Challenges in Quantifying Greenhouse Gas Impacts of Waste-Based Biofuels in EU and US Biofuel Policies: Case Study of Butanol and Ethanol Production from Municipal Solid Waste

Fanran Meng, and Jon McKechnie

Environ. Sci. Technol., Just Accepted Manuscript • DOI: 10.1021/acs.est.9b04286 • Publication Date (Web): 25 Sep 2019

Downloaded from pubs.acs.org on September 28, 2019

Just Accepted

“Just Accepted” manuscripts have been peer-reviewed and accepted for publication. They are posted online prior to technical editing, formatting for publication and author proofing. The American Chemical Society provides “Just Accepted” as a service to the research community to expedite the dissemination of scientific material as soon as possible after acceptance. “Just Accepted” manuscripts appear in full in PDF format accompanied by an HTML abstract. “Just Accepted” manuscripts have been fully peer reviewed, but should not be considered the official version of record. They are citable by the Digital Object Identifier (DOI®). “Just Accepted” is an optional service offered to authors. Therefore, the “Just Accepted” Web site may not include all articles that will be published in the journal. After a manuscript is technically edited and formatted, it will be removed from the “Just Accepted” Web site and published as an ASAP article. Note that technical editing may introduce minor changes to the manuscript text and/or graphics which could affect content, and all legal disclaimers and ethical guidelines that apply to the journal pertain. ACS cannot be held responsible for errors or consequences arising from the use of information contained in these “Just Accepted” manuscripts.
Challenges in Quantifying Greenhouse Gas Impacts of Waste-Based Biofuels in EU and US Biofuel Policies: Case Study of Butanol and Ethanol Production from Municipal Solid Waste

Fanran Meng †,* and Jon McKechnie †

† Sustainable Process Technologies Group, Faculty of Engineering, University of Nottingham, Nottingham NG7 2RD, U.K.

* Corresponding author. E-mail: Fanran.Meng@nottingham.ac.uk
Abstract

Conversion of wastes to biofuels is a promising route to provide renewable low-carbon fuels, based on a low- or negative-cost feedstock whose use can avoid negative environmental impacts of conventional waste treatment. However, current policies that employ LCA as a quantitative measure are not adequate for assessing this type of fuel, given their cross-sector interactions and multiple potential product/service streams (energy, fuels, materials, waste treatment service). We employ a case study of butanol and ethanol production from mixed municipal solid waste to demonstrate the challenges in using life cycle assessment to appropriately inform decision-makers. Greenhouse gas emissions results vary from -566 gCO$_2$eq./MJ$_{\text{biofuel}}$ (under US policies that employ system expansion approach), to +86 gCO$_2$eq./MJ$_{\text{biofuel}}$ and +23 gCO$_2$eq./MJ$_{\text{biofuel}}$ (under initial and current EU policies that employ energy-based allocation), relative to gasoline emissions of +94 gCO$_2$eq. LCA methods used in existing policies thus provide contradictory information to decision-
makers regarding the potential for waste-based biofuels. A key factor differentiating life cycle assessment methodologies is the inclusion of avoided impacts of conventional waste treatment in US policies, and their exclusion in EU policies. Present EU rules risk discouraging the valorisation of wastes to biofuels, and thus forcing waste towards lower-value treatment processes and products.

Graphical abstract
1 Introduction

Liquid biofuels can play a key role in the decarbonisation of the transport sector, and have been studied extensively with life cycle assessment (LCA) tools to quantify their net contribution to addressing greenhouse gas (GHG) emissions associated with conventional, fossil fuels. LCA methodologies have been developed as a quantitative element of transport fuel policies globally, wherein they are used to determine a fuel's eligibility (US Energy Independence and Security Act; EU Renewable Energy Directive) or to calculate its contribution to reducing emissions related to fuel use (e.g., California Low Carbon Fuel Standard). The development of waste-based fuels has received significant attention as they can avoid land use implications of crop-based biofuels (e.g., carbon stock reductions in biomass and soil pools; biodiversity impacts) while also contributing to waste treatment objectives in the perspective of a more circular economy. However, waste-to-energy systems are complex in view of their multi-functional nature: they provide a waste treatment service and can produce a diverse range of material and energy co-products, as well as the primary liquid fuel.
LCA frameworks employed in existing policies which were developed to principally consider crop- and agriculture/forestry residue-based biofuels. These approaches face challenges in evaluating biofuels produced from more complex, mixed waste feedstock streams and in accounting for interactions between the waste treatment and energy sectors.

LCA plays a central and quantitative role in global policies aimed at reducing GHG emissions of transport fuels. In the EU, the Fuel Quality Directive regulates a minimum of 6% reduction of the life cycle GHG intensity of transport fuels by 2020 compared to 2010 level, which can be achieved through the use of biofuels as one means. In order to be considered as renewable biofuels, life cycle GHG emissions must be at least 50% lower than from the fossil fuel they replace and 60% for newer installations from January 2018. Similar thresholds are present in US policy: the Energy Independence and Security Act (EISA) requires biofuels to achieve a life cycle GHG reduction threshold as compared to a 2005 petroleum baseline for different types of biofuels (e.g., 60% reduction for cellulosic biofuel, 50% reduction for advanced biofuel from 2005 petroleum baseline).
renewable biomass, and 20% reduction for conventional biofuels). Low Carbon Fuel
Standards (LCFS), which have been implemented in California and other North
American jurisdictions,²,³ employ a GHG-intensity target to encourage low-carbon
transport fuels.

LCA-based biofuel policies differ substantially in their life cycle GHG emission
calculation methodologies, which has substantial impact on the assessed GHG
emissions of fuels. Prior studies have demonstrated how LCA study factors, including
definition of system boundaries, co-product allocation methods, and selection of
functional units, can return very different results for the same feedstock/fuel pathway.⁴-

The EU RED and FQD policies are based on an attributional LCA methodology,
which attempts to isolate the impact of fuel production and use from connected
systems. Where fuel production processes result in multiple outputs, environmental
impacts are allocated between the primary fuel product and co-products on an energy
basis,¹¹,¹² and therefore the broader impacts of fuel production on co-product markets
is not considered. Numerous prior studies have evaluated biofuels using the EU
methodology and have identified that this approach risks underestimating the
environmental benefit of biofuel systems by ignoring co-product use and
corresponding displacement of production elsewhere (e.g.,7), particularly if co-
products do not have an energy content and therefore cannot be allocated an
environmental impact under the prescribed allocation method.13 This limitation is
particularly relevant for waste-based biofuels, the production of which may encourage
recovery of materials with no energy content (e.g., scrap metal and/or glass for
recycling). Further, wastes are attributed zero GHG emissions;11, 12 as such, avoided
emissions due to diverting wastes from conventional treatment routes (e.g., landfill)
are not credited to the biofuel product.14 In contrast, the US EISA and North American
LCFS policies employ a partially consequential LCA methodology that aims more to
evaluate the change in GHG emissions arising from adoption of alternative fuels.
These policies employ system expansion to deal with multiple products, wherein the
primary fuel product is “credited” with avoided emissions by assuming that co-products
would displace production elsewhere in the economy. Further, benefits of avoided
waste treatment processes, such as landfilling, are also credited to the biofuel product (e.g.,\textsuperscript{15}). With credits from co-products considered, biofuels can in some cases be attributed with negative emissions: credits from co-products exceed the total emissions associated with producing and using the fuel (e.g.,\textsuperscript{16,17}). Such results can be misleading, as the assessed biofuels do not achieve an absolute reduction of atmospheric GHGs, but rather a relative reduction in GHG emissions considering the production displaced by co-products. For waste-derived biofuels, such distortions may be amplified given the potential for a wider range and greater quantity of co-products.

Overall, while existing policies on the surface have similar GHG emissions thresholds, fuel eligibility is dependent on the specific assessment methodologies employed. Ultimately, these methodologies diverge in terms of the “question” they are asking, and therefore whether fuels are evaluated in terms of the overall environmental impacts of the system producing biofuels, or a share of impacts that can be directly attributed to the fuel product in isolation.
Waste-based biofuels can provide policy-relevant benefits beyond provision of low-carbon transport fuels. By diverting waste feedstocks from conventional treatment routes (landfilling; incineration), the high cost of disposal by these routes can be avoided. This is particularly relevant in jurisdictions such as the UK where landfill tax, currently £91.35/tonne, or approximately $120 USD/tonne, greatly increases the cost of disposal by this route. Waste utilisation for fuel production can also encourage the recovery of other materials (e.g., scrap metal, plastic for recycling), and avoiding significant GHG emissions associated with landfilling biogenic wastes (e.g.,) or incinerating plastic-based wastes (e.g.,).

Specific support for waste-derived biofuels varies greatly between regions. The EU RED requires 10% renewable energy share in transport fuel consumption by 2020. A cap limiting first-generation biofuels to 7% share indirectly supports second- and third- generation fuels from non-crop feedstocks, including waste-based biofuels. In the UK, the Renewable Transport Fuel Obligation provides a stricter limitation on crop-based fuels and further incentivises waste-based fuels by awarding double Renewable
Transport Fuel Certificates (RTFC) per litre of liquid renewable fuels derived from certain waste or residue feedstocks; these credits are tradeable and have a market value of £0.18 to £0.24 per RTFC, or approximately $0.25 to $0.30 USD per RTFC, thus financially supporting waste-based fuels. Under the US EISA and California LCFS, there is no specific support for waste-derived fuels.

Waste-based biofuel production systems are complex to evaluate due to their cross-sector interactions (waste and energy/transport sectors) and the wide range of potential co-products. In addition to producing a fuel output, any system producing biofuels from wastes may: 1) avoid current waste treatment processes; 2) enable the recovery of recyclable materials; and 3) co-produce other energy outputs (e.g., excess electricity; heat; fuels). For policies to be comprehensively informed, and for business to make appropriate decisions in response to policies, an appropriate LCA framework is needed to account for this complexity. Therefore, we have developed a case study to explore the implications of LCA methodology decisions on assessed GHG emissions and primary energy demand and to reflect on how these varying model
outputs are capable of answering different questions about waste-based biofuels. The case study considers a MSW to acetone-butanol-ethanol conversion process based on an autoclave mechanical heat treatment process and subsequent fermentation of the biomass fibre to liquid biofuels (butanol, ethanol) and other co-products. Alternative system boundaries and allocation approaches are applied in the context of LCA frameworks within EU and US policies. The results are compared and integrated to more meaningfully inform policymakers and industry on the net GHG implications of waste to biofuel systems.

2 Methods

In this study, we compare life cycle methodologies to evaluate waste-to-biofuels systems and consider how information from the differing approaches can help to inform decision-making. We map these methodology decisions to current and recent biofuels policies in Europe and North America to consider how LCA methodologies
influence the assessed GHG emissions of biofuels. A case study scenario of liquid biofuel (butanol, ethanol) production from municipal solid waste (MSW) is employed.

2.1 LCA methodologies

The overall environmental performance of converting the organic content of MSW to biofuels and concurrently avoiding current waste treatment practices is evaluated. Given the wide range of potential products/co-products (energy outputs; recovered metals/glass/plastics) with diverse materials and energy market applications (see Figure 1), a set of LCA methodologies are deployed to better understand how decisions on how to allocate impacts between liquid biofuel product and the energy and material co-products influence results. We consider the following set of LCA methodologies:

1) US EISA / California LCFS: Avoided waste treatment processes included (credit to primary biofuel product); all co-products evaluated with system expansion (credit to primary biofuel product) (see Section 1.1.1 in SI)
2) EU RED I (original policy): Avoided waste treatment excluded; electricity co-product evaluated with system expansion (credit to primary biofuel product); all other co-products evaluated by energy allocation (see Section 1.1.2.1 in SI)

3) EU RED II (current policy): Avoided waste treatment excluded; exergy allocation of electricity and heat co-products; all other co-products evaluated by energy allocation (see Section 1.1.2.2 in SI)

4) Mass-based allocation alternative: Avoided waste treatment may or may not be included; all co-products evaluated by mass-based allocation (see Section 1.1.3 in SI)

5) Economic allocation alternative: Avoided waste treatment may or may not be included; all co-products evaluated by economic value allocation (see Section 1.1.4 in SI)

The LCA models are developed in GaBi 8.2 using Ecoinvent 3.3 inventory databases, following ISO Standards 14040 and 14044. Two environmental
impacts are quantified: global warming potential (GWP), based on the most recent IPCC 100-year GWP factors to quantify GWP in terms of CO$_2$ equivalents (CO$_2$ eq.)$^{25}$ and primary energy demand (PED) in terms of MJ.

Figure 1 Schematic representation of life cycle assessment of butanol and ethanol from MSW.
The scope and functional unit LCA models are developed to evaluate the case study scenario of MSW conversion to acetone-butanol-ethanol based on an autoclave pre-treatment. Autoclave pre-treatment converts biogenic content to a biofibre material, and enables the recovery of sterilised metal, glass, and plastic materials. The biomass fibre is subsequently converted to liquid biofuels (ethanol, butanol), hydrogen, acetone via hydrolysis and fermentation, and heat/power from combustion of unconverted residual biomass material. The functional unit is one MJ of liquid biofuel (butanol and ethanol), denoted as $\text{MJ}_{\text{biofuel}}$. Results are also considered on the basis of 1 tonne MSW treated. A schematic process flow diagram defining the system boundaries is shown in Figure 1. The system boundaries start from the sorting and transportation of MSW. Prior energy use and environmental burdens of the processes and products that generated MSW are excluded in this study.

2.2 Waste composition and avoided treatment

The waste composition used is representative of the UK MSW with the following wet composition by mass: paper and cardboard (22%), food waste (17%), wood (8.7%),
plastic (22%), glass (1%), garden waste (3%), metals (4%), textiles (6.6%) and others (15.7%). The lignocellulosic content of total MSW is 53% on the wet basis and its moisture content is 40%.

By diverting wastes to biofuel production, conventional treatment processes are avoided. We assume that incoming waste would otherwise be treated by incineration (71%) and landfilling (29%), based on current practices in UK. Considerations of credits related to inclusion/exclusion of avoided waste treatment under different allocation approaches are detailed in Section 2.2. Implications of considering avoided waste treatment are discussed in Section 3.1. For landfill and incineration options, we draw on the results from the Ecoinvent database and literature. For landfill gas recovery, it is assumed that 62% of biogas is recovered (52% for energy recovery and 10% for flaring) and 38% is emitted.

2.3 Waste-to-biofuel process

The MSW-to-biofuel production process has been previously modelled based on a demonstration plant operation and further details on the process are available in the
Supporting Information (SI). The system starts with the pretreatment autoclave process, based on a working facility developed by Wilson BioChemical. MSW is input to the autoclave and treated with steam at moderate temperature (160°C for two hours). The organic content of the MSW is converted into a biomass fibre within the autoclave, which is then recovered to be used as feedstock in ABE production via enzymatic hydrolysis and fermentation. Energy recovery from unconverted biomass and biogas generated in wastewater treatment is sufficient to provide all heat and power requirements of the integrated autoclave/biofuel production process, with excess electricity exported to the grid. Recyclable material streams (ferrous & non-ferrous metals, glass, plastic, wood and textiles) are sterilised within the autoclave and separated from the output stream for subsequent material recovery. All remaining material is classed as waste and sent to incineration/landfill at the same proportions as current waste treatment (see Section 2.2).

Biogenic fibre derived from MSW differs significantly from more conventional biofuel feedstocks, exhibiting a comparatively low total sugar content (~45% glucose and 5%
xylose) and the presence of contaminants that inhibit enzymatic hydrolysis. These factors ultimately limit the liquid biofuel yield. Correspondingly, greater quantities of residual biomass material are available for energy recovery, resulting in a comparatively high output of co-product electricity. Table S1 in the SI details the outputs of the process and their respective destinations.

2.4 Co-product allocation

Co-products arising from the conversion of MSW to liquid biofuel can be classified as energy products (hydrogen; excess electricity); chemicals (acetone); and scrap materials (metal, plastic and glass) (Figure 1). Allocation methods differ between each of the LCA methodologies considered.

The US EISA and California LCFS employ a system expansion approach, wherein co-products are assumed to displace production elsewhere, with associated avoided impacts credited to the primary biofuel product. We assume direct displacement for co-product electricity (avoiding average UK grid generation), hydrogen (avoiding production from fossil fuel sources), and acetone (avoiding primary production). Scrap
materials require further processing before they displace alternative production in a market; these downstream processes to convert scrap to saleable materials are included in the model. Plastic waste, of average composition\textsuperscript{31} is input to a mechanical recycling process to recover, per 1000kg input, 236 kg PET, 63 kg PP, 122 kg PE, and 1 kg PVC.\textsuperscript{32} Recovered materials are assumed to displace primary production. Unrecyclable materials (films, wastes and residues, 580kg) are disposed of by incineration (71%) and landfill (29%).\textsuperscript{27} For metals recycling, we use inventory data from Gabi and Ecoinvent database.\textsuperscript{28, 33} Glass is assumed to be recovered to replace aggregates and result in negligible net change in GHG emissions and PED.\textsuperscript{34} The EU RED methodologies are based on allocating impacts between primary and co-products on an energy content basis. The original policy, EU RED I, requires allocation based on the lower heating value of the products, with the exception of excess electricity which addressed by system expansion. For the EU RED I scenario we assume co-product electricity displaces average UK grid generation. EU RED II employs allocation for co-generated heat and electricity based on their respective
exergy content, which accounts for the temperature (i.e., quality) of the heat product.

All other co-products are considered with energy allocation. Table S4 in the SI presents characteristics and values of exergy allocation. For both EU RED I and II, there is no allocation to non-energy products (recovered metal, plastic). Partitioning ratios are shown in Table 1.

Two additional allocation methods are considered that are able to account for non-energy co-products. Mass allocation distributes the GHG emissions associated with main products and co-products based on their respective mass. A two-stage mass allocation is employed: first, upstream processes and waste pre-treatment are allocated between the biofibre and non-biogenic content on a mass basis (see Figure S5 in the SI). Second, we allocate a share of biofuel production impacts to co-product acetone and hydrogen (electricity and heat have no allocation as they have no mass). Finally, economic allocation apportions impacts between co-products on the basis of their financial value. We conduct the allocation considering the overall production
outputs, as intermediate product (biofibre) does not have a financial value (see Table 1).

Table 1 Partitioning ratio for mass, energy, and economic allocation.

| Allocation | 1-Autoclave | 2-Biorefinery | Recovered plastic, metal, glass |
|------------|-------------|---------------|---------------------------------|
| Biogenic   | Butanol and ethanol | Acetone | Hydrogen | Electricity and heat |
| Energy value-EU RED I | 30.7% | 69.3% | 61.5% | 22.3% | 16.1% | - |
| Energy value-EU RED II | 30.7% | 69.3% | 8.5% | 3.1% | 2.2% | 86.3% |
| Mass value-general | 53% | 47% | 3.2% | 1.3% | 0.2% | 95.3% (as biomass fuel) |
| Economic value | - | - | 23.8% | 6.1% | 2.3% | 48% |
|             |             |               |                                   |                                | Recovered plastic, metal, glass |

Page 21 of 44

Environmental Science & Technology
3 Results and Discussion

3.1 Greenhouse gas emissions evaluated under current policies

Overall, the production of liquid biofuels (butanol, ethanol) from MSW achieves lower GWP than the reference gasoline product. However, quantified impacts vary substantially between the LCA methodologies considered.

Waste-derived biofuels achieve substantial reductions in GHG emissions and PED relative to gasoline when employing the system expansion approach, as in US EISA and CA LCFS policies. Negative GHG emissions (-600% relative to gasoline) are achieved, due in large part to the significant credit for avoiding landfilling and incineration in current waste treatment (-576 gCO$_2$/eq./MJ$_{\text{biofuel}}$). Excluding avoided waste treatment would still result in very low GHG emissions under system expansion approach (11 gCO$_2$/eq./MJ$_{\text{biofuel}}$) as a result of significant co-product credits for electricity export and metals recovery (-166 and -202 gCO$_2$/eq./MJ$_{\text{biofuel}}$, respectively).

Recovery of plastics does not provide a significant net reduction in GHG emissions, as recycling and residual waste disposal incurs similar emissions (418
gCO$_2$eq./MJ$_{\text{biofuel}}$ as those associated with avoided primary plastic production (-446 gCO$_2$eq./MJ$_{\text{biofuel}}$), assuming 100% displacement with recovered plastics. If recycled plastics are used in other markets, due to their potentially reduced quality relative to primary plastics, this co-product credit may moderately decrease.\textsuperscript{32} Co-products of acetone and hydrogen only contributes to 4% of the total credits (-30.34 gCO$_2$eq./MJ$_{\text{biofuel}}$) (see Figure S7). It is noted in this case study, the relatively low sugar yield by hydrolysis and correspondingly low biofuel yield results in larger quantities of residual biomass available for co-product electricity production than with conventional feedstocks. Major GHG emissions sources arise from the manufacture of enzymes (187 gCO$_2$eq./MJ$_{\text{biofuel}}$), included in the total biorefinery emissions indicated in Figure 2a (also see Table S6). Other process inputs (pH control; fermentation nutrients; microorganism) have smaller impacts, totalling 20.87 g CO$_2$eq./MJ$_{\text{biofuel}}$. Treatment of residual waste from autoclave has a large GHG emission of 141.01 g CO$_2$eq./MJ$_{\text{biofuel}}$. Collection and transport accounts for about 4% while fuel distribution and use accounts for less than 1% of the total PED and GHG emissions (see Figure S7 in the
On balance, with substantially negative GHG emissions, the MSW-derived biofuels would by far surpass the eligibility requirements for the US EISA policy. GHG emissions are substantially higher when allocation is used to evaluate the MSW-derived biofuels. The initial RED I policy employs energy allocation between products, with the exception of co-product electricity: excess electricity is evaluated by system expansion, and the credit from displacing generation elsewhere is allocated between the biorefinery products. No impacts are allocated to the recovered metal and glass co-products, as these material do not have an energy content. RED I results in GHG emissions of 86 gCO$_2$ eq./MJ$_{biofuel}$, achieving only a minor reduction of 9% relative to gasoline and therefore would not qualify as an eligible biofuel under the policy.

Enzyme production$^{29}$ represents approximately 85% of net emissions allocated to biofuel production. The higher net GHG emissions, relative to the system expansion approach, result from the exclusion of avoided waste treatment and the apportioning of the co-product electricity credit between biofuels and other products: of the total 102 gCO$_2$ eq./MJ$_{biofuel}$ credit, only 31 gCO$_2$ eq. is credited to the biofuel product. Thus,
although the production of biofuels from MSW would achieve significant overall GHG reductions when all products are considered, this pathway would not be eligible under the original RED policy.

In contrast, under the revised RED II policy, the MSW-derived biofuels would be eligible, with overall GHG emissions of $23 \text{ gCO}_2 \text{eq/MJ}_{\text{biofuel}}$, a reduction of 75%. With exergy allocation applied to the co-product electricity and heat, a large share of biorefinery emissions (86%) are applied to these outputs; correspondingly, fewer emissions are attributed to the biofuel product. Excess electricity is attributed with GHG emissions of $86 \text{ gCO}_2 \text{eq/MJ}$, which represents a 12% reduction compared to UK grid electricity mix\textsuperscript{35} (see Table S5 in the SI). Enzyme production still contributes the largest share of GHG emissions attributed to the biofuel outputs (68%).

By excluding avoided waste treatment impacts, the RED I and RED II policies ignore an important service provided by waste valorisation systems of diverting and treating waste that would otherwise be destined to landfill/incineration. Inclusion of avoided waste treatment would reduce the GHG emissions assessed under RED I and RED
Apportioning credits related to avoided landfilling and incineration processes would result in net GHG emissions of -22 gCO$_2$eq./MJ$_{\text{biofuel}}$ and 8 gCO$_2$eq./MJ$_{\text{biofuel}}$ for RED I and RED II, respectively. In both cases, biofuel products would achieve eligibility, with net emissions more completely quantified by including the impact of waste diversion from conventional routes to input to the production system.

### 3.1.1 Greenhouse gas emissions evaluated under alternative allocation methods

Mass and economic allocation are considered as alternatives to system expansion and energy allocation approaches, as these allow allocation to non-energy products (recovered metal, glass) (see Figure 2a and Table S6). With mass allocation, only a small fraction of impacts are allocated to the biofuel products, which represent only 3% of product outputs by mass. As a consequence, biofuels are attributed a small net GHG emission of 9.2 gCO$_2$eq./MJ$_{\text{biofuel}}$, or -0.4 gCO$_2$eq./MJ$_{\text{biofuel}}$ if avoided waste treatment is considered. A higher share of emissions are attributed to the biofuel products under economic allocation (36%) due to the comparatively high value of
these outputs relative to other products, resulting in net GHG emissions of 56 gCO$_2$eq./MJ$_{biofuel}$ or -81 gCO$_2$eq./MJ$_{biofuel}$ if avoided waste treatment is considered.

3.2 Primary energy demand

MSW-derived biofuels are associated with lower PED than conventional gasoline fuel. As with GWP, however, the calculated PED varies substantially between LCA methodologies considered (see Figure 2b and Table S6). Applying system expansion, as in the US EISA and CA LCFS policies, returns a strongly negative value, with PED at -1,238% relative to gasoline. The large co-product credit associated with recovered plastics is principally responsible, by avoiding both the consumption of feedstock and process energy required for plastics manufacture (-12.9 MJ/MJ$_{biofuel}$). Further, the disposal of most residual plastic waste by incineration provides useful energy outputs (heat, electricity) which are credited to the primary biofuel product. Thus plastic recycling is much more beneficial from a PED perspective than when considering GHG emissions as in Section 3.1. Electricity co-product also contributes to the strongly negative PED value (-6.85 MJ). The largest PED source is the manufacture of
enzymes, contributing approximately 2.31 MJ/MJ\textsubscript{biofuel} (total biorefinery demands 3.50 MJ/MJ\textsubscript{biofuel}). Excluding waste treatment results in lower impacts being assessed for the biofuel products, in contrast with the GWP results. Incineration of residual wastes provides useful energy outputs, which are forgone when waste is diverted to the biorefinery process. As such, if avoided waste treatment is excluded from the analysis, net PED increases to -17.8 MJ/MJ\textsubscript{biofuel}.

Results for the allocation approaches (RED I and II, mass, and economic allocation) follow a similar pattern as those presented in Section 3.1 for GWP. Under RED I, a reduction in PED of 23% relative to gasoline is achieved, as only the electricity co-product credit is applied to products. For RED II, a significant share of energy use is allocated to the heat and electricity co-products, and thus only a small PED consumption is attributed to liquid biofuels, resulting in a 58% reduction relative to gasoline. Similarly, mass allocation returns a PED reduction of 75% as only a small share of production impacts are attributed to the biofuels. From an economic allocation perspective, however, the biofuel products represent a large share of value of the
product outputs (24%) and are correspondingly attributed a large share of life cycle PED, resulting in a net increase relative to gasoline of 60%.
**Figure 2** Life cycle global warming potential (a) and primary energy demand (b) of MSW-derived liquid biofuel relative to reference fossil fuel based on different allocation methods.

### 3.3 Comparison with other waste treatment routes

Presenting study results on the basis of 1 tonne MSW treated enables comparison between waste management options. For this analysis, system expansion is employed...
for all waste treatment processes to understand the total impacts of the treatment process and product outputs (including liquid biofuel use in place of gasoline). We compare current case study results with conventional treatment processes: landfill without biogas recovery, landfill with biogas recovery and incineration for energy recovery (electricity only or CHP generation) (Figure 3). MSW conversion to liquid biofuels is the superior option for both categories, achieving significantly greater reductions in GHG emissions and PED than conventional waste treatment routes. Without landfill gas capture, sanitary landfill operation emits the highest GHG emissions of 573 kg CO$_2$eq/t MSW; with landfill gas capture, the GWP of landfills for the electricity only and CHP options are 240 and 223 kg CO$_2$eq/t MSW, respectively. Increasing capture rate of biogas has been reported to be key in reducing the GWP of the landfill option$^{20}$. In comparison, incineration is a net source of GHG emissions, as fossil CO$_2$ emissions, largely from plastics combustion, exceed avoided emissions associated with displacing heat and electricity production elsewhere. This results in emissions of 174 and 58 kg CO$_2$eq/t MSW for the electricity-only and CHP incinerators,
respectively. Incineration is able to recover useful energy from waste, indicated with negative PED for both electricity and CHP scenarios (-3070 and -4880 MJ/t MSW, respectively. MSW conversion to liquid biofuels, alongside electricity, acetone, hydrogen, and recyclates, delivers a far greater reduction in GWP (-30 kg CO₂eq/t MSW) and PED (-10357 MJ/t MSW) than conventional waste treatment options (disaggregated inputs can be found in Figure S8 in the SI). Further improvements could be realised by finding markets for excess heat. In the current study, we assume excess heat has no use. However, if the autoclave/biorefinery is integrated with other industrial processes, district heating, or finds other uses (sterilization; cooling generation), this would result in further reductions in GWP (110 kgCO₂eq./t MSW) and PED (1,700 MJ/t MSW).
3.4 Discussion

The study evaluates alternative allocation methodologies for the life cycle evaluation of waste-derived biofuels, considering a case study of butanol and ethanol production from MSW. While biofuel production from MSW is demonstrated to reduce GHG emissions and PED relative to gasoline, the magnitude of these reductions are dependent on the allocation method employed. In practice, LCA researchers and
policy makers have to select one allocation method that is most appropriate for the analysis of biofuel systems. To do so requires careful consideration, as alternative allocation methods are ultimately answering very different questions.

System expansion aims to understand the overall impact of introducing a new product system. This approach benefits from its comprehensiveness in evaluating the overall impact of the product system, but is based on a clear identification of biofuels as the primary product and all other outputs as secondary. Where there are large co-product outputs, associated credits can distort the results and risk not reflecting stakeholder values or decision criteria. In the current study, significant electricity and recovered metal co-products contribute to very negative emissions; avoided waste treatment benefits are also solely attributed to the primary product. Whether it is appropriate to consider liquid biofuels as a primary product is questionable, given that this output represents only 24% of the financial value of system outputs, and substantially less on energy (2.6%) and mass (1.7%) bases.
An alternative approach to evaluate the overall impacts of waste conversion to biofuels may be from the perspective of a waste treatment system – using a functional unit of one tonne MSW, or equivalent – and thus taking into account all of the diverse outputs of the system without having to artificially identify a single primary product. Such results are not useful in biofuels policies that require a specific impact be attributed to the biofuel product. However, such a framework could be appropriate for a waste treatment sector-focused approach to evaluating and supporting higher value products from waste (biofuels, bulk and high value chemicals, others), while concurrently supporting diversion from conventional treatment routes.

In contrast, allocation approaches aim to attribute impacts to a specific, single product. Allocation is, in theory, effective at isolating the impact of biofuel products from the other outputs of the waste biorefinery system. However, the diversity of products poses a challenge, as some cannot be addressed with energy allocation (e.g., recovered metal, glass), and others cannot be addressed by mass allocation (e.g., electricity, heat). Economic allocation may be more appropriate considering
these issues, with further benefit of being able to better consider the motivations of
producers. However, this approach faces challenges including fluctuation of results
with market prices, and challenges of including non-monetisable goods within the
analysis.

A key question facing the analysis of waste-derived fuels is how avoided waste
treatment should be included within LCA calculations. Avoided waste treatment is
excluded in EU policy, but this approach ignores the “co-service” of waste treatment
provided by biofuel production and thus overestimates the impacts of waste-based
fuels. In contrast, system expansion gives full credit to biofuels for waste diversion,
despite this being but one product of the biorefinery system, and ignoring any other
changes occurring in the waste treatment sector, including those in response to policy
drivers to limit or reduce waste to landfill (and increasingly, to incineration). In future,
multiple viable opportunities may exist to utilise MSW, and therefore the role of a single
use in avoiding conventional waste treatment would be questionable. Sector
interactions are notoriously challenging for LCA to address (for example, induced land
use change arising from crop-based biofuels), but should be pursued in future work to ensure that the contexts of the energy and waste sectors are properly considered. At present, by excluding the benefits of diverting wastes from landfill and incineration, EU policy disadvantages biofuel production relative to other, lower-value uses of waste streams.

Biofuel production from mixed wastes poses specific challenges to LCA practitioners and policymakers. As illustrated in the current study, methodology decisions dramatically influence results, with waste-derived biofuels either reducing GHG emissions by 9% relative to gasoline under the EU’s RED I policy, or by 700% using system expansion as in US EISA and CA LCFS policies. Development of a relevant LCA framework that can account for the complexities of waste biorefining is essential to provide appropriate policy support for waste-derived fuels.
Associated Content

The Supporting Information is available free of charge on the ACS Publications website at DOI: XXX.

Supporting Information includes additional details on the allocation method, figures and tables that support the modelling and the results interpretation. Figures S1–S6 show the boundaries, flows and processes considered in the allocation methods. Figures S7-9 shows the environmental efficiency of waste to biofuel, comparison with other waste treatment routes and sensitivity analysis results. Table S1 summarises the outputs of the autoclave and biorefinery process. Table S2 is an overview of current biofuel regulations in the EU and US. Table S3 characterises the system expansion method for MSW to ABE pathway. Table S4 presents characteristics and values of exergy allocation for RED II methodology. Table S5 displays GHG emissions of co-products under different allocation methods compared to Ecoinvent 3.3 values. Table S6 presents life cycle GWP and PED of MSW-derived liquid biofuel relative to reference fossil fuel based on different allocation methods corresponding to Figure 2.
Author Information

Corresponding Author

* E-mail: Fanran.Meng@nottingham.ac.uk; phone: +44 0115 74 87191

ORCID

Fanran Meng: 0000-0002-9014-1231

Jon McKechnie: 0000-0001-5656-1649

Notes

The authors declare no competing financial interest.
Acknowledgment

This work was supported by the funding from the Bioenergy Sustaining the Future 2 MSWBH Project 620103. It was also funded by the Industrial Biotechnology Catalyst (Innovate UK, BBSRC, EPSRC) to support the translation, development and commercialisation of innovative Industrial Biotechnology processes.

References

1. European Union Fuel Quality Directive. Fuel quality. <https://ec.europa.eu/clima/policies/transport/fuel_en>, (accessed November 2018).
2. Oregon Department of Environmental Quality (Oregon DEQ). Oregon Clean Fuels Program. <https://www.oregon.gov/deq/aq/programs/Pages/Clean-Fuels.aspx>, (accessed April 2019).
3. British Columbia (BC). Renewable & Low Carbon Fuel Requirements Regulation. <https://www2.gov.bc.ca/gov/content/industry/electricity-alternative-energy/transportation-energies/renewable-low-carbon-fuels>, (accessed April 2019).
4. Luo, L.; van der Voet, E.; Huppes, G.; Udo de Haes, H. A. Allocation issues in LCA methodology: a case study of corn stover-based fuel ethanol. Int. J. Life Cycle Ass. 2009, 14 (6), 529-539.
(5) McKechnie, J.; Pourbafrani, M.; Saville, B. A.; MacLean, H. L. Exploring impacts of process technology development and regional factors on life cycle greenhouse gas emissions of corn stover ethanol. *Renew. Energ.* **2015**, *76*, 726-734.

(6) Murphy, C. W.; Kendall, A. Life cycle inventory development for corn and stover production systems under different allocation methods. *Biomass Bioenerg.* **2013**, *58*, 67-75.

(7) Cai, H.; Han, J.; Wang, M.; Davis, R.; Biddy, M.; Tan, E. Life-cycle analysis of integrated biorefineries with co-production of biofuels and bio-based chemicals: co-product handling methods and implications. *Biofuel Bioprod. Bior.* **2018**, *12* (5), 815-833.

(8) Buchspies, B.; Kaltschmitt, M. A consequential assessment of changes in greenhouse gas emissions due to the introduction of wheat straw ethanol in the context of European legislation. *Appl. Energ.* **2018**, *211*, 368-381.

(9) Ahlgren, S.; Björklund, A.; Ekman, A.; Karlsson, H.; Berlin, J.; Börjesson, P.; Ekvall, T.; Finnveden, G.; Janssen, M.; Strid, I. Review of methodological choices in LCA of biorefinery systems - key issues and recommendations. *Biofuel Bioprod. Bior.* **2015**, *9* (5), 606-619.

(10) Liu, W.; Wang, J.; Richard, T. L.; Hartley, D. S.; Spatari, S.; Volk, T. A. Economic and life cycle assessments of biomass utilization for bioenergy products. *Biofuel Bioprod. Bior.* **2017**, *11* (4), 633-647.

(11) European Commission. *Directive of the European Parliament and of the Council on the Promotion of the Use of Energy from Renewable Sources (Recast)*; 2017.

(12) European Union. Directive 2009/28/EC of the European Parliament and of the Council of 23 April 2009 on the promotion of the use of energy from renewable sources and amending
and subsequently repealing Directives 2001/77/EC and 2003/30/EC. *Off. J. Eur. Union* **2009**, *140*, 16-62.

(13) Buchspies, B.; Kaltschmitt, M. Life cycle assessment of bioethanol from wheat and sugar beet discussing environmental impacts of multiple concepts of co-product processing in the context of the European Renewable Energy Directive. *Biofuels* **2016**, *7* (2), 141-153.

(14) Oconnell, A.; Edwards, R. Personal communication in RED rules with researchers from Joint Research Centre, European Commission. In 2018.

(15) Unnasch, S. *Avoided Life Cycle GHG Emissions from MSW Disposal, Life Cycle Associates, Report No. LCA6060.120.2015*; 2015.

(16) McKechnie, J.; Zhang, Y.; Ogino, A.; Saville, B.; Sleep, S.; Turner, M.; Pontius, R.; MacLean, H. L. Impacts of co-location, co-production, and process energy source on life cycle energy use and greenhouse gas emissions of lignocellulosic ethanol. *Biofuel Bioprod. Bior.* **2011**, *5* (3), 279-292.

(17) D'Avino, L.; Dainelli, R.; Lazzeri, L.; Spugnoli, P. The role of co-products in biorefinery sustainability: energy allocation versus substitution method in rapeseed and carinata biodiesel chains. *J. Clean Prod.* **2015**, *94*, 108-115.

(18) HM Revenue & Customs. *Policy paper- Landfill Tax: increase in rates*; April 2019, 2018.

(19) Kalogo, Y.; Habibi, S.; MacLean, H. L.; Joshi, S. V. Environmental Implications of Municipal Solid Waste-Derived Ethanol. *Environ. Sci. Technol.* **2007**, *41* (1), 35-41.

(20) Jeswani, H. K.; Azapagic, A. Assessing the environmental sustainability of energy recovery from municipal solid waste in the UK. *Waste Manage.* **2016**, *50*, 346-363.
(21) European Commission. Biofuels. <https://ec.europa.eu/energy/en/topics/renewable-energy/biofuels>, (accessed January 2018).

(22) Department for Transport. Renewable Transport Fuel Obligation Annual Report 2016-17. <https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/695185/rtfo-annual-report-2016-2017-web.pdf>, (accessed November 2018).

(23) International Organization for Standardization. ISO 14040: Environmental Management: Life Cycle Assessment: Principles and Framework. 2006.

(24) International Organization for Standardization. ISO 14044: Environmental Management, Life Cycle Assessment, Requirements and Guidelines. 2006.

(25) Stocker, T.; Qin, D.; Plattner, G.; Tignor, M.; Allen, S.; Boschung, J.; Nauels, A.; Xia, Y.; Bex, B.; Midgley, B. IPCC, 2013: climate change 2013: the physical science basis. Contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change. 2013.

(26) Department for Environmental Food and Rural Affairs (Defra). Digest of Waste and Resource Statistics – 2017 Edition. <https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/607416/Digest_of_Waste_and_Resource_Statistics__2017_rev.pdf>, (accessed August 2018).

(27) Department for Environmental Food and Rural Affairs (Defra). Statistics on waste managed by local authorities in England in 2016-17. <https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/664594/LACW_mgt_annual Stats_Notice_Dec_2017.pdf>, (accessed November 2018).
(28) Wernet, G.; Bauer, C.; Steubing, B.; Reinhard, J.; Moreno-Ruiz, E.; Weidema, B. The ecoinvent database version 3 (part I): overview and methodology. *Int. J. Life Cycle Ass.* **2016**, *21*(9), 1218–1230.

(29) Meng, F.; Ibbett, R.; de Vrije, T.; Metcalf, P.; Tucker, G.; McKechnie, J. Process simulation and life cycle assessment of converting autoclaved municipal solid waste into butanol and ethanol as transport fuels. *Waste Manage.* **2019**, *89*, 177-189.

(30) Wilson Bio-Chemical. &lt;http://wilsonbio-chemical.co.uk/the-wilson-system/&gt;, (accessed December 2017).

(31) WRAP (Waste & Resources Action Programme). *Composition of plastic waste collected via kerbside*; 2018.

(32) Shonfield, P. LCA of management options for mixed waste plastics. *WRAP, UK* **2008**.

(33) Gabi. Gabi Extension Database VII Plastics. In 2014.

(34) Flanagan, J.; Davies, M. Glass recycling–life cycle carbon dioxide emissions. *Sheffield: British Glass, 45p 2003*.

(35) Digest of UK Energy Statistics (DUKES). DUKES 2016 Chapter 5: Electricity. In 2017.