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To cite this article: S J H M Yusof *et al* 2018 *IOP Conf. Ser.: Mater. Sci. Eng.* **368** 012004

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Environmental performance of bioethanol production from oil palm frond petiole sugars in an integrated palm biomass biorefinery.

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Abstract. Generation of biofuels from renewable resources such as lignocellulosic biomass is a promising approach to reduce the sole reliability on the depleting fossil fuel. The aim of this work is to assess the potential environmental impact of bioethanol production from oil palm frond in a conceptual oil palm based biorefinery model, utilizing wet disc milling as a pretreatment method. A cradle-to-gate approach was selected, beginning with the harvesting and transportation of the frond petiole from the plantation, followed by production of oil palm frond petiole sugars via pretreatment and saccharification prior to bioethanol fermentation, and finally purification of the fermentation products to obtain anhydrous bioethanol. A life cycle assessment was performed using CML 2 baseline 2000 method (SimaPro v8.0), where ten impact categories were evaluated. It was found that the most significant environmental impact was from sugar recovery process with contribution of more than 90 %. This is mainly due to high power consumption by wet disc milling during pretreatment. Apart from that, production of enzyme and chemicals which were used during saccharification consumes high energy thus contributing major problems to the surrounding. Finding of this study helps to identify the hotspot which can be improved to establish a more energy efficient and greener system for bioethanol production from oil palm frond petiole sugars.



1. Introduction

Oil palm frond (OPF) is among the major biomass produced in the oil palm industry with generation of 54.17 and 54.24 million tonnes in 2010 and 2011, respectively [1]. It is obtained during replanting, pruning and harvesting, hence is available throughout the year. In the common practice, pruned frond is left in the plantation for nutrient recycling and soil conservation [2-3]. However, it was discovered that the basal (lower) part of OPF is rich in cellulosic materials and sugars, which are needed in the production of the biofuels and biobased chemicals [4-5]. Therefore, it is suggested that this part of OPF to be collected and serve as a raw material for biobased products whereas the remaining two third that contains high nutrients still serve as fertilizers in the plantation. With high carbohydrates and nutritive contents, OPF petiole can be converted into value added products such as biofuels, biobased chemicals, biofertilizer and animal feed [6-9]. Moreover, OPF juice was found to have high amount of free sugars with 70 % of glucose, and can be easily obtained by simple pressing method [10].

First generation bioethanol is generated using sugars and starch from feedstocks such as sugarcane and corn [11-14]. However, bioethanol production from lignocellulosic materials including crop residues, forestry and municipal waste is getting much attention these days to overcome the limitation in first generation biofuel production [15-17]. Apart from eliminating competition with food source, most of these wastes are available in large quantity and cheap. Examples of potential crop residues are wheat straw, sweet sorghum, cane bagasse, rice straw and corn stover [12,18-22]. However, the conversion of carbohydrate polymers to biofuel is a challenge due to the complex structure of lignocellulose. Pretreatments to alter its original structure is necessary prior to saccharification and fermentation [6,11-12,22].

Previously, feasibility study on bioethanol production from OPF petiole sugars was conducted within an integrated palm biomass biorefinery [6]. It was demonstrated that integration of a biorefinery to an existing palm oil mill was possible and has high potential for scaling up. Nevertheless, the environmental aspect should also be considered in promoting a sustainable biofuel production process [12]. Life cycle assessment (LCA) is a common tool to evaluate the impacts of a product system to the surrounding through its life cycle and have been extensively used to measure the environmental performance of bioethanol production [11,13,16,23-25]. Therefore, a LCA for bioethanol production from OPF petiole sugars was conducted in this study to evaluate the potential harm of the process to the environment.

2. Material and methods

2.1 Process description

2.1.1 OPF collection and transportation. Model used in this study was as reported by Abdullah et al.[6]. Bioethanol was produced from OPF petiole sugars collected from 4 neighboring oil palm mills. Approximately 57 600 tonnes of OPF petioles can be obtained from 240 000 tonnes of fresh fruit bunches (FFB) harvested every year. The harvested OPF petioles were transported to the mills which located approximately 15 km from the plantation. It was proposed that an additional cart for carrying OPF petioles to be attached to the existing truck carrying the harvested FFB [6]. The sugar production was primarily conducted at the individual mills using the surplus energy from the existing cogeneration system. Then, concentrated OPF petiole sugars from every mill was combined at a biorefinery which is located at one of the four mills. Each mill was assumed to be located 80 km radius from each other [6].

2.1.2 OPF sugar production. Cleaned petiole were milled to obtain the OPF juice which contain free sugars. It was reported that OPF juice comprise of 54 g/L, 21 g/L and 2 g/L of glucose, sucrose and fructose respectively [10]. Approximately 50 % (w/w) of OPF juice can be obtained, leaving another 50 % of OPF pressed fiber. The OPF pressed fiber was then subjected to wet disc milling (WDM) pretreatment at conditions described by [26]. 80 % of holocellulose was recovered after pretreatment, where 95 % of it was converted to glucose and xylose following saccharification.

Saccharified sugars and the OPF juice were subsequently concentrated using evaporators to remove 50 % of their water content before transmitted to the selected mill which locates the bioethanol refinery. A total of 13 900 tonnes/year/mill of sugars can be generated.

2.1.3 Bioethanol production. The concentrated sugars were sterilized prior to inoculation of *S. cerevisiae* culture at 10 % (v/v). Urea was added to the fermentation media as nitrogen source. The fermentation was conducted in a reactor at 30 °C for 24 hours. Separation of solids and supernatant in the fermentation broth was subsequently performed by centrifugation. The supernatant which contains bioethanol was further purified using a continuous distillation, rectification, stripping and molecular sieves drying to obtain 98.9 % of anhydrous bioethanol. Whereas the solid residue was subjected to the waste treatment system for production of dried distiller grains with soluble (DGGs).

2.2 Life cycle assessment

LCA is a method to assess the environmental impacts of a product through its life cycle from the raw material acquisition and production, to end-use and disposal, with a generic framework provided by ISO 14040 and 14044 [16,27]. According to ISO 14040 (1997), LCA consists of 4 different phases, starting with goal and scope definition, inventory analysis, life cycle impact assessment and finally interpretation. Each step was conducted based on ISO standards as guidelines to ensure precise decision can be concluded. In this study, the characterization model CML 2 baseline 2000 v2.05 incorporated in software SimaPro v8.0 (Pre Consultants 2014) was adopted, based on the previous LCA study on bioethanol production which is almost similar to the present case [27-28].

2.2.1 Goal and scope of study. The aim of this study is to quantify the environmental impact of bioconversion of OPF petiole sugars for bioethanol production. The impacts were assessed according to all processes involved, starting from the harvesting of the OPF petiole at the plantation, transportation of petiole to the mill where it will undergo pretreatment and saccharification processes for sugar production, followed by the sugar conversion to bioethanol via fermentation and finally purification of bioethanol to obtain anhydrous bioethanol (cradle-to-gate). The evaluated impact was based on functional unit of 1 tonne of anhydrous bioethanol. Figure 1 shows the life cycle assessment system boundaries for the case studied.

2.2.2 Inventory analysis. Data for harvesting and transportation of OPF petiole were obtained from Pusat Penyelidikan Tun Razak, Pahang. While data on material input and output for bioethanol production were attained from the earlier study [6] and also calculated based on findings from the previous works [6, 9, 26, 29]. Most of emission data were generated from simulation by Superpro Designer software v9.5. Since the life cycle data for cellulase production is not available, the material and energy demands were estimated from the inventory data generated using values from previous study utilizing similar enzyme, assuming comparable enzyme production steps were adopted [30-31]. Inventory for urea, sodium and citric acid were obtained from the EcoInvent 3.1 database. Table 1 listed data use for the Life Cycle Impact Assessment.

2.2.3 Assumptions. In order to facilitate the evaluation of the impact arising from the proposed model, few assumptions were implemented. Steam required to run the bioethanol biorefinery is solely provided by the oil palm mill, through cogeneration system. Approximately 140 053 tonne/ year of steam was needed and this requirement can be fulfilled by the mill using the existing excess steam left following FFB processing [6]. Hence, impact of steam was excluded. Whereas the electricity is obtained from both the cogeneration and the national grid. This is because only 7.72 GWh of electricity can be produced by the cogeneration system based on boiler and steam efficiency of 77.4% [6]. 4.08 GWh of electricity per tonnes FFB was required for FFB processing, leaving an excess energy of 3.64 GWh of electricity. On the other hand, total electricity required for bioethanol production using WDM pretreatment method was 1 076 GWh, beyond the surplus electricity available. Therefore, an extra electricity supply of 1 072 GWh must be obtained from the external source and the

value was considered in the inventory.

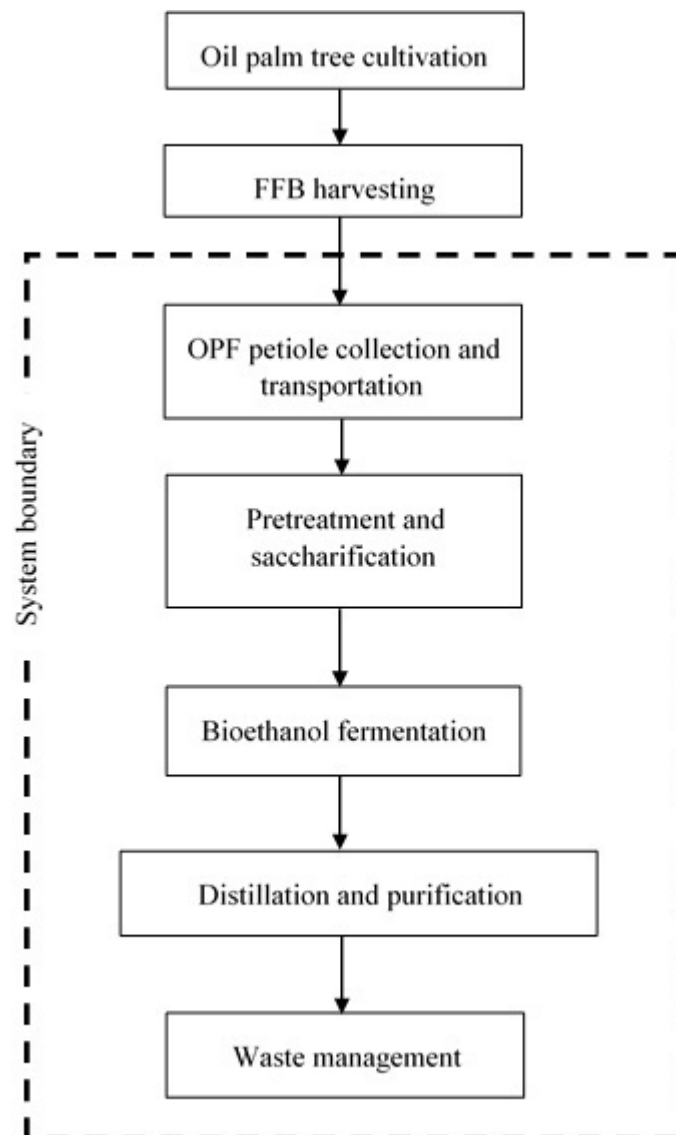


Figure 1 Life Cycle Assessment system boundaries for the bioethanol production from OPF petiole sugars.

Table 1 Inventory data for bioethanol production from OPF petiole sugars.

	Values	Reference
Transportation		
<i>Input</i>		
Trucks, 6 tonnes capacity, tkm/year	7 199 985	This study
Trucks, 20 tonnes capacity, tkm/year	3 324 480	This study
Bioethanol production		
<i>Input</i>		
OPF petiole, tonnes/year/mill	57 600	[6]
OPF pressed fiber, tonnes/year/mill	20 160	[6]
OPF petiole sugars, tonnes/year/mill	13 900	[6]

Citric acid, tonnes/year	2327	This study
Sodium citrate, tonnes/year	4688	This study
Enzyme, tonnes/year	2685	This study
Urea, tonnes/year	1286	This study
Air, tonnes/year	1 121 681	This study
Water, tonnes/year	2 707 952	This study
Electricity, MWh/year	1 072 110	This study

Output

Anhydrous bioethanol, tonnes/year	26 700	This study
Stillage, tonnes/year	521 800	This study
DGGS, tonnes/ year	2611	[6]
Saccharified fiber residue, tonnes/year	33 600	[6]
Energy from OPF residue, MJ/year	1767	[6]

Emission

CO ₂ , tonnes/year	27 138	This study
N ₂ , tonnes/year	861 012	This study
O ₂ , tonnes/year	261 387	This study
Water vapor, tonnes/year	34 560	This study
Ethanol, tonnes/year	82	This study
Waste water, tonnes/year	270 000	[6]
Water for recycle, tonnes/year	57 600	[6]

Enzyme production**Input**

Cellulosic hydrolysate, kg	7600	[30]
Water, kg	4474	[30]
Maize steep water, kg	119	[30]
Diammonium phosphate (nutrient), kg	47	[30]
Electricity, kWh	2236	[30]

Output

Enzyme broth, kg	11 455	[30]
CO ₂ , kg	879	[30]
Furfural, kg	8.2	[30]
Acetic acid (VOC), kg	2.8	[30]

2.2.4 *Characterization model and impact categories.* CML 2 baseline 2000 model (SimaPro v8.0) was applied to perform the Life Cycle Impact Assessment. Ten mid-point impacts was evaluated: abiotic depletion potential (ADP), acidification potential (AP), eutrophication potential (EP), global warming potential (GWP100), ozone layer depletion potential (ODP), human toxicity potential (HTP), fresh water ecotoxicity potential (FETP), marine ecotoxicity potential (METP), terrestrial ecotoxicity potential (TETP), and photochemical oxidation potential (PCOP).

3. Results and discussion

Physical or mechanical pretreatment such as ball milling and WDM aims to increase the surface area by reducing the biomass particle size, together with defibrillation and reduction in crystallinity degree [32]. This provides higher accessibility of enzymes to substrate, thus escalating the saccharification rates and yield. Previous study showed that with WDM application, 95 % of holocellulose was successfully converted into simple sugars following saccharification [26]. Nevertheless, despite of its efficiency in enzymatic hydrolysis, WDM has high energy consumption and application in a larger

scale was somehow difficult [29,33-34]. It was reported that approximate energy required were 48 MJ/kg for sugarcane bagasse and 39.6 MJ/kg of sugarcane straw for 20 cycles [29]. Whereas for OPMF, 18.9 MJ/kg was consumed for 9 cycles [35].

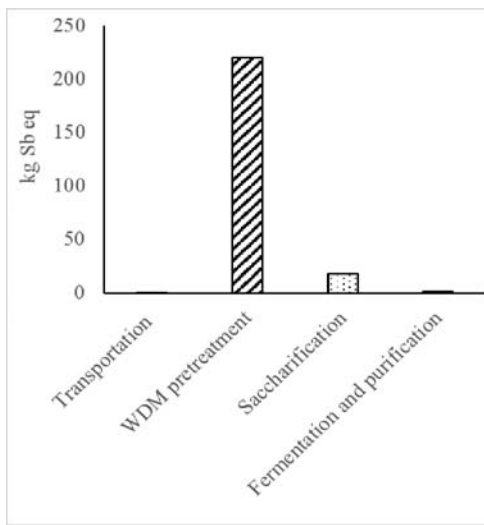
Figure 2 depicted impact values for each category according to major process involved. The process was classified into 4 sub-processes: transportation, WDM pretreatment, saccharification and ethanol fermentation and purification. By comparing contribution of each sub-process to the environmental burden, the process which needs to be improved can be identified.

In general, life cycle impact assessment demonstrated that pretreatment step by wet disc milling had a major impact to all categories, except for EP (Figure 2d), ODP (Figure 2e) and TETP (Figure 2j) where saccharification impact dominates. WDM consumed high electricity thus requires high usage of fossil fuel. This explains its significant impact in ADP (92%). ADP (Figure 2a) which refers to the decrease of non-living natural resources (including energy resources) such as minerals or crude oil, is a very widely used impact category reflecting fossil fuel energy use [27]. Similarly, application of enzyme and chemicals (citric acid and urea) during saccharification and fermentation, involved high utilization of fossil fuel during their production and diesel consumption for transportation.

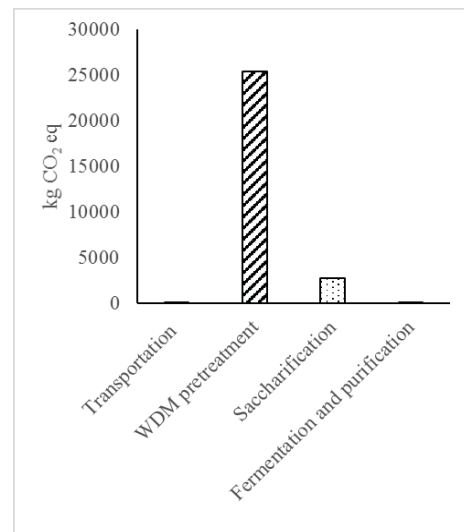
GWP (Figure 2b) is a quantification of an equivalent amount of carbon dioxide released to the surroundings. Due to high electricity consumption, wet disc milling contributed the highest impact to global warming potential (90%) as atmospheric emissions are directly related to energy and resource consumption [36]. Whereas saccharification has the second highest impact with almost 10%, due to enzyme application. Previously, enzyme production was found to be the main hotspot for carbon dioxide emission [27, 36]. This is because enzyme production consumes a substantial amount of fossil or combustion electricity for air compression which also generates a considerable amount of carbon dioxide emission in the chain [16]. Utilization of fossil fuel energy by all transport and during production of urea used in fermentation resulted in carbon dioxide emission. Additional contribution comes from carbon dioxide produced during fermentation.

AP refers to environmental damage cause by acidic gas such as sulfur dioxide (SO₂). Since fossil fuel consumption resulted in SO₂ emissions [27], thus WDM produced a significant impact in AP (Figure 2c). Similarly, high usage of fossil fuel explained the reason for saccharification, fermentation and transportation contribution in this impact category.

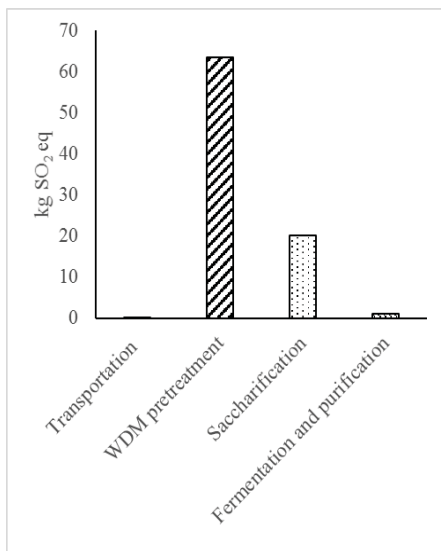
For EP category, it was demonstrated that the impact from saccharification and pretreatment steps were almost equal, which was approximately 50 % (Figure 2d). Eutrophication potential is associated with environmental impacts of excessively high nutrients (N and P) in the river and lakes. Consequences are like shifting in species composition, increased biological productivity such as algal bloom and subsequent oxygen consuming degradation processes [19, 27]. Significant burden from saccharification step was due to application of diammonium phosphate which serves as a nitrogen source in enzyme production, contributing to nutrients emission. High eutrophication effect was



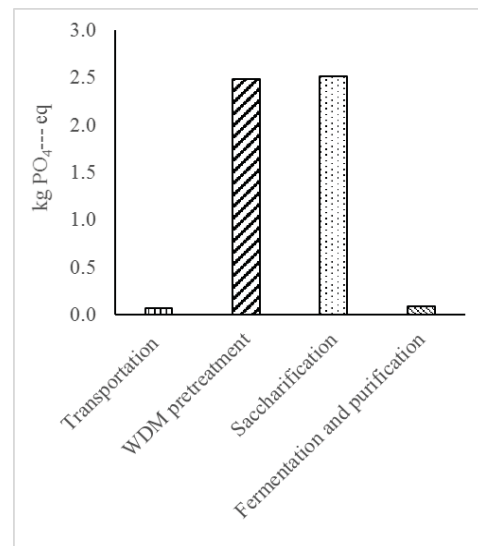
(a)



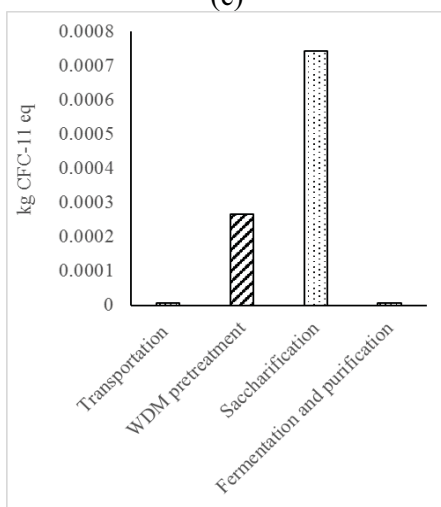
(b)



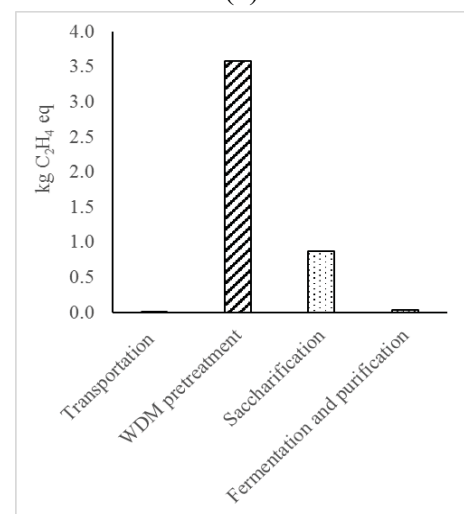
(c)



(d)



(e)



(f)

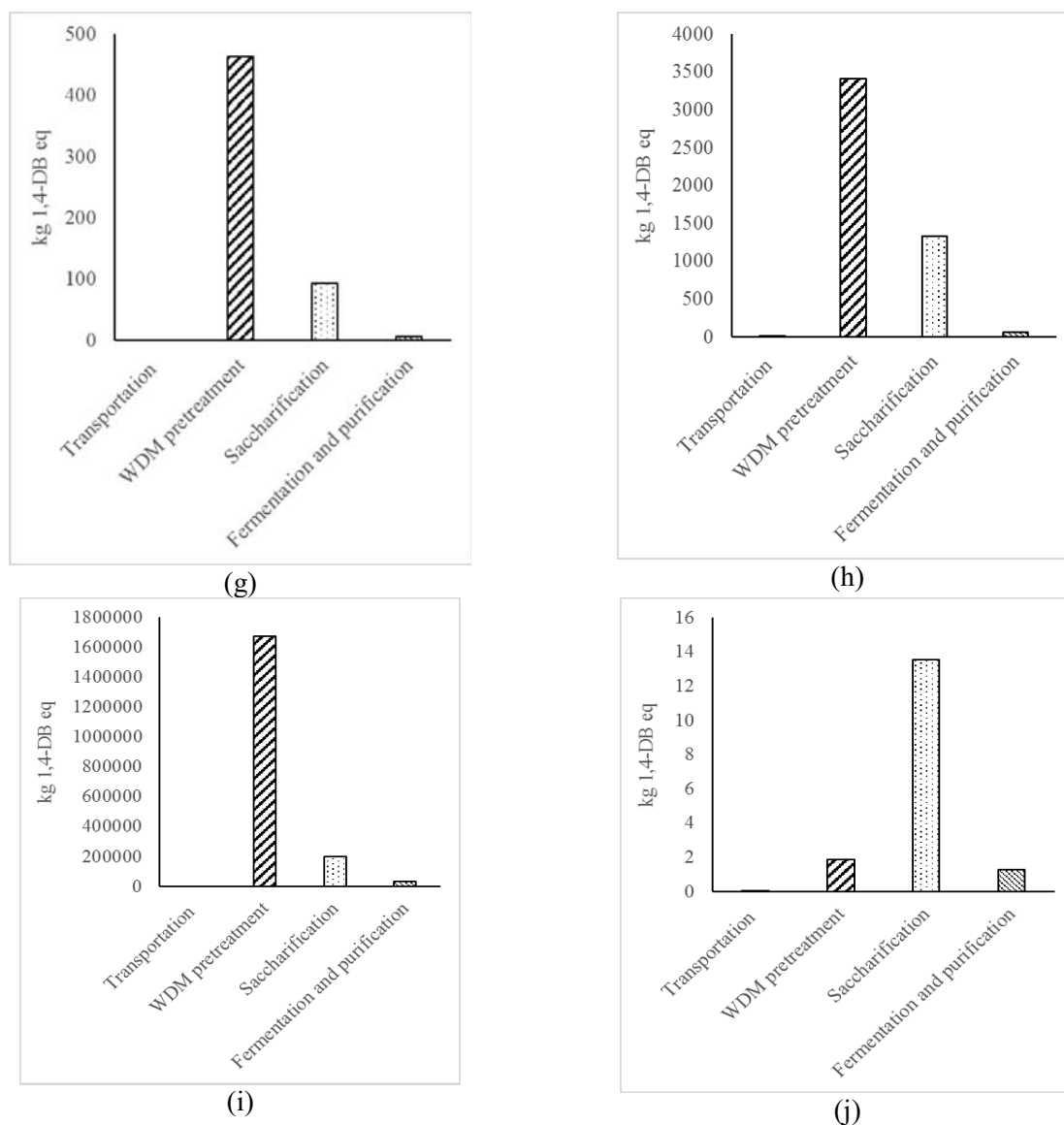


Figure 2 Characteristic values of impact categories for 1 tonnes of bioethanol production from OPF petiole sugars. (a) ADP, (b) GWP, (c) AP, (d) EP, (e) ODP, (f) POCP, (g) FAETP, (h) HTP, (i) MAETP and (j) TETP.

observed due to NH_3 and NO_x emissions from nitrogen based fertilizer production and application and diesel use in agricultural machinery and tractors [16]. Whereas effect from WDM was due to fossil fuel consumption which contributed to atmospheric deposition. Nitrogen in the form of NO_x , was released and later deposited in the water source as wet or dry deposition [37-38].

ODP is referring to the decrease in the total volume of ozone in the Earth's stratosphere and is caused by various chlorinated and bromated substances [27, 39]. It was found that saccharification was the major contributor, followed by WDM and transportation at 73 % and 26 %, each (Figure 2e). Earlier study reported that production of carbon source and electricity use during cellulase production contributed to a significant impact in ODP [40]. Halons which are used as fire suppressant and coolant in the gas pipeline distribution system was emitted during electricity generation, increasing the risk of ozone depletion [41].

Whereas for POCP, WDM has the highest impact with 80% and saccharification was the second highest with 19 % (Figure 2f). The photochemical oxidation, also referred as summer smog, is the result of reactions between NO_x and hydrocarbons or volatile organic compounds (VOC) [27]. Intensity of impact increased with increment of amount of emissions from fossil fuel consumption. Previous study showed that bioethanol conversion unit produced the highest POCP, with SO₂ as major pollutant, generating from coal combustion [19, 39].

HTP, FAETP, MAETP and TETP characterization compares a large number of chemicals that can contribute to cancer or have other negative effects on human health, fresh aquatic ecosystem health, marine aquatic ecosystem health and terrestrial ecosystem health, respectively [19]. Coal which is commonly use as fossil energy for electricity and steam production contains high composition of heavy metals such as As, Hg, Pb and Cr, thus contributes to high impact in ecotoxicity potential. This clarifies the significant impact from high energy requirement process such as WDM and saccharification (Figure 2g-2j).

Based on these findings, several approaches can be suggested to reduce the environmental impact. Other pretreatment methods with higher or comparable performance could be used to replace WDM. Hydrothermal pretreatment, including liquid hot water, hot compressed water and superheated steam are among potential methods as it is more environmental friendly since it use only water and heat (chemical free) [42]. With process optimization it was demonstrated that application of hot compressed water of oil palm frond prior to enzymatic hydrolysis can generate glucose yield of 93% [43]. Furthermore, few studies on LCA of pretreatment methods in biofuel production have shown that liquid hot water pretreatment produced lower carbon dioxide or GHG emission [27,39,44]. Apart from that, with combination of WDM and hot compressed water, the amount of energy consumed was reduced up to 21 % [35]. Nonetheless, there are few drawbacks that also needs to be considered. Hydrothermal pretreatment consumed substantial amount of water which resulted in more waste water that have to be treated [44]. Moreover, the process also generated significant amount of inhibitors which needs to be removed as they could interrupt the saccharification and fermentation process [45]. Therefore, further investigation should be conducted prior to selection of the most appropriate method.

Modifications related to enzyme production and hydrolysis can helps to improve the environmental performance cause by enzyme application. Among alternatives that can be considered are to decrease the enzyme quantity, improving the enzyme production process, applying enzyme recycling and utilizing recombinant yeast which produce hydrolytic enzymes, thus eliminating the requirement of enzyme production step [46]. Other than that, the environmental performance can also be improved by using the waste stream such as COD in the stillage for energy recovery [47-48].

4. Conclusions

A cradle-to-gate LCA on the conversion of OPF petiole sugars to bioethanol reveals that pretreatment and saccharification steps were the hotspots which needs to be improved. Despite of its high efficiency in sugar production, application of WDM is somehow environmentally unfavorable. Selection of proper pretreatment method, as well as improving enzyme and chemicals utilization, are therefore crucial to reduce burden to the environment. This study could serve as a base case for comparison with other LCA work on bioethanol production from OPF petiole sugars utilizing other pretreatment technique in the future.

Acknowledgements

The authors would like to thank Science and Technology Research Partnership for Sustainable Development (SATREPS) under Japan Science and Technology Agency (JST) and Japan International Cooperation Agency (JICA) for partly funding this work. The authors also thank Universiti Putra Malaysia for technical assistance and Universiti Kuala Lumpur for the permission to use the SimaPro v8.0 (PreConsultant 2014) during this study. Appreciation also goes to Universiti Malaysia Perlis and Ministry of Higher Education, Malaysia for the provision of study leave and scholarship for the first author.

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