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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

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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 $-566 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$ (under US policies that employ system expansion approach), to $+86 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$ and $+23 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$ (under initial and current EU policies that employ energy-based allocation), relative to gasoline emissions of $+94 \text{ gCO}_2\text{eq.}$ 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

42 product. LCA frameworks employed in existing policies which were developed to
43 principally consider crop- and agriculture/forestry residue-based biofuels. These
44 approaches face challenges in evaluating biofuels produced from more complex,
45 mixed waste feedstock streams and in accounting for interactions between the waste
46 treatment and energy sectors.

47 LCA plays a central and quantitative role in global policies aimed at reducing GHG
48 emissions of transport fuels. In the EU, the Fuel Quality Directive regulates a minimum
49 of 6% reduction of the life cycle GHG intensity of transport fuels by 2020 compared to
50 2010 level, which can be achieved through the use of biofuels as one means.¹ In order
51 to be considered as renewable biofuels, life cycle GHG emissions must be at least
52 50% lower than from the fossil fuel they replace and 60% for newer installations from
53 January 2018. Similar thresholds are present in US policy: the Energy Independence
54 and Security Act (EISA) requires biofuels to achieve a life cycle GHG reduction
55 threshold as compared to a 2005 petroleum baseline for different types of biofuels
56 (e.g., 60% reduction for cellulosic biofuel, 50% reduction for advanced biofuel from

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⁻

¹⁰ 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

72 methodology and have identified that this approach risks underestimating the
73 environmental benefit of biofuel systems by ignoring co-product use and
74 corresponding displacement of production elsewhere (e.g.,⁷), particularly if co-
75 products do not have an energy content and therefore cannot be allocated an
76 environmental impact under the prescribed allocation method.¹³ This limitation is
77 particularly relevant for waste-based biofuels, the production of which may encourage
78 recovery of materials with no energy content (e.g., scrap metal and/or glass for
79 recycling). Further, wastes are attributed zero GHG emissions;^{11, 12} as such, avoided
80 emissions due to diverting wastes from conventional treatment routes (e.g., landfill)
81 are not credited to the biofuel product.¹⁴ In contrast, the US EISA and North American
82 LCFS policies employ a partially consequential LCA methodology that aims more to
83 evaluate the change in GHG emissions arising from adoption of alternative fuels.
84 These policies employ system expansion to deal with multiple products, wherein the
85 primary fuel product is “credited” with avoided emissions by assuming that co-products
86 would displace production elsewhere in the economy. Further, benefits of avoided

87 waste treatment processes, such as landfilling, are also credited to the biofuel product
88 (e.g.,¹⁵). With credits from co-products considered, biofuels can in some cases be
89 attributed with negative emissions: credits from co-products exceed the total
90 emissions associated with producing and using the fuel (e.g.,^{16, 17}). Such results can
91 be misleading, as the assessed biofuels do not achieve an absolute reduction of
92 atmospheric GHGs, but rather a relative reduction in GHG emissions considering the
93 production displaced by co-products. For waste-derived biofuels, such distortions may
94 be amplified given the potential for a wider range and greater quantity of co-products.
95 Overall, while existing policies on the surface have similar GHG emissions thresholds,
96 fuel eligibility is dependent on the specific assessment methodologies employed.
97 Ultimately, these methodologies diverge in terms of the “question” they are asking,
98 and therefore whether fuels are evaluated in terms of the overall environmental
99 impacts of the system producing biofuels, or a share of impacts that can be directly
100 attributed to the fuel product in isolation.

101 Waste-based biofuels can provide policy-relevant benefits beyond provision of low-
102 carbon transport fuels. By diverting waste feedstocks from conventional treatment
103 routes (landfilling; incineration), the high cost of disposal by these routes can be
104 avoided. This is particularly relevant in jurisdictions such as the UK where landfill tax,
105 currently £91.35/tonne, or approximately \$120 USD/tonne,¹⁸ greatly increases the cost
106 of disposal by this route. Waste utilisation for fuel production can also encourage the
107 recovery of other materials (e.g., scrap metal, plastic for recycling), and avoiding
108 significant GHG emissions associated with landfilling biogenic wastes (e.g.,¹⁹) or
109 incinerating plastic-based wastes (e.g.,²⁰).

110 Specific support for waste-derived biofuels varies greatly between regions. The EU
111 RED requires 10% renewable energy share in transport fuel consumption by 2020. A
112 cap limiting first-generation biofuels²¹ to 7% share indirectly supports second- and
113 third- generation fuels from non-crop feedstocks, including waste-based biofuels.. In
114 the UK, the Renewable Transport Fuel Obligation provides a stricter limitation on crop-
115 based fuels and further incentivises waste-based fuels by awarding double Renewable

116 Transport Fuel Certificates (RTFC) per litre of liquid renewable fuels derived from
117 certain waste or residue feedstocks; these credits are tradeable and have a market
118 value of £0.18 to £0.24 per RTFC,²² or approximately \$0.25 to \$0.30 USD per RTFC,
119 thus financially supporting waste-based fuels. Under the US EISA and California
120 LCFS, there is no specific support for waste-derived fuels.

121 Waste-based biofuel production systems are complex to evaluate due to their cross-
122 sector interactions (waste and energy/transport sectors) and the wide range of
123 potential co-products. In addition to producing a fuel output, any system producing
124 biofuels from wastes may: 1) avoid current waste treatment processes; 2) enable the
125 recovery of recyclable materials; and 3) co-produce other energy outputs (e.g., excess
126 electricity; heat; fuels). For policies to be comprehensively informed, and for business
127 to make appropriate decisions in response to policies, an appropriate LCA framework
128 is needed to account for this complexity. Therefore, we have developed a case study
129 to explore the implications of LCA methodology decisions on assessed GHG
130 emissions and primary energy demand and to reflect on how these varying model

131 outputs are capable of answering different questions about waste-based biofuels. The
132 case study considers a MSW to acetone-butanol-ethanol conversion process based
133 on an autoclave mechanical heat treatment process and subsequent fermentation of
134 the biomass fibre to liquid biofuels (butanol, ethanol) and other co-products.
135 Alternative system boundaries and allocation approaches are applied in the context of
136 LCA frameworks within EU and US policies. The results are compared and integrated
137 to more meaningfully inform policymakers and industry on the net GHG implications
138 of waste to biofuel systems.

139 2 Methods

140 In this study, we compare life cycle methodologies to evaluate waste-to-biofuels
141 systems and consider how information from the differing approaches can help to
142 inform decision-making. We map these methodology decisions to current and recent
143 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)

158 2) EU RED I (original policy): Avoided waste treatment excluded; electricity co-
159 product evaluated with system expansion (credit to primary biofuel product);
160 all other co-products evaluated by energy allocation (see Section 1.1.2.1 in
161 SI)

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

165 4) Mass-based allocation alternative: Avoided waste treatment may or may not
166 be included; all co-products evaluated by mass-based allocation (see Section
167 1.1.3 in SI)

168 5) Economic allocation alternative: Avoided waste treatment may or may not be
169 included; all co-products evaluated by economic value allocation (see Section
170 1.1.4 in SI)

171 The LCA models are developed in GaBi 8.2 using Ecoinvent 3.3 inventory
172 databases, following ISO Standards 14040 and 14044.^{23, 24} 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 CO₂ equivalents (CO₂ 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%).²⁶ 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.²⁷ 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.^{20, 28} 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.¹⁹

2.3 Waste – to – biofuel process

The MSW-to-biofuel production process has been previously modelled based on a demonstration plant operation²⁹ and further details on the process are available in the

211 Supporting Information (SI). The system starts with the pretreatment autoclave
212 process, based on a working facility developed by Wilson BioChemical.³⁰ MSW is input
213 to the autoclave and treated with steam at moderate temperature (160°C for two
214 hours). The organic content of the MSW is converted into a biomass fibre within the
215 autoclave, which is then recovered to be used as feedstock in ABE production via
216 enzymatic hydrolysis and fermentation. Energy recovery from unconverted biomass
217 and biogas generated in wastewater treatment is sufficient to provide all heat and
218 power requirements of the integrated autoclave/biofuel production process, with
219 excess electricity exported to the grid. Recyclable material streams (ferrous & non-
220 ferrous metals, glass, plastic, wood and textiles) are sterilised within the autoclave and
221 separated from the output stream for subsequent material recovery. All remaining
222 material is classed as waste and sent to incineration/landfill at the same proportions
223 as current waste treatment (see Section 2.2).

224 Biogenic fibre derived from MSW differs significantly from more conventional biofuel
225 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

241 materials require further processing before they displace alternative production in a
242 market; these downstream processes to convert scrap to saleable materials are
243 included in the model. Plastic waste, of average composition³¹ is input to a mechanical
244 recycling process to recover, per 1000kg input, 236 kg PET, 63 kg PP, 122 kg PE, and
245 1 kg PVC.³² Recovered materials are assumed to displace primary production.
246 Unrecyclable materials (films, wastes and residues, 580kg) are disposed of by
247 incineration (71%) and landfill (29%).²⁷ For metals recycling, we use inventory data
248 from Gabi and Ecoinvent database.^{28, 33} Glass is assumed to be recovered to replace
249 aggregates and result in negligible net change in GHG emissions and PED.³⁴

250 The EU RED methodologies are based on allocating impacts between primary and
251 co-products on an energy content basis. The original policy, EU RED I, requires
252 allocation based on the lower heating value of the products, with the exception of
253 excess electricity which addressed by system expansion. For the EU RED I scenario
254 we assume co-product electricity displaces average UK grid generation. EU RED II
255 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				
	Biogenic	Non-biogenic	Butanol and ethanol	Acetone	Hydrogen	Electricity and heat	Recovered plastic, metal, glass
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)	(allocated at autoclave)
Economic value	-	-	23.8%	6.1%	2.3%	48%	37.6%

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 gCO₂eq./MJ_{biofuel}). Excluding avoided waste treatment would still result in very low GHG emissions under system expansion approach (11 gCO₂eq./MJ_{biofuel}) as a result of significant co-product credits for electricity export and metals recovery (-166 and -202 gCO₂eq./MJ_{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

288 $\text{gCO}_2\text{eq./MJ}_{\text{biofuel}}$) as those associated with avoided primary plastic production (-446
289 $\text{gCO}_2\text{eq./MJ}_{\text{biofuel}}$) , assuming 100% displacement with recovered plastics. If recycled
290 plastics are used in other markets, due to their potentially reduced quality relative to
291 primary plastics, this co-product credit may moderately decrease.³² Co-products of
292 acetone and hydrogen only contributes to 4% of the total credits (-30.34
293 $\text{gCO}_2\text{eq./MJ}_{\text{biofuel}}$) (see Figure S7). It is noted in this case study, the relatively low sugar
294 yield by hydrolysis and correspondingly low biofuel yield results in larger quantities of
295 residual biomass available for co-product electricity production than with conventional
296 feedstocks. Major GHG emissions sources arise from the manufacture of enzymes
297 ($187 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$), included in the total biorefinery emissions indicated in Figure
298 2a (also see Table S6). Other process inputs (pH control; fermentation nutrients;
299 microorganism) have smaller impacts, totalling $20.87 \text{ g CO}_2\text{eq./MJ}_{\text{biofuel}}$. Treatment of
300 residual waste from autoclave has a large GHG emission of $141.01 \text{ g CO}_2\text{eq./MJ}_{\text{biofuel}}$.
301 Collection and transport accounts for about 4% while fuel distribution and use
302 accounts for less than 1% of the total PED and GHG emissions (see Figure S7 in the

SI). 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 $86 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$, achieving only a minor reduction of 9% relative to gasoline and therefore would not qualify as an eligible biofuel under the policy. Enzyme production²⁹ 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 $102 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$ credit, only $31 \text{ gCO}_2\text{eq.}$ is credited to the biofuel product. Thus,

318 although the production of biofuels from MSW would achieve significant overall GHG
319 reductions when all products are considered, this pathway would not be eligible under
320 the original RED policy.

321 In contrast, under the revised RED II policy, the MSW-derived biofuels would be
322 eligible, with overall GHG emissions of 23 gCO₂eq/MJ_{biofuel}, a reduction of 75%. With
323 exergy allocation applied to the co-product electricity and heat, a large share of
324 biorefinery emissions (86%) are applied to these outputs; correspondingly, fewer
325 emissions are attributed to the biofuel product. Excess electricity is attributed with
326 GHG emissions of 86 gCO₂eq/MJ, which represents a 12% reduction compared to UK
327 grid electricity mix³⁵ (see Table S5 in the SI). Enzyme production still contributes the
328 largest share of GHG emissions attributed to the biofuel outputs (68%).

329 By excluding avoided waste treatment impacts, the RED I and RED II policies ignore
330 an important service provided by waste valorisation systems of diverting and treating
331 waste that would otherwise be destined to landfill/incineration. Inclusion of avoided
332 waste treatment would reduce the GHG emissions assessed under RED I and RED

II. Apportioning credits related to avoided landfilling and incineration processes would result in net GHG emissions of $-22 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$ and $8 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$ 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 $9.2 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$, or $-0.4 \text{ gCO}_2\text{eq./MJ}_{\text{biofuel}}$ 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 gCO₂eq./MJ_{biofuel}, or -81 gCO₂eq./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_{biofuel} (total biorefinery demands 3.50 MJ/MJ_{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_{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

389 for all waste treatment processes to understand the total impacts of the treatment
390 process and product outputs (including liquid biofuel use in place of gasoline). We
391 compare current case study results with conventional treatment processes: landfill
392 without biogas recovery, landfill with biogas recovery and incineration for energy
393 recovery (electricity only or CHP generation) (Figure 3). MSW conversion to liquid
394 biofuels is the superior option for both categories, achieving significantly greater
395 reductions in GHG emissions and PED than conventional waste treatment routes.
396 Without landfill gas capture, sanitary landfill operation emits the highest GHG
397 emissions of 573 kg CO₂eq/t MSW; with landfill gas capture, the GWP of landfilling for
398 the electricity only and CHP options are 240 and 223 kg CO₂eq/t MSW, respectively.
399 Increasing capture rate of biogas has been reported to be key in reducing the GWP of
400 the landfill option²⁰. In comparison, incineration is a net source of GHG emissions, as
401 fossil CO₂ emissions, largely from plastics combustion, exceed avoided emissions
402 associated with displacing heat and electricity production elsewhere. This results in
403 emissions of 174 and 58 kg CO₂eq/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).

Figure 3 Life cycle primary energy demand (left axis) and global warming potential (right axis) comparing MSW-derived liquid biofuels with landfill and incineration options.

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

426 policy makers have to select one allocation method that is most appropriate for the
427 analysis of biofuel systems. To do so requires careful consideration, as alternative
428 allocation methods are ultimately answering very different questions.

429 System expansion aims to understand the overall impact of introducing a new
430 product system. This approach benefits from its comprehensiveness in evaluating the
431 overall impact of the product system, but is based on a clear identification of biofuels
432 as the primary product and all other outputs as secondary. Where there are large co-
433 product outputs, associated credits can distort the results and risk not reflecting
434 stakeholder values or decision criteria. In the current study, significant electricity and
435 recovered metal co-products contribute to very negative emissions; avoided waste
436 treatment benefits are also solely attributed to the primary product. Whether it is
437 appropriate to consider liquid biofuels as a primary product is questionable, given that
438 this output represents only 24% of the financial value of system outputs, and
439 substantially less on energy (2.6%) and mass (1.7%) bases.

440 An alternative approach to evaluate the overall impacts of waste conversion to
441 biofuels may be from the perspective of a waste treatment system – using a functional
442 unit of one tonne MSW, or equivalent – and thus taking into account all of the diverse
443 outputs of the system without having to artificially identify a single primary product.
444 Such results are not useful in biofuels policies that require a specific impact be
445 attributed to the biofuel product. However, such a framework could be appropriate for
446 a waste treatment sector-focused approach to evaluating and supporting higher value
447 products from waste (biofuels, bulk and high value chemicals, others), while
448 concurrently supporting diversion from conventional treatment routes.

449 In contrast, allocation approaches aim to attribute impacts to a specific, single
450 product. Allocation is, in theory, effective at isolating the impact of biofuel products
451 from the other outputs of the waste biorefinery system. However, the diversity of
452 products poses a challenge, as some cannot be addressed with energy allocation
453 (e.g., recovered metal, glass), and others cannot be addressed by mass allocation
454 (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

470 use change arising from crop-based biofuels), but should be pursued in future work to
471 ensure that the contexts of the energy and waste sectors are properly considered. At
472 present, by excluding the benefits of diverting wastes from landfill and incineration, EU
473 policy disadvantages biofuel production relative to other, lower-value uses of waste
474 streams.

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

482 **Associated Content**

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

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

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504 **Notes**

505 The authors declare no competing financial interest.

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