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**Bioethanol from corn stover – Integrated environmental impacts of alternative biotechnologies**

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16 **Abstract**

17 The environmental impact potentials of producing bioethanol from corn stover were systematically assessed  
18 by life cycle assessment using datasets reported in the literature. The 15% best datasets with respect to  
19 global warming potential for seven different technological configurations were extracted from a published  
20 database reviewing 474 publications on converting corn stover to bioethanol. A total of 10 impact categories  
21 were evaluated. The impacts included both environmental loads and savings, and generally ranged from  
22  $-0.1$  to  $0.1$  person-equivalent per ton of dry corn stover for most non-toxic impacts and  $-0.2$  to  $0.5$  person-  
23 equivalent for toxic impacts. Fossil fuel substitution with bioethanol provided savings in most impact  
24 categories, and so did the energy recovered from the residues, while enzyme production was a significant  
25 load. The treatment and discharge of effluent from the liquid residues may constitute a significant load to  
26 the environment. Based on the cumulative probabilities of overall environmental performance together with  
27 the bioethanol amount produced, the prioritisation of technologies for further development should be as  
28 follows: steam explosion (S4) and ammonia-based (S6) technologies as the highest priorities with  
29 approximately 100% and 40% probabilities to have savings in non-toxic and toxic impacts, respectively;  
30 acid (S1), alkaline (S2) and fungi (S7) technologies as medium priorities and solvent-based (S3) and liquid  
31 hot water (S5) technologies have the lowest priorities. We suggest the integration of life cycle assessment  
32 modelling to the research and development of biofuel production from biomass waste to ensure that the  
33 technologies being developed for full-scale applications are sustainable.

34  
35 **Keywords**

36 bioethanol; life cycle assessment; integrated environmental impacts; probability analysis; sustainability  
37 quantification; corn stover

38

## 39 **1. Introduction**

40 Bioethanol is used as green fuel in many countries, such as the USA, Brazil and India (Morales et al.,  
41 2015, Soam et al., 2016, Wojtusik et al., 2016). It is often used as an amendment to gasoline to reduce the  
42 fossil content of the fuel and thereby lessen the global warming impact of transportation. To date, nearly all  
43 bioethanol is produced by first-generation biotechnologies using corn, sugarcane or molasses as feedstocks  
44 (Zabed et al., 2016). Given that these types of feedstocks are either grown primarily for producing  
45 bioethanol or could have served as animal fodder, their label of being green and contributing to savings in  
46 fossil CO<sub>2</sub> emissions has been severely questioned (Jung et al., 2017, Abo et al., 2019). This has led to  
47 interest in using waste such as agricultural residues for producing bioethanol, and significant savings in  
48 global warming potential have been supposed when compared to fossil fuels (Dutta et al., 2014, Chang et  
49 al., 2017). Using corn stover, the residue generated by corn processing, has gained great interest in the field  
50 of bioethanol production because of its availability in very large quantities and at low costs, though corn  
51 stover has also been partly used for tilling down or as animal fodder. However, the technologies for  
52 producing bioethanol from corn stover are primarily at the experimental level (Zhao et al., 2018); the  
53 literature barely revealed any scientific reports on full scale plants in operation (Zhao et al., 2018). The  
54 challenge is that the corn stover is not easily converted to bioethanol because of its structure and high lignin  
55 content, and many approaches have been reported on how to pretreat and hydrolyse corn stover prior to  
56 saccharification and fermentation to bioethanol (Loow et al., 2016).

57 Several pretreatment processes have been developed and investigated for the disruption of  
58 lignocellulosic structures in order to make the polysaccharides available for further conversion (Solarte-  
59 Toro et al., 2019). This includes physical, chemical, biological and other processes (Capolupo and Faraco,  
60 2016, Kumar et al., 2016). However, given the overall aim of supporting sustainable development by  
61 substituting fossil fuels with bioethanol and thereby reducing greenhouse gas emissions from the transport  
62 sector, balancing of the ethanol production and the overall environmental performance including global  
63 warming potential is crucial for technology selection and optimisation. Life cycle assessment (LCA) has  
64 been acknowledged as an effective approach for addressing the potential environmental impacts of the  
65 technological management systems for products, services and solid waste. In recent years, many studies  
66 have conducted LCA on systems of bioethanol production with specific focuses, including different  
67 feedstocks (Chang et al., 2017, Rathnayake et al., 2018), typical conversion technologies (Zhu et al., 2015,  
68 Mandegari et al., 2017, Olofsson et al., 2017) and local biorefinery strategies (Farahani and Asoodar, 2017,  
69 Zucaro et al., 2018). However, relevant studies often investigated only one or few technologies for  
70 converting lignocellulosic waste to bioethanol (Hong et al., 2015, Guerrero and Munoz, 2018). In many  
71 cases, the studied LCA systems did not cover the full processes including pretreatment, hydrolysis,  
72 fermentation and ethanol substitution; in addition, only one dataset or inconsistent data sources for  
73 individual processes were often used (Trivedi et al., 2015, da Silva et al., 2019). Considering the full

74 technologies for converting corn stover to bioethanol, residue treatment and estimating the uncertainty and  
75 robustness are critical and a prerequisite for comparison of technologies (Bairamzadeh et al., 2018). These  
76 aspects have been involved in only few studies (Pourhashem et al., 2013, Murphy and Kendall, 2015).  
77 Despite the many papers addressing the environmental impacts and uncertainties of bioconversion  
78 technologies for cellulosic bioethanol production (Spatari et al., 2010, Neupane et al., 2017), the current  
79 literature leaves a very scattered picture of which technologies for cellulosic bioethanol production are most  
80 successful in ethanol production and environmental performance given the significant variations in  
81 technical approaches and process parameters (Zhao et al., 2019).

82 After reviewing 474 scientific publications on corn stover conversion to bioethanol, Zhao et al. (2019)  
83 recently determined the global warming potential of 141 research-based datasets with relatively high  
84 completeness and/or consistency on producing bioethanol from corn stover, including upstream  
85 contributions (electricity, chemicals, enzymes, etc.), energy use and process emission during bioethanol  
86 production as well as savings obtained from utilising the residues for energy purposes and from bioethanol  
87 substituting for fossil gasoline. The experimental datasets were grouped into seven technological  
88 configurations according to the pretreatment approaches (acid, alkaline, solvent-based, steam explosion,  
89 liquid hot water, ammonia-based and fungi). Interestingly, only half of the datasets would provide savings  
90 in global warming potential. Moreover, when the energy recovery from solid and liquid residues was not  
91 included, only a quarter of the datasets provided savings in global warming. This suggested that not only  
92 selecting the right technology (which pretreatment to use) but also optimising the individual technology is  
93 important. Zhao et al. (2019) modeled global warming potentials by parametrising all key processes and  
94 establishing statistical distributions of the key parameters based on the reported data. This method enabled  
95 focusing on the best datasets, and considering the data representing the 15% best cases for each technology  
96 in terms of providing savings in global warming, Zhao et al. (2019) concluded that steam explosion and  
97 ammonia-based pretreatment statistically were the most promising technologies for converting 18%–22%  
98 of the corn stover into bioethanol (dry weight) and providing potential global warming savings of 850–  
99 1050 kg CO<sub>2</sub>-equivalence per 1000 kg of dry corn stover.

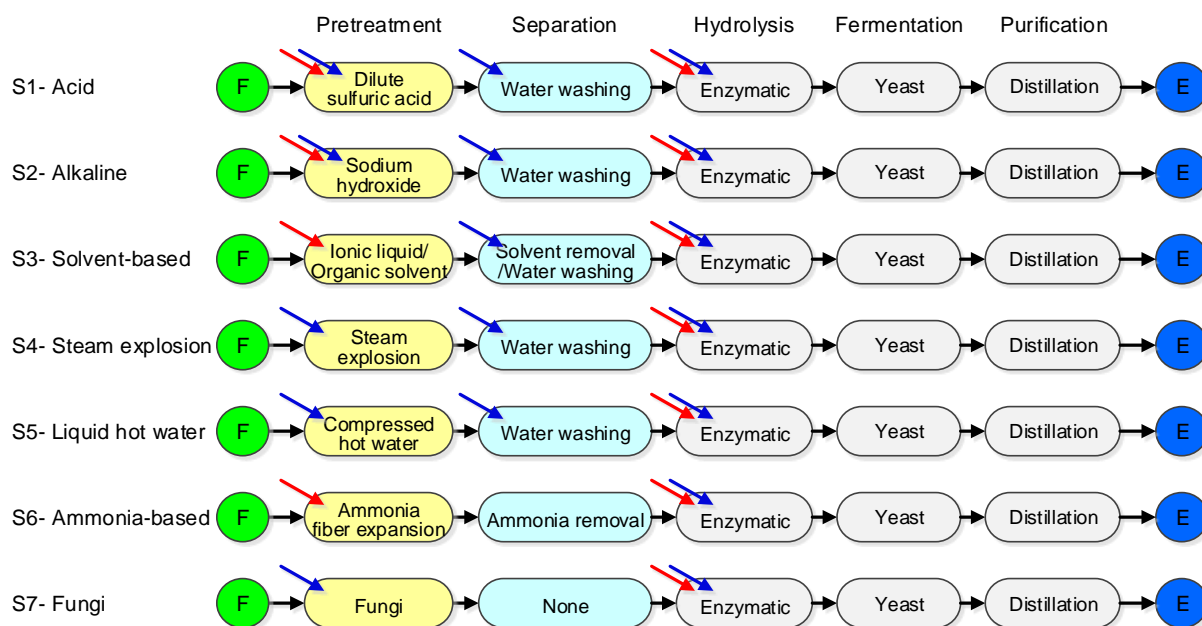
100 Savings in global warming contribution when producing bioethanol for substituting fossil fuels is a  
101 key environmental issue, and this issue was addressed by Zhao et al. (2019). However, to address  
102 sustainability in a broad context, all potential environmental impacts must be addressed. This aspect is  
103 particularly important to bioethanol production from corn stover because experimentally documented  
104 pretreatment technologies are considerably different in terms of approach and energy, chemical and enzyme  
105 consumption. These technologies all have highly variable environmental rucksacks associated with their  
106 production processes. This work provides for the first time a stringent LCA considering 10 potential  
107 environmental impacts of seven different technologies for producing bioethanol from corn stover. We  
108 conduct the LCA on the large dataset reported by Zhao et al. (2018) and the statistical parameter  
109 distributions extracted from the 141 most consistent datasets by Zhao et al. (2019). We focus on the 15%

best datasets within each technology with respect to global warming potential (GWP) established by Zhao et al. (2019), because we acknowledge that the key experimental parameters vary significantly and we do believe that technology development is always focusing on the best performing solutions. Here ‘best’ means providing the largest savings in GWP, which does not necessarily mean yielding the highest bioethanol output. The LCA study provides a thorough assessment of biorefinery technologies under development in terms of ethanol produced per ton corn stover and environmental impact potentials in global warming, acidification, nutrient enrichment, photochemical ozone formation, particulate matter, ecotoxicity and human toxicity. In addition, the process contribution to the impacts, the global sensitivity analysis of parameters and the probability analysis of integrated impact potentials were performed to provide comprehensive understandings on the alternative biotechnologies. The size and quality of the parametrised datasets established allow us to provide estimates of the robustness of the quantifications and an analysis of the most sensitive parameters governing the environmental aspects of producing bioethanol from corn stover. This contribution provides unprecedented quantitative insights into the technologies being developed for producing bioethanol from corn stover.

## 2. Approach and methods

### 2.1 Technological configurations and model system

Typical biotechnological configurations for converting corn stover to ethanol were identified in our previous review as combinations of pretreatment, hydrolysis, fermentation and purification (Zhao et al., 2018). According to the pretreatment step, the seven technological configurations involved in this study are briefly illustrated in Figure 1. The details are provided in a previous work (Zhao et al., 2018).



**Figure 1.** Main processes of the seven technological configurations for producing ethanol from corn stover (F: feedstock, i.e. corn stover; E: ethanol produced. The red arrows indicate the addition of chemicals

134 or enzymes; the blue arrows indicate water addition. The side streams for liquid and solid residue treatment  
135 are included in the life cycle system but not shown here.).

136 For the assessment of the integrated environmental impacts of the seven identified technological  
137 configurations, the technological systems for producing ethanol from corn stover were established as  
138 described in our previous study, and the functional unit was defined as the ‘biorefinery of 1 t (1000 kg dry  
139 matter) of corn stover with bioethanol as the major product’ (Zhao et al., 2019). The system boundary  
140 includes the collection, transportation and mechanical preparation of the corn stover, all the major processes  
141 of bioethanol production (including use of water, chemicals, enzymes and energy), the treatment of solid  
142 and liquid residues (including water, heat and power recovery from incineration and anaerobic digestion)  
143 and the downstream use of the produced ethanol (system diagram shown in the Supplementary Information)  
144 (Zhao et al., 2019). All the details of process description and system assumptions including ethanol  
145 substitution, energy consumption in common processes and treatment of liquid and solid residues are  
146 provided in a previous work (Zhao et al., 2019).

## 148 **2.2 Modelling tool and method for life cycle impact assessment**

149 LCA was performed with EASETECH, a specialised LCA model for assessing the environmental  
150 technologies of complex systems handling heterogeneous material flows (Clavreul et al., 2014). The  
151 method for life cycle impact assessment used in this study was in accordance with the recommendation of  
152 the International Reference Life Cycle Data System (ILCD) Handbook and considered the following impact  
153 categories: climate change (global warming potential), stratospheric ozone depletion, acidification,  
154 eutrophication (terrestrial), eutrophication (freshwater), eutrophication (marine), photochemical ozone  
155 formation (human health), ecotoxicity (freshwater), human toxicity (cancer effects), human toxicity (non-  
156 cancer effects), particulate matter, ionizing radiation (human health), and abiotic resource depletion (fossil  
157 and mineral). Land use and resource depletion of water were not included because of the strong  
158 geographical uncertainty; abiotic resource depletion (fossil and mineral) was also not included because of  
159 its irrelevance (mineral) and partial coverage in climate change (fossil). For comparison across categories,  
160 the results in this study were expressed in personal equivalents whereby the results are normalised by the  
161 annual impact from an average person. The normalisation factors of all the impact categories of the ILCD  
162 methods are listed in Table 1 (Laurent et al., 2013). Given the different recommendation levels of the impact  
163 categories on the basis of the quality of the characterisation model for each impact category (Hauschild et  
164 al., 2013), the results in this study are presented in terms of non-toxic and toxic categories separately.

165 The modelling was carried out as an attributional LCA. In addition, life cycle inventory data for  
166 background processes, including chemicals, heat, electricity and fuel production, were mainly selected from  
167 the Ecoinvent database (allocation at the point of substitution) (Ecoinvent, 2016). Global processes were  
168 used where possible because the study does not apply to any specific region. For processed used in multiple

169 scenarios, we employed the same process across the different scenarios. The details are found in (Zhao et  
 170 al., 2019).

171 **Table 1** Normalisation factors and impact categories of the ILCD methods used in this study (Hauschild  
 172 et al., 2013, Laurent et al., 2013)

ILCD Impact category	Unit	Normalisation factor	Recommendation level*
Climate change	kg CO <sub>2</sub> eq./PE/year	$8.10 \times 10^{+03}$	I
Stratospheric ozone depletion	kg CFC-11 eq. /PE/year	$4.14 \times 10^{-02}$	I
Acidification	mol H <sup>+</sup> eq. /PE/year	$4.96 \times 10^{+01}$	II
Eutrophication (terrestrial)	mol N eq. /PE/year	$1.15 \times 10^{+02}$	II
Eutrophication (freshwater)	kg P eq. /PE/year	$6.20 \times 10^{-01}$	II
Eutrophication (marine)	kg N eq. /PE/year	$9.38 \times 10^{+00}$	II
Photochemical ozone formation (human health)	kg NMVOC eq. /PE/year	$5.67 \times 10^{+01}$	II
Ecotoxicity (freshwater)	CTUe/PE/year	$6.65 \times 10^{+02}$	II/III
Human toxicity (cancer effects)	CTUh/PE/year	$5.42 \times 10^{-05}$	II/III
Human toxicity (non-cancer effects)	CTUh/PE/year	$1.10 \times 10^{-03}$	II/III
Particulate matter	kg PM <sub>2.5</sub> eq. /PE/year	$2.76 \times 10^{+00}$	I/II
Ionising radiation (human health)	kBq U <sup>235</sup> eq. (to air) /PE/year	$1.33 \times 10^{+03}$	II

173 \* Recommendation level: I, recommended and satisfactory; II, recommended but in need of some  
 174 improvements; III, recommended, but to be applied with caution (Hauschild et al., 2013).

175

### 176 2.3 Key process parameters and data distribution

177 To evaluate and compare the integrated environmental impacts of different technological  
 178 configurations, a parameterisation approach was applied to provide consistent flows (mass, solids, key  
 179 substrates and water) and material and energy accounts. A set of parameters was identified for the seven  
 180 technological configurations and used as input in the modelling in accordance with (Zhao et al., 2019). The  
 181 parameter descriptions are listed in Table 2.

182 In our previous work on global warming footprint of alternative biotechnologies for bioethanol  
 183 production from corn stover, the probability distribution of each parameter was obtained on the basis of the  
 184 141 individual datasets with relatively high completeness and consistency selected from 474 publications  
 185 (Zhao et al., 2019). The data quality and completeness were reported in Zhao et al. (2018, 2019). From  
 186 Monte Carlo simulations, ‘best-practice’ cases with the top 15% cumulative probability in terms of GWP



were identified to represent potential targets for improving the technologies of each configuration. The key parameters identified from global sensitivity analysis can be optimised according to the parameter probability distribution in the top 15% ‘best-GWP’ cases.

**Table 2** Parameters used in modelling the technological configurations for the bioconversion of corn stover into ethanol

Classification	Parameter	Unit	Description
Raw material composition	GPP_RM	%TS	Glucan proportion in raw materials
	XPP_RM	%TS	Xylan proportion in raw materials
	LPP_RM	%TS	Lignin proportion in raw materials
	APP_RM	%TS	Ash proportion in raw materials
	OPP_RM	%TS	Other proportion in raw materials
Solid loading	SL_PT	%wt	Total solid content in PT*
	SL_PW	%wt	Total solid content in post-wash
	SL_HL	%wt	Total solid content in HL*
	SL_FT	%wt	Total solid content in FT*
Conversion coefficient	GEF_PT	%	Glucan yield to solid phase in PT
	GEF_HL	%	Glucose yield to liquid phase in HL
	GEF_FT	%	Ethanol yield from glucose in FT
	XEF_PT	%	Xylan yield to solid phase in PT
	XEF_HL	%	Xylose yield to liquid phase in HL
	XEF_FT	%	Ethanol yield from xylose in FT
	LEF_PT	%	Lignin yield to solid phase in PT
Operational condition	T_PT	°C	Reaction temperature in PT
	MS_PT	Rpm	Mixing speed in PT
	MS_HL	Rpm	Mixing speed in HL
	MS_FT	Rpm	Mixing speed in FT
	RT_PT	H	Reaction time in PT
	RT_HL	H	Reaction time in HL
	RT_FT	H	Reaction time in FT
	PS		Pretreatment severity
Material consumption	CC_PT	kg/kg	Chemical consumption in PT
	CR_PT	%	Chemical recovery ratio in PT
	BA_HL	kg/kg	Enzyme consumption in HL
Energy consumption	EC_PT	kWh/kg	Electricity consumption in PT
	HC_PT	MJ/kg	Heat consumption in PT
	EC_HL	kWh/kg	Electricity consumption in HL
	HC_HL	MJ/kg	Heat consumption in HL
	EC_FT	kWh/kg	Electricity consumption in FT
	HR_PT	MJ/kg	Heat recovery in PT

\* PT, HL, and FT stand for pretreatment, hydrolysis, and fermentation, respectively.

With regard to the environmental concern of bioethanol production from corn stover, global warming reduction is of the highest priority in substituting fossil fuel. In this study investigating the integrated environmental impacts of alternative technologies with the best GWP performance, a specific probability distribution of each parameter was adopted on the basis of the parameter values of the top 15% ‘best-GWP’ cases. Normal (ND), lognormal (LD), uniform (UD) and triangular (TD) distributions were implemented by the statistical software Minitab 17.1 to appropriate parameters and entered into the EASETECH LCA

model (Bisinella et al., 2016). The cumulative distribution and fitting curve of each parameter in each technological configuration are shown in the Supplementary Information. The corn stover composition was identical across the technological configurations as follows: glucan 36.2%±3.2%, xylan 22.7%±4.2%, lignin 18.5%±3.7%, ash 5.0%±2.7% and others 17.6%±7.9% (Zhao et al., 2019).

## 2.4 Statistical approach and global sensitivity analysis

Based on the parameter distribution obtained from the top 15% ‘best-GWP’ cases, Monte Carlo method was implemented in EASETECH to randomly sample 5000 values within each parameter distribution and calculate the environmental impact results of each scenario. Apart from the statistical data including average value and deviation, the calculated results were further used to present the uncertainty describing the overall variation and the robustness of the results (Mullins et al., 2011) by constructing a frequency histogram and computing a probability distribution within a 95% confidence level (Zhao et al., 2019). Lognormal fitting was found to be appropriate for describing the distribution of all the results. Discernibility analysis was performed accordingly to show statistically how frequently one technology is better than another when both technologies are subject simultaneously to the same variations in common parameters (Bisinella et al., 2016). Priority classification was then provided for all the studied technologies according to their sustainability performance from a probability perspective, supplemented by considerations on the ethanol production.

Sensitivity was expressed by the sensitivity ratio calculated for each parameter in each scenario, by changing one parameter by 1% once at a time while keeping all the other parameters fixed at their basic values (Bisinella et al., 2016). The mode values calculated from the distribution of each parameter were used as the basic values of the tested parameters in sensitivity analysis. The global sensitivity analysis (also known as global importance analysis), which includes an analysis of the fundamental connections between the sensitivity and the uncertainty of individual parameters, was applied to present the contribution of each independent parameter to the result variance (Bisinella et al., 2016).

## 2.5 Scenario sensitivity analysis with different wastewater treatment processes

Compared with the other processes and data applied in this study, the selection of external processes of wastewater treatment was highly uncertain because no dataset exists specifically for wastewater from corn stover treatment. In addition, the contributions of wastewater treatment to eutrophication (freshwater, marine) and ecotoxicity were found extremely significant in some technological configurations as shown in Section 3.2. The choice of wastewater treatment process may thus significantly affect the results and conclusions of this study. This issue was addressed by performing a scenario sensitivity analysis using four different datasets for wastewater treatment as listed below. The mode values calculated from the distribution of each parameter were used as the basic values of the tested scenarios. Except for the external process of

wastewater treatment, all the parameters, including biogas production and energy recovery from AD, remained identical for all the scenarios.

- Scenario A applied an external process for municipal wastewater treatment from the Ecoinvent database (treatment of wastewater, average, capacity  $1 \times 10^9$  L/year). This process was the default external process applied in all assessments.

- Scenario B applied an external process from the Ecoinvent database representing the average wastewater treatment plant in the market, (market for wastewater, average). This process aimed at a general situation for wastewater treatment.

- Scenario C applied an external process from the Ecoinvent database for treating wastewater with a low level of pollutants (market for wastewater, unpolluted). Assuming that the effluent from the AD of the liquid residue from corn stover contains fewer pollutants than what is found in municipal wastewater, this process may appropriately represent the wastewater treatment.

- Scenario D assigned no external process of treating wastewater and no effluent (the biogas generation and energy recovery remained the same). With no wastewater treatment, this scenario represents a best case scenario because our inventory does not contain any information about the effluent content. In addition, no energy nor chemicals are used in the treatment. This scenario is no real alternative but a modelling step to assess the importance of wastewater treatment.

### 3. Results and discussion

Based on the parameter distribution from ‘best-GWP’ cases with the top 15% cumulative probability in terms of GWP, five levels of assessment were performed to reveal and compare the different process configurations for biorefining of corn stover to bioethanol in terms of (1) ethanol production and GWP, (2) environmental impact potentials in different categories, (3) the contribution of the main processes within a configuration to the different impact categories, (4) the global sensitivity analysis of parameters for different impact categories and (5) the probability analysis of integrated impact potentials. In addition, scenario sensitivity of wastewater treatment and process optimisation for alternative technologies are further discussed. The results provide a comprehensive understanding and potential guidance of a state-of-art biorefinery producing bioethanol, from an integrated environmental perspective.

#### 3.1 Ethanol production and GWPs for ‘best-GWP’ technologies

Large variation in parameters within and across the technologies for bioethanol production was observed in our previous study with 141 individual datasets (Zhao et al., 2018). It has been verified that using parameter distribution can mitigate the effects of parameter outliers reported in individual cases and provide general understanding on the bioethanol production and environmental impacts of alternative technologies (Zhao et al., 2019). By selecting the datasets that encompass the best 15% of the probability

distribution with respect to GWP, the parameter distribution can be narrowed in terms of GWP performance for the potential optimisation of each technological configuration. We believe that this method is a sound approach because focusing on the best parameter combinations to obtain the best results is common in any research and development activity. In this case the GWP performance is in focus as representing the best result.

Table 3 summarises the bioethanol production and carbon footprint of alternative technologies in terms of ‘best-GWP’. The full parameter distributions from the 141 datasets are available in our previous work (Zhao et al., 2019). The ‘best-GWP’ cases of all the alternative technologies showed significant savings ranging from  $-537$  to  $-1078$  kg CO<sub>2</sub>-eq /t dry corn stover in average. Technological configurations with steam explosion and ammonia-based pretreatment seem the best in reducing carbon footprint of bioethanol production from corn stover. The bioethanol production was also improved by 3% to 15% for all the alternative technologies in the ‘best-GWP’ cases. Technological configurations with ammonia-based and alkaline pretreatment show the highest ethanol production per ton of corn stover, consistent with the results of the individual cases (Zhao et al., 2019).

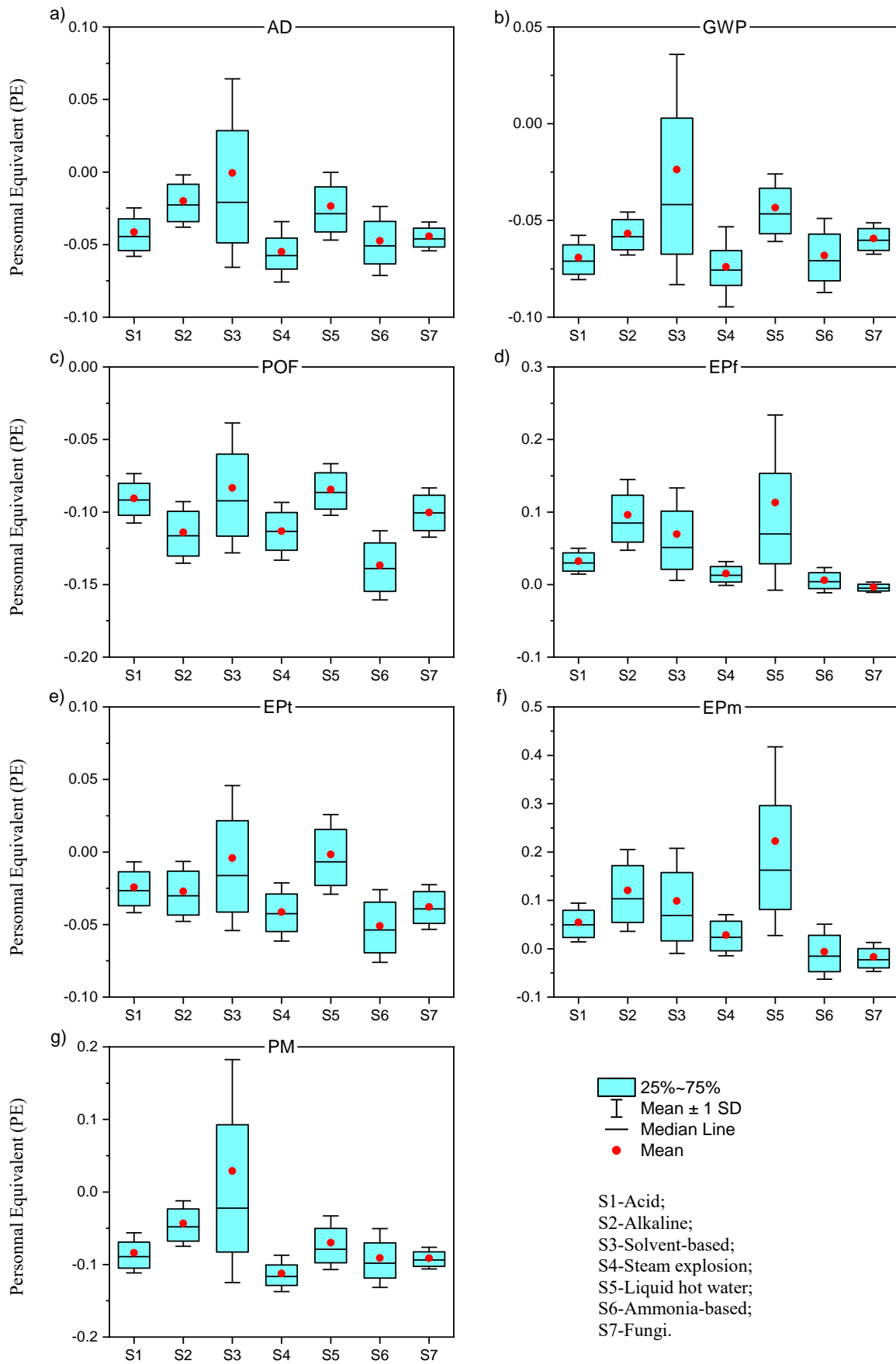
Given that the purpose of bioethanol production is to substitute fossil fuel and thus reduce the global warming impacts, the balancing of ethanol production and the overall carbon footprint is the key question for the development of the bioethanol production technologies. However, the potential impacts on other environmental categories are also important given complex technologies, use of chemicals and enzymes and the exchange with the energy system.

**Table 3** Bioethanol production and GWPs of alternative technologies in terms of best practice in GWP

Technological configuration	Bioethanol production (kg/t dry corn stover)	Carbon footprint (kg CO <sub>2</sub> -eq /t dry corn stover)
S1-Acid	149±39	-796±42
S2-Alkaline	195±35	-772±39
S3-Solvent based	175±67	-548±202
S4-Steam explosion	178±52	-1078±166
S5-Liquid hot water	148±28	-537±55
S6-Ammonia based	216±46	-945±92
S7-Fungi	155±24	-742±36

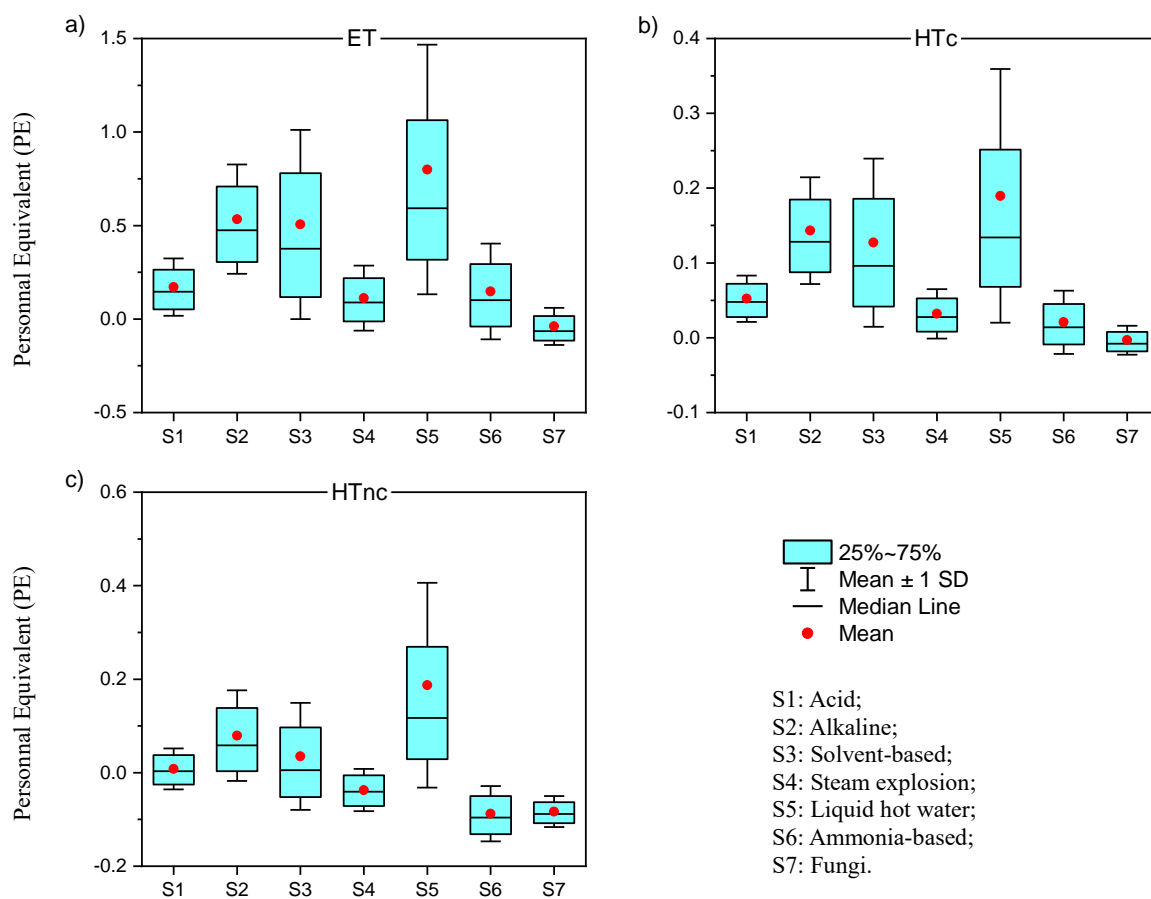
### 3.2 Environmental impact potentials in 10 categories of the technological configurations with parameter distribution of ‘best practice in GWP’

The normalised impacts of non-toxic and toxic categories with a 95% confidence level are shown in box figures with mean and median values and standard deviations (Figures 2 and 3, respectively) for ‘best-GWP’ cases. The detailed histograms of Monte-Carlo simulation for each category and each configuration can be found in the Supplementary Information.



297 **Figure 2.** Normalised environmental impact potentials in non-toxic categories of the seven  
 298 technological configurations with parameter distribution of ‘best practice in GWP’. a) AD: acidification; b)  
 299 GWP: global warming potential; c) POF: photochemical ozone formation; d) EPf: eutrophication,  
 300 freshwater; e) EPT: eutrophication, terrestrial; f) EPM: eutrophication, marine; g) PM: particulate matter.

301 In general, the normalised results (personal equivalents) of the toxic categories are numerically larger  
 302 than the results of the non-toxic categories. However, ILCD assigns a different recommendation level to  
 303 the toxic categories indicating less reliability due to associated uncertainty (Hauschild et al., 2013). The  
 304 normalised results of non-toxic categories are moderate and all within the same order of magnitude (Figure  
 305 2). Thus, we chose to present the integrated environment impacts with non-toxic and toxic categories  
 306 separately, as shown in Figures 2 and 3, respectively. Stratospheric ozone depletion and ionising radiation  
 307 are not included because the personal equivalents of these two categories are marginal with at least one  
 308 order of magnitude lower than the other categories (details can be found in the Supplementary Information).



309 **Figure 3.** Normalised environmental impact potentials in toxic categories of the seven technological  
 310 configurations with parameter distribution of ‘best practice in GWP’. a) ET: ecotoxicity, freshwater; b) HTc:  
 311 human toxicity, cancer effects; c) HTnc: human toxicity, non-cancer effects.

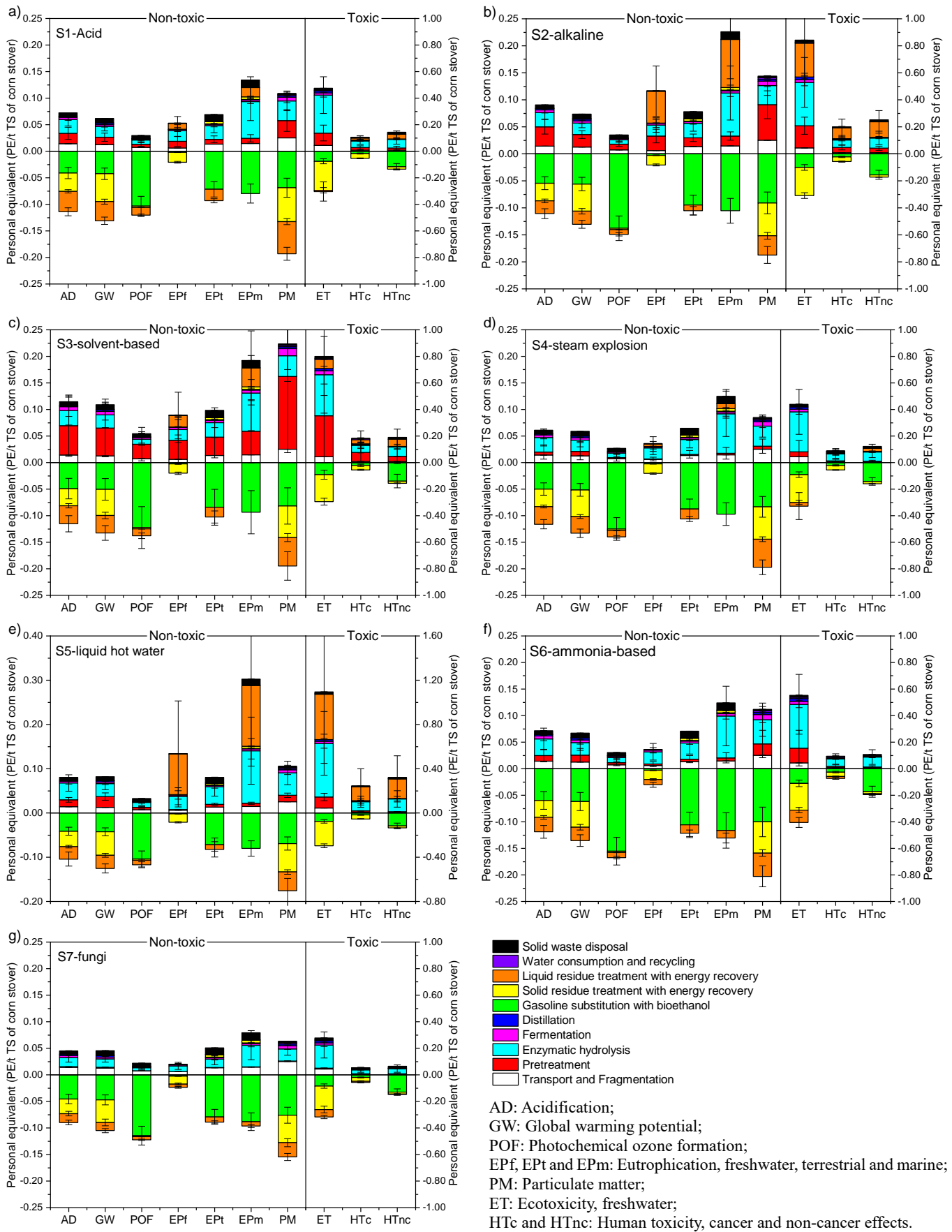
312  
 313 Specifically, the normalised impacts in non-toxic categories show good environmental benefits for  
 314 most categories and most configurations. This is mainly attributed to the net savings from ethanol  
 315 substituting fossil fuel and energy recovered from the residues. An exception to this is the results of  
 316 eutrophication to freshwater and eutrophication to marine water, which both show loads to the environment.  
 317 Tracing back to the life cycle inventory, the potential adverse impacts to water environment are derived  
 318 from the discharged pollutants in the treated liquid residues. However, due to the lack of data in liquid  
 319 residue treatment during bioethanol production, the wastewater treatment used in all the scenarios was an

external process for municipal wastewater treatment available in Ecoinvent (see Section 2.5). Of the same reason, the potential impacts to ecotoxicity to freshwater are significantly higher than those to other categories. The effects of the wastewater treatment are discussed in Section 3.6 based on the scenario sensitivity analysis. Among the technological configurations, S3 (solvent-based) show significant variations in most categories, and S5 (liquid hot water) display high variations in eutrophication to freshwater and eutrophication to marine and in the three toxic categories. The reason is that only a few very important parameters in S3 and S5 (e.g. solvent recovery ratio and enzyme consumption in hydrolysis) were optimised with the top 15% cumulative probability in terms of GWP, whereas some other parameters still held distributions with significant variation. This result contributed to the high variations in normalised impacts in the other categories.

### **3.3 Process contribution to environmental impact potentials in 10 categories**

The contributions of individual processes within each technological configuration to the environmental impact potentials are shown in Figure 4 (detailed data with average values and deviations can be found in the Supplementary files of data source). In general, pretreatment, enzymatic hydrolysis, gasoline substitution with bioethanol, as well as solid and liquid residue treatment with energy recovery are major contributors to most impact categories. The other processes including transport and fragmentation, fermentation, distillation, water consumption and recycling, as well as solid waste disposal show insignificant contributions to the impact potentials because of low energy and material consumption and few emissions.

Particularly, pretreatment shows minor contributions to POF, EPt, EPm and HTnc, but significant contributions to PM, AD, GW and ET. The reason is that its major impact to the environment is energy or material consumption rather than direct emissions. The environmental loads of pretreatment are significantly different between the technological configurations due to the diversity in pretreatment. Figures grouped in terms of environmental impact categories, for each technology configuration, can be found in the Supplementary Information. Huge amount of solvent consumption is the main reason for the loads of the technology based on solvent pretreatment (S3) to all the categories despite that the solvent recovery ratio has already been optimised with respect to GWPs. Enzymatic hydrolysis also contributes as a load to all the categories for all the technological configurations, especially to EPm and ET because of the life cycle inventory for enzyme production. Enzyme production data are scarce in literature but important for the results. It is supposed that, in full scale operations, the enzymes can be produced with a lower impact than what is seen for general enzyme datasets published.



353 **Figure 4.** Process contribution to normalised environmental impact potentials in non-toxic and  
 354 categories of the seven technological configurations. a) S1-Acid; b) S2-Alkaline; c) S3-Solvent-based; d)  
 355 S4-Steam explosion; e) S5-Liquid hot water; f) S6-Ammonia-based; g) S7-Fungi. (Please note that S5 uses  
 356 different axis scales from the others.)



357 Gasoline substitution with bioethanol provides large savings in POF, EPt, EPm and HTnc. In addition,  
358 its contributions to the savings in AD, GW and PM are moderate and comparable with those of energy  
359 recovery from solid and liquid residue treatment. All these savings are from avoiding production and  
360 utilisation of fossil fuel. Gasoline substitution with bioethanol shows minimal impacts to EPf and HTc,  
361 attributed to the irrelevant or low emissions to be avoided.

362 Liquid residue treatment with energy recovery contributes to the savings in AD, GW, POF, EPt and  
363 PM to a certain extent. However, this process results in impact loads in EPf, EPm and ET in most  
364 technological configurations, especially in S2 (alkaline) and S5 (liquid hot water). The reason is that  
365 phosphate, nitrate and heavy metals from the wastewater treatment are potentially discharged into water  
366 environment. The significance of the wastewater treatment is discussed in Section 3.5.

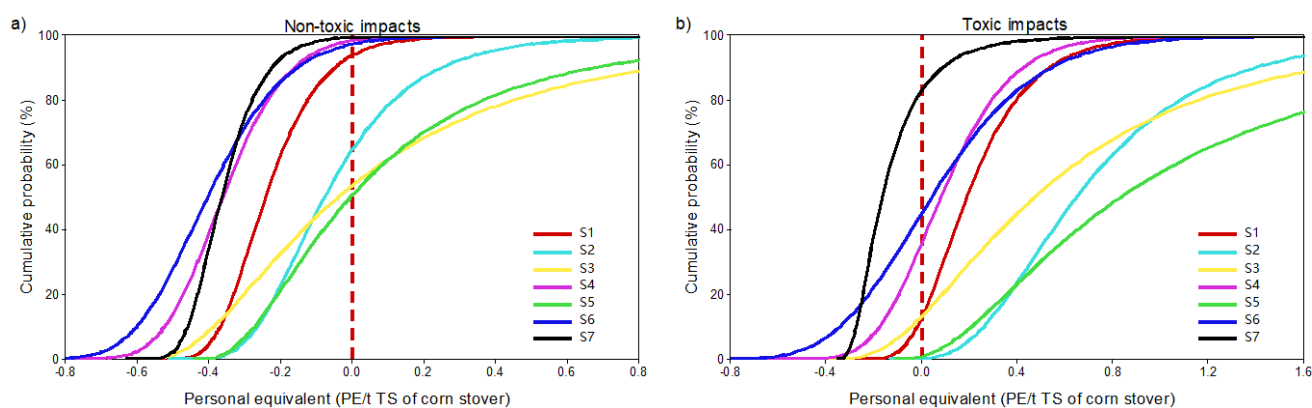
367 The substance contribution to environmental impact potentials is provided in the Supplementary files  
368 of data source. The detailed data on impact potentials of each substance to each impact category were  
369 obtained with mode parameters for the seven technological configurations. In general, the impacts to AD,  
370 POF and EPt were mainly caused by nitrogen oxides. Meanwhile, the impacts to EPm and EPf were due to  
371 phosphate and nitrate, respectively. These substances were associated with energy consumption and  
372 substitution. The impacts to GWP and PM were attributed to CO<sub>2</sub> (fossil) and particulates (<2.5 μm),  
373 respectively, which were also related to energy consumption or substitution. By contrast, the potential  
374 impacts to ET, HTc and HTnc mainly originated from heavy metals to soil and water, including Zn, Cr and  
375 Pb. These substances were mainly from wastewater treatment and energy and material consumption. All  
376 data were highly uncertain and the impact categories have relatively low recommendation levels.

377 Global sensitivity analysis was performed by combining the sensitivity of the results to the parameters  
378 and the uncertainty of each parameter. The results reveal how and which parameters affect the variance of  
379 the results. The distributions of some important parameters including conversion yields and chemical and  
380 enzyme consumptions had been narrowed into much lower variations than in their full distributions because  
381 the parameter distributions of 'best-GWP' were applied. In this instance, some parameters which had not  
382 been optimised according to GWPs now emerge in global sensitivity due to their high variations, including  
383 solid loadings in post wash (SL\_PW), lignin and xylan yields to solid phase in pretreatment (LEF\_PT and  
384 XEF\_PT, respectively). The detailed results are available in the Supplementary Information. Solid loadings  
385 in post wash, pretreatment and hydrolysis become important, indicating that after process optimisation from  
386 global warming perspective, the side streams carrying huge amount of water should receive priority from  
387 the perspective of environmental impacts in further optimisation.

### 389 **3.4 Probability analysis of integrated impact potentials with Monte Carlo simulations**

390 The results within the different impact categories show that the different alternative configurations for  
391 bioethanol production from corn stover have highly different environmental impact potentials. For

392 comparing across the technological alternatives, the normalised personal equivalents of the different impact  
 393 categories are summed up for each round of Monte-Carlo simulation with equal weighting to present the  
 394 integrated impact potentials of each technological configuration. The results are presented separately for  
 395 non-toxic and toxic categories. On the basis of the 5000 Monte-Carlo simulations, the cumulative  
 396 probabilities of the integrated impact potentials were obtained for each configuration, with a lognormal  
 397 distribution in 95% confidence level (Figure 5, the histograms of distribution fitting are available in the  
 398 Supplementary Information).



399  
 400 **Figure 5.** Cumulative probability of integrated impact potentials in a) non-toxic and b) toxic categories  
 401 of the seven technological configurations. (S1-Acid; S2-Alkaline; S3-Solvent-based; S4-Steam explosion;  
 402 S5-Liquid hot water; S6-Ammonia-based; S7-Fungi.)

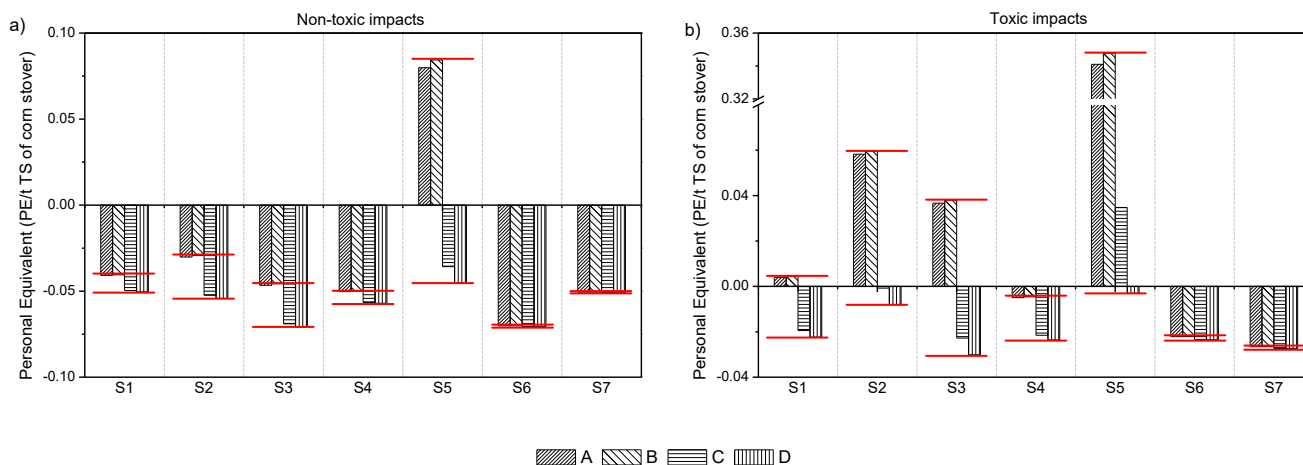
403  
 404 The integrated impact potentials of non-toxic categories were mainly negative for most configurations  
 405 with probabilities of 50%–100%. This result indicates that producing ethanol from corn stover provides  
 406 overall benefits to the environment when the GWP optimised (15% best) technologies are used. Particularly,  
 407 configurations S6 (ammonia-based), S4 (steam explosion) and S7 (fungi) are more likely than the other  
 408 configurations to provide savings in impacts (–0.8 to –0.2 PE/t TS of corn stover) in non-toxic categories.  
 409 This is attributed to more ethanol substitutions and less chemical and energy consumption according to the  
 410 process contribution results (Figure 3). S7, S1 (acid) and S4 have relatively narrow distributions in non-  
 411 toxic impacts because of their low variation in the parameters optimised according to ‘best-GWP’. However,  
 412 this fact also means that further improvement of these technologies will be limited and difficult. In addition,  
 413 S5 (liquid hot water) and S3 (solvent-based) are likely to be less attractive in non-toxic impacts because of  
 414 their very broad distributions.

415 In toxic impact categories, the integrated impact potentials are negative for only S7, S6 and S4, with  
 416 probabilities of 40%–80%. S7 has the highest probability for avoiding toxic impacts because of very limited  
 417 consumption of water, energy and chemicals. S1, S3, S2 and S5 have only 0%–15% probability for  
 418 obtaining savings in the toxic categories. These results may again be affected by the external process  
 419 selection for anaerobic wastewater treatment, as discussed in Section 3.5.

### 3.5 Scenario sensitivity analysis of wastewater treatment process

As suggested by the biorefinery framework proposed by NREL, USA (Tao et al., 2017), we assume that the liquid residues separated from the bioethanol processes is treated by AD with energy recovery from the generated biogas. The details on the AD process and biogas utilisation are described in the Supplementary Information. However, because of lacking accurate data for liquid residue treatment in bioethanol production, a life cycle inventory for the wastewater treatment process was selected from the Ecoinvent database (Ecoinvent, 2016), as described previously. However, this wastewater treatment process appeared to be crucial to the integrated environmental impacts according to the process contribution results discussed previously.

Figure 6 shows the changes of integrated impact potentials in terms of the different options in wastewater treatment process, introduced by using three different options for wastewater treatment (details in the Supplementary Information) and one unrealistic option modelled with no energy consumption or effluent from wastewater treatment. In the technological configuration S5, which uses huge amount of water for pretreatment and postwash, wastewater treatment presented significant sensitivity to the integrated environmental impacts of both non-toxic and toxic categories. By contrast, the results with different processes of wastewater treatment are quite stable for S6 and S7 because of the minimum water consumption during pretreatment with no need for postwash. In general, wastewater treatment processes indicate more significant sensitivity to toxic impacts than to non-toxic impacts.



**Figure 6.** Normalised environmental impact potentials with different wastewater treatment processes in a) non-toxic and b) toxic categories of the seven technological configurations. See text for explanation of wastewater treatment options A: treatment of wastewater, average, capacity  $1 \times 10^9$  L/year; B: market for wastewater, average; C: market for wastewater, unpolluted; D: modelled as no energy consumption or effluent. (S1-Acid; S2-Alkaline; S3-Solvent-based; S4-Steam explosion; S5-Liquid hot water; S6-Ammonia-based; S7-Fungi. Red lines show the maximum and minimum ranges)

448 Shifting the wastewater treatment options from Scenarios A and B to Scenarios C and D can sometimes  
449 change the ranking of the technologies according to their sensitivities, especially for the technological  
450 configuration S3. Therefore, accurate data with little uncertainty regarding the wastewater treatment is  
451 desirable for reducing the uncertainty of the technology comparison. However, the technological  
452 configurations S6, S4 and S7, which were identified in the previous section as promising in avoiding  
453 environmental impacts, still perform satisfactorily with different options of wastewater treatment process.  
454 This result indicates that the technological selection from an integrated environmental perspective basically  
455 is robust in spite of wastewater treatment uncertainty, and is thus valuable for further technological  
456 development.

### 457 458 **3.6 Process optimisation for alternative technologies**

459 According to the probability perspective of the integrated environmental impacts, only some  
460 technologies are of interest for further development. However, technological selection is a comprehensive  
461 task that must pay attention to the yield of bioethanol obtained, the environment impacts, economy, and the  
462 technological feasibility and robustness. The comparison of the technological alternatives for bioethanol  
463 production from corn stover was for the first time made in the current study by quantitatively integrating  
464 the performance with respect to ethanol production, GWP and overall environmental performance from a  
465 statistical perspective. Suggestions for technology optimisation were also provided according to their global  
466 sensitivity to the integrated environmental impacts. The outcome is summarised below, where we also offer  
467 our views on prioritising the technological alternatives for further development:

468 • S1: Technological configuration with acid pretreatment, medium priority. This technology has been  
469 studied extensively, but its ethanol production is low, and its performance in GWP reduction is just fair. It  
470 performs fair with respect to non-toxic impacts and poor with respect to toxic impacts. However,  
471 optimisation of water consumption (solid loading) in all the processes, enzyme and acid consumption, and  
472 xylose recovery in pretreatment has room for improvement.

473 • S2: Technological configuration with alkaline pretreatment, medium priority. This technology has a  
474 high ethanol production and a fair to good performance in GWP reduction. It performs fair with respect to  
475 non-toxic impacts but is not satisfying with respect to toxic impacts. This technology has great room for  
476 improvement, including optimization of enzyme and alkaline consumption as well as water consumption  
477 (solid loading) in all the processes.

478 • S3: Technological configuration with solvent-based pretreatment, low priority. This technology has  
479 a fair ethanol production. However, the GWP reduction performance is poor due to its use of solvent in  
480 spite of the high solvent recovery. The result shows high uncertainty with respect to almost all the  
481 environmental impacts. In addition, the average performance is not satisfying enough. The major concerns

482 in further study include optimisation of water consumption (solid loading) in all processes, solvent  
483 recovery and enzyme consumption.

484 • S4: Technological configuration with steam explosion, high priority. This technology is fairly good  
485 in both ethanol production and GWP reduction. This technology is also good in avoiding non-toxic impacts  
486 and fair with respect to toxic impacts. Technological optimisation should focus on lignin conversion in  
487 pretreatment, enzyme consumption and solid loadings during the process.

488 • S5: Technological configuration with liquid hot water pretreatment, low priority. This technology  
489 performs poorly in both ethanol production and GWP reduction. Particularly, this technology shows high  
490 uncertainty with respect to almost all the environmental impacts, and the average performance is poor. The  
491 water and enzyme consumption during postwash and hydrolysis as well as the overall ethanol production  
492 require significant improvement for further optimisation.

493 • S6: Technological configuration with ammonia-based pretreatment, high priority. This technology  
494 produces a high amount of ethanol and performs well in GWP reduction. This technology is also good at  
495 avoiding non-toxic impacts and fair with respect to the toxic impacts. Optimisation of this technology  
496 should focus on lignin conversion in pretreatment, enzyme consumption and ammonia recycling.

497 • S7: Technological configuration with fungi pretreatment, medium priority. This technology performs  
498 poorly in ethanol production, and its performance in GWP reduction is fair. However, after optimization,  
499 this technology can be good at avoiding integrated non-toxic and toxic impacts, but the room for  
500 improvement is considered limited. Further optimisation should focus on enzyme consumption and solid  
501 loadings in hydrolysis.

502 In general, technological configurations S4 and S6 seem to have potential for further technological  
503 development and application according to the available data from laboratory experiments reported in  
504 scientific literature. S1, S2 and S7 may have potential but further research on the abovementioned key  
505 issues should be undertaken. However, S3 and S5 do not seem attractive from an environmental perspective,  
506 including GWP and integrated non-toxic and toxic categories. These two technologies require substantial  
507 improvements at the laboratory scale before they can be considered for technological development and  
508 practical application.

## 509 **4. Conclusions**

511 The potential environmental impacts, namely, climate change (global warming potential),  
512 stratospheric ozone depletion, acidification, eutrophication (terrestrial), eutrophication (freshwater),  
513 eutrophication (marine), photochemical ozone formation (human health), ecotoxicity (freshwater), human  
514 toxicity (cancer effects), and human toxicity (non-cancer effects), of producing bioethanol from corn stover  
515 were systematically assessed by LCA for selected relevant experimental datasets reported in literature. The  
516 15% best datasets with respect to savings in GWP for seven different technological configurations were

517 extracted from recently reviewed 474 publications on converting corn stover to bioethanol. These datasets  
518 were considered relevant for developing technologies of bioethanol production from corn stover because  
519 of the overall purpose to reduce the greenhouse gas footprint of fuel consumption by amending gasoline  
520 with bioethanol. The assessed impacts were complex and included both loads to the environment (positive  
521 numbers) and savings to the environment (negative values), generally from  $-0.1$  PE to  $0.1$  PE per t of dry  
522 corn stover for most non-toxic impact categories. In general, the technologies with pretreatment using  
523 solvent (S3) or liquid hot water (S5) had the highest environmental loads and the highest uncertainty due  
524 to the high solvent consumption and water and energy consumption, respectively. The toxic impacts with  
525  $-0.2$  PE to  $0.5$  PE per t of dry corn stover were in general higher than the non-toxic impacts, but this result  
526 was consistent across technologies. The fossil fuel substitution with bioethanol provided environmental  
527 savings, and so did the energy recovered from the liquid and the solid residues. Meanwhile, the production  
528 of the enzymes used for most technologies was a significant load to the environment. Each process  
529 throughout the whole technology shows different sensitivities to different environmental impact categories.  
530 The effluent from the anaerobic digestion treating the liquid residue was not well characterised in the  
531 datasets available and should receive considerable attention in future development work. The reason is that  
532 our analysis indicates that the effluent treatment and discharge in nearly all the technologies could be a  
533 significant load to the environment and particularly important to toxic impacts. Based on cumulative  
534 probabilities of the overall environmental performance together with the amount of bioethanol produced,  
535 we consider the technologies with pretreatment by using steam explosion (S4) or ammonia (S6) as the  
536 highest priority for further development with approximately 100% and 40% probabilities to have savings  
537 in non-toxic and toxic impacts, respectively. By contrast, the technologies with pretreatment by using  
538 solvents (S3) or liquid hot water (S5) have the lowest priorities with only 50% and less than 10%  
539 probabilities to have savings in non-toxic and toxic impacts, respectively. The technologies with  
540 pretreatment by using acids (S1), alkali (S2) and fungi (S7) are of medium priority because they need  
541 significant improvements either in their bioethanol yield or environmental performance. From this point of  
542 view, the technologies with steam explosion and ammonia pretreatment should receive priority in  
543 development of technology for producing bioethanol from corn stover. Lignin conversion in pretreatment,  
544 enzyme consumption, solid loadings during the process and chemical recycling are suggested to be  
545 carefully optimized during industrial application from a sustainability perspective. In addition, we suggest  
546 that assessment of the potential environmental impacts to be an integrated part of the further research and  
547 development for producing bioethanol from corn stover in order to ensure the development of sustainable  
548 technologies.

## 550 **Conflicts of interest**

551 The authors declare no conflicts of interest.

552

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558

## 559 **Electronic Supplementary Information**

560 Electronic Supplementary Information associated with this article can be found in the online version:  
561 system boundary; parameter distribution of the top 15% ‘best-GWP’ cases; histograms of Monte-Carlo  
562 simulation; selected external processes of wastewater treatment; process contribution to environmental  
563 impact potentials; sensitivity coefficients of parameters; global sensitivity analysis of parameters;  
564 uncertainty contribution of parameters; original sources of the extracted datasets; data of process  
565 contributions to each impact category; data of impacts in terms of substance with mode parameters.

566

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