Environmental assessment of the production of itaconic acid from wheat straw under a biorefinery approach

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Abstract: This study performs the environmental assessment of itaconic acid (IA) production from wheat straw. The Life Cycle Assessment (LCA) methodology is used to determine the environmental hotspots, considering impact categories such as Global Warming (GW), Fossil Resource Scarcity (FRS), Water Consumption (WC), among others. A sensitivity analysis was performed considering an optimization of the steam explosion process and 100% renewable energy. The results report an impact of about 14.33 kg CO_2 eq in GW, 4.15 kg of oil eq in FRS, for each kg of IA produced for the baseline scenario. Moreover, the pretreatment and fermentation stages constitute hotspots of the IA production. In addition, using a renewable energy source in production would reduce the impact by 82% in GW, 71% in PM and 82% in FRS categories. The optimization of the steam explosion process presents a better performance in GW and FRS but also lies in an increase in WC.

Keywords: Life Cycle Assessment, Biorefinery, Industrial Biotechnology, Lignocellulosic residues, Itaconic acid.

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1. Introduction

The circular economy is not an environmental philosophy that emerged as a response to global challenges such as climate change, sustainability and biodiversity loss, but in addition to the environmental benefits, it is a driver of green growth, proving to be an excellent opportunity to strengthen growth and social welfare, through the creation of quality jobs, thanks to the innovative ecosystemic potential, while decoupling development and welfare from the consumption of natural resources and the environmental impacts associated. In short, the circular economy is based on factors such as efficiency, resilience and systems thinking, which require a metabolic approach, integrating biological and technological material cycles, to give greater importance to reuse and recycling than to consumer goods (Dyjakon, 2018; Guardia et al., 2019).

Biomass corresponds to a diverse, renewable, and recyclable resource in nature, with multiple uses like a renewable source or as a feedstock for biomaterials and biochemicals production (Moussa et al., 2016; González-García et al., 2018a). The combination of two principles: exploiting the residual biomass use and reducing the associated emissions are approached in the biorefinery concept (Cherubini, 2010). Biorefinery techniques harness the advantages of biomass to serve as a sustainable strategy for the emerging circular bioeconomy, known as the application of biomaterials in the technical and production cycles (Ubando et al., 2020; Pinales-Márquez et al., 2021). Furthermore, biorefining has potential benefits such as reducing greenhouse gas (GHG) emissions and dependence on fossil energy generation, maximum utilization of natural resources and security of food resources, and low generation of toxics (Duan et al., 2020; Yadav et al., 2020). In this regard, the valorization of biomass as a potential feedstock is receiving attention, as new policies in several countries, promoting the reuse, valorization, and sustainability of this resource. The main aim is to elaborate sustainable bio-based products such as biofuels, biochemicals, biopolymers, bioenergy, among others (Ubando et al., 2020).

There is a broad belief that bio-based products are always a better environmental alternative to fossil-based materials, however, this statement requires further analysis, e.g., considering the application of life cycle assessment (LCA) methodology, as the term "bio-based" does not always imply "environmental-friendly" (Mirabella et al., 2013). This is particularly relevant in impact categories less popular than climate change, such as eutrophication, acidification, land use, and water depletion (Moussa et al., 2016).

If the potential of lignocellulosic biomass is pursued in the biorefinery concept, the most exploited feedstocks are wheat and barley straw, coconut husk, corn stover, empty fruit bunch, rice, sugar cane bagasse, sorghum stalks, and wood residues. Notably, wheat is the most extensively cultivated crop worldwide (Le Gouis et al., 2020), playing a vital crop in food security (Erice et al., 2019). A challenge for wheat production is associated with the ineffective management of the residual biomass generated globally. (Li and Chen, 2020) estimated that 354 Mt of wheat straw is generated each year worldwide, which corresponds to the first of the world's four major agricultural residues. In this sense, European agricultural production is the largest source of wheat straw (132.59 $Mt \cdot yr^{-1}$). In this context, a closed-loop approach is required to support the implementation of a circular bioeconomy built upon bio-based resources to sustainably develop value-added products, valorizing what was previously wasted (Ingrao et al., 2021). Wheat straw represents an agricultural waste stream with high fractions of cellulose and hemicellulose (Li et al., 2018), and it has been considered as a potential feedstock for producing bioethanol (Wang et al., 2013), bioplastics (Yang et al., 2019) and biochemicals (Câmara-Salim et al., 2021).

IA has been applied in different sectors such as agricultural, pharmaceutical, and medical (Kuenz et al., 2012), being used as a co-monomer to generate thermoplastics, surfactants, polymers and polyester resins (Okabe et al., 2009). The IA production from agricultural biomass can be a viable pathway in biorefineries (Magalhães et al., 2017, 2019, 2020; Yang et al., 2020) as a renewable choice to classical monomers (acrylic and methacrylic acids) from petrochemical industry (Okabe et al., 2009). In the industry, IA is manufactured by sucrose fermentation through *Aspergillus terreus* fungus (Klement and Büchs, 2013). The biotechnological IA production is industrially established, however, significant research on improvements in fermentation and recovery processes is ongoing (Wu et al., 2017; Nieder-Heitmann et al., 2018; Magalhães et al., 2019).

This study assesses the environmental impacts of the valorization of wheat straw to produce itaconic acid under a biorefinery approach to promote a new circular bioeconomy strategy. Thus, this is the first study that analyses the life-cycle environmental impacts of bio-based IA production. Through this study, it is expected to determine those aspects that may limit the environmental viability of the production system and thus establish improvement strategies in an early stage of design. In addition, a contribution analysis of the life-cycle stages is carried out, aiming at detecting the environmental hotspots in the IA production system. In this way, improvement strategies can be proposing to reduce the overall environmental profile in future research and development activities.

2. Materials and methods

LCA is a well-known and comprehensive methodology for the assessment of the environmental impacts of a service or production system throughout all stages of its life cycle (ISO, 2006a). This tool has been widely applied to consumer products, industrial and chemical processes at various scales in recent years (Moussa et al., 2016; González-García et al., 2018a; Câmara-Salim et al., 2021). In this research, the main goal is to provide new data information relate to the life-cycle environmental impacts of the bio-based IA production process at full scale. In this way, it is offered insights to researchers, industry, and decision-makers to promote the recovery of agricultural waste, specifically wheat straw, under a joint biorefinery and circular bioeconomy approaches.

2.1. System description

To assess the environmental profile of IA based on wheat straw, this study follows the LCA methodology agreeing to the ISO 14040 and 14044 guidelines, considering a cradle-to-gate approach, i.e., the system boundary are delimited with those productive processes from wheat cultivation to IA production at factory gate (ISO, 2006a, 2006b). The functional unit (FU) corresponds to 1 kg of IA production with a 99% purity. Moreover, the wheat cultivation activities take place in Puglia, Italy, therefore, it was assumed that the biorefinery plant is located close to the feedstock supply (Sandén and Pettersson, 2014). Consequently, the wheat straw transport to the biorefinery platform was disregarded (Câmara-Salim et al., 2021). Moreover, the system boundary established in this study is presented in **Figure 1**.

Insert Figure 1

2.1.1. Wheat cultivation system

Following a monoculture regimen, durum wheat (*Triticum turgidum ssp. durum desf.*) is grown on an area of approximately 0.25 ha. The cultivation process starts with soil preparation in August and ends with harvesting in June. As the main outputs of this stage, yields of 5.5 t·ha^{-1} for wheat grain and 3.5 t·ha^{-1} for wheat straw were obtained. Traditionally, there are two uses for wheat straw: it can be left on the field as a soil amendment or sold as a low-value animal feed. The inventory data of this stage is summarized in Table 1.

Insert Table 1 here

2.1.2. IA production system design

In order to simulate the IA production system, the Superpro designer® v11 software was used (see supplementary material). A capacity of approximately 2,212 t·yr^{-1} of IA product in 330 days of production (7,920 hr·yr^{-1}) was considered for the process modelling. Moreover, an amount of 26,605 t·yr^{-1} of straw wheat (dry basis) was required.

2.1.2.1. S1: Pretreatment section

The process starts with a storage silo to feed the biorefinery plant continuously. Two milling equipment are used to homogenize the wheat straw at 20°C consisting of the following fractions, expressed as % dry weight in decreasing percentage: cellulose (30.2); hemicellulose (22.3); acid-insoluble lignin (15.3); acid-soluble lignin (1.7); acetyl groups (2.6); ash (4.7); and extractives (14.7) (Ballesteros et al., 2006). The steam explosion process is applied for a milled straw at high pressure (1.3 MPa) for 5 minutes at 198°C (Qiu and Chen, 2012) considering a biomass ratio of 2:1 (Al-Zuhair et al., 2013). A fraction of the water evaporated (25%) from the flash tank is condensed and sent to the enzymatic hydrolysis to reduce water consumption. After the steam explosion, a decanter separates the liquid phase from the solid fraction. The liquid phase goes through a reverse osmosis process to return water to the hydrolysis reactor. The solution flow is then cooled to 45°C for enzymatic hydrolysis (Al-Zuhair et al., 2013), in which 20 $FPU \cdot g^{-1}$ cellulases (dry wheat straw) and 1 $U \cdot g^{-1}$ β-glucosidase (dry wheat straw) were added at pH 5 (Chen et al., 2021).

Furthermore, a hydrolysis yield of 53% was assumed after the steam explosion (Leitner and Lindorfer, 2016). Following (Al-Zuhair et al., 2013), the bioreactor is coupled with ultrafiltration membranes to separate the diluted glucose from the additional lignocellulosic material. Finally, the flat-bottom tank collects the diluted glucose from the extractor and stores it for supply to the fermenters operating in batch mode.

2.1.2.2. S2: Inoculum and fermentation section

The fermentative production process is based upon the aerobic culture of *Aspergillus terreus* (Klement and Büchs, 2013). The process starts with medium sterilization, which is heated to 134°C and then cooled to 35°C (Magalhães et al., 2019). The sterilized broth is moved to the first bioreactor to be inoculated, and then, it is transported to a second one, which will generate the starter culture for the main fermenters. Inoculum production is considered to take 48 hours (Okabe et al., 1993; Magalhães et al., 2019). For microbial growth, the culture medium contains three main factors: glucose (carbon source), ammonia (nitrogen source), and a mixture of minerals (KH_2PO_4, NH_4NO_3, and $CaCl_2$). A working volume of 3.3 m^3 and 30.6 m^3 for the first and second reactor are used to produce 10 $g \cdot L^{-1}$ of biomass, considering 80% of useful volume. Since this is an aerobic fermentation, filters are used to sterilize the air and then its pressure is increased by 2 bar using a centrifugal compressor, and it is sent to the fermenters.

The inoculum obtained is sent to the main fermenters: three stirred-tank reactors operating in parallel. This configuration is chosen because it presents the highest concentration and productivity according to the IA production literature (Magalhães et al., 2017). These reactors operate in a fed-batch mode so that glucose is continuously fed into the fermenters. The fermentation process is performed at 35°C, and it takes 168 h to complete (Magalhães et al., 2019). After the fermentation, the broth contains around 80 $g \cdot L^{-1}$of IA and 9.4 $g \cdot L^{-1}$ of biomass. The broth produced is transferred to a buffer tank, which operates in batch mode. From this tank, the broth is sent to a disk-stack centrifuge to remove biomass, with a IA recovery yield of 95% (Magalhães et al., 2019). Even though most IA is present in the supernatant (278 kg of $IA \cdot h^{-1}$), the centrifuge sludge has a considerable quantity of IA, almost 15 kg of $IA \cdot h^{-1}$. Therefore, the sludge is further diluted with water using a custom mixer and then centrifuged in a second disc-stack centrifuge, under the same conditions as the first one. The supernatants from both centrifuges are then combined using a flow mixer and sent to the recovery section.

2.1.2.3. S3: Recovery section

At this stage, two recovery processes are applied to obtain the IA crystal. The first one starts with a triple-stage evaporator to concentrate solution to 350 $g \cdot L^{-1}$. Then, crystals are produced by cooling the solution to 15°C, considering a continuous crystallizer with a yield of 80% (Magalhães et al., 2017). The suspension obtained is filtered by a rotary vacuum filter, which retains 99% of the IA crystals (da Gama Ferreira and Petrides, 2020). However, the filtrate contains a significant quantity of uncrystallized IA (58 $kg \cdot h^{-1}$). Therefore, a second recovery process is performed again with the same conditions for the three mentioned processes (evaporation, crystallization, and filtration). Additionally, the steam produced by evaporators is condensed to obtain 1.5 t·h^{-1} of water. About 67% is recycled to a holding tank and then sent to the fermentation section, while the remaining 33% is mixed with the filter cake to produce a solution with 350 g·L^{-1} of IA crystals.

2.1.2.4. S4: Purification section

To purify the IA crystals, the solution is heated until 80°C using a stirred tank. Then, the IA solution is sent to a granular activated carbon column to remove residual organic acids and other impurities. Then, the flow is crystallized at 15°C and IA crystals are recovered using a rotary vacuum filter. Next, the crystals are sent to a rotary dryer, which eliminates the residual moisture with hot air. In this process, a final loss on drying of 1% was specified. Again, the filtrate contains a significant amount of uncrystallized IA (68 kg·h^{-1}), therefore, uncrystallized IA is recycled to the crystallizer considering a belt filtration with 95% IA recovery. The final product contains 99% of IA crystals and 1% of moisture, at a rate of 279,3 kg·h^{-1} IA crystals.

2.1.2.5. Waste management

All sections of IA production (i.e. pretreatment, fermentation, recovery and purification) have a waste stream (see supplementary materials). The residues from sections S1, S2, and S3 are sent to the anaerobic digestion (AD) process to produce biogas, which will feed a gas turbine-generator and steam generator to produce power and heat (Achinas et al., 2019). Meanwhile, the effluent obtained from the purification section is transferred to a wastewater treatment plant (see **Figure 1**). The methane generated from the AD process is burned in a gas turbine for energy generation, considering efficiency of 28% (Bove and Lunghi, 2006). Nearly 85% of the digestate is re-distributed to the AD process, and the remaining fraction 15% is transferred to a filtration process to separate the liquid from the solid waste. The liquid waste is dispatched to a wastewater plant to be treated, while the solid waste, which is an organic stream is assumed to be left on the field as a soil amendment. In this sense, it is considered that a fraction of the carbon content of the biomass (16%) will be stored in the land as soil organic carbon in a long term (Fang et al., 2019), with a carbon content of 44% of its dry matter (González-García et al., 2021). Thus, the growth in soil carbon content was considered as an environmental credit in the results. The remaining 84% should be emitted as CO_2, however, it is assumed to be equivalent to the carbon sequestration since both flows occur in the same year (González-García et al., 2021). Finally, the electricity generated (4,637 MWh) is used in recovery and waste management sections, while the high-pressure steam (21,188 t) is sent to the pretreatment section, specifically to the steam explosion process.

2.2. Inventory data

Wheat straw inventory data (see **Table 1**) were collected through interviews with Italian farmers located in Puglia (Southern Italy). In this crop cultivation stage, an economic allocation was followed to determine the environmental impacts of wheat grain and straw. For the IA production, inventory data were obtained through mass and energy balance modeled in Superpro designer® v11 software (see **Table 2**). Moreover, the Ecoinvent® v3.6 database (Wernet et al., 2016) was used to model the background processes (see supplementary data).

Insert Table 2 here

2.3. Life cycle impact assessment

In order to assess the environmental burdens of IA production, the characterization factors of the ReCiPe midpoint 1.04v hierarchist method global (Huijbregts et al., 2017) were considered, and the software SimaPro 9.1 was used. The environmental impact categories analyzed for the IA production correspond to Global Warming - GW (kg CO_2 eq); Particulate Matter – PM (kg PM2.5 eq); Freshwater Eutrophication – FE (kg P eq); Marine Eutrophication – ME (kg N eq); Human Carcinogenic Toxicity – HT (kg 1,4-DCB); Land Use – LU (m^2a crop eq); Fossil Resource Scarcity – FRS (kg oil eq); and Water Consumption – WC (m^3).

2.4. Sensitivity analysis

Considering the baseline scenario of IA production described in section 2.1.2, a sensitivity analysis was performed to evaluate how changes in the manufacturing processes may alter the environmental profile of IA product. In this way, two assumptions were established: *i*) Improvement of the steam production process (SE1) and *ii*) Electricity mix based on renewable energy sources (CE2).

2.4.1. Improvement consumption in steam explosion process

Scientific literature has demonstrated that a critical point of the biorefinery processes is the pretreatment stage of lignocellulosic resources for their transformation into high-added-value products (Bello et al., 2018; Câmara-Salim et al., 2021; Gonzalez-Garcia et al., 2018; González-García et al., 2018b). In this sense, steam production is one of the main hotspots. Therefore, an optimization of the steam explosion procedure is performed, where the produced steam fraction is recirculated to the process and then high-pressure saturated water is added to reduce the required quantity of steam. For this, a gas compressor and a heating process were considered to reach the required conditions of 1.3 MPa and 198°C (see supplementary materials). The tap water consumption (see supplementary materials) is then considered in the inventory for the environmental assessment.

2.4.2. Change in the electricity mix

An average of the European electricity mix is considered from the Ecoinvent® v3.6 database for the baseline scenario (see supplementary materials). This inventory process considers different energy sources, such as thermal, nuclear, natural gas, among others. Regarding the energy sources contribution in 2019, according to data from (EUROSTAT, 2021), conventional thermal electricity production decreased and accounted for 42.8 % of total production, meanwhile, nuclear energy source accounted for 26.7%. Wind energy increased by 14.9%, reaching 13.3%; hydro accounted for 12.3%, and solar energy increased by 4.1%, accounting for 4.4% of the overall production. Thus, increasing the energy generation based on renewable sources represent a crucial strategy to reduce GHG emissions and take on the Paris Agreement on Climate Change 2015 and the European Union's goal by 2030 for emissions reduction by leastwise 40% below the levels of 1990 (European Parliament, 2018). Therefore, in this scenario, IA production considers the generation of electricity only from clean-renewable sources. For this purpose, the Ecoinvent® v3.6 database was utilized for the inventory of the production and transmission of renewable energy, considering the three most important sources in Europe distributed as follows: 40% wind (onshore wind farms), 30% hydro, and 30% solar. In addition, the optimization in the steam explosion process of the SE1 scenario is also considered. Finally, since the high energy demand in enzyme production, this process was also supplied with the same share of renewable energy.

3. Results and discussion

3.1. Environmental analysis

The environmental results of IA production according to the three scenarios evaluated are presented in **Table 3**. Regarding the baseline scenario, the results indicated that the inoculum and fermentation section and pretreatment section are the life-cycle stages that most contribute to the environmental impacts of bio-based IA production (see **Figure 2**). The fermentation section is the main contributor in five of the eight categories: GW, PM, FE, HT, and FRS. Meanwhile, the pretreatment stage is the major contributor in ME, LU, and WC. The factor contribution in each stage of IA production is presented in **Figure 3**. Furthermore, the main results regarding every impact category are described next.

Insert Table 3 here

Insert Figure 2 here

Insert Figure 3 here

3.1.1. Global Warming

GW measures the integrated infrared radiative forcing increase of greenhouse gas (GHG) expressed in kg CO_2 eq (Huijbregts et al., 2017). In this impact category, the inoculum and fermentation section is the main contributor with 40% of the total impacts, followed by the pretreatment section with 35% (see **Figure 2**). In the inoculum and fermentation stage, the electricity demand is the main contributor to GHG emissions with 86% of the total **(Figure 3b**). The steam explosion process represents the largest contribution with 66% of impacts in the pretreatment stage, due to steam production (**Figure 3a**). Wheat straw production presents a less contribution of 11% of the pretreatment impacts. Wheat environmental impacts are due to the production (background process) and application (field emission) of fertilizer, mainly. The GHG emissions of IA production are distributed as follows: 85%, for CO_2, 9% for N_2O, and 5% for CH_4. Regarding the CH_4 emissions, these happen mainly in the fermentation stage, due to electricity production.

3.1.2. Particulate matter formation

As in the GW, in the PM category, the inoculum and fermentation section is the main responsible for this impact category with about 42% of the total (**Figure 2**), due to electricity production (**Figure 3b**). The pretreatment process appears as the second main contributor with 26%, due to steam production (**Figure 3a**). The release of sulfur dioxide (SO_2) corresponds to 52% of the total PM formation into the air. Regarding $PM_{2.5}$ particles and nitrogen oxides (NO_x) emissions, these are responsible for 28.8% and 13.6% of the total PM impacts, respectively.

3.1.3. Freshwater and marine eutrophication

Regarding the FE category, 55% of the impacts are caused by the inoculum and fermentation section, followed by the recovery section with 22% (see **Figure 2**). In addition, these impacts come from phosphate in water corresponding to about 98%. Concerning the ME impact category, 67% of the environmental impacts depend on the pretreatment stage due to the milling process, i.e., wheat straw production (**Figure 3a**). The contribution of milling depends on the production of wheat straw, mainly due to the application of fertilizers. In addition, the environmental impacts come from the nitrates released into water bodies, which correspond to about 94%.

3.1.4. Human carcinogenic toxicity

In IA production, the “inoculum and fermentation” and recovery stages are principally accountable for 54% and 21% of the total impacts, respectively (**Figure 2**). Both are due to electricity production (**Figures 3b and 3c**). In addition, chromium VI in water causes 92% of the impacts, due to electricity production from fossil sources.

3.1.5. Land use

The pretreatment section represents the main contribution with 54% of the impacts related to the LU category (**Figure 2**), due to potato starch needed for the enzyme production (**Figure 3a**). Moreover, the milling process appears as the second main contributor with 24% of the overall impacts on land occupation, due to wheat cultivation.

3.1.6. Fossil resource scarcity

The pretreatment and "inoculum and fermentation" stages are the hotspots of the system with approximately 38% each one (**Figure 2**). This occurs because of the heat production needed for the steam explosion process (**Figure 3a**), and electricity production for fermentation (**Figure 3b**). Moreover, natural gas consumption contributes 41.6% of the overall impacts, followed by hard coal with 26.9% and coal brown with 16.9%.

3.1.7. Water consumption

The pretreatment stage is responsible for the highest water consumption for IA production (**Figure 2**), due to enzymatic hydrolysis (**Figure 3a**). In this sense, tap water consumed for the hydrolysis process and for enzyme production (i.e., irrigation process for potato starch) stands out. In addition, the negative contribution in this category represents the water returned to the Technosphere through wastewater treatment.

3.2. Sensitivity analysis

Figure 4 shows the results of the sensitivity analysis considering a) the base scenario, which follow the production process described in section 2.1.2; b) the optimization in the steam explosion process that recovery the steam and added the complementary saturated hot water (SE1), and c) the change in the electricity mix with 100% of renewable energy sources and the steam process optimization (CE2). In this sense, a significant reduction in most impact categories is observed when electricity production is based on renewable sources (scenario CE2). However, this is different for the water consumption category, where both sensitive scenarios are higher than the base case. In scenario SE1, the processes necessary to obtain steam at the required conditions are considered in the simulation, therefore, tap water is the input to the process to meet the required amount after the recovered steam fraction. Otherwise, the scenario CE2 considers hydropower with a contribution of 30% in electricity production. Hence, this renewable production has a water consumption 33% higher than the average European electricity production process of the Ecoinvent® database. Consequently, a rise in water consumption for IA production is obtained.

As for the SE1 scenario, although it presents better values for GW and FRS, it also represents a worse environmental profile in FE and HT categories. While the differences with respect to the baseline scenario in the PM, ME, and LU categories are marginal, with 1%, 3%, and 3%, respectively. As for the FE category, this increase is motivated by an increase in the contribution of the pretreatment stage from 14% to 26% in the environmental profile, due to the increase in water consumption explained above. With respect to the HT category, the pretreatment stage also increases almost twice its contribution (13% to 24%) due to the water consumption in the steam explosion. Otherwise, it is important to point out that the decrease in steam production in the SE1 scenario represents a reduction of 11% and 16% in the GW and FRS categories, respectively. Regarding the CE2 scenario, the purification stage appears as the main contributor in categories such as GW (45%), PM (55%), FE (59%), HT (74%), and FRS (53%). This occurs due to the production of sodium hydroxide, but more importantly, due to the significant reduction in the contribution of the remaining processes in the system. For example, the fermentation process has an average contribution of 40% in all categories in the baseline scenario, but reaches 19% in the CE2 scenario, while purification has an average contribution of 2% in the baseline scenario and reaches 38% in the CE2 scenario. Otherwise, the pretreatment stage is the most contributor in ME (76%), LU (70%), and WC (46%). Overall, the results show that the energy sources that make up the electricity matrix have a significant influence, both positive and negative, on the environmental performance of IA production.

Insert Figure 4 here

3.3. Limitations of the study and future research

In this study, the environmental profile of the IA production considering a baseline scenario and two improvement strategies were carried out. As far as is known, there is no other LCA study related to IA production through either a chemical or a bio-based pathway. As consequence, this represents a limitation for a deeper analysis, since it is not possible to compare the results obtained in this study with other IA environmental profiles proposed in the scientific literature.

As future research lines, the first case would be evaluating the chemical production of IA to compare and determine whether this bio-based production, represents an improvement from an environmental perspective, motivating the promotion of the wheat straw valorization into IA product, through a joint approach of biorefinery and circular bioeconomy. Moreover, since lignocellulosic resources can be acquired from different biomasses, there are numerous possibilities to use them as feedstocks for IA production. Thus, it may be possible to determine which feedstock obtains the best environmental performance and production yield. Furthermore, to analyze the environmental implication of different downstream processes in the IA production would be another interesting research, following for example those scenarios proposed by (Magalhães et al., 2019). Otherwise, since the pretreatment stage is an important contributor to the environmental profile of IA production, it would also be interesting to analyze alternative techniques that aim to obtain a better environmental performance. These techniques could contribute to reduce the consumption of inputs (energy, steam, enzymes, and water). Attention could be paid into the introduction of genetic engineering, which could allow the utilization of cheaper alternative substrates, the increase of IA yield and the reduction of the production cost (Gopaliya et al., 2021).

Finally, other dimensions of sustainability can be studied to realize the triple bottom line of IA production. In a previous study by (Magalhães et al., 2019), the authors evaluated the techno-economic characteristic of the downstream processes of IA production through the comparison of adsorption, reactive extraction, and electrodialysis, to assess the most advantageous process considering the production costs. In this sense, a life cycle cost (LCC) of IA production can be attractive to determine the hotspots of this economic dimension. Finally, social analysis is also recommended to obtain the full sustainability performance of a bio-based IA production.

4. Conclusions

This study evaluates the environmental impacts of bio-based IA production from wheat straw valorization through the LCA methodology. Results indicate an impact of about 2.64 and 14.33 kg CO_2 eq in GW, 0.76 and 4.15 kg of oil eq in FRS, per 1 kg of IA produced for CE2 and base scenarios, respectively. Moreover, pretreatment and fermentation are the most impactful stages in baseline and SE1 scenarios, due to steam and electricity production.

Furthermore, considering an electricity production based on fully renewable energy a decrease of 82% in GW and FRS and, 71% in PM is obtained.

E-supplementary data can be found in the e-version online.

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Data Availability

The authors confirm that the data supporting the findings of this study are available within the article [and/or] its supplementary materials.

References

1. Achinas, S., Martherus, D., Krooneman, J., Euverink, G.J.W., 2019. Preliminary assessment of a biogas-based power plant from organic waste in the North Netherlands. Energies 12. https://doi.org/10.3390/en12214034
2. Al-Zuhair, S., Al-Hosany, M., Zooba, Y., Al-Hammadi, A., Al-Kaabi, S., 2013. Development of a membrane bioreactor for enzymatic hydrolysis of cellulose. Renew. Energy 56, 85–89. https://doi.org/10.1016/j.renene.2012.09.044
3. Ballesteros, I., Negro, M.J., Oliva, J.M., Cabañas, A., Manzanares, P., Ballesteros, M., 2006. Ethanol production from steam-explosion pretreated wheat straw. Appl.

Biochem. Biotechnol. 130, 496–508. https://doi.org/10.1385/ABAB:130:1:496

4. Bello, S., Ríos, C., Feijoo, G., Moreira, M.T., 2018. Comparative evaluation of lignocellulosic biorefinery scenarios under a life-cycle assessment approach. Biofuels, Bioprod. Biorefining 12, 1047–1064. https://doi.org/10.1002/bbb.1921
5. Bove, R., Lunghi, P., 2006. Electric power generation from landfill gas using traditional and innovative technologies. Energy Convers. Manag. 47, 1391–1401. https://doi.org/10.1016/j.enconman.2005.08.017
6. Câmara-Salim, I., González-García, S., Feijoo, G., Moreira, M.T., 2021. Screening the environmental sustainability of microbial production of butyric acid produced from lignocellulosic waste streams. Ind. Crops Prod. 162. https://doi.org/10.1016/j.indcrop.2021.113280
7. Chen, J., Wang, X., Zhang, B., Yang, Y., Song, Y., Zhang, F., Liu, B., Zhou, Y., Yi, Y., Shan, Y., Lü, X., 2021. Integrating enzymatic hydrolysis into subcritical water pretreatment optimization for bioethanol production from wheat straw. Sci. Total Environ. 770, 145321. https://doi.org/10.1016/j.scitotenv.2021.145321
8. Cherubini, F., 2010. The biorefinery concept: Using biomass instead of oil for producing energy and chemicals. Energy Convers. Manag. 51, 1412–1421. https://doi.org/10.1016/j.enconman.2010.01.015
9. da Gama Ferreira, R., Petrides, D., 2020. Production of Biobased Itaconic Acid Modeling and Evaluation with SuperPro Designer®. Intelligen.
10. Duan, Y., Pandey, A., Zhang, Z., Awasthi, M.K., Bhatia, S.K., Taherzadeh, M.J., 2020. Organic solid waste biorefinery: Sustainable strategy for emerging circular bioeconomy in China. Ind. Crops Prod. 153, 112568.

https://doi.org/10.1016/j.indcrop.2020.112568

11. Dyjakon, A., 2018. Harvesting and baling of pruned biomass in apple orchards for energy production. Energies 11. https://doi.org/10.3390/en11071680
12. Erice, G., Sanz-Sáez, Á., González-Torralba, J., Méndez-Espinoza, A.M., Urretavizcaya, I., Nieto, M.T., Serret, M.D., Araus, J.L., Irigoyen, J.J., Aranjuelo, I., 2019. Impact of elevated CO2 and drought on yield and quality traits of a historical (Blanqueta) and a modern (Sula) durum wheat. J. Cereal Sci. 87, 194–201. https://doi.org/10.1016/j.jcs.2019.03.012
13. European Parliament, 2018. Directive (EU) 2018/2001 of the European Parliament and of the Council on the promotion of the use of energy from renewable sources. Off. J. Eur. Union 2018, 82–209.
14. EUROSTAT, 2021. Electricity generation statistics – first results [WWW Document]. URL https://ec.europa.eu/eurostat/statistics-explained/index.php?title=Electricity_generation_statistics_–_first_results#Electricity_supplied_to_the_market (accessed 6.29.21).
15. Fang, Y., Singh, B.P., Cowie, A., Wang, W., Arachchi, M.H., Wang, H., Tavakkoli, E., 2019. Balancing nutrient stoichiometry facilitates the fate of wheat residue-carbon in physically defined soil organic matter fractions. Geoderma 354, 113883. https://doi.org/10.1016/j.geoderma.2019.113883
16. González-García, S., Almeida, F., Moreira, M.T., Brandão, M., 2021. Evaluating the environmental profiles of winter wheat rotation systems under different management strategies. Sci. Total Environ. 770. https://doi.org/10.1016/j.scitotenv.2021.145270
17. González-García, S., Argiz, L., Míguez, P., Gullón, B., 2018a. Exploring the

production of bio-succinic acid from apple pomace using an environmental approach. Chem. Eng. J. 350, 982–991. https://doi.org/10.1016/j.cej.2018.06.052

18. Gonzalez-Garcia, S., Gullón, B., Moreira, M.T., 2018. Environmental assessment of biorefinery processes for the valorization of lignocellulosic wastes into oligosaccharides. J. Clean. Prod. 172, 4066–4073. https://doi.org/10.1016/j.jclepro.2017.02.164
19. González-García, S., Morales, P.C., Gullón, B., 2018b. Estimating the environmental impacts of a brewery waste–based biorefinery: Bio-ethanol and xylooligosaccharides joint production case study. Ind. Crops Prod. 123, 331–340. https://doi.org/10.1016/j.indcrop.2018.07.003
20. Gopaliya, D., Kumar, V., Khare, S.K., 2021. Recent advances in itaconic acid production from microbial cell factories. Biocatal. Agric. Biotechnol. 36. https://doi.org/10.1016/j.bcab.2021.102130
21. Guardia, L., Suárez, L., Querejeta, N., Rodríguez Madrera, R., Suárez, B., Centeno, T.A., 2019. Apple Waste: A Sustainable Source of Carbon Materials and Valuable Compounds. ACS Sustain. Chem. Eng. 7, 17335–17343. https://doi.org/10.1021/acssuschemeng.9b04266
22. Huijbregts, M.A.J., Steinmann, Z.J.N., Elshout, P.M.F., Stam, G., Verones, F., Vieira, M., Zijp, M., Hollander, A., van Zelm, R., 2017. ReCiPe2016: a harmonised life cycle impact assessment method at midpoint and endpoint level. Int. J. Life Cycle Assess. 22, 138–147. https://doi.org/10.1007/s11367-016-1246-y
23. Ingrao, C., Matarazzo, A., Gorjian, S., Adamczyk, J., Failla, S., Primerano, P., Huisingh, D., 2021. Wheat-straw derived bioethanol production: A review of Life

Cycle Assessments. Sci. Total Environ. 781, 146751. https://doi.org/10.1016/j.scitotenv.2021.146751

24. ISO, 2006a. ISO 14040 - Environmental management - Life cycle assessment - Principles and framework. International Organization Standardization.
25. ISO, 2006b. ISO 14044 - Environmental management - Life cycle assessment - Requirements and guidelines. https://doi.org/10.1007/s11367-011-0297-3
26. Klement, T., Büchs, J., 2013. Itaconic acid - A biotechnological process in change. Bioresour. Technol. 135, 422–431. https://doi.org/10.1016/j.biortech.2012.11.141
27. Kuenz, A., Gallenmüller, Y., Willke, T., Vorlop, K.D., 2012. Microbial production of itaconic acid: Developing a stable platform for high product concentrations. Appl. Microbiol. Biotechnol. 96, 1209–1216. https://doi.org/10.1007/s00253-012-4221-y
28. Le Gouis, J., Oury, F.X., Charmet, G., 2020. How changes in climate and agricultural practices influenced wheat production in Western Europe. J. Cereal Sci. 93, 102960. https://doi.org/10.1016/j.jcs.2020.102960
29. Leitner, V., Lindorfer, J., 2016. Evaluation of technology structure based on energy yield from wheat straw for combined bioethanol and biomethane facility. Renew. Energy 87, 193–202. https://doi.org/10.1016/j.renene.2015.09.037
30. Li, S., Chen, G., 2020. Agricultural waste-derived superabsorbent hydrogels: Preparation, performance, and socioeconomic impacts. J. Clean. Prod. 251, 119669. https://doi.org/10.1016/j.jclepro.2019.119669
31. Li, S., Chen, G., Anandhi, A., 2018. Applications of emerging bioelectrochemical technologies in agricultural systems: A current review. Energies 11, 1–21. https://doi.org/10.3390/en11112951

32. Magalhães, A.I., de Carvalho, J.C., Medina, J.D.C., Soccol, C.R., 2017. Downstream process development in biotechnological itaconic acid manufacturing. Appl. Microbiol. Biotechnol. 101, 1–12. https://doi.org/10.1007/s00253-016-7972-z

33. Magalhães, A.I., de Carvalho, J.C., Thoms, J.F., Medina, J.D.C., Soccol, C.R., 2019. Techno-economic analysis of downstream processes in itaconic acid production from fermentation broth. J. Clean. Prod. 206, 336–348. https://doi.org/10.1016/j.jclepro.2018.09.204

34. Magalhães, A.I., de Carvalho, J.C., Thoms, J.F., Souza Silva, R., Soccol, C.R., 2020. Second-generation itaconic acid: An alternative product for biorefineries? Bioresour. Technol. 308, 123319. https://doi.org/10.1016/j.biortech.2020.123319

35. Mirabella, N., Castellani, V., Sala, S., 2013. Life cycle assessment of bio-based products: A disposable diaper case study. Int. J. Life Cycle Assess. 18, 1036–1047. https://doi.org/10.1007/s11367-013-0556-6

36. Moussa, H.I., Elkamel, A., Young, S.B., 2016. Assessing energy performance of bio-based succinic acid production using LCA. J. Clean. Prod. 139, 761–769. https://doi.org/10.1016/j.jclepro.2016.08.104

37. Nieder-Heitmann, M., Haigh, K.F., Görgens, J.F., 2018. Process design and economic analysis of a biorefinery co-producing itaconic acid and electricity from sugarcane bagasse and trash lignocelluloses. Bioresour. Technol. 262, 159–168. https://doi.org/10.1016/j.biortech.2018.04.075

38. Okabe, M., Lies, D., Kanamasa, S., Park, E.Y., 2009. Biotechnological production of itaconic acid and its biosynthesis in Aspergillus terreus. Appl. Microbiol. Biotechnol. 84, 597–606. https://doi.org/10.1007/s00253-009-2132-3

39. Okabe, M., Ohta, N., Park, Y.S., 1993. Itaconic acid production in an air-lift bioreactor using a modified draft tube. J. Ferment. Bioeng. 76, 117–122. https://doi.org/10.1016/0922-338X(93)90067-I

40. Pinales-Márquez, C.D., Rodríguez-Jasso, R.M., Araújo, R.G., Loredo-Treviño, A., Nabarlatz, D., Gullón, B., Ruiz, H.A., 2021. Circular bioeconomy and integrated biorefinery in the production of xylooligosaccharides from lignocellulosic biomass: A review. Ind. Crops Prod. 162. https://doi.org/10.1016/j.indcrop.2021.113274

41. Qiu, W., Chen, H., 2012. Enhanced the enzymatic hydrolysis efficiency of wheat straw after combined steam explosion and laccase pretreatment. Bioresour. Technol. 118, 8–12. https://doi.org/10.1016/j.biortech.2012.05.033

42. Sandén, B., Pettersson, K., 2014. Systems Perspectives on Biorefineries, Applied Physics Letters.

43. Talebnia, F., Karakashev, D., Angelidaki, I., 2010. Production of bioethanol from wheat straw: An overview on pretreatment, hydrolysis and fermentation. Bioresour. Technol. 101, 4744–4753. https://doi.org/10.1016/j.biortech.2009.11.080

44. Ubando, A.T., Felix, C.B., Chen, W.H., 2020. Biorefineries in circular bioeconomy: A comprehensive review. Bioresour. Technol. 299. https://doi.org/10.1016/j.biortech.2019.122585

45. Wang, L., Littlewood, J., Murphy, R.J., 2013. Environmental sustainability of bioethanol production from wheat straw in the UK. Renew. Sustain. Energy Rev. 28, 715–725. https://doi.org/10.1016/j.rser.2013.08.031

46. Wernet, G., Bauer, C., Steubing, B., Reinhard, J., Moreno-Ruiz, E., Weidema, B., 2016. The ecoinvent database version 3 (part I): overview and methodology. Int. J. Life

Cycle Assess. 21, 1218–1230. https://doi.org/10.1007/s11367-016-1087-8

47. Wu, X., Liu, Q., Deng, Y., Li, J., Chen, X., Gu, Y., Lv, X., Zheng, Z., Jiang, S., Li, X., 2017. Production of itaconic acid by biotransformation of wheat bran hydrolysate with Aspergillus terreus CICC40205 mutant. Bioresour. Technol. 241, 25–34. https://doi.org/10.1016/j.biortech.2017.05.080

48. Yadav, B., Chavan, S., Atmakuri, A., Tyagi, R.D., Drogui, P., 2020. A review on recovery of proteins from industrial wastewaters with special emphasis on PHA production process: Sustainable circular bioeconomy process development. Bioresour. Technol. 317, 124006. https://doi.org/10.1016/j.biortech.2020.124006

49. Yang, J., Ching, Y.C., Chuah, C.H., 2019. Applications of Lignocellulosic Fibers and Lignin in. Polymers (Basel). 11, 1–26.

50. Yang, J., Xu, H., Jiang, J., Zhang, N., Xie, J., Zhao, J., Bu, Q., Wei, M., 2020. Itaconic acid production from undetoxified enzymatic hydrolysate of bamboo residues using Aspergillus terreus. Bioresour. Technol. 307, 123208. https://doi.org/10.1016/j.biortech.2020.123208

Table 1. Inventory data for agricultural process and remarkable information associated with wheat straw cultivation

Key parameters for wheat straw production	Value	Unit
Wheat grain yield	5.5	t·ha^{-1}
Wheat straw yield	3.5	t·ha^{-1}
Wheat grain price	0.29	€·kg^{-1}
Wheat straw price	0.07	€·kg^{-1}
Allocation factor grain	85.5	%
Allocation factor straw	14.5	%
Grain moisture content	13	%
Straw moisture content	10	%
Life Cycle Inventory data	Value	Unit
Nitrogen application, as N	104.4	kg·ha^{-1}
Phosphorus application, as P	40.2	kg·ha^{-1}
Pesticides application (active ingredient)	2.01	kg·ha^{-1}
Seed	180	kg·ha^{-1}
Diesel	42.3	l·ha^{-1}
Field emissions		
N_2O	1.64	kg·ha^{-1}
NO_2	4.18	kg·ha^{-1}
NH_3	2.96	kg·ha^{-1}
NO_3^- *leaching*	114.64	kg·ha^{-1}
P-PO_4^{-3} *leaching*	70	g·ha^{-1}
P-PO_4^{-3} *runoff*	220	g·ha^{-1}

Table 2. Global Life Cycle Inventory data for IA production

Inputs from Technosphere		Inputs from Nature		Outputs to Technosphere	
Pretreatment section					
Straw wheat (t)	26,605				
Water (t)	62,903				
Steam (t)	27,496				
Enzyme (t)	118				
Electricity (MWh)	44,084				
Fermentation section					
NH_4NO_3 (t)	89	Air (t)	234,383	CO_2 biogenic (t)	781
$CaCl_2$ (t)	148				
KH_2PO_4 (t)	3				
NH_3 (t)	33				
Glucose (t)	668				
Electricity (MWh)	29,515				
Recovery section					
Water (t)	158				
Electricity (MWh)	16,385				
Purification section					
Water (t)	7,338	Air (t)	2,259		
NaOH (t)	2,928				
Electricity (MWh)	115				
Waste management					
Biomass residues (t)	745,433	Air (t)	375,705	Organic amendment (t)	23,995
Water (t)	69,814			Effluent water (m^3)	167,807
Electricity (MWh)	2,230				
IA product (t)	2,203				

Table 3. Comprehensive comparative results among scenarios evaluated

	Process	Baseline scenario	Scenario SE1	Scenario CE2
Scenarios characteristics	Steam explosion process (SE)	A fraction of steam obtained from AD process, the remaining based on Ecoinvent database	Water recovery	Water recovery
		Water recovered from SE process sent to enzymatic hydrolysis		
	Electricity production	Electricity mix based on Ecoinvent database (fossil + renewable energy)	As in baseline scenario	Full renewable energy sources (wind, solar and hydro)
	Enzyme production for enzymatic hydrolysis	Based on Ecoinvent database (fossil + renewable energy)	As in baseline scenario	Based on renewable energy sources

Impact category	Unit	Base Scenario	Scenario SE1	Scenario CE2
GW	kg CO_2 eq	14.33	12.72	2.64
PM	g PM2.5 eq	21.64	21.83	6.22
FE	g P eq	10.10	11.66	1.27
ME	g N eq	4.36	4.47	3.74
HT	kg 1,4-DCB	0.56	0.63	0.07
LU	m^{2}a crop eq	1.05	1.09	0.73
FRS	kg oil eq	4.15	3.47	0.76
WC	m^3	0.29	0.33	0.40

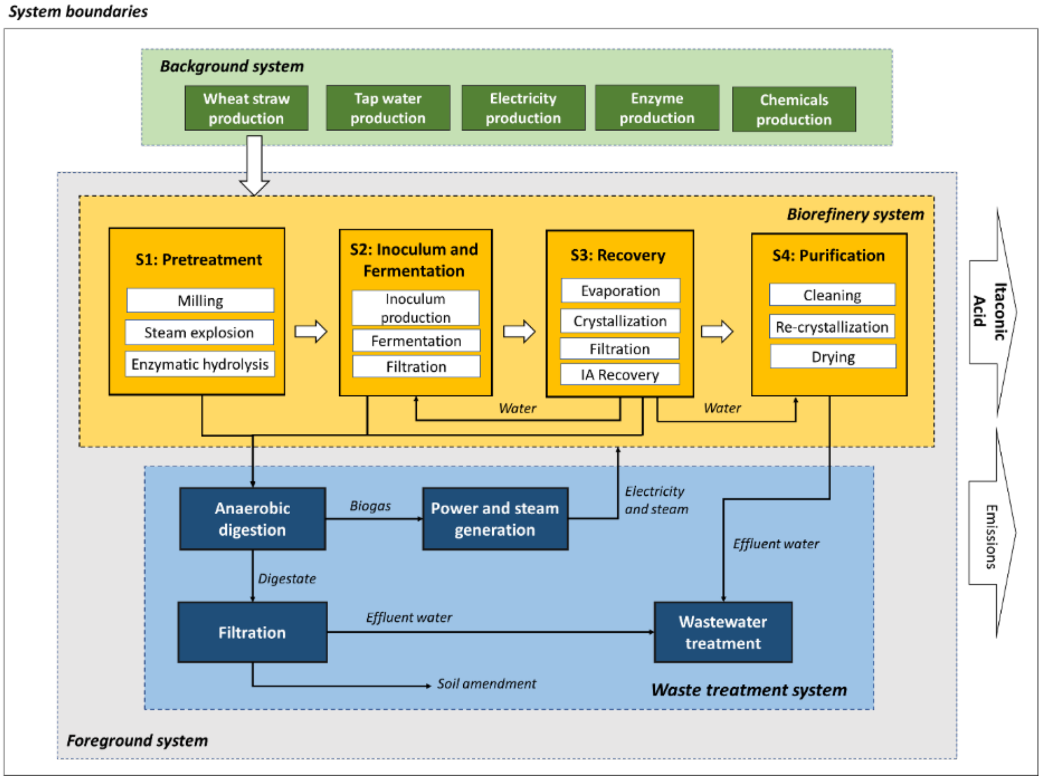


Figure 1. System boundary for IA production under biorefinery approach

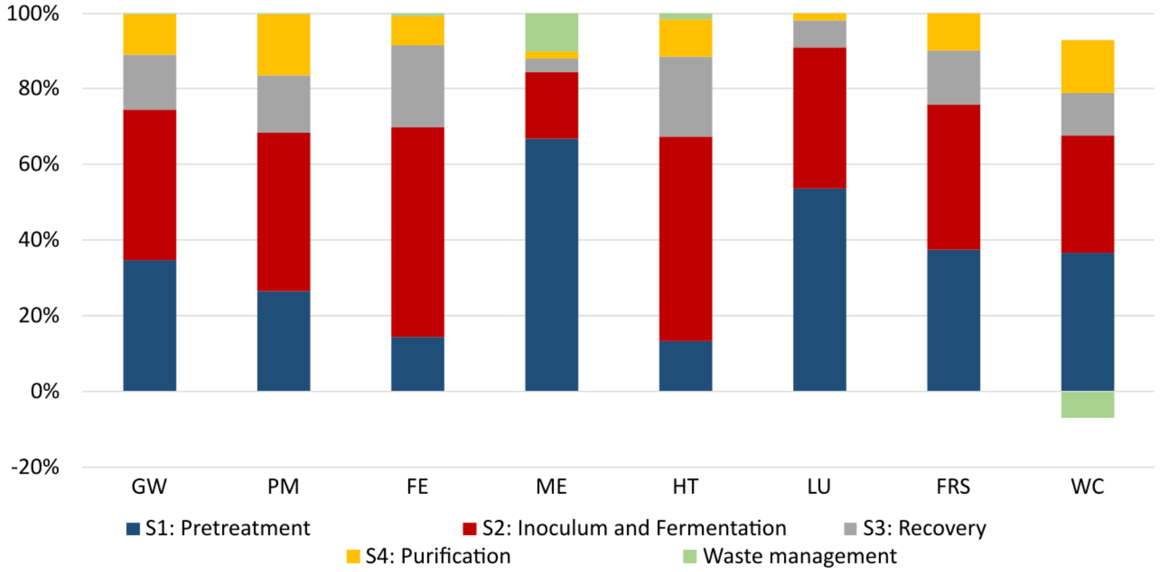
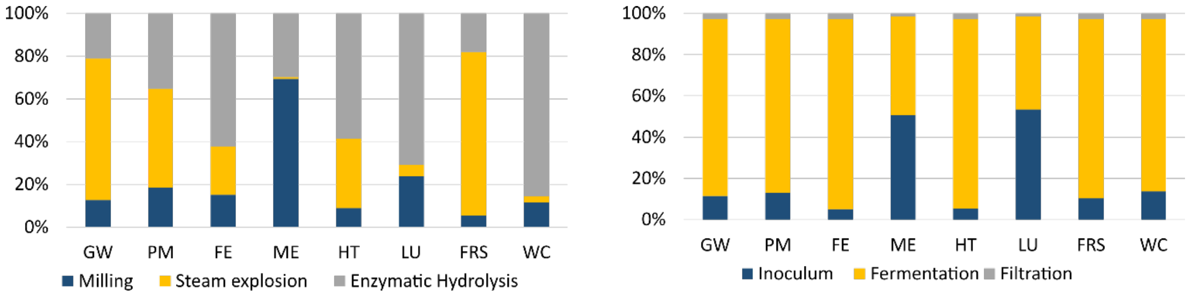
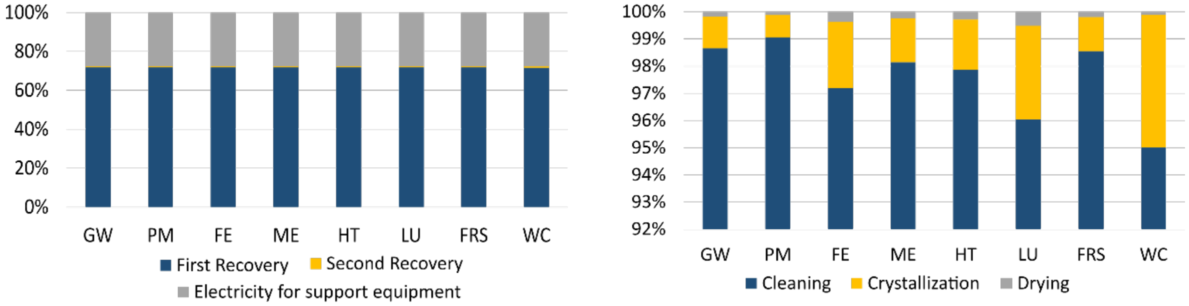


Figure 2. Distribution of environmental burdens of IA production (base scenario) per impact categories and production stages.



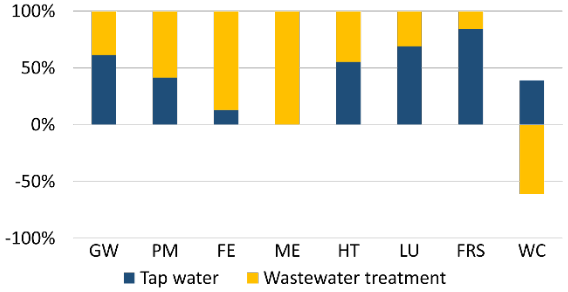
a) S1: Pretreatment

b) S2: Fermentation



c) S3: Recovery

d) S4: Purification



e) Waste treatment

Figure 3. Distribution of burdens per activities involved in the different stages (base case).

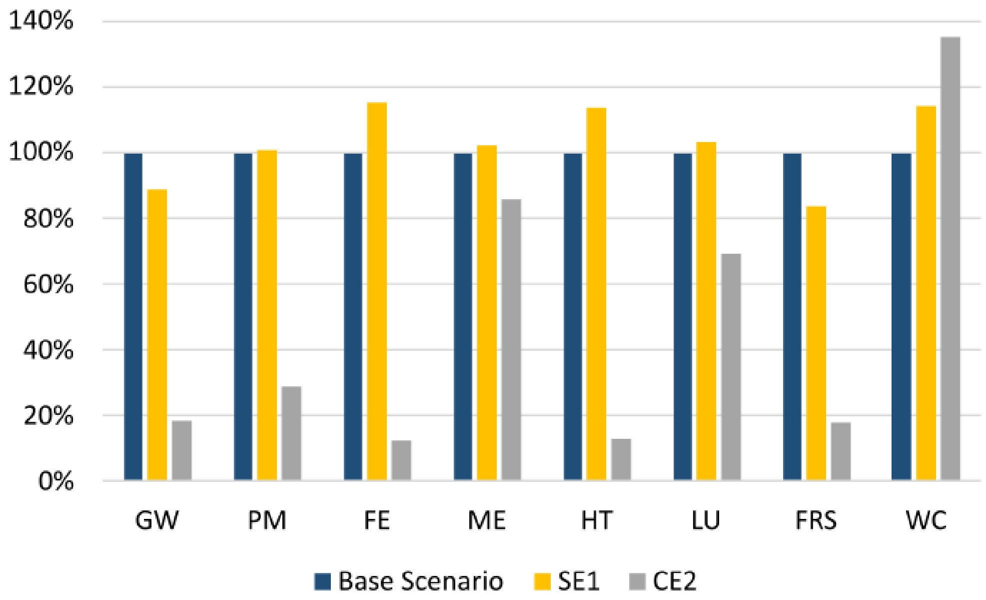


Figure 4. Sensitive analysis: comparative profile of IA production in base scenario; IA production of steam explosion optimization (SE1); IA production with 100% of renewable energy sources and steam explosion optimization (CE2)