Renewable carbon opportunities in the production of succinic acid applying attributional and consequential modelling.

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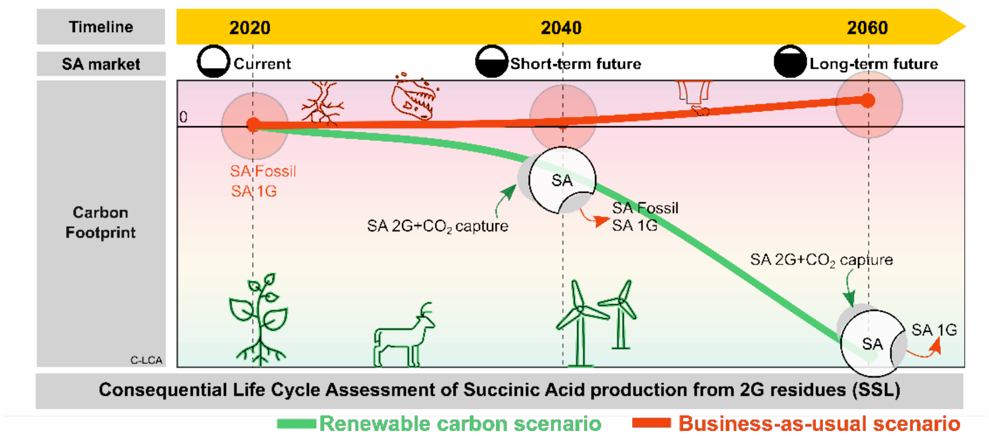
Abstract

Succinic acid (SA) is a top biobased chemical with numerous opportunities in the field of circular economy for climate neutrality. The objective of this work is to environmentally analyze the bio-production of SA from residual sugar-based streams from the pulp and paper industry (SSL). In this study we have complemented attributional life cycle assessment (A-LCA) with consequential life cycle assessment (C-LCA) analyzing the effect of mass versus economic allocation in the first method, and the potential of net reductions of carbon emissions in the chemical industry in the latter. The results present an analysis of the environmental effects of producing SA with two operation modes: fed-batch and continuous fermentation as well as the influence of assuming different geographical locations of the bio-SA production plant through the assessment of the effect of the electricity mix. On the other hand, utilizing the facultative anaerobic and capnophilic bacterium *Basfia succiniciproducens* in the fermentation and thus being CO_2 an input, brings up the opportunity of assessing the carbon capture and utilization potential of the bio-SA value chain. An assessment of the upstream section and origin of CO_2 was performed by studying the effect of capturing CO_2 from industrial static point sources (cement industry and bioethanol production from fermentation). The carbon footprint attributional results suggest

that SA from SSL provides a reasonable substitution for the SA fossil alternative although not reaching the same results when comparing against first generation SA produced from sorghum, which is 62% better. From the consequential perspective, substituting the current market of SA (fossil and 1st generation SA) by SA from SSL will provide improvements of up to 1465% by 2060.

Graphical abstract



Keywords: consequential LCA, biobased succinic acid, sulfite spent liquor, renewable carbon, carbon capture and utilization, climate neutrality

1. Introduction

In the road to decarbonize the chemical industry, the production of chemicals from sugars through microbial fermentation provides opportunities for the implementation of value chains yielding sustainable building blocks from biomass (biorefinery-based processes) [1]. These biochemical building blocks aim to replace petrochemical counterparts achieving lower environmental impacts within their life cycle. Within this approach, the retrofitting of waste streams into processes makes it possible to contribute to the benefits of the circular economy. This concept would technically help reduce the fossil and mineral resources needed while simultaneously maintaining the chemical production sector at a competitive level [2].

The systemic approach of considering bio-based chemicals automatically more sustainable than the petrochemical production may be a correct assumption in the environmental protection areas of climate change and resource scarcity. However, in many instances, issues such as acidification, eutrophication or land use change arise as secondary environmental concerns derived from substitution of the business-as-usual approach (BAU) with a bioeconomy-based scenario. Here, environmental life cycle evaluations (i.e. life cycle assessment methodology, LCA) play a relevant role in determining potential environmental impacts and possible hotspots in newer processing technologies [3].

The "Top ten bio-based building blocks'' list reported by the US Department of Energy includes the bio-based production of succinic acid (SA). This biochemical excels in having a growing niche market and in its potential as platform to produce petrochemicals such as 1,4-butanediol, tetrahydrofuran, γ-butyrolactone, and polymers such as polybutylene succinate (PBS) [4]. On the other hand, its feasibility to replace the petrochemical BAU production process from maleic anhydride presents the potential of achieving reductions in the life cycle environmental impacts, making SA more attractive than other fossil-based precursors. There is wide interest in producing biochemicals such as SA because of their application in many areas such as food, feed, pharmaceuticals, surfactants and detergents, plastics, fibers, solvents, etc. [5].

The market of SA has experienced exponential growth over the past two decades and a turnover in which bio-based SA has become the primary form of production and petrochemical SA has mostly plateaued [6]. The world's consolidated commercial volume of production of bio-SA reached an annual 65 kt in the year 2019 [7] and is expected to grow up to 245 kt produced annually [4], with some degree of uncertainty due to the recent decline in production. The limited market growth of end-products and high failed investment of the company BioAmber, which experienced bankruptcy contribute to this uncertainty [8]. SA is a product with high selling price, in the range of 2.2 €/kg to 2.6 €/kg. In the case of the bio-based production, the production costs and market price tend to be in the upper range [9]. Thus, lowering the cost of production of biobased SA is one of the areas of improvement expected for full deployment of bio-based routes. Its full deployment is also closely related to the price evolution of glucose or the sugar source in fermentation [5,6].

The production of bio-SA from diverse nutrient sources has been extensively studied in literature. Research mainly focuses on first-generation feedstocks or wastes from food [10–15]. However, the production through this type of biomass arises the food competition issue. Implementing the utilization of a residual biomass-based stream rather than crops could be a potential way to decrease production costs as well as environmental effects in direct and indirect land utilization changes [16]. Alternative renewable resources used as feedstocks for the fermentation, such as sugar-rich residual streams from the pulp and paper industry, may put in place measures for sustainable production of bio-SA at lower cost nearing the predicted 0.9-1 €/kg [17]. The pulp and paper industry (specifically sulfite pulping) is a large producer of spent sulfite liquor (SSL), which is a residual lignocellulosic-based stream containing sugars fit for SA fermentation. This sector processes one of the largest amounts of forest biomass, producing around 400 million metric tons of paper annually in around 5000 paper mill facilities worldwide for 2015 [18,19]. SSL has been traditionally incinerated for energy recovery when possible or used for the production of vanillin [20] or bioethanol [21,22]. Diverting the SSL stream to a bio-factory producing SA presents potential for sustainability improvement.

The production of SA through microbial fermentation presents another great sideline opportunity for further decarbonization of the chemical industry. It arises from the conditions of the fermentation, in which the capnophilic microorganism *B. succiniciproducens* requires CO_2 fixation for SA production [17,23]. Thus, CO_2 is fed as a raw material into the bioreactor and fixed into the bio-SA produced. This CO_2 fixation introduces to the value chain of SA the concept of carbon capture and utilization (CCU) [24]. CCU together with carbon capture and storage (CCS) have been acknowledged as feasible routes for sustainable industrialization and decarbonization by the Intergovernmental Panel for Climate Change (IPCC) [25]. The embodiment of carbon in the bioproduct allows a delay in the global warming emissions to the atmosphere, which do not account as net credits to climate change but aid in the overall climate change mitigation efforts. Efforts directed towards implementation of technologies that are able to capture CO_2 emissions from industrial sources, and either store them or utilize them, have at stake the objective of limiting the global temperature rise predicted for the next decades [26]. While some studies have mentioned this as an advantage, to our knowledge the assessment of the potential origin of CO_2 and its environmental implications in the production of bio-SA has not been analyzed yet with detail [27].

In this work, we investigate the environmental implications of the production of bio-SA using SSL from the pulp and paper industry. There are multiple studies addressing the LCA of bio-SA [5,28–31], however, to the best of our knowledge, none have analyzed the effect of carbon accounting within the life cycle of the bio-SA product due to CCU implementation. Furthermore, when including CCU into the modelled system, the evaluation through consequential LCA (C-

LCA), in combination with attributional LCA (A-LCA) becomes key to analyze the broad environmental consequences within the chemical industry.

This study does not aim to focus on the experimental feasibility of SA production involving the use of CO_2 —there are many studies that thoroughly analyze and optimize its manufacture [32–34]. This study aims to verify whether such production would actually be aligned with the environmental targets set by policy-making bodies. This is even more critical when analyzing the rising trends in the number of studies on bio-SA production. The quantification of environmental impacts along the value chain of production allows to analyze whether there is a *quid pro quo* when producing bio-SA and implementing CCU. It also allows to determine whether these kinds of systems are truly sustainable and aligned with decarbonization strategies. In any novel biotechnological process, there is a balancing act between the potential carbon reductions achieved, and the potential increase in impacts in other areas of protection affecting other natural systems and cycles. The specific novelties sought through our study are described below.

1) In the manufacture of bioproducts there is a knowledge gap on the environmental impacts of novel systems framed within the circular economy trend (e.g., utilization of SSL). The results of this assessment aim to shed light on their environmental particularities and complexities and to draw science-based conclusions valuable for stakeholders.

2) While some studies analyze the economic implications and cost-effectiveness of substituting fossil alternatives, in this study we carry out a tailor-made LCA implementing the consequential perspective, going beyond standard LCAs. This is especially critical in products involving interconnected systems or value chains, which will have a change in their environmental impacts as a consequence of a change in the foreground system. Covering the consequential perspective introduces the possibility of assessing future scenarios, through different timelines, by looking at the potential

behavior of the SA market share. While this is not intended to predict the future, it gives valuable insight to the interested parties on whether potential actions are aligned with environmental objectives. C-LCA provides the study with a context that attributional studies may lack.

3) CCU and carbon accounting have been included in the system, extending the inventory to the upstream value chain, exploiting the opportunity to obtain CO_2 from a point emission source. Experts state that any reduction in the production of fossil-based products, principles of collaboration and integration should apply, making the possibility that these kinds of systems give with regards to CCU very attractive. The objective is to be able to draw conclusions of the weaknesses and opportunities that these kinds of synergies bring to the chemical industry.

2. Methods

The environmental assessment for the production of bio-SA was addressed through the LCA methodology [35,36]. This methodology allows to evaluate the whole life cycle of a product systematically and provides quantitative measure of its environmental sustainability. All the compulsory phases in LCA have been addressed: goal and scope definition, life cycle inventory (LCI), life cycle impact assessment and interpretation. We have approached this LCA from two modelling perspectives: attributional and consequential, each of which is further detailed.

2.1 Goal and scope definition

The goal of this study is to determine whether bio-based SA produced from SSL is environmentally competitive when compared to the petrochemical production of SA. The second objective lies in analyzing which are the hot spots, pitfalls, and strengths of SA production through fermentation when SSL is the main carbon source. On the other hand, to analyze whether the mode of operation in fermentation (i.e., continuous fermentation or fed-batch fermentation) and other variables influence in a great deal the overall impact of the system under study.

The common methodological assumptions in LCA for the determination of the carbon footprint (CF) of the product will be closely addressed and examined. Different modelling choices when complex systems —forestry growth, CCU and temporary storage of CO_2— are included in the life cycle of biobased products, will be assessed. Finally, the environmental consequences (C-LCA) of the implementation of bio-SA production from SSL will be addressed through system expansion and an analysis of the marginal products within the system boundaries considered. The objective for C-LCA is to further analyze the effect of multifunctionality without incurring in the uncertainty of the somewhat arbitrary allocation procedures implemented. The functional unit of the study is 1 kg of bio-SA produced from SSL as main nutrient of fermentation and using CO_2 from an industrial point source.

2.2 System boundary definition

The LCA methodology was implemented in a cradle-to-gate perspective, considering, thus, environmental impacts associated to the production of bio-SA from the production of raw materials to the gate of the bio-SA production plant. This includes an assessment of the silviculture activities involved in eucalyptus wood production, the sulfite pulping process, the origin of CO_2 used to guarantee strongly anaerobic conditions in the fermentation, as well as the fermentation and downstream of bio-SA.

The scope of the analysis was not extended to grave because bio-SA is a direct substitute of fossil SA, thus, the use and end of life phases are likely to be the same, resulting, in a comparative assessment, in the same relative differences.

The system (Figure 1) was divided into several processing subsections: SS1. Eucalyptus woodchips, SS2. Pulp and paper industry, SS3. Pretreatment, SS4. Sterilization, SS5. Fermentation (including carbon capture) and SS6. Downstream. The fossil-based production of

SA from maleic anhydride was considered for comparative purposes as the BAU fossil scenario. The production of SA from sorghum was considered as the BAU biobased scenario [6]. The detailed description of each subsystem and the BAU scenario is provided in the Supplementary Material file (Section 1).

In SS5. Fermentation, CO_2 was supplied continuously to the bioreactor. In this work, we have considered the origin of the CO_2 as a relevant factor to the system. Therefore, rather than cutting off the system's boundary, the sourcing and conditioning of the stream was assessed. Including CO_2 within the model is in agreement with the "At the point of substitution, APOS'' modelling approach [37] depicted in the Ecoinvent 3 database [38]. This modelling option assumes that CO_2 is allocated with some impacts from the producer process, rather than considering zero impact. This approach highlights the transformation of the stream from emission to technical flow [39]. CO_2 was considered to be captured at source and used locally, therefore the transport (through pipelines, up to 200 km without recompression) was not included in the subsystem [40]. Furthermore, some authors have analyzed the environmental contribution of the pipeline distribution in CO_2 capture, being negligible in most cases [41].

Two scenarios for the CCU section were considered, assuming the capture of 90% of CO_2 emissions [42]. In the first scenario, CO_2 was obtained from a high volume of emission fossil point source: the production of clinker for the cement industry. A post combustion plant with monoethanolamine was included in the system boundaries for the fossil CO_2 capture and purification [43]. In the second scenario, biogenic CO_2 was sourced from bioethanol production industries. Although many studies present that fermentation processes using CO_2 do not require further purification or processing, we considered the most unfavorable scenario, to obtain conservative results, in which CO_2 is dehydrated [44]. Both scenarios allow to study under which circumstances SA from SSL has the potential to be a temporal carbon fixation system. They represent two opposite cases, referencing the potential use of different CO_2 sources in industry.

For the near term future it is projected that CO_2 will be used from systems in which it exits at higher purity (e.g. fermentation off-gas), while in the long term it is expected that the increase in demand will introduce less pure, post combustion sources for which capture infrastructure must be set in place (e.g. cement industry emissions) [45].

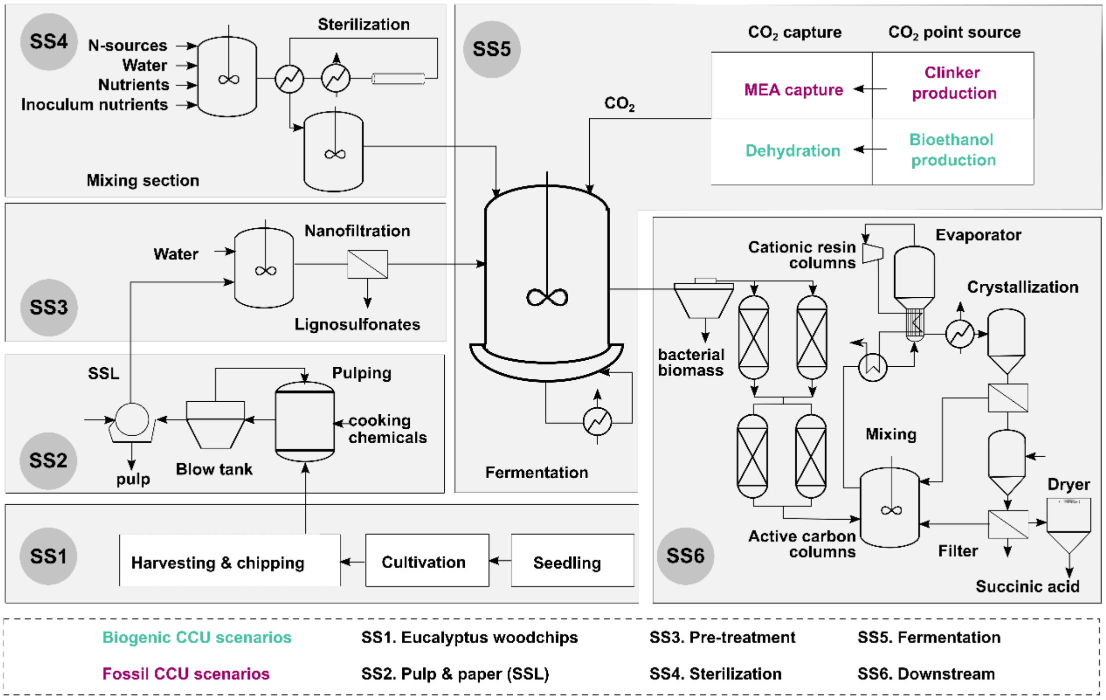


Figure 1. Cradle-to-gate system boundaries for production of bio-based SA from SSL with CCU (functional unit: 1 kg bio-SA)

2.3 LCI assumptions, and limitations

The LCI data, provided in the Supplementary material file Tables S1-S14, were implemented under the following assumptions, for the most part opting for the most conservative decision.

Eucalyptus globulus short rotation plantations were considered for the eucalyptus silviculture subsystem [46], considered to be extensible to European locations [46,47]. A distance of 100 km was considered from the wood site to the pulp and paper industry for preprocessing, with 5% losses during transportation [48,49]. Data for calcium bisulfite pulping was considered for SS2 [50]. This available data for spruce pulping (softwood), was considered to be adaptable to eucalyptus (hardwood) since it was estimated that the energy consumption did not show significant differences [51]. Two main products exit SS2, the pulp and the sugar-rich liquor. Pretreatment in SS3 allows to separate lignosulfonates as a valuable coproduct to the cement industry, utilized in many instances as additives.

The SA plant was considered to be located nearby the point source of CO_2 emissions for both alternatives (biobased and fossil CO_2). Data of mass and energy balances for the bio-SA production, including SSL pretreatment, fermentation and downstream (SS3-SS6) stages was based on process simulation results from UniSim process design and simulator software. The inputs for the simulation of the process were derived from the experimental results of bio-SA production [17,52]. Disodium hydrogen phosphate in the culture medium was modelled through sodium phosphate as proxy. Carnallite was used as proxy for magnesium chloride. Wastewater (with low salt concentrations) from the system, exiting in SS6 was treated with in a generic wastewater plant. Bacterial biomass (including condensate water from evaporation with trace amounts of organic acids) was considered as biowaste treated through incineration. The use of ion exchange columns implies the result of spent activated carbon and ion exchange resin, which were treated as waste. For evaporation in the downstream section of the continuous operation, steam was only needed in a negligible amount for start-up, and the operation could be maintained with electricity.

Capture and conditioning of CO_2 from clinker production and bioethanol fermentation was considered from bibliographic published data assuming that 90% of the emissions were captured [42]. Data on the actual clinker and bioethanol processes was considered as an adapted background process from the Ecoinvent database [38].

2.4 Methodology

2.4.1 Modelling approach and scenario definition

This assessment was approached from two LCA perspectives, attributional and consequential (Figure 2), implemented with the ReCiPe 1.1 hierarchist method in the SimaPro 9.0 software. The Ecoinvent 3.5 database [38] was used for the modelling of the background processes. For the attributional approach, the “at the point of substitution” database set was used while for the consequential assessment the database was modified accordingly [37].

A-LCA is a modelling approach in which there is an exclusive accounting of the share of impacts assigned to our system, per functional unit, from the overall anthropogenic activity. In the boundaries in which the system is interconnected with external systems, system expansion or allocation can be performed, to partition impacts. For the attributional perspective, allocation is the recommended choice to deal with multifunctionality in diverse subsystems along the life cycle of SA (Figure 2). Due to the controversy among physical or causal relationships between products both mass and economic allocation (MA and EA respectively) have been studied [53]. For the biobased BAU alternative, ammonium sulphate was a relevant co-product, and for the purpose of comparability, MA and EA were applied as well. Details of the allocation approaches taken in each subsystem are provided in the Supplementary material file, Section 2.1.

The scenarios (8 in total) considered in the implementation of the attributional approach are a combination of the following variables: continuous and fed batch operation, CO_2 from bio-based point source and CO_2 from fossil point source, MA and EA. Mass and economic allocation factors as well as prices considered for EA are presented in Tables S15 and S16 in the Supplementary material file. A sensitivity assessment of the electricity mix was performed considering two boundaries of action that give a range of possible results. For the improvement scenario, Norway was substituted as electricity of foreground processes. As far as the worsening scenario, Poland was considered to represent the most carbon-based mix among European Union (EU) countries [54]. Results for any other EU country mix were considered to fall within this range of plausible operation.

In C-LCA a causal relationship between our system and limiting external systems is studied. In this sense, the action of producing 1 kg of SA (functional unit) has a positive or negative effect in delimiting systems. In this modelling approach, these consequences are quantitatively accounted [58]. In C-LCA multifunctionality is approached differently, through system boundary expansion (Figure 2). While C-LCA starts also with the definition of the functional unit (1 kg bio-SA) and the scope of system boundaries (cradle-to-gate), the latter includes those activities that change as a response of a change derived from the functional unit. The objective is to determine what unit processes are affected by a change introduced through the functional unit and their causal relationships, assuming an elastic supply-demand relationship —not large price variations should be expected as a result of demand variations [67].

Determining which are the reference products of the system, the constrained markets and marginal suppliers through the 5-step procedure developed by Weidema et al. [55], are key steps of the C-LCA approach depicted with further detail in Section 2.2 of the Supplementary Material file. In summary the cause-effect events analyzed through C-LCA are: i. System boundaries are expanded to include the co-product (lignosulfonates) which is considered as an avoided burden of another market (i.e., plasticizer additive in the cement industry) [56] ii) Bioethanol is the main product derived from SSL —stream rich in sugars apt for any fermentation. The shift of producing SA from SSL rather than the state-of-the-art production of bioethanol results in the need to compensate the bioethanol produced by SSL. iii) Obtaining CO_2 from point sources, to produce bio-SA from SSL would lead to an intrinsic change of the clinker and bioethanol industries. iv) The production of SA with SLL as carbon source will lead to displacing the BAU production methods in the current market. The affected suppliers in this study are part of the selection of a marginal supply mix obtained by extrapolating available market trends.

Depending on the possible SA market behaviors in the future, the forecasted market displacements are studied through different scenarios. The premise is that trends of production are considered to favor bioproduction in the mid to long term, having a transition that works towards decarbonization goals and a reduction of the dependence in fossil fuels. In the short term the displacement of the SA market would be that of a mix of biobased SA produced from sorghum (1st generation) and the fossil production (from maleic anhydride) [6]. The market that is more fit to absorb a diminution in the production in the short term (next 20 years) is a mix of both technologies. Two divergent scenarios have been considered for the short term. In the first hypothetic timeline (TL1), the SA market is expected to go in a direction of growth in which fossil is not completely substituted by bio-based production considering a mix of 50.9% fossil SA and 49.1% SA from sorghum. For the second hypothetical short-term timeline, the growth of bio-based SA is considered to upkeep nowadays market and moves away from the fossil alternative (17.2% fossil, 82.8% bio-based from sorghum). For long term effects (next 40 years), the future scenario is based on a bio-based generalized economy, in which the SA will completely be produced through biogenic sources (SA from sorghum). Thus, the long-term introduction of SA from SSL in the market is expected to substitute 100% BAU bio-based SA production for both timelines. The estimations related to the market growth of SA and calculations of market mixes are included in the Supplementary material (Table S17, Tables S19-S21) according to recommendations in methodology [57,58]. The inventories considered for C-LCA are available as well in the Supplementary material file, Tables S22-S25. A summary of the scenarios considered for C-LCA is provided in Table 1.

Table 1. Selection of C-LCA scenarios attending to the marginal SA mix, the timeline of application and the source of CO_2 (biogenic or fossil). These scenarios were applied to both the continuous and fed-batch modes of operation.

Scenario	**Timeframe**	**Year**	**CO_2 source**	**Substituted SA mix (%)**	
				Fossil SA	Sorghum SA
Current	present	2020	-	-	

B-ST-TL1	short-term, timeline 1	2040	biogenic	50.9	49.1
F-ST-TL1	short-term, timeline 1	2040	fossil	50.9	49.1
B-ST-TL2	short-term, timeline 2	2040	biogenic	17.2	82.8
F-ST-TL2	short-term, timeline 2	2040	fossil	17.2	82.8
B-LT	long-term	2060	biogenic	0	100
F-LT	long-term	2060	fossil	0	100

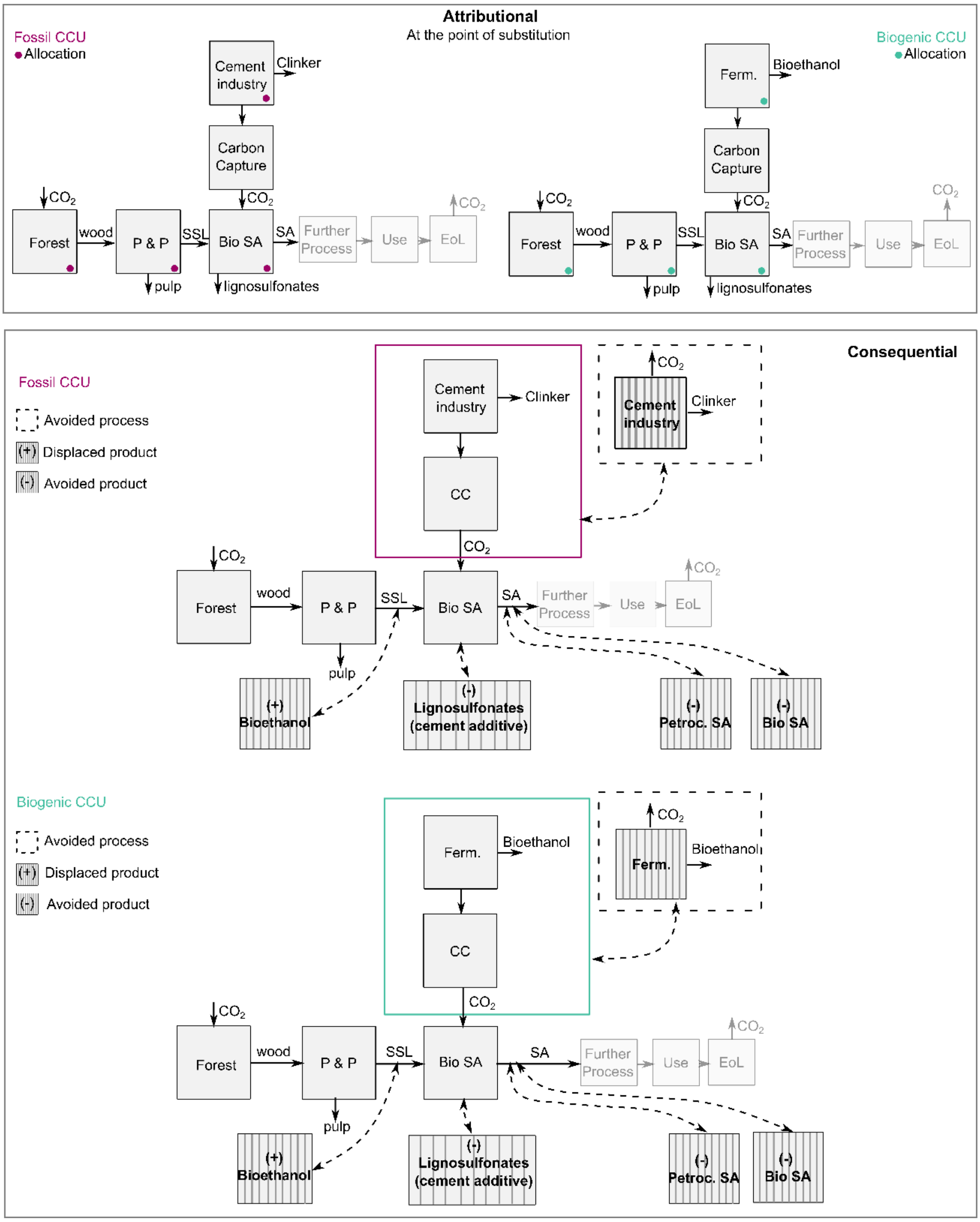


Figure 2. Methodological and system boundary framework for SA production system under attributional and consequential perspectives. In attributional systems boxes with a purple or

turquoise circle have undergone allocation. In consequential systems boxes with a discontinued line display avoided systems. Boxes with (+) display products that have been displaced by the production of 1 kg of bio-SA. Boxes with (-) display products in which their production has been avoided by the production of 1 kg of bio-SA.

2.4.2 Carbon accounting and selection of impact categories

The evaluation of the CF in biogenic systems has been assessed through the consideration of carbon uptake during the growth phase of the trees, adapting the global warming potential impact category in ReCiPe [59]. On the other hand, in this study, the production of SA acts as a carbon fixation process due to the use of CO_2 as feedstock in fermentation. Carbon storage in bio-based systems through CCU allows for a delay in the emissions to the atmosphere. In this study we consider the delay of emissions embodied in SA through the temporal closure of the carbon cycle, which leads to the accounting of the carbon uptake by forest growth as negative emissions. This is feasible since the carbon is chemically stored in SA and because this will be a comparative study in which differences in scenarios can be analyzed in relative terms [60]. Furthermore, the further processing and end-of-life of the compared systems (i.e. SA in this study and BAU SA) are considered to be identical [39,61]. Although the delay of emissions is not discounted over time in our assessment, the time delay of a pulse emission of CO_2 in the framework of emissions with a time horizon of 100 years could be considered to absorb radiation only for the remaining time after the delay. This concept would introduce the time-dependent correction of the Global Warming (GW) emissions over time, which has been proposed by von der Assen et al. [62].

Other depicted impact categories in the assessment were ozone depletion, OD in kg CFC11 eq, ozone formation, OF in kg NO_x eq, terrestrial acidification, TA in kg SO_2 eq, freshwater eutrophication, FE in kg P eq, marine eutrophication, ME in kg N eq, freshwater ecotoxicity, FET in kg 1,4-DCB eq, marine ecotoxicity, MET in kg 1,4-DCB eq, human toxicity, HT in kg 1,4-DCB eq, land use, LU in m^2a crop eq, fossil scarcity, FS in kg oil eq and water consumption, WC in m^3.

3. Results and discussion

The results are presented in two main sections, first dealing with the A-LCA, and then delving into C-LCA conclusions, where an analysis of the implications of the environmental assessment approach is included. Nowadays, the need to make improvements based on science-based targets, makes the quantification of impacts key in the development of technologies based in environmental awareness and abatement of impacts. Thus, providing insight through two perspectives is expected to enrich the conclusions from the study.

3.1 A-LCA perspective for SA production and sensitivity to parameters

Figure 3 shows the comparative profile for the most relevant impact categories studied against the BAU fossil SA and the BAU biobased SA with EA and MA. The CF category (Figure 3-A) displays low dispersion of results among scenarios for SA produced from SSL. The best case regarding the CF is the continuous production, utilizing fossil CO_2 considering MA (1.92 kg CO_2 eq). This scenario improves the BAU fossil alternative by 40.84%, however, it does not present better results when compared to the BAU biobased alternative which is 3.78 times better (1.62 times better when considering EA). The worst alternative is the fed-batch fermentation utilizing fossil CO_2 and considering EA (3.47 kg CO_2 eq). While being very close to the 3.24 kg CO_2 eq of the fossil BAU, this scenario shows significantly worse results when compared to the biobased BAU, being 2.92 times higher (EA). Regarding the carbon intensity of the electricity used within foreground processes, the range of CF variation is significant, showing the relevance of the location of the processing plant, when energy is supplied with the country's electricity mix and the importance of favoring renewable energy. While the European average presents reductions of the CF for all scenarios, a different picture is shown if the Polish electricity mix is analyzed. All scenarios experience an increase of their CF overreaching the fossil production of SA when electricity is very carbon driven. An increase in the equivalent CO_2 emissions in the range of 56.91 to 107.75% is expected for the analyzed scenarios, presenting no improvement with respect to any of the BAU options. However, when a less carbon intensive mix (i.e., Norwegian electricity) substitutes the European average, the drops in the CF are observable for all scenarios in a range of 39.24 to 74.30%. The MA alternatives with Norwegian electricity experience a decrease that results in making an improvement with respect to the bio-based SA with EA alternative. Although fed-batch operation presents slightly differing results to continuous operation, it is safe to say that the environmental results for the studied categories are not sensitive to the mode of operation. The differences among MA and EA, however, are more pronounced.

When the OD impact category is analyzed (Figure 3-B), the worse scenario among the BAU alternatives is the biobased production of SA with EA. None of the SA scenarios from SSL result in higher impacts than the benchmark. It becomes apparent that OD is not affected as much as CF (Figure 3-A) by the electricity of the system varying only in the short range of 7.27 to 10.23% when comparing the percent change from Poland-to-Norway sensitivity scenarios. Regarding the scenarios studied in this work, the best alternative for OD is the fed-batch production with fossil CO_2 and EA, while the worse scenario becomes the opposite combination (continuous operation, biobased CO_2, and MA). In OD, the differences among scenarios with MA due to the origin of the CO_2 utilized are more pronounced. MA produces an effect in which the assigned impacts to the CO_2 (as technosphere flow) from its value chain are increased. Here, the involvement of nitrogenous emissions in the plantation of biomass for bioethanol production makes contributions to the OD category that are slightly more pronounced than those for fossil CO_2. The mentioned differences suppose a decrease of 13 and 16% (for continuous and fed-batch fermentation respectively), not reaching further improvement with respect to the best BAU alternatives.

In OF (Figure 3-C), the different scenarios present quite uniform results, with small variations present in the change from EA to MA —around 1.1 times higher results when allocating with the mass criterion. Although with a smaller range than that of CF, the effect of the electricity mix is remarkable in this impact category. However, even with the greatest reductions achieved through the Norwegian electricity scenarios, none of the alternatives experience further improvement than that of the base-case (European electricity), which is better than bio-based BAU SA (considering EA) exclusively.

Biobased transformations are penalized in TA (Figure 3-D) which is more affected by CO_2 utilized in SA production that originates in fermentation. In this case, the processing of biomass (additionally to SSL from wood) results in slight increases in the impacts of TA similarly to OF (Figure 3-C). However, in all, the scenarios are quite stable. The greatest difference is found in scenarios concerning continuous operation and biobased CO_2 ranging from $1.70 \cdot 10^{-2}$ kg SO_2 eq (EA) to $2.10 \cdot 10^{-2}$ kg SO_2 eq (MA). All scenarios are better than the fossil BAU alternative and the economic-allocated-biobased BAU.

All scenarios, in the FE category, are worse than any of the BAU alternatives (Figure 3-E). The results are anywhere from 67.60 to 79.68% worse than the fossil BAU alternative. They are only slightly improved with the low carbon intensive electricity mix reaching a maximum 16.70% improvement when comparing the biobased BAU (with EA) and the scenario with continuous fermentation, utilization of biobased CO_2 and EA. The ME indicator (Figure 3-F) presented lower dependability on the electricity mix than FE, reaching a low margin of improvement (10.02-20.46%) through the less carbon intensive electricity supply.

Toxicity categories (FET, MET, HT) results (Figure 3-G, Figure 3-H, Figure 3-I) are mostly constant throughout scenarios when observing each of the analyzed parameters (allocation, CO_2 origin and mode of operation). In these categories, most of the contributions have their origin in the use of harsh chemicals, toxic to water bodies as well as to the human health. These chemicals are mostly utilized during fermentation and are not affected by allocation procedures (as otherwise are CO_2 or SSL inputs). They also remain mostly constant when it comes to changes in the system related to the mode of operation. For the three studied indicators, the present study displays worse contributions than the fossil and biobased BAU alternatives. FET and MET (Figure 3-G, Figure 3-H) could reach better results than the fossil BAU option only with improvements regarding the electric mix, with 49.15 and 48.52% improvement margin on average for FET and MET, respectively.

LU is the impact category most affected by allocation (Figure 3-J). When MA is the principle applied, results worsen in the range of 126.23 to 175.24% with respect to EA. LU is very sensitive to allocation because SSL bears impacts directly related to forestry activities. Land occupation of forest systems is being allocated quite differently with respect to the pulp in the cases where mass or economic criteria are selected —56.10 versus 15.65% allocation factor respectively— which goes to prove the volatility of allocation criteria in multifunctional systems.

Although FS (Figure 3-K) behaves usually in a trend like that of CF, in this case there are slight differences due to the accounting of negative emissions related to the growth of biomass and uptake of CO_2 in CF. While the sensitivity to the electricity mix varies in similar ranges, the differences in results among scenarios, especially when modifying the source of CO_2 in fermentation, are mostly unobservable. The FS category improves the BAU fossil alternative by a range of 1.15 to 1.40 times, being the fed-batch production of SA (EA) with biobased CO_2 the best alternative. In WC, as expected, the electricity does not have a direct effect in the results (Figure 3-L). The allocation method changes the results up to 22.78%. The production of SA from SSL consumes more water than the fossil BAU and the biobased BAU (MA), penalization that happens at the expense of reducing the overall CF.

In all, the results show a marked dispersion among categories and parameters. This suggests that, as expected, it is not straight forward to conclude whether fossil SA production or biobased production should be recommended from the environmental perspective. The production of SA from SSL presents results in the same order of magnitude as the benchmarked studies [6,30]. While there is an improvement in the CF category with respect to the fossil production of SA [6], this improvement comes at the expense of burden shifting. Other categories of protection appear to worsen when compared to the fossil alternative, especially OD, eutrophication of waters, WC and LU. First generation biomass —sorghum— provides good expectations with respect to the CF of SA along with some other indicators such as FE, and toxicity categories [30]. However, first generation feedstocks compete with food and feed in land and resource utilization [63]. Utilizing the residual SSL fraction along with CO_2 emissions from point sources provides good potential for the circularity of the process. Circular economy processes allow for the full exploitation of resources, limiting the exacerbation of bioavailability. Accordingly, it is expected that European policies recommend a reduction of consumption, the adoption of circular design standards and the creation of relationships within industrial sites [64] to foster sustainable growth.

Some authors have analyzed the attributional results from different systems producing SA. Foulet et al. [65] evaluate bio based SA synthesized from municipal solid waste, observing an improvement of the climate change indicator with respect to SA produced from sorghum or sugar beet. However, burden shifting also occurs in their study —ecotoxicity (water and terrestrial) and OD categories experience worsening with respect to the BAU. They highlight the need of awareness towards the methodology and assumptions selected in LCA which be carefully considered and reported. Other studies have also evaluated the climate change impact and non-renewable energy use, showing improvements with respect to the petrochemical production of SA [30,66]. However, the disparity of results is again highlighted by the authors, when addressing the influence that the allocation approach has to the system.

In all, a marked dispersion is found in literature, which confirms the need of going one step beyond A-LCA in complex systems involving circularity of residues, capture and use of CO_2 and carbon accounting. This will be addressed in the next section of the manuscript, through C-LCA analysis.

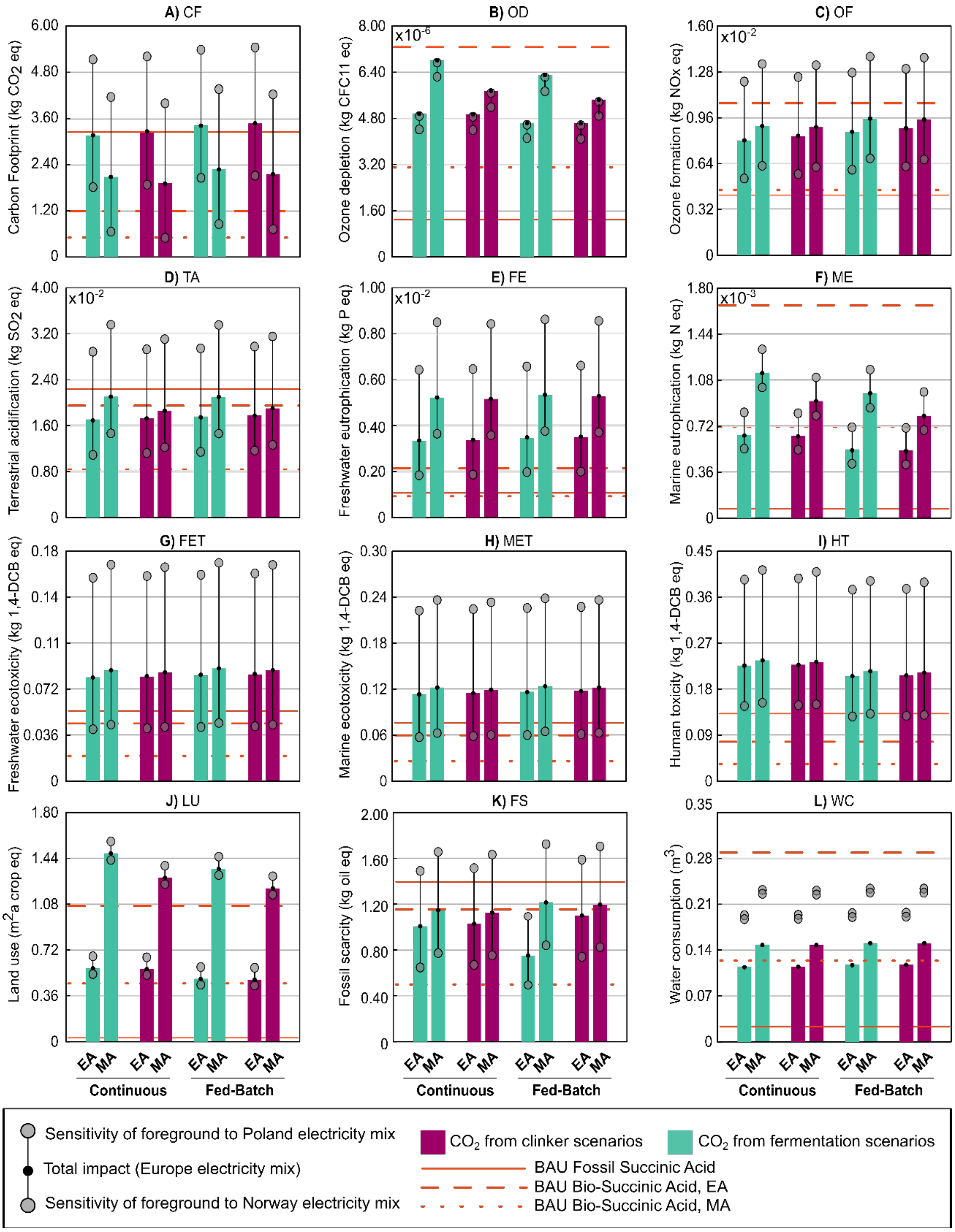


Figure 3. Comparative evaluation of environmental profiles through A-LCA for SA production scenarios from SSL per functional unit (1 kg SA). A) Carbon footprint B) Ozone depletion C) Ozone formation D) Terrestrial acidification E) Freshwater eutrophication F) Marine eutrophication G) Freshwater ecotoxicity H) Marine ecotoxicity I) Human toxicity J) Land use K) Fossil scarcity L) Water consumption. EA: energy allocation, MA: mass allocation

The impact breakdown for the CF in Figure 4 allows to analyze the impact contributions from input flows and emissions in the system, going beyond the net environmental results shown in Figure 3. The contribution analysis in the production of SA is interesting in order to pinpoint which areas of the system contribute the most to the overall CF of 1 kg of SA. Figure 4A, shows that, in the production of SA with biogenic CO_2, the main contributor to impact is SS5, fermentation (53.8%) followed by the downstream operations in SS6 (28.23%). In the case of both subsystems, electricity is responsible for most of the impacts, being also the greatest input flow contribution to all subsystems. From this substantial share of impacts stems the variability found when analyzing the effect of the carbon intensity in the grid. Other hotspot of the system, and the main contributor to SS5 is the utilization of sodium hydroxide, having an effect very comparable to that of the electricity. Sulfuric acid is relevant to SS6, accounting for 36.80% of the CF in the subsystem.

The burdens from the value chain of CO_2 from a biogenic source are accounted for, however, in this case (Figure 4-A), the contributions are not substantially relevant in CF. This is also due to the consideration of EA, which gives more weight to the added value of bioethanol rather than to CO_2 with a 97.30% allocation factor. In EA, also the purchase value of SSL is lower than that of the paper pulp, leading to contributions from the fixation of CO_2 during plant growth of -14.35%.

Better CF is reached in the fed-batch production mode (Figure 4-B) totaling 2.15 kg CO_2 eq. Differences arise in the CO_2 uptake contribution, -33.98%, which accounts for one of the greatest shares in this scenario due to the MA calculations. This goes to show the importance of the methodological approaches in LCA. In this scenario SS5 and SS6 are, again, the greatest burdening subsystems. The fed-batch production depicts a slightly greater impact contribution from steam, being used in SS2, SS4 and SS6, and a relative reduction in the contributions from electricity and sodium hydroxide which originates in changes on the allocation procedure mainly. Along the same lines, the use of CO_2 from cement shows no relevant shares to the total CF.

Other studies have shown contribution assessments in the production of SA from apple pomace, with hotspots found in the downstream section, being very electricity and chemical intensive [28]. In González-García et al. [28], results of 5.30 kg CO_2 eq were reported, showing a benchmark above other results found in the literature which were presented in our study as BAU alternatives. Any of the scenarios in our study present promising results in CF showing 59% improvement with respect to the aforementioned production from apple pomace.

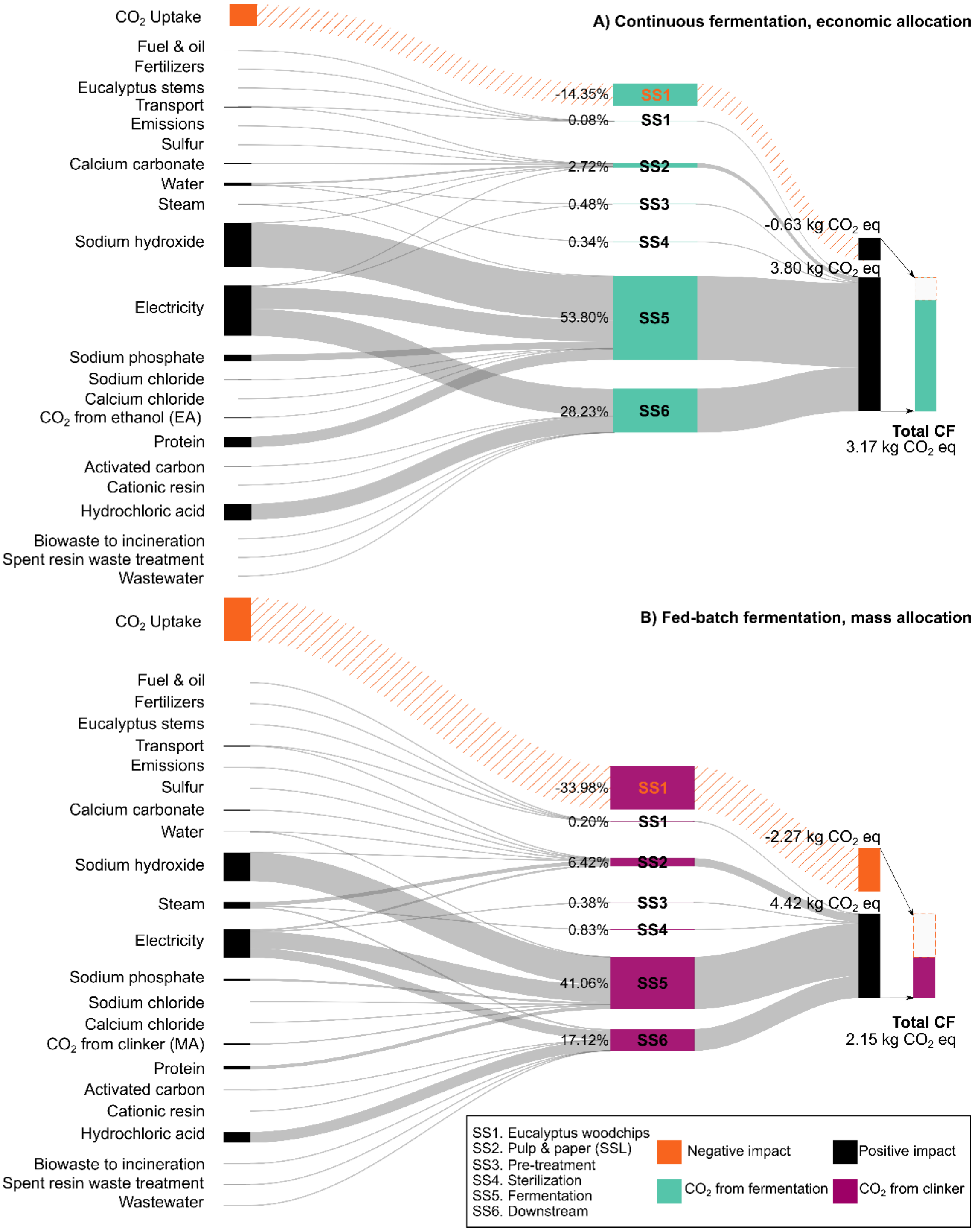


Figure 4. Sankey diagrams depicting the CF breakdown calculated through A-LCA for A) continuous fermentation considering economic allocation and biobased CO$_2$ and B) fed-batch fermentation considering MA and fossil CO$_2$. Each breakdown presents the activity contribution to each subsystem as well as the subsystem contribution to the total CF. Note that A) and B) are scaled differently and should not be directly compared.

3.2 Analyzing the consequences of SA production in the prospect of a future bioeconomy.

The consequential modelling alternative allows not only to account for decarbonization synergies in multifunctional systems, and renewable carbon but also to study potential future projections in the implementation of bioeconomic routes of production. Figure 5 shows the encouraging potential that SA production from renewable carbon and circular value chains has. The CF over the next 40 years is depicted for a projected exponential increase of the SA market production (Figure 5B). It is also depicted as percent improvement per functional unit with respect to the current mix of SA production (12% fossil SA, 88% biobased SA) in Figure 5A. The implications of these results sustain the capabilities of CCU and biomass utilization (i.e., renewable carbon) in the decarbonization of the chemical industry, to the contrary of what was expected when analyzing the attributional model.

The current mix of SA production is expected to experience very little increase of CF even considering that its production will exponentially grow. This is due to the main contribution from non-carbon-intensive production methods (SA from sorghum), with respect to the declining market of fossil SA production. However, these results are still unparalleled by the potential of producing SA from SSL and implementing CCU. The CF for the next 40 years is expected not only to break through the carbon neutral barrier, but to deliver a negative CF in the cradle to gate approach. One could expect improvements of to up to 1465% with respect to the current market per functional unit for SA with fossil-based CO_2. Improvements of up to 1022% could be expected for the biogenic CO_2 SA scenario (See Section 6 of the Supplementary Material for further details).

In these results, the effect of using renewable carbon in the form of CO_2 is far clearer than in the attributional approach. There is an actual measurable effect for the introduction of carbon capture technologies in the studied sectors. Consequential modelling allows to measure how much the change in the emissions profile of SA and its ancillary sectors (cement industry, bioethanol industry) can be expected to vary, and in which direction. Also, differences between using biobased and fossil CO_2 from each industry are visible here. It is more beneficial, when analyzing the CF, to produce SA with captured fossil CO_2 than with biogenic CO_2. For an increase in production, fossil CO_2 could potentially provide a 1.5 times greater sink of carbon emissions than the production with CO_2 from fermentation in the next 40 years.

Here, we not only analyze the potential effects of reducing CO_2 emissions to the atmosphere by coupling the SA production system with emitting industries, but we have also included the effect of multiproduction (production of ammonium sulfate as co-product) in the BAU options, making both alternatives comparable. The results suggest that increasing the resource potential of biomass through the production of multiple products [30] is not as beneficial as reducing carbon emissions from other industries, especially when those are fossil CO_2 emissions.

Due to the complexity of the intertwined systems and data acquisition in the analysis of these industrial synergies, to the knowledge of the authors there is no comparable study to the one presented here. However, the enlightening results suggest that finding areas of improvement by means of finding synergistic approaches to biochemical production is by far the route with most potential to stay within temperature warming scenarios below 2°C [25].

The effect of the carbon intensity of the electricity is well depicted in Figure 5A. The good potential for improvement is illustrated in the figure, following the conclusions drawn from the attributional model. The CF of the fossil CO_2 alternative could be improved 1.3 times additionally in the Norwegian grid, while the effect of a very carbon intensive mix could suppose a 0.8 times decay with respect to the biogenic CO_2 scenario.

Regarding the two temporal approaches taken —TL1 and TL2— their effect is not very pronounced in the results. In terms of market substitutions by the SA proposed in this study, it is not as important whether the decarbonization of the SA feedstock happens at a higher or lower rate, considering that in the long run a complete biobased market is predicted. As expected, the CF prediction is slightly better for the substitution of a SA mix of production that is more fossil-based (TL1) than bio-based (TL2). Improvements in the CF will be greater if the starting point includes higher shares of fossil markets.

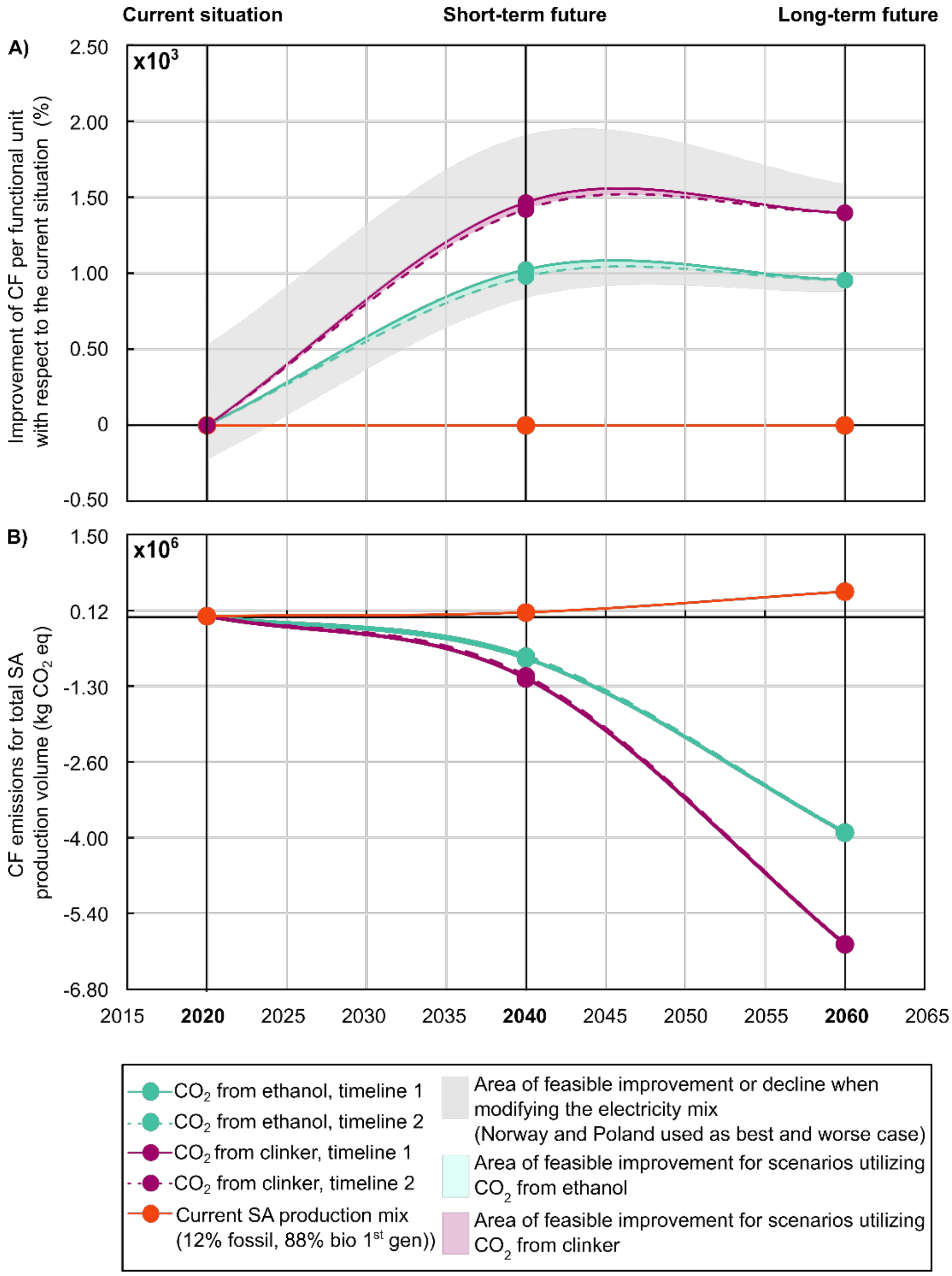


Figure 5. Projection of C-LCA impacts in CF to 20 and 40 years from the current situation. SA is produced in continuous operation. A) Displays the percent improvement of the CF with respect to the current SA scenario per functional unit B) Displays the total CF of SA market considering a growth in the SA production volume

When expanding the indicator portfolio to other environmental impact categories (Figure 6) the results show that, although being a great decarbonization system, the production of SA still presents burden shifting to other natural systems. In CF, the substitution of clinker production and bioethanol production systems with systems involving CCU, as well as the accounting of lignosulfonates as avoided product, provide credits to the system (Figure 6-A). In the current scenario, the inclusion of ammonium sulfate as credits to the system has the same impact. However, as mentioned earlier, the first is a better contribution to decarbonization strategies than the latter.

In the same way, all scenarios present better results than the current alternative of production in impact categories such as TA (Figure 6-C), OF (Figure 6-D), ME (Figure 6-E), FS (Figure 6-K), and WC (Figure 6-L). In these impact categories, the improvements are mainly due to the avoided burdens that arise in the substitution of the current market of SA by our SA from SSL. In the attributional perspective it was harder to depict which scenario was best in categories other than CF (Figure 6-A). On few occasions, the allocation scenarios and methodological implications did not help in providing a clear answer as to which alternative was better. Going back to Figure 6, for OD (Figure 6-B), FE (Figure 6-F), MET (Figure 6-G), FET (Figure 6-H), HT (Figure 6-J), and LU (Figure 6-I) the proposed scenarios of SA production from SSL are worse than the current mix of production. For these categories, the substitution of SA markets has an inverse effect, affecting mostly toxicity-related impact categories. A similar behavior is encountered for the fed-batch operation mode, which is presented in Figures S2 and S3 in the Supplementary material file.

The decision of the modelling approach comes early in LCA, and the attributional approach is usually selected, especially for the comparison of scenarios [67]. However, the results in this study suggest that for systems clearly involving effects outside the foreground boundaries, further assessment of the modelling choice should be made. In this category should fall bioenergy systems, land use change for bioproduction, carbon utilization strategies and circular economy processes. Future research should be focused on analyzing from a holistic perspective —be it C-LCA modelling, or other methodologies— the real effects of implementing novel bioeconomy-based routes of chemical production. This does not take away from the fact that more complex mathematical and modelling approaches come with higher uncertainty, especially when data is not readily available [68]. The effect of sensitivity to free parameters in the consequential model should be addressed and studied. For example, in this assessment, we have evaluated different timelines, SA market mixes and different CO_2 point sources to account for uncertainty. Standardization bodies such as the ISO standard [69] recommend performing sensitivity, consistency, and/or completeness checks within the compulsory interpretation phase. The World Business Council for Sustainable Development (WBSD) also recommends sensitivity checks when analyzing systems involving delayed emissions and carbon storage [70]. The GHG protocol standard also highlights the need to perform a sensitivity check, to analyze in qualitative and quantitative terms which parameters of the study are most sensitive to changes in assumptions [71]. In general, the recommendations show that performing analysis of sensitivity of different variables that may affect the results of the model is highly encouraged.

The intrinsic advantage of temporary storage is to delay emissions (*buying time*) so that other mitigation and fossil resource reduction strategies can be developed. In general, all efforts are made towards eluding the tipping points in the global warming trends that would make the climate situation irreversible. Looking over the next 100 or 150 years, a sustained delay in the emissions to the atmosphere may prove valuable, as the peaks in radiation resulting in the consequential warming of the atmosphere would be substantially reduced. This, in turn reduces the high probability of overreaching the Earth's biocapacity, and further destabilization of the natural carbon cycles. The urgency of meeting reduction targets in the next 10 years makes a delay in the emissions in periods of high atmospheric concentration of GHG emissions overall valuable [25]. Of course, there is still controversy about carbon accounting and how this delay in emissions should be considered, especially depending on the time horizon. Also, controversy lies in the actual benefit of temporary storage, in which opposers argue that the reversibility of temporary carbon storage would result in increased emissions to the atmosphere in the future [72].

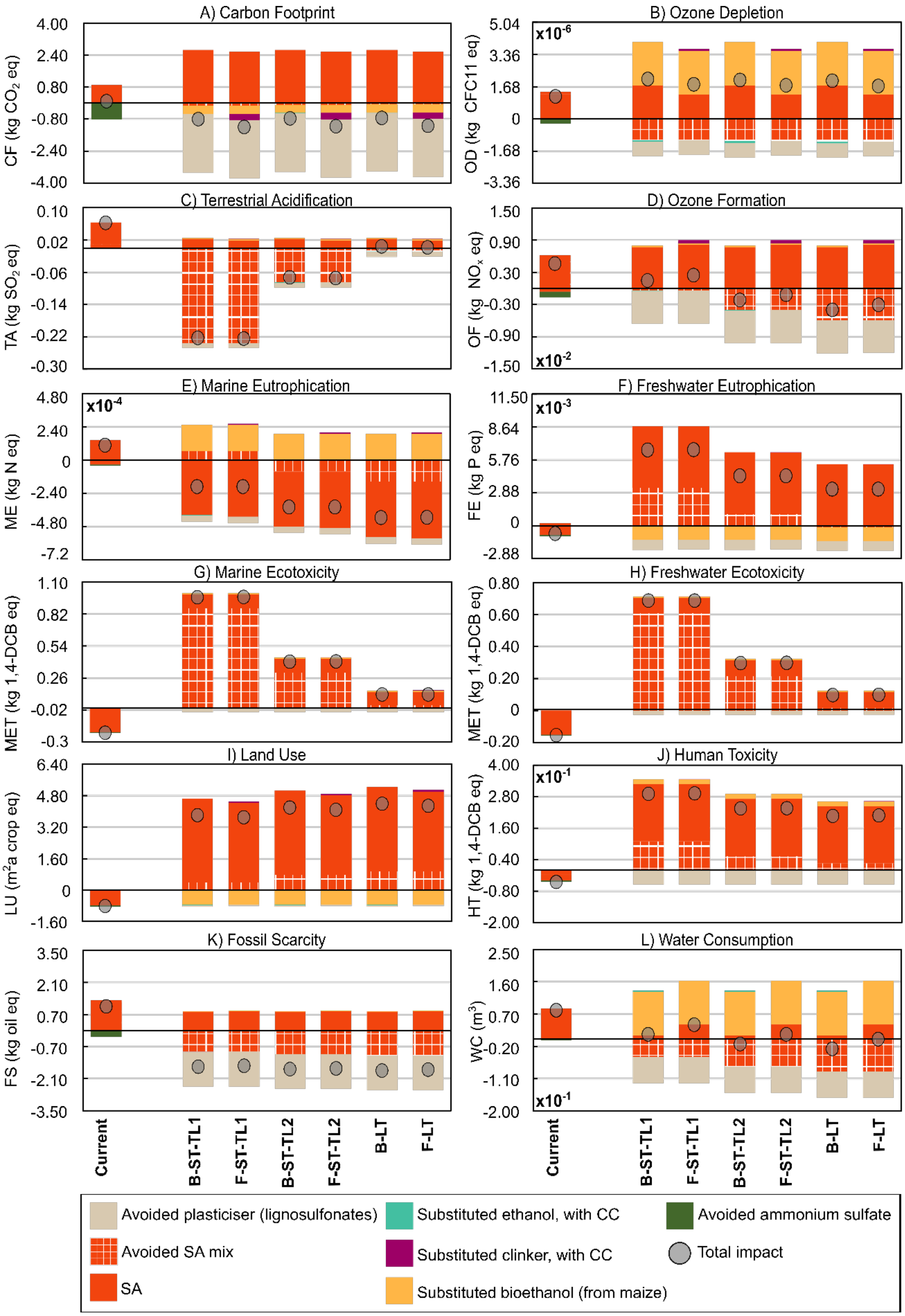


Figure 6. Comparative evaluation and breakdown of environmental profiles through C-LCA for SA produced in continuous operation. A) Carbon footprint B) Ozone depletion C) Terrestrial

acidification D) Ozone formation E) Marine eutrophication F) Freshwater eutrophication G) Marine ecotoxicity H) Freshwater ecotoxicity I) Land use J) Human toxicity K) Fossil scarcity L) Water consumption.

4. Conclusions

A-LCA results, especially of complex systems present a great dispersion leading to troublesome decision making. Many times, the need of assumptions hinders the study, leaving LCA practitioners and stakeholders even more so undecided in the selection of the best processing alternative or best available technique environmentally. When carbon capture —being it for storage or utilization— comes into play, the complexity of emission and technical flow accounting adds to the uncertainty and dispersion of the conclusions drawn from any study. Biomass adds to this, driving the need of more complex and detailed evaluations such as C-LCA. Although not fit for all systems, in this case, the proposed C-LCA has shed light to the study of alternatives in SA production. In all, relying on fossil chemicals (SA from maleic anhydride) is not an option to encompass the objectives of decarbonization set in the Paris agreement. Beyond the *status quo*, the utilization of CO_2 emissions as valuable carbon (i.e., renewable carbon) presents promising results for the reduction of the CF when analyzing the system through a synergistic approach.

The attributional approach depicts results that appear to fit within the state of the art in the literature, showing improvements 41% in the CF of the product when compared against the fossil alternative. However, SA from SSL and CCU does not show improvements with respect to the biobased BAU alternative, which presents 62% better results in CF, when the market value of the co-product ammonium sulfate is considered through EA.

However, in decision-based consequential approach, the projections to the next 20 and 40 years of the SA market, show the great carbon sinking potential of SA produced from SSL and CCU. The production of SA has a cascade of effects, starting from the reduction of the CO_2 emissions in high volume emitting industries such as that of cement or bioethanol. The use the SSL residual stream also presents great potential to decarbonize the future bio-chemical industry. Substitutions of the current market mix of SA by SA from SSL depict up to 1465% improvement with respect to the current market. However, this great potential in CF comes at the expense of degrading other impact categories, such as toxicity-related indicators or LU which are worsened with respect to BAU alternatives.

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