to obtain estimates of specific energy use and GHG emissions for all products. Figure 3 shows that the energy use and GHG emissions vary widely depending on the allocation method adopted. Under the partitioning approach (i.e., mass, energy, economic, HMEN), the energy use of bioethanol from straw ranged from 1.7 to 2.9 GJ t⁻¹ (Fig. 3c), while its GHG emissions varied from 149 to 247 kg CO₂ t⁻¹ (Fig. 3a). For straw-bioethanol, the substitution approach computed values of -15.9 GJ t⁻¹ for energy use (Fig. 3d) and 155.6 kg CO₂ t⁻¹ for GHG emissions (Fig. 3b), while the energy use and GHG emission values calculated using the surplus method were 4.8 GJ t⁻¹ and 412.6 kg CO₂ t⁻¹, respectively (Fig. 3b,d).

The energy use of bioethanol from mixed biomass sources ranged from 0.4 to 2.3 GJ t⁻¹ (Fig. 3c), while the GHG emissions varied from 32.9 to 181.8 kg CO₂ t⁻¹ (Fig. 3a) under partitioning approaches. Under the surplus method, the energy use was 6.3 GJ t⁻¹ (Fig. 3d) and GHG emissions were 503.2 kg CO₂ t⁻¹ (Fig. 3b). Negative values were obtained for energy use (-66.1 GJ t⁻¹) and GHG emissions (-2358.6 kg CO₂ t⁻¹) when the substitution method was chosen (Fig. 3d,b). Under the partitioning approach, sugar beet bioethanol consumed 1.4–1.9 times more nonrenewable energy and emitted two to 2.8 times more GHGs than straw-based bioethanol (Fig. 3a,c). When the surplus

method was selected, the energy used was 20.3 GJ t⁻¹ (Fig. 3a), while the GHG emissions were 2555.9 kg CO₂ t⁻¹ (Fig. 3b). Estimates of energy use and GHG emissions for sugar beet bioethanol were -43.8 GJ t⁻¹ and 1426.2 kg CO₂ t⁻¹, respectively, when the substitution method was adopted (Fig. 3b,d).

Overall, the variability in energy use and GHG emissions across the allocation methods suggested the latter had a strong influence on the environmental performance of biorefinery products, but the degree of the influence depended on the biomass feedstock utilized. Some trends were, however, uncovered: In all cases, bioethanol from mixed biomass sources (i.e., manure + straw + food waste) consumed less nonrenewable energy and emitted less GHGs than both bioethanol from straw and sugar beet. Bioethanol from straw had better environmental performances than sugar beet bioethanol. In general, biofuels from cellulosic crops (i.e., bioethanol, biogas, biomethane) used less energy and emitted less GHGs than bioethanol from sugar beet or bio-oil from rapeseed (Fig. 3). Likewise, protein products from cellulosic crops performed better than those produced from sugar beet and rapeseed. The same observation was true for fiber from cellulosic and sugar beet crops. Finally, digestates from cellulosic biomass had a lower impact both in terms of energy use and GHG emissions than digestate from sugar beet (Fig. 3).

Relative to gasoline and under the partitioning methods, all bioethanol reduced the energy use (82–99%) and GHG emissions (72–98%). The largest reduction in energy used (93–99%) occurred when mixed biomass was the feedstock, while the lowest reduction in energy use (82–93%) was linked to sugar beet. Bioethanol from mixed biomass also achieved the largest reduction in GHG emissions (93–98%), while bioethanol from sugar beet had the lowest reduction in GHG emissions (72–88%). Small savings in energy use (37%) and a small increase in GHG emissions (4%) relative to gasoline were observed when the surplus method was used, but under the substitution method, savings in energy use and reduction in GHG emissions reached 304% and 196%, respectively. Compared to natural gas and under the partitioning methods, all biogas reduced the energy use by 80–98% and GHG emissions by 70–97%. As with bioethanol, little to no reduction in either energy use or GHG emissions was achieved when the surplus method was adopted as allocation method. However, when the substitution approach was chosen, the maximum saving in energy used was 2855%, while the maximum reduction in GHG emissions was 1366%. These results showed that allocation methods influenced the savings of biofuel relative to conventional fuels.

Discussion

The HMEN method presented in this study overcomes the limitations of allocation methods based solely on the mass or energy content of biorefinery products, which both suffer from drawbacks. In particular, this new, hybrid method was able to assign environmental impacts to the products with no mass (such as electricity) or no energy content (e.g., earth-foam), which is a major improvement over classical allocations (Table 2). This means that LCA practitioners using these schemes run the risk of overlooking important environmental burdens for certain biorefinery products, a risk which the new method (HMEN) can handle and mitigate (Table 2). Compared to the economic allocation, another widely used method for biorefineries, HMEN is still based on the physical relationship between products and is therefore independent of price fluctuations. Because it uses only the physical flows within biorefineries to split burdens between products, it can limit the freedom of choice and thus the risks of controversy around the outcomes of the evaluation. The use of dispatch factors (i.e., energy content) as a weighting factor is not only consistent with the hypothesis that energy consumption is tied to the amount of mass transported, but also consistent with the conservation of energy during biorefining processes. However, such use of energy content as a weighting factor does not provide information on the degradation of energy or resources during a process, nor does it quantify the usefulness or quality of the various material streams flowing through a system and exiting as products and/or wastes (Wang *et al.*, 2004). Nevertheless, the HMEN method is more robust than the mass and energy allocation currently used in LCA of biorefineries (Table 2). Like the HMEN approach, the economic allocation method distributed impacts among biorefinery products. Differences in estimates between the economic allocation and the HMEN method were in most cases insignificant; for bioethanol, for example, the differences in allocated energy use and GHG emissions were less than a factor of 1.5 (Table 2). Although the economic allocation captures some underlying motivation to produce different biorefining products, the potential market fluctuations (Malça & Freire, 2006), the lack of a physical basis for using market values as weighting factors (Wang *et al.*, 2004), the difficulty of applying this method when no market experience exists, and the uncertainties inherent in this approach favor the use of the HMEN method for attributional LCAs of biorefining products.

The HMEN method performs at a high level of resolution (i.e., at biorefinery level) with regard to process streams in biorefineries. This means it does not allocate

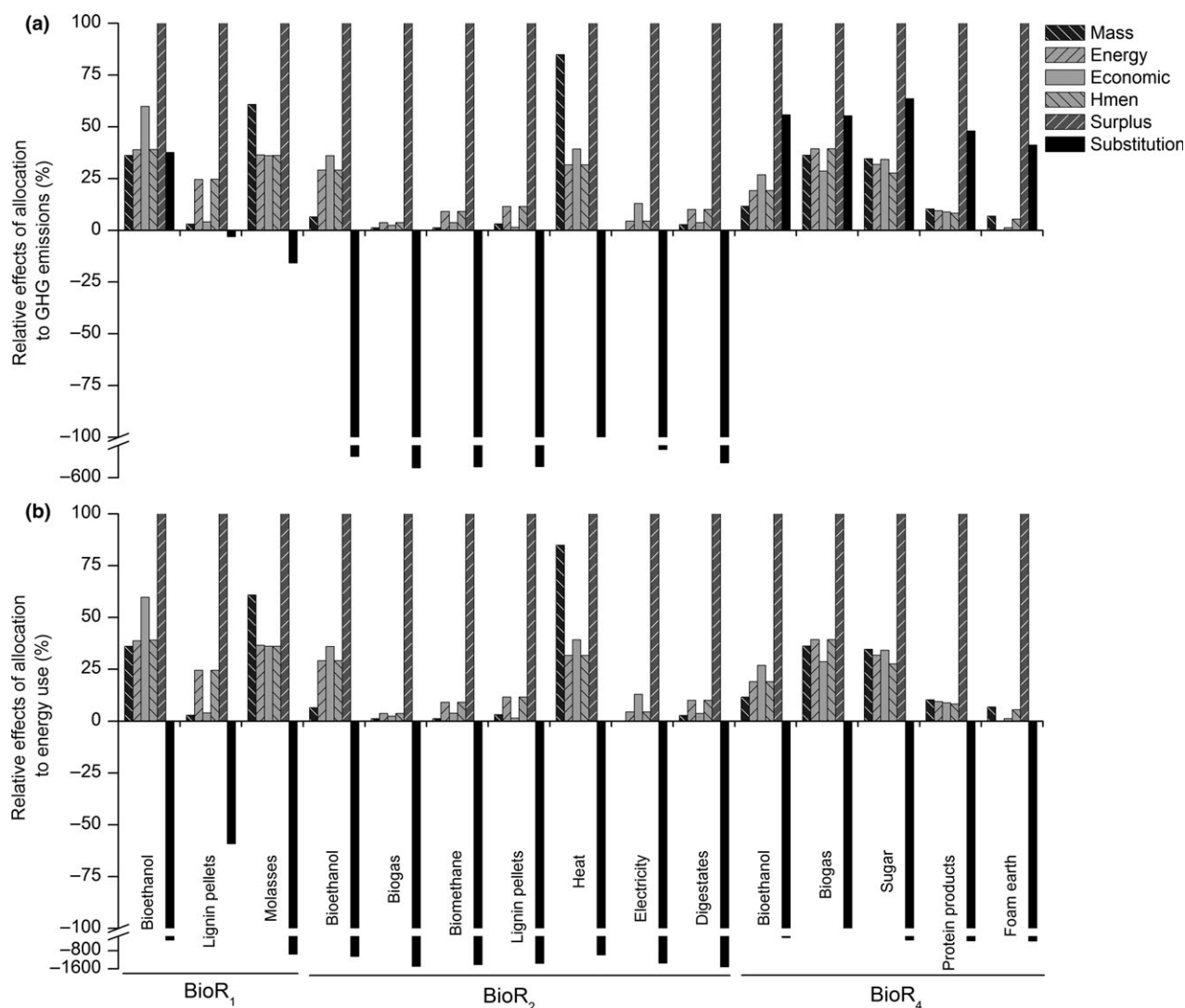


Fig. 2 Comparison of the influence of allocation methods on specific energy use (b) and GHG emissions (a).

emissions to each intermediate product as the subdivision or the LP method does. Although it has been demonstrated that differences in energy use and GHG emissions between allocation at refinery level and the refining at process level can be up to 40% (Furuholt, 1995), our opinion is that intermediate products in biorefineries are often used for internal processes within biorefineries and not sold as final products. Moreover, except for the subdivision and the linear programming, most of the allocation models in LCA perform at a high level of resolution because of a data availability issue. Finally, it has to be acknowledged that the HMEN method does not accommodate the indirect market-mediated effects, a situation which can only be dealt with through substitution. However, this approach is not exempt from shortcomings: Substitution fails to quantify the environmental impacts of a specific

product with sufficient accuracy, as the uncertainties of added system can be overriding (Pawelzik *et al.*, 2013).

We showed that estimates of the energy use and GHG emissions of biorefinery systems varied strongly depending on the allocation method adopted (Fig. 3), which is in line with several previous findings that report effects of allocation method on LCA results and conclusions (Azapagic & Clift, 1999; Heijungs & Guinée, 2007). We found that very few allocation methods arrived at the same prioritization of biorefinery products. This contrasts with the conclusions of Curran (Curran, 2007) that different allocation methods lead to same prioritization of products (Table 2). HMEN results in the same prioritization as the energy allocation, and similar ranking as the economic or mass allocation. Overall, these results reiterate the need for additional and more stringent guidance on allocation issues for

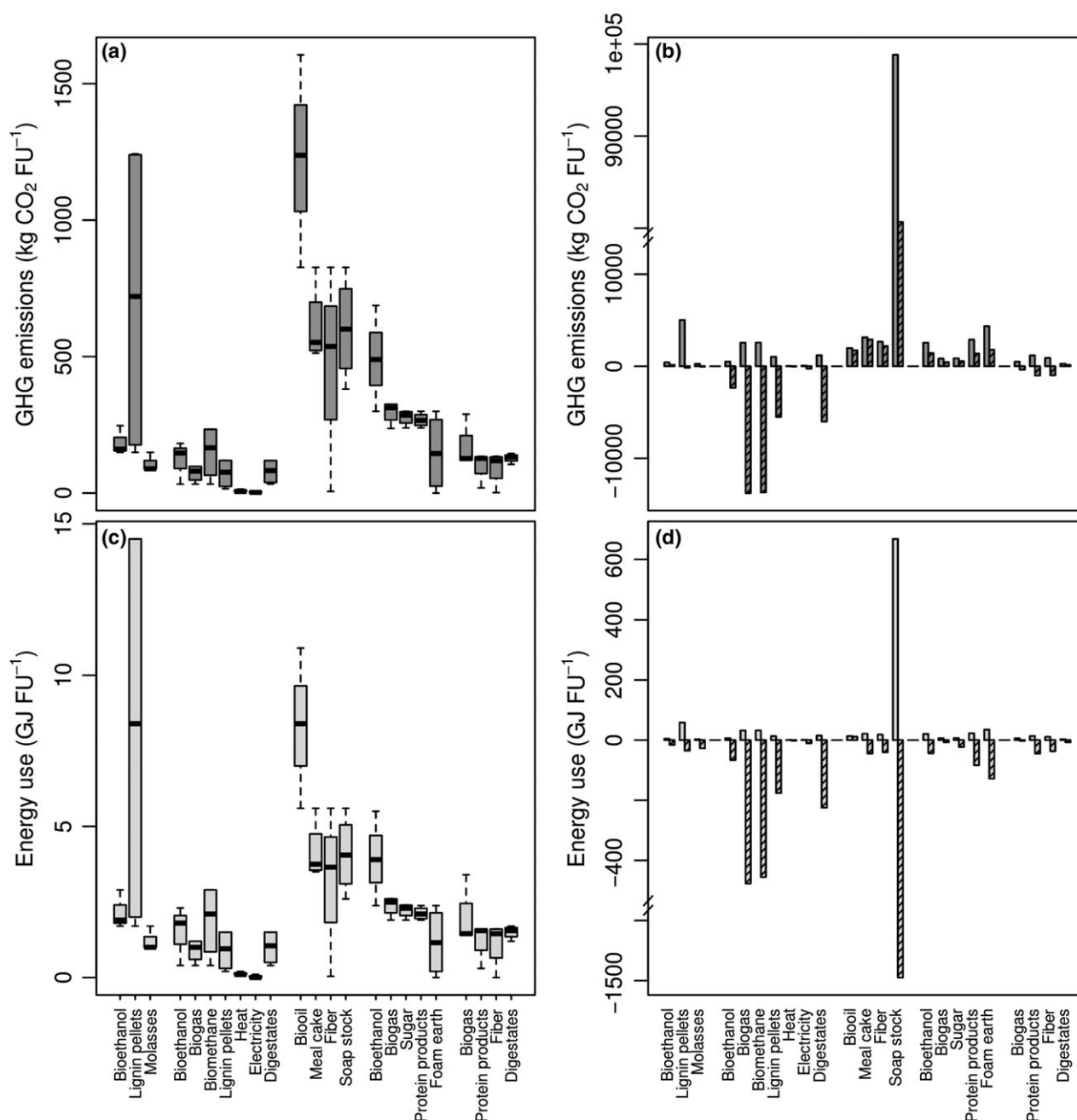


Fig. 3 Variability in specific energy use and GHG emissions of various biorefinery products, depending on type of allocation method applied: (a,c) mass, energy, and economic allocation; and (b,d) surplus (straight) and substitution (striped) methods. Functional unit is 1 t (tonne) for fuels and materials and 1 GJ for heat and electricity.

LCA practices. Without additional guidance, inconsistencies will not be reduced, and this will limit the usability of information from LCA studies in decision-making (Pelletier *et al.*, 2015).

Existing directives are not very helpful for practitioners as they provide different recommendations for assessments of multifunctional systems (Wardenaar *et al.*, 2012). The EU-RED (RED, 2009) suggests the use

of energy allocation, the US-EPA (EPA, 2010) and IEA (Jungmeier *et al.*, 2014) adopt the system expansion method, while the PAS2050 (BSI, 2011) suggests the use of economic allocation where system expansion is not possible. The ISO (ISO 14044, 2006) and the ILCD book (EC-JRC, 2010) recommend firstly avoiding allocation, and secondly applying physical causality-based allocation methods when allocation is unavoidable. Allocation

problems in biorefining can be solved in many ways depending on the research questions (Reap *et al.*, 2008; Cherubini *et al.*, 2011), but any method used for solving this problem must reduce risks of controversy, have the fewest unintended consequences, and generate solutions that are consistent with the principles of ISO standards for LCA. Although substitution is the preferred method in ISO, we showed that it may result in negative estimates of energy use and GHG emissions for the main products because of the credits from the exported energy products of the biorefinery systems. This observation suggests that substitution should not be applied universally without examining the individual situation (Wang *et al.*, 2011). Moreover, the application of the substitution method requires the identification of the main product, which in the case of the biorefinery is difficult (Parajuli *et al.*, 2015), especially if the products are given equal importance. Guidance on identification of the main products exists (Weidema, 2000), but its importance in biorefineries is limited because they generate several marketable products (Bozell & Petersen, 2010). These, coupled to uncertainties about the displaced products, further limit the suitability of this method for biorefineries.

Direct comparison of results from this work with those of other studies on allocation in biorefineries is difficult due to differences in methods used, system boundaries, feedstock and technologies investigated, generated products, in addition to other assumptions. In their analysis of bioethanol from sugar beet refinery, Malça & Freire (2006) assigned between 28% and 37% of energy use to bioethanol. Allocation factors for energy use attributable to sugar beet bioethanol in this study ranged from 12% to 27%. Pulp and sugar were the coproducts in Malça & Freire (2006), while in our study, biogas, protein products, foam earth, and sugar were produced, and this may explain the difference observed. The allocation factors (63–99%) for GHG emissions assigned to straw-bioethanol in Cherubini *et al.* (2011) are higher than those for GHG emissions (36–60%) attributed to straw-based bioethanol in this study (Table 2). Also, the hybrid method of Cherubini *et al.* (2011) assigned 84% of GHG emissions to bioethanol. This latest allocation factor is more than double that for straw-based bioethanol estimated by the HMEN method (39%). Differences in the conceptual framework as well as the type and amount of coproducts explain the difference in results of the two approaches. The hybrid method of Cherubini *et al.* (2011) is a combination of the partitioning and substitution approaches, while the HMEN in this study is a mass- and energy-based allocation. Electricity, heat, and phenols were the coproducts in Cherubini *et al.* (2011), while molasses and lignin pellets were the coproducts in our study

(Table 2). For switchgrass bioethanol in the United States, allocation factors for energy use (62–98%) and GHG emissions (69–98%) (Wang *et al.*, 2011) were also higher than the calculated factors for energy use (36–329%) and GHG emissions (36–60%) in this study. In the US study, switchgrass was converted into bioethanol and electricity, but in our study, straw was converted into bioethanol, molasses, and lignin pellets. Finally, the comparison of bioethanol to gasoline, like that of biogas and biomethane to natural gas, showed that, in general, these biofuels save GHG emissions relative to their fossil fuel counterparts. Reduction in GHG emissions ranged from 42% to 196% for bioethanol, 58% to 1365% for biogas, and from 98% to 616% for biomethane. This reinforces the generally accepted conclusion that biofuels reduce GHG emissions (Jury *et al.*, 2010; Bühle *et al.*, 2011; Monti *et al.*, 2012; Muñoz *et al.*, 2014). However, for bioethanol, the saving in GHG emissions would be much lower because only 5% bioethanol is blended with gasoline in many countries.

By integrating mass and energy allocation methods, the HMEN approach solves the dilemma of multifunctional allocation in biorefineries. HMEN is based on physical relationships between the products and factors in the amounts of biomass used to produce each biorefinery product. It allows environmental burdens to be distributed to all biorefinery products without significant computational difficulties. The application of HMEN to a set of diverse case studies showed that HMEN surpassed existing mass and energy allocation methods. HMEN is thus preferable for complex systems such as biorefineries. Some weaknesses of the HMEN method were highlighted, and future work will explore the possibility to overcome them. Although we have limited the test to biorefineries, the approach can be used in other systems where multifunctionality problems occur. LCA practitioners are thus encouraged to test the method in other case studies.

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Supporting Information

Additional Supporting Information may be found online in the supporting information tab for this article:

Table S1. Lower heating value and market prices of biorefinery products and coproducts.

Table S2. Avoided (i.e. credits) energy use and GHG emissions due to replacement of conventional products by biorefinery coproducts.