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Pursuing single or combined wheat straw based poly(butylene succinate) production routes: A life cycle approach of first- and second-generation feedstocks

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ABSTRACT

The depletion of fossil resources and the climate change crisis call for an urgent shift to production pathways based on renewable and low-carbon sources. In addition, plastic pollution worldwide motivates the identification of new sources for their bio-based counterparts, which have an increasing demand. This research aims to evaluate the environmental feasibility of different cereal-based feedstocks for the production of poly(butylene succinate) (PBS), which is obtained from the polymerisation of succinic acid (SA) and 1,4 butanediol (BDO) monomers. The baseline scenario analysed corresponds to the use of wheat straw as a source of the fermentable sugars. Furthermore, five other cereal-based production routes combining first-generation (1G) feedstocks such as wheat and maize grain, and second-generation (2G) feedstocks, such as sorghum, barley straw, and maize stover, combined with wheat straw, were evaluated. The Life Cycle Assessment (LCA) methodology was used to identify the main hotspots of these valorisation routes at the early stage of the biorefinery design, considering all the burden categories provided by the ReCiPe impact method. The results showed that the straw-based PBS profile reached a Global Warming Potential of 3.43 kg CO_2eq, whereas a range value from 2.34 to 7.27 kg CO_2eq was estimated when wheat straw is combined with sorghum and barley straw, respectively. The pre-treatment stage represents a substantial impact on the strategy considered to produce fermentable sugars, particularly, for barley straw. Therefore, improvements are still required to reduce the energy demand and increase the sugar yield.

1. Introduction

About 140 million tonnes of petrochemical polymers are produced annually worldwide, and the growing petrochemical plastics production is expected to reach a yearly output of 850 million tonnes of plastic waste by 2050 [42]. In Europe, each year, nearly 26 million tons of post-consumer plastic waste is generated, where about 40% are sent to incineration [30].

Biopolymers have emerged to promote ecological conservation and reduce dependence on fossil resources in the current economic model. From them, it is possible to produce bioplastics with various uses, e.g., in biomedical devices such as wound dressings, bioresorbable implants, and drug delivery systems [3]. Bioplastics are an emerging market, representing only about 1.5% of the total plastics market with an estimated value of 390 Mt. (Plastics [33]). Accordingly, in terms of production levels, bioplastics are not comparable with their fossil-based counterpart, since bio-based production is not enough to fully meet the demand of the sector [16]. However, its world production is expected to increase from 2.2 to 6.3 million tons in a 5-year time frame [14]. In this regard, there is a need to identify diverse bio-based sources and production pathways to meet the anticipated growing demand for bioplastics in order to encourage the transition to a low-carbon economy.

Bioplastics encompass two types, on the one hand, those that are durable and non-degradable from a biological source, and on the other hand, those that are biodegradable [41]. Regarding the latter, they can be both bio-based and petrochemical plastics, where poly(butylene succinate) (PBS) biopolymer is one of them [7]. PBS has emerged as a substitute for polyethylene (PE) and polypropylene (PP), making it the

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most suitable option for packaging [34,35]. PBS is a white crystalline thermoplastic polymer with suitable mechanical and processability properties in textile filaments, infection molds, and extruded and blown products [2]. This biopolymer is obtained from the polycondensation of succinic acid (SA) and 1,4-butanediol (BDO), and it can be produced both by monomers derived from petroleum source and by bacterial fermentation [3]. With respect to other biopolymers (e.g., polylactic acid - PLA and polyhydroxyalkanoates - PHA), PBS presents eco-efficient end-of-life management routes, when mechanical recycling, incineration and composting were evaluated [13].

To design the process modelling for the potential production system of this bioproduct, biorefinery arise as the key concept to transform different bio-based feedstocks under an environmental viability perspective. In this regard, biopolymers (and subsequently bioplastics) can be obtained from first (1G) and second (2G) generation renewable sources. The 1G materials are mainly food crops, such as oil vegetables, sugarcane, and grain crops (e.g., sugarcane, barley, wheat, corn, cassava, soybean, sugar beet). The 2G feedstocks correspond to mainly lignocellulosic energy crops (e.g., sweet sorghum), perennial grasses (e.g., switchgrass), and biomass residues (e.g., straw) [26].

Cereals are relevant components in a healthy reference diet [44]. Among them, wheat is the most widely grown crop worldwide and is considered a staple crop for the food security challenge [9]. In 2020, about 119 Mt. of common wheat were produced in Europe, distantly followed by grain maize with 68 Mt. [15]. During cereal crops cultivation, the use of residual biomass after harvesting is a challenge that farmers should address. For instance, about 58 Mt. of wheat crop residues will be produced by 2030 in Europe [38]. Cereal straw has different commercial or agronomic uses: it can be sold as feed or bedding for livestock, left in the field as organic fertiliser, or burned to reduce pests and weeds in the next crop [17].

The environmental impacts of the bio-based production PBS have been addressed with different renewable feedstocks. For instance, Tecchio et al. [43] presented an evaluation protocol to predict the sustainability of new materials in the market, using PBS as a case study, and considering maize starch, sugar cane, and lignocellulosic biomasses for the bio-SA production. Patel et al. [32] studied the bio-based PBS production from corn (1G) and 2G feedstocks such as corn stover, wheat straw, miscanthus and hardwood, considering only cumulative energy demand (the non-renewable energy indicator) and greenhouse gas emissions. Following the agricultural waste utilisation initiative, Ioannidou et al. [20] evaluated the bio-based production of PBS from sugar beet pulp, corn glucose syrup, and corn stover.

This research encompasses two aims: i) to conduct an environmental assessment of PBS production from wheat straw using the Life Cycle Assessment (LCA) methodology to identify critical points from a life cycle perspective; and ii) to evaluate different production pathways for obtaining PBS based on both 1G and 2G cereal feedstocks to identify a variety of alternatives for its production. To do this, five scenarios are analysed, where wheat straw is combined with 1G (wheat and maize grain) and 2G (barley straw, maize stover and sorghum) cereal feedstocks. Thus, the environmental viability of different raw materials and pre-treatment methods for obtaining SA and BDO monomers is also evaluated.

2. Methods

2.1. Aim and scope definition

The aim of this research is to evaluate the production of PBS from wheat straw from an attributional environmental perspective. Then, this route will be compared with other cereal-based feedstocks to identify advantages and drawbacks in the valorisation route analysed at an early stage of the biorefinery design. To do this, the LCA methodology was applied following the International Organization for Standardization ISO 14040 and 14,044 guidelines [21,22]. The functional unit selected was 1 kg of PBS product. The scope of the research is limited to a cradle to biorefinery gate (see Fig. 1), i.e., processes are selected from the extraction of raw materials to obtaining the biochemical product for subsequent use.

2.2. Wheat grain and straw cultivation stage

Durum wheat (*triticum durum*) was grown on an area of approximately 0.25 ha in a monoculture regime in Apulia, Italy, the most important producer region. The process begins in August with soil preparation and ends with grain harvesting in June. Wheat grain (13% moisture) and straw (10% moisture) reach yields of about 5.5 and 3.5 $t{\cdot}ha^{-1}$, respectively. The market prices considered for grain and straw were 0.29 and 0.07 $€{\cdot}kg^{-1}$, respectively [39].

2.3. Process modelling of straw-based PBS

The biorefinery plant operates annually for 330 days with a plant capacity of 40 kt of PBS. The process modelling was performed using the Superpro designer ® software v11, and the literature studies used for the modelling, especially for bioconversion stages, were selected based on parameters such as final concentration, fermentation yield, and productivity.

2.3.1. Pre-treatment of wheat straw

The wheat straw composition (in dry matter) considered was 38.9% cellulose, 23.5% hemicellulose, 18% lignin, 4.5% protein, 14.5% extractives, 9.7% ash and 5.5% others [12]. The process begins by shredding the straw and sending it to a hydrothermal process for cellulose and hemicellulose fractionation. The process is carried out under high pressure (19 bar) at 210 °C with a solid-steam ratio of 1:2 [4]. The solid and liquid fractions obtained are separated by filtration. Then, the enzymatic hydrolysis process is performed using 20 $mg{\cdot}g^{-1}$ cellulose of Cellic® CTec3 cellulase (Novozyme, Denmark) at 50 °C for 72 h [24]. The solid lignin stream is recovered by filtration and passed to the steam generation stage to reduce the energy demand of the facility. The glucose-rich stream is sent to the SA and BDO production section. The process modelling is presented in **Fig. SM1** of the **Supplementary Materials.**

2.3.2. Succinic acid production

The SA production stage is modelled based on the work of Ma et al. [25] and Ioannidou et al. [20] to reach a SA concentration of 101.0 $g{\cdot}L^{-1}$ and a yield of 0.78 $g{\cdot}g^{-1}$. The production process consists of two sections: the bioconversion and the downstream separation and purification (DSP). The first consists of mixing water with the carbon source and nutrients (e.g., nitrogen sources, minerals like magnesium carbonate -$MgCO_3$), followed by thermal sterilization of the fermentation medium. The stream is then cooled to fermentation temperature (37 °C) before addition to the bioreactors. The inoculum is 10% (*v*/v) of the fermentation broth in the reactor. The pH is maintained at 6.7 by adding a 10 M sodium hydroxide (NaOH) solution. During fermentation, carbon dioxide (CO_2) is used due to the metabolic needs in the reductive cycle of tricarboxylic acid (TCA) to produce SA.

The second stage (i.e., the DSP) comprises separation and purification processes [1]. In the DSP stage, the biomass is removed by centrifugation of the fermented broth, which is later sent to activated carbon columns for the removal of impurities. The effluent is fed into cation exchange resin columns and the acidified liquid stream is then mixed with the stream coming from the crystallisation stage before being concentrated by evaporation (SA concentration of 214 $kg{\cdot}m^{-3}$). The concentrated liquid is treated by two-stage crystallisation at 4 °C. The SA wet crystals are dried up to a purity of 99.5%, and the overall SA recovery yield in the DSP is about 95% (*w*/w).

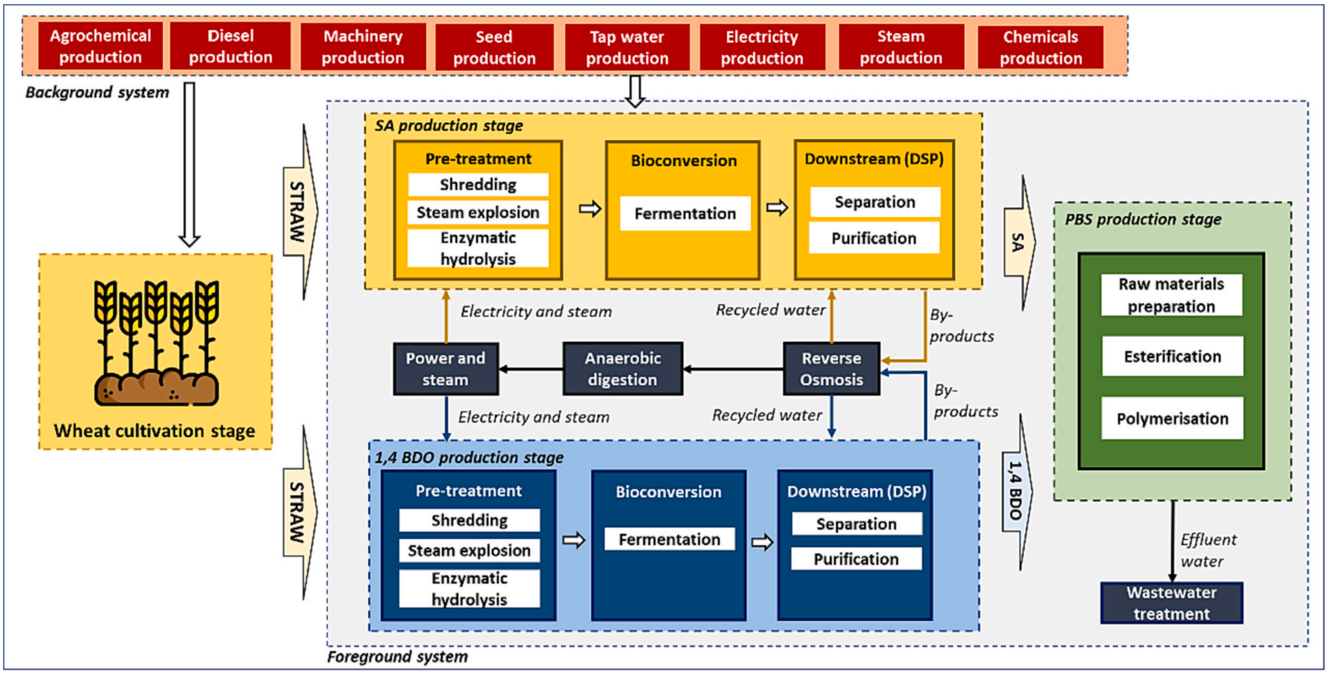


Fig. 1. System boundary of the straw-based PBS production.

2.3.3. BDO production

The process design of the BDO production was based on the fermentation efficiency reported by Burgard et al. [8], which used a genetically engineered *E. coli* strain, and the procedure carried out by Ioannidou et al. [20], which obtain a final BDO concentration of 125 $g\cdot L^{-1}$ with a yield of about 0.4 $g\cdot g^{-1}$. This production process is also divided into two sections: bioconversion and DSP. The bioconversion process was designed following a procedure like that of SA, where aerobic conditions are maintained in the fermentation stage. In the DSP stage, the biomass is removed by centrifugation and the liquid stream is processed through a series of cation and anion exchange resin columns to remove minerals and organic acid salts from the fermentation broth. The output liquid stream is concentrated to about 633 $g\cdot L^{-1}$ and then purified by distillation at 180 °C to separate the water and γ-butyrolactone (GBL). The BDO product achieves a purity of about 99.7% with a recovery yield of 92%. The residual biomass obtained from both BDO and SA production was used for steam generation.

2.3.4. PBS polymerisation

The process conditions for the PBS polymerisation were taken from Kamikawa et al. [23]. This step can be divided into three sections: raw material preparation, esterification, and polymerisation. In the first, the SA and BDO monomers are mixed at 80 °C using a molar ratio of 1.3:1. In the second, the liquid flow goes to the esterification reactor, which is carried out at 230 °C for 3 h. Then, the polymerisation is a polycondensation reaction requiring titanium tetrabutoxide as catalyst and is carried out at 240 °C for 16 h. The gaseous stream from the esterification and polymerisation reactors is distilled to recover the remaining BDO fraction. The PBS product is then cooled and can be granulated and stored (not considered here).

2.4. Comparison of PBS production with other cereal-based feedstocks

One of the potential barriers in the promotion of straw-based PBS production could be to secure feedstock supply [18]. Straw may be a valuable resource to farmers, and according to Giannoccaro et al. [17], some of the benefits of its current use are i) pest and weed reduction for the following growing season; ii) sales revenue; and iii) state subsidy under agri-environmental schemes when it is used as soil amendment. Consequently, other cereal-based feedstocks were combined with wheat straw to produce PBS with the objective of determining the potential of a variety of feedstocks to reduce the dependence on wheat straw. As a result, five additional bio-based pathways were considered (see Fig. 2). The scenarios were: i) WS + BS: wheat straw (WS) for 1,4 butanediol and barley straw (BS) for succinic acid; ii) WS + MS: wheat straw for 1,4 butanediol and maize stover (MS) for succinic acid; iii) S + WS: sorghum grain (S) for succinic acid and wheat straw for 1,4 butanediol; iv) WS + W: wheat straw for succinic acid and wheat grain (W) for 1,4 butanediol; v) WS + M: wheat straw for succinic acid and maize grain (M) for 1,4 butanediol. These scenarios will be compared with the wheat straw-based PBS (baseline) to identify possible advantages or disadvantages in the environmental performance of these cereal-based biorefineries. In addition, the implications of feedstock selection and pre-treatment strategies to obtain the same product will be identified.

2.4.1. Maize grain and Stover cultivation

The cultivation process of maize grain and stover used for the modelling is obtained from the work performed by Noya et al. [31], where the grain is destined for the food market. They evaluated the production of different classes of maize crops in Italy, following a cradle-to-gate perspective. Here, the 600 class of maize grain was selected as having the best environmental performance in almost all the categories studied, due to the low fertilisation requirement and the highest biomass yield of the sample analysed. The activities of this system can be grouped in three stages: i) land preparation, which consist of organic fertilisation, ploughed, harrowed, and sown; ii) herbicide control and mineral fertilisation that is carried out in the crop growth stage; and finally, iii) biomass harvesting. The system obtains a production yield of about 6.71 and 8.59 $t\cdot ha^{-1}$ of grain and straw, respectively. The prices considered were 247 €$\cdot t^{-1}$ for maize grain and 72.4 €$\cdot t^{-1}$ for the straw. Furthermore, following the work of Câmara-Salim et al. [10], it was considered that 30% of the stover is collected as it is a recommended harvest rate. For further details of life cycle characteristics of this cultivation stage, please see Noya et al. [31]. In addition, the allocation factors to distribute the burdens were obtained from the abovementioned study.

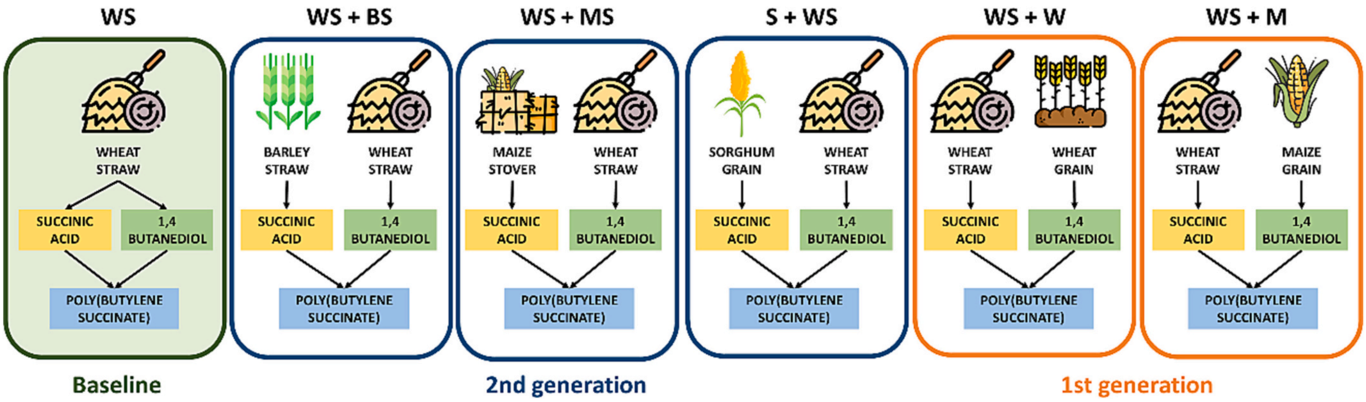


Fig. 2. Comparison of valorisation routes for producing PBS.

2.4.2. *Pre-treatment of barley straw and maize stover*

The pre-treatment of barley straw to obtain the fermentable sugars is based on the work performed by Raud et al. [37]. This process corresponds to a sudden decompression pre-treatment of the biomass. The barley straw contains cellulose (45.7%), hemicellulose (32.6%), lignin (5.2%), ash (3.8%) [37]. Briefly, in this process the biomass is dried to a moisture content of less than 10%. The biomass is then mixed with distilled water and added to the reactor, which is pressurised with nitrogen N_2 gas at a pressure of 10 bar. The temperature is then increased to 150 °C. Once the temperature is reached, the reactor is cooled to 80 °C, and the pressure is released through a valve. After this process, the flow is cooled to a temperature of 50 °C for enzymatic hydrolysis. This is carried out with an enzyme mixture (Accellerase 1500) at a rate of 0.3 ml per g of biomass. The process lasts 24 h at a temperature of 50 °C. From this pre-treatment process, a glucose yield of 338 g per kg of biomass is obtained. The flow diagram of this process is presented in **Fig. SM2** in the Supplementary Materials. Concerning the barley cultivation stage, the inventory data, considering an Italian context production, was obtained from the Ecoinvent® database v.3.8.

Regarding the pre-treatment of maize stover, the same procedure and operative conditions as for the pre-treatment of the wheat straw were carried out. The characterisation of the maize stover used was cellulose 40.2%, hemicellulose 28.5%, lignin 20.8%, protein 2.9%, extractives 1.5%, other 6.1%, which is obtained from the research of Lopes et al. [24].

2.4.3. *Pre-treatment of wheat and maize grain*

The pre-treatment process of the 1G feedstocks such as wheat and maize grains is obtained from the work of Bello et al. [6]. Accordingly, the grains are transformed in starch through a wet milling treatment process to split cereal starch and gluten components. Then, an enzymatic hydrolysis is performed to breaks down starch into glucose through the enzymatic activity. This treatment process produces co-products for food and feed industries such as gluten, meal, and bran in the case of wheat, as well as meal, oil, and gluten for the maize grain processing.

2.4.4. *Succinic acid from sorghum*

The business-as-usual production route of SA is based on the study addressed by ([5,27]. These authors used primary data on bio-SA production provided by Myriant Corporation®, an industrial biotechnology company in Quincy, Massachusetts. This company produces multiple chemical building blocks with proprietary technologies based on a one-step anaerobic fermentation process [27]. The process consists of fermentation, salt separation, evaporation, and crystallisation. For this process, the sorghum grain, a non-food energy crop in the United States, and dextrose are used as feedstocks. Furthermore, ammonium sulphate is produced as co-product (about 1.4 kg per kg of SA). For addressing the multifunctionality of the system, a system expansion approach was applied as was performed by Moussa et al. [27].

2.5. *Life cycle inventory and impact assessment method*

The life cycle inventory (LCI) of all systems evaluated are presented in the Supplementary Materials. The data of the cultivation stage of wheat grain and straw is presented in **Table SM1.** The LCI related to the PBS production based on wheat straw is presented in Tables **SM2-SM5**. The LCI of the maize grain and stover cultivation stage is showed in **Table SM6.** The inventory data of the pre-treatment method of barley straw and maize stover is available in **Table SM7** and **SM8**, respectively. Furthermore, the LCI of the pre-treatment of 1G feedstocks wheat and maize grains is presented in **Table SM9** and **SM10**, respectively. Finally, the inventory data of succinic acid production from sorghum feedstock is showed in **Table SM11.** On the other hand, to estimate the potential environmental burdens of straw-based PBS, all 18 midpoints categories (see Table 1 and their acronyms) from the ReCiPe 2016 (H) V1.07 / World (2010) (H) method [19] were considered.

2.6. *Sensitivity analysis of the baseline scenario*

The environmental profile of the PBS may vary depending on the assumptions considered in the modelling. Thus, three variations on the assumptions were considered as follows:

i. Characterisation of the straw: the cellulose content in wheat straw has a value between 28% and 39% [11]. Accordingly, the feedstock composition of Nabarlatz et al. [29] was considered,

Table 1
Environmental profile of 1 kg of PBS product in the baseline scenario.

Impact category	Acronym	Unit	Total
Global warming	GW	kg CO_2 eq	3.43
Stratospheric ozone depletion	SOD	mg CFC_{11} eq	13.55
Ionizing radiation	IR	kBq Co-60 eq	0.81
Ozone formation, Human health	OF, HH	g NOx eq	9.32
Fine particulate matter formation	PM	g $PM_{2.5}$ eq	6.24
Ozone formation, Terrestrial ecosystems	OF, TE	g NOx eq	9.44
Terrestrial acidification	TA	g SO_2 eq	17.60
Freshwater eutrophication	FE	g P eq	2.25
Marine eutrophication	ME	g N eq	3.03
Terrestrial ecotoxicity	TET	kg 1,4-DCB	12.32
Freshwater ecotoxicity	FET	kg 1,4-DCB	0.39
Marine ecotoxicity	MET	kg 1,4-DCB	0.53
Human carcinogenic toxicity	HT	kg 1,4-DCB	0.22
Human non-carcinogenic toxicity	HNT	kg 1,4-DCB	12.89
Land use	LU	m^2a crop eq	0.27
Mineral resource scarcity	MRS	g Cu eq	28.26
Fossil resource scarcity	FRS	kg oil eq	0.87
Water consumption	WC	m^3	0.22

which corresponds to a cellulose content of 28.4%, hemicellulose of 22.5%, and lignin of 15.9%.

ii. Market price of the straw: Given that the straw market is restricted, a higher demand may increase the value of this feedstock. Considering the baseline straw price of 0.07 €·kg^{-1}, an increase of two (0.14 €·kg^{-1}) and four (0.28 €·kg^{-1}) times was considered (maintaining the grain price). Thus, the allocation factors for the straw increased to 25.3% and 40.3%, respectively.

iii. Renewable electricity generation: Since electricity generation is evolving towards renewable sources, a potential scenario was assumed in which the electricity production mix is based solely on renewable sources such as wind (30%), hydro (30%), and solar (40%).

3. Results and discussion

3.1. Environmental profile of straw-based PBS

The environmental profile of the straw-based PBS for all impact indicators is presented in Table 1. In order to identify the processes that contribute most to the environmental performance of the PBS, an analysis of the critical points was carried out. Fig. 3a shows that the first stage of PBS production (i.e., the preparation of materials) is the main contributor with an average of 99% of the burdens, because of the production of SA and BDO (see Fig. 3b). On average, SA and BDO are responsible of about 54.7% and 44.9% of the total impacts. The profile of both monomers in the GW category was 2.31 kg CO_2eq per kg SA and 1.30 kg CO_2eq per kg BDO.

In the case of the SA production (see Fig. 3c), the critical process depends on the category analysed. For instance, the pre-treatment and DSP stages were the main hotspots in seven categories, ranging from 46.2% (FET) to 87.7% (ME) in the first one, and from 42.8% (FRS) to 65.2% (HT) in the second one. On the other hand, the bioconversion process stood out in four categories with contributions that range from 42.3% (GW) to 73.9% (MRS). As it can be noted from Fig. 3c, both in PM and FRS categories, bioconversion and DSP stages sharing similar contributions of about 41–43%, respectively. Furthermore, wheat straw and electricity in shredding were mainly responsible for loads in the straw pre-treatment stage, as well as ash treatment in toxicity-related categories (see Fig. 4a). The burdens of the bioconversion stage were motivated by the requirement of magnesium sulphate and electricity (see Fig. 4b), meanwhile, those related to the DSP stage were caused mainly by the separation process (see Fig. 4d), which is due to the hydrochloric acid (HCl) required for the resin regeneration. In the purification process, the electricity demand is the principal responsible.

Concerning the contribution analysis of BDO production (see Fig. 3d), the pre-treatment process was the critical process in almost all the impact categories, where the contributions of the streams were distributed as the SA monomer (since it is the same process). Nevertheless, the BDO bioconversion played a more relevant role, especially in the FET and HT categories, where this process was the most impactful accounting for about 54.6% and 66.5% of the impacts, respectively, due to the consumption of phosphorous nutrients (see Fig. 4c). Besides phosphorous minerals, the electricity demand also highlighted as a hotspot in the bioconversion stage of BDO (see Fig. 4c); whereas, in the DSP stage (see Fig. 4e), the purification process was the main responsible for the burdens due to electricity or water demand, depending on the impact category.

3.2. Sensitivity analysis of straw-based PBS

Fig. 5 shows the environmental profiles of the sensitivity analysis with respect to the straw-based PBS production scenario. It shows that the use of renewable sources for electricity generation reduces loads in almost all impact categories. The exception was the water consumption category, with marginal growth (3%) due to the higher share of hydroenergy in the generation mix. For the GW and FRS categories, loads decreased by 27% and 29%, respectively, while the largest reductions

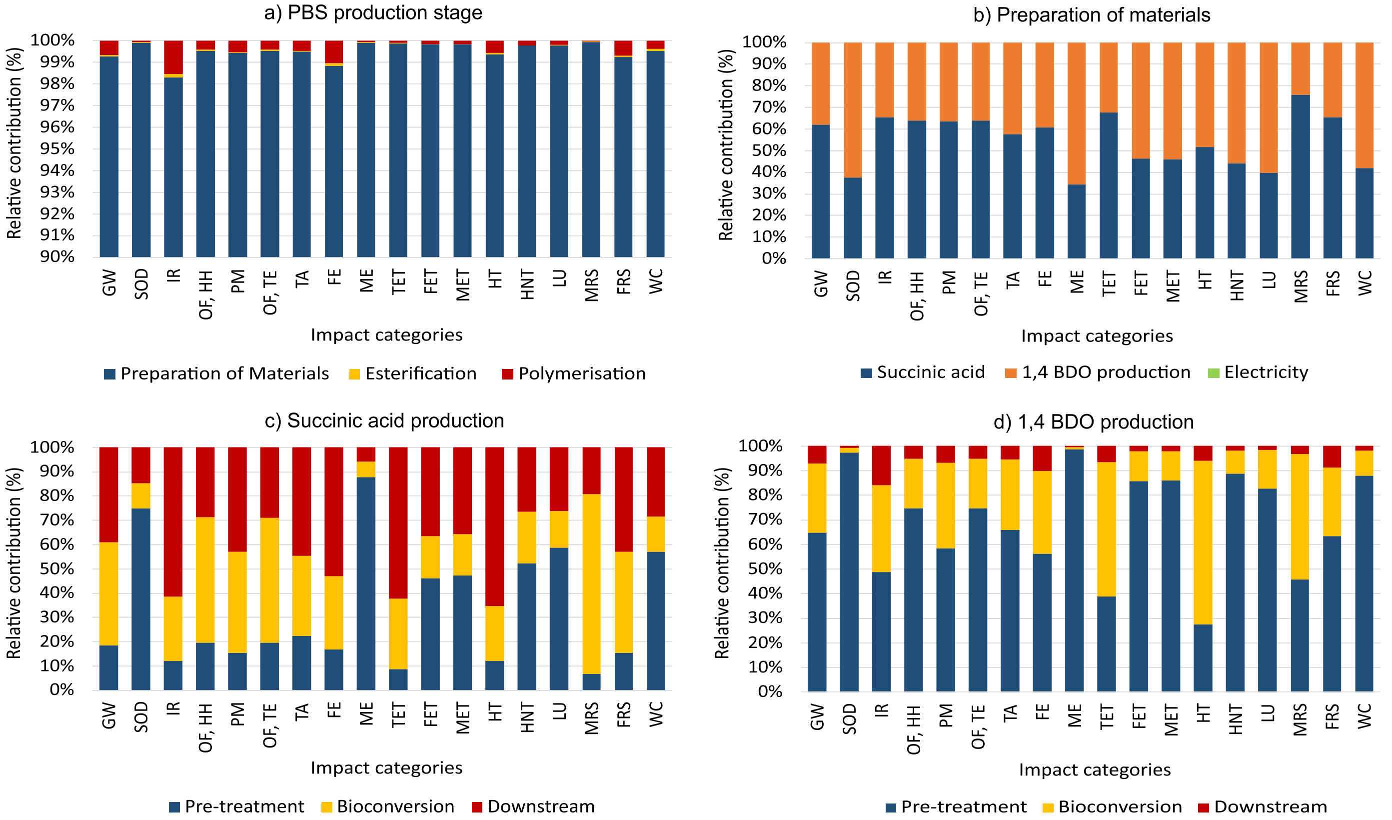


Fig. 3. Process contribution to the straw-based PBS in the baseline scenario.

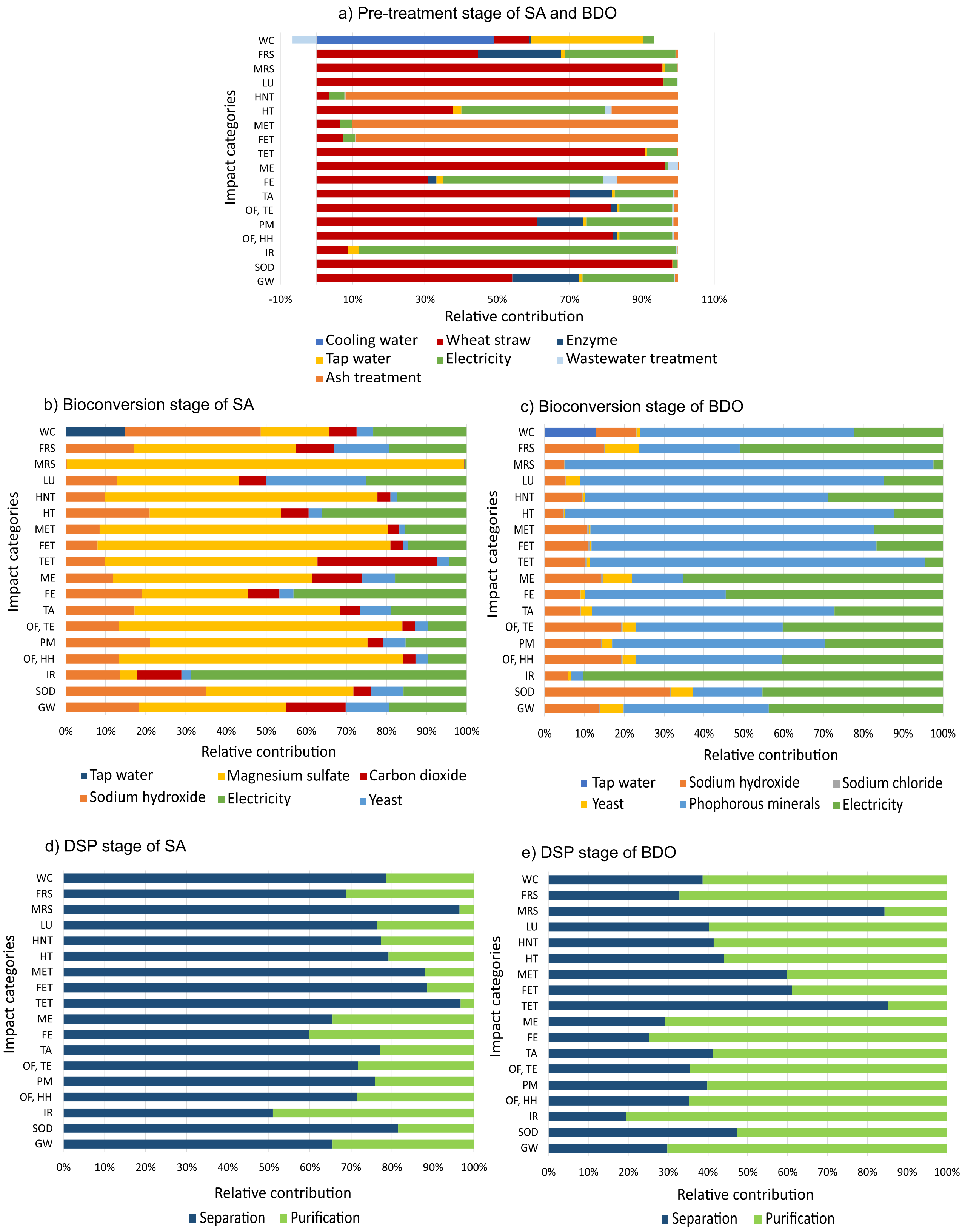


Fig. 4. Impact contributions to each category in SA and BDO stages.

were observed in the IR (68%) and FE (45%) categories.

If the straw contains a lower proportion of cellulose, the impacts grow in all categories by about 20% on average. Furthermore, if the market price of straw increased by two and four times compared to the reference value, the loads increased by about 19% and 46% on average, respectively.

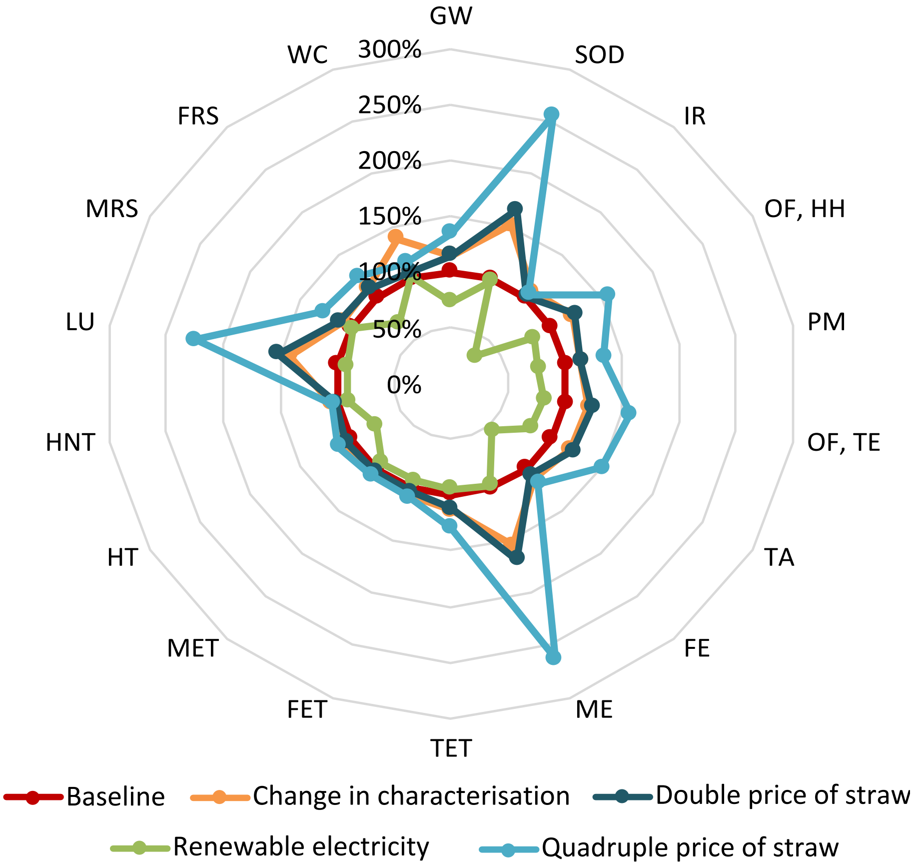


Fig. 5. Sensitivity analysis of the baseline scenario.

3.3. Comparison with other cereal-based PBS production routes

Fig. 6 shows the relative comparison of the PBS production from wheat straw (baseline) and other valorisation routes combining 1G and 2G cereal-based feedstocks. Results indicated that according to the impact category evaluated different PBS production routes represent the best or the worst option from an environmental perspective. For instance, the best scenario was the combination of wheat straw for BDO

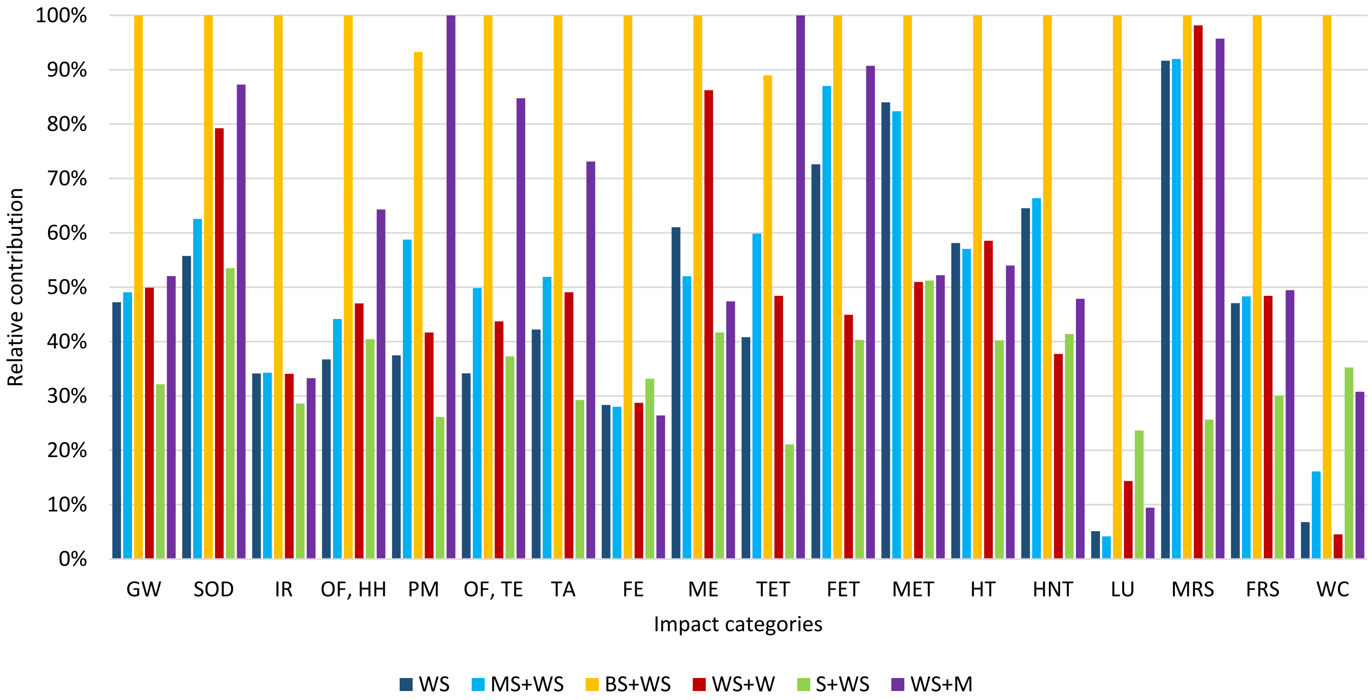


Fig. 6. Comparison of the environmental burdens of PBS in the scenarios analysed.

and sorghum for SA (i.e., S + WS), which presented the lowest burdens in 13 out of 18 impact categories analysed. The scenario BS + WS presented the highest burdens in almost all categories, thus, it corresponds to the worst alternative except for the PM and TET categories. The baseline scenario (i.e., PBS from wheat straw) is the second-best production route, as it ranks second in 12 burden indicators. MS + WS production obtained the lowest impacts in LU category, which is very close to the scenario WS (1% of difference), and WS + M has the lowest impact in the FE category, also close to the scenario WS (2% of difference). The wheat-based PBS production (i.e., WS + W) reached the lowest impact in MET, HNT and WC categories.

From Fig. 6 is also possible to observe that the greatest differences among the scenarios were observed in WC, LU, TET, PM categories. In WC, the scenarios BS + WS (the highest burdens) and WS + W (the lowest burdens) have a difference of about 95%. In the LU category, scenarios BS + WS (the highest burdens) and MS + WS (the lowest burdens) reached a value of 96%. In TET and PM, the scenario WS + M (the worst route) was 79% and 74% higher than the scenario S + WS (the best alternative), respectively.

Whether the baseline scenario is compared with the combination of wheat straw with 1G feedstocks (maize and wheat grain), the latter ones have higher burdens in 12 and 13 out of 18 categories, respectively. From them, the burden indicators that highlighted were ME (41%), SOD (42%) and LU (180%) in the WS + W, and WC (352%), PM (167%), TET (145%) and OF, TE (148%) in the WS + M. Concerning the GW category, per kg of product, the scenario BS + WS (7.27 kg CO_2eq) was three times greater than route S + WS (2.34 kg CO_2eq). Meanwhile, scenarios WS + M, WS + W, and MS + WS obtained values of 3.79, 3.63, and 3.57 kg CO_2eq, respectively. The specific values of all remaining impact categories for the six scenarios analysed are presented in the **Table SM12** in the Supplementary Materials.

As in the baseline scenario, the production of SA and BDO are the main contributors to the environmental burdens of PBS in these production routes. In the case of the best route (i.e., S + WS), SA has a GW profile of 1.46 kg CO_2eq per kg of product, and it represents about 43.0% of total GW impacts. On average, the total contribution of BDO from wheat straw was about 57.5% and sorghum-based SA was about 41.1%. This was different to the baseline scenario, where the SA accomplished (on average) the highest contribution. Consequently, if SA profiles from both feedstocks (sorghum and wheat straw) are compared, the greatest reductions with sorghum were obtained in MRS (95%), FET (96%), and ME (92%) impacts with respect to the straw. In the case of MRS, sorghum grain and ammonia production represent about 68% of the impacts. Meanwhile, sorghum grain was the main contributor in ME (82%) and electricity was the hotspot in the FET and MET categories (54–56%). On the other hand, in the straw-based SA production, the bioconversion was the main responsible of MRS impacts and the pre-treatment method was the reason in FET, MET and ME categories. The first one was due to $MgCO_3$ demand for the fermentation process; in FET and MET categories the loads were caused due to the ash disposal from the steam generation and in the ME impact because of wheat cultivation. Nevertheless, the impacts of PBS from S + WS related to FE, LU and WC categories were about 1.2, 4.6 and 5.2 times higher compared to the straw-based PBS, respectively. This is motivated due to the higher impacts caused by the succinic acid production based on sorghum.

The worst production route (i.e., BS + WS) is mainly due to the burdens attributed to the SA production from barley straw, which is responsible, on average, of about 78.7% of the burdens of the preparation of material stage (see Fig. 7a) (or 78.3% of the total profile). For instance, the GW profile of SA from barley straw is about 8.24 kg CO_2eq per kg of product. The main responsible of this performance is the pre-treatment process of barley straw in almost all categories (see Fig. 7b). Although the nitrogen gas (N_2) explosive decompression pre-treatment could seem like a green strategy because it avoids using catalysts or chemicals in the process, the environmental performance suggests another point of view. Fig. 7c shows the process contribution of this stage, the N_2 explosive decompression process is the main contributor in 13 out of 18 indicators. This occurs, mainly, due to the N_2 demand for pressuring the stream in the reactor, and the subsequent requirement of steam to obtain the desired temperature (150 °C).

The combination use of maize stover and wheat straw (scenario MS + WS) has a similar burdens distribution of SA (59.1%) and BDO (40.9%) monomers as the baseline scenario, on average. Regarding the maize stover use for SA production, the pre-treatment process stood out in 10 categories. The main contributors for them were the maize stover cultivation (in categories such as SOD, PM, OF(TE), TA, ME, TET, FET, and WC) and the ash disposal obtained from the steam generation using the residual biomass (in categories such as MET and HNT).

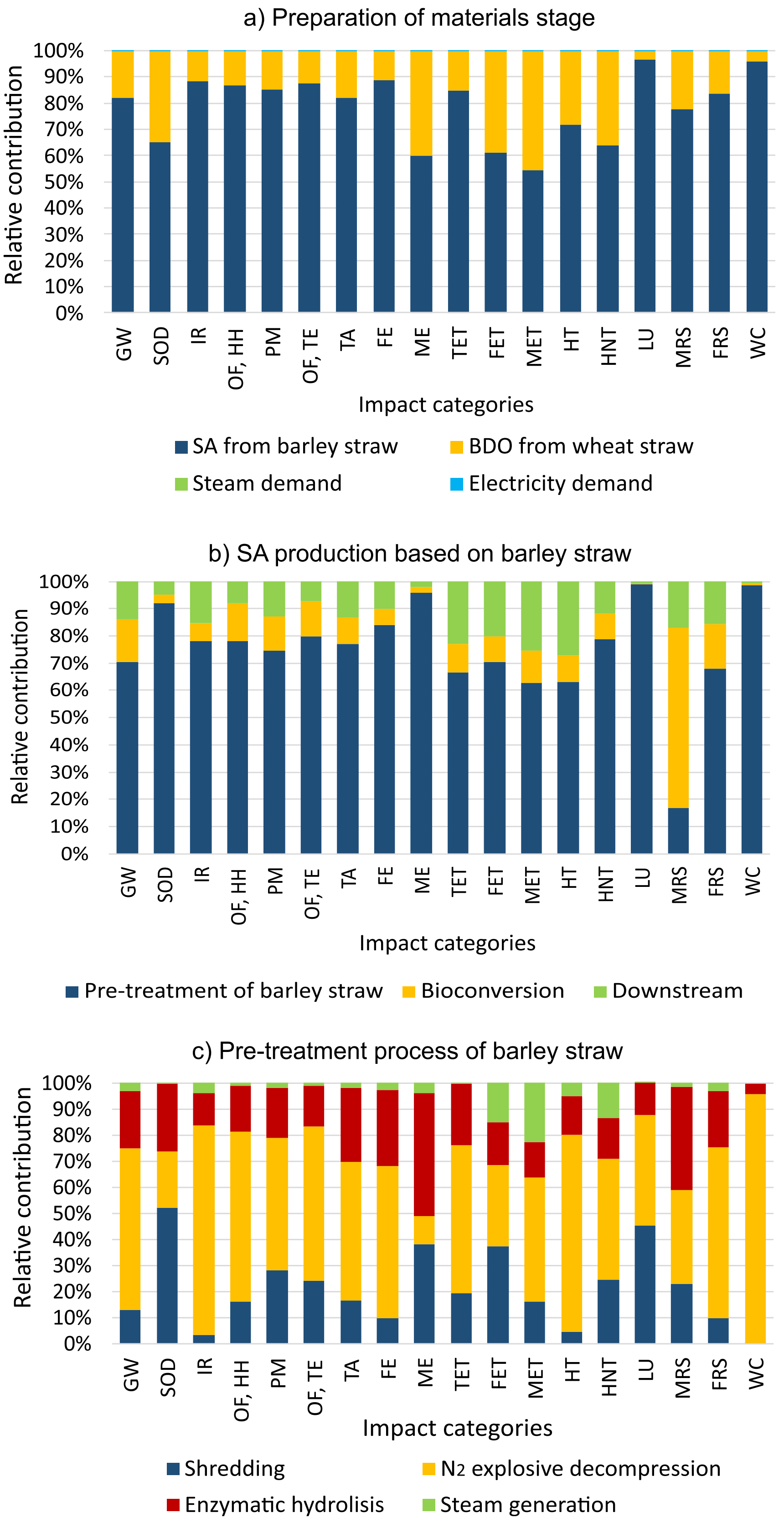
In the instance of the combination of wheat straw with 1G feedstocks to produce PBS polymer, the scenario WS + W showed a similar distribution of the burdens between SA and BDO monomers that the baseline scenario, although their difference in the WC category was higher. In this category, the production of SA represents about 63.1% of the loads, whereas in the baseline was 41.8%. This is due to the reduction of the water consumption per kg of BDO, where the wheat grain consumes about 0.08 m^3 and the straw close to 0.19 m^3. The cooling water demand for reducing the temperature of the stream after the thermal hydrolysis was the main reason behind this.

Finally, the integration of maize grain for BDO production with wheat straw (i.e., WS + M scenario) leads to higher burdens with respect to the straw-based PBS in 12 categories, where the exceptions were reduction in categories such as IR (1%), FE (2%), ME (14%), MET (32%), HT (4%), and HNT (17%). In the LU category, per kg of PBS, the route WS + M (0.49 m^2a crop eq) is almost twice the impacts of straw-based route (0.27 m^2a crop eq). The main impacts of BDO from maize grain were related to the pre-treatment method, which is mainly motivated by the burdens of the grain cultivation. Here, it is relevant to highlight that the glucose from maize grain were affected by the burdens allocation with other products generated in this process (maize oil, gluten feed and meal).

3.4. Comparison with other bio-based PBS production from literature

Different studies have been presented in the literature evaluating the environmental burdens of bio-based PBS to promote the production of bioplastics. Here, the comparison was addressed related to the GW category, since it was the indicator most analysed. Tecchio et al. [43] performed an ex-ante LCA analysis to evaluate only the Cumulative Energy Demand (CED) and greenhouse gas emissions (GHG) indicators. They estimated mean values of about 139 MJ and 6.4 kg CO_2eq at industrial scale for CED and life cycle GHG categories, respectively. They considered feedstocks such as maize starch, sugar cane and lignocellulosic with crystallisation and electrodialysis as extraction processes. The best alternative they identified was the use of sugar cane with electrodialysis extraction process. For this, they obtained a GW value of 4.17 kg CO_2eq per kg of PBS, which is higher to the value reached with the baseline scenario (3.43 kg CO_2eq). A difference with the study of Tecchio et al. [43] was that they evaluated a partly bio-based PBS, i.e., only SA monomer comes from bio-based source and the BDO was produced from fossil resources.

Patel et al. [32] also reported only the non-renewable energy use (through the CED indicator) and GHG emissions (based on Intergovernmental Panel on Climate Change - IPCC 2013, 100a method) of full bio-based and partly bio-based PBS tray (bio-SA and fossil fuel-based BDO). They examined corn as 1G raw material, and corn stover, wheat straw, miscanthus and hardwood as 2G feedstocks. Their research obtained a net value of about 3 kg CO_2eq per kg of PBS tray based on wheat straw production. Since the authors followed a cradle-to-grave approach, they assumed municipal solid waste incineration with energy recovery as an end-of-life option. Thus, environmental credits from the avoided emissions of heat and electricity generation were tacked into account in the modelling. These authors also reported a GW profile,


a) Preparation of materials stage

b) SA production based on barley straw

c) Pre-treatment process of barley straw

Fig. 7. Contribution analysis of BS + WS focused on SA from barley straw.

per kg PBS, of approximately 2.2 kg CO_2eq from corn grain.

Furthermore, Ioannidou et al. [20] assessed the life cycle environmental burdens of the bio-PBS from sugar beet pulp (SBP), corn stover (CS) and from corn glucose syrup (GS). They estimated a GW profile per kg of product of about 2.35 kg CO_2eq from SBP, 0.25 kg CO_2eq from CS and − 0.24 from GS. The negative profile reached with the glucose syrup feedstock was caused due to the authors considered the biogenic CO_2 of corn cultivation (−2.55 kg CO_2 per kg of PBS). This assumption was also considered for corn stover and sugar beet cultivation. Whether their results do not consider biogenic CO_2 from corn cultivation, the GHG emissions were 2.31 kg CO_2eq per kg of PBS. Recently, Rajendran and Han [36] evaluated the environmental burdens of PBS based on food waste. In the case of GW category, they obtained a profile per kg of biopolymer of 5.19 kg of CO_2eq. In their study, they used a 1,4 butanediol produced from corn bioethanol and corn stover succinic acid performed by Satam et al. [40].

3.5. *Fossil counterpart comparison*

To determine the environmental viability of a full bio-based PBS production, studies considered different fossil counterparts of this product. For instance, Tecchio et al. [43] considered the polyethylene terephthalate (PET) as fossil equivalent with a carbon footprint of 2.54 kg CO_2eq per kg of product at industrial scale. Whereas the GW profile of granulate PET from the Ecoinvent® v3.8 database is 2.81 kg CO_2eq. On the other hand, Ioannidou et al. [20] compared the bio-based PBS against general-purpose polystyrene, which has a value of 3.84 kg CO_2eq per kg of product according to the Ecoinvent® v3.8 database. In addition, the GW profile per kg of the fossil-derived PBS is about 6.6 kg CO_2eq [28], which is greatly higher than the full straw-based PBS (3.45 kg CO_2eq), and 2.8 times higher than the profile of PBS obtained from the scenario S + WS (the best one from all the routes analysed). In this regard, the combination of different cereal feedstocks for producing PBS can represents an interesting alternative to decrease the dependence of fossil resources for plastics production. Nevertheless, the pre-treatment method employed to obtain the fermentable sugars plays a critical role in the environmental viability of the bio-based production.

4. Conclusions

This research evaluates the environmental feasibility of different routes of biological production of PBS polymer, focusing on the use of wheat straw as the main sugar source, as well as its combination with 1G and 2G cereal-based feedstocks to obtain the SA and BDO monomers required for the polymerisation of this product. Thus, this research innovates by offering a life-cycle perspective of different alternatives to produce PBS in order to avoid exclusive dependence on a single feedstock whose supply could be limited by various factors or uses (e.g., organic fertilisation or animal feed), as well as by its seasonality. It is also of interest to identify how feedstock selection and pre-treatment strategy for biomass fractionation determine the environmental viability of bio-based production.

Although the environmental profile of wheat straw-based PBS was lower than its of fossil-based counterpart and the general-purpose polystyrene, the fact that the pre-treatment, and separation and post purification (DSP) or bioconversion stages are those processes that contributed most to the environmental impacts of the monomers production indicates that there is still room for improvement to reduce the overall environmental burdens of this valorisation route.

Regarding the combination of wheat straw with other cereal feedstocks, in general, its integration with 1G crops presented higher loads compared to 2G PBS. Combining the use of wheat straw for BDO and sorghum (a non-food crop) for SA was the best alternative in all alternatives analysed. In addition, further studies on the environmental consequences of these PBS production pathways can be conducted to identify the potential effect associated with the products displaced by these valorisation routes.

CRediT authorship contribution statement

Ricardo Rebolledo-Leiva: Conceptualization, Investigation, Methodology, Formal analysis, Software, Visualization, Writing – original draft. **Dimitrios Ladakis:** Supervision, Writing – review & editing, Validation. **Sofia-Maria Ioannidou:** Software, Formal analysis, Data curation, Writing – review & editing. **Apostolis Koutinas:** Supervision, Writing – review & editing, Validation. **Maria Teresa Moreira:** Supervision, Writing – review & editing, Visualization. **Sara González-García:** Supervision, Writing – review & editing, Visualization, Funding acquisition.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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