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RESEARCH ARTICLE

Evaluation of sugar feedstocks for bio-based chemicals: A consequential, regionalized life cycle assessment

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Abstract

Fermentable sugars are an attractive feedstock for the production of bio-based chemicals. However, little is known about the environmental performance of sugar feedstocks when demand for sugars increases, and when local conditions and sensitivities of receiving ecosystems are taken into account. Production of monosaccharides from various first- and second-generation feedstocks (sugar beet, sugar cane, wheat, maize, wood, residual woodchips, and sawdust) in different geographic locations was assessed and compared as feedstock for monoethylene glycol (MEG) using consequential, regionalized life cycle assessment. Sugar cane grown in Thailand performed best in all three areas of protection, that is, for life cycle impacts on human health, ecosystem quality, and resources (respectively, equal to -7.6×10^{-5} disability-adjusted life years, -1.2×10^{-8} species-years and −0.046 US dollars per amount of feedstock needed to produce 1 kg of MEG). This was mainly due to benefits from by-products—incineration of sugar cane bagasse generating electricity and use of sugar cane molasses for the production of bioethanol. The wood-based feedstocks and maize performed worse than sugar cane and sugar beet, but their evaluation did not consider that sugar extraction technology from lignocellulose is immature, while identification of marginal suppliers of the marginal crop is particularly uncertain for maize. Wheat grown in Russia performed the worst mainly due to low agricultural yields (with impacts equal to 8.9×10^{-5} disability-adjusted life years, 6.9×10^{-7} species-years, and 1.8 US dollars per amount of feedstock required to produce 1 kg of bio-based MEG). Our results suggest that selection of sugar feedstocks for bio-based chemicals should focus on (i) the intended use of by-products and functions they replace and (ii) consideration of geographic differences in parameters that influence life cycle inventories, while spatial differentiation in the life cycle impact assessment was less influential.

KEYWORDS
biochemicals, ethylene glycol, life cycle assessment, PET, sugars, sustainability

1 | INTRODUCTION

Sugars are becoming an attractive raw material for the production of biochemicals, but the sustainability implications of using sugars in the chemical industry are still debated (Bello et al., 2021; Salim et al., 2019). One biochemical of particular interest to the chemical industry is bio-based monoethylene glycol (MEG), which can be used in the production of polyester fibers and film and for polyethylene terephthalate (PET) resins (Rosenboom et al., 2022; Spekreijse et al., 2019). Sugars can be converted to bio-based MEG by cracking (hydrous pyrolysis) to glycolaldehyde intermediate, followed by its catalytic hydrogenation to the final MEG product (Schandel et al., 2021). Recent life cycle assessment (LCA) studies showed that environmental performance of sugars extracted from first-generation feedstocks (i.e., sugar and starchy crops) might be comparable with that of sugars extracted from second-generation feedstocks (i.e., lignocellulosic biomass; Dammer et al., 2017, 2019). We contribute to this debate by (1) assessing the environmental consequences of increasing demand for sugars and (2) quantifying related environmental impacts while considering geographical differences in sensitivities of ecosystems to the extraction of natural resources (e.g., water) or to receive emissions (e.g., ammonia). Both aspects are highly relevant to consider in the sustainability evaluation of sugars extracted from biomass feedstocks but have not adequately been addressed until now (Bello et al., 2021; Moncada et al., 2018; Morales et al., 2017; Renouf et al., 2008; Salim et al., 2019).

Bio-based chemicals can have a widespread potential to replace fossil-based alternatives, potentially resulting in environmental consequences which cannot be evaluated just by using attributional LCA (Ögmundarson et al., 2020). The consequential life cycle inventory (LCI) modeling framework is more appropriate to use when the analyzed decision (e.g., to increase demand for sugar) is able, via market effects, to cause an increase in capacity to meet the additional demand, triggering long-term investments and larger-scale changes with consequences beyond the studied product system (Bjørn et al., 2018; EC-JRC, 2010). Increasing demand for sugars may lead to competition with food, thereby indirectly contributing to a loss of natural land elsewhere (Bjørn et al., 2017). This effect, known as indirect land-use change (iLUC) has been recognized as an important contributor to climate impacts of biofuels (Naik et al., 2010; Pawelzik et al., 2013; Searchinger et al., 2008). It is, therefore, important to know how sugar feedstocks perform if potential environmental consequences, including impacts from iLUC, are considered.

Spatial differentiation in life cycle impact assessment improves realism of environmental impacts, leading to more accurate results and potentially better decisions (Anton et al., 2014; Heidari et al., 2017; Henderson et al., 2017; Owsianiak et al., 2018; Potting & Hauschild, 2006). Some of the more recent LCIA methodologies provide sets of spatially differentiated factors at country scale, next to generic sets of factors that should be valid on a global scale (Bulle et al., 2019; Huijbregts et al., 2017; Verones et al., 2020). Except water-use impacts, however, implementation of the spatially differentiated sets of characterization factors into LCA modeling software is limited; furthermore, input flows of resource consumptions (again, except water flows) and output flows of emissions are usually not regionalized in unit process inventories. This makes execution of regionalized LCAs challenging. Thus, most LCAs, including LCAs on extraction of sugars from biomass (Bello et al., 2021; Moncada et al., 2018; Morales et al., 2017; Renouf et al., 2008; Salim et al., 2019), have been executed using generic LCIA methods.

The environmental performance of sugar feedstocks is expected to be determined by three main factors. First, the type and source of biomass (e.g., sugar and starchy crops or wood) will determine efforts related to sugar extraction from the feedstock. Sugar extraction results in either sucrose (for sugar crops) or glucose (for starchy crops) or a mixture of glucose with minor fractions of xylose, galactose, arabinose, and mannose (for lignocellulosic biomass). While the extraction technologies from sugar and starchy biomass are well established, extraction processes from lignocellulosic materials are still under development. These processes often rely on pre-treatment of the wood to enhance accessibility of the cellulosic substrate, conversion (through hydrolysis) of cellulose and hemicellulose into their constituting monosaccharides (primarily glucose, xylose, and mannose), and removal of unconverted fractions, such as lignin, and of process chemicals (e.g., solvents; Zhu & Pan, 2010). Second, the geographic location of feedstock production and sugar extraction determines the environmental burden from agricultural production and supply of energy and water. The third main factor is the use of by-products from feedstock production or extraction (e.g., sugar beet leaves used as fertilizers, sugar cane straw used for energy production or sugar cane molasses used for the production of bioethanol) as it determines credits (impact offsets) for replaced products or avoided processes.

Little is known about the sustainability performance of sugar feedstocks when environmental consequences of increasing demand for sugars are considered and local conditions and sensitivities of receiving ecosystems are taken into account, and it was hence the objective of our study to assess and compare environmental consequences of increased demand for sugars from different feedstocks. The comparison was framed around first- and second-generation feedstocks (sugar beet sugar cane, wheat, maize, wood, residual woodchips, and sawdust), geographic locations (considering selected countries from Europe, South America, and Asia), and uses of agricultural by-products (e.g., for bioenergy production or use as fertilizer).

2 | MATERIALS AND METHODS

2.1 | Sugar, starchy, and lignocellulosic feedstocks

A set of 21 scenarios was defined, considering different feedstocks produced in different countries and considering different uses of biomass residues (by-products) from the feedstock production (Table 1). Selection of regions was based on information retrieved from the Danish chemical industry, which needs to decide where to source sugar feedstocks for bio-based chemicals (Schandel et al., 2021). The sugar feedstocks sugar beet and sugar cane represent, respectively, 20% and 80% of the world's sugar production globally (~180 million tons produced in 2021; USDA, 2021). The starchy feedstocks are represented by wheat (~780 million tons produced in 2022), which is used in various food products, and maize (~1 billion tons produced in 2022) which is used to make food, feed, and various industrial products like biodegradable foams, plastics, and adhesives (Scott & Emery, 2015; USDA, 2022). Lignocellulosic feedstocks include wood, residual woodchips, and sawdust. Residual woodchips include various forest chips (from forested areas) and wood residue chips (from untreated wood residues, recycled wood, and off-cuts). About 4 billion m^3 of wood were extracted globally in 2020, of which nearly half was used for energy production (FAO, 2021).

2.2 | Life cycle assessment

The LCA study was carried out according to the guidelines of the ILCD Handbook of the European Commission, in line with the ISO 14044 standard (EC-JRC, 2010; ISO, 2006).

2.2.1 | Functional unit

The main function of the compared feedstocks is to provide monosaccharide syrup of sufficient purity to serve as a substrate for the production of MEG, chosen as an exemplar bio-based chemical for which demand may substantially increase in the future due to its use for the production of polyester fibers and PET resins. The composition of monosaccharides and amounts of syrup per unit of feedstock vary. Thus, to allow for a fair comparison between different feedstocks, the functional unit was defined as "delivery of monosaccharide syrup for the production of 1 kg MEG at a biochemical plant." It was assumed that water content of the syrup is equal to 29%, and that purification steps provide syrups with at least 95% of monosaccharides and <0.1% ash content, considered as a sufficient purity level to enable direct use for MEG production. Assumed yield of MEG is 50% on mass basis, that is, 1 kg of monosaccharide syrup yields 0.5 kg of polymer-grade bio-MEG. We assumed that all feedstocks have a widespread potential to replace fossil-based alternatives.

2.2.2 | System boundaries

Figure 1 shows system boundaries for all feedstocks. They include all the cradle-to-gate processes from the production (if relevant) and supply of the feedstocks, through sugar extraction, hydrolysis (if relevant), and purification, to supply of resulting syrup to the biochemical plant. Residual woodchips and sawdust are waste products and, thus, no impacts are attributed to their production. However, system boundaries consider their (replaced) conventional use (that is, incineration for energy production). Similarly, in accordance with the consequential perspective taken in the LCA, impact offsets are given for replaced products or avoided processes by agricultural or extraction by-products. Note that the production of MEG is outside the system boundary because the current definition of the functional unit allows for a meaningful comparison between feedstocks without considering impacts from the biochemical production since there is no difference in the processing of the different monosaccharide syrups.

During feedstock cultivation, the crop residues that are not used for sugar extraction and are not disposed of as waste are considered as waste products. These include straw (for wheat and sugar cane), maize stover, sugar beet leaves and forest residues (for virgin wood production). Extraction by-products include wheat bran, wheat gluten feed and meal (for wheat) maize oil, maize gluten feed and meal (for maize); furfural and lignin (for the lignocellulosic feedstocks); molasses, residual limestone and beet pulp (for sugar beet); molasses, residual filter cake, bagasse, ashes from bagasse combustion, and vinasse (for sugar cane).

TABLE 1 Overview of the compared sugar feedstocks

Scenario no.	Feedstock	Geographic location of feedstock production[a]	Biomass residues use[b]
1	Sugar beet	NL[c]	Fertilizer
2		DK	Fertilizer
3	Sugar cane	BR-SP	Energy production
4		BR-PE[d]	Energy production
5		TH	Energy production
6			Burning in the field
7	Wheat	DE	Animal feed
8			Fertilizer
9		DK	Animal feed
10			Fertilizer
11		RU	Animal feed
12			Fertilizer
13	Maize	US-IA	Animal feed
14			Fertilizer
15		US-NE	Animal feed
16			Fertilizer
17	Wood	DE	Energy production
18		FI[c]	Energy production
19		PL[c]	Energy production
20	Residual woodchips	DE	No residues
21	Sawdust	DE	No residues

[a]Country/region codes: DK = Denmark, DE = Germany, RU = Russia, US-NE = United States—Nebraska, US-IA = United States—Iowa, NL = The Netherlands, BR-SP = Brazil—São Paulo state, BR-PE = Brazil—Pernambuco state, TH = Thailand, FI = Finland, PL = Poland.

[b]Biomass residues are wheat straw, maize stover, forest residues (e.g., branches, foliage, roots), sugar beet leaves, and sugar cane straw for the wheat, maize, wood, sugar beet, and sugar cane scenarios, respectively.

[c]Location of the MEG plant is Denmark.

[d]Location of sugar cane production is BR-SP (Brazil—São Paulo state).

2.2.3 | Modeling framework

The substitution of PET bottles made from fossil-based MEG with bottles from bio-MEG is expected to have relatively large-scale consequences on the sugar market, potentially requiring the construction of new sugar plants or the conversion of portions of natural land into agricultural land. Therefore, according to the ILCD guidelines, the results of this study are intended for meso-macro-level decision support (situation B) and the consequential approach is applied as LCI modeling framework (Bjørn et al., 2018; EC-JRC, 2010). Consistently with ILCD recommendations, the consequences of a change in demand or supply are modeled by considering long-term marginal data, that is, the supplier (marginal supplier) and/or the technology (marginal technology) that are actually affected by the change for processes that are expected to experience structural changes as a consequence of the decision. In case of multifunctional processes, system expansion was applied as it avoids by-product allocation, and only unconstrained systems capable of adapting to the change in demand (or supply) are considered for modeling the substituted products.

Table 2 summarizes the main methodological aspects of the consequential LCI modeling framework. It shows that (1) the study addresses the increase in the demand for the feedstock (rather than increasing supply of the feedstock), (2) the market is constrained for residual woodchips and sawdust, but not for other feedstocks, and, therefore, increased demand for residual woodchips or sawdust is met by the supply of virgin woodchips, (3) increased demand for wheat, maize, sugar beet, or sugar cane is met by increasing production of barley in Canada (for wheat and sugar beet), oat in Russia (for maize) and either maize in the United States or rice in India (for

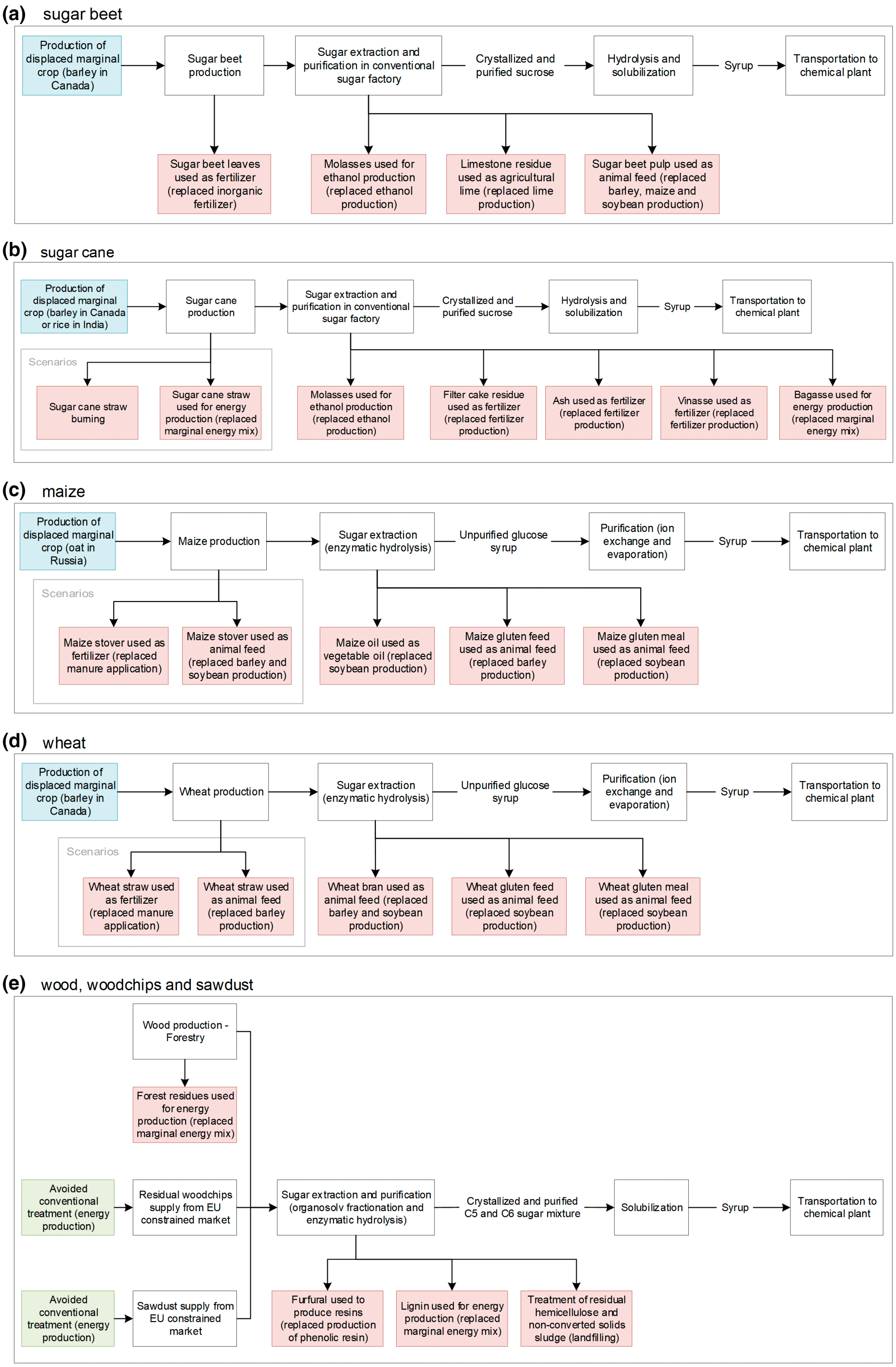

FIGURE 1 System boundaries for the functional unit "delivery of monosaccharide syrup for the production of 1 kg MEG at a biochemical plant" for different sugar feedstocks: sugar beet (a); sugar cane (b); maize (c); wheat (d); and wood, residual woodchips, and sawdust (e). Processes in blue, green, and red represent, respectively: (i) production of displaced marginal crop as a consequence of increased demand for feedstock; (ii) avoided incumbent treatment of waste streams; and (iii) replaced products or avoided processes by agricultural or extraction by-products or treatment of waste.

sugar cane), which are identified as marginal crops and marginal supplying regions. For the wood scenarios, potential consequences from increased demand, which could involve intensification of forest management or replacement of unmanaged forests with intensive forestry were not modeled due to limited knowledge about

TABLE 2 Main methodological aspects of the consequential life cycle inventory modeling framework

Steps	Feedstock: Sugar beet	Sugar cane	Wheat	Maize	Wood	Residual woodchips	Sawdust
1. Change in demand or supply?	This LCA addresses the question: "what are the consequences of increasing demand for sugar feedstocks"?						
2. Identify constraints in the market	No constraints	No constraints	No constraints	No constraints	No constraints	Market is constrained	Market is constrained
3. Identify product substitutions	Not relevant for unconstrained market	Not relevant for unconstrained market	Not relevant for unconstrained market	Not relevant for unconstrained market	Not relevant for unconstrained market	Increased demand for residual woodchips and sawdust is met by increased supply of virgin woodchips	
4. Identify affected production technology	Increased demand for sugar beet and sugar cane is met by displacing marginal crop		Increased demand for wheat and maize is met by displacing marginal crop		Not considered	Virgin woodchips are supplied by the marginal European market mix	
4.1 Marginal crop and marginal supplier	Barley, Canada	1. Maize, USA (for Brazil) 2. Rice, India (for Thailand)	Barley, Canada	Oat, Russia	Not considered	Not relevant for residual feedstock	Not relevant for residual feedstock

actual practices and their implications. Similar studies on wood-based biofuels and construction products applying consequential modeling have acknowledged lack of sufficient data and disregarded modeling of this aspect (De Rosa et al., 2018; Earles et al., 2013). Details of the identification of consequences from changes in demand or supply, identification of marginal suppliers, and identification of by-product substitutions, are presented in the Supporting Information, Sections S1 and S2; Fabbri et al. (2022).

2.2.4 | Model parameters and unit processes

Production and supply of sugar and starchy biomass feedstocks was modeled taking existing unit processes ecoinvent database (version 3.7.1, consequential version) as starting point for further adaptations (Steubing et al., 2016; Wernet et al., 2016). The adaptation considered country- or region-specific (i) agricultural yields, (ii) consequences of increased feedstock demand, (iii) utilization of by-products, (iv) unit processes for water supply, (v) unit processes for energy mixes, and (vi) transport means and distances. Supply of residual woodchips and utilization/treatment of biomass residues was based on existing ecoinvent processes. Sugar extraction from sugar beet and sugar cane was modeled as conventional sugar refinery process, which yields purified white sugar and, thus, includes the purification step based on existing processes in ecoinvent. Unit processes for sugar beet production and sugar extraction in Denmark were adapted using primary data from sugar producer Nordic Sugar A/S. Extraction of monosugars from the lignocellulosic feedstocks was modeled as organosolv (OV) pre-treatment (using numerous organic or aqueous solvent mixtures and a small amount of acid catalyst to solubilize the lignin and hemicellulose fractions) followed by enzyme-mediated hydrolysis based on a large-scale process (Moncada et al., 2018) This is a promising route for sugar extraction from lignocellulose (Bello et al., 2021; Moncada et al., 2018). Hydrolysis of sucrose was modeled based on the process for obtaining invert sugar (Asadi, 2006). Sugar extraction from wheat, maize, and lignocellulosic feedstocks was modeled based on data from studies assessing extraction of sugars for the production of fermentation products or biochemicals (Moncada et al., 2018; Renouf et al., 2008; Salim et al., 2019). Purification reduces ashes or other impurities (such as proteins and pigments) and allows for adjustment of the water content to obtain a syrup of either pure glucose (for wheat, maize), or a mix of glucose and minor fractions of other monosaccharides (for lignocellulosic feedstocks) or a mix of glucose and fructose (for sugar beet and sugar cane). It was modeled as

TABLE 3 Characterized impact scores for all midpoint impact categories per functional unit ("delivery of monosaccharide syrup for the production of 1 kg MEG at a biochemical plant") expressed in category-specific units for different feedstocks, geographic locations of feedstock production, and use of biomass residues. The ranking between scenarios is illustrated within each column with different colors. Red shading indicates the highest impact scores and green the lowest impact scores. All impact categories are from ReCiPe 2016, except marine eutrophication and water scarcity, which are from the IMPACT World+ methodology

Scenario no.	Feedstock	Geographic location[a]	Biomass residues	Global warming	Stratospheric ozone depletion	Ionizing radiation	Ozone formation, human health	Particulate matter	Ozone formation, terrestrial ecosystems
				kg CO_2 eq	kg CFC11 eq	kBq Co-60 eq	kg NO_x eq	kg $PM_{2.5}$ eq	kg NO_x eq
1	Sugar beet	NL	Fertilizer	−0.36	0.000004	0.012	−0.00049	0.0019	−0.00039
2		DK	Fertilizer	−0.23	0.00001	0.013	0.002	0.0011	0.0022
3	Sugar cane	BR-SP	Energy	0.39	0.00001	−0.054	0.0048	−0.00033	0.0011
4		BR-PE	Energy	0.46	0.00001	−0.054	0.0059	0.000043	0.0022
5		TH	Energy	−1	−0.000005	−0.059	0.0056	−0.00028	−0.0022
6			Burning	−1	−0.000005	−0.061	0.0046	−0.00084	−0.0029
7	Wheat	DE	Animal feed	5	0.00016	0.043	0.031	0.02	0.033
8			Fertilizer	−3.3	0.00010	0.11	0.015	0.01	0.016
9		DK	Animal feed	2.5	0.00015	0.046	0.032	0.016	0.033
10			Fertilizer	−5.7	0.00009	0.11	0.016	0.0064	0.016
11		RU	Animal feed	19	0.00038	−0.13	0.066	0.037	0.071
12			Fertilizer	11	0.00032	−0.062	0.05	0.027	0.054
13	Maize	US-IA	Animal feed	5.8	0.00003	−0.066	0.0037	0.031	0.0040
14			Fertilizer	−3	−0.000005	0.024	−0.0076	0.024	−0.0078
15		US-NE	Animal feed	5.8	0.00003	−0.067	0.0039	0.032	0.0043
16			Fertilizer	−3	−0.000003	0.024	−0.0074	0.025	−0.0076
17	Wood	DE	Energy	5.5	0.000006	0.071	0.0038	0.0012	0.0042
18		FI	Energy	5.5	0.000006	0.032	0.0037	0.0012	0.0038
19		PL	Energy	5.7	0.000006	0.072	0.004	0.0013	0.0044
20	Residual woodchips	DE	No residues	9.2	0.000006	−0.053	0.012	0.0081	0.012
21	Sawdust	DE	No residues	9.2	0.000006	−0.051	0.012	0.0082	0.012

[a]Country/region codes: BR-SP (Brasil-São Paulo state); Br-PE (Brasil-Pernambuco state) DE (Germany), DK (Denmark), FI (Finland), NL (the Netherlands), PL (Poland), TH (Thailand), US-IA (United States of America—Iowa), US-IA (United States of America—Nebraska), RU (Russia).

an ion-exchange process where hydrochloric acid, sodium hydroxide, and ammonia are used to regenerate resins. Data for modeling purification of the syrup from wheat and maize are based on technical reports on sugar refining through ion exchange technology (Purolite, 2009, 2021). Purification of sugars from lignocellulosic biomass is included as part of the enzymatic extraction procedure and hence was not modeled separately. It was assumed that all resulting monosugars meet food consumption standards (>0.05% ashes). The product systems were modeled using SimaPro, version 9.2.0.2 (PRé Sustainability bv). Model parameters and unit processes are documented in the Supporting Information, Section S3; Fabbri et al. (2022).

2.2.5 | Life cycle impact assessment

Environmental impact scores were calculated using ReCiPe 2016 (Huijbregts et al., 2017), complemented by IMPACT World+ (Bulle et al., 2019) LCIA methodologies. They were chosen because they (i) generally consider spatial differentiation where relevant (i.e., for ozone formation, particulate matter (PM), freshwater eutrophication and terrestrial acidification in ReCiPe 2016, and marine eutrophication and water availability in IMPACT World+) and (ii) allow environmental impact scores to be calculated at endpoint (damage) levels consistently for all impact categories. Both midpoint impact scores and damage scores were, therefore, calculated. The latter allows for weighting of impact categories contributing to total damage in three key areas of protection in LCIA: (i) human health, where impacts are expressed in disability-adjusted life years, DALY, (ii) ecosystem quality considering terrestrial, freshwater, and marine ecosystems, where impacts are expressed as loss of biodiversity (in species-years), and (iii) resources, where impacts are expressed in US dollars (as of 2013). The IMPACT World+ damages scores, expressed in PDF · m^2 · year (where PDF is the Potentially

Terrestrial acidification	Freshwater eutrophication	Marine eutrophication	Terrestrial ecotoxicity	Freshwater ecotoxicity	Marine ecotoxicity	Human carcinogenic toxicity	Human non-carcinogenic toxicity	Land use	Marine resource scarcity	Fossil resource scarcity	Water scarcity
kg SO_2 eq	kg P eq	kg N eq	kg 1,4-DCB eq	kg 1,4-DCB eq	kg 1,4-DCB eq	kg 1,4-DCB eq	kg 1,4-DCB eq	m^2a crop eq	kg Cu eq	kg oil eq	m^3 world eq
0.013	−0.00002	0.001	3	−0.038	−0.043	−0.059	−4.9	3.3	−0.018	−0.027	−4.9
0.022	0.00012	0.0011	0.89	0.0065	0.01	−0.034	−4.3	3.7	−0.014	−0.15	−1.8
0.016	0.00031	0.00033	5.4	0.01	0.018	0.021	1.4	1.4	0.0032	0.2	1.5
0.017	0.00031	0.00036	6.1	0.011	0.02	0.022	1.4	1.4	0.0034	0.22	1.5
−0.0044	−0.00076	−0.0003	1.8	−0.045	−0.054	−0.045	−1	−0.71	−0.0015	−0.21	−20
−0.0046	−0.00075	−0.00031	1.7	−0.039	−0.046	−0.041	−0.78	−0.79	−0.0013	−0.21	−20
0.12	0.00081	0.0043	20	0.42	0.57	0.29	−13	29	0.045	2.6	15
0.1	−0.0014	0.0021	0.31	0.16	0.24	0.087	−19	29	−0.035	0.63	10
0.11	0.00038	0.005	23	0.46	0.62	0.3	−9	29	0.042	1.7	14
0.091	−0.0018	0.0029	3.6	0.2	0.28	0.1	−16	29	−0.038	−0.29	10
0.18	0.0045	0.0061	38	0.82	1.1	0.49	−7	66	0.1	5.2	44
0.16	0.0023	0.004	18	0.56	0.74	0.28	−13	65	0.024	3.2	39
0.19	0.00059	0.0059	6.4	−0.034	−0.08	−0.18	1.0	−0.05	0.008	1.7	18
0.18	−0.0021	0.004	−14	−0.28	−0.41	−0.39	−4.6	9.4	−0.085	−0.41	51
0.2	0.00062	0.0061	6.7	−0.029	−0.075	−0.18	1.1	0.23	0.0085	1.7	20
0.19	−0.002	0.0042	−13	−0.28	−0.4	−0.39	−4.5	9.7	−0.084	−0.4	53
0.0044	−0.00003	0.053	7.4	0.16	0.2	−0.069	3.6	4.4	0.0041	0.36	1.9
0.0047	−0.0001	0.053	5.6	0.17	0.21	−0.067	3.5	6.1	0.0041	0.33	1.9
0.0048	−0.00001	0.053	10	0.17	0.21	−0.067	3.7	4.4	0.0044	0.4	2
0.025	0.0055	0.053	14	0.35	0.45	0.25	10	6	0.0044	1.2	2.4
0.025	0.0055	0.053	14	0.35	0.46	0.25	10	6	0.0044	1.2	2.4

Disappeared Fraction of species) were converted into species-years assuming terrestrial species density equal to 1.48×10^{-8} species/m^2 (Goedkoop et al., 2009). Spatially differentiated characterization factors for impact categories other than water use (which are already implemented in SimaPro) were imported into SimaPro software and matched with those input and output flows, which can be regionalized and contributed most to total midpoint impacts.

Greenhouse gas (GHG) accounting in ReCiPe 2016 considers emissions of biogenic CO_2 and removal of CO_2 from the air as carbon neutral. In addition to GHG process emissions, we accounted for GHG emissions stemming from land-use change, LUC (i.e., the process of transforming the original use of a land into a different one), differentiating between direct land-use change, dLUC (i.e., change in the use or management of land by humans that may lead to a change in land cover) and iLUC (i.e., a shift in land use caused indirectly somewhere else in the world as a consequence of a direct LUC). Different methods have been developed for the estimation of GHG emissions linked to land-use change. An overview of existing iLUC and dLUC methods is presented in the Supporting Information, Section S4; Fabbri et al. (2022).

In this study, the normative specification PAS2050 of the British Standards Institution was used to account for dLUC emissions (BSI, 2011). It was chosen because it (i) is in line with the IPCC guidelines (IPCC, 2006, 2019), (ii) is recommended as a reference methodology in the Product Environmental Footprint guide of the European Commission (2013, 2021) and the GHG-Protocol (WRI WBCSD, 2011), and (iii) has already been implemented, with some adaptations, into ecoinvent 3.7.1 (Blonk Consultants, 2021; Donke et al., 2020; Reinhard et al., 2017).

Indirect LUC are relevant for consequential LCA. The estimation of iLUC emissions is generally more complex and uncertain because indirect emissions cannot be directly measured and because of difficulties in establishing a relationship between the demand for agricultural products and land-use changes in the context of global land-use dynamics (De Rosa et al., 2016). While different methods for estimating iLUC emissions are available, there is no consensus on which method provides the most appropriate estimates of iLUC emissions (Ahlgren & Di Lucia, 2014; De Rosa et al., 2016; Delzeit et al., 2016). In this study, the method of Schmidt et al. (2015) was used to account for iLUC emissions. It was chosen because it (i) has been applied in different studies on biofuels and biorefinery products (Corona et al., 2018; Lask et al., 2021; Prapaspongsa & Gheewala, 2017) and (ii) was previously used to support decision making by the Danish Environmental Protection Agency and the Danish Energy Agency (Høst-Madsen et al., 2014; Schmidt & Munoz, 2014).

3 | RESULTS

Table 3 shows the ranking of feedstocks according to their characterized scores in each of the considered midpoint impact categories. Four main trends are observed. First, the production of monosaccharide syrups can lead to both environmental burdens (i.e., positive scores) and environmental benefits (i.e., negative scores), depending on the scenario and impact category. Second, the environmental performance largely depends on the use of biomass residues and the functions that they substitute. Impact scores are generally lower when biomass residues replace conventional fertilizers, than when they replace animal feed. This is true for all feedstocks except maize, for which land-use and water scarcity impacts are lower when animal feed is replaced. Third, the geographic location influences the environmental performance, depending on the feedstock and impact category. For example, sugar cane produced in Thailand had lower impacts in 16 out of 18 midpoint impact categories, compared with sugar cane produced in Brazil. The fourth main observation is, that with some exceptions, sugarcane and sugar beet feedstocks generally perform the best, while wheat produced in Russia (again, with some exceptions) generally performs the worst. Ranking of other feedstocks and other countries depends on impact category and trade-offs between different environmental impacts occur.

To weigh different impact categories in terms of their contribution to potential damages and ultimately facilitate comparison between scenarios, damage scores were computed (Table 4). They showed that the exceptions from general trends at the midpoint level did not propagate to damage scores. The damage assessment confirmed that sugarcane produced in Thailand and wheat produced in Russia are indeed the best and the worst sugar feedstocks, respectively. The largest contributors to damages to human health were impacts stemming from water availability (up to ~95% contribution for sugar beet and sugar cane scenarios), impacts from global warming (up to ~35% contribution for wood scenarios), and impacts from exposure to PM (up to ~25% contribution for wheat scenarios). The largest contributor to damages to ecosystem quality across all feedstocks was land use (from ~50% to ~90% contribution to total damage), except for two maize scenarios for which damage scores were dominated by impacts related to terrestrial acidification (~70% contribution to total damage). Damages on resources were generally dominated by impacts from fossil resource scarcity (up to 99% contribution to total damage).

4 | DISCUSSION

4.1 | Environmental burdens and benefits

Negative scores can be explained by benefits of using biomass by-products. Figure 2 illustrates this for global warming, chosen as an exemplar impact category given the strong focus on climate in the ongoing debate on sustainability of bio-based chemicals. For sugar beet and maize, agricultural residues like beet leaves and maize stover replace conventional fertilizers like manure, which brings climate benefits (note that in the consequential modeling framework, the decrease in demand for manure, whose market is constrained, was modeled as avoided production of inorganic fertilizers). For sugar cane and wheat, benefits stem mainly from the use of extraction by-products, like sugar cane bagasse, which avoids electricity production from the grid when the bagasse is incinerated, sugar cane molasses which are used as feedstock for the production of ethanol (replacing ethanol production from dedicated sugarcane cultivation), or wheat bran and wheat gluten which replace conventional animal feed (soybean and barley). For other scenarios, climate burdens associated with cultivation of feedstock or extraction of sugar were, however, higher than benefits from replaced energy or animal feed. This was the case of incineration of sugar cane straw with energy recovery in Brazil, use of maize stover as animal feed, which replaces the production of barley, and use of wheat bran, gluten feed, and gluten meal as animal feed, which replace the production of barley and soybean meal (Figure 2e–g). Trade-offs between burden and benefits occur in all other impact categories, albeit contribution patterns of different processes can be different compared with climate change.

TABLE 4 Damage to the areas of protection of human health, ecosystems, and resources per functional unit ("delivery of monosaccharide syrup for the production of 1 kg MEG at a biochemical plant") for different feedstocks, geographic locations of feedstock production, and use of biomass residues. The ranking between scenarios is illustrated within each column with different colors. Red shading indicates the highest damage scores and green the lowest damage scores

Scenario no.	Feedstock	Geographic location[a]	Biomass residues	Human health DALY	Ecosystems species · year	Resources USD2013
1	Sugar beet	NL	Fertilizer	−1.3E-05	3.1E-08	0.033
2		DK	Fertilizer	−5.E-06	3.7E-08	−0.024
3	Sugar cane	BR-SP	Energy	2.1E-06	1.7E-08	0.074
4		BR-PE	Energy	2.5E-06	1.8E-08	0.084
5		TH	Energy	−7.6E-05	−1.1E-08	−0.047
6			Burning	−7.6E-05	−1.2E-08	−0.046
7	Wheat	DE	Animal feed	5.E-05	3.1E-07	0.93
8			Fertilizer	3.E-05	2.7E-07	0.31
9		DK	Animal feed	4.5E-05	2.9E-07	0.6
10			Fertilizer	2.5E-05	2.6E-07	−0.014
11		RU	Animal feed	8.9E-05	6.9E-07	1.8
12			Fertilizer	6.9E-05	6.5E-07	1.2
13	Maize	US-IA	Animal feed	−4.5E-05	5.7E-08	0.82
14			Fertilizer	4.5E-05	1.1E-07	0.18
15		US-NE	Animal feed	−4.4E-05	6.1E-08	0.82
16			Fertilizer	4.7E-05	1.2E-07	0.19
17	Wood	DE	Energy	1.4E-05	5.6E-08	0.2
18		FI	Energy	1.4E-05	7.1E-08	0.19
19		PL	Energy	1.4E-05	5.7E-08	0.22
20	Residual woodchips	DE	No residues	2.4E-05	9.E-08	0.21
21	Sawdust	DE	No residues	2.4E-05	9.E-08	0.21

Abbreviation: DALY, disability-adjusted life years.

[a]Country/region codes: BR-SP (Brasil-São Paulo state); Br-PE (Brasil-Pernambuco state) DE (Germany), DK (Denmark), FI (Finland), NL (the Netherlands), PL (Poland), TH (Thailand), US-IA (United States of America—Iowa), US-IA (United States of America—Nebraska), RU (Russia).

The three lignocellulosic feedstocks sourced from managed forests or from residues of forest or industrial activities performed worse than sugar beet and sugar cane, better than wheat, and comparable with maize (Table 4). Although some studies have found that second-generation feedstocks were environmentally less burdensome or comparable with food crops (Bello et al., 2021; Moncada et al., 2018), conversion of woody biomass into sugars is not without challenges (Ahorsu et al., 2018; Mohr & Raman, 2013). These relate especially to the use of harsh and energy-intensive pretreatment methods necessary to overcome the high recalcitrance of the wood, resulting in larger environmental impacts compared with first-generation biomass (Dammer et al., 2017, 2019; Morales et al., 2017). Indeed, the largest environmental problem with using lignocellulosic feedstocks in our study was the actual sugar extraction step, driven by the input of enzymes, steam, and the disposal (through landfilling) of the residual fraction from the extraction process (Figure 2f,g). Another factor that explains the modest performance of lignocellulosic feedstocks is the underlying consequential modeling framework, and especially consistent use of system expansion (as opposed to allocation applied in attributional LCA studies). For wood, a significant environmental burden is associated with replaced bioenergy generation, which has to be produced from conventional sources. For residual woodchips and sawdust, the consequence of increased demand for these constrained feedstocks is the indirect increase in virgin wood supply. The influence of the modeling approach on the performance of feedstocks for biochemical or biofuel production has previously been recognized (Brandão et al., 2021; Prapaspongsa & Gheewala, 2017). Our consequential LCA results show that at the current level of technology development use of lignocellulosic materials as a source of sugars is not environmentally

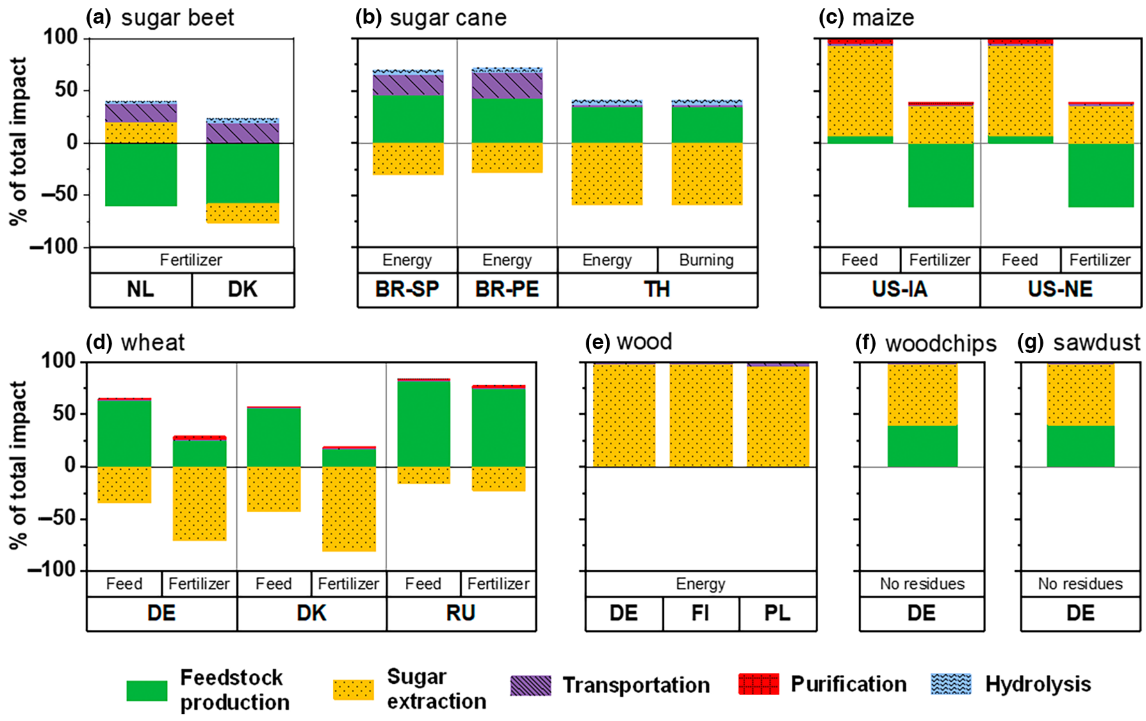

FIGURE 2 Contribution of life cycle processes to global warming impacts for different feedstocks, geographic locations of feedstock production, and use of biomass residues. The feedstocks are: sugar beet (a); sugar cane (b); maize (c); wheat (d); wood (e); residual woodchips (f); and sawdust (g). The final purification step for sugar beet, sugar cane, and the lignocellulosic scenarios is nested into the sugar extraction. Below each bar, the use of biomass residues (by-products) from the feedstock production is listed (i.e., as fertilizer, as animal feed, for energy recovery or burned on site). Country/region codes are the same as in Table 1.

competitive compared with sugar extraction from sugar-rich crops.

4.2 | Influence of geographic location

Largest differences in impact scores between geographic locations were observed for freshwater eutrophication (by a factor of 3 to 8 across all feedstocks) and freshwater ecotoxicity (by up to a factor of 5 for wheat). These differences are mainly caused by geographic variability in input and output flows, as determined by differences in crop yields or intensity of electricity grid mixes between countries (see Table S5; Fabbri et al., 2022). For sugar beet, differences in global warming impact scores between the Netherlands and Denmark are due to differences in GHG emission savings caused by different amounts of manure (here, modeled as inorganic fertilizers) which are replaced by the sugar beet leaves (Figure 2a). For sugar cane, differences between Thailand and two states of Brazil can be explained by the fact that the Thai electricity mix is less clean than the Brazilian mix (returning larger benefits when replaced by electricity resulting from bagasse combustion), and by higher sugar cane yield in Thailand than in Brazil (Figure 2b). By contrast, differences in energy mixes, water supply processes, and transportation efforts are not high enough to influence the comparison between the two Brazilian states. For wheat, slightly lower global warming impact scores for Denmark compared with Germany are due to a slightly cleaner long-term marginal energy mix in Denmark (mainly based on wind power and biomass). Wheat produced in Russia generally performs worst because of low agricultural yield (about three times lower compared with Germany or Denmark), which translates to significantly larger agricultural efforts per unit of syrup produced. The same processes, albeit to a different extent depending on the impact category, explain differences in impact scores for other impact categories.

Although spatial variability in sensitivities of ecosystems was considered in this study where possible and relevant (i.e., for impacts stemming from ozone formation, PM formation, freshwater, terrestrial and marine eutrophication, and water availability), for the majority of impact categories spatial differentiation in life cycle impact assessment was less important than geographic differences in life cycle inventories. With two exceptions, the ranking of feedstocks was not influenced by the consideration of spatial differentiation in the life cycle impact assessment (see Supporting Information, Section S5; Fabbri et al., 2022). The first exception is human health impacts stemming from PM, where impact scores become lower for sugar beet in Denmark (compared with the Netherlands) when spatial differential in life cycle impact assessment was considered. This is because ammonia, which is a precursor of PM and an important contributor to related human health impacts, is about eight times more problematic (per unit emission) in the region comprising the Netherlands compared with the region comprising Denmark. Indeed, dispersion characteristics of ammonia and resulting PM and population levels which determine PM intake, are significantly different in the Netherlands compared with Denmark (van Zelm et al., 2016). The second example concerns freshwater eutrophication, where maize in the United States performed worse relative to other countries when spatial differentiation in eutrophication impacts was considered. Regionalized characterization factors for phosphorus emitted to freshwater, which is a major contributor to freshwater eutrophication impacts, are four times higher in the United States than the generic factor for waterborne phosphorus. Comparison between regionalized and generic impacts could not be made for water use because water use inventories are fully regionalized in ecoinvent 3.7.1 (which presents a challenge for implementing generic factors for water extraction). Water scarcity indicators vary by three orders of magnitude across the globe (Boulay et al., 2018), suggesting that ranking of scenarios could be influenced by consideration of spatial differentiation in this impact category. Nevertheless, our findings indicate that spatial differentiation in impact assessment may not necessarily lead to changed conclusions in LCA, which is consistent with an earlier LCA study where large benefits were obtained from collecting site-specific inventories, as opposed to potential benefits from implementing spatially differentiated LCIA method (Owsianiak et al., 2018).

The iLUC emissions generally did not influence ranking of feedstocks (see Figure 3; Supporting Information, Section S5; Fabbri et al., 2022). Emissions of CO_2 associated with iLUC increase global warming impacts by 0.13 to 2.9 kg CO_2 eq per functional unit. The smallest increases were for sugar beet in the Netherlands and Denmark (0.13 and 0.15 kg CO_2 eq, respectively). The largest increases were for wheat in Denmark and Germany (2.1 kg CO_2 eq in both countries). For sugar cane in Thailand, there was a positive contribution from iLUC. This is because burdens from iLUC are outweighed by the avoided iLUC when ethanol made from the sugar cane by-product molasses replaces ethanol made from the whole sugar cane plant.

4.3 | Limitations

The insights into the environmental impacts of using sugar feedstocks for bio-based chemicals that are offered by this study need to be interpreted in the light of a number of limitations. First, consequential modeling relies on our ability to understand and model market mechanisms, and this understanding can be limited. For example, for maize scenarios, oat is the crop with the lowest returns in the United States and, thus, it has previously been assumed to be the marginal crop (Ash et al., 2018). Meanwhile, more recent agricultural development projections identified Russia and Ukraine among the key producers who are expected to increase the production of oats over the next decade (OECD-FAO, 2021). Consistently with those projections, Russia was the marginal supplier of oat in this study. However, these projections naturally do not consider the war in Ukraine, making the consequential LCA results particularly uncertain for maize scenarios. Similarly, the influence of the war on long-term energy mixes in Europe is not obvious (Tollefson, 2022). Finally, consequential LCA results are very sensitive to the intended use of by-products and functions they replace, which triggers the question about which by-product use should be considered as default.

Second, there are uncertainties in damage modeling. Although damage-based characterization factors are

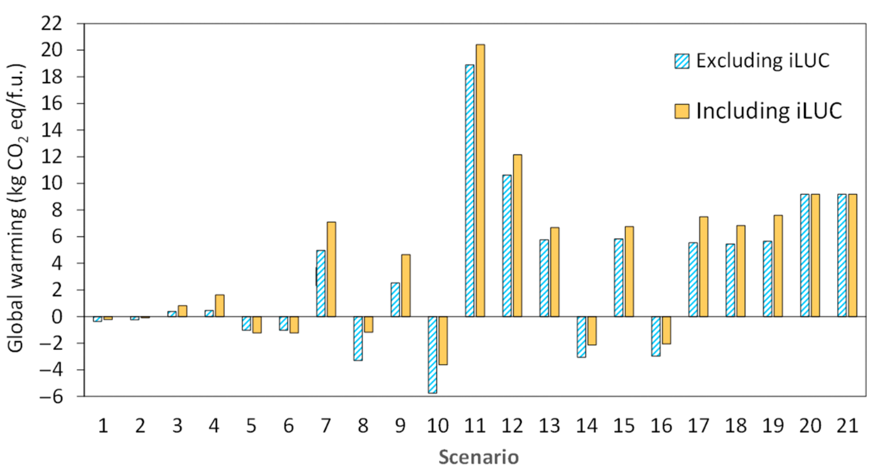

FIGURE 3 Influence of indirect land-use impacts (iLUC) on the ranking of different feedstocks, geographic locations of feedstock production, and use of biomass residues in terms in the midpoint impact category global warming. Impacts are per functional unit (i.e., delivery of monosaccharide syrup for the production of 1 kg MEG at a biochemical plant). Numbering of scenarios is the same as for Table 1.

generally more environmentally relevant compared with normalization and weighing steps of the LCIA phase of the LCA, they are statistically uncertain, and these uncertainties are different for different impact categories (Hauschild & Huijbregts, 2015). This may influence ranking of feedstocks, particularly so when differences in damage scores are caused by those impact categories that are certain, rather than by those categories that do not seem to contribute substantially but are uncertain.

Third, we sought to apply regionalization in the characterization modeling where relevant and where possible and showed that in most cases it did not influence the ranking of feedstocks compared with generic LCIA methods. The land use impact category was, however, not regionalized in this study, which is a limitation. Latest LCIA methodologies offer regionalized approaches for land use (Verones et al., 2020), and it is, therefore, relevant to check whether our conclusions would hold if spatially differentiated characterization factors for land use were used.

Fourth, accounting for climate impacts from land-use change depends on the method chosen and its underlying assumptions. For example, following IPCC recommendations most direct LUC methods, including the method used in the ecoinvent 3.7.1 database that was used in the present study, recommend a 20-year amortization period (i.e., the time horizon over which the direct LUC emissions are linearly distributed for accounting; BSI, 2011; European Commission, 2015; WRI WBCSD, 2011), and this has previously been identified as a limitation (Kløverpris & Mueller, 2012; Schmidt et al., 2015). A practical implication of this limitation is that impacts from direct LUC are accounted for Brazil only because in all other geographic locations the land use change occurred more than 20 years before present.

Finally, the inventory modeling of the pretreatment and conversion of lignocellulosic biomass depends on the modeling of the technology for the sugar extraction step (Bello et al., 2021; Morales et al., 2017; Tao et al., 2014); furthermore, the available technologies are generally less mature compared with sugar extraction from sugar and starchy biomass. This makes the comparison with crop-based feedstock less fair, as technological and environmental learning may improve the performance of the lignocellulosic feedstocks when experience with technology is gained with time (Thomassen et al., 2020).

5 | CONCLUSIONS

Using regionalized LCA and following a consequential modeling framework we showed that the most sustainable sources of sugar, environmentally speaking, are sugar cane and sugar beet. This suggests that these feedstocks should be further evaluated in terms of their economic and social performance to understand better potential societal implications and ultimately support the decision about their large-scale implementation as a source of sugar for the production of biochemicals. The environmental performance of sugar feedstocks in consequential LCA depends largely on (1) the amount of by-products in the agriculture or extraction steps; (2) the intended use of by-products and extent of environmental benefits from replaced functions; and (3) geographic variability in parameters determining life cycle inventories (e.g., agricultural yields or environmental intensity of marginal energy mixes). Consideration of geographical differences and spatial differentiation in the life cycle impact assessment was less influential.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are openly available in Zenodo https://doi.org/10.5281/zenodo.7225167.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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