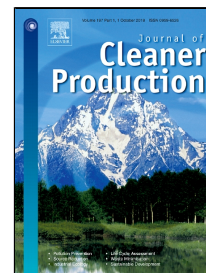# Accepted Manuscript

Life-cycle Assessment on Food Waste Valorisation to Value-added Products

Chor-Man Lam, Iris K.M. Yu, Shu-Chien Hsu, Daniel C.W. Tsang

Please cite this article as: Chor-Man Lam, Iris K.M. Yu, Shu-Chien Hsu, Daniel C.W. Tsang, Life-cycle Assessment on Food Waste Valorisation to Value-added Products, *Journal of Cleaner Production* (2018), doi: 10.1016/j.jclepro.2018.07.199

1   **Life-cycle Assessment on Food Waste Valorisation to Value-added Products**

2   Chor-Man Lam[a], Iris K.M. Yu[a], Shu-Chien Hsu[b,*], Daniel C.W. Tsang[c,*]

3   **Abstract**

4   Food waste can serve as a potential substitute for fossil-derived feedstocks for producing value-added chemicals,

5   such as hydroxymethylfurfural (HMF), yet their environmental performance has not yet been evaluated, thus

6   impeding informed decision-making. This study aims to develop a life-cycle assessment (LCA) framework to

7   guide decisions on selecting the most environmentally favourable food waste valorisation option to produce HMF.

8   A LCA framework was developed to assess the environmental performance of eight food waste valorisation

9   scenarios with different combinations of solvents, catalysts, and experimental conditions. The environmental

10   impacts associated with the use of water solvent, organic co-solvents, metal catalysts, as well as the reaction

11   temperature and time were estimated. Experimental data were analysed for building the life-cycle inventory. The

12   conversion of bread waste using water-acetone medium with the catalyst aluminium chloride (AlCl$_3$), at 140°C

13   for 30 minutes, was revealed to be the most environmentally favourable food waste valorisation option, due to the

14   utilization of less polluting co-solvent (acetone) and catalyst (aluminium chloride) as well as the relatively high

15   yield of HMF (27.9 Cmol%). It is expected that when the development of large-scale valorisation systems become

16   more mature and information is more readily available, the decision-supporting tool could be expanded to (1)

17   evaluate the pilot-scale and the industrial-scale of food waste valorisation to HMF, and (2) include the economic

18   performance of the scenarios so that more comprehensive results could be provided to assist decision-making.

19

[a] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong.
[b] Co-corresponding author. Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong. Tel: 2766-6057. Email address: mark.hsu@polyu.edu.hk
[c] Co-corresponding author. Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong. Tel: 2766-6072. Email address: dan.tsang@polyu.edu.hk

1. Introduction

The continuous increase of global demand for energy and petroleum-derived chemicals has been exerting pressure on global climate change as well as the world's fossil resources reserves. This increase is mostly driven by the thriving economies such as China and India, which are estimated to have a total of 51% growth in energy demand in the year 2015-40 (van Putten et al. 2013; USEIA 2017). The depleting fossil reserves have aroused much attention, since petrochemicals have been vital for the production of various important chemicals and products such as polymers, solvents, pesticides, pharmaceuticals, and asphalt (Rout et al. 2016; Sheldon, 2016; Matsakas et al. 2017). Renewable biomass appears to be an ideal and sustainable substitute for fossils as it is a great carbon source with high contents of carbohydrates, lignocellulose, and fatty acids (van Putten et al. 2013). The need for biochemical production has been reflected by its remarkable global market value of $1 trillion USD, which could create 200,000 job opportunities in the US (Golden and Handfield 2014).

Previous studies have investigated conversion of biomass to biofuels and value-added chemicals via different biological technologies (e.g., fermentation) and chemical processes (e.g., pyrolysis and catalysis) (Schwartz et al. 2014; Karka et al. 2017). Carbohydrate-rich feedstock such as corn and sugarcane are widely used for ethanolfatty acid methyl ester can be generated from bread waste and waste cooking oils (Xiong et al., 2017; Yu and Tsang, 2017).

The valorisation of biomass to hydroxymethylfurfural (HMF) has been extensively studied as it is a versatile platform chemical, which has been listed as one of the top 10 bio-based chemicals by the US Department of Energy (Rout et al. 2016; Chen et al., 2017; Yu and Tsang, 2017). Derivatives of HMF are the building blocks of various products including pharmaceuticals, polymers, antifungal substances, and ligands (van Putten et al. 2013; Sheldon 2016). At present, commercial HMF production is primarily produced from sugarcane syrup (Kläusli 2014). The process of growing the energy crops, however, is suggested to pose threats to the environment because of soil and water contamination by pesticides and fertilisers (Karka et al. 2017). As a result, the call for a more sustainable way to synthesise HMF is imperative. A vast variety of valorisation methods with different substrates, catalysts, and solvents are demonstrated in the literature (Table 1). However, these studies focused on technological advancements for high-yield production of HMF without assessing the environmental impacts induced in the production process, which possibly hinders decision makers from evaluating the novel options in a holistic perspective. Thus, to identify and evaluate the environmental impacts of the HMF production process from waste biomass, a decision-supporting tool is required.

51   A number of studies evaluated the environmental feasibility of different biomass waste conversions via

52   life cycle assessment (LCA). For instance, Woon et al. (2016) studied the environmental impacts of using food

53   waste to generate biogas through anaerobic digestion, while another study carried out by Vandermeersch et al.

54   (2014) also considered the possibility of producing heat and energy through food waste (Scenario 1), with another

55   scenario of animal feed production (Scenario 2) for comparison. Karka et al. (2017) conducted a large-scale LCA

56   of the production of 23 products including various chemicals, biodiesel, and biogas using five different types of

57   biomass waste (wood chips, municipal solid waste, rapeseed oil, wheat straw, and waste cooking oil). The majority

58   of these LCA studies targeted the environmental impacts on biogas or energy production, with only a few on high

59   value-added chemicals. To the best of our knowledge, LCA on biomass waste valorisation to HMF has not been

60   conducted. Thus, this paper intends to fill this research gap by entirely focusing on the production process of this

61   high-valued chemical.

62   In this study, food waste is chosen as a representative of waste biomass for evaluation because food waste

63   accounts for nearly one-third of municipal solid wastes (MSW) in Hong Kong (HK EPD 2016) and other densely

64   populated cities. Globally, one-third of the total food production that amounts to approximately 1.3 billion tons is

65   being disposed per year, generating 3.3 billion tons of CO_2 equivalent and causing an annual economic loss of

66    $750 billion USD (Gustavsson et al., 2011; FAO, 2013). Catalytic conversion approaches have been recently

67   developed to produce HMF from food waste (Yu et al., 2016; Yu et al., 2017a). Yet, their environmental

68   performances have not yet been evaluated. Trade-off between the technological performance and environmental

69   compatibility often renders decision making difficult. For instance, using water as the reaction medium is

70   environmentally benign, but the resultant HMF yield is usually inferior to that in organic solvent-containing

71   medium (Yu and Tsang, 2017).

72   Therefore, the primary aim of this study is to develop an LCA framework to assess the environmental

73   significance of various system components (solvents, catalysts, reaction temperature, reaction time, etc.) in the

74   food waste-to-HMF process, by comparing the environmental impacts arising from eight laboratory-scale

75   conversion systems as the pilot scheme is yet to be established. This study could assist researchers in designing

76   green catalytic systems for biomass waste valorisation, as well as advise decision makers in sustainable

77   management of biomass waste in long term.

78

79   **2. Methodology**

80   LCA is known as a 'cradle to grave' approach for evaluating the entire life-cycle environmental impacts

81   of a process, service, or product (Nanda et al. 2015). The associated energy and resource consumption, together

82   with the emissions for the whole system are being assessed (Gonzalez-Garcia et al. 2017). To systemically carry

83   out a LCA, four steps have to be followed according to ISO 14040 standard (ISO 14040 2006; Parajuli et al.

84   2017), which are 1) goal and scope definition: defining the objective, system boundary, and the functional unit;

85   2) data collection and inventory analysis: explaining and presenting the life-cycle inventory data for the

86   assessment; 3) Life cycle impact assessment: selecting the impact categories for the energy and resource, as well

87   as the emission generated, and; 4) Results interpretation and presentation: reporting the results as detailed as

88   possible with the suggestions on impact reduction.

89   **2.1   Goal and scope definition**

90   The goal of this LCA is to assess and compare the environmental performance of eight experimental

91   methods for producing HMF from food waste, which significantly differ in terms of the operating parameters (i.e.,

92   process inputs) and product yields (i.e., process outputs). The LCA is conducted in this study to evaluate the

93   environmental burdens and benefits originated from the raw material acquisition, material processing, production

94   and the yield of final products. The functional unit (FU) is defined as the conversion of 1 g of food waste substrates.

95   The scope of this LCA covers eight scenarios of catalytic conversion of food waste to HMF, including

96   processes of the use of solvent and co-solvents, the addition of catalysts, heating, and yielding of HMF. Water

97   was used as the solvent in all scenarios, while various organic solvents were used as the co-solvents. Either tin

98   (IV) chloride ($SnCl_4$) or aluminum chloride ($AlCl_3$) were used as the catalysts. Microwave reactor was used for

99   the heating process. The environmental consequences related to such processes were included in the system

100   boundary, which determines what processes and activities are included in the LCA (Figure 1).

101   *2.1.1   Descriptions of LCA scenarios*

102   Scenario 1 (S1) used bread waste as the substrate of the conversion process. $SnCl_4$ was used as the

103   catalyst and added to the organic solvent-water medium, which was prepared using water and dimethyl sulfoxide

104   (DMSO). The mixture was heated to 160°C with a reaction time of 20 minutes.

105   Scenario 2 (S2) used the same substrate, organic solvent-water medium, catalyst and reaction time as S1.

106   The reaction temperature was 140°C in S2.

107   Scenario 3 (S3) was the conversion of bread waste substrate to HMF using DMSO-water medium and

108   $SnCl_4$ catalyst. The reaction temperature and time were 140°C and 60 minutes respectively.

109   Scenario 4 (S4) was the conversion of bread waste substrate to HMF using a medium of water and

110   tetrahydrofuran (THF), and $SnCl_4$ as the catalyst. The mixture was heated to $140^{o}C$ and the reaction time was 120

111   minutes.

112   Scenario 5 (S5) used an acetone-water medium with the addition of $SnCl_4$ catalyst for the conversion of

113   bread waste to HMF. The reaction temperature and time were $140^{o}C$ and 10 minutes respectively.

114   Scenario 6 (S6) converted bread waste substrate to HMF using an acetone-water medium and $AlCl_3$ as

115   the catalyst. The mixture was heated to $140^{o}C$ and allowed for reaction for 30 minutes.

116   Scenario 7 (S7) used rice as the food waste substrate. A DMSO-water medium was used, and $SnCl_4$ was

117   used as the catalyst. The reaction temperature and time were $140^{o}C$ and 40 minutes respectively.

118   Scenario 8 (S8) used kiwi fruit as the food waste substrate. The $SnCl_4$ catalyst was added to the DMSO-

119   water medium. The mixture was heated to $140^{o}C$ and allowed for reaction for 20 minutes.

120   *2.1.2 Descriptions of food waste conversion experiments*

121   *Food waste and chemicals*

122   Food waste produced from the Hong Kong International Airport, including bread, rice and kiwifruit

123   wastes, were collected for the experiments. The samples were freeze-dried, ground, and sieved through a 0.2 mm

124   mesh for storage in an airtight storage container at $4^{o}C$ before use. Drying is only employed for experimental

125   investigations and is not needed in the scaled-up application, where the indigenous water content of food waste

126   will be used as a water solvent. Co-solvents including dimethyl sulfoxide (DMSO; ≥99.9%) and tetrahydrofuran

127   (THF; ≥99.8%) from RCI Labscan, and acetone (99.5%) from Duksan Pure Chemicals were used to prepare the

128   reaction medium. The metal chloride catalysts, $SnCl_4 \cdot 5H_2O$ (98%) and $AlCl_3 \cdot 6H_2O$ (ACS grade), were purchased

129   from Sigma Aldrich and Anaqua, respectively. All the chemicals were used as received.

130   *Process of Catalytic conversion*

131   The process of catalytic conversion of food waste was reported in our previous studies (Yu et al., 2016;

132   Yu et al., 2017a, 2017b, 2017c). In brief, 1 g of dried food waste as the substrate (equivalent to the concentration

133   of 5 wt/v%), 10 ml of organic solvent and 10 ml of water as the reaction medium, and 0.39 or 0.27 g of metal

134   chloride catalyst ($SnCl_4$ or $AlCl_3$, equivalent to the concentration of 55.5 mM) were mixed. The mixture was

135   heated to $140-160^{o}C$ in an Ethos Up Microwave Reactor (Milestone, maximum power 1900 W), with the reaction

136   time held between 10-120 min under magnetic stirring. The inputs of the process include food waste, organic

137   solvent, water, metal chloride, and microwave energy (flow quantity shown in Table 2). Diluted and filtered

138   samples were analysed by high-performance liquid chromatography (HPLC) to reveal the HMF yield, i.e., the

139 process output, resulted from each scenario, using the Chromaster (Hitachi) equipped with an Aminex HPX-87H

140 column (Bio-rad). All the trials were conducted in duplicate.
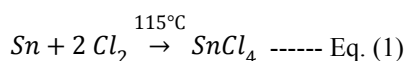
2.2 Life-cycle inventory analysis

142 Life-cycle inventory (LCI) analysis is the phase of accounting the inputs and outputs of materials and

143 energy of the system being studied. The full account of inputs and outputs provides the fundamental information

144 for further estimation of environmental impacts in the next phases. In this study, the details of the conditions of

145 the laboratory conversion process of food waste to HMF were organized and used to build the LCI (Table 2). The

146 electricity consumption for heating the reaction mixture was calculated based on the power of the microwave

147 reactor and the duration of heating using the equation $E = P \times t$ (E denotes energy, P denotes power and t denotes

148 time). The amounts of solvent, co-solvents, catalysts, and HMF were measured from the experiments.
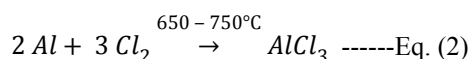
2.3 Life-cycle impact assessment

150 Based on LCI analysis of the inputs and outputs of the systems, the induced environmental consequences

151 were estimated in the life-cycle impact assessment (LCIA) phase. The ReCipe Endpoint method was adopted for

152 conducting the LCIA. Instead of the individualist (short-term and optimistic) or the egalitarian (long-term and

153 precautionary) models, this study adopted the hierarchist model (PRé Sustainability; Huijbregts et al. 2016), which

154 has been recognized as the consensus model. The hierarchist model was selected as the temporal boundary and

155 the mechanisms of the impacts used in such model are based on scientifically credible data (PRé Sustainability;

156 Huijbregts et al. 2016). Eighteen midpoint indicators and three endpoint indicators are analysed in the ReCipe

157 Endpoint method. The software SimaPro 8.3.0.0, which is a widely recognized LCA tool, was used in this study.

158 The solvents and electricity were produced through relatively common processes, thus the information

159 on the associated environmental emissions is available in databases. Such information was adopted from the

160 EcoInvent database in this study. To fit the local situation, emissions associated with electricity consumption were

161 estimated by considering the fuel mix for electricity generation in Hong Kong: 53% coal, 22% natural gas, 23%

162 nuclear and 2% renewable energy (HK ENB 2014).

163 The environmental emissions originated from the production process of $SnCl_4$ and $AlCl_3$ have not yet

164 been documented in the databases, thus such emissions were estimated according to the method used by the

165 EcoInvent for building life-cycle inventories of chemicals in order to ensure the consistency (Althaus et al., 2007).

166 The reactions of metals with chlorine gas at elevated temperature were assumed to be the synthesis method to

167 produce the metal chlorides, as represented by Equation 1 for $SnCl_4$ and Equation 2 for $AlCl_3$.

168
$$Sn + 2\,Cl_2 \xrightarrow{115°C} SnCl_4 \text{ ------ Eq. (1)}$$

6

169
$$2\,Al + \,3\,Cl_2 \quad \overset{650-750°C}{\rightarrow} \quad AlCl_3 \quad \text{------Eq. (2)}$$

170 Stoichiometric calculations were used to estimate the quantity of elemental metals and chlorine gas required. The

171 energy for elevating the reaction temperature was also included in the analysis.

172   The conventional approach to produce HMF was neither documented in the previous LCA studies nor

173 the databases. In this study, the reaction between sugar syrup (fructose) and sulfuric acid at a temperature of 166°C

174 was assumed (Fachri et al. 2015). Based on such study, 2.55 g of sugar syrup, 0.07 g of sulfuric acid and 0.21

175 kWh of electricity are required for producing 1 g of HMF (Fachri et al. 2015). The environmental impacts of sugar

176 production and sulfuric acid were obtained from the Agri-footprint (Blonk Consultants 2015a, b) and the U.S.

177 LCI (Franklin Associates 2011) databases, respectively. The Hong Kong fuel mix for electricity generation was

178 considered during the estimation of impacts associated with electricity consumption. The avoidance of starting

179 materials (sugar syrup and sulfuric acid) usage and energy consumption for heating via food waste valorisation

180 were considered in the evaluation of environmental performance.

181   The LCIA includes the steps of classification, characterization, normalization, weighting and aggregation.

182 The environmental emissions were classified into different categories according to the eighteen midpoint

183 indicators, and then characterized into common equivalent units which reflects their contributions to the midpoint

184 impacts. The characterized impacts were then normalized against the world average impacts, so that the relative

185 importance of the impacts in different categories could be considered in the LCA. The normalized results were

186 weighted and aggregated to provide single score LCA results, which are inclusive and convenient indicators to

187 guide decision-making.

188

189 **3. Results and discussion**

190   The overall LCA results showed that S6 performed the best among the eight scenarios, while S4 was the

191 least environmentally favourable option. To understand the details on the environmental implications, the results

192 of different LCIA phases, namely characterisation, normalisation, and weighting and aggregation, were analysed

193 and discussed in the following sections. The single-score results were further segregated according to the

194 contributions of different processes.

195 *3.1   Characterisation results*

196   The environmental impacts of the scenarios in different impact categories are presented in Figure 2. In

197 each impact category, scenarios with the highest impact were assigned the value 1, while other scenarios with

198 lower impact were assigned the values between 0 and 1 by referring to the highest impact. Negative values refer

7

199   to the environmental benefits achieved. The scenarios with relatively lower impacts, i.e., points plotted closer to

200   the centre of the graph, have better environmental performance.

201   Among the scenarios, S4 performed the most unfavourably in all impact categories, which was possibly

202   attributed to the use of THF as the co-solvent and the corresponding less favourable HMF yield compared to

203   majority of the studied systems (Table 2). In comparison, S6 generated apparently lower impacts in natural land

204   transformation (NLT) and metal depletion (MD) compared with the other scenarios because S6 was the only

205   scenario that used $AlCl_3$ instead of $SnCl_4$ as the catalyst. The production of $AlCl_3$ requires the utilization of

206   aluminium mines, while $SnCl_4$ production requires the mining of tin ores. The major reason for the difference was

207   the abundance of metal resources: aluminium is one of the most abundant metals, but tin is a relatively limited

208   natural resource (Smith and Huyck, 1999). The utilisation of aluminium, therefore, causes much lower

209   environmental stress in terms of NLT and MD impacts than that of tin.

210   The yield of HMF from food waste avoided the environmental impacts of the industrial HMF production

211   process from sugar syrup, thus offering notable environmental benefits. When the environmental savings could

212   compensate for the negative impacts, net environmental benefits could be accomplished, as demonstrated by the

213   negative values in marine eutrophication (ME) categories achieved by S1, S5, S6, S7, and S8 (Figure 2).

214   *3.2   Normalization results*

215   Most scenarios except S6 showed the highest impact in the MD impact category (Figure 3). The major

216   reason is the high environmental stress associated with the use of the limited resource of tin metal for catalyst

217   production in most of the studied scenarios, underscoring that catalyst selection was critical for sustainable

218   management of metal resources. Following the MD impact, marine ecotoxicity (MX), freshwater ecotoxicity (FX),

219   and human toxicity (HT) emerged as the significant impacts. The most significant contributors to such impact

220   categories were the production of co-solvents, including THF, DMSO, and acetone, which are listed in descending

221   order of toxicity impacts and consistent with the findings of Capello et al. (2007). The storage of THF produces

222   organic peroxides (Guan and Guan 2012), which are toxic chemicals that could cause human fatality through

223   inhalation, ingestion and dermal contacts (Faggian and Faggian 2014; Government of Canada 2017). The

224   inhalation of THF has been identified to be carcinogenic in laboratory tests on rodents (Faggian and Faggian 2014).

225   Previous studies have revealed that the lethal dosages of DMSO exposure in tests on rodents and fish were much

226   higher than those of THF, indicating that DMSO has lower level of toxicity on human and aquatic ecosystems

227   (Faggian and Faggian 2014). Both DMSO and acetone have been recognised as environmentally friendly solvents

228   with low toxic potential to human (Soroko et al. 2011). Yet, DMSO exposure has been observed to induce higher

229   levels of stress proteins (indicators for toxic response) than acetone exposure in fish embryos, implying that

230   DMSO has higher aquatic ecotoxicity than acetone (Hallare et al. 2006). Production of co-solvents was also

231   identified to be the major contributor to the freshwater eutrophication (FE) impacts. The impacts on fossil

232   depletion (FD), particulate matter formation (PMF), terrestrial acidification (TA), climate change (CC) and

233   photochemical oxidant formation (POF) were attributed to the energy consumption.

234   ***3.3   Single score LCA results***

235   The above characterisation and normalisation results revealed the environmental performance of the

236   scenarios in different mid-point categories, yet single and all-inclusive indicators to guide decision-making were

237   not provided. Thus, the normalized results were assigned weightings and aggregated using the ReCipe Endpoint

238   methodology so that single score results could be obtained. The single score LCA results of the eight scenarios of

239   food waste valorisation are shown in Figure 4. The environmental impacts are presented in milli-points (mPt),

240   which reveal the overall impacts of the scenarios. The results indicate that S6 is the most environmentally friendly

241   option, while S4 is the most polluting scenario.

242   The environmental impacts were categorised into the Human health, Ecosystems and Resources aspects.

243   The impacts in Resources aspect were the highest, contributing to 74% to the overall impacts on average (Figure

244   4). The high environmental stress on resources depletion is mainly attributed to the use of the relatively limited

245   tin resources for producing the metal chloride catalyst. Human health impacts ranked after impacts on Resources

246   with an average contribution of 25% to the overall impacts. The production of organic solvents, especially THF,

247   and the metal mining process caused adverse impacts to human health. As mentioned above (Section 3.2),

248   exposure to organic solvents through different routes has been revealed to cause human health threats, such as

249   cancer and fatality. The excavation activity of tin mining and the disposal of tailings change the radionuclide

250   compositions in soil, thus increasing the chance of radiological exposure of mine workers and nearby residents

251   (Arogunjo et al. 2009; Aliyu et al. 2015). During bauxite mining for AlCl$_3$ catalyst production, the excavation

252   activities release air pollutants, such as dust and fine particulate matters, that harm the respiratory and

253   cardiovascular systems after inhalation (Abdullah et al. 2016). Drinking water could also be polluted by the

254   discharge of bauxite washing water. Chronic ingestion of metal-containing water may increase cancer risks. The

255   environmental emissions from energy consumption and the production processes of organic solvents and metal

256   catalysts caused eutrophication, toxicity, and climate change impacts to the ecosystem. The scenarios in this study

257   had relatively low impacts on the Ecosystems aspect (0.27% - 2.78% of the overall environmental impacts).

9

258   *3.4*   *Single score LCA results with process contributions*

259   To investigate the significance of different system parameters (i.e., reaction temperature, reaction time,

260   solvents, etc.) in determining the total environmental impacts, the single score LCA results are presented to

261   illustrate the individual process contributions (Figure 5). The processes involved in the conversion of food waste

262   to HMF included the utilisation of solvent, co-solvents, catalysts, energy, and the yield of HMF. The use of

263   solvents, catalysts, and energy contributed to the adverse environmental impacts, while the production of HMF

264   recovers untapped value from food waste to synthesise high-value products and presents an alternative to

265   petroleum refinery, thus providing environmental benefits that should be properly recognized and quantified. In

266   all the scenarios, the use of water as solvent only contributed to trivial environmental impacts (only accounted for

267   0.003% of the overall impacts on average). The water use was assumed to obtain from conventional potable water

268   treatment methods, which presented significantly lower environmental impacts compared to other processes, such

269   as the production of organic solvents and metal chlorides. It should be noted that in industrial applications, water

270   solvent should come from the indigenous water content of food waste where additional water demand can be

271   avoided or minimised.

272   In the following sections, the environmental impacts of the scenarios are compared based on the choices

273   of reaction temperature and reaction time, as well as the selection between various organic co-solvents and the

274   two types of catalysts.

275   3.4.1   Energy input

276   The HMF production efficiency can be improved with higher energy input, which can be achieved by

277   increasing the reaction temperature or lengthening the retention time. S1 and S2 are bread waste conversion to

278   HMF in DMSO-water medium with the $SnCl_4$ as the catalyst. The reactions were allowed for 20 minutes. S1 used

279   a higher temperature (160°C) than S2 (140°C), so that a higher yield of HMF could be obtained. The energy

280   consumption for S1 was a double of that for S2, yet the HMF yield of S1 was only 70% higher than S2. The LCA

281   results indicated that although S2 yielded less amount of HMF, the reduced energy consumption still provided it

282   the environmental advantage over S1. S2 and S3 are scenarios with the same reaction conditions except the

283   reaction time. With a threefold length of reaction time, S3 produced 0.199g HMF/g substrate, which was 58%

284   higher than the yield in S2. However, the environmental benefits from HMF production could not compensate the

285   extra energy consumption for the longer heating time, so S3 was less environmentally favourable than S2. These

286   results highlighted that increasing the energy input (either reaction temperature or time) may not be a feasible

287   approach for improving the HMF yield from the environmental perspective. Among all the scenarios, energy

10

288 consumption-related impacts were the highest in S4 that involved THF as the co-solvent (Figure 5), which

289 disfavoured catalytic conversion and, thus, resulted in a high energy demand for noticeable HMF production

290 (Table 2). This suggested the indirect effects imposed by other system components on the magnitude of

291 environmental impacts associated with energy inputs during the conversion process.

292 3.4.2   Organic co-solvents

293   Three types of organic co-solvents, including THF, DMSO and acetone, were used in the different

294 scenarios. THF used in S4 was the co-solvent that contributed the most significantly to the environmental burden

295 of the scenarios, while the use of DMSO (in S1, S2, S3, S7 and S8) and acetone (in S5 and S6) presented much

296 lower environmental impacts than THF. The environmental impacts associated with the co-solvents were

297 attributed to two major factors: the toxicity of the organic co-solvents and the energy requirement in the solvent

298 production processes. The solvents with descending order of toxicity levels are THF > DMSO > acetone and the

299 details were discussed in Section 3.2. The solvents with descending order of energy-associated impacts (including

300 climate change, photochemical oxidant formation, acidification and fossil fuel depletion) are THF > acetone >

301 DMSO. The production of THF has been revealed to be incurring the highest environmental impacts among the

302 three solvents because the petrochemical production process involves numerous conversion steps with high energy

303 and resource requirements (Capello et al., 2007). Methanol, the starting material for THF production, is oxidized

304 to formaldehyde. The Reppe synthesis process used to convert formaldehyde to butylene glycol is energy-

305 intensive, thus is one of the major contributors to the environmental impacts (Capello et al. 2009). Yet, the most

306 dominant factors for the impacts were the cyclization and dehydration processes for the conversion from butylene

307 glycol to THF, owing to the high energy and resource demand (Capello et al. 2009). Acetone could be produced

308 from benzene in BTX (mixture of benzene, toluene and xylene). Benzene is alkylated to cumene, which is an

309 intermediate chemical, and then converted to acetone through the Hock process (ACS 2017). Such production

310 process incurred relatively low environmental impacts because of the low requirements for ancillary chemicals

311 and energy (Capello et al. 2009). Using the same starting chemical as THF, the production of DMSO presented

312 much lower energy-related impacts because DMSO could be simply produced by the oxidation of dimethyl

313 sulphide, which is produced from reacting methanol with hydrogen sulphide (Capello et al. 2009; Bertau et al.

314 2014).

315   Accounting for the toxicity, energy-associated impacts and other relatively minor impacts, the overall

316 environmental impacts per mass of solvents are THF > acetone > DMSO. However, scenarios using acetone (S5

317 and S6) in this study produced lower solvent-related impacts than those using DMSO (S1, S2, S3, S7 and S8),

11

318 because the former scenarios required larger mass of co-solvents (acetone; 7.85g) than the latter ones (DMSO;

319 11.00g).

320 3.4.3   Catalysts

321   The catalyst $AlCl_3$ used in S6 showed trivial environmental impacts, while, in contrast, the $SnCl_4$ catalyst

322 was the most or the second-most dominant polluter in other scenarios. One of the factors contributing to the

323 difference was the different abundance levels of the metal resources:aluminium resource is abundant while tin is

324 a relatively limited metal resource (Smith and Huyck, 1999). The preparation of elemental tin has also been

325 identified to be more polluting than that of elemental aluminium. Elemental tin is refined from tin metal, which

326 could be prepared from cassiterite mining and further processing. Elemental aluminium is prepared from primary

327 and secondary aluminium metal. To obtain primary aluminium, bauxite is mined and concentrated into aluminium

328 hydroxide, which is then purified to aluminium oxide. Primary aluminium could be yielded by refining aluminium

329 oxide. Secondary aluminium metal, which is recovered from scrap, is another source of elemental aluminium. The

330 LCA study on metals conducted by Nuss and Eckelman (2014) revealed that the preparation of elemental tin

331 performed less environmentally favourable than that of elemental aluminium in global warming potential,

332 cumulative energy demand, acidification, eutrophication and human toxicity. Such findings corroborated the

333 higher environmental impacts from $SnCl_4$ production than that from $AlCl_3$ in this study.

334 3.4.4   HMF production

335   The conversion of food waste to HMF avoided the impacts of conventional HMF production process,

336 which involved the inputs of crop-derived fructose syrup , acid catalyst, and energy supply (Fachri et al., 2015).

337 The different yields of HMF obtained offered environmental benefits that compensated for 3.86% of the

338 environmental burdens in the most polluting scenario and 36.86% of those in the most environmentally favourable

339 scenario, respectively. The HMF yield could be increased by increasing the reaction temperature (S1 compared

340 to S2), lengthening the reaction time (S3 compared to S2) and using acetone as the co-solvent (S5 and S6).

341 However, in S1 and S3, the environmental benefits from higher HMF yield could not compensate for the increased

342 energy consumption for heating. In S5, the use of $SnCl_4$ caused significant environmental drawbacks that

343 overwhelmed the environmental benefits of the scenario. After balancing the damages and benefits, S4 was

344 revealed to be the most environmentally favourable option.

345

4. Conclusions

To inform the decision-making on the selection of the best food waste valorisation option, this study developed an LCA framework for evaluating the environmental performance of the food waste valorisation scenarios by including the major processes of the utilization of solvent, co-solvents, catalysts, energy and the recovery of HMF. The LCA conducted in this study assessed eight scenarios of food waste valorisation via catalytic conversion, and concluded that S4 is the most polluting scenario while S6 is the most environmentally favourable option. The use of a less polluting catalyst ($AlCl_3$) and co-solvent (acetone), as well as the relatively high yield of HMF (27.9 Cmol%), provided S6 the superior environmental performance. Metal depletion impacts, which were attributed mainly to the production of metal chlorides catalyst, were the highest among the categories, followed by the toxicity impacts (marine ecotoxicity, freshwater toxicity, and human toxicity) which were contributed mostly by the production of organic co-solvents. The energy and $SnCl_4$ catalyst consumptions were the most dominant factors of the environmental impacts in most of the scenarios.

To keep the consistence of the framework while the detailed economic information about catalyst recycling was unavailable, only the environmental aspect was considered in this study. However, when the development of such valorisation process become more mature, and the information is more readily available in the future, the inclusion of the economic aspect is expected, so that a more comprehensive decision-supporting tool could be developed. The LCA in this study acts as an early milestone for guiding the selection of the best valorisation process, thus contributing to the development of the waste valorisation systems.

5. Acknowledgement

13

References

368 Abdullah NH, Mohamed N, Sulaiman LH, et al (2016) Potential Health Impacts of Bauxite Mining in Kuantan.
370 Malays J Med Sci MJMS 23:1–8

371 ACS (2017) What's New in Phenol Production? In: Am. Chem. Soc.
372 https://www.acs.org/content/acs/en/pressroom/cutting-edge-chemistry/what-s-new-in-phenol-
373 production-.html. Accessed 27 Dec 2017

374 Aliyu AS, Mousseau TA, Ramli AT, Bununu YA (2015) Radioecological impacts of tin mining. Ambio 44:778–
375 787. doi: 10.1007/s13280-015-0677-1

376 Althaus H-J, Hischier R, Osses M, et al (2007) Life Cycle Inventories of Chemicals. Dubendorf

377 Arogunjo AM, Höllriegl V, Giussani A, et al (2009) Uranium and thorium in soils, mineral sands, water and food
378 samples in a tin mining area in Nigeria with elevated activity. J Environ Radioact 100:232–240. doi:
379 10.1016/j.jenvrad.2008.12.004

380 Bardhan SK, Gupta S, Gorman ME, Haider, MA (2015) Biorenewable chemicals: Feedstocks, technologies and
381 the conflict with food production. Renew Sust Energ Rev 51:506–520

382 Bertau M, Offermanns H, Plass L, et al (2014) Methanol: The Basic Chemical and Energy Feedstock of the Future:
383 Asinger's Vision Today. Springer Science & Business Media

384 Blonk Consultants (2015a) Agri-footprint 2.0 - Part 1: Methodology and basic principles

385 Blonk Consultants (2015b) Agri-footprint 2.0 - Part 2: Description of data

386 Capello C, Fischer U, Hungerbühler K (2007) What is a green solvent? A comprehensive framework for the
387 environmental assessment of solvents. Green Chem 9:927–934. doi: 10.1039/B617536H

388 Capello C, Wernet G, Sutter J, et al (2009) A comprehensive environmental assessment of petrochemical solvent
389 production. Int J Life Cycle Assess 14:467–479. doi: 10.1007/s11367-009-0094-4

390 Chen SS, Maneerung T, Tsang DCW, Ok YS, Wang CH (2017) Valorization of biomass to hydroxymethylfurfural,
391 levulinic acid, and fatty acid methyl ester by heterogeneous catalysts. Chem Eng J 328:246-273.

14

392   Dashtban M, Gilbert A, Fatehi P (2014) Recent advancements in the production of hydroxymethylfurfural. RSC

393   Adv 4:2037–2050

394   Domínguez de María P, Guajardo N (2017) Biocatalytic valorization of furans: Opportunities for inherently

395   unstable substrates. ChemSusChem 10:4123–4134

396   Fachri BA, Abdilla RM, Bovenkamp HH van de, et al (2015) Experimental and kinetic modeling studies on the

397   sulfuric acid catalyzed conversion of D-Fructose to 5-Hydroxymethylfurfural and levulinic acid in water.

398   ACS Sustain Chem Eng 3:3024–3034

399   Faggian F, Faggian V (2014) Combining the European chemicals regulation and an (eco)toxicological screening

400   for a safer membrane development. J Clean Prod 83:404–412

401   FAO, 2013. Food Wastage Footprint: Impacts on Natural Resources—Summary Report. Food and Agriculture

402   Organization of the United Nations.

403   Franklin Associates (2011) Cradle-to-gate Life Cycle Inventory of Nine Plastic Resins and Four Polyurethane

404   Precursors

405   Golden JS, Handfield RB (2014) Why biobased? Opportunities in the emerging bioeconomy.

406   http://www.biopreferred.gov/files/WhyBiobased.pdf. Accessed 3 Jan 2018

407   Gonzalez-Garcia S, Gullon B, Moreira MT (2017) Environmental assessment of biorefinery processes for the

408   valorization of lignocellulosic wastes into oligosaccharides. J Clean Prod.

409   http://dx.doi.org/10.1016/j.jclepro.2017.02.164

410   Government of Canada CC for OH and S (2017) Organic Peroxides - Hazards : OSH Answers.

411   http://www.ccohs.ca/. Accessed 20 Dec 2017

412   Guan G, Guan G (2012) Development of Advanced Biodiesel Fuel Production Process. J Jpn Pet Inst 55:171–181

413   Gustavsson, J., Cederberg, C., Sonesson, U., van Otterdijk, R., Meybeck, A., 2011. Global Food Losses and Food

414   Waste. Food and Agriculture Organization of the United Nations, Rome.

415   Hallare A, Nagel K, Köhler H-R, Triebskorn R (2006) Comparative embryotoxicity and proteotoxicity of three

416   carrier solvents to zebrafish (Danio rerio) embryos. Ecotoxicol Environ Saf 63:378–388. doi:

417   10.1016/j.ecoenv.2005.07.006

418   HK ENB (2014) Future Fuel Mix for Electricity Generation Consultation Document

419   HKEPD (2016) Monitoring of solid waste in Hong Kong – waste statistics for 2015. Hong Kong Environmental

420   Protection Department. https://www.wastereduction.gov.hk/sites/default/files/msw2015.pdf. Accessed

421   11 November 2017

422   Huijbregts MA., Steinmann ZJN, Elshout PMF, et al (2016) ReCiPe 2016 A harmonized life cycle impact

423   assessment method at midpoint and endpoint level Report I: Characterization. Netherlands

424   ISO 14040 (1997) Environmental management – life cycle assessment – principles and framework. International

425   Standards Organisation, Geneva

426   ISO 14040 (2006) Environmental management - life cycle assessment – principles and framework. International

427   Standards Organisation, Geneva

428   ISO 14044 (2006) Environmental management - life cycle assessment – requirements and guidelines. International

429   Standards Organisation, Geneva

430   Kargbo DM (2010) Biodiesel production from municipal sewage sludges. Energy Fuels 24:2791–2794

431   Karka P, Papadokonstantakis S, Kokossis A (2017) Cradle-to-gate assessment of environmental impacts for a

432   broad set of biomass-to-product process chains. Int J Life Cycle Assess 22:1418–1440

433   Kläusli T (2014) AVA Biochem: commercialising renewable platform chemical 5-HMF. Green Process Synth

434   3:235–236

435   Matsakas L, Gao Q, Jansson S, Rova U, Christakopoulos P (2017) Green conversion of municipal solid wastes

436   into fuels and chemicals. Electron J Biotechn 26:69–83

437   Mohan SV, Nikhil GN, Chiranjeevi P, Reddy CN, Rohit MV, Kumar AN, Sarkar O (2016) Waste biorefinery

438   models towards sustainable circular bioeconomy: Critical review and future perspectives. Bioresour

439   Technol 215:2–12

440   Mukherjee A, Dumont MJ, Raghavan V (2015) Review: Sustainable production of hydroxymethylfurfural and

441   levulinic acid: Challenges and opportunities. Biomass Bioenergy 72:143–183

442   Nanda S, Azargohar R, Dalai AK, Kozinski JA (2015) An assessment on the sustainability of lignocellulosic

443   biomass for biorefining. Renew Sust Energ Rev 50:925–941

444   Nuss P, Eckelman MJ (2014) Life Cycle Assessment of Metals: A Scientific Synthesis. PLOS ONE 9:e101298.

445   doi: 10.1371/journal.pone.0101298

446 Parajuli R, Knudsen MT, Birkved M, Djomo SN, Corona A, Dalgaard T (2017) Environmental impacts of

447   producing bioethanol and biobased lactic acid from standalone and integrated biorefineries using a

448   consequential and an attributional life cycle assessment approach. Sci Total Environ 598:497–512

449 PRé Sustainability ReCiPe | PRé Sustainability. In: ReCiPe PRé Sustain. https://www.pre-

450   sustainability.com/recipe. Accessed 20 Dec 2017

451 Rout PK, Nannaware AD, Prakash O, Kalra A, Rajasekharan R (2016) Synthesis of hydroxymethylfurfural from

452   cellulose using green processes: A promising biochemical and biofuel feedstock. Chem Eng Sci 142:318-

453   346

454 Schwartz TJ, O'Neill BJ, Shanks BH, Dumesic JA (2014) Bridging the chemical and biological catalysis gap:

455   challenges and outlooks for producing sustainable chemicals. ACS Catal 4:2060–2069

456 Sheldon, RA (2016) Green chemistry, catalysis and valorization of waste biomass. J Mol Catal A: Chem 422:3-

457   12

458 Smith KS, Huyck HL (1999) An overview of the abundance, relative mobility, bioavailability, and human toxicity

459   of metals. Environ Geochem Miner Depos 6:29–70

460 Soroko I, Bhole Y, Livingston AG (2011) Environmentally friendly route for the preparation of solvent resistant

461   polyimide nanofiltration membranes. Green Chem 13:162–168. doi: 10.1039/C0GC00155D

462 USEIA (2017) International energy outlook 2017. U.S. Energy Information

463   Administration.https://www.eia.gov/outlooks/ieo/. Accessed 11 November 2017

464 van Putten RJ, van der Waal JC, Rasrendra CB, Heeres HJ, de Vries JG (2013) Hydroxymethylfurfural, A versatile

465   platform chemical made from renewable resources. Chem Rev 113:1499–1597

466 Vandeeersch T, Alvarenga RAF, Ragaert P, Dewulf J (2014) Environmental sustainability assessment of food

467   waste valorization options. Resour Conserv Recy 87:57–64

468 Woon KS, Lo IMC, Chiu SLH, Yan DYS (2016) Environmental assessment of food waste valorization in

469   producing biogas for various types of energy use based on LCA approach. Waste Manage 50:290–299

470 Xiong, X., Yu, I.K.M., Cao, L., Tsang, D.C.W., Zhang, S., Ok, Y.S. A review of biochar-based catalysts for

471   chemical synthesis, biofuel production, and pollution control. Bioresource Technol., 2017, 246, 254-270.

472 Yu, I.K.M. and Tsang, D.C.W. (2017d) Conversion of biomass to hydroxymethylfurfural: A review of catalytic

473   systems and underlying mechanisms. Bioresource Technol. 238, 716-732

474 Yu, I.K.M., Tsang, D.C.W., Chen, S.S., Wang, L., Hunt, A.J., Sherwood, J., Vigier, K.D.O., Jérôme, F., Ok, Y.S.
475   and Poon, C.S. (2017a) Polar aprotic solvent-water mixture as the medium for catalytic production of
476   hydroxymethylfurfural (HMF) from bread waste. Bioresour. Technol. 245, 456-462.

477 Yu, I.K.M., Tsang, D.C.W., Yip, A.C.K., Chen, S.S., Ok, Y.S., Poon, C.S. (2016) Valorisation of food waste into
478   hydroxymethylfurfural: Dual role of metal ions in successive conversion steps. Bioresour. Technol. 219,
479   338-347.

480 Yu, I.K.M., Tsang, D.C.W., Yip, A.C.K., Chen, S.S., Ok, Y.S., Poon, C.S. (2017b) Valorisation of starchy,
481   cellulosic, and sugary food waste into hydroxymethylfurfural by one-pot catalysis. Chemosphere 184,
482   1099-1107.

483 Yu, I.K.M., Tsang, D.C.W., Yip, A.C.K., Chen, S.S., Wang, L., Ok, Y.S., Poon, C.S. (2017c) Catalytic
484   valorisation of starch-rich food waste into hydroxymethylfurfural (HMF): controlling relative kinetics
485   for high productivity. Bioresour. Technol. 237, 222–230.

486

487   Table 1 Valorisation of Lignocellulosic biomass to HMF

Biomass	Solvent	Catalyst	Temp (°C)	Time	HMF yield (%)	References
Corn stover	DMA-LiCl; [EMIM]Cl	$CrCl_2$; HCl	140	2 h	48	Binder & Raines, 2009
Corn stover	DMA-LiCl; [EMIM]Cl	$CrCl_2$	140	6 h	16	Binder & Raines, 2009
Cassava waste	Acetone; DMSO; Water	Sulfonated carbon-based catalyst	250	1 min	12.1	Daengprasert et al., 2011
Corn stalk	[BMIM][Cl]	corn stalk-derived carbonaceous solid acid catalyst	150	30 min	44.1	Yan et al., 2014
Raw corn stalk	GVL; Water	PTSA-POM	190	100 min	19.5	Xu et al., 2015
Corn stalk	GVL	SC-CCA	210	1 h	2.2	Zhang et al., 2016
Sugarcane bagasse	NMP	SPPTPA-1	140	1 h	19.8	Mondal et al., 2015
Sugarcane bagasse	DMA-LiCl	SPPTPA-1	140	20 min	5.7	Mondal et al., 2015
Sugarcane bagasse	DMSO	SPPTPA-1	140	1 h	18.8	Mondal et al., 2015
Rice straw	[BMIM][Cl]	$CrCl_3$	200	3 min	47	Zhang, 2010
Rice straw	[MIM]HSO_4	N/A	160	30 min	7.9	Yokohama & Miyafuji, 2016
Bread waste	ACN; Water	$SnCl_4$	140	10 min	26	Yu et al., 2017
Bread waste	Acetone; Water	$SnCl_4$	140	10 min	26	Yu et al., 2017
Bread waste	DMSO; Water	$SnCl_4$	140	1 h	27.4	Yu et al., 2017
Bread waste	THF; Water	$SnCl_4$	140	2 h	8	Yu et al., 2017

488

489

19

490

Table 2 LCI of food waste valorisation

	Process input					Process output
	Food waste (1 g)	Solvent (10 mL)	Co-solvent (10 mL)	Catalyst	Electricity (Wh)	HMF yield (g)
S1	Bread waste	Water	DMSO	$SnCl_4$; 0.289g	100.0	0.214
S2	Bread waste	Water	DMSO	$SnCl_4$; 0.289g	50.0	0.126
S3	Bread waste	Water	DMSO	$SnCl_4$; 0.289g	150.0	0.199
S4	Bread waste	Water	THF	$SnCl_4$; 0.289g	400.0	0.109
S5	Bread waste	Water	Acetone	$SnCl_4$; 0.289g	33.3	0.191
S6	Bread waste	Water	Acetone	$AlCl_3$; 0.148g	100.0	0.203
S7	Rice waste	Water	DMSO	$SnCl_4$; 0.289g	100.0	0.227
S8	Fruit waste	Water	DMSO	$SnCl_4$; 0.289g	50.0	0.137

491

492
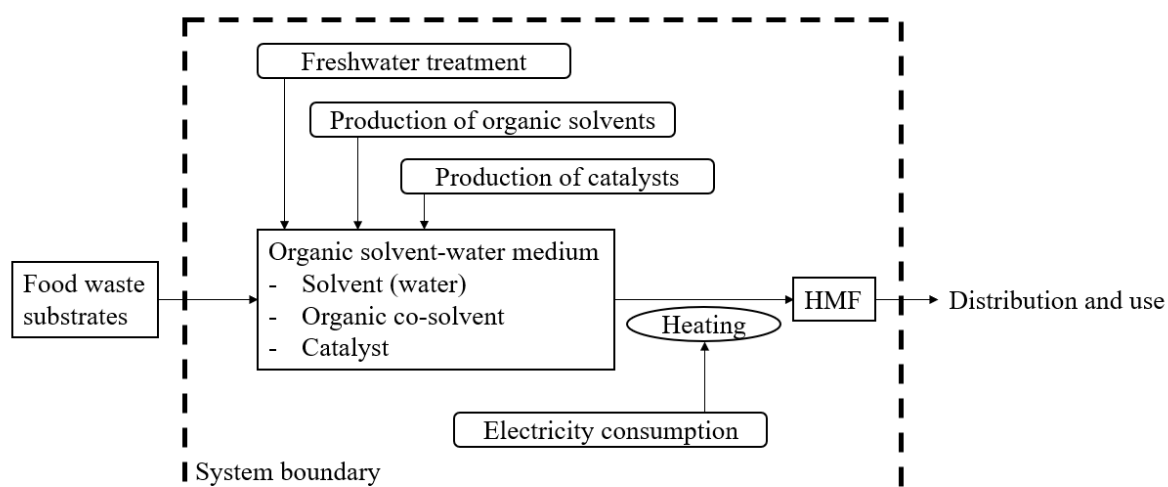
493

494   Figure 1 System boundary of food waste valorisation LCA
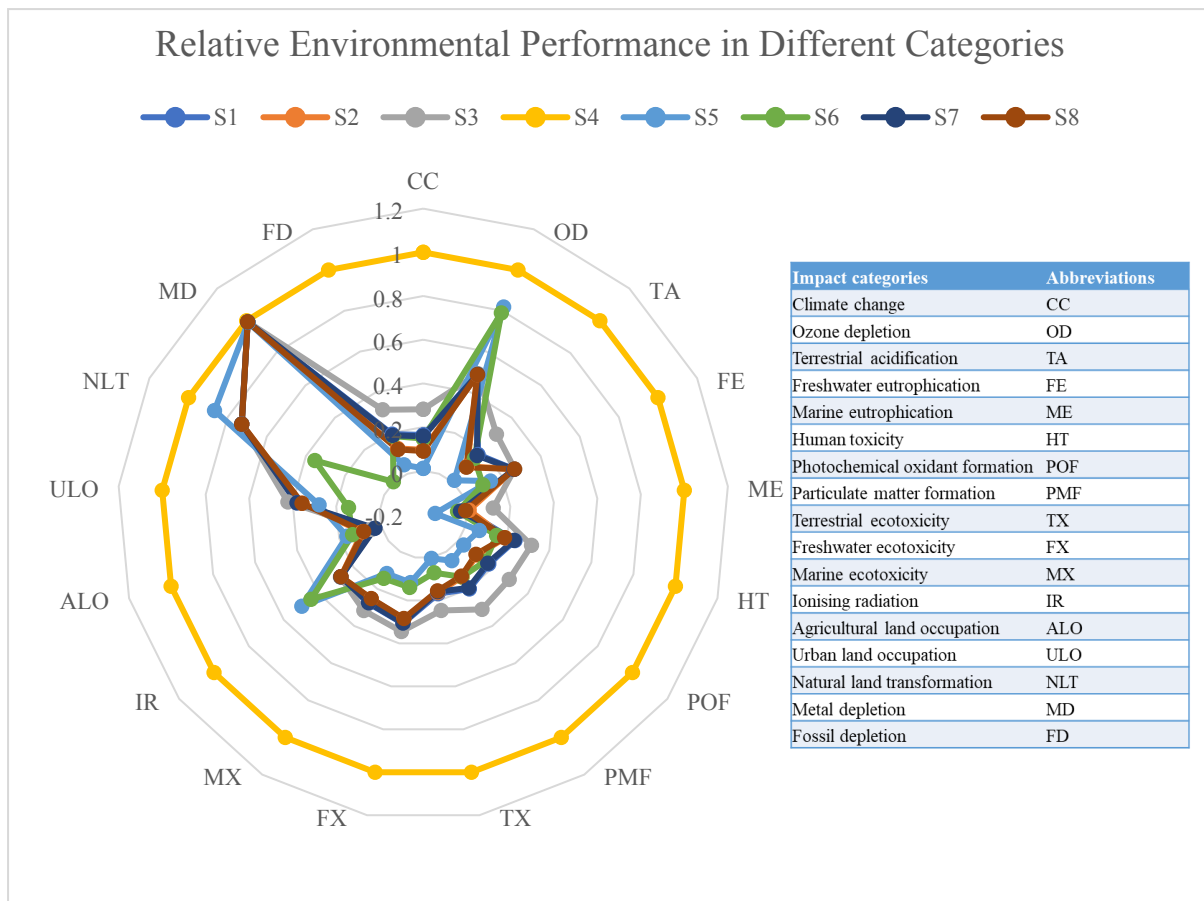
495

496

497

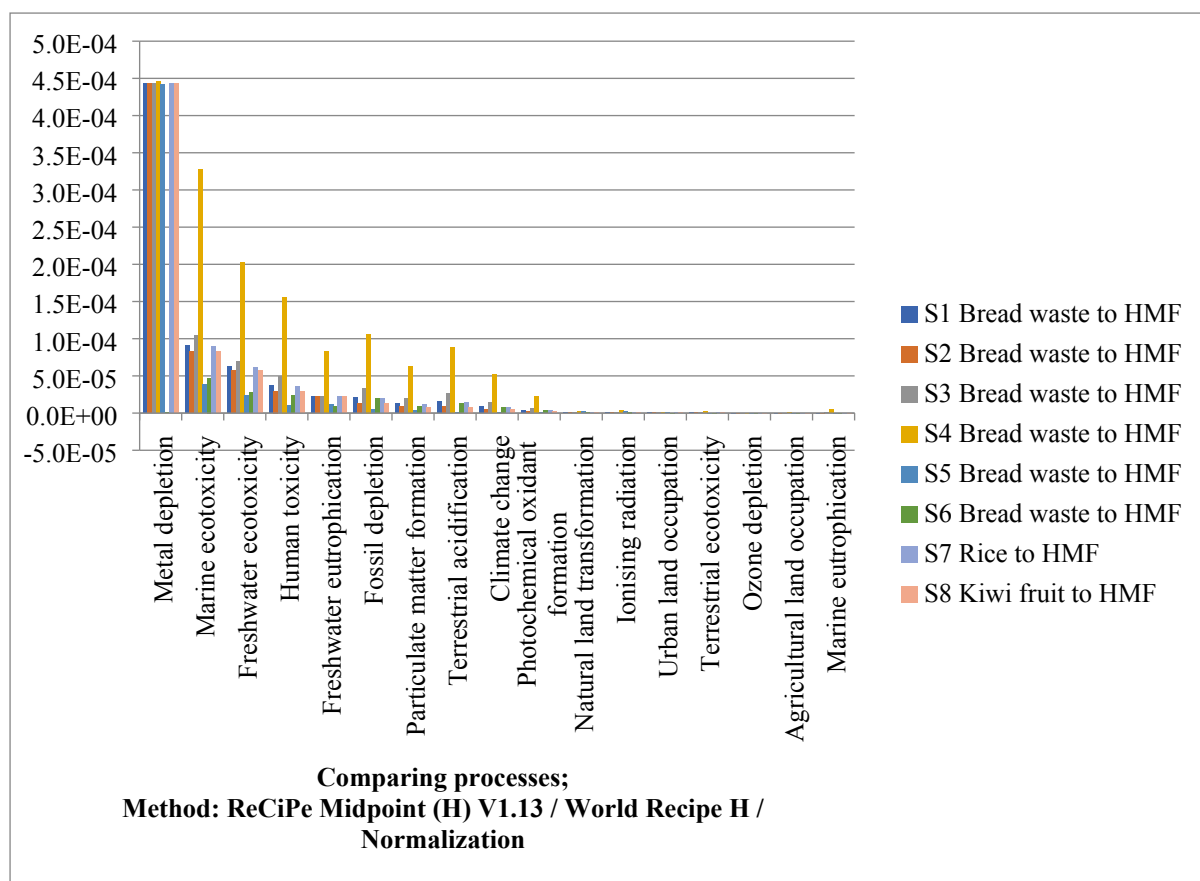**Relative Environmental Performance in Different Categories**

Legend: S1 · S2 · S3 · S4 · S5 · S6 · S7 · S8

Impact categories	Abbreviations
Climate change	CC
Ozone depletion	OD
Terrestrial acidification	TA
Freshwater eutrophication	FE
Marine eutrophication	ME
Human toxicity	HT
Photochemical oxidant formation	POF
Particulate matter formation	PMF
Terrestrial ecotoxicity	TX
Freshwater ecotoxicity	FX
Marine ecotoxicity	MX
Ionising radiation	IR
Agricultural land occupation	ALO
Urban land occupation	ULO
Natural land transformation	NLT
Metal depletion	MD
Fossil depletion	FD

498   Figure 2 Relative environmental performance in LCA categories

499

500

501   Figure 3 Normalized LCA results
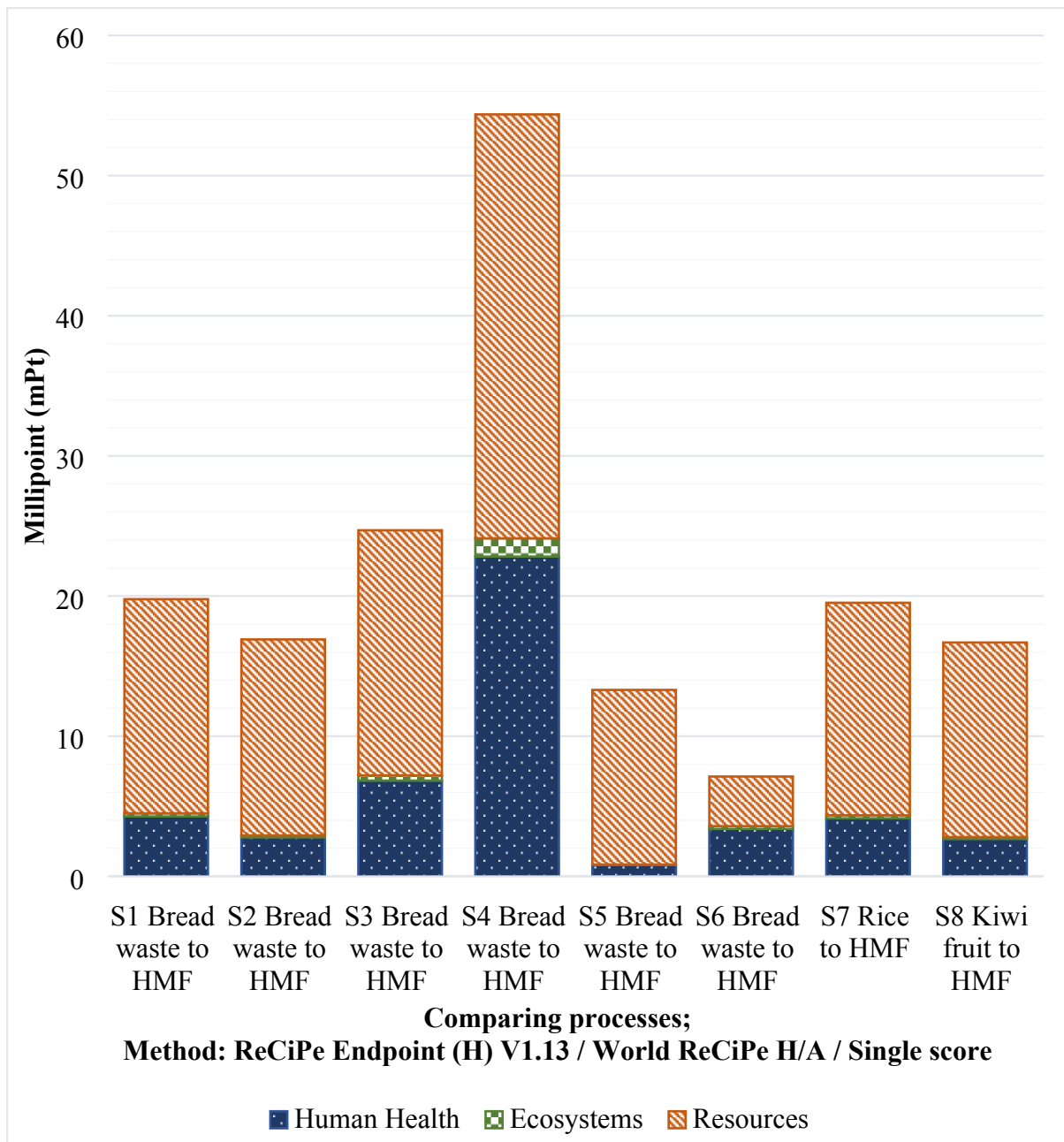
502

503

504   Figure 4 Single score LCA results
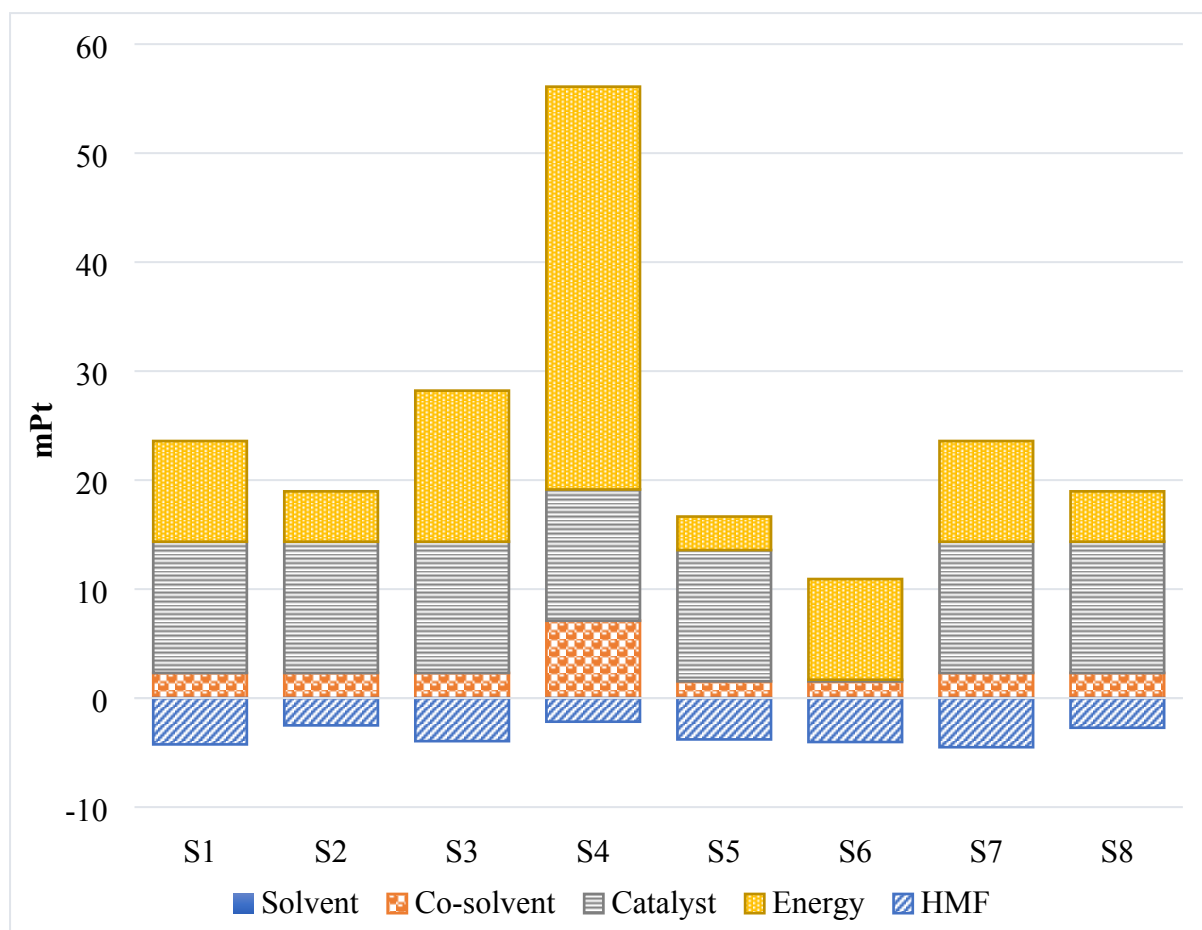
505

506

507 Figure 5 Process contributions to LCA results

508

Highlights

1. A Life-cycle analysis framework was developed to assess the environmental performance of eight food waste valorisation scenarios.

2. Experimental data were analysed for building the life-cycle inventory.

3. The conversion of bread waste using water-acetone medium was revealed to be the most environmental friendly option.

4. The energy consumption and the utilisation of $SnCl_4$ catalyst were the predominant contributors to the environmental impacts.