

15 **Abstract**

16 In 2015/2016, the total municipal solid waste (MSW) collected by local authority in the U.K.
17 was 26 million tonnes and over 57% is still put into landfill or incinerated. MSW is a promising
18 feedstock for bio-butanol production as it has a high lignocellulosic fibre content such as paper,
19 wood, and food waste, about 50 wt% of a typical MSW stream. The study evaluates acetone,
20 butanol, ethanol and hydrogen production from autoclaved municipal solid waste feedstock.
21 Life cycle assessment is undertaken to evaluate the acetone, butanol, ethanol and hydrogen
22 production process, considering cogeneration of heat and power from residual biogenic waste
23 based on experimental data and process modelling. Acetone, butanol, and ethanol product yield
24 can be achieved at 12.2 kg butanol, 1.5 kg ethanol, 5.7 kg acetone, and 0.9 kg hydrogen per
25 tonne MSW. The product yield is relatively low compared to other lignocellulosic feedstocks
26 primarily because of the lower hydrolysis yield (38% for glucose) achieved in this study;
27 however, hydrolysis yields could be improved in future optimisation work. The conversion
28 shows a net primary energy demand of -1.11 MJ/MJ liquid biofuels (butanol and ethanol) and
29 net greenhouse gas emission of -12.57 g CO₂eq/MJ liquid biofuels, achieving a greenhouse gas
30 reduction of 115% compared to gasoline comparator.

31 **Keywords**

32 Municipal solid waste, Waste autoclaving, Enzymatic hydrolysis, ABE fermentation, Life
33 cycle assessment

34 **1 Introduction**

35 The EU transport sector accounted for 25.8% of the total greenhouse gas (GHG) emissions
36 in the EU in 2015 at 1.05 Gt/yr including international aviation and maritime emissions
37 (European Environment Agency, 2017). The EU's climate change targets have already stated
38 transport emissions must be cut by 60% by 2050 compared with 1990 levels (The European
39 Union, 2014). Renewable energy sources can contribute to climate change mitigation through
40 the reduction of GHG emissions and achieve sustainable development, driving the increased
41 demand for renewable fuels. The EU issued the Renewable Energy Directive (RED), for
42 instance, requires that renewable energy content should account for at least 10% of the energy
43 used in transportation by 2020 (European Commission, 2018).

44 Butanol (C_4H_9OH) is an attractive renewable liquid transport fuel. Its superior properties
45 have been well documented: butanol fits the existing fuel infrastructure as it can be stored in a
46 mixture with traditional gasoline and diesel at a varied ratio; it has a better energy density (30%
47 greater than ethanol) and combustion performance due to similar air: fuel ratio to that of
48 gasoline than ethanol and can be used in higher quantities in a standard petrol engine (up to
49 20% without any engine modifications) but ethanol is limited to 15% (Wu et al., 2007); and
50 exhibits low solubility in water which reduces the risk of groundwater contamination from
51 unintended release. Butanol thus has the potential to substitute both ethanol and biodiesel in
52 the biofuel market to reach \$185.3 billion in 2021 as estimated by Pike Research (Microbiology
53 Society, 2013). In addition to being a potential biofuel, butanol is also a valuable C4 feedstock
54 for chemical synthesis (e.g., methacrylate esters, butyl glycol ethers, butyl acetates, and
55 plasticizers) and an industrial solvent or co-solvent for surface coatings (Bankar et al., 2013).
56 However, challenges still need to be addressed, such as high feedstock costs and associated
57 high operational cost requiring cheaper and sustainable feedstocks.

58 Municipal solid waste (MSW) is a promising feedstock for butanol production as it has a
59 high lignocellulosic fibre content such as paper, wood, and food waste, about 50 wt% of a
60 typical MSW stream. Unlike other cellulosic feedstocks, MSW has low/negative feedstock
61 prices, immediate availability, and high potential in reducing GHG emissions of biofuel
62 production compared to first and second generation biofuels from food crop and agricultural
63 or forest residue wastes. Further, diverting the organic content of MSW for biofuel production
64 is also beneficial in addressing MSW waste management issues (Gharfalkar et al., 2015;
65 Jeswani and Azapagic, 2016). In 2015/2016, the total MSW collected by local authority in the
66 U.K. was over 26 million tonnes and over 57% is still put into landfill or incinerated
67 (Department for Environmental Food and Rural Affairs (Defra), 2017b; Science and
68 Technology Select Committee, 2014). The gate fee charged by landfilling in the UK reveal a
69 cost of £19/tonne excluding landfill tax and £102/tonne including landfill tax, while tipping
70 fees for incineration are £86/tonne in 2015/2016 (WRAP (Waste & Resources Action
71 Programme), 2016). Therefore, the production of biofuel from MSW is extremely attractive in
72 terms of environmental and economical perspective.

73 Life cycle assessment (LCA) provides a transparent methodology that can be used to
74 examine lignocellulosic biofuel production as it examines the environmental burdens over the
75 entire life, from production, through use and on to disposal or recycling (McKechnie et al.,
76 2011). Existing LCAs of biofuel production (e.g., ethanol) from various feedstocks including
77 corn stover, wheat straw, poplar, eucalyptus and waste papers amongst others have been widely
78 reported in the literatures (Borrion et al., 2012; Michael et al., 2012; Mohammad et al., 2013;
79 Zhang et al., 2010b). Few studies by Chester & Martin (Chester and Martin, 2009), Schmitt et
80 al. (Schmitt et al., 2012) and Kalogo et al. (Kalogo et al., 2007) evaluated ethanol production
81 from MSW via dilute acid pretreatment and hydrolysis and presented the life cycle

82 environmental impacts of the conversion. It showed MSW derived ethanol can reduce energy
83 use and GHG emissions compared to gasoline and other cellulosic ethanol production. But
84 environmental impacts are highly dependent on conversion technology, process conditions
85 used, and waste classification and its overall impact of MSW to ethanol is limited due to
86 availability of MSW.

87 Autoclaving is a new pretreatment technology replacing conventional dilute acid
88 pretreatment to process unsorted MSW. Prior LCA study considered autoclaving unsorted
89 MSW with subsequent composting in tunnels/in confined windrow/ in turning windrow, or
90 anaerobic digestion of biogenic fibres (Quirós et al., 2015). The results showed that autoclaving
91 with sorting, digesting anaerobically and composting had the lowest environmental impact
92 values for eutrophication and global warming potential. However, there is no consideration of
93 higher value uses for this biogenic fibre materials, such as liquid biofuels (e.g., butanol).

94 To address the gaps in technical and environmental aspects of acetone, butanol, ethanol
95 (ABE) production from pretreated MSW, in this study, we develop process models for ABE
96 production from autoclaved MSW feedstock. The overall technical performance and
97 environmental impacts (i.e., primary energy demand and global warming potential) are
98 evaluated across the integrated processes, including autoclave, hydrolysis, fermentation, and
99 distillation.

100 **2 Method**

101 The study models in detail the production of ABE and hydrogen from autoclaved municipal
102 solid waste feedstock. The overall process design converts MSW into ABE and hydrogen by
103 autoclave, hydrolysis, fermentation and distillation. Product recovery, energy recovery,
104 wastewater treatment and utilities are also included in the design. The recovery of non-biogenic
105 content of MSW after autoclave, including plastics, metal, and glass materials, are excluded in

106 this study. The input and output mass and energy flow of the system are extracted from
107 experimental data and process modelling as discussed below. Inventory data is also
108 supplemented by Ecoinvent database (Wernet et al., 2016) and literature data where available,
109 e.g., enzyme production for hydrolysis (Nielsen et al., 2006) (see Table 1). The LCA is
110 undertaken in GaBi 8.2 (2017) using Ecoinvent 3.3 inventory databases. Two environmental
111 impacts are quantified: primary energy demand (PED) in terms of MJ and global warming
112 potential (GWP), based on the most recent IPCC 100-year GWP factors to quantify GWP in
113 terms of CO₂ equivalents (CO₂ eq.) (Stocker et al., 2013). Direct CO₂ emissions from the
114 fermentation and combustion of butanol and ethanol fuel are excluded as the emitted carbon
115 from renewable biomass resources can be counted as “carbon neutral”. Avoided emissions
116 from diverting waste to butanol and ethanol production from other conventional waste
117 treatment routes (e.g., landfill, incineration) are also excluded.

118 **2.1 Scope and functional unit**

119 We develop an LCA model of MSW-ABE following the ISO Standards 14040 and 14044
120 (International Organization for Standardization, 2006a, b). The functional unit is defined as
121 one MJ of liquid biofuels (butanol and ethanol)) used as a reference to quantify all inputs and
122 outputs of the process steps. A schematic process flow diagram defining the system boundaries
123 is shown in Fig. 1. The system boundary begins with the sorting and separation of MSW
124 (energy use and environmental burdens of the processes and products generating MSW are
125 excluded from the study) and ends with the combustion of the fuel in a light duty vehicle.
126 System expansion method is used to consider the co-products’ benefits where liquid biofuels
127 (butanol and ethanol) are the main product and acetone, hydrogen, and excess electricity are
128 considered as co-products thereby allocating their impacts to main product liquid biofuels.

129

130 **2.2 Waste composition**

131 The waste composition used is representative of the UK MSW with the following wet
132 composition by mass: paper and cardboard (22%), food waste (17%), wood (8.7%), plastic
133 (22%), glass (1%), garden waste (3%), metals (4%), textiles (6.6%) and others (15.7%)
134 (Department for Environmental Food and Rural Affairs (Defra), 2017a).

135 **2.3 Life Cycle inventory**

136 **2.3.1 Autoclave Pretreatment**

137 The autoclave system is a mechanical heat treatment process developed by Wilson Bio-
138 Chemical (Wilson Bio-Chemical, 2017), based on existing autoclaving and steam boiler
139 technologies. The autoclave is a front-loading and rotating pressure vessel in which the MSW
140 is treated with high pressure saturated steam in a batch process. The organic fraction is broken
141 down to a cellulose-rich fibre, which has a high sugar composition (40–50%) and thus could
142 be suitable for biofuel production via fermentation. Untreated wastes such as plastic, glass,
143 textiles and metals are sterilised and able to be recovered post-autoclave.

144 Based on the best performance running of the autoclave process, processing parameters of
145 160°C for two hours for each batch have been determined. Thus the energy requirement of 43
146 MJ electricity and 274 MJ natural gas and 245 L water is determined from plant operation to
147 pretreat one tonne of MSW (Wilson Bio-Chemical, 2017).

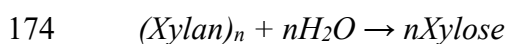
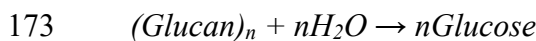
148 **2.3.2 Hydrolysis**

149 The waste stream has been subjected to high temperature processing and this is similar to the
150 pretreatment normally required to overcome the inherent recalcitrance of biomass feedstocks
151 to subsequent enzymatic saccharification to sugars. Preliminary studies have previously
152 demonstrated that between 30-40% of the fibre sugar can be released by subsequent enzyme
153 hydrolysis (Ibbett, 2018). A commercially available enzyme cocktail - Novozymes Cellic

154 CTec2 (Novozymes, 2018) - is used in this study. Samples of the MSW fibre are milled to a
 155 consistent particle size (0.5mm) and then loaded into the hydrolysis vessel where it is diluted
 156 with water from the mains water tank into a dilute slurry (20% solids assumed in this study but
 157 a range of 20–30% solids are evaluated in the sensitivity analysis). At this point, adjustments
 158 may also be made to process conditions such as pH. The slurry is then dosed with an enzyme
 159 solution (5% wt/wt) (15–60 filter paper units (FPU) per gram cellulase) which is stored in its
 160 own separate storage tank, causing hydrolysis of the biogenic fibre to a solution of fermentable
 161 sugars. There will also be solids remaining in suspension which are either un-hydrolysable or
 162 have not been fully hydrolysed. All of this mixture is transferred downstream to the filtration
 163 unit. The efficiency of hydrolysis is assessed by monitoring glucose release into the media,
 164 over a period of 48h at the temperature of 50°C, by high pressure ion chromatography.

165 The composition of biogenic fibre transported from the Wilson System as stated above can
 166 be evaluated by analysis of the monosaccharides liberated by total acid hydrolysis of the
 167 polysaccharides present in the fibre cell-wall matrix (Ibbett et al., 2011). This method gave a
 168 monosaccharide content as follows: glucose (40-45%), xylose (4-5%), galactose (0.7%) and
 169 arabinose (2.9%), as shown in Fig. 2, where glucose is mainly derived from cellulose and the
 170 other three monosaccharides are derived from hemicellulose.

171 The net enthalpy change for each reaction (see the reactions outlined below) is calculated
 172 using the heat of formation (ΔH_f) (Humbird et al., 2011), see eq (1) as below.



$$Q_{reaction} = \sum \frac{\Delta H_f \times (m_b cy)}{M_w} \quad (1)$$

175 where ΔH_f is the heat of formation (kJ/mol), m_b is the number of moles of sugar formed from
176 1 kg MSW fibre (mol), c is the sugar composition (%), y is sugar hydrolysis conversion yield
177 (%), M_w is the molecular weight of the sugar (kg/mol).

178 Mixing is achieved by an agitator and heat control is achieved by a centrifugal pump which
179 pumps the hydrolysate around a loop through a heat exchanger cooled with cooling water, as
180 is assumed in previous biorefinery models (Humbird et al., 2011).

181 **2.3.3 Fermentation**

182 After hydrolysis, the sugary solution is adjusted (as required) with antifoam, pH altering
183 agents and other required additions (such as nutrients) in preparation for fermentation. The
184 microorganism is *Clostridium acetobutylicum* ATCC 824 and the medium of all fermentations
185 contain (per L) KH_2PO_4 1 g, K_2HPO_4 0.76 g, $\text{CH}_3\text{COONH}_4$ 2.9 g (of which CH_3COO^- is 2.2
186 g), yeast extract (Duchefa) 2.5 g, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ 6.6 mg, $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ 1 g, and *p*-aminobenzoic
187 acid (*p*-ABA) 0.1 g. Following this, the vessel is inoculated with a previously prepared
188 *Clostridium* culture from the seed culture vessel. The fermentation is conducted at 37°C over
189 a duration of 48 hours per batch. We assume that the inventory data of producing *Clostridium*
190 *acetobutylicum* would be similar to that of producing *Z. mobilis* as in the design (Dunn et al.,
191 2012; Humbird et al., 2011). Inventory data of nutrients are obtained from publicly available
192 data (Adom and Dunn, 2015; Edwards, 2016).

193 Fermentation then proceeds, producing ABE as well as hydrogen and CO_2 . As the butanol
194 concentration builds, it inhibits the growth of the *Clostridium*. Therefore, nitrogen gas stripping
195 is used to selectively remove the ABE which escapes as a vapour to the condenser along with
196 CO_2 , hydrogen, nitrogen and some water vapour. The hydrogen and CO_2 are collected during
197 the fermentation and purified by Pressure swing adsorption in the subsequent distillation stage.
198 Totally, ABE fermentation yield can be achieved at 33% of which has been validated

199 experimentally by a 30% yield and hydrogen yield is 1.6% based on molar ratio shown in Table
200 2. The relative ratio of produced solvents acetone, butanol and ethanol is 28%, 62% and 10%.

201 The reactions represent a qualitative measure rather than stoichiometric quantitative
202 relationships for the ABE conversion process. Based on eq (1) above, reaction energy shows
203 the conversion is mildly exothermic; as such, we assume fermentation unit does not require
204 thermal management (i.e., heat loss to surroundings is balanced by heat release from
205 fermentation).

206 **2.3.4 Distillation**

207 The acetone, butanol, ethanol and water vapours are condensed in the beer column and are
208 subsequently transferred to rectification column for separation. The un-condensable hydrogen,
209 nitrogen and CO₂ continue through the condenser to pressure swing adsorption. Pressure swing
210 adsorption technology (HyGear, 2017) is used to purify hydrogen from the gaseous mixture
211 leaving the condenser. This hydrogen can be pressurised and stored in a tank, whilst the
212 remaining N₂ and CO₂ are discarded. The acetone, butanol, ethanol and water are distilled
213 based upon their relative volatilities into pure streams of each component by a gas stripping
214 procedure. This concentrated solution of solvent will then be distilled to obtain the three distinct
215 compounds: butanol, acetone and ethanol to meet the requested biofuel specifications.
216 Recirculation of the stripping gas will reduce the costs. Firstly, acetone is assumed to be
217 extracted in the first distillation column with a purity of 99 wt%. After separation of acetone,
218 the remaining liquid goes through a decanter before going to the second distillation column to
219 separate ethanol which is then dehydrated through the molecular sieve adsorption to a purity
220 of 99.5 wt%. The bottom stream which is mainly butanol and water after separation of ethanol
221 goes to the third distillation for butanol separation (99.7 wt%) (Baral and Shah, 2016; Grisales
222 Díaz and Olivar Tost, 2017). These pure products are finally stored in tanks, ready for

223 distribution or analysis. The waste biofuels and water mixture are assumed to be discharged to
224 the lignin separator and wastewater treatment system mixed with stillage from hydrolysis and
225 fermentation units.

226 **2.3.5 Wastewater Treatment**

227 Wastewater treatment process in previous model (Humbird et al., 2011) is adapted in this
228 study. After hydrolysis, fermentation and distillation, the stillage contains unfermented sugars,
229 organic acids and solid residuals, e.g., lignin. They are sent to a press filter to separate solids
230 (mainly lignin) to the combustor while the liquid fraction is sent to wastewater treatment .
231 Wastewater treatment consists of anaerobic and aerobic digestion treating and recycling the
232 wastewater to minimise the amount of water discharged and fresh water requirement. The total
233 chemical oxygen demand is assumed to be proportional to the solid content concentration in
234 the wastewater and is calculated to be 245 g/L. In anaerobic digestion, 91% of organic content
235 is converted into biogas (86%) and microorganism cell mass (5%). The biogas from the digester
236 has a composition of 51% CH₄/49% CO₂ on a dry molar basis. Methane is produced on the
237 basis of the organic content at a yield of 228 g biogas/kg chemical oxygen demand while the
238 cell mass is produced at 45 g cell mass/kg chemical oxygen demand (Humbird et al., 2011).

239 The liquid after anaerobic digestion is further treated in aerobic digestion where 96% of the
240 remaining soluble organic content is digested, with 76% converting into water and carbon
241 dioxide and 22% producing cell mass. Chemicals such as caustic soda must be added to adjust
242 the pH for digestion. After this step, the liquid is sent to a membrane bioreactor clarification
243 system where the aerobic biomass sludge is separated to sludge centrifuge mixed with
244 anaerobic sludge for dewatering. The centrifuge solid is dewatered and sent to the combustor
245 for energy recovery and the remaining water is recycled to aerobic digester for additional
246 treatment. The clarified water is pumped to the reverse osmosis for salt removal. About 79%

247 water is assumed to pure and recycled to the process and 21% of the water rejected from reverse
248 osmosis is further concentrated in an evaporator system. In the evaporator, about 7% dry brine
249 content (mainly sodium nitrate) is sent for disposal while the concentrated water is assumed to
250 be clean and recycled to the process.

251 **2.3.6 Energy recovery**

252 The lignin from hydrolysis and fermentation residue and biogas and sludge produced from
253 wastewater treatment are fed to the combustor for heat and power generation. A grate
254 incinerator was modelled with electricity and heat production efficiency of 18.7% and 21.6%
255 of the waste's lower heating value, respectively (Veolia, 2012). The cogenerated heat is
256 assumed to be used to meet process heat requirements; the excess could be used for sterilization
257 or for cooling generation via absorption refrigeration plant on site, or exported via a heat
258 network, but such uses are not considered in the present study and excess heat is assumed to
259 not have a practical use. The electricity is used to supply the process and any surplus electricity
260 is assumed to be exported to the grid. The electricity system is assumed to be representative of
261 the UK average mix in 2015, composed of nuclear (20.9%), coal (22.5%), natural gas (29.7%),
262 hydro (1.9%), heavy fuel oil (0.6%), wind (14.2%), other renewables (8.7%), and other (1.4%)
263 (Digest of UK Energy Statistics (DUKES), 2017).

264 **2.4 Sensitivity analysis**

265 **2.4.1 Sensitivity analysis of background systems**

266 Relative environmental performance of MSW derived liquid biofuels depends on reference
267 fuels system, enzyme production and which electricity types (i.e., renewable content of the
268 electricity) the exported electricity is displacing. It arises from regional variability of electricity
269 generation sources and associated impacts. Thus we conducted sensitivity analyses to study the
270 influence of background systems on life cycle emissions of MSW derived liquid biofuels.

271 **2.4.2 Sensitivity analysis of process variables**

272 The overall yield of ABE from MSW depends on the biogenic fibre content of MSW, the
273 conversion efficiencies from cellulose to glucose and xylose during hydrolysis, glucose and
274 xylose yield and glucose and xylose conversion during fermentation. The ultimate ABE
275 concentration is also dependant on the solid concentration during hydrolysis. Distributions are
276 fitted where sufficient data are available (e.g., hydrolysis yields) or assigned based on
277 minimum/maximum values to model parameters. Monte Carlo simulations (Ruth and Jechura,
278 2003; Tu and McDonnell, 2016) enable an investigation into how input uncertainty propagates
279 through the mass and energy balance model and LCA model. These distributions and
280 underlying data sources are summarised in Table 4 for processing parameters including
281 hydrolysis yield and solid content. Triangular distributions are used for these parameters where
282 the peak values – indicating highest probability - are those from experiments, with the
283 probability decreasing linearly until reaching zero at the upper and lower bounds considered.
284 The number of iterations for the Monte Carlo analysis to estimate the probability of different
285 results was set to 10,000 as a general rule of thumb (Barreto and Howland, 2005) using Crystal
286 Ball (Oracle, 2016). Results are shown in Section 3.3.

287 **3 Results and Discussion**

288 **3.1 Material and Energy Balance**

289 Fig. 3 indicates the flows of MSW into the main processing system including mass and
290 energy balance. The total input is one tonne MSW, of which 53% of wet mass is lignocellulosic
291 content. It has a moisture content of 40%, with remaining components broken down by sugar
292 dry content as 45% glucose and 5% xylose as measured experimentally (totally 31.8% dry
293 convertible lignocellulosic content). Sugar levels are key as they are the molecules that react
294 within the hydrolysis and fermentation stages to produce ABE. Hydrolysis yield was previously

295 found to be 38% for glucose from experimental measurement and 70% for xylose. Thus the
296 lignocellulosic feedstock can produce 54.4 kg glucose and 11.1 kg xylose per tonne MSW after
297 hydrolysis reaction. The outputs from the conversion are 12.2 kg butanol (7.6 kg gasoline
298 equivalent on energy basis), 1.5 kg ethanol (0.9 kg gasoline equivalent), 5.7 kg acetone, and
299 0.9 kg hydrogen as well as 0.3 kg acetic acid and 1.0 kg butyric acid. ABE product yield
300 achieved in this study is relatively low compared to other lignocellulosic feedstocks which have
301 been subjected to higher severity hydrothermal deconstruction pretreatments (Ibbett et al.,
302 2011) (3.2 wt% in this study versus 11.8–14.9 wt%, on a dry substrate basis) (Baral and Shah,
303 2016; Montano, 2009), primarily because of the lower hydrolysis yield (38% for glucose)
304 achieved. However, it should be noted that hydrolysis yields could be improved up to 85% and
305 more details are discussed in Section 3.3.2. These mass transfers from MSW to biorefinery
306 products (ABE and hydrogen) are used as the bases of LCA analysis.

307 The energy recovery and process energy (i.e., electricity and heat) required for each step of
308 ABE production system are also shown in Fig. 3, which are obtained from plant operation data
309 and process modelling. Enthalpy of materials in hydrolysis and fermentation depends on the
310 mass and temperature of the materials. The autoclave process is the most energy intensive step
311 accounting for about 42% of the total heat requirement and 41% total electricity requirement.
312 The hydrolysis requires about 32% of the heat requirement and only 0.4% electricity use for
313 mixing compared to 0.7% electricity use for fermentation. About 26% of the total heat and 4%
314 of the total electricity use are required for distillation process for ABE product recovery.
315 Wastewater treatment process also consumes about 54% of the total electricity to recover
316 process water, separate lignin solid, produce biogas via equipment such as water pump, digester
317 blower, anaerobic basin, sludge centrifuge and evaporator.

318 Wastewater produced during the MSW-to-ABE process can be treated into 94% clean water
319 through anaerobic and aerobic digestion. The wastewater treatment unit can produce 389 kg
320 lignin (35% moisture content) and 21 kg biogas and 8 kg sludge (79% moisture content) to the
321 energy recovery unit. Considering the calorific values of lignin, biogas (51% CH₄/49% CO₂),
322 and sludge (16 MJ/kg (Demirbas, 2017), 14 MJ/kg, and 4 MJ/kg (Humbird et al., 2011),
323 respectively), energy recovery unit totally produces 266 kWh electricity and 1108 MJ
324 heat (Veolia, 2012). These energy sources are used to provide energy use for autoclave and
325 ABE biorefinery onsite. Excess electricity can be sold to the grid which can contribute to the
326 grid to reach the target of 15% electricity using renewable sources by 2020 as regulated by the
327 Renewable Obligation produced from (Department for Environmental Food and Rural Affairs
328 (Defra), 2013). Due to lack of integrated district heating system in UK, excess heat is assumed
329 to be discarded to the environment.

330 As shown in Fig. 3, the biogenic portion of MSW has a calorific value of 13018 MJ. For a
331 plant capacity of 150000 t MSW/yr with operating hours of 8000, the total energy content is
332 28.4 MW/yr. The total products have energy values of 3.6 MW/yr from main products of ABE
333 (37.3 MJ/kg butanol, 31.8 MJ/kg acetone and 29.7 MJ/kg ethanol) and hydrogen (141.7 MJ/kg)
334 (The Engineering Toolbox, 2019) and 4.5 MW/yr from excess electricity. Therefore, the total
335 energy efficiency of 28% can be obtained through the MSW derived ABE conversion system
336 (see Table 3). In this study, electricity co-product is higher compared to other feedstocks, as a
337 greater share of biomass is not converted to fuels and thus is available for energy recovery.
338 Excess heat is available and, if it could be utilised (e.g., by co-located industrial process and/or
339 district heating, for sterilization, or for cooling generation) then total energy yield could
340 improve to 37% compared to 47% presented for ethanol production from corn stover (Humbird
341 et al., 2011).

342 3.2 Life Cycle Assessment Results

343 LCA evaluation based on the mass and energy balance data is performed to quantify the life
344 cycle PED and GWP associated with converting one tonne of MSW feedstock into ABE (see
345 Fig. 4). Excluding the impact of further treating the non-biogenic content of MSW, the ABE
346 production from MSW shows a net PED of -1.11 MJ/MJ liquid biofuels (-559.69 MJ/t MSW)
347 and net GHG emission of -12.57 g CO₂eq/MJ liquid biofuels (-6.32 kg CO₂eq/t MSW),
348 including co-products credits of acetone, excess electricity, and hydrogen. MSW to BE can
349 thus achieve better PED as bioethanol produced from agricultural residues (-0.1 – 0.6 MJ/MJ
350 fuel) (Michael et al., 2012), achieving reductions in fossil energy use by 2.3 MJ/MJ fuel
351 compared to conventional gasoline (1.2 MJ/MJ) (Michael et al., 2012). If excess heat could be
352 utilised, additional credits would result in net PED of -2.13 MJ/MJ liquid biofuels and net GWP
353 of -17.56g CO₂eq/MJ liquid biofuels (40% further emission reduction).

354 As seen in Fig. 4, the largest energy and GHG emissions source arising from ABE production
355 is the manufacture of enzymes, contributing approximately 2.31 MJ PED/MJ liquid biofuels
356 and 186.7 g CO₂eq./MJ liquid biofuels. Other process inputs (pH control; *C. acetobutylicum*
357 bacterium) and fermentation nutrients have substantially smaller impacts, totalling
358 approximately 10% of autoclave/biorefinery GHG emissions. Process energy (electricity and
359 heat) related emissions have been avoided through the energy recovery from the unconverted
360 biomass (primarily lignin solids), biogas generated by the wastewater treatment plant and waste
361 sludge. Transport of ABE to depot and to filling station account for less than 1% of the total
362 emission.

363 It is noted that a low glucose hydrolysis yield of 38% has been considered, based on the
364 experimental evidence from this study. However, a lower sugar hydrolysis and ABE yield
365 results in more residual biomass available for co-product electricity production, thereby

366 providing a larger co-product GHG credit when displacing grid electricity. The impacts of these
367 process variables on product yield and environmental impacts are discussed further in Section
368 3.3.

369 **3.3 Sensitivity Analysis**

370 **3.3.1 Sensitivity analysis of background systems**

371 The life cycle GHG emissions per MJ MSW derived liquid biofuels is compared to results
372 of gasoline (the EU fossil fuel comparator, US Environmental Protection Agency (EPA) and
373 US oil shale based on Energy Research Architecture (ERA)), and bioethanol (i.e., ethanol
374 produced from ethanol, sugarcane, corn stover, switchgrass, and miscanthus). Various life
375 cycle GHG emissions are also present for various electricity replacement of electricity mix,
376 electricity from hard coal, natural gas, hydro power, wind power in UK and electricity mix in
377 US and China (see Fig. 5).

378 Overall GHG emission of MSW derived liquid biofuels provides 115%, 114% and 109%
379 reduction compared to the transportation gasoline based on EU RED (84 g CO₂eq/MJ) for the
380 base case, US EPA (93 g CO₂eq/MJ) and US ERA (139 g CO₂eq/MJ) (Pieprzyk et al., 2009),
381 respectively. MSW to bioethanol can give 14.5–56.4 g CO₂eq/MJ ethanol (Chester and Martin,
382 2009; Kalogo et al., 2007; Schmitt et al., 2012; Zhang et al., 2010a); therefore MSW to BE can
383 achieve over 100% relative GHG reduction than MSW to bioethanol only. The difference is
384 primarily because of the different system boundaries (e.g., waste collection and hauling,
385 classification, coproducts considerations, and landfill treatment of residual waste) and
386 assumption (e.g., enzyme production and fermentation microorganism) made in the studies.
387 MSW derived liquid biofuels has smaller GHG emissions than other lignocellulosic ethanol
388 pathways (corn stover, switchgrass, miscanthus) and reduction of over 100% relative to corn
389 and sugarcane (Michael et al., 2012).

390 The GHG emissions assigned to enzyme production in the current study are 5.9 g CO₂eq/g
391 of produced enzyme (commercially Novozymes Cellic CTec2) (Kløverpris, 2018) while some
392 onsite enzyme production process using glucose as a feedstock have been reported to emit 4.1–
393 11.5 g CO₂eq/g cellulose (Hsu et al., 2010; McKechnie et al., 2015). However, the newly
394 produced Cellic 1.0 series (0.64 g CO₂eq/g enzyme) have achieved significant reduction in
395 GHG emissions. We also considered different Novozymes Cellic CTec series enzyme
396 products: Cellic CTec in 2009 (7.25 g CO₂eq/g), Cellic CTec3 in 2012 and 2014 (5.1 g
397 CO₂eq/g), and assumed Cellic X in 2022 (0.32 g CO₂eq/g) but their respective dosage is not
398 described here due to commercial confidentiality. Their relative impacts have been shown in
399 Fig. 5 (MSW- liquid biofuels 2), indicating a range of –135 to 22.13 g CO₂eq/MJ liquid
400 biofuels. Furthermore, improved potency of enzyme will lead lower dosages of enzyme in
401 hydrolysis in the future, achieving further GHG emission reductions but beyond the scope of
402 this study.

403 Due to the low GHG intensity of 1.7 g CO₂eq per MJ electricity produced from wind power,
404 1 MJ MSW derived liquid biofuels emits 201 g CO₂eq compared to –248.88 g CO₂eq using
405 coal electricity source of which the GHG intensity is 266.3 g CO₂eq per MJ electricity (see Fig.
406 5). This is due to different credits of fixed excess electricity can achieve via electricity
407 displacement. GHG intensity of China electricity mix (240.8 g CO₂eq/MJ electricity) lead the
408 largest GHG reduction of –205.54 g CO₂eq/MJ liquid biofuels compared to –96.74 g CO₂eq/MJ
409 liquid biofuels using US electricity mix (176.9 g CO₂eq/MJ electricity) and -12.57 g CO₂eq/MJ
410 liquid biofuels using UK electricity mix (112.1 g CO₂eq/MJ electricity).

411 **3.3.2 Sensitivity analysis of process variables**

412 Across various parameters as listed in Table 4, ABE product yield can be achieved at a range
413 of 10.73–82.14 kg/t MSW(see Fig. 6a) while a certainty of 90% in the range of 18.3–48.8 kg

414 /t MSW. The composition of incoming MSW feedstock varies but the biogenic content
415 including paper, card, food waste, garden waste, wood, and other organic lies in the range of
416 30-85%. The maximum 85% of biogenic content is assumed for residue MSW exiting material
417 recycling facility where non-biogenic content such as plastics, ferrous and non-ferrous metals
418 are removed. By applying Defra average data, lignocellulosic content of MSW is about 82%
419 (paper 21.6%, card 17.3%, food waste 31%, garden waste 5.3%, wood 3.2%, and other organic
420 3.5%) after going through material recycling facility. A hypothetical lower bound of 30% is
421 considered to evaluate the impact of low biogenic content on the product yield. We have
422 conservatively assumed a glucose hydrolysis yield of 38% based on the current experimental
423 evidence, but we also analyse scenarios where the efficiencies of theoretical glucose ranges of
424 30–85% and xylose ranges of 60–90% (Kalogo et al., 2007). Solid concentration rate ranges
425 from 20% to 30%; the base case is 20%. It should be noted that the variability with product
426 yield would have a large impact on associated financial aspects, where the trade-offs need to
427 be addressed in future optimisation. But before considering the optimisation, how process
428 parameters affect the product yield needs to be understood as follows in the next section.

429 The impacts of process parameter variability on the product yield are evaluated as in Fig. 7a.
430 As biogenic fibre content and glucose hydrolysis yield determines the quantity of total inputs
431 of the conversion, it has the largest impact on the product yield. It indicates the importance of
432 selection of higher biogenic content of MSW from collection facilities or materials recycling
433 facilities. Also optimisation of hydrolysis yield is also the key in future research focus as this
434 is the main barrier to maximising product output. Solid concentration in hydrolysis does not
435 have any direct impact on the absolute product yield but affects the final product concentration
436 as discussed below.

437 Under these scenarios, the ABE product concentration lies in the range of 15.1–79.6 g ABE/L
438 (9.4–47.5 g butanol/L) while the base case is 17.8 g ABE/L (11.2 g butanol/L) (see Fig. 6b)
439 compared to 6.2–64.2 g ABE/L (~10 g/L) as reported (Bankar et al., 2013; Grisales Díaz and
440 Olivar Tost, 2017). However, the high concentration of substrate and ABE products can be
441 toxic to microorganisms that the fermentation can be inhibited completely at butanol
442 concentration of approximately 15 g/L (Grisales Díaz and Olivar Tost, 2017). Glucose
443 hydrolysis yield and solid concentration contribute to the largest variance of the results (88%)
444 (see Fig. 7b). Therefore, future optimisation also needs to consider trade-offs amongst
445 increasing product yield and product concentration level and reducing growth inhibition effect,
446 for instance, by continuous removal of solvents (Brito and Martins, 2017).

447 Under the parameters considered as in Table 4, the net GWP ranges from –26.68 to 6.61 g
448 CO₂eq/MJ liquid biofuels while the base case is -12.57 g CO₂eq/MJ liquid biofuels (see Fig.
449 6c). With 90% certainty the GWP of conversion from MSW to liquid biofuels lies in the range
450 of -16.82–2.60 g CO₂eq/MJ liquid biofuels. Compared to the gasoline, MSW derived liquid
451 biofuels can reduce GHG emissions by 92%% to over 100%, which can significantly contribute
452 to GHG emission reductions of transport fuels by a minimum of 6% by 2020 compared to the
453 2010 level as regulated by EU Fuel Quality Directive (European Union Fuel Quality Directive,
454 2009), and meet the sustainability criteria for biofuels by EU Renewable Energy Directive
455 (GHG savings of at least 60% in comparison to fossil fuels in 2018) (European Union
456 Renewable Energy Directive, 2009). Glucose hydrolysis yield and biogenic fibre content has
457 the largest impact on the quantity of total GWP as in Fig. 7c.

458 Unlike the impact on product yield, uncertainty of process parameters shows different
459 impacts on GHG emissions. Although the increase of sugar yield/sugar content can be
460 beneficial to the increase of product yield, it is detrimental to the GWP reduction. High

461 biogenic content with relatively low ABE product yield (e.g., low sugar content/yield) can
462 minimise overall GWP per MJ liquid biofuels. This is primarily because more unconverted
463 biogenic fibre residue goes to energy recovery unit and generates more co-product electricity
464 as discussed in Section 3.2.

465 **4 Conclusions**

466 The study presents the conversion of ABE from municipal solid waste feedstocks via
467 autoclave, enzymatic hydrolysis, and ABE fermentation based on experimental data and
468 process modelling, demonstrating the conversion to be a promising way to valorise organic
469 wastes in the production of liquid biofuels. The mass and energy balance results show the
470 overall conversion process is energy self-sufficient as process energy demand (steam and
471 electricity) is fully covered by the energy recovery from lignin from hydrolysis and
472 fermentation residue and biogas and sludge produced from wastewater treatment while surplus
473 electricity be exported to the grid as co-product benefits. In total, the integrated process can
474 produce 12.2 kg butanol (7.6 kg gasoline equivalent), 1.5 kg ethanol (0.9 kg gasoline
475 equivalent), 5.7 kg acetone, and 0.9 kg hydrogen as well as 0.3 kg acetic acid and 1.0 kg butyric
476 acid from each tonne raw MSW. It should be noted that hydrolysis yield can be potentially
477 improved up to 85% for glucose conversion as discussed which would increase ABE solvent
478 output to about 39 kg.

479 MSW-derived liquid biofuels can contribute to reduce GHG emissions by over 100% %
480 compared to gasoline base on EU RED standard and conventional bioethanol. Monte Carlo
481 simulations indicate the ABE product yield with a certainty of 90% in the range of 18.3-48.8
482 kg /t MSW. Correspondingly, the net GWP ranges from -16.82 to 2.60 g CO₂eq/MJ liquid
483 biofuels with a 90% certainty while the base case is -12.57 g CO₂eq/MJ liquid biofuels.

484 Sensitivity analysis results indicate high biogenic content with relatively low ABE product
485 yield (e.g., low sugar content/yield) can minimise overall GWP per MJ liquid biofuels.

486 However, key technology challenges still need to be addressed in improving hydrolysis yield
487 which has already showed the potential to be up to 90% as this is the main barrier to maximising
488 product output for future commercialisation. Ongoing investigation is studying the non-
489 biogenic content of MSW recycling such as metal and plastic recycling benefits in order to
490 comprehensively quantify the waste management of MSW. Future work also goes for waste
491 supply chain investigation and facility design optimisation (e.g., capacity; colocation).

492

493 **Nomenclature**

GHG	Greenhouse gas
MSW	Municipal solid waste
LCA	Life cycle assessment
ABE	Acetone, butanol, ethanol
PED	Primary energy demand
GWP	Global warming potential
RED	Renewable Energy Directive
US EPA	US Environmental Protection Agency
US ERA	US Energy Research Architecture

494

495 **Notes**

496 The authors declare no competing financial interest.

497 **Acknowledgment**

498 This work was supported by the funding from the Bioenergy Sustaining the Future 2
499 MSWBH Project [grant number 620103].

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626 **Figure Captions**

627 Fig. 1 Overall diagram for MSW to ABE conversion (dashed line means outside the system
628 boundary)

629 Fig. 2 Sugar composition in MSW fibre by total acid hydrolysis as measured by ion
630 chromatography (weights determined in the monomer form, against total dry weight of
631 sample). Residual is difference from 100, which accounts for all other organic and non-organic
632 material present in the biofiber, which is not suitable for fermentation.

633 Fig. 3 Diagram showing the inputs into and outputs from the process of ABE production
634 from MSW. Dashed line shows the excluded flow in this analysis

635 Fig. 4 a) Primary energy demand and b) Global warming potential of MSW derived liquid
636 biofuels

637 Fig. 5 Comparison of life cycle global warming potential per MJ fuels amongst gasoline,
638 MSW derived liquid biofuels, and bioethanol.

639 Fig. 6 Monte Carlo analysis of the values of a) ABE product yield; b) ABE product
640 concentration; c) life cycle GWP of MSW derived liquid biofuels; at various process
641 parameters. Dashed lines show the base case.

642 Fig. 7 Relative contribution of processing parameters on the sensitivity of a) ABE product
643 yield; b) ABE product concentration; c) life cycle GWP of MSW derived liquid biofuels.

644 Table 1 Process data of MSW to ABE conversion.

645 Table 2 Parameters of ABE fermentation of MSW hydrolysates.

646 Table 3 Overall mass and energy balance of ABE production from MSW.

647 Table 4 Parameters for Monte Carlo simulation.

648 **Tables and Figures**649 **Table 1**

650 Process data of MSW to ABE conversion.

	Input		Quantity of product	
Feedstock	MSW	1	t MSW	
	Moisture content of MSW	40%		
Autoclave	Fibre yield ex autoclave+ sieve	53%		Autoclaved fibre dry 318 kg
	Energy consumption			
	Electricity	43	MJ/t MSW	
	Heat	274	MJ/t MSW	
	Process water	245	kg/t MSW	
Hydrolysis	Glucose content	45%		Sugar 65.51 kg
	Xylose content	5%		
	Glucose hydrolysis yield	38%		
	Xylose hydrolysis yield	70%		
	Solid content in hydrolysis slurry	20%		
	Energy consumption			
	Electricity	0.41	MJ/t MSW	
	Heat	214.63	MJ/t MSW	
	Enzyme loading, w enzyme/w sugar	15.90	kg/t MSW	
	volume 6M H2SO4 (for pH control)	8.59	kg/t MSW	
	Weight of water added for slurry	1035.51	kg	
Clean water usage	5366.25	kg		
Fermentation	Sugar use efficiency	90%		
	ABE yield	33%		
	Energy consumption			
	Electricity	0.68	MJ/t MSW	
	<i>Clostridium acetobutylicum</i>	0.16	kg	
	Monopotassium phosphate	1.09	kg	
	Dipotassium phosphate	0.83	kg	
	Ammonium acetate	3.17	kg	
	Yeast extract (Duchefa)	2.73	kg	
	FeSO4.7H2O	0.01	kg	
	MgSO4.7H2O	1.09	kg	
	Process water	4509.87	kg	

Distillation	Energy consumption			Acetone	5.716	kg
	Electricity	3.31	MJ/t MSW	Butanol	12.24	kg
	Heat	125.30	MJ/t MSW	Ethanol	1.52	kg
				Acetic acid	0.28	kg
				Butyric acid	1.02	kg
Gas stripping	Energy consumption			H ₂	0.93	kg
	Electricity	0.56	MJ/t MSW	CO ₂	30.66	kg
	Heat	46.15	MJ/t MSW			
	Process water	10.75	kg			
Wastewater Treatment	Energy consumption			Clean water recovered	10871.18	kg
	Electricity	56.55	MJ/t MSW	Net water required	-296.20	kg
	Heat	0.02	MJ/t MSW			
Energy recovery	Electricity	959.03	MJ/t MSW			
	Heat	1107.76	MJ/t MSW			
	Excess Electricity	854.51	MJ/t MSW			
	Excess Heat	447.66	MJ/t MSW			
Transport of ABE to depot	Transport per					
	Truck for liquids (diesel)	150	km	2.92	ton·km / t MSW	
	Fuel	Diesel				
	Energy consumption depot					
	Electricity UK mix	0.00084	MJ/MJ liquid biofuels			
Transport to filling station	Transport per					
	Truck for liquids (diesel)	150	km	2.92	ton·km / t MSW	
	Fuel	Diesel				
	Energy consumption depot					
	Electricity UK mix	0.0034	MJ/MJ liquid biofuels			

652 **Table 2**

653 Parameters of ABE fermentation of MSW hydrolysates.

Type	Formula	Molar ratio	Weight percentage
Glucose	C ₆ H ₁₂ O ₆	1	-
Acetic acid	CH ₃ COOH	0.014	0.5%
Butyric acid	C ₄ H ₈ O ₂	0.035	1.7%
Acetone	C ₃ H ₆ O	0.3	9.8%
Butanol	C ₄ H ₉ OH	0.5	20.8%
Ethanol	C ₂ H ₅ OH	0.1	2.6%
Carbon dioxide	CO ₂	2.1	52.0%
Hydrogen	H ₂	1.4	1.6%
Microbial cells	CH _{1.8} O _{0.5} N _{0.2}	-	11%

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Table 3
Overall mass and energy balance of ABE production from MSW.

Inputs	Tonne/year	MW
MSW (40% moisture)*	150000	28.4/yr
Total input		28.4/yr
Outputs		
Acetone	947	0.9/yr
Butanol	2028	0.2/yr
Ethanol	252	2.4/yr
Hydrogen	139	0.001/yr
Heat generation		5.8/yr
Electricity generation		5.0/yr
Total output		14.4/yr
Process heat demand		3.2/yr
Process electricity demand		0.5/yr
Net heat surplus		2.4/yr
Net electricity surplus		4.5/yr
Energy efficiency (main product + net electricity surplus)		28%
Energy efficiency(main product + net electricity surplus)- ethanol from corn stover (Humbird et al., 2011)		47%

658 * (Excluding non-biogenic content)

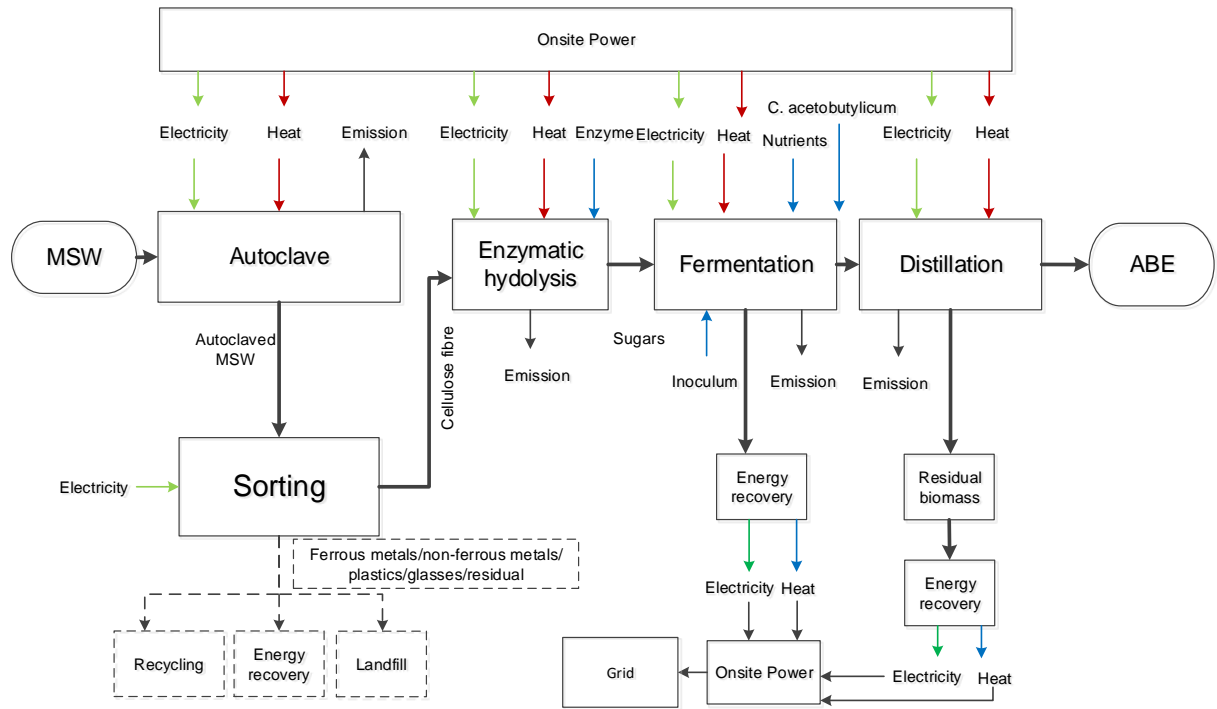
659

660 **Table 4**
 661 Parameters for Monte Carlo simulation.

Parameter	Distribution
Biogenic fibre content	Triangular (Min: 30%, Max: 85%, Mode: 53%)
Glucose content	Triangular (Min: 40%, Max: 65%, Mode: 45%)
Xylose content	Triangular (Min: 5%, Max: 15%, Mode: 5%)
Glucose hydrolysis yield	Triangular (Min: 30%, Max: 85%, Mode: 38%)
Xylose hydrolysis yield	Triangular (Min: 60%, Max: 90%, Mode: 70%)
Solid concentration in hydrolysis	Triangular (Min: 20%, Max: 30%, Mode: 20%)

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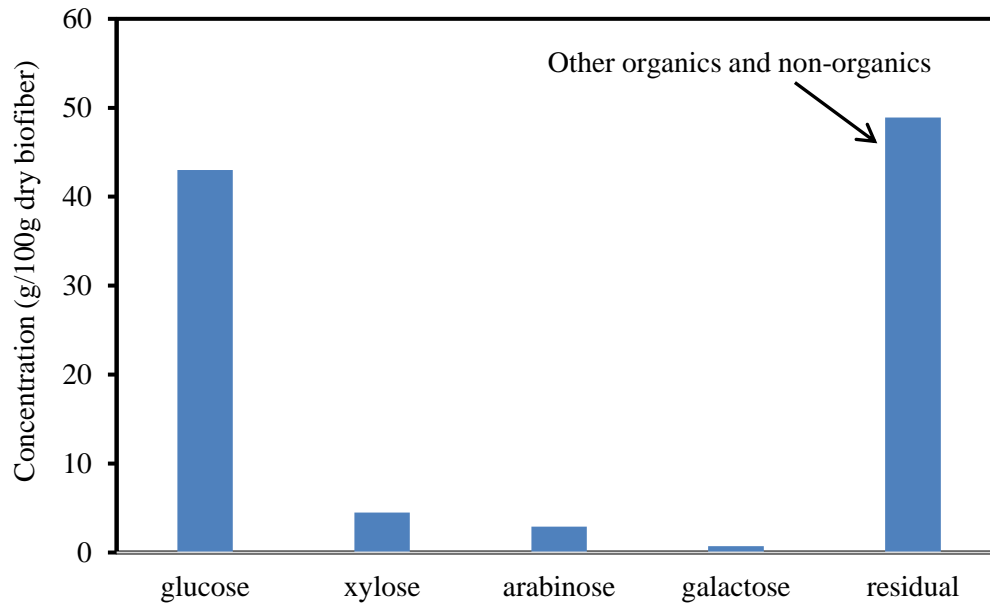


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665 Fig. 1 Overall diagram for MSW to ABE conversion (dashed line means outside the system
666 boundary)

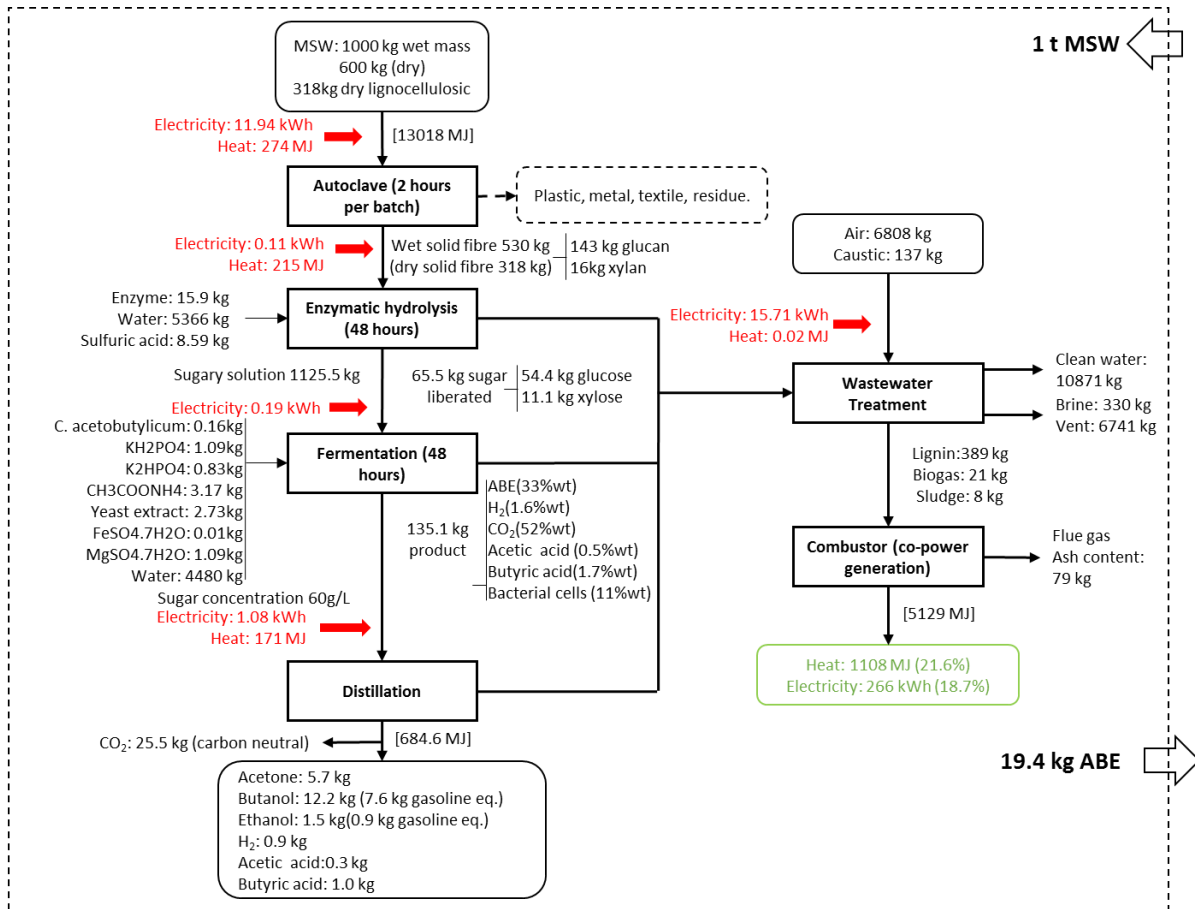
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671 Fig. 2 Sugar composition in MSW fibre by total acid hydrolysis as measured by ion
672 chromatography (weights determined in the monomer form, against total dry weight of
673 sample). Residual is difference from 100, which accounts for all other organic and non-organic
674 material present in the biofiber, which is not suitable for fermentation.



675

676 Fig. 3 Diagram showing the inputs into and outputs from the process of ABE production

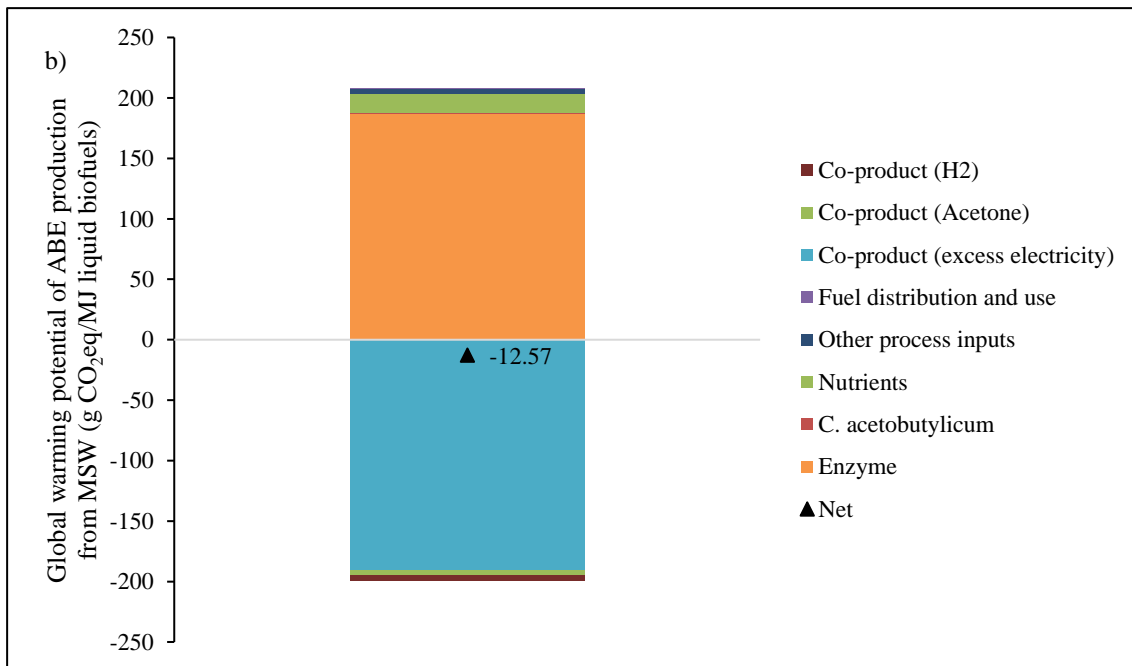
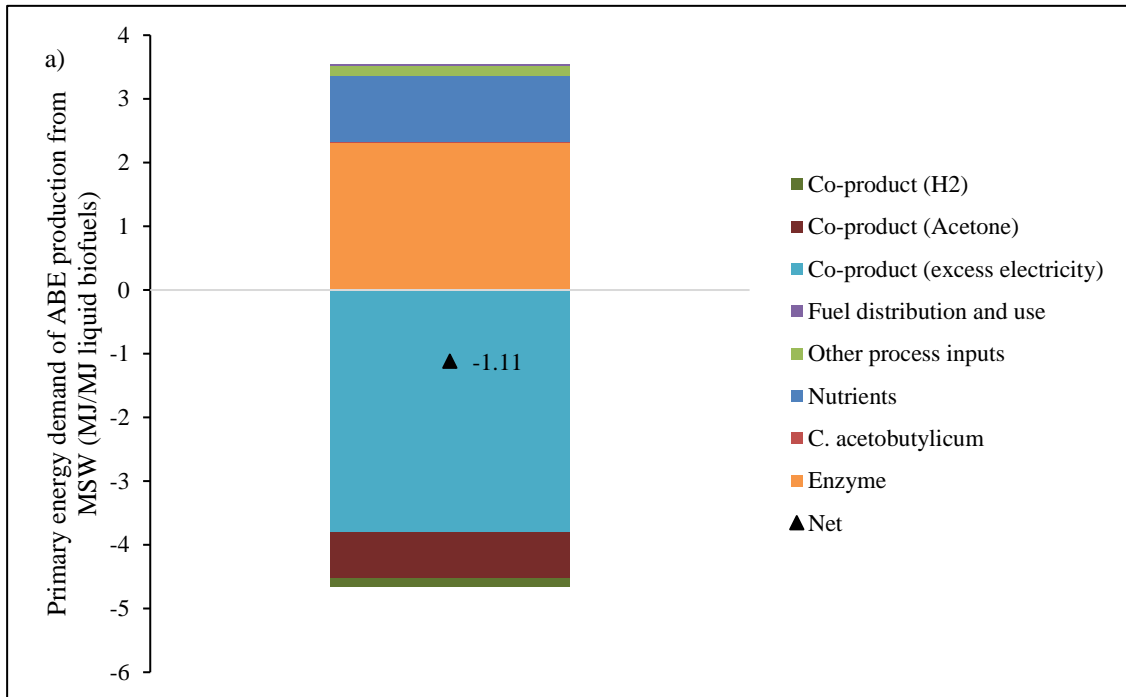
677 from MSW. Dashed line shows the excluded flow in this analysis.

678

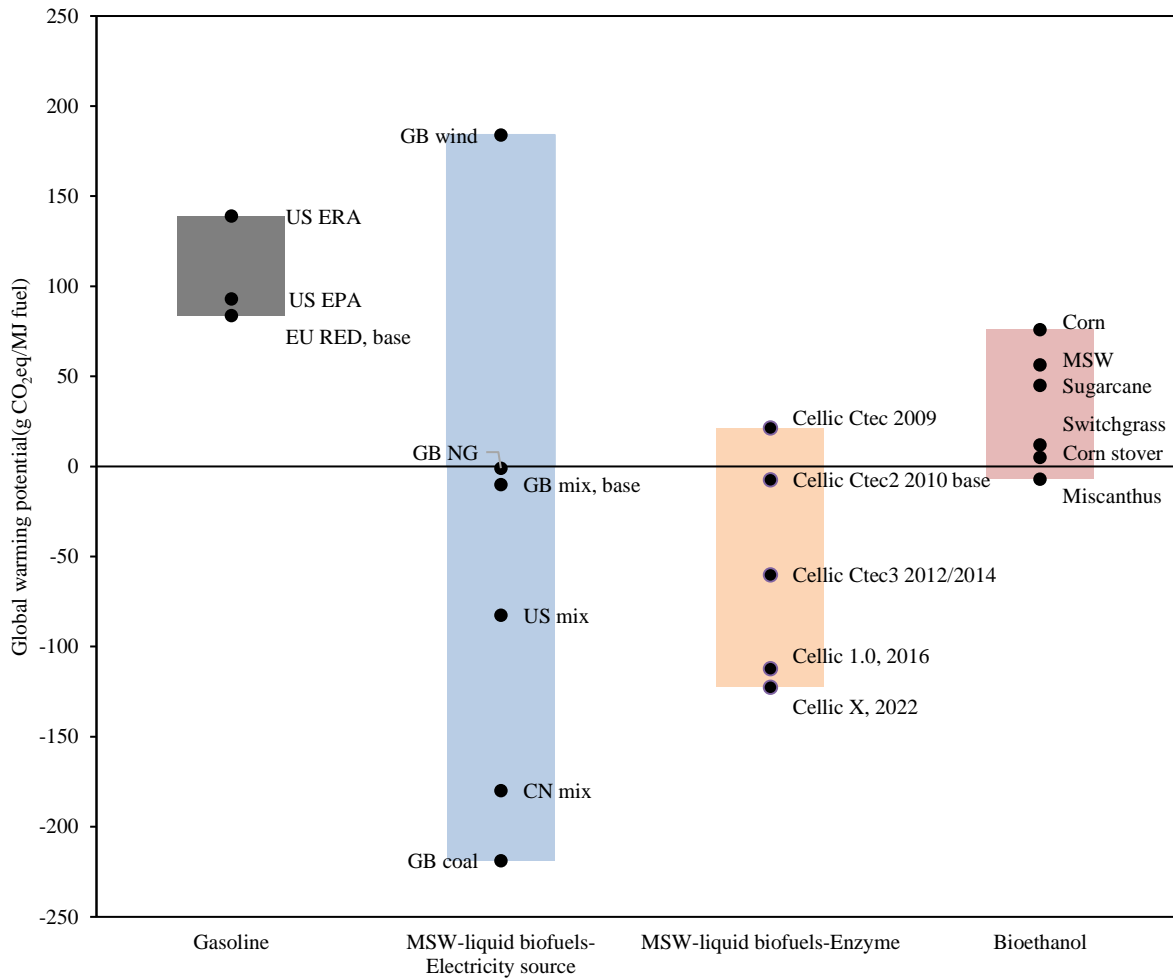
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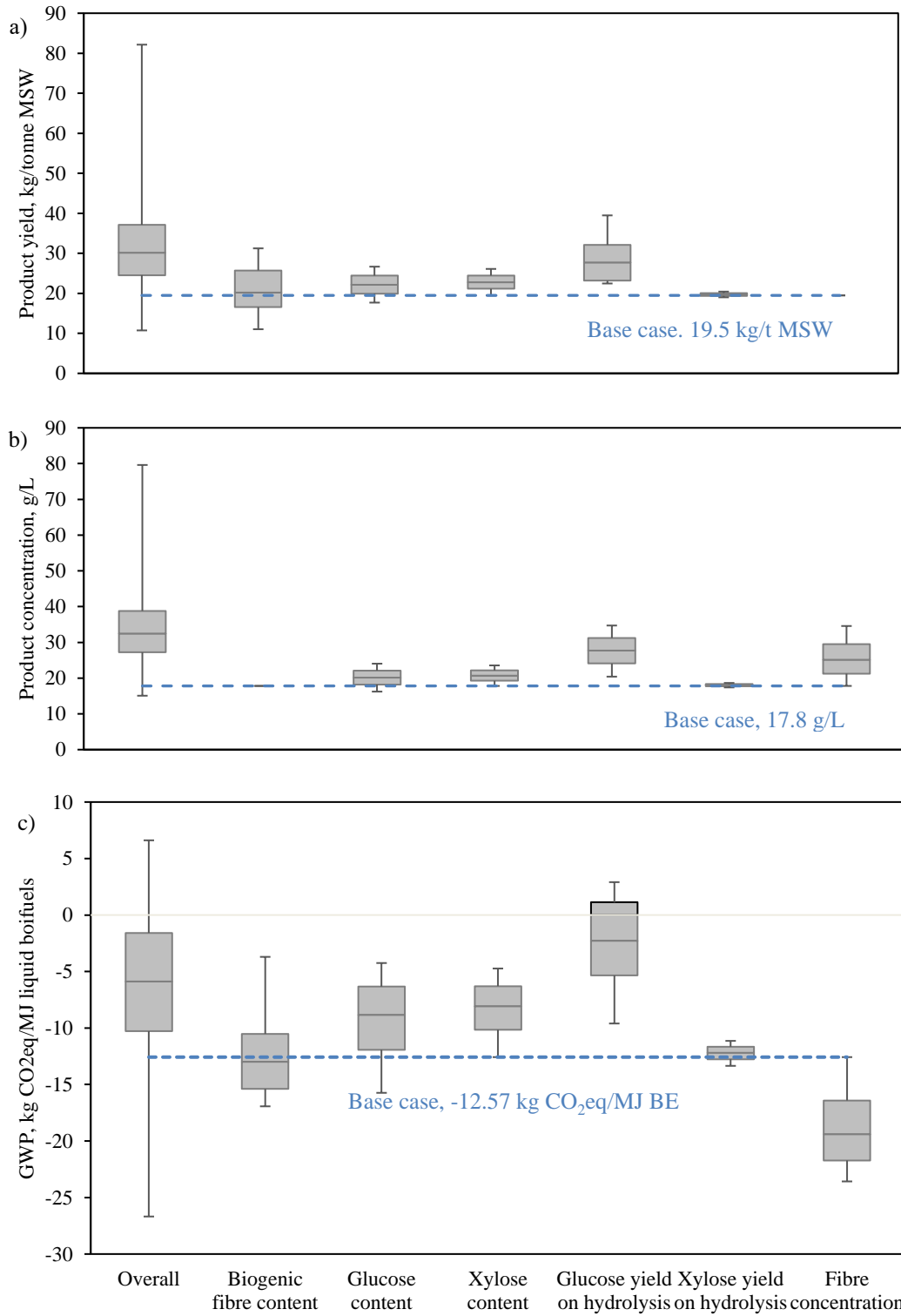
682 Fig. 4 a) Primary energy demand and b) Global warming potential of MSW derived liquid
 683 biofuels.
 684



685

686 Fig. 5 Comparison of life cycle global warming potential per MJ fuels amongst gasoline,
 687 MSW derived liquid biofuels, and bioethanol¹.

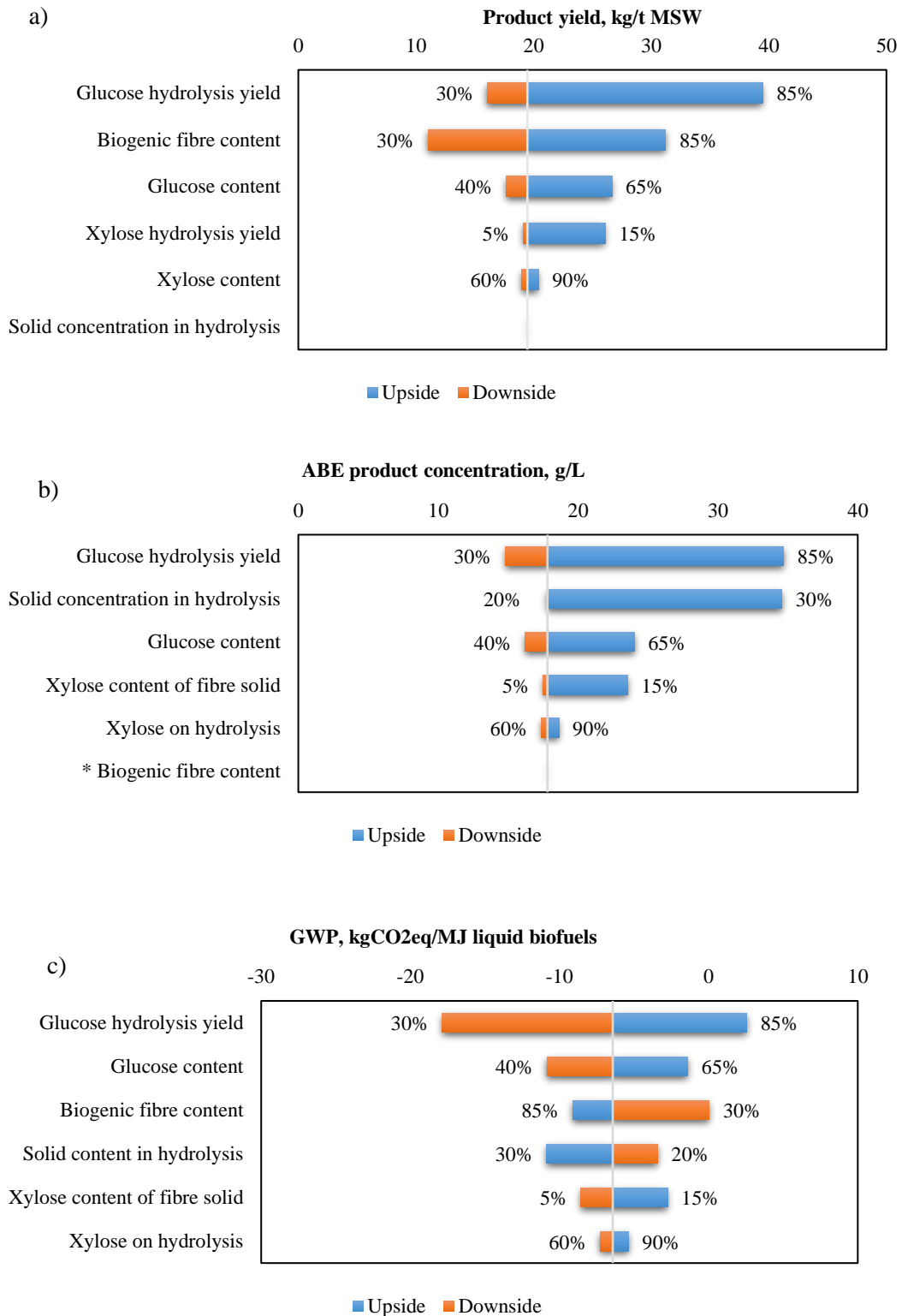
¹ Notes: MSW-BE Electricity source (sensitive coproduct credits from electricity replacement of electricity mix, electricity from hard coal, natural gas (NG), wind power in UK and electricity mix in US and China); MSW-BE Enzyme (various series of enzyme production); gasoline (the EU fossil fuel comparator, US EPA and US oil shale based on ERA); bioethanol (cellulosic ethanol produced from ethanol, sugarcane, corn stover, switchgrass, and miscanthus)



688 Fig. 6 Monte Carlo analysis² of the values of a) ABE product yield; b) ABE product
689 concentration; c) life cycle GWP of MSW derived liquid biofuels; at various process
690 parameters. Dashed lines show the base case.

691

² Note: For the box plot, the ranges are calculated as follows: The box, from bottom to top, indicates the 25th (Q1), 50th and 75th (Q3) percentile values from the data plotted. The interquartile range (IQT) is $Q3 - Q1$. The whisker below is defined as minimum and the whisker above is defined as maximum.



692 Fig. 7 Relative contribution of processing parameters on the sensitivity of a) ABE product
 693 yield; b) ABE product concentration; c) life cycle GWP of MSW derived liquid biofuels.