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Life cycle analysis of fermentative production of succinic acid from bread waste

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ABSTRACT

According to the US Department of Energy, succinic acid (SA) is a top platform chemical that can be produced from biomass. Bread waste, which has high starch content, is the second most wasted food in the UK and can serve as a potential low cost feedstock for the production of SA. This work evaluates the environmental performance of a proposed biorefinery concept for SA production by fermentation of waste bread using a cradle-to-factory gate life cycle assessment approach. The performance was assessed in terms of greenhouse gas (GHG) emissions and non-renewable energy use (NREU). Waste bread fermentation demonstrated a better environmental profile compared to the fossil-based system, however, GHG emissions were about 50% higher as compared to processes using other biomass feedstocks such as corn wet mill or sorghum grains. NREU for fermentative SA production using waste bread was significantly lower (~ 46%) than fossil-based system and about the same as that of established biomass-based processes, thus proving the great potential of waste bread as a valuable feedstock for bioproduction of useful chemicals. The results show that steam and heating oil used in the process were the biggest contributors to the NREU and GHG emissions. Sensitivity analyses highlighted the importance of the solid biomass waste generated in the process which can potentially be used as fish feed. The LCA analysis can be used for targeted optimization of SA production from bread waste, thereby enabling the utilization of an otherwise waste stream and leading to the establishment of a circular economy.

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1. Introduction

The non-renewable nature of fossil fuels and their negative impact on the environment has necessitated the search for alternative, sustainable, and environment friendly routes, based on the use of renewable feedstock. Depletion of petroleum products along with the emission of greenhouse gases has been exacerbated by the exponentially growing human population, which has led to an increasing amount of food production, and consequently, the generation of an enormous amount of food waste (Haroon et al., 2016; Corrado and Sala, 2018). About one-third of the food produced in the world (~ 1.3 billion tonnes, costing around $750 billion) is wasted or lost through the food chain every year (Gustavsson, 2011; Paritosh et al., 2017; Pham et al., 2015). In the UK alone, an estimated 10 million tonnes of food and drink are wasted annually after the farm gate, worth around £20 billion, and the carbon footprint from these wastes is estimated to be equivalent to one-fifth of the UK's total emissions (WRAP, 2020; HM-Government, 2018). In addition to being a significant loss of valuable materials, these enormous quantities of waste result in serious management problems, both economically and environmentally. Presently, most of this highly perishable food waste forms a significant part of the municipal solid waste, which leads to bad odour, creates air pollution, contaminates ground water, and serves as a breeding ground for pathogenic microbes (FAO, 2012; Kumar and Longhurst, 2018). This problem of food waste has been intensified due to the slow progress made in the development of efficient technologies for waste treatment and disposal.

The microbial conversion of renewable biomass into fuels and chemicals is a green and clean approach (Haroon et al., 2016; de Jonge et al., 2020). The commonly used methods for dealing with food wastes are composting, anaerobic digestion or landfills. Food waste is attractive in terms of its nutrient content, i.e. 30–60% starch/sugars, 5–10% proteins and 10–40% lipids (Kumar and

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Longhurst, 2018) and thus the high quantity of food waste can serve as the potential feedstock for global bioproduction of large quantities of chemicals with high market value. A more profitable way of channelizing food waste could be the efficient transformation of this renewable organic carbon source to a spectrum of industrially important chemicals and fuels via a greener route (Tuck et al., 2012). With 44% of all bread being thrown away, bread is the second most wasted food in the UK after potatoes (WRAP, 2020; Ventour, 2008). Every day about 20 million whole slices of bread are binned by the UK households which equates to annual bread wastage of 328,000 tons (WRAP, 2020; Ventour, 2008). Similar to other food waste streams, bread waste consists of complex carbohydrates (50–70%), proteins (8–10%), lipids (1–5%) and traces of phosphorus (Adessi et al., 2018; Haroon et al., 2016; Leung et al., 2012). The carbohydrate in the bread is mostly in the form of starch, which can be easily hydrolyzed to fermentable sugars in comparison to other bulk, crude, or renewable sources such as recalcitrant lignocellulose wastes where extraction of sugars is cumbersome. This composition makes bread waste a high potential feedstock for the fermentative production of chemicals (Melikoglu and Webb, 2013).

Succinic acid (SA) ($C_4H_6O_4$), is a dicarboxylic acid, which has been identified as one of the twelve platform chemicals produced from renewable resources by the U.S Department of Energy (US DOE) (Werpy and Petersen, 2004). It showcases a wide range of applications in industries such as pharmaceuticals, food, polymers, plasticizers, and green solvents (Nghiem et al., 2017). The presence of two carboxyl groups makes SA a precursor molecule for the synthesis of a variety of chemical compounds (Prabhu et al., 2020). The commercial market of SA is expanding with a demand of 50,000 metric tons in 2016, which is anticipated to double by 2025 (Chinthapalli et al., 2018). The fermentative production of SA has several advantages over the chemical route such as mild operating conditions, environmentally friendly approach, reduced greenhouse gas (GHG) emission, biodegradable biocatalysts, substrates, intermediates, and by-products. Currently, bio-based SA constitutes a significant fraction of the total market (Cheng et al., 2012; Stylianou et al., 2020).

The higher cost of SA production through the biological route reduces the economic viability of the process, which limits the commercial scale production. Like any other bio-derived product, one of the factors strongly influencing the process economics of bio-based SA production is the cost of feedstock (Stylianou et al., 2020). The process economics can be improved using some waste or low-cost feedstocks (Pateraki et al., 2016; Stylianou et al., 2020).

Leung et al. (2012) investigated bread waste, and Zhang et al. (2013) studied cake and pastry waste as feedstock for fermentative SA production by *Actinobacillus succinogenes*. Fig. 1 shows the biochemical pathway for SA production from glucose as a carbon source by *A. succinogenes*. The SA titer obtained with bread, cake and pastry wastes were 47.3, 24.8 and 31.7 g/L with a yield of 1.16, 0.80 and 0.67 g/g sugar, respectively. The yield of SA per gram of bread, cake and pastry wastes were 0.55, 0.28 and 0.35 g, respectively. The SA productivity achieved on bread, cake and pastry wastes were 1.12, 0.79 and 0.87 g/L.h, respectively. These studies conducted by our research groups successfully demonstrated the use of bakery wastes for biological SA production at laboratory scale. The results obtained are comparable to even SA accumulated on pure glucose by *A. succinogenes* in terms of titer and productivity with much better yield. For example, Liu et al. (2008) achieved SA titer, yield and productivity of 60.2 g/L, 0.75 g/g and 1.3 g/L.h. Similarly, Bradfield and Nicol (2014) amassed 48.5 g/L SA with a conversion yield of 0.91 g/g on glucose. Following these experimental demonstrations, Lam et al. (2014) conducted a detailed techno-economic analysis to estimate process economics on commercial scale fermentative production of SA using bread waste. The process was found to be economically feasible with a return on investment of 12.8% and a payback period of 7.2 years.

Although the economic feasibility of the process has been demonstrated, it is critical to evaluate the environmental impact of the process to determine the process sustainability. Life cycle assessment (LCA) is a common and well-established methodology used to determine the environmental impact of the processes. Some researchers have investigated the sustainability of SA production using renewable feedstock such as dextrose (corn), sorghum grains (a non-food crop), apple pomace, mixed food waste, etc. (Cok et al., 2014; Ögmundarson, 2018; Moussa et al., 2016).

Cok et al. (2014) performed a cradle-to-gate LCA study for biological SA production through three different processes; the first one was based on yeast fermentation at low pH, the second one on anaerobic fermentation to succinate salt at neutral pH, and third was an analogous process that led to co-production of ammonium sulfate in the downstream processing. They also compared the performance of these bio-processes with the production of maleic anhydride, SA, and adipic acid through the petrochemical route. The results of the impact assessment, which was characterized using non-renewable energy use (NREU) and GHG emissions, showed that low pH yeast fermentation with direct crystallization had the lowest environmental impact when compared to other bio-processes or the petrochemical routes for SA production.

Smidt et al. (2015) performed a cradle-to-gate LCA analysis for fermentative SA production in an European plant using corn wet mill as the starting material at low pH with direct crystallization for product recovery. It was shown that this particular bio-based process had lower Global Warming and Resource Depletion impact, however, it showed a higher impact in Land Use and Dust & Particulate Matter categories, as compared to the fossil-based SA production. Smidt et al. (2015) also showed that employing Brazilian sugar cane feedstock in place of corn could result in an overall lower impact due to its efficient refining process and lower land usage.

Moussa et al. (2016) used real production data from a Myriant corporation facility in the USA and assessed the environmental performance of bio-based SA production process (based on a non-food crop, Sorghum grains) using a cradle-to-gate LCA. This analysis showed that the impact parameters like GHG emissions and non-renewable fossil cumulative energy demand were lower than the petrochemical alternative as well as the bio-based process which employs dextrose as the feedstock.

González-García et al. (2018) used a cradle-to-gate LCA and determined the environmental performance of a fermentative SA production process based on apple pomace as feedstock. Extraction and distillation operations were identified as the major environmental hotspots. Global warming potential (GWP) of the proposed process per kg SA produced, was found to be significantly higher than other bio-based processes (like those using corn, or sorghum grains or sugar cane as feedstock) as well as that of fossil-based process.

Brunklaus et al. (2018) conducted a cradle-to-gate LCA study comparing the environmental performance of two different valorisation options for mixed food waste, one leading to the production of biogas and the other for the synthesis of SA. Here, waste bread was used as a proxy for mixed food waste, based on the results of Lam et al. (2014). However, instead of *A. succinogenes* (that were used in the original study by Lam et al. (2014)), fermentation was assumed to be carried out using *E. coli*, with the yield estimated from a lab-scale study for mixed food waste (Sun et al., 2014), and whey fermentation used as a model for *E. coli* process (Jungbluth et al., 2007). Data used for yield calculation was based on fermentation of mixed food waste from restaurants (rice, noodles, meat, and vegetables) and not waste bread. Impact assessment results from this study showed that the GWP for biogas

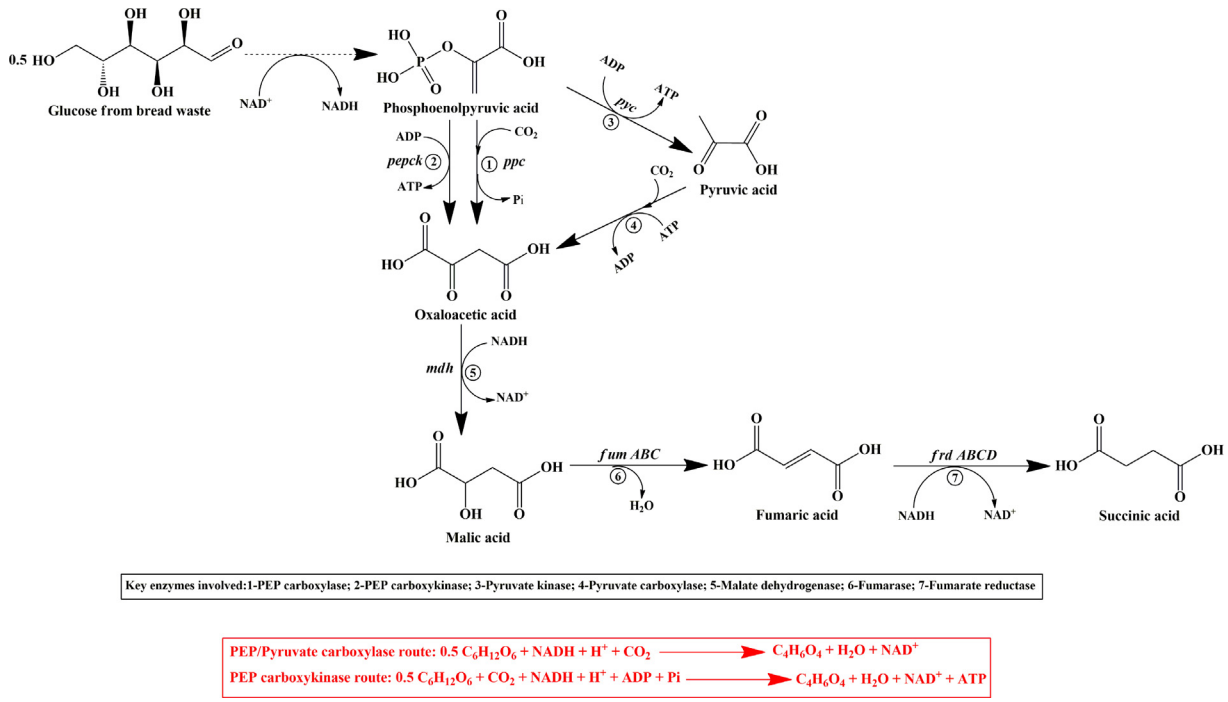

Fig. 1. Biochemical pathway for succinic acid production from glucose in *Actinobacillus succinogenes*.

was only 22 CO_2 kg eq./ ton of food waste. This was significantly lower than other similar studies based on food waste. On the other hand, GWP for SA production was 667 kg CO_2 eq./ ton SA. Brunklaus et al. (2018) also estimated the GWP for yeast-based fermentation of corn to SA, and it was found to be 2340 kg CO_2 eq./ ton SA. They concluded that while the environmental impact was lower when using food waste from a valorisation perspective (biogas production); when considering it as a feedstock for SA production, food waste was found to be more favourable than the established feedstock, corn. It is to be noted though, while Brunklaus et al. (2018)'s study predicts GHG emissions of 2340 kg CO_2 eq./ ton SA for corn-based SA production, Cok et al. (2014)'s study, based on which these results are calculated, predicts a maximum impact of 1700 kg CO_2 eq./ ton SA for same feedstock. Brunklaus et al. (2018) have not provided any explanation for this inconsistency in results.

Albizzati et al. (2021) also presented an LCA study to describe SA production based on the fermentation of food waste. Similar to Brunklaus et al. (2018), SA production was modeled on the feasibility study by Lam et al. (2014), however, the yield used in the study was not derived from the results of Lam et al. (2014), but was assumed to be based on the glucose content of the feedstock (0.08 kg SA kg^{-1} w/w). As opposed to previous studies, Albizzati et al. (2021) also included indirect land-use changes, which have a significant impact on bio-based production processes that demand or displace crops. Along with LCA, this study also presented conventional and societal life cycle costing of the process. Albizzati et al. (2021) found that GWP for bio-based SA production using food waste as feedstock was 2.2 ± 0.03 kg CO_2-eq./kg SA higher than the reference SA. The analysis also suggests few areas of improvement, like the use of burden-free steam, recirculation of oil and NaCl, decrease in the use of potassium chloride, and increase in product yield. However, Albizzati et al. (2021) concluded that while these changes may reduce the GHG emissions, the overall economic and societal costs would be comparable to the reference product, only after increasing the plant capacity by 43%.

As can be seen from the brief literature review presented above, though a couple of studies have investigated the environmental performance of fermentative production of SA using waste bread, these studies have either been based on *E. coli* based fermentation as microbial cell factory (Brunklaus et al., 2018) or calculated the yield based on glucose content and not directly based on waste bread (Albizzati et al., 2021). The objective of this study is to evaluate the environmental performance of fermentative SA production using waste bread as feedstock, where fermentation is carried out using *A. succinogenes* and the productivity and yield values are directly calculated based on the weight of the feedstock. A cradle-to-gate LCA is implemented and environmental impacts in terms of GHG emissions and non-renewable energy demand of SA production have been calculated and compared with the established SA production processes through biological and petrochemical routes.

The analysis presented in this work is focused on assessing the answers to three main questions. First, what are the environmental hotspots or the largest contributors to environmental impact among the different raw materials and energy streams that are used in the production process. Second, how do the impact findings of NREU and GHG emissions for SA produced from bread waste compare to SA synthesis from well-established fermentative and petrochemical route. Finally, how does the allocation approach used to account for the by-product influence the environmental performance of SA production.

2. Process description

Fig. 2 illustrates the flowchart of the fermentative SA production using bread waste. All the key steps in the conversion process remain similar to those considered by Lam et al. (2014). The process starts with bread waste collection and transportation to the pilot plant, followed by grinding of bread waste into smaller pieces $< 1\ cm^3$. The small pieces were then blended with water. This was followed by enzymatic hydrolysis which was carried out at 55^{0}C

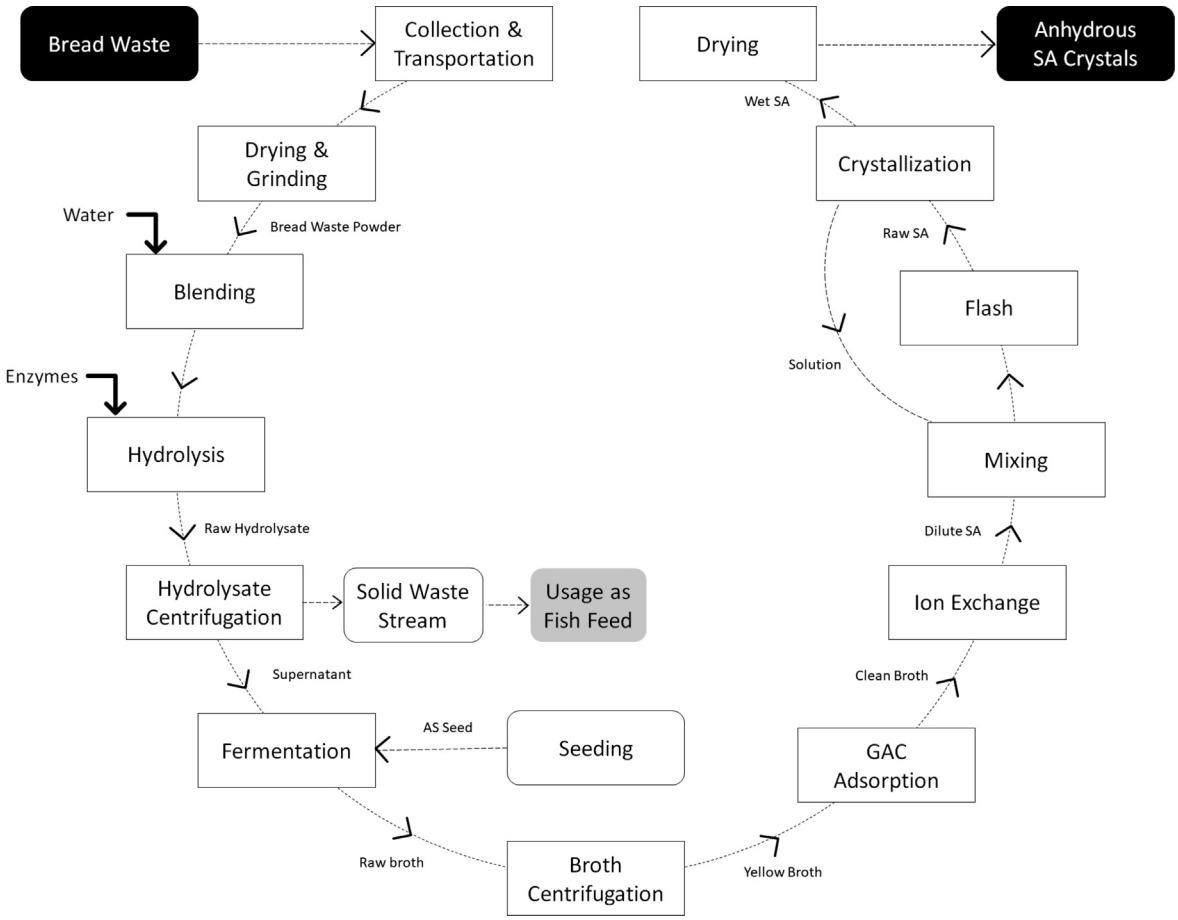

Fig. 2. Flowsheet of the fermentative SA production process from bread waste.

for 24 h using industrial grade glucoamylase and protease to release glucose and free amino nitrogens, respectively. The solid and liquid fractions were separated after the hydrolysis, and the supernatant was filtered before transferring to the bioreactor for fermentation. The supernatant was supplemented with additional nutrients and SA fermentation was performed with *A. succinogenes* using an inoculum of 5% (v/v) at 37 °C. The carbon dioxide was continuously sparged and pH was controlled using $MgCO_3$ and NaOH. Complete details of the production process can be found in previous studies (Leung et al., 2012; Zhang et al., 2013; Lam et al., 2014).

3. LCA methodology

The LCA is conducted in four phases: (i) goal and scope definition (ii) inventory analysis; (iii) impact assessment and (iv) interpretation, in line with ISO standards: 14040–14044 (ISO, 1997; ISO, 1998; Finkbeiner et al., 2006).

3.1. Goal and scope of study

The goal of this study is to assess the potential environmental impacts and benefits of fermentative SA production using bread waste. The functional unit used to report the environment profile is 1 kg industrial grade (99.5% wt.) SA. The scope of the LCA spans from the 'cradle' (raw material extraction) to the production gate before its distribution for end use. SA is a chemical intermediate and can be further used in a wide range of applications, and therefore product logistics, use and end of life, have been left out of the scope of this study. The study analyses the NREU and GHG emissions from the SA production process using bread waste, and the results were compared with the SA production processes using the conventional fossil-based route and other renewable feedstock to determine the relative environmental performance of the process.

3.2. Life cycle inventory

The inventory data used in this work is based on the primary data obtained for the SA production pilot plant described previously by Lam et al. (2014), who developed process simulation models and conducted an analysis for the plant processing 1 tonne/day of bread waste with 312 operating days annually. The use of process simulation data for LCA is a common approach used for the processes that are tested only at the laboratory or pilot scale (Kumar and Murthy, 2012; Shemfe et al., 2018; Sajid et al., 2016; Sadhukhan et al., 2019). While Lam et al. (2014)'s study is based on bread waste collection and treatment in Hong Kong, this work is based in the UK context. The primary data on chemicals, co-products, and utilities used in the production process were obtained from the various reports generated from the process simulations. Based on this information, the inventory data for producing 1 kg SA is calculated and presented in Table 1. Data on the corresponding waste emissions has been obtained from Carlsson (2016). Ecoinvent LCI database version 3.6 (Wernet et al., 2016) was used to extract majority of the background life cycle inventory data. However, some other databases, such as USLCI and Industry data 2.0 were also used for some inputs/processes (steam, sodium

Table 1
Life cycle inventory (LCI) data for the production of 1 kg SA (data compiled based on information from Lam et al. (2014) and Carlsson (2016)).

Inputs	Amount	Unit	Process/Data source
Chemicals			
Hydrochloric acid	0.079	kg	Hydrochloric acid, without water, in 30% solution state {RER}/Ecoinvent 3
Magnesium carbonate	0.273	kg	Potassium carbonate {GLO}/Ecoinvent 3
Sodium hydroxide	0.197	kg	Sodium hydroxide, production mix, at plant/RNA/USLCI
NaCl brine	15.007	kg	Sodium chloride, brine solution {GLO}/ Ecoinvent 3
Enzyme, Glucoamylase	0.007	kg	Enzyme, Glucoamylase, Novozyme Spirizyme/kg/RER/USLCI
Utilities			
Steam	40.531	kg	On-site steam average/Industry data 2.0
Electricity	1.010	kWh	Electricity, medium voltage {GB}/ Ecoinvent 3
Heating oil	3.385	kg	Kerosene {Europe without Switzerland}/Ecoinvent 3
Process water	117.142	kg	Tap water {Europe without Switzerland}/Ecoinvent 3
Energy for CO_2 capture, compression and storage	0.100	kWh	Christodoulou et al. (2017)
Output			
Succinic acid crystals	1	kg	
Solid biomass (fish feed)	10.639	kg	
Waste Emissions			
Nitrogen oxides	2.223	g	ecoinvent data on emissions to air
Ammonia	0.279	g	
Sulfur dioxide	2.264	g	
Carbon dioxide, fossil	722.460	g	
Carbon dioxide, biogenic	36.123	g	
Methane, fossil	3.717	g	
Methane, biogenic	0.041	g	
Chromium VI	0.074	mg	
Arsenic	0.477	mg	
PAH	0.421	mg	

hydroxide, and enzyme). Database used for different inputs is also highlighted in Table 1. As the plant location is assumed to be in the UK, data specific for electricity production mix, chemicals, and other secondary data was based on the UK or European Union averages, when available.

Certain assumptions have been made in the LCI to simplify the analysis. Bread waste is assumed as a post-consumer waste and thus the environmental impact associated with the generation of the bread waste is excluded from this analysis. Also, typically bread waste is collected and transported to a landfill site (along with other food waste); therefore the additional energy use and emissions due to transportation & collection of the bread waste to the biorefinery are assumed to negligible, and hence not considered to be part of this analysis. This assumption is in accordance with similar studies in literature, where transportation impacts are negligible (Moussa et al., 2016; Zah et al., 2007; Khoo et al., 2010; Gironi and Piemonte, 2010; Brunklaus et al., 2018; Bjarnadottir et al., 2002). For confirmation, a maximum distance of 1000 km was assumed (in the UK scope), and the respective contribution towards GHG emissions and NREU were calculated. Considering a transport, lorry > 32 metric ton, RER process from ecoinvent, with EURO6, and assuming that based on feed requirement and all wastage, 2.5 kg waste bread is transported for each kg of SA, GHG emissions and NREU were estimated to be only 0.018 kg CO_2 eq./kg SA and 0.317 MJ/kg SA, respectively. As it is shown later in the results and discussion section, these values account for less than 1.5% of the total impacts, and hence the assumption remains valid even after assuming a very high transport distance and over estimation of feed. The impact of waste bread separation is assumed to be negligible compared to all other inputs and utilities for the process, and hence not included in the analysis (Elginoz et al., 2020).

Manufacturing and maintenance of the plant infrastructure are considered to have an extended life and large throughput, and therefore environmental impacts of infrastructure are also not included. Smidt et al. (2015) showed that infrastructure impacts were negligible for large scale processes except for the effect on metal depletion. Bread waste based fermentative production of SA using *A. succinogenes* is assumed to follow the theoretical optimal stoichiometry as presented by Gunnarsson et al. (2014), where 1 mol of CO_2 is consumed for producing 1 mol of SA. For the analysis, it is assumed that CO_2 is captured from a coal fired power plant and transported 100 km to the SA pilot plant, and then compressed and stored on-site for continuous use. Herein, a transport distance of 100 km is an approximation, and this may vary depending on the actual plant location.

All supplied CO_2 was assumed to be converted to SA. As the CO_2 was captured from a fossil-source, an equivalent amount of fossil CO_2 is deducted from the total CO_2 emissions. This way environmental credit for CO_2 sequestration is accounted in the impact assessment calculations. Environmental impacts of the transportation of supplementary chemicals to the pilot plant are assumed to be negligible (Cok et al., 2014; Moussa et al., 2016).

3.3. Allocation procedure

Most of the bio-processes produce some co-products along with the main product and the method used to allocate energy use and emissions among the main product and co-product can significantly impact the results (Kumar and Murthy, 2012). In addition to SA, the current process produces large quantities of solid wastes streams that can be used as fish feed (Lam et al., 2014). This solid biomass has some value and is thus not perceived as a waste but as a by-product in this analysis. Various methods based on either mass, energy or economics are used to allocate environmental impacts among the main product and co-products. The system expansion is another common approach used to account for the impact of additional products, where co-product is assumed to replace some other product in the market and the environmental impacts of that product are credited to the current process (Hermann et al., 2007). ISO 14044 standard recommends that allocation should be avoided whenever possible (Mackenzie et al., 2017). This leaves us with system expansion; however, this approach is typically only used when the co-product can also be independently produced by a stand-alone process (Hermann et al., 2007). As there is no independent process for producing the particular solid waste stream as obtained in the current SA production process, we have used allocation to account for its impact. For this study, the mass allocation has been performed for the solid biomass (fish feed), as economic allocation led to unreasonably high environmental impact. Based on mass balance, allocation factors used were 0.086 and 0.914 for SA and solid biomass, respectively. To highlight the large difference in results based on alternative allocation approaches, a sensitivity analyses is presented in Section 4.3.

3.4. Life cycle impact assessment (LCIA)

The life cycle impact assessment (LCIA) methodologies adopted for this study were single-issue LCIA methods, namely, Cumulative Energy Demand (CED) (v 1.11) and IPCC 2013 GWP 100a (v 1.03), as implemented in SimaPro 9.1. CED describes "total quantity of primary energy which is necessary to produce, use and dispose a product", and provides characterization factors for the energy resources (non-renewable and renewable). IPCC 2013 GWP 100a was developed by the Intergovernmental Panel on Climate Change (IPCC) and describes the global warming potential with a time horizon of 100 years. Results are presented using two impact categories calculated from the above methods, GHG emissions and NREU, expressed as kg CO_2 equivalent and MJ of primary units of fossil energy resource depletion per functional unit, respectively. The system model, Allocation at the point of substitution or APOS, was used in the analysis.

4. Results and Discussion

4.1. Contribution analysis

Fig. 3A illustrates the impact on GHG emissions in terms of kg CO_2 eq, as a function of different chemicals and utilities used in the production of 1 kg SA by fermentation of bread waste. As shown in Fig. 3A, the biggest contribution of GHG emissions is due to steam with 0.98 kg CO_2 eq/kg SA, which is about 76% of the total 1.3 kg CO_2 eq generated in the fermentative production of SA using bread waste. Heating oil is the second biggest contributor to GHG emissions with 0.14 kg CO_2 eq, which is about 11% of the total GHG emissions produced from the process. All the remaining supplementary chemicals and utilities each contribute less than 5% GHG emissions. Thus, it can be concluded that unit operations involving steam use are the main environmental hotspots during SA production from bread waste.

Environmental impact can be reduced if the steam is derived from a parallel process in the biorefinery which generates steam as a co-product, thereby minimizing the environmental burden for generating the on-site steam at the plant (Hermann et al., 2007). It should be noted that steam was also found to be the biggest contributor to utility cost in the economic feasibility study performed by Lam et al. (2014). Therefore, focusing on abetting the impact of steam on GHG emissions will also help in reducing the overall cost of production.

Fig. 3B shows the impact on NREU of different supplementary chemicals and utilities used in the production of 1 kg SA by fermentation of bread waste. Results show that heating oil is responsible for almost half, about 50%, of the total 31.55 MJ used in the production of 1 kg SA. In addition to heating oil, steam is also a large contributor to NREU, requiring 14.21 MJ per kg of SA, which is equivalent to 45% of the total. All the remaining chemicals and utilities contribute less than 1%. In summary, there are two major hotspots, heating oil and steam in terms of non-renewable fossil energy use.

It should be noted that in the current analysis we have considered on-site steam generation and instead of choosing any one particular fuel, we have used the average data set (considering inputs from different fuels such as coal, oil, natural gas, biomass, solar, etc.) from Industry data 2.0 database. As steam is found to be the primary environmental hotspot for SA production influencing both GHG emissions as well as NREU, large scale implementation of the proposed biorefinery approach should be focussed on reducing the impact of steam. One possible strategy can be making alternative use of waste agricultural biomass for steam generation. For example, the production of steam through the burning of sugar cane bagasse in the boiler is common practice in sugar mills, which has a much lower impact than those using traditional fossil fuels (Zhang et al., 2020). Overall, focussing the optimization of the process in ways that reduces or replaces the use of steam and heating oil with renewable alternatives can help in making the process greener and more sustainable.

4.2. Comparison with other biomass-based and fossil-based SA production processes

As mentioned previously, different feedstock such as dextrose (derived from corn wet mill) and sorghum grains (a non-food crop), have been employed to produce SA (Cok et al., 2014; Moussa et al., 2016). Other than these substrates, the majority of SA is being produced by the conventional petrochemical route, which involves catalytic oxidation of n-butane into maleic anhydride (Smidt et al., 2015). In this section, we compare the impact of SA production from different feedstocks and routes on GHG emissions and NREU. Data regarding the GHG and NREU impact categories for fossil-based and other biomass-based SA production is obtained from previous LCA studies by Cok et al. (2014),Smidt et al. (2015), and Moussa et al. (2016), who have used data from operating plants of Reverdia (dextrose) and Myriant Corporation (sorghum) for the sustainability analyses.

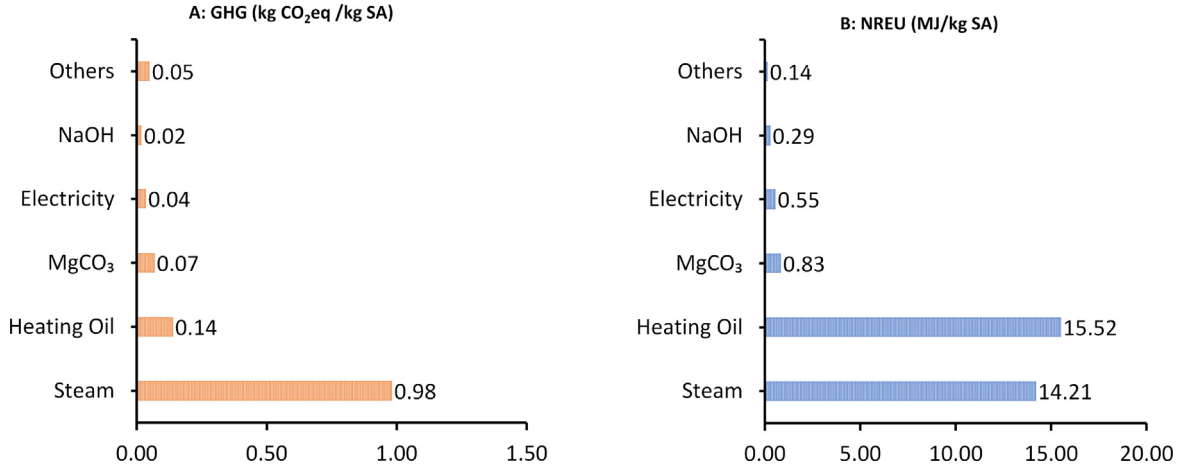

Fig. 3. Individual contribution of supplementary chemicals and utilities towards (A) GHG Emissions and (B) NREU during SA production from fermentation of bread waste.

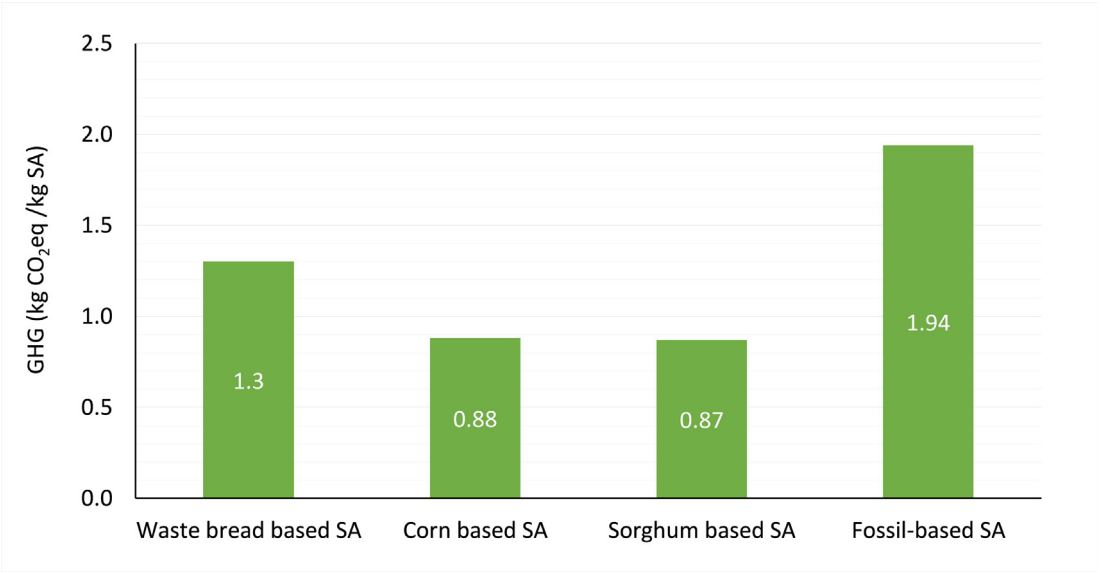

Fig. 4. Comparison analysis of GHG emissions (kg CO_2 eq) during 1 kg SA production from fermentative (with three renewable substrates, bread waste, dextrose (Cok et al., 2014) and sorghum grains (Moussa et al., 2016)), and traditional petrochemical route (Cok et al., 2014).

Fig. 4 shows the total GHG emissions generated from the production of 1 kg SA by using processes with different biomass- and fossil-based SA production. SA produced from biomass-based substrates such as dextrose or sorghum grains, generates 0.88 and 0.87 kg CO_2 eq GHG emissions, respectively, which are also the lowest emissions reported among the different processes in Fig. 4. Compared to these two established processes that are being operated by Reverdia and Myriant Corporation respectively, SA production by bread waste fermentation generates 1.3 kg CO_2 eq GHG emissions, which is around 47% higher than Reverdia's process. Fossil-based SA production process generates even higher GHG emissions, 1.94 kg CO_2 eq, which is 120% higher than Reverdia's process and around 50% higher than SA generated from bread waste. Here we can see that although the SA production process from the fermentation of bread waste is still not as clean as the production processes that use dextrose or sorghum grains, it has a much lower environmental impact when compared to fossil-based SA production.

It should also be noted that both Reverdia and Myriant Corporation have optimized their processes over many years for maximum production with minimum impact. Similarly, there is a vast scope for further optimization of the SA production process from bread waste fermentation, particularly, the unit operations involving product purification, in which the current process consumes a large amount of energy (Lam et al., 2014). Also, the fact that waste bread is a post-consumer waste and does not require any new arable land for its production, would work in its favour when considering land-use changes.

Fig. 5 shows the NREU for the production of 1 kg SA by using processes with different biomass-based substrates and for SA produced from fossil resources. It can be observed that among these four options, the process using sorghum grains as substrate has the minimum non-renewable energy demand, 6.89 MJ/kg SA. This was observed mainly because of the environmental credit added in the LCA (using system expansion approach) due to the co-production of ammonium sulfate (a high value co-product) (Moussa et al., 2016). Other bio-based production processes using dextrose and bread waste, demand almost similar quantities of non-renewable energy per kg of SA, 32.7 MJ and 31.55 MJ, respectively. In comparison, fossil-based production of SA has much a higher non-renewable energy requirement, 59.2 MJ/kg SA, which is ~ 760%, 81% and 88% higher demand than that used by sorghum, dextrose, and bread waste based SA production, respectively.

Here it can be highlighted that the energy demand of the fermentative production process of SA using bread waste is not only lower than the petrochemical route, but it is also closely matching the industry standard when compared to Reverdia's dextrose-based process. Optimization and scaling of the different unit operations of the pilot plant could potentially help in reducing the non-renewable energy demand further, making it environment friendly and lower the overall production cost.

In addition to the established processes based on dextrose (corn) and sorghum grains, the environmental impact of fermentative SA production using waste bread can also be compared to other biogenic feedstocks. For example, LCA results obtained by Brunklaus et al. (2018) show that fermentation of mixed food waste from restaurants (rice, noodles, meat, and vegetables), leads to GHG emissions of 0.667 kg CO_2 eq./kg SA and NREU of 10.2 MJ/kg SA. These results are very encouraging, as they show that using mixed food waste for SA production can lead to 50% less GHG emissions and would require only 1/3 of non-renewable energy, compared to that using waste bread as the feedstock. These impact parameters are even better than the established processes, except for NREU result when using sorghum grains (6.89 MJ/kg SA), as reported by Moussa et al. (2016). However, it must be noted that the results by Brunklaus et al. (2018) are based on a small laboratory-scale experiment and the process would need to be optimized for pilot/large scale operations to consistently maintain such high productivity.

González-García et al. (2018), reported that SA production using apple pomace fermentation results in GHG emissions of 5.30 kg CO_2 eq./ kg SA, which are 4 times higher than the emissions obtained in the current study for SA production using fermentation of waste bread. González-García et al. (2018) did not report NREU values, however, they calculated the cumulative energy demand (this includes both renewable and non-renewable energy usage), which was found to be 227 MJ/ kg SA. As non-renewable energy is typically the bigger contributor to total energy demand, it can be confirmed that energy usage for fermentative SA production using apple pomace was higher than that using waste bread.

The closest study to the current work is the analysis by Albizzati et al. (2021), who also used waste bread as a feedstock for the fermentative production of SA. However, since they assumed a much lower yield, 0.08 kg SA kg^{-1} w/w, GHG emissions from this process per kg SA was found to be significantly higher than the fossil-based process, and therefore much higher than the current work. Albizzati et al. (2021) did not report NREU for the process.

A European Commission report (De Matos et al., 2016) presented the detailed cradle-to-gate LCA analysis results from a previous study by Patel et al. (2006). This study provided the predictions of how improvements in fermentation broth concentrations and productivities, along with improved product separation and purification schemes, could lead to increased environmental savings for the different fermentation based bio-processes, about 2–3 decades in the future. These scenarios specifically looked at bio-based SA production using anaerobic continuous fermentation of dextrose and calculated the environmental impact of the process based on sugar extracted from different sources, such as starch, sugar cane or lignocellulose. Predictions for GHG emissions and NREU obtained from this analysis were 0.3 kg CO_2 eq., −0.6 kg CO_2 eq., and −0.2 kg CO_2 eq., and 28.0 MJ, 9.1 MJ and 17.5 MJ per kg SA, for sugar extracted from starch, sugar cane and lignocellulose, respectively (Patel et al., 2006). While these results were calculated for dextrose, considering the comparable impacts as shown in the current analysis, the future scenarios for waste bread as feedstock could also be commensurate. The prospective results are ambitious but show promise that with effective optimization bio-processes can sustainably replace their fossil-based counterparts.

4.3. Sensitivity analyses, how does the allocation decision influence the LCA results?

In this LCA study, the solid biomass generated in the production process is accounted for by using mass allocation. Because of the large amount of solid biomass produced (10.69 kg/kg SA), it had a significant impact on the results. Herein, we present a sensitivity analysis comparing the mass allocation approach to three other scenarios: economic allocation, no-allocation, and system expansion approach.

Lam et al. (2014) postulated that the biomass generated in the production process could be sold as fish feed at a price of $0.45/kg. Compared to this, the average price of SA fluctuates between $3–8/kg (Pais et al., 2016). In this study, the selling price of SA was assumed to be $5/kg. When considering economic allocation for the first alternate scenario, based on the individual price per unit, the economic allocation factors for SA and the solid biomass were estimated to be 0.51 and 0.49, respectively. For the second scenario, where we do not account for solid biomass at all, 100% allocation is assigned to SA.

Obtaining data for the third scenario is challenging, mainly because there is no separate stand-alone process that generates the same type and quality of fish feed, which is equivalent to the solid biomass generated from the bread waste fermentation. Fish feed typically consists of two main types of ingredients, crop-derived and fish derived ingredients in approximately 50:50 ratio by weight (Pelletier and Tyedmers, 2007). As the solid biomass generated in the SA production process is mainly derived from the fermentation of bread waste, for this analysis we assumed it can replace the crop derived component of the fish feed. To implement the system expansion approach, the energy used (in MJ) and GHG emissions (in kg CO_2 eq) for producing 1 kg crop derived ingredients of fish feed were calculated, and equivalent NREU and GHG emissions were subtracted from the same values generated during production of SA by fermentation of bread waste (Pelletier and Tyedmers, 2007; Pelletier et al., 2009).

Fig. 6 shows the results of the sensitivity analyses, comparing GHG emissions and NREU for the four LCA scenarios discussed above, mass allocation, economic allocation, no allocation and system expansion approach. As expected, LCA with no allocation resulted in the worst in both impact categories, generating 15.2 kg CO_2 eq. and non-renewable energy demand of 379.8 MJ per kg SA. This is mainly because the total impact of the used raw materials and energy consumed in the process were assigned to the single product, i.e. SA. The GHG emissions for the economic allocation and system expansion approach were identical (7.5 kg CO_2 eq. in both cases) which were about half of that generated with no allocation, but these were still significantly (about 475%) higher than that obtained from LCA with mass allocation.

Results for NREU also show a much higher impact in all three alternate scenarios when compared to the mass allocation

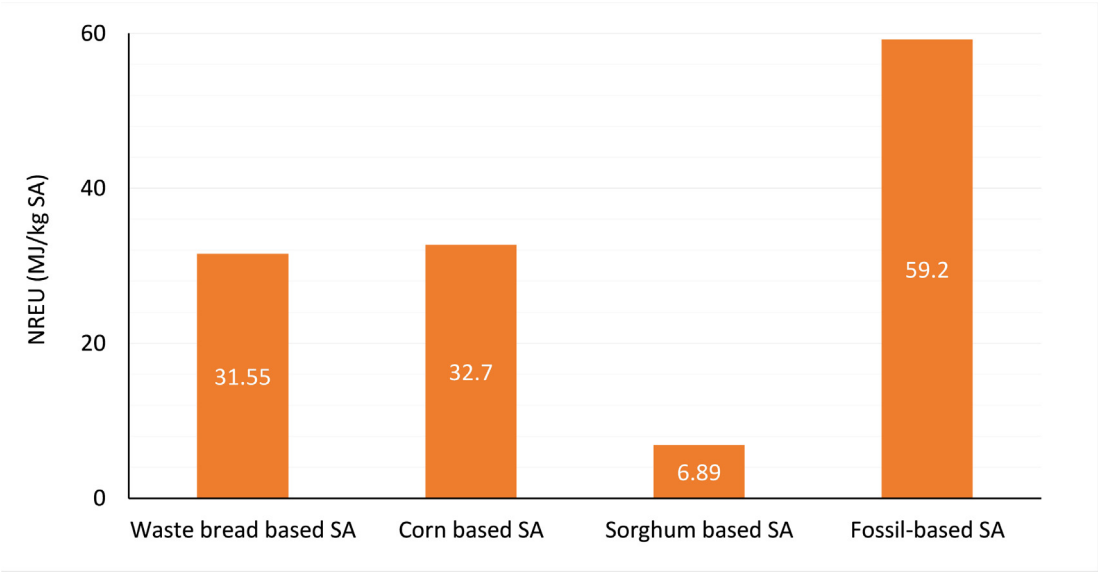

Fig. 5. Comparison analysis of NREU (MJ) of 1 kg SA production from three biomass-based substrates, bread waste, dextrose and sorghum grains, and the traditional petrochemical route.

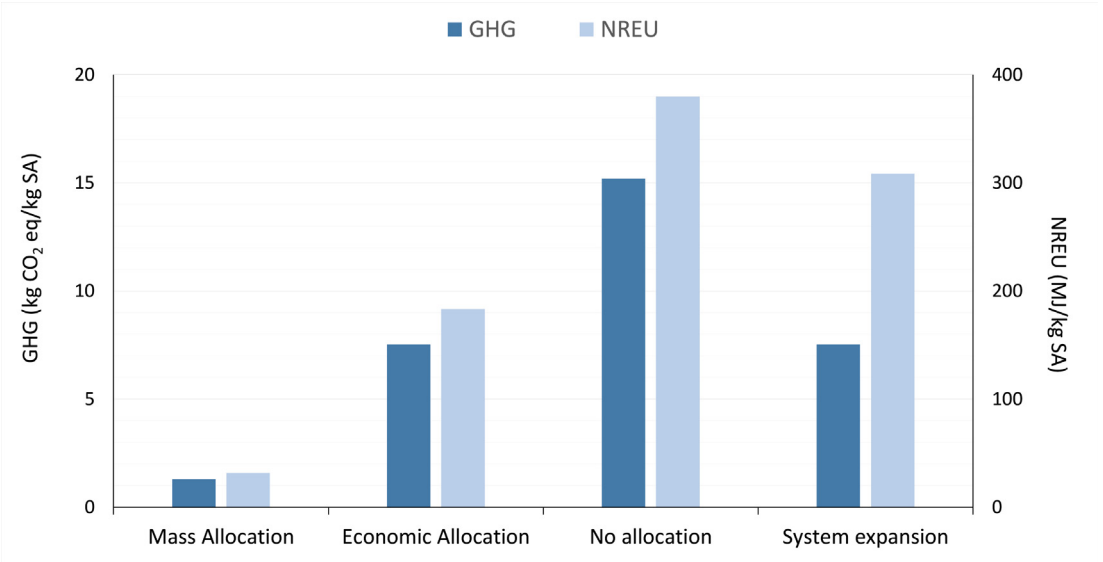

Fig. 6. Life cycle impact assessment results in terms of GHG emissions and NREU of SA production by fermentation of bread waste using different approaches to account for by-product.

approach, with 480%, 1,100% and 880% higher for economic allocation, no allocation and system expansion approaches respectively. These results highlight, how the attribution of the by-product can influence the sustainability of the SA production process from bread waste fermentation.

LCA with economic allocation results in higher impacts on GHG and NREU because of the large difference in the pricing of the two products. It should also be noted that because the by-product is produced almost 10 times per kg of SA, the combined effect of the difference in price and production amount, assigns a much higher impact on SA production in this approach. Here, we have assumed the fish feed cost as $0.45/kg, however, this varies a lot, depending on the type of fish and the country where it is sold. All the above factors need to be finalized to negate the distorting effect of market prices before the use of economic allocation can be deemed feasible for this LCA.

Similarly, the system expansion approach as explained before is also based on lot of assumptions, and it would be incorrect to use this approach before a detailed comparison between the composition of the solid biomass and the crop-based ingredients of fish feed is performed. If instead of only accounting for the crop-based ingredients, the solid biomass can replace the complete fish feed, the impact of the process on GHG and NREU will be reduced. Nevertheless, LCA results with a system expansion approach may give quite unrealistic results before such assumptions are based in some justification. Therefore, in addition to improve the yield of the main product, it is very important to exploit the full value of the by-product and use the correct approach in performing the impact calculation of the production process.

Looking from the UK perspective, the total 328,000 tonnes of bread waste generated annually can be used as a feedstock for the production of SA (replacing the petrochemical route) in this proposed biorefinery. The SA yield achieved by Leung et al. (2012) was 0.55 g SA per g of bread waste, and based on this calculation, nearly 50% of bread waste (about 164 kiloton) can be recovered in the form of top platform chemical, SA. Furthermore, it can result in GHG emission savings of 17,354 tonnes of CO_2 eq. and non-renewable energy savings of 738,232 GJ per year. These numbers highlight the enormous potential of bread waste as a feedstock for second generation (2G) biorefinery which is based on non-edible/waste biomass. While this study is focused on just one platform chemical, SA, the high starch content of bread waste and easy extraction of fermentation sugars in comparison to alternative biomass based feedstocks makes it an attractive substrate for fermentation based biorefinery processes for the production of a wide range of biochemicals and biofuels such as lactic acid, ethanol, etc. It is envisaged that the positive environmental profile for fermentative SA production using bread waste, as highlighted in this study, would lead to further studies towards profitable waste bread based fermentative SA production, eventually contributing towards the establishment of a circular economy. In this way, it can make a notable impact on bioeconomy of the UK and contribute towards UK's goal to achieve zero carbon emission by 2050.

5. Conclusions

This study presents the environmental impact in terms of GHG emissions and non-renewable energy demand of SA production based on fermentation of bread waste. Subprocesses requiring steam and heating oil were found to be the environmental hotspots of the process. The environmental impact of SA production from fermentation of bread waste was found significantly lower compared to the fossil-based SA production. And, although the emissions and fossil energy use were relatively higher compared to the SA production from dedicated crops like corn and sorghum grains, the use of the waste material (bread) avoids the use of food crops and the need for arable land. Sensitivity analysis demonstrated the importance of correct allocation of the solid biomass generated as a by-product in the system, as it can significantly alter the LCA results. Energy requirement and GHG emissions from the proposed process can be reduced in a targeted way by focusing on the largest contributors identified in this analysis.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Life cycle analysis of fermentative production of succinic acid from bread waste

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