



SUPPORTING INFORMATION FOR:

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Summary

This Supporting Information S1 provides the description of all input data of the model, the assumptions made and the literature and database references. The environmental data is based on life cycle assessments, with full information on the life cycle inventory and the results of the life cycle impact assessment based on the ReCiPe method. It also includes additional figures and analyses on the results.

Content

1	General data, parameters, and assumptions	S1-3
1.1	Transportation-related data.....	S1-3
1.2	Reference products.....	S1-4
1.3	Feedstock and process-related.....	S1-4
2	Economic parameters.....	S1-7
2.1	Revenue.....	S1-7
2.2	Biorefinery-related cost.....	S1-7
2.3	Feedstock cost.....	S1-8
2.4	Transportation cost	S1-9
3	Environmental parameters	S1-10
3.1	Reference products.....	S1-11
3.2	Biorefinery-related impacts	S1-12
3.3	Feedstock impacts	S1-13
3.4	Transportation impacts	S1-14
4	Modeling.....	S1-15
4.1	Formulation of the MILP model.....	S1-15
4.2	Scenarios.....	S1-17
4.3	Model computation.....	S1-17
4.4	Linearization of transport costs.....	S1-17
4.5	Linearization of capacity dependent parameter	S1-19
5	Results.....	S1-21
5.1	Selection of environmental categories	S1-21
5.2	Opportunity costs	S1-22
5.3	Prices per liter	S1-23
5.4	Sensitivity analysis.....	S1-24
5.5	Pareto optimization	S1-26
	References	S1-27

1 General data, parameters, and assumptions

This section provides the nomenclature in Table S1-1, as well as a detailed description of general data and parameters of the model, their background, the underlying assumptions, calculations, and references in the following sub-sections. Values for these parameters are given in the Supplementary Information S2 (sheet Data 1).

Table S1-1: General data and parameters

Data	Unit	Definition
$\delta_{r,s}$	km	distance between regions $r \in R$ and $s \in R$
ζ_r^{1G}	t	current demand for 1G bioethanol in region $r \in R$
ζ_r^{FF}	t EtOH eq.	current demand for petrol in region $r \in R$ expressed in metric tons EtOH equivalent
$\psi_{r,f}$	t	total available feedstock $f \in F$ in region $r \in R$
$\mu_{r,f}$	t / km ²	specific feedstock potential for feedstock $f \in F$ in region $r \in R$
θ_f	t EtOH / t	transformation factor from feedstock $f \in F$ to bioethanol
θ_p^{byp}	t / t EtOH	co-production factor from bioethanol to by-product $p \in P$
σ_c	t EtOH	capacity of a biorefinery with capacity level $c \in C$
τ_r		regional road network tortuosity factor of region $r \in R$
ρ	km	width of sourcing annuli
v_m		geometric factor for the average distance between the center and annulus $m \in M$

1.1 Transportation-related data

$\delta_{r,s}$: is a distance matrix of all 98 NUTS-1 regions $r, s \in R$ of the EU-28. The distances are aggregated from NUTS-3 to NUTS-1 based on Eurostat (2018a) which gives road distance for each pair of the 1453 regions on NUTS-3 level. The distance between two NUTS-1 regions is calculated as the average distance between each NUTS-3 region of the one NUTS-1 region and all NUTS-3 regions of the other NUTS-1 region. For the calculation of the average distance *within* one region (i.e. the diagonal of the distance matrix), we use the disk line picking method (Weisstein, 2018).

τ_r : represents the tortuosity of the regional road network in NUTS-1 region $r \in R$. The tortuosity factor is calculated as the average road distance between two NUTS-3 regions derived from Eurostat (2018a), divided by the average beeline between two points in the respective NUTS-1 region. We assume each NUTS-1 region to be a circle and calculate the *average beeline* by the disk line picking method (Weisstein, 2018).

$$\tau_r = \frac{\text{average road distance between NUTS3 regions of NUTS1 region}_r}{\text{average beeline}_r} \quad (1)$$

As this calculation is a rough estimation, we set a lower bound of 1.2 (corresponds to a well-developed road network) and an upper bound of 2 (corresponds to a less developed road network).

ρ : is the width of a sourcing annulus for sourcing within a region in km. This auxiliary parameter is required for the stepwise linearization of the non-linear transport costs (see 0) when sourcing from “around” a biorefinery in a region. A low value for ρ means a more accurate resolution of the linearized curve, which, however, entails high computation times of the mixed-integer linear programming (MILP) model. A high value for ρ means a more imprecise resolution of the linearization, but has the advantage of shorter computation times. Experiments showed a reasonable tradeoff between accuracy and computation times for $\rho = 5$ km.

v_m : represents a geometric factor for the average distance between the center and an annulus m , see Eq. (13).

1.2 Reference products

ζ_r^{1G} : is the current demand for first-generation bioethanol (1G EtOH) in each region $r \in R$, and thus the upper bound for the substitution of 1G EtOH by second-generation bioethanol (2G EtOH). The consumption of bioethanol on NUTS-1 level is disaggregated from NUTS-0 level, based on population share. The whole fuel and industrial bioethanol demand is considered and the values are based on the Biofuels Barometer (2019) and ePure (2018).

ζ_r^{FF} : is the current demand for fossil petrol in each region $r \in R$, and thus the upper bound for the substitution of petrol by 2G EtOH. The demand for petrol on NUTS-1 level is disaggregated from NUTS-0 level, based on population share. The NUTS-0 petrol demand is based on the year 2018 (Eurostat, 2019a)

1.3 Feedstock and process-related

$\psi_{r,f}$ and $\mu_{r,f}$: are the total and specific available bioeconomic feedstock potential of feedstock $f \in F$ of each NUTS-1 region $r \in R$ for the year 2018 in metric tons and metric tons/km² respectively. In the 2030 scenario in section 3.3 of the manuscript, these parameters assume the 2030 values. Both the 2018 and the 2030 values are based on Wietschel et al. (2019). According to Wietschel et al. (2019), regional feedstock potentials could theoretically also be negative due to a high demand of other straw applications compared to the supply. However, in the study at hand, the supply is assumed to be zero in these regions.

θ_f : is the transformation factor from feedstock $f \in F$ to bioethanol. As straw is mainly composed of cellulose (30 – 40%), hemicellulose (20 – 30%), and lignin (20%) (see Table S1-2), we assume 35% cellulose and 25% hemicellulose in the dry matter content. The considered feedstocks are wheat straw, maize stover, barley straw, and rapeseed straw. This work assumes the same transformation factor for all considered feedstocks. The transformation efficiencies of cellulose to glucose and hemicellulose to xylose, as well as the fermentation efficiency are assumed to be 85%. These assumptions translate to 0.2211 t ethanol per t straw, which is congruent with literature sources (Clariant, 2018; Santibañez-Aguilar et al., 2011; Wang et al., 2013) (Further information on the process can be found in 3.2).

Table S1-2: Chemical composition of crop residues (common wheat straw, maize stover, barley straw, rapeseed straw)

Material	Average fraction
Cellulose	35% ± 5%
Glucan (C6)	35% ± 5%
Hemicellulose	25% ± 5%
Xylan (C5)	20%
Arabinan (C5)	2%
Galactan (C6)	0.75%
Mannan (C6)	0.41%
Lignin	20%
Acids	4.84%
Extractives	4.78%
Ash	8.59%
C	46.7%
H	5.49%
O	38.4%
N	0.67%
S	0.10%
LHV	17.6 MJ/kg

θ_p^{byp} : is the co-production factor for the by-products $p \in P$ in relation to 1 metric ton of produced bioethanol. The hydrothermal cracking separates cellulose, hemicellulose, and lignin. In the enzymatic hydrolysis, lignin and other unconverted solids are separated from the carbohydrates and can either be combusted for the production of electricity and heat (Gupta & Verma, 2015) or sold for the production of other bio-based products. This study assumes a co-generation of electricity and heat to supply the whole energy demand of the biorefinery and the sale of excess electricity to the regional grid. Lignin is separated in the enzymatic hydrolysis (0.2 kg dry matter (d.m.) lignin per kg straw) and burned in a co-generation plant to produce electricity and heat. The lower heating value of lignin amounts to about 25 MJ/kg (d.m.) and the combustion releases all chemically available energy of lignin. The hot gases are converted in a boiler to steam at an efficiency of 88% (McKendry, 2002). The steam either can directly be used in the production process or further converted into electric energy by a turbo generator (45% efficiency). We assume a co-production of 2.513 kWh electric energy per kg EtOH. 1.5 kWh are consumed by the biorefinery and 1.013 kWh per kg EtOH are available as excess electricity (also see 3.2). This assumption is congruent with Humbird et al. (2011). The excess electricity is sold to the regional grid, where the regional electricity mix of the same function is substituted. Other high added value applications based on residual lignin are assessed and developed currently by different projects, which would also broaden the range of possible co-products of EtOH production (Rehap, 2018).

The second by-product considered is stillage as residual from the fermentation process, which can further be utilized as either animal fodder, fertilizer, or for the production of biogas (Tonneini et al., 2016). We assume biogas production from the by-product stillage. According to Kaparaju et al. (2009), between 10.2% and 17.8% versatile solids (VS) of wheat straw is converted to stillage in bioethanol production. We assume 10.2% VS, as this value was obtained from an industrial EtOH producer from wheat straw. This translates to 1 kg straw stillage from 9.804 kg straw. With an ultimate CH₄ yield of 0.324 ± 0.03 (m³/kg of VS), 30.2592 kg straw, i.e. 6.69 kg EtOH, yield 1 m³ of biogas (Kaparaju et al., 2009). The production of 1 kg EtOH thus yields 0.14947 m³ biogas as by-product (0.033048 m³/kg straw), which we assume to be sold to the regional gas grid. The methane (CH₄) content of the biogas is 61.6% compared to about 95% methane content of high quality natural gas (Kaparaju et al., 2010) and we assume a substitution of the equivalent natural gas in the regional grid. A methane

content of 61.6% in biogas thus translates to a substitution of 0.65 m³ natural gas per 1 m³ biogas. The revenue for selling biogas is derived from the natural gas price by taking the methane content ratio of both gases. The resulting values for environmental and economic parameters on a regional level and the respective modeling can be found in Supporting Information S2.

2 Economic parameters

This section provides the nomenclature in Table S1-3, as well as a detailed description of the economic parameters of the model, their background, the underlying assumptions, calculations, and references in the following sub-sections. Values for the economic parameters are given in the Supporting Information S2 (sheet Data 2).

Table S1-3: Economic parameters

Parameter	Unit	Definition
α_r^{1G}	€ / t	competitive revenues of 2G EtOH in region $r \in R$ for substitution of 1G EtOH
α_r^{FF}	€ / t	competitive revenues of 2G EtOH in region $r \in R$ for substitution of fossil petrol
$\alpha_{r,p}^{byp}$	€ / t	revenues of by-products $p \in P$ in region $r \in R$ when substituting references
κ_c^{depr}	€ / BR	annual costs (depreciated) of installing a biorefinery with capacity level $c \in C$
κ_c^{pers}	€ / BR	annual costs for personnel of a biorefinery with capacity level $c \in C$
κ_c^{oper}	€ / BR	annual costs fix costs of operating a biorefinery with capacity level $c \in C$
κ^{var}	€ / t	variable costs of refining bioethanol at a biorefinery
$\varphi_{r,f}$	€ / t	costs of feedstock $f \in F$ at farms in region $r \in R$
ε_t^{fix}	€ / t	fixed costs of transporting feedstock with transport mode $t \in T$
ε_t^{var}	€ / tkm	variable costs of transporting feedstock with transport mode $t \in T$
η_t^{fix}	€ / t	fixed costs of transporting bioethanol with transport mode $t \in T$
η_t^{var}	€ / tkm	variable costs of transporting bioethanol with transport mode $t \in T$
γ_r^{inv}		investment cost factor in region $r \in R$
γ_r^{lab}		labor cost factor in region $r \in R$
γ_r^{tra}		transport cost factor in region $r \in R$

$\alpha_r^{FF}, \varepsilon_t^{var}, \eta_t^{var}$ vary depending on the tax scenario (see section 2.4 of the manuscript)

2.1 Revenue

α_r^{1G} : is the competitive revenue in region $r \in R$ when second-generation bioethanol (2G EtOH) substitutes first-generation bioethanol (1G EtOH).

α_r^{FF} : is the competitive revenue in region $r \in R$ when second-generation bioethanol replaces fossil petrol. Fuel revenues and excise taxes for fossil petrol and biofuels are derived from European Commission (2020) and are averaged from the beginning of 2018 to July 2019. Petrol prices are very volatile, wherefore this parameter should be treated with caution.

$\alpha_{r,p}^{byp}$: are the revenues of by-products electricity and biogas in region $r \in R$. The prices are derived from Eurostat (2019) and Eurostat (2019c)

2.2 Biorefinery-related cost

κ_c^{depr} : are the annualized investment costs of installing a biorefinery with capacity level $c \in C$. We assume a depreciation time of 20 years and a constant depreciation rate per year. The reference value for Romania is derived from the German Federal Government (2014) and Sunliquid (2019) and then regionalized for every region $r \in R$ by the regionalization factor γ_r^{inv} ,

which is based on the Purchasing Power Parity for gross fixed capital formation derived from (Eurostat, 2020b).

κ_c^{pers} : annual costs for personnel of a biorefinery with capacity level $c \in C$. The number of employees is derived from Sunliquid (2019). The costs (reference value for Germany) are regionalized by the regionalization factor γ_r^{lab} which is based on the Labor Cost Index (compensation of employees plus taxes minus subsidies) for the activity industry (except construction) (Eurostat, 2020a)

κ_c^{oper} : are the annual costs of operating a biorefinery with capacity level $c \in C$. It includes 2% maintenance cost, 0.5% administration cost, 1% insurance cost, and 0.75% for unforeseen expenditures of the total investment cost for a biorefinery with capacity $c \in C$ (German Federal Government, 2012).

The aforementioned parameters κ_c^{depr} , κ_c^{pers} , and κ_c^{oper} include a cost degression with a scaling coefficient λ of 0.8, based on literature values (Lauven et al., 2018; Towler & Sinnott, 2013). The relation between costs and capacity is illustrated by the following equation. For example, doubling the capacity leads to about 1.7411 times the investment costs and construction impacts:

$$Investment\ costs_c = \left(\frac{capacity_c}{capacity_0} \right)^\lambda \times Investment\ costs_0 \quad (2)$$

κ^{var} : are the variable costs (per metric ton) of refining bioethanol at a biorefinery (German Federal Government, 2012).

2.3 Feedstock cost

$\varphi_{r,f}$: are the costs for feedstock $f \in F$ at farms in region $r \in R$. The feedstock-specific nutrient price is calculated based on the specific nutrient mass and the specific nutrient price. Table S1-4 shows nutrient prices in June 2018 and the feedstock-specific content (Harms, 2018). As economic incentive, we assume, according to Harms (2018), a profit margin of 50% based on the nutrient price.

Table S1-4: Feedstock specific nutrient prices

Nutrients	Price (€/kg)	Wheat straw (kg/t)	Maize stover (kg/t)	Barley straw (kg/t)	Rapeseed straw (kg/t)
Nitrogen	0.82	5.00	9.00	5.00	11.00
Phosphorus	0.56	3.00	2.00	3.00	6.00
Potassium*	0.60	7.00	10.00	9.00	12.50
Magnesium	1.19	2.00	2.00	2.00	2.00
Sulfur	0.05	1.80	2.00	1.80	1.80
Humus	0.04	100.00	80.00	100.00	80.00
Sum (€/t)		17.90	21.92	19.20	28.40
50% profit margin (€/t)		8.23	10.09	8.83	12.78

*Potassium specific values are based on the assumption that half of the potassium is washed out, wherefore only half of the specific potassium mass is used in the calculation (Harms, 2018).

The harvesting and baling costs consist of labor and machinery costs for baling straw into square bales (1.2m x 0.9m x 2.4m). We assume country-specific labor costs (Eurostat, 2020b).

Additionally, machinery costs arise for a hauler and the press machine, which we assume to be independent from the sourcing country. As the properties of the different feedstock types are comparable, labor and machinery costs are independent of the feedstock type. The logistics costs again comprise labor and machinery costs. Labor costs for logistics on field and farm are also affected by country-specific labor costs (Eurostat, 2020a). Machinery costs for field-to-farm logistics comprise in-field loading (telescopic handler with trailer), transportation (telescopic handler with trailer; the assumed average travelled distance from field to farm is 3 km) and unloading at the farm (telescopic with pliers for square bales).

2.4 Transportation cost

ε_t^{fix} : are the fixed costs (per metric ton) of transporting feedstock with transport mode $t \in T$, derived from Henrich et al. (2009), and inflation-adjusted to the year 2019. The fixed transportation costs for $t = rail$ include a 30km pendulum tour truck transport.

ε_t^{var} : are the variable costs (per ton kilometer) of transporting feedstock with transport mode $t \in T$. The variable feedstock transportation cost are derived from Henrich et al. (2009) for Germany and inflation-adjusted to the year 2019. The costs are further regionalized by the regionalization factor γ_r^{tra} , which is based on the Purchasing Power Parity index for transportation derived from (Eurostat, 2020b).

η_t^{fix} : are the fixed costs (per metric ton) of transporting bioethanol with transport mode $t \in T$, derived from Henrich et al. (2009), and inflation-adjusted to the year 2019. We assume that the biorefinery has as direct access to the railway system.

η_t^{var} : variable costs (per ton kilometer) of transporting bioethanol with transport mode $t \in T$. The variable ethanol transportation cost are derived from Henrich et al. (2009) and inflation-adjusted to the year 2019.

3 Environmental parameters

This section provides the nomenclature of the 21 environmental categories in Table S1-5, as well as a detailed description of the environmental parameters of the model (Table S1-6), their background, the underlying assumptions, calculations, and references in the following sub-sections. Values for the environmental parameters as well as the Life Cycle Inventory are given in the Supporting Information S2 (sheets Data 3 and Data 4 respectively). The Life Cycle Inventory (LCI), i.e. the inputs and emissions associated with the environmental parameters, is modeled in SimaPro 9, assessing the ecoinvent database version 3.5 (Wernet et al., 2016). For the impact allocation, the procedure "allocation at the point of substitution" (APOS) is chosen. The Life Cycle Impact Assessment (LCIA), i.e. the characterization of the life cycle inventory in terms of impact and damage categories, is carried out using ReCiPe 2016 (H) v1.1 (Huijbregts et al., 2017).

Table S1-5: Environmental midpoint and endpoint categories (set L), according to Huijbregts et al. (2017)

Tag	Impact category / midpoint	Impact unit	Endpoint aggregation	Midpoint-to-endpoint factor
M1	Global warming	kg CO ₂ eq	E1 & E2	9.28E-07 & 2.80E-09
M2	Stratospheric ozone depletion	kg CFC11 eq	E1	5.31E-04
M3	Ionizing radiation	kBq Co-60 eq	E1	8.50E-09
M4	Ozone formation, Human health	kg NO _x eq	E1	9.10E-07
M5	Fine particulate matter formation	kg PM _{2.5} eq	E1	6.29E-04
M6	Ozone formation, Terrestrial ecosystems	kg NO _x eq	E2	1.29E-07
M7	Terrestrial acidification	kg SO ₂ eq	E2	2.12E-07
M8	Freshwater eutrophication	kg P eq	E2	6.71E-07
M9	Marine eutrophication	kg N eq	E2	1.70E-09
M10	Terrestrial ecotoxicity	kg 1,4-DCB	E2	1.14E-11
M11	Freshwater ecotoxicity	kg 1,4-DCB	E2	6.95E-10
M12	Marine ecotoxicity	kg 1,4-DCB	E2	1.05E-10
M13	Human carcinogenic toxicity	kg 1,4-DCB	E1	3.32E-06
M14	Human non-carcinogenic toxicity	kg 1,4-DCB	E1	2.28E-07
M15	Land use	m ² a crop eq	E2	8.88E-09
M16	Mineral resource scarcity	kg Cu eq	E3	2.31E-01
M17	Fossil resource scarcity	kg oil eq	E3	4.57E-01
M18	Water consumption	m ³	E1 & E2	2.22E-06 & 6.04E-13
Tag	Area of protection / endpoint	Damage unit		
E1	Human health	DALY		
E2	Ecosystem quality	Species.yrs		
E3	Resource scarcity	USD2013		

Table S1-6: Environmental parameters

Parameter ($\forall l \in L$)	Unit	Definition
$\tilde{\alpha}_l^{1G}$	impact* / t	saved impact when substituting 1 metric ton of 1G bioethanol
$\tilde{\alpha}_l^{FF}$	impact* / t	saved impact when substituting 1 metric ton of fossil petrol
$\tilde{\alpha}_{l,r,p}^{byp}$	impact* / MJ; impact* / m ³ gas	saved impact when substituting 1 MJ conventional electricity or 1 m ³ conventional gas application by by-product $p \in P$ in region $r \in R$
$\tilde{\omega}_l^{CO_2}$	benefit* / t	benefit from CO ₂ stored in 1 metric ton of 2G EtOH
$\tilde{\omega}_l^{CH_4}$	benefit* / t	benefit from CO ₂ stored in CH ₄ per metric ton of produced 2G EtOH
$\tilde{\kappa}_{l,c}^{depr}$	impact* / BR	annual impacts of installing a biorefinery with capacity level $c \in C$
$\tilde{\kappa}_l^{var}$	impact* / t	variable impacts of refining 1 metric ton of 2G bioethanol at a biorefinery
$\tilde{\phi}_{l,f}$	impact* / t	impacts of 1 metric ton of feedstock $f \in F$ at farm (dry matter)
$\tilde{\varepsilon}_{l,t}^{fix}$	impact* / t	fixed impacts of transporting 1 metric ton of feedstock with transport mode $t \in T$
$\tilde{\varepsilon}_{l,t}^{var}$	impact* / tkm	variable impacts of transporting 1 metric ton of feedstock with transport mode $t \in T$ over 1 km
$\tilde{\eta}_{l,t}^{fix}$	impact* / t	fixed impacts of transporting 1 metric ton of bioethanol with transport mode $t \in T$
$\tilde{\eta}_{l,t}^{var}$	impact* / tkm	variable impacts of transporting 1 metric ton of bioethanol with transport mode $t \in T$ over 1 km

*All environmental parameters exist for each of the 21 environmental categories, $\forall l \in L$, see Table S1-5. The unit of the 'impact' depends on the respective category and the given parameter; for example, for global warming and the parameter $\tilde{\kappa}_l^{var}$, the unit is kg CO₂ equivalents per metric ton of 2G bioethanol, and for human health, it is DALY per metric ton of 2G bioethanol.

3.1 Reference products

$\tilde{\alpha}_l^{1G}$: is the impact of first-generation bioethanol equivalent for the calculation of the benefits through substitution. Based on the EU Agricultural Outlook 2018 (European Commission, 2018), the EU's average ethanol consists of about 24% sugar beet and molasses, 32% wheat grain, and 44% other cereals (mainly maize). First-generation bioethanol is modelled based on this information and data from the ecoinvent database (*Ethanol, without water, in 99.7% solution state, from fermentation, at service station {CH} | market for | APOS, U*).

$\tilde{\alpha}_l^{FF}$: is the impact of the fossil petrol equivalent for the calculation of the benefits through substitution. The LCA is based on the respective ecoinvent process (*Petrol, low-sulfur {Europe without Switzerland} | market for | APOS, U*).

$\tilde{\alpha}_{l,r,p}^{byp}$: is impact of application substituted by by-product $p \in P$ in region $r \in R$. As described in section 1.3, this study considers co-produced electricity and biomethane. Ecoinvent provides Life Cycle Inventories (LCI) for the average electricity mix of each European country. We assume that each MJ of co-produced electricity substitutes one MJ of the regional electricity mix. Ecoinvent provides the LCIs for natural gas for most EU countries and we assume that a co-produced 1 m³ biogas substitutes the energy equivalent of natural gas (0.65 m³). For all missing countries, this study assumes the average European natural gas mix provided by ecoinvent.

$\tilde{\omega}_l^{CO_2}, \tilde{\omega}_l^{CH_4}$: benefit from stored CO₂ in second-generation biofuel and biomethane. To enable a fair comparison between fossil based and bio-based fuels, CO₂ stored in the bio-based fuel (released during combustion) flows negatively into the carbon balance.

Table S1-7 shows the carbon balance of the 2G bioethanol production. We assume the same balance for every type of straw, and values are based on the average feedstock composition. The intake of biogenic carbon takes place during the growth phase of the plant. Carbon dioxide

is released in the two process steps fermentation and combustion. The remaining carbon is assumed to be stored in the bioethanol and its by-product biomethane. To allow a comparison with the two fossil references petrol and natural gas, the biogenic carbon stored in the products is modeled as carbon dioxide 'credit'. While combusting petrol releases carbon that has been locked up for millions of years, burning bioethanol and biomethane emits carbon that is part of the biogenic carbon cycle.

Table S1-7: Biogenic carbon balance of 2G bioethanol production

C biogenic	kg C kg ⁻¹ straw	kg C MJ ⁻¹ EtOH	kg C kg ⁻¹ EtOH
Input			
C in straw	4.00E-01	6.77E-02	1.81E+00
Output			
C stored in EtOH	1.23E-01	1.95E-02	5.21E-01
<i>Co-fermentation</i>			
C-CO ₂ (released CO ₂ in fermentation)	7.24E-02	1.23E-02	3.28E-01
<i>Stillage</i> (upgraded to bio-methane)	6.01E-02	1.02E-02	2.72E-01
<i>Combustion in CHP (total)</i>	1.44E-01	2.44E-02	6.51E-01
C-CH ₄	6.00E-07	1.02E-07	2.71E-06
C-CO	6.00E-06	1.02E-06	2.71E-05
C-non-methane organic compounds	1.20E-05	2.03E-06	5.43E-05
C-CO ₂	1.44E-01	2.44E-02	6.51E-01

3.2 Biorefinery-related impacts

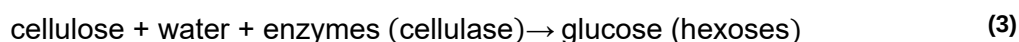
$\tilde{\kappa}_{l,c}^{depr}$: are the annual impacts of installing a biorefinery with capacity level $c \in C$, the affiliated co-generation system, and the affiliated biogas system. The biorefinery is modelled according to Jungbluth et al. (2007). This work assumes a biorefinery operation time of 20 years, wherefore the installing impacts are evenly allocated annually over 20 years. Analogous to the biorefinery costs, the impacts include a "cost degression", using the scale coefficient λ (see 2.2).

$\tilde{\kappa}_l^{var}$: are variable impacts of refining bioethanol at a biorefinery. The following paragraph summarizes the biorefinery production process:

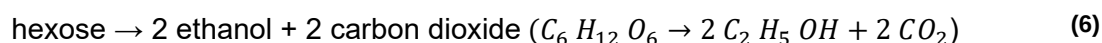
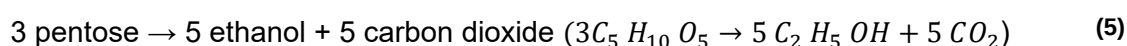
Pretreatment: The cleaning and mechanical shredding of the fibers to between 1 and 3 mm large particles is the first processing step. The energy needed for the shredding is provided by the co-produced electricity. It is followed by the hydrothermal cracking (autohydrolysis) to disrupt lignin and other non-cellulose components to separate cellulose and hemicellulose in subsequent steps (Gupta & Verma, 2015). Most LCA studies on cellulose ethanol still adopt the US Department of Energy's National Renewable Energy Laboratory's (NREL) process dilute acid pretreatment, followed by steam explosion (Gerbrandt et al., 2016). Processes that use acids or bases as catalysts for the pretreatment require a neutralization step, which increases the waste production and the process energy requirements, although research identified pretreatment options for commercial plants with fewer environmental impacts and costs (Gerbrandt et al., 2016). The study at hand assumes chemical free steam explosion pretreatment, which can be considered the state-of-the-art production process in commercial plants (Sunliquid, 2019).

Enzyme production: Enzymes, which are required for the enzymatic hydrolysis, are produced from the pre-treated feedstock in an integrated step of the production process, wherefore the enzymes are optimized to the type of straw. All inputs for the enzyme production are taken from Humbird et al. (2011).

Saccharification: Subsequently, the enzyme mixture hydrolyzes cellulose and hemicellulose chains to form sugar monomers (hexoses and pentoses) in the saccharification process.



Fermentation: Hexose (C6 sugars) and pentose (C5 sugars) are fermented to a fermentation broth and CO₂. The maximum theoretical fermentation efficiency is 0.51 kg ethanol per kg hexose/pentose.



Purification: Ethanol is then recovered from the fermentation broth by micro-sieves to a purity of 99.7 vol.%. The transformation efficiency of cellulose to glucose and hemicellulose to xylose, as well as the fermentation efficiency is assumed to be 85%. These assumptions translate to 0.2211 t ethanol per t straw, which is congruent with literature sources on similar technologies (Clariant, 2018; Santibañez-Aguilar et al., 2011; Wang et al., 2013).

Cogeneration: The energy (electric and thermal) consumed by the biorefinery is produced by the combustion of lignin in a co-generation plant. Emissions from the co-generation to produce process energy are the main sources for environmental impacts.

3.3 Feedstock impacts

$\tilde{\varphi}_{l,f}$: are the impacts of feedstock $f \in F$ at farm. The effect of the agricultural residue removal and the associated environmental impacts is a controversial topic in the scientific literature. Many studies on energy use of agricultural residues ignore the environmental consequences of residue removal (Cherubini & Strømman, 2011). The work of Cherubini and Ulgiati (2010) and Nguyen et al. (2013) investigated the effects of crop residue removal on the soil quality and observed long-range consequences like decreasing yield due to nutrient removal, a change in soil N₂O emissions, and a decrease of soil organic carbon. Also Lal (2005) showed that a complete removal of straw has a negative impact on the soil organic carbon (SOC) and soil erosion. Table S1-8 shows the SOC loss and the resulting CO₂ emissions in case of 100% residue removal (Cherubini & Ulgiati, 2010). Different studies suggest a so called *sustainable removal rate* at which the SOC share is not significantly changed over time due to saturation effects. Sheehan et al. (2004) stated a removal of 40% straw under conventional mulch till and 70% under no-till as sustainable. The bioeconomic feedstock potential applied in this work takes into account the sustainable removal rate with the assumption that a removal of 40% (and 50% for maize) does not affect the SOC (Scarlat et al., 2010; Wietschel et al., 2019).

Table S1-8: Loss of soil organic carbon and its impact on CO₂ emissions.

Material	Residue removal rate	SOC loss	CO ₂ emissions	Source
Cereal straw	100%	0.039 t C/t _{straw}	0.143 t CO ₂ /t _{straw}	Cherubini and Ulgiati, 2010
Wheat straw	40%	-	-	Scarlat et al., 2010
Maize stover	50%	-	-	Scarlat et al., 2010
Barley straw	40%	-	-	Scarlat et al., 2010
Rapeseed straw	40%	-	-	Scarlat et al., 2010

In addition to the SOC balance, straw removal leads to a removal of the nutrients nitrogen, phosphorous and potassium, which entails a decrease in grain yields (Cherubini & Ulgiati, 2010). To account for additional environmental impacts by straw removal, we compare a 100% straw incorporation without additional fertilization with a residue removal of 40 to 50% plus additional fertilization to compensate for the loss of the nutrients nitrogen, phosphorous and potassium. Table S1-9 features the nutrient requirement per removed metric ton of straw.

Table S1-9: Additional fertilizer requirement per removed metric ton of straw (based on Cherubini & Ulgiati, 2010; Harms, 2018)

Material	Wheat straw	Maize stover	Barley straw	Rapeseed straw	Comment
N - CAN*	1.5 kg/t	2.7 kg/t	1.5 kg/t	3.3 kg/t	30% of total N in straw
P – P ₂ O ₅	3.0 kg/t	2.0 kg/t	3.0 kg/t	6.0 kg/t	100% of total P in straw
K – K ₂ O	7 kg/t	10 kg/t	9 kg/t	12.5 kg/t	50% of total K in straw

* calcium ammonium nitrate fertilizer

3.4 Transportation impacts

$\tilde{\varepsilon}_{l,t}^{fix}$: are fixed impacts (per metric ton) of transporting feedstock with transport mode $t \in T$. For $t = tractor$, we do assume no additional fixed impacts, for $t = truck$, we assume a single bale loading. For $t = rail$, we assume a 30 km pendulum tour truck transport and two bale loadings (loading of the transport truck and loading of the train).

$\tilde{\varepsilon}_{l,t}^{var}$: are variable impacts (per ton kilometer) of transporting feedstock with transport mode $t \in T$. For $t = truck$, we assume 50% with EURO5 and 50% EURO6 transport. According to the European average rail system, we assume a share of 44% diesel train and 56% electric train. Tractor, truck, and rail transport is modeled with the respective ecoinvent LCI processes.

$\tilde{\eta}_{l,t}^{fix}$: fixed impacts (per metric ton) of transporting bioethanol with transport mode $t \in T$. The fixed bioethanol transport impacts include all activities needed to offer the bioethanol at the market.

$\tilde{\eta}_{l,t}^{var}$: variable impacts (per ton kilometer) of transporting bioethanol with transport mode $t \in T$. The transportation with $t = truck$ assumes a 32 t truck.

4 Modeling

4.1 Formulation of the MILP model

Table S1-10: Index sets

Index set	Definition	Description
$R = \{1 \dots 98\}$	regions	NUTS-1 level regions in 28 EU member states
$C = \{1 \dots 37\}$	capacity levels	25,000, ..., 3,000,000 metric tons (output), see σ_c (Table S1-1)
$F = \{1 \dots 4\}$	feedstock	wheat straw, maize stover, barley straw, rapeseed straw
$P = \{1 \dots 2\}$	by-products	surplus energy, biomethane
$M = \{1 \dots 100\}$	sourcing annulus	5 km, ..., 500 km
$T = \{1 \dots 3\}$	transport modes	farm tractor, truck, rail
$L = \{1 \dots 21\}$	LCIA categories	ReCiPe 2016 endpoints (E1-3) and midpoints (M4-21), see Table S1-5

Table S1-11: Decision variables

Variable	Domain	Definition
$B_{r,c}$	$\in \{0,1\}$	construction of a biorefinery in region $r \in R$ with capacity level $c \in C$
$F_{r,m,f,t}^{in}$	$\in \mathbb{Q}_0^+$	transported amount of feedstock $f \in F$ within region $r \in R$ from sourcing sector $m \in M$ to biorefinery with transport mode $t \in T$
$F_{r,s,f,t}^{out}$	$\in \mathbb{Q}_0^+$	transported amount of feedstock $f \in F$ from region $r \in R$ to biorefinery in region $s \in R$ with transport mode $t \in T$
$P_{r,s,t}^{1G}$	$\in \mathbb{Q}_0^+$	transported amount of 2G bioethanol from biorefinery in region $r \in R$ to demand in region $s \in R$ with mode $t \in T$, substituting 1G bioethanol
$P_{r,s,t}^{FF}$	$\in \mathbb{Q}_0^+$	transported amount of 2G bioethanol from biorefinery in region $r \in R$ to demand in region $s \in R$ with mode $t \in T$, substituting petrol (fossil fuel, FF)

Table S1-12: Objective functions and decision expressions

Function	Expression	Formulation
$\max O^{econ}$	= revenues (1G)	$= \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} P_{r,s,t}^{1G} \alpha_s^{1G}$
	+ revenues (FF)	$+ \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} P_{r,s,t}^{FF} \alpha_s^{FF}$
	+ revenues (by – products)	$+ \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} \sum_{p \in P} (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) \alpha_{r,p}^{byp} \theta_p^{byp}$
	– refinery costs installation	$- \sum_{r \in R} \sum_{c \in C} B_{r,c} \kappa_c^{depr} \gamma_r^{inv}$
	– refinery costs personnel	$- \sum_{r \in R} \sum_{c \in C} B_{r,c} \kappa_c^{pers} \gamma_r^{lab}$
	– refinery costs others	$- \sum_{r \in R} \sum_{c \in C} B_{r,c} \kappa_c^{oper}$
	– refinery costs process	$- \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) \kappa^{var}$
	– feedstock costs	$- (\sum_{r \in R} \sum_{m \in M} \sum_{f \in F} \sum_{t \in T} F_{r,m,f,t}^{in} \varphi_{r,f} + \sum_{r \in R} \sum_{s \in R} \sum_{f \in F} \sum_{t \in T} F_{r,s,f,t}^{out} \varphi_{r,f})$
	– collection costs fixed	$- (\sum_{r \in R} \sum_{m \in M} \sum_{f \in F} \sum_{t \in T} F_{r,m,f,t}^{in} \varepsilon_t^{fix} + \sum_{r \in R} \sum_{s \in R} \sum_{f \in F} \sum_{t \in T} F_{r,s,f,t}^{out} \varepsilon_t^{fix})$
	– collection costs variable	$- (\sum_{r \in R} \sum_{m \in M} \sum_{f \in F} \sum_{t \in T} 2 F_{r,m,f,t}^{in} \varepsilon_t^{var} \gamma_r^{tra} v_m \rho \tau_r + \sum_{r \in R} \sum_{s \in R} \sum_{f \in F} \sum_{t \in T} 2 F_{r,s,f,t}^{out} \varepsilon_t^{var} \gamma_s^{tra} \delta_{r,s})$
	– distribution costs fixed	$- \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) \eta_t^{fix}$
	– distribution costs variable	$- \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} 2 (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) \eta_t^{var} \delta_{r,s}$
$\max O_l^{env}$	= benefit (1G)	$= \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} P_{r,s,t}^{1G} \tilde{\alpha}_l^{1G}$
	+ benefit (FF)	$+ \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} P_{r,s,t}^{FF} \tilde{\alpha}_l^{FF}$
	+ benefit (by – products)	$+ \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} \sum_{p \in P} (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) \tilde{\alpha}_{l,p}^{byp} \theta_p^{byp}$
	+ benefit (carbon storage)	$+ \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) (\tilde{\omega}_l^{CO2} + \tilde{\omega}_l^{CH4} \theta_2^{byp})$
	– refinery impacts installation	$- \sum_{r \in R} \sum_{c \in C} B_{r,c} \tilde{\kappa}_{l,c}^{depr}$
	– refinery impacts process	$- \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) \tilde{\kappa}_l^{var}$
	– feedstock impacts	$- (\sum_{r \in R} \sum_{m \in M} \sum_{f \in F} \sum_{t \in T} F_{r,m,f,t}^{in} \tilde{\varphi}_{l,r,f} + \sum_{r \in R} \sum_{s \in R} \sum_{f \in F} \sum_{t \in T} F_{r,s,f,t}^{out} \tilde{\varphi}_{l,r,f})$
	– collection impacts fixed	$- (\sum_{r \in R} \sum_{m \in M} \sum_{f \in F} \sum_{t \in T} F_{r,m,f,t}^{in} \tilde{\varepsilon}_{l,t}^{fix} + \sum_{r \in R} \sum_{s \in R} \sum_{f \in F} \sum_{t \in T} F_{r,s,f,t}^{out} \tilde{\varepsilon}_{l,t}^{fix})$
	– collection impacts variable	$- (\sum_{r \in R} \sum_{m \in M} \sum_{f \in F} \sum_{t \in T} 2 F_{r,m,f,t}^{in} \tilde{\varepsilon}_{l,t}^{var} v_m \rho \tau_r + \sum_{r \in R} \sum_{s \in R} \sum_{f \in F} \sum_{t \in T} 2 F_{r,s,f,t}^{out} \tilde{\varepsilon}_{l,t}^{var} \delta_{r,s})$
	– distribution impacts fixed	$- \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) \tilde{\eta}_{l,t}^{fix}$
	– distribution impacts variable	$- \sum_{r \in R} \sum_{s \in R} \sum_{t \in T} 2 (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) \tilde{\eta}_{l,t}^{var} \delta_{r,s}$
$\forall l \in L$		

Table S1-13: Constraints

Constraint	
(1) $\sum_{m \in M} \sum_{f \in F} \sum_{t \in T} F_{s,m,f,t}^{in} \theta_f + \sum_{r \in R} \sum_{f \in F} \sum_{t \in T} F_{r,s,f,t}^{out} \theta_f \geq \sum_{u \in R} \sum_{t \in T} (P_{s,u,t}^{1G} + P_{s,u,t}^{FF})$	$\forall s \in R$
(2) $\sum_{s \in R} \sum_{t \in T} (P_{r,s,t}^{1G} + P_{r,s,t}^{FF}) \leq \sum_{c \in C} B_{r,c} \sigma_c$	$\forall r \in R$
(3) $\sum_{m \in M} \sum_{t \in T} F_{r,m,f,t}^{in} + \sum_{s \in R} \sum_{t \in T} F_{r,s,f,t}^{out} \leq \psi_{r,f}$	$\forall r \in R, f \in F$
(4) $\sum_{t \in T} F_{r,m,f,t}^{in} \leq \pi \rho^2 \mu_{r,f} (2m - 1)$	$\forall r \in R, m \in M, f \in F$
(5) $\sum_{r \in R} \sum_{t \in T} P_{r,s,t}^{1G} \leq \zeta_s^{1G}$	$\forall s \in R$
(6) $\sum_{r \in R} \sum_{t \in T} P_{r,s,t}^{FF} \leq \zeta_s^{FF}$	$\forall s \in R$
(7) $F_{r,r,f,t}^{out} = 0$	$\forall r \in R, f \in F, t \in T$
(8) $P_{r,s,1}^{1G} + P_{r,s,1}^{FF} = 0$	$\forall r \in R, s \in R$
(9) $\sum_{c \in C} B_{r,c} \leq 1$	$\forall r \in R$

(1) Flow preservation input to output, (2) Sufficient BR capacity, (3) Limited feedstock per region, (4) Limited feedstock per segment, (5, 6) Demand is limited, (7) F^{out} is zero for transport within the same region, as this is done by F^{in} , (8) Bioethanol cannot be distributed by farm tractor, (9) Only one BR per region.

4.2 Scenarios

Table S1-14: Scenarios

Scenario	#	Verbal definition	Affected parameters
Tax scenario	T1	Current tax situation per country	α_r^{FF}
	T2	Half (50%) excise tax abatement in every country	α_r^{FF}
	T3	Full (100%) excise tax abatement in every country	α_r^{FF}
	T4	Carbon tax of €50 per metric ton of CO ₂ eq.	$\alpha_r^{FF}, \varepsilon_t^{var}, \eta_t^{var}$
	T5	Carbon tax of €375 per metric ton of CO ₂ eq.	$\alpha_r^{FF}, \varepsilon_t^{var}, \eta_t^{var}$
Feedstock availability (default)	2018	Feedstock potential as determined for 2018 (Wietschel et al., 2019)	$\psi_{r,f}, \mu_{r,f}$
Only for section 3.3 of the manuscript	2030	Feedstock potential as forecasted for 2030 (Wietschel et al., 2019)	$\psi_{r,f}, \mu_{r,f}$

4.3 Model computation

The MILP model is implemented in IBM ILOG CPLEX Optimization Studio 12.8.0.0. The optimization is carried out on an Intel® Core™ i7-2600 CPU with 4 physical cores at 3.40 GHz and 32 GB RAM, and an Intel® Xeon® CPU E5-2690 with 8 cores at 2.90 GHz and 64 GB RAM. The set MIP gap tolerance is 10^{-6} for single-objective optimization (exception: economic T5 runs out of memory at around 10^{-2}). For epsilon-constraint optimization, the gap can be higher to ensure solvability on the used systems.

4.4 Linearization of transport costs

With increasing capacities, biorefineries require a larger catchment area around the biorefinery. The catchment area of a biorefinery is modelled as a circle with the assumption of evenly distributed feedstock within a region. The amount of collectable feedstock grows with the square of the radius. $\psi_{r,f}$ (in metric tons) is the total regional feedstock potential, $\mu_{r,f}$ (in

$\frac{t}{km^2}$) is the specific feedstock potential of feedstock $f \in F$ in region $r \in R$, and A_r is the area of the region r :

$$\mu_{r,f} = \frac{\psi_{r,f}}{A_r} \quad (7)$$

$$\psi_{r,f} = A_r \times \mu_{r,f} = \pi \times \rho^2 \times \mu_{r,f} \quad (8)$$

If the whole feedstock of a region were sourced, the average sourcing distance would be:

$$average\ sourcing\ distance_{r,f}^{circle} = \sqrt{\frac{\psi_{r,f}}{\pi \mu_{r,f}}} \times v \times \tau_r \quad (9)$$

Where τ_r is the regional tortuosity factor, and $v = \frac{2}{3}$ corresponds to the geometric factor for the average distance from the center of a circle to each area point within the circle. Eq. (10) depicts this geometric relationship (in Eq. (10) R and r correspond to *radius*):

$$\frac{\int dA\ r}{\int dA} = \frac{\int_0^R dr\ r \int_0^{2\pi} d\varphi\ r}{\int_0^R dr\ r \int_0^{2\pi} d\varphi\ 1} = \frac{2\pi \int_0^R dr\ r^2}{2\pi \int_0^R dr\ r} = \frac{2}{3}R \quad (10)$$

To calculate the transport costs, the *average sourcing distance* is multiplied by the amount of collected feedstock (decision variable $F_{r,m,f,t}^{in}$), the variable transport cost ε_t^{var} , and the factor 2 to factor in pendulum tours. Equation (11) shows that this relation results in non-linear transport cost with degressive specific transportation costs and progressive total collection costs (Lauven et al., 2018; Wright & Brown, 2007). The term is linearized step-wise in annuli of the same width ρ (step-width = 5 km). Index $m \in M$ is introduced to count the annuli. The total transport costs are calculated as the sum over all annuli of which feedstock is sourced (12).

$$transport\ cost_{r,f,t} = \sqrt{\frac{F_{r,f,t}^{in}}{\pi \mu_{r,f}}} \times v \times \tau_r \times F_{r,f,t}^{in} \times \varepsilon_t^{var} \times 2 \quad (11)$$

$$transport\ cost_{r,f,t} = \sum_{m \in M} v_m \times \rho \times \tau_r \times F_{r,m,f,t}^{in} \times \varepsilon_t^{var} \times 2 \quad (12)$$

Fig. S1-1 exemplifies the step-wise linearization of the transport cost. Transportation distances are assumed to be fixed for feedstock sourcing within one “sourcing annulus”. The incremental annulus width ρ allows the adjustment of the granularity of the linearization, index $m \in M$ counts the annuli and v_m represents the geometric factor for the average distance between the center and the annulus m (in Eq. (13) r correspond to *radius*):

$$v_m = \frac{2\pi \int_{(m-1)\rho}^{m\rho} dr\ r^2}{2\pi \int_{(m-1)\rho}^{m\rho} dr\ r} = \frac{2}{3} \frac{3m^2 - 3m + 1}{2m - 1} \quad (13)$$

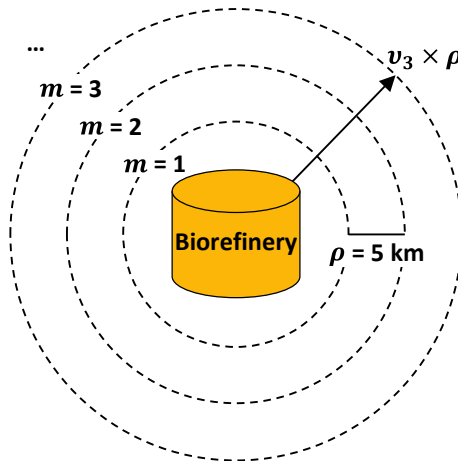


Fig. S1-1: Step-wise linearization of transport cost

The feedstock supply per annulus is limited, which requires one additional constraint. Equation (14) shows the relation between the radius of the whole region, the annulus width ρ , and the maximum number of annuli n . This equation is solved for ψ .

$$\sqrt{\frac{\psi_{r,n,f}}{\pi \mu_{r,f}}} = n \times \rho \quad (14)$$

$$\psi_{r,n,f} = \rho^2 n^2 \pi \mu_{r,f} \quad (15)$$

$$\text{amount per annulus}_{r,m,f} = \rho^2 m^2 \pi \mu_{r,f} - \rho^2 (m-1)^2 \pi \mu_{r,f} = \pi \rho^2 \mu_{r,f} (2m-1) \quad (16)$$

In the next step, this relation is transformed into constraint 4 (see Table S1-13) applied in the model. Decision variable $F_{r,m,f,t}^{in}$ must be smaller than the available feedstock in each annulus.

$$\sum_t T F_{r,m,f,t}^{in} \leq \pi \rho^2 \mu_{r,f} (2m-1) \quad (17)$$

4.5 Linearization of capacity dependent parameter

Due to economies of scale, we assume non-linearly decreasing marginal investment costs and environmental impacts with increasing biorefinery capacity. Towler and Sinnott (2013, page 308) give a range for the scale parameter λ in chemical plants from 0.7 to 0.9, and as cellulose ethanol production requires physical pre-treatment, this work uses 0.8 as scale parameter for the construction-based investment costs and environmental impacts. We assume the same scale parameter for environmental impacts, as scale effects are attributable to a more efficient use of construction resources.

$$\text{Investment costs}_c = \left(\frac{\text{capacity}_c}{\text{capacity}_0} \right)^\lambda \times \text{Investment costs}_0 \quad (18)$$

We then introduced discrete capacity steps $c \in C$. The step-width of capacity levels C strongly affects the model run time. To adequately address the model run time and model precision tradeoff, the step-width is 25,000 t for small capacities (25,000 to 300,000 t), 50,000 t for medium capacities (300,000 t to 1,000,000 t), 100,000 t for high capacities (1,000,000 to 1,500,000 t), and 250,000 t to account for outliers of maximum optimal capacity (1,500,000 to 3,000,000 t). The smallest capacity level matches the capacity of a pilot plant (Clariant, 2018),

the maximum capacity of 3 Mt is twice the optimal biorefinery capacity found by other studies (Lauven et al., 2018).

5 Results

5.1 Selection of environmental categories

Each midpoint contributes to its respective endpoint differently. These contributions are the result of the parameters used (resulting from Life Cycle Inventories and Life Cycle Impact Assessment) and decisions taken in this application case on the one hand, and the aggregation factors between midpoints and endpoints given by the ReCiPe 2016 method (cf. Table S1-5) on the other hand. For example, if in one optimization run, the values of the midpoints mineral resource scarcity (M16) and fossil resource scarcity (M17) are (in the unit of their respective endpoint resource scarcity (E3)) $[-8 \times 10^6]$ and $[3320 \times 10^6]$ USD₂₀₁₃ respectively, the sum of magnitudes of all contributions to E3 is 3328×10^6 USD₂₀₁₃, which means a relative contribution by M16 and M17 of -0.25% and 99.75% respectively to the endpoint value. Fig. S1-2 shows the maximum (over all optimization runs) of magnitudes of relative contribution of each midpoint to its respective endpoint (i.e. to the total of magnitudes of relative contributions). In our opinion, this provides a reasonable estimate of the relevance of each midpoint in this application case, and a set of midpoints that this work should focus on. The main body of this work thus includes only those categories which contribute at least 5% (positively or negatively) to the respective endpoint in any optimization run. These are most notably global warming (M1), particulate matter formation (M5), land use (M15), and fossil resource scarcity (M17), and to a lesser degree terrestrial acidification (M7), human non-carcinogenic toxicity (M14), and water consumption (M18) (see Fig. S1-2). Fossil resource scarcity (M17) is almost identical to its endpoint resource scarcity (E3); thus, only E3 is displayed in the result section of the manuscript.

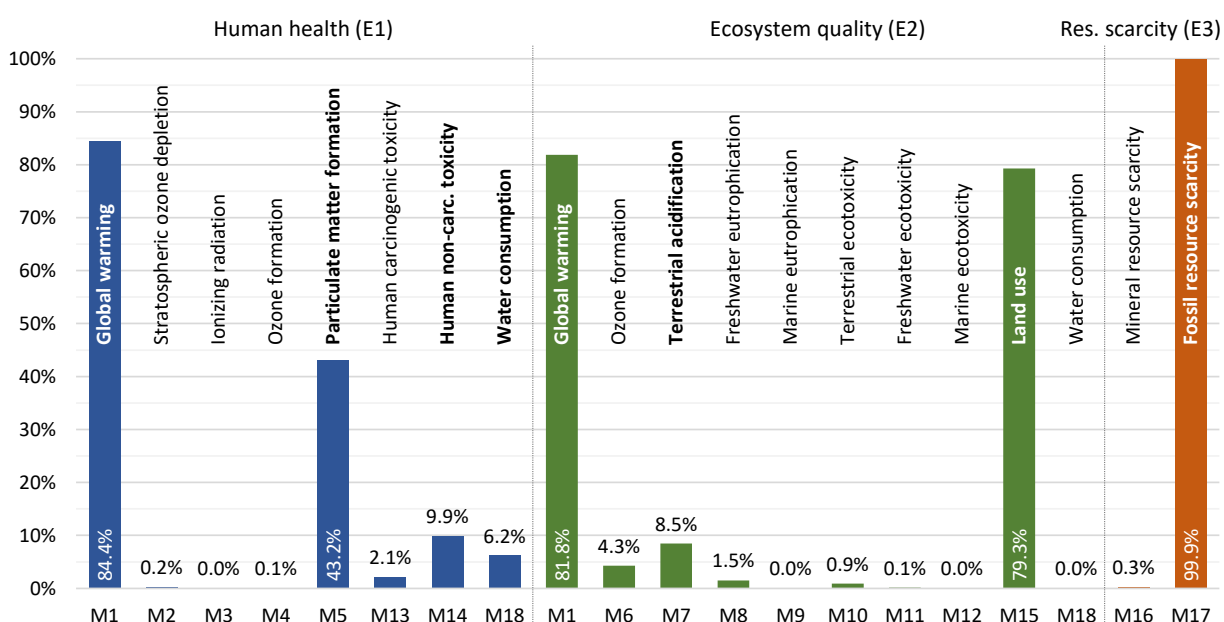


Fig. S1-2: Maximum relative contribution of each midpoint to the respective endpoint (i.e. share of each midpoint to the sum of magnitudes of contributions to the respective endpoint; maximum over all optimization runs). Underlying data used to create this figure can be found in the Supporting Information S2.

5.2 Opportunity costs

Environmental “opportunity costs” refer to the percentual deterioration of one optimal objective value when optimizing another objective. Eq. (19) generically displays the calculation of the percentual opportunity cost between two objectives l and k ($l, k \in L$), with O_l^{env} being the optimal objective value of l , and $V_{l,k}^{env}$ being the value for l when optimizing k . This is then exemplified for the categories human health (E1) and land use (M15) in equations 20-21:

$$\text{opportunity costs}_{l,k} = (V_{l,k}^{env} - O_l^{env}) / O_l^{env}, \forall l, k \in L \quad (19)$$

$$\text{opportunity costs}_{\text{human health, land use}} = (V_{\text{human health, land use}}^{env} - O_{\text{human health}}^{env}) / O_{\text{human health}}^{env} \quad (20)$$

$$\text{opportunity costs}_{\text{human health, land use}} = (1.20E4 \text{ DALY} - 4.95E4 \text{ DALY}) / 4.95E4 \text{ DALY} = -76\% \quad (21)$$

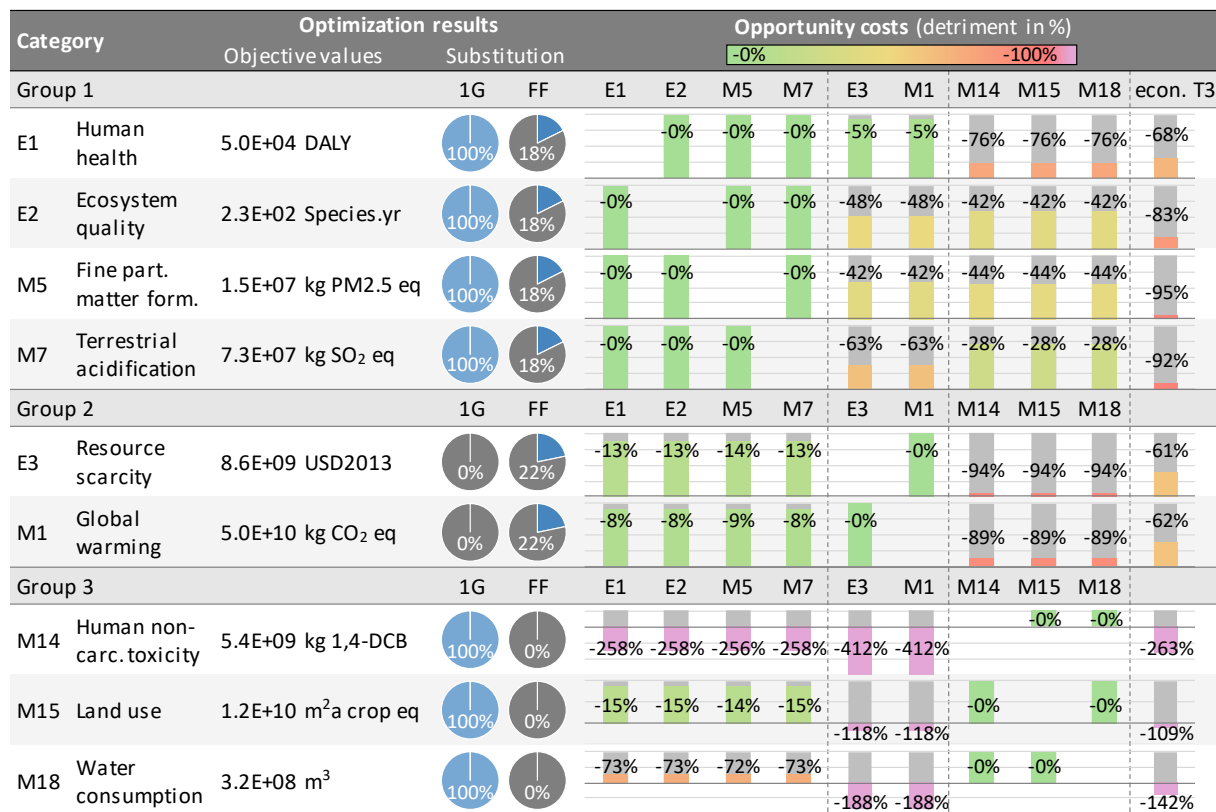


Fig. S1-3: Endpoints and relevant midpoints with their objective value, clustered in groups of mutual congruencies (i.e. with similar substitution decisions, in % of the total demand for 1G/FF), as well as opportunity costs per category for the optimization towards the other environmental categories (= Fig. 5 in the manuscript). Underlying data used to create this figure can be found in the Supporting Information S2.

The midpoints not displayed in Fig. S1-3 can be assigned to the three groups as follows, according to their optimal substitution decisions:

Group 1:

- ionizing radiation (M3)

Group 2:

- fossil resource scarcity (M17)

Group 3:

- stratospheric ozone depletion (M2)

- ozone formation human health (M4)
- ozone formation terrestrial ecosystems (M6) (with high opportunity costs)
- freshwater eutrophication (M8)
- marine eutrophication (M9)
- terrestrial ecotoxicity (M10)
- freshwater ecotoxicity (M11)
- marine ecotoxicity (M12)
- human carcinogenic toxicity (M13)
- mineral resource scarcity (M16)

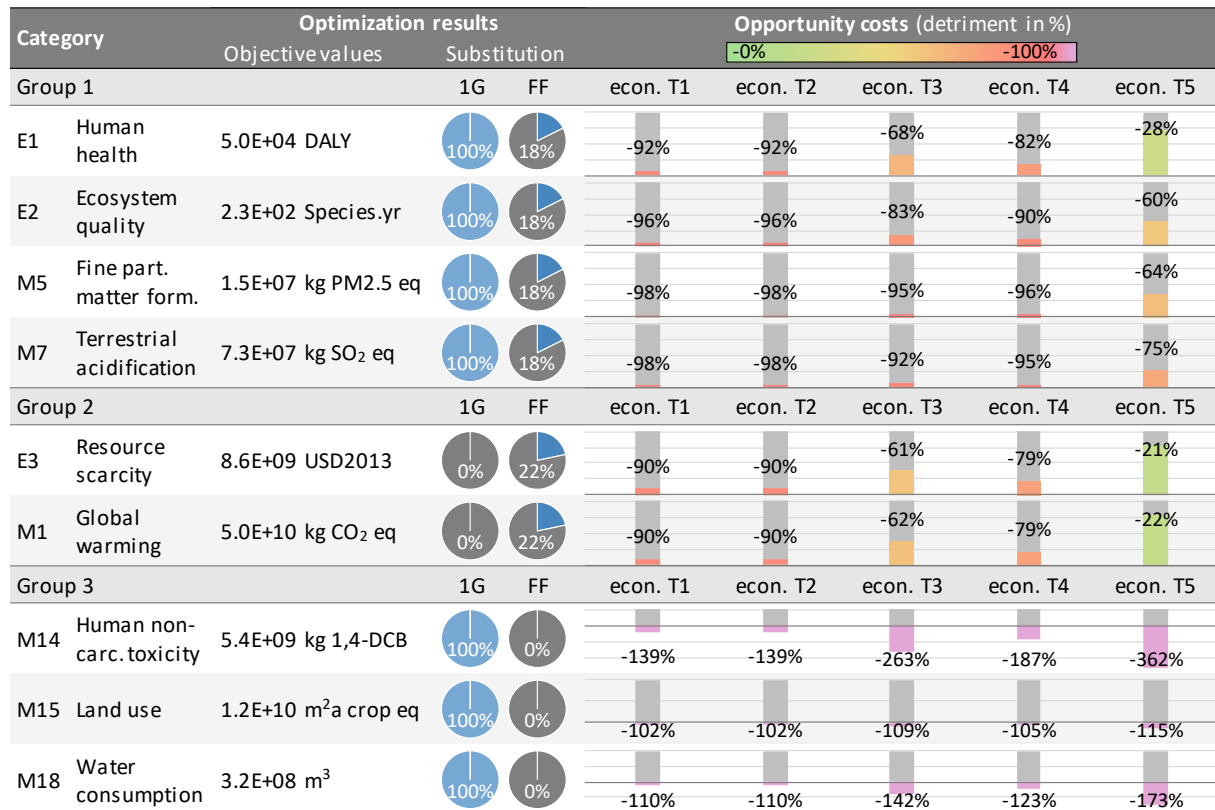


Fig. S1-4: Endpoints and relevant midpoints with their objective value, clustered in groups of mutual congruencies (i.e. with similar substitution decisions, in % of the total demand for 1G/FF), as well as opportunity costs per category for the optimization towards the economic objective function in the five tax scenarios. Underlying data used to create this figure can be found in the Supporting Information S2.

5.3 Prices per liter

Fig. S1-5 depicts the total costs of optimal networks per liter of produced second-generation bioethanol for all objectives and scenarios in ascending order (grey columns). They are to be regarded as averages, as the particular costs per liter *per biorefinery* are subject to deviations due to different costs structures in the different regions. The evaluation shows that within the three congruency groups, the per-liter costs are almost identical, and that the objectives of Group 1 and Group 2 also lead to comparable per-liter costs (0.87-0.88 €/l 2G EtOH).

For comparison, the figure also shows the range of parameter values for competitive prices when substituting fossil petrol (α_r^{FF}) in the five tax scenarios. Above and below the colored bars, the maximum and minimum values respectively with the corresponding country tag are indicated, as well as the median value of the 28 countries (i.e. average between the 14th and 15th highest values). This illustrates, for example, why Sweden is the only country in scenario

T1 (status quo) with high enough competitive revenues to offset the network costs. It also shows that scenario T4 (€50 carbon tax) features a higher range of revenues between the countries and a higher maximum revenue. However, overall profitability is higher in scenario T3 (100% excise tax abatement; cf. Fig. 4 of the manuscript), as the tax abatement has a larger effect on those country in which bioethanol is currently very uncompetitive (e.g. UK), which is indicated by the higher minimum and median values. Scenario T5 (€375 carbon tax) is able to resolve many of the conflicts between the economic and various environmental objectives.

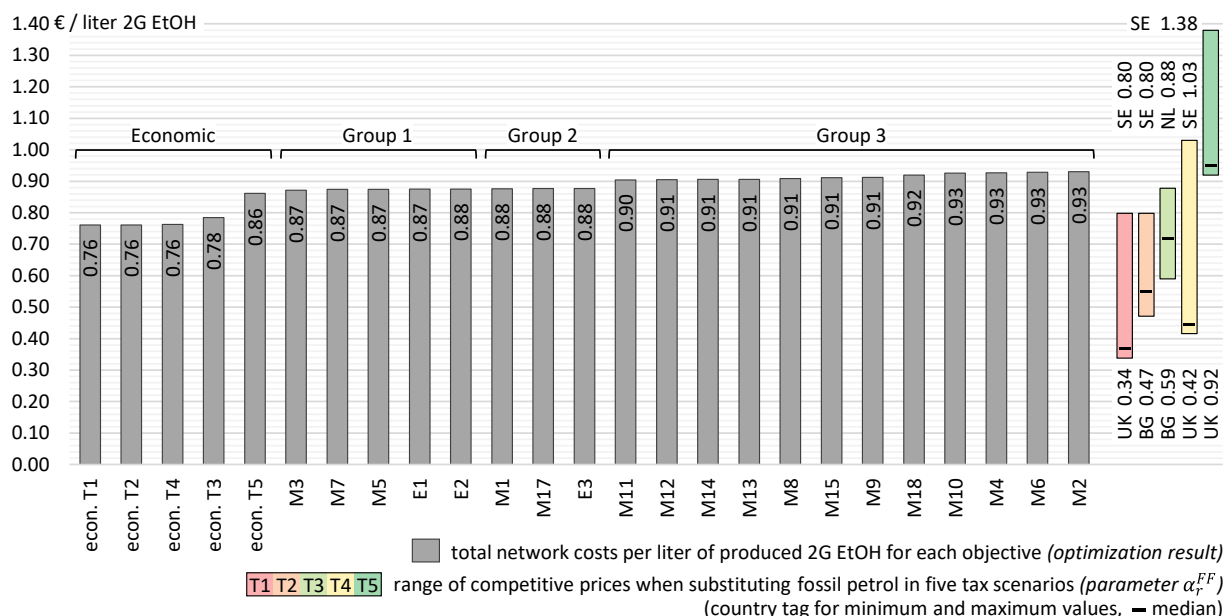


Fig. S1-5: Total network costs per liter of produced second-generation bioethanol for the different objectives in ascending order. Additionally, the figure depicts the range of petrol liter prices (in €/EtOH eq.) in the five tax scenarios with a country tag for the minimum and maximum as well as the median price. Underlying data used to create this figure can be found in the Supporting Information S2.

5.4 Sensitivity analysis

Many of the parameters used in this model are subject to uncertainty in practice. This section analyses the repercussion of variations in key economic and environmental parameters in terms of optimal decisions and objective values. A full Monte Carlo simulation of environmental parameters, which is common practice in LCA studies, and their impact on the outcome of the optimization is not practical due to computational times of the MILP model. An analysis of the contribution of the different revenue/benefit and cost/impact elements towards the respective objective value is given in Fig. S1-6. Feedstock impacts, impacts from production processes, and feedstock transportation impacts are the most relevant elements of the environmental objective functions. For economic objective functions, feedstock costs, biorefinery depreciation, and feedstock transportation are most relevant. Together with the revenue/benefits parameters in each objective function, the model is suspected to be most sensitive towards changes in these parameters, which is why they are chosen for a sensitivity analysis. Moreover, the economic function is suspected to be more sensitive than the environmental ones, as the total revenues compared to the cumulative magnitude of all cost elements ($\triangleq$ return on sales of 4.7% in T1) is much smaller than analogously in the environmental cases.

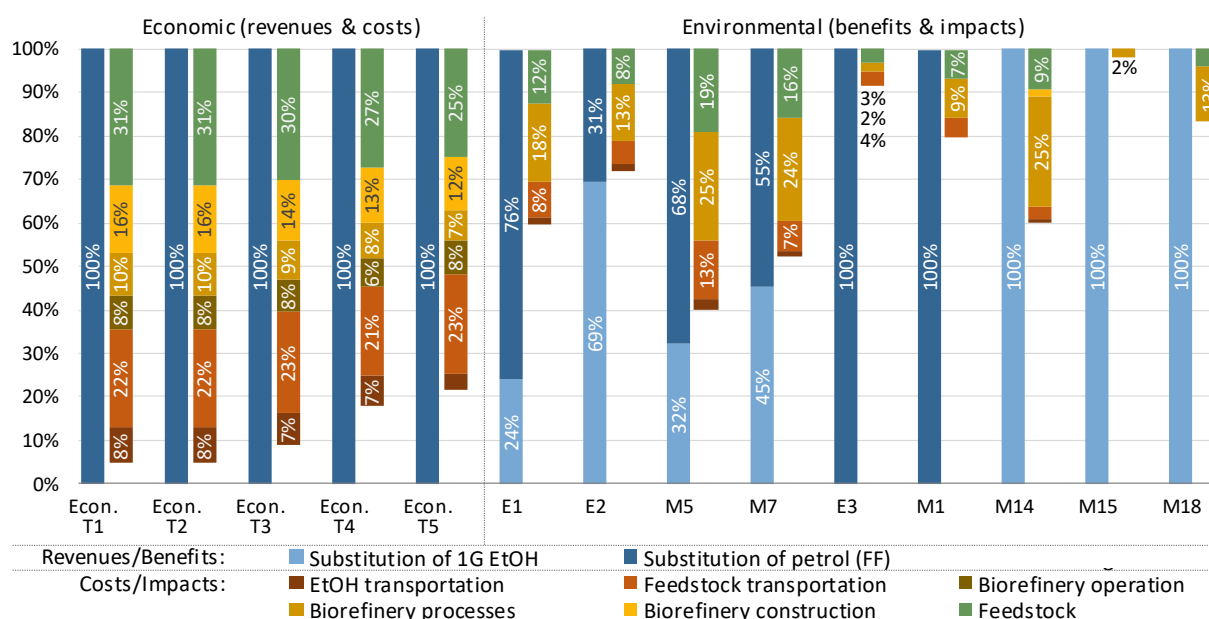


Fig. S1-6: Composition (revenues/benefits vs. costs/impacts) of objective values, and relative contribution of different types of revenues/benefits and costs/impacts. Underlying data used to create this figure can be found in the Supporting Information S2.

Table S1-15 lists the altered economic and environmental parameter changes (by $\pm 10\%$ and $\pm 20\%$) and the resulting changes in objective values and in the number of biorefineries, displayed exemplary for economic (tax scenario T3; tax scenarios T1-T4 behave similarly, only T5 is much more robust) and ecosystem quality (E2) optimization. It shows that economic results are expectedly sensitive towards changes in the revenues, and the whole network turns unprofitable for reductions by 10% or 20%. Vice versa, revenues of +10% and +20% have the potential to increase the objective values by up to +359%. The high economic sensitivity towards the revenue (which is, to a significant degree, beyond the control of a company) could be one reason for why only few market participants have been active in the field of advanced bioethanol production to date. Decreasing feedstock and feedstock transportation cost have a large contribution to the economic objective value as well. A reduction of feedstock cost by 20% increases the network profit by +81%, and a reduction of transportation cost by 20% increases it by 62%, but likewise, increasing feedstock and feedstock transportation cost can diminish the objective value significantly. The environmental results are expectedly much more robust to alteration of parameter values, and the most sensitive parameters are the environmental impacts of the substituted reference products, with an objective value decline of -11% when environmental impacts of 1G EtOH are decreased by 20%.

To account for uncertainties and possible changes in the feedstock availability in the future, we computed the model with the feedstock volumes forecasted on NUTS-1 basis for 2030 (Wietschel et al., 2019). Overall, the 2030 feedstock scenario assumes an increase of feedstock potentials by 12.5% on average, which results in distinctly higher economic objective values of up to 31% (for tax scenario T1, and 27% in T3). The impact of feedstock scenario 2030 with higher feedstock potentials leads to higher environmental objective values of up to 12.4% (for the objectives global warming and resource scarcity). Higher feedstock potentials would slightly reduce conflicts between the economic and most environmental categories. The comprehensive results of the sensitivity and scenario analyses for different objective functions can be found in the Supporting Information S2 (sheets Results 5 and Results 1 respectively).

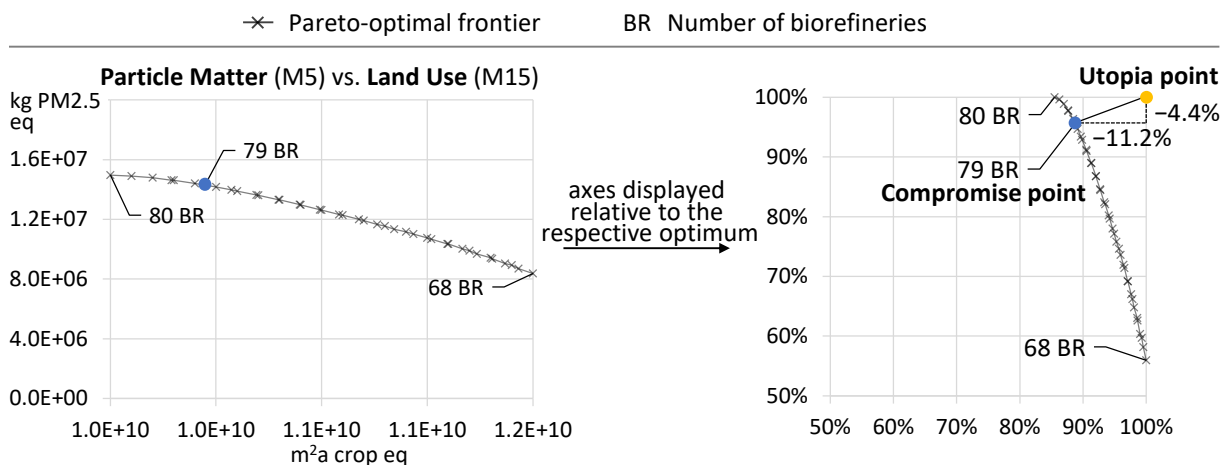
Table S1-15: Minimum and maximum changes in economic and environmental solutions (objective values and number of biorefineries) when altering model parameters by $\pm 20\%$ and $\pm 10\%$

Altered economic parameters								Altered environmental parameters						
Parameter alteration	1) α_r^{1G}	2) α_r^{PF}	3) $\varphi_{r,f}$	4) κ_c^{depr}	5) κ^{var}	6) $\varepsilon_t^{fix}, \varepsilon_t^{var}$	7) $\eta_t^{fix}, \eta_t^{var}$	1) $\tilde{\alpha}_l^{1G}$	2) $\tilde{\alpha}_l^{PF}$	3) $\tilde{\varphi}_{l,f}$	4) $\kappa_{l,c}^{depr}$	5) $\tilde{\kappa}_l^{var}$	6) $\tilde{\varepsilon}_{l,t}^{fix}, \tilde{\varepsilon}_{l,t}^{var}$	7) $\tilde{\eta}_{l,t}^{fix}, \tilde{\eta}_{l,t}^{var}$
Resulting changes in economic (T3) objective value								Resulting changes in environmental (E2) objective value						
-20%	±0%	-100%	+81%	+35%	+21%	+62%	+17%	-11%	-5%	+2%	+0%	+4%	+1%	+1%
-10%	±0%	-84%	+37%	+16%	+10%	+29%	+8%	-5%	-2%	+1%	+0%	+2%	+1%	+0%
+10%	±0%	+141%	-32%	-15%	-10%	-24%	-8%	+5%	+2%	-1%	-0%	-2%	-1%	-0%
+20%	±0%	+359%	-59%	-28%	-20%	-45%	-16%	+11%	+5%	-2%	-0%	-4%	-1%	-1%
Resulting changes of the economic (T3) solution in the number of biorefineries								Resulting changes of the environmental (E2) solution in the number of biorefineries						
-20%	± 0	- 24	+ 10	+ 8	+ 4	+ 4	± 0	± 0	± 0	± 0	± 0	± 0	± 0	± 0
-10%	± 0	- 13	+ 5	+ 6	+ 2	+ 4	± 0	± 0	± 0	± 0	± 0	± 0	± 0	± 0
+10%	± 0	+ 17	- 2	- 4	± 0	± 0	± 0	± 0	± 0	± 0	± 0	± 0	± 0	± 0
+20%	± 0	+ 26	- 4	- 5	- 1	± 0	± 0	± 0	± 0	± 0	± 0	± 0	± 0	± 0

Parameters: 1) revenues/saved impacts of 1G EtOH, 2) revenues/saved impacts of 1G EtOH and fossil petrol, 3) feedstock costs/impacts, 4) construction and depreciation costs/impacts of biorefineries, 5) variable costs/impacts from processes in biorefineries, 6) fixed/variable costs/impacts of feedstock transport, 7) fixed/variable costs/impacts of 2G EtOH transport.

5.5 Pareto optimization

Fig. 7 of the manuscript shows Pareto-optimal frontiers for six pairs of different environmental and economic objective functions. Every point of the curves could be considered optimal, depending on the decision maker's preference. The figure also gives the optimum number of biorefineries at three points of the curves: At the two optima for pure optimization of the two objectives, and at a third point, which is a suggestion for one possible compromise point. This compromise point is defined as the shortest relative Euclidean distance from the so called "Utopia point", i.e. an unreachable point where both objectives are at their optimal values. This is visualized in Fig. S1-5.

**Fig. S1-7: Visualization of an exemplary Pareto-optimal frontier and a theoretical "Compromise point", i.e. point of shortest relative Euclidean distance from the "Utopia point"**

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