ORIGINAL RESEARCH

Lignocellulosic ethanol production combined with CCS—A study of GHG reductions and potential environmental trade-offs

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Abstract
The combination of bioethanol production and carbon capture and storage technologies (BECCS) is considered an indispensable method for the achievement of the targets set by the Paris agreement. In Croatia, a first-of-its-kind biorefinery project is currently underway that aims to integrate a second-generation ethanol plant into an existing fossil refinery. The goal is to replace the fossil fuel production by second-generation ethanol production using miscanthus. In the ethanol fermentation, CO2 is emitted in highly concentrated form and this can be directly compressed, injected and stored in exploited oil reservoirs. This study presents an assessment of the greenhouse gas (GHG) reduction potential of miscanthus ethanol produced in combination with CCS technology, based on data from the planning process of this biorefinery project. The GHG reduction potential is evaluated as part of a full environmental life cycle assessment. This is of particular relevance as a lignocellulosic ethanol industry is currently emerging in the European Union (EU) and LCAs of BECCS systems have, so far, often omitted environmental impacts other than GHG emissions. Overall, the ethanol to be produced in this planned biorefinery project would clearly achieve the EU’s global warming potential (GWP) reduction target for biofuels. Depending on the accounting approach applied for the biological carbon storage, reduction potentials between 104% and 138% relative to the fossil comparator are likely. In addition, ethanol can reduce risks to resource availability. As such, the results generated from data based on the intended biorefinery project support the two major rationales for biofuel use. However, these reductions could come at the expense of human health and ecosystem quality impacts associated with the combustion of lignin and biogas. In order to prevent potential environmental trade-offs, it will be imperative to monitor and manage these emissions from residue combustion, as they represent significant drivers of the overall environmental impacts.

KEYWORDS
BECCS, bioenergy, carbon capture and storage, environmental impacts, ethanol, LCA, miscanthus
1 | INTRODUCTION

The Paris Agreement stipulates that global warming needs to be kept below 1.5°C above preindustrial levels. As the transportation sector is a major contributor to global greenhouse gas (GHG) emissions, new approaches are necessary to mitigate related negative impacts. Biofuels, in particular fuels from lignocellulosic biomass, are considered a valid option for reducing the sector's GHG emissions (Morales et al., 2015). However, due to the shortage of time for action, carbon capture and storage technologies (CCS) are often considered an indispensable complement to other mitigation efforts. The combination of bioethanol production and CCS (BECCS) is seen as a promising approach that could contribute to the removal of carbon dioxide (CO₂) from the atmosphere (Edwards & Celia, 2018). For a number of reasons, including land use aspects and competition with food production, the European Union favours the supply of advanced biofuels from feedstocks such as lignocellulose (Directive (EU), 2018/2001). Miscanthus is a promising lignocellulosic feedstock due to its high productivity and potential to grow on marginal land. Until now, miscanthus has not been used for commercial ethanol production and the environmental performance of miscanthus ethanol has only been analysed based on lab-scale experiments or techno-economic models (Boakye-Boaten et al., 2017; Lask et al., 2019). However, the potential for commercial ethanol production has been recently proven in precommercial refinery trials (e.g. as assessed within the EU-financed demonstration project GRACE [grant agreement no 745012]). In Croatia, a first-of-its-kind biorefinery project is currently under development. The aim is to integrate a miscanthus ethanol plant into an existing fossil oil refinery in close proximity to exploited oil reservoirs, which are suitable for carbon storage.

The fossil fuel production is to be replaced by ethanol production based on miscanthus. The ethanol fermentation generates a stable flow of CO₂ as off-gas. In contrast to CO₂ sources in other BECCS systems (mainly combustion processes), this is emitted in a highly concentrated form and can be directly compressed, injected and stored in the given deposits. The feedstock for the refinery will be cultivated on unused arable land, which is abundantly available in the region surrounding the refinery. Ongoing commercial-scale field trials show the potential of these areas for miscanthus production and provide valuable information on agronomic operations. These practical experiences of feedstock cultivation as well as data from the planning and design phase of the refinery can be used for a representative assessment of the contribution of this low-input feedstock to the sustainable advancement of the transportation sector. This study presents an evaluation of the GHG reduction potential of miscanthus ethanol produced in combination with CCS technology. The GHG reduction potential is evaluated as part of a full environmental life cycle assessment, which compares ethanol’s environmental performance with that of fossil petrol. This is of particular relevance as a lignocellulosic ethanol industry is currently emerging in Europe and LCAs of BECCS systems have, so far, often omitted environmental impacts other than GHG emissions (Gough et al., 2018; Sanchez et al., 2018).

2 | MATERIALS AND METHODS

2.1 | Goal and scope

This study has two objectives. First, it aims to determine the GHG reduction potential of miscanthus ethanol produced in a BECCS system in comparison with a fossil alternative. For this purpose, the GWP per mega joule (MJ) is evaluated and the results are benchmarked with the EU’s RED2 reduction targets for liquid biofuels. In line with the RED2 calculation methodology, impacts from car manufacture and maintenance as well as road construction are not included in these results.

The second objective is the identification of a potential environmental burden shifting when replacing petrol by ethanol. This is achieved through an assessment of both the production of ethanol within a BECCS system and its use in a medium-sized passenger car. It is also performed to provide information on optimization potential in terms of environmental performance. A cradle-to-grave approach is taken and a comparison with petrol, the fossil reference, is included. The analysis is based on a functional unit of 1 km driven in a medium European passenger car (vehicle weight 1600 kg). In contrast to the assessment in line with the RED2 calculation methodology, car manufacture and maintenance as well as road construction and maintenance are included in these results.

2.2 | Methods

The environmental performance is assessed by conducting an LCA according to the ISO standards 14040 and 14044 (ISO 14040, 2006; ISO 14044, 2006), using the life cycle impact assessment method collection ReCiPe2016 v1.1 (Huijbregts et al., 2017). ReCiPe 2016 v1.1 is chosen as it is up-to-date and allows endpoint results to be derived. Results for all endpoints—damage to human health, ecosystems and resource availability—are presented and relevant midpoint indicator results are discussed. Relevant midpoint indicator categories are identified as those which contribute at least 80% of the total impacts at endpoint level. The ecoinvent database v3.5 cut-off is taken for background data and openLCA 1.9 for modelling and impact calculation (Wernet et al., 2016).
2.3 | System boundaries

A Croatian biorefinery project currently under development serves as a case study. The facility is to be incorporated into an existing oil refinery in Sisak. It is designed for an annual biomass intake of 243 kt (15% moisture content) and an expected production output of 55 kt ethanol. The location has two major advantages: First, the refinery will be in close proximity to depleted crude oil reservoirs, which can be used for the storage of CO₂. Experience with enhanced oil recovery ensures the feasibility of carbon sequestration in this area and provides reliable data for the assessment. Second, land for miscanthus cultivation is abundantly available in Croatia. In total, 2,149,080 ha of land can be utilized for agriculture. In 2018, only 69.1% of this area was cultivated, while 30.9% (663,435 ha) remained unused. Large sections of these areas had been used for agricultural production in the past but were abandoned during the war in the 1990s (Tomić, 2020). A substantial proportion of this unused land lies within a 75–100 km radius of Sisak. It has been estimated that around 60,000 ha of unused agricultural land is located in Sisačko–moslavačka, the county surrounding Sisak (Bilandžija et al., 2016). These lands hold substantial potential for the cultivation of energy crops without affecting other agricultural production structures.

Figure 1 presents the system assessed in the study. It includes preparation of the unused agricultural land, miscanthus cultivation, transport to and processing in the nearby refinery in Sisak, capture and sequestration of fermentation off-gas, product distribution and the use phase. A 20 year cultivation period for miscanthus, including the production and transport of inputs as well as agricultural procedures were considered. It was assumed that miscanthus reaches its full yield potential from the third cultivation year onwards. For the second cultivation year, only half the amount was taken.

Input and output data for the processing stage are derived from the precommercial tests using miscanthus as a feedstock and basic refinery engineering. The modelling of the fossil reference system is based on an established dataset from ecoinvent v3.5 (Wernet et al., 2016).

2.4 | Life cycle inventory

2.4.1 | Land clearing

Unused agricultural land for miscanthus production is abundantly available in the region under study, as described in Section 2.3. This land has not been in use for at least 20 years and is densely vegetated with bushes, shrubs and small trees (above-ground biomass ~47 t FM ha⁻¹; 50% DM content; 50% carbon content in dry matter; INA d.d., personal communication, March 2020). For this reason, land clearing and biomass removal is required prior to the miscanthus establishment. 90% of the above-ground biomass is removed for bioenergy production. The remaining 10% decay, releasing carbon. The clearing activities require 130 L diesel per hectare (INA d.d., personal communication, March 2020). The impacts of the diesel consumption and the biomass decay are fully allocated to the miscanthus cultivation. The removed biomass is used for energy production. For this reason, the transportation and combustion impacts are not imputed to the miscanthus cultivation but allocated to the energy production.

2.4.2 | Agricultural system

A rhizome-based miscanthus establishment and cultivation is considered. Required agronomic operations include soil
preparation, establishment, fertilization, harvest and recultivation of the plantation. Harvest includes mowing, swathing, baling and loading of the bales. Data for the harvesting procedures, e.g. quantities and diesel consumption, are derived from commercial miscanthus cultivation. Table 1 presents all procedures with corresponding frequencies over the cultivation period. Fertilizers and herbicides are summed up over the entire period including the establishment phase and divided by the total biomass yield over 20 years. A full biomass yield of 22 t DM ha\(^{-1}\) year\(^{-1}\) is assumed. In addition, lower- (19 t DM ha\(^{-1}\) year\(^{-1}\)) and higher-yielding (25 t DM ha\(^{-1}\) year\(^{-1}\)) scenarios were tested. Experience from commercial miscanthus plantations indicates that approximately 4 t DM ha\(^{-1}\) remain on the field after harvest (Terravesta Ltd, personal communication, March 2020). For this reason, the harvestable biomass yield amounts to 7 (or 5.5/8.5) t DM ha\(^{-1}\) year\(^{-1}\) for the second year and 18 (or 15/21) t DM ha\(^{-1}\) year\(^{-1}\) for the subsequent years. Table 2 presents annual quantities and biomass properties. As nitrogen fertilization is not typically applied in commercial miscanthus plantations, only phosphorus and potassium fertilization was considered in this study. Quantities of phosphorus and potassium fertilizers as well as application of herbicides and other agricultural procedures are based on experiences of commercial-scale miscanthus cultivation (Terravesta Ltd, personal communication, March 2020). All input data are shown in Table 2. Field emissions associated with the miscanthus cultivation are modelled using established models. These are summarized in Table 3. Pesticides are assumed to be released to agricultural soil (Nemecek & Kägi, 2007). As the miscanthus cultivation takes place in a specific region, location-specific characterization factors (CF) for land use are considered as suggested in the ReCiPe2016 methodology (Huijbregts et al., 2017). The global CF for permanent crops is replaced by the ecoregion-specific CF given in Baan et al. (2013). The baled biomass is assumed to be transported from field to refinery by truck over a distance of 40 km.

Miscanthus captures substantial amounts of carbon in the below-ground plant parts. From here on, this is referred to as biological carbon storage and is based on the amount of carbon which can be stored in the below-ground biomass of established Miscanthus × giganteus. An average ratio of 0.52 for the above-/below-ground biomass distribution was derived from the literature (Davey et al., 2017; Kahle et al., 2001; Richter et al., 2015). Assuming a biomass carbon content of 48%, the total below-ground carbon storage was calculated as 17.54, 20.31 and 23.08 t C ha\(^{-1}\), respectively, for a full yield of 19, 22 and 25 t ha\(^{-1}\) (for detailed calculation, see Section 1.1 in Supplementary Material). As the fate of the soil carbon sequestered is uncertain, a conservative accounting approach is applied. In accordance with the ILCD handbook, a temporary carbon storage for the cultivation period is assumed (European Commission, 2010). Following this approach, the carbon is assumed to be completely released after the miscanthus cultivation. However, such flash emissions after miscanthus removal are unlikely (Mangold et al., 2019) and for this reason, indefinite biological carbon storage is considered as a maximal contrast in a sensitivity analysis.

**Table 1** Agricultural procedures during a 20-year miscanthus cultivation period

| Agricultural procedure                          | Number per cultivation period |
|-----------------------------------------------|------------------------------|
| Ploughing—prior to establishment             | 1                            |
| Rotary harrowing                              | 1                            |
| Planting                                      | 1                            |
| Rolling                                       | 1                            |
| Herbicide spraying                            | 5                            |
| Phosphorus and potassium fertilisation        | 20                           |
| Mowing                                        | 19                           |
| Swathing                                      | 19                           |
| Baling                                        | 19                           |
| Bale loading                                  | 19                           |
| Ploughing—final year                          | 1                            |

**Table 2** Main inputs of 20-year miscanthus cultivation period and average properties of harvested biomass

| Inputs/outputs | Quantity | Unit                  |
|----------------|----------|-----------------------|
| Rhizomes       | 15,000   | pieces ha\(^{-1}\)    |
| Phosphorus, in form of triple superphosphate—56 kg P\(_2\)O\(_5\) ha\(^{-1}\) once in 4 years | 14 | kg P\(_2\)O\(_5\) ha\(^{-1}\) year\(^{-1}\) |
| Potassium, in form of potassium chloride—119 kg K\(_2\)O ha\(^{-1}\) each year | 119 | kg K\(_2\)O ha\(^{-1}\) year\(^{-1}\) |
| Herbicides     | 11       | L ha\(^{-1}\)         |
| Dry matter (DM) harvested (full yield)        | 18,000   | kg ha\(^{-1}\)        |
| DM harvested (over cultivation period)        | 16,650   | kg ha\(^{-1}\) year\(^{-1}\) |
| DM content                                          | 85       | %                     |

**Table 3** Considered field emissions and their primary sources with references

| Emission | Source | Reference          |
|----------|--------|--------------------|
| Nitrous oxide (N\(_2\)O) | Harvest residues | Bouwman et al. (2002); IPCC (2019) |
| Phosphorus/Phosphate (P, P\(_2\)O\(_5\)) | P fertiliser | Prasuhn (2006) |
| Heavy metals | Fertilisers/pesticides | Freiermuth (2006) |
2.4.3 | Refinery

Chemical and nutrient inputs are derived from the planning phase of the lignocellulose refinery. A conversion efficiency of 22.6% is assumed for the analysis, meaning that 226 g of ethanol is produced per kilogram of dry miscanthus (INA d.d., personal communication, March 2020). Figure 2 presents an overview of the refinery processes.

Lignin and vinasse are by-products of ethanol production. Vinasse is anaerobically digested and the digestate sent to an on-site wastewater treatment (WWT) plant. Emissions from the WWT plant are estimated based on (Doka, 2009). The biogas is combusted alongside the lignin in an adjacent boiler for heat and power generation, covering the refinery’s entire steam and electricity demand (including the electricity for the WWT as well as for CO₂ compression and transport). Carbon-, nitrogen- and sulphur-related components in the flue gas and ash are modelled in accordance with transfer coefficients for solid waste incineration (Doka, 2013). Flue gas cleaning is necessary to meet the European legal requirements for emissions of NOₓ, SO₂ and particulates (see Table S3). Quicklime is applied for the desulphurization of the flue gas. The resulting calcium sulphate is assumed to be disposed of to landfill. NOₓ reduction is achieved using ammonia as catalyst and particulate matter reduction by electrostatic precipitation. Non-carbon-, nitrogen- and sulphur-related emissions from the residue combustion are modelled using emission factors for wood chip combustion in a cogeneration unit as surrogate (Wernet et al., 2016). Presently, a recast of the European industry emission limits is anticipated. For this reason, lower legal limits were tested in a sensitivity analysis (see Table S3).

2.4.4 | Carbon capture and storage

The fermentation off-gas is highly concentrated CO₂ and is processed without further refinement. It is compressed to 30 bar, liquefied and transported to the injection facility via pipelines. Here, it is further compressed to 190 bar for injection into the former oil wells. Electricity requirements for compression and pumping are taken from real-life data in Croatia (0.30 kWh kg⁻¹ CO₂ for compression and 0.15 kWh kg⁻¹ CO₂ for injection; INA d.d., personal communication, March 2020). The electricity requirements for initial compression and transport are covered by the refinery’s electricity generation. For injection, the Croatian electricity mix is considered (Wernet et al., 2016).

2.4.5 | Use phase

Information on the use phase, including the car manufacture and maintenance, road construction and maintenance as well as emissions from driving are based on established processes from the life cycle inventory database ecoinvent 3.5 with EURO5 standard (Wernet et al., 2016). For carbon dioxide and carbon monoxide, the source of the emissions is changed from fossil to biogenic/non-fossil. It should be noted that car manufacture and maintenance (of car and road) were only included in the full environmental assessment.

2.4.6 | Fossil reference system

The fossil reference system is based on standard ecoinvent processes for crude oil extraction and refining as well as the distribution of petrol and its use in a medium-sized European car with EURO5 standard (Wernet et al., 2016).

3 | RESULTS

3.1 | GWP reduction potential

European legislation defines mandatory GWP reduction targets for biofuels (European Parliament, Council of the
European Union, 2018). For lignocellulosic ethanol production facilities starting operation after January 2021, 65% reductions need to be achieved relative to the fossil fuel comparator (94 g CO₂ eq MJ⁻¹). Figure 3 presents the GWP of miscanthus ethanol from the assessed production for three biomass yield scenarios. In line with the REDII calculation methodology, the results are presented in g CO₂ eq MJ⁻¹ and do not include impacts from car manufacture and maintenance or road construction. For all yield levels, lignocellulosic ethanol from miscanthus exceeded the reduction targets when combined with CO₂ capture and storage (CCS). The conservative assumption of temporary storage of the biologically captured CO₂ led to emission reductions of more than 104% relative to the fossil comparator, i.e. net-negative emissions are achieved. Indefinite storage gave substantially higher emission savings of approximately 138% (Figure 3). Detailed contribution analyses of the GWP results are provided in Figure S1.

3.2 Endpoint indicator results

The following sections present the results for the three endpoints, damage to human health, ecosystem quality and resource availability, calculated for the baseline scenario of driving 1 km in a medium-sized European car running on either ethanol or petrol. A full biomass yield of 19, 22 and 25 t DM ha⁻¹ (corresponding to 15, 18 and 21 t DM ha⁻¹ harvestable biomass), temporary storage of carbon in the below-ground miscanthus plant parts over the cultivation period (17.5, 20.3 and 23.1 t C ha⁻¹) as well as capture and storage of CO₂ from the fermentation off-gas are considered.

3.2.1 Damage to human health

Figure 4 presents impacts on human health associated with driving a medium-sized car over a distance of 1 km. Under the
present assumptions and irrespective of the miscanthus yield, ethanol results in higher human health impacts than the fossil reference. These are dominated by fine particulate matter formation (FPMF), which accounts for 66% of total impacts and mainly stems from residue combustion and associated SO2 and NOx emissions. Further relevant impact categories include human non-carcinogenic toxicity (HNCT), GWP and human carcinogenic toxicity (HCT), contributing 13%, 11% and 8% respectively. Impacts from other categories (water consumption, stratospheric ozone depletion, ionizing radiation) are negligible, as the four categories FPMF, HNCT, GWP and HCT together account for 98% of the human health impacts. Major differences in impact characteristics between ethanol and petrol are observed for GWP and FPMF: GWP impacts are only a quarter of those of petrol, whereas FPMF impacts of ethanol are twice as high and mainly stem from the residue combustion in the refinery. Detailed contribution analyses are provided in Section 3.2.4.

3.2.2 | Damage to ecosystem quality

Ecosystem quality impacts for ethanol are slightly higher (<5%) than for petrol. Figure 5 presents potential ecosystem damage associated with the reference flows. Similar to human health impacts, variations in biomass yield influence the results only slightly. The most relevant midpoint impact categories are terrestrial acidification (TA), land use (LU), GWP and ozone formation (OF), contributing 33%, 24%, 17% and 14% respectively. In sum, these four categories account for 88% of the total ecosystem impacts. Major differences in the impact patterns between the alternatives are found for TA, LU and GWP. While ethanol results in substantially lower GWP, both TA and LU impacts are considerably higher than for the fossil reference. A detailed contribution analysis of the relevant impact categories is given in Section 3.2.4.

3.2.3 | Damage to resource availability

Figure 6 presents the damage to resource availability associated with the reference flows. The results indicate lower resource consumption for ethanol than for petrol. Mineral resource impacts are negligible for both alternatives, with the midpoint indicator fossil resource scarcity accounting for 97% (ethanol) and 99% (petrol) of total impacts. For ethanol, the largest proportion of impacts is associated with refinery chemicals (mainly ammonia) and diesel consumption in the biomass provision.

3.2.4 | Contribution analyses of most relevant impact categories

The most relevant impact categories for human health and ecosystem quality were identified from the results presented in Figures 4 and 5. These include FPMF, GWP, HNCT, LU, OF and TA. Figure 7 presents contribution analyses for each of these midpoint impact categories (for the medium-yield scenario: 22 t DM ha⁻¹ full biomass yield/18 t DM ha⁻¹ harvestable yield).

Five of the six categories, FPMF, GWP, HNCT, OF and TA, are only slightly influenced by impacts from the miscanthus cultivation, with contributions ranging from 14% for to −6.8% for HNCT. Negative values for the HNCT contributions result from the generic heavy metal balance, which indicates an export of heavy metals from the field through the harvested biomass. In contrast to the patterns for these five impact categories, miscanthus cultivation is a major driver of LU impacts, accounting for 27% of the total impacts. Refinery operation contributes to all examined impact categories, with contributions ranging from 12% for HNCT to 51% for LU and 69% for GWP. The LU impacts derive predominantly from the agricultural production of major refinery inputs. Although the
absolute area used for miscanthus cultivation is higher than for the provision of the refinery inputs, the impacts on species losses and ecosystem quality are lower due to the ecoregion-specific characterization factors for permanent crops. The GWP impacts are dominated by the upstream impacts of major refinery inputs as well as emissions from the wastewater treatment. These impacts are however counterbalanced by credits from the CO$_2$ storage, which represents the major individual contributor in the GWP category. Residue combustion and related emissions constitute the major driver in three of the six impact categories. This stage accounts for 50%, 56% and 58% of the impacts for FPMF, OF and TA, respectively, and is predominantly due to the emissions of NO$_x$, PM$_{<2.5}$, NH$_3$ and SO$_2$. Sulphur-containing refinery inputs are the major source of the latter. Car production and maintenance contribute substantially to all impact categories considered except land use. It is a major individual driver of the total impacts for both GWP and HNCT. It should however be emphasized that these impacts are less relevant for the ethanol-petrol comparison, as the absolute impacts per kilometre driven are identical for both alternatives.

3.3 | Accounting for soil carbon sequestration

The endpoint indicator results presented above are based on a conservative accounting approach for soil carbon storage. It assumes a temporary carbon storage, i.e. all the carbon is released at the end of the cultivation period. A sensitivity analysis considers indefinite storage and compares it to the default assumption. The endpoint indicator results for damage to ecosystem quality are presented in Figure 8. These indicate lower impacts for the indefinite-storage scenario. In this scenario, impacts on ecosystem quality are lower than for petrol. Similarly, total human
health impacts of ethanol are reduced but still exceed the ones for petrol (Figure S2).

4 | DISCUSSION

Ethanol production from miscanthus in combination with CCS can contribute substantially to the reduction of GHG emissions in the European transportation sector when replacing petrol. Irrespective of biological carbon storage and yield variations, the reduction targets set by the European Union can be achieved in the given context. Depending on the accounting approach for the biological carbon storage, reduction potentials between 104 and 138% are likely relative to the EU’s fossil fuel comparator. Both accounting approaches taken in this study are extreme alternatives. Carbon stored in the below-ground biomass is neither fully and spontaneously released when the miscanthus cultivation ends, nor is the whole amount stored indefinitely. In practice, a reduction potential in between these worst- and best-case scenarios is to be expected. Whatever the actual case, net-negative emissions are likely.

In addition, it was shown that ethanol use affects resource availability to a smaller extent than petrol. Accordingly, the major rationale for biofuel use—mitigation of climate change impacts and fossil resource dependence—can be effectively achieved. However, it should be noted that these positive effects could potentially be accompanied by environmental trade-offs, as indicated by the endpoint indicator results in Figures 4 and 5. Under the baseline conditions and assumptions, ethanol indicates higher potential impacts on human health and ecosystem quality than the fossil alternative petrol. Given a relative difference of less than 15% (human health impacts) and 5% (ecosystem quality) between the ethanol and the petrol scenarios, these trade-offs are not significant. However, the results clearly emphasize the need to actively assess and manage potential risks. In order to support this, the contribution of the major life cycle stages (primary production, processing, CCS and use phase) and associated uncertainties are discussed below. In line with the study’s objective, optimization potentials are also identified.

4.1 | Feedstock production and supply

The impact contributions of feedstock production and supply, including land clearing and miscanthus cultivation, are relatively small. In addition, the analyses revealed that the endpoint results and related conclusions are consistent for the considered yields ranging between 19 and 25 t DM ha\(^{-1}\) (harvestable yield; 15–21 t DM ha\(^{-1}\)). Irrespective of the yield level, miscanthus cultivation has inherent land use impacts. These constitute a major driver of the impacts on ecosystem quality and are substantially higher than for petrol. This is a trade-off typical for land-based bioenergy production. It should be emphasized that the land use impacts are due to assumptions for potential species losses related to the cultivation of perennial (permanent) crops in Europe. However, biodiversity impacts vary strongly depending on context and location (Elshout et al., 2014). The actual situation in Croatia may not be ideally represented by the applied characterization factors as the area in question is currently experiencing a rapid spread of *Amorpha fruticosa* L., an invasive plant originating from North America (Krpan et al., 2014). Established miscanthus cultivations effectively suppress the growth of weeds. For this reason, large-scale cultivation of miscanthus is considered one option to reduce the proliferation of this invasive species and support local species diversity (Cossel et al., 2019).

As shown in Figure 8, the endpoint results and associated conclusions are strongly influenced by the assumed storage duration. The assumption of indefinite carbon storage...
storage reduces the total human health impacts of ethanol to below the level of petrol. Although an indefinite storage of the whole carbon amount is unlikely, this scenario emphasizes the need to ensure biological carbon sequestration over long periods. Regardless of the accounting approach, it should be noted that a relatively conservative approach was taken for the estimation of the biological carbon storage potential. Only carbon stored in the below-ground biomass was considered and further soil carbon dynamics were neglected. Approximately 1 t C ha$^{-1}$ year$^{-1}$ was assumed to be stored, which is in the lower range of estimates for miscanthus cultivation. Soil carbon sequestration potentials between 0.7 and 2.2 t C ha$^{-1}$ year$^{-1}$ have been previously reported for cultivation on arable land (McCalmont et al., 2017). However, the area intended for the miscanthus cultivation has not been in use for the last 20 years and has had a constant vegetation cover in this period. Thus, a strong soil carbon accumulation exceeding the carbon stored in the below-ground biomass seems questionable. Nevertheless, even without net soil carbon accumulation, ethanol achieves a substantial GHG reduction potential. The cultivation of the area could potentially even result in emissions following the initial soil preparation. Nevertheless, the utilization of this previously unused land has further implications. Bioenergy production is often related to the displacement of existing agricultural production systems. This displacement is referred to as indirect land use change (iLUC) and is associated with substantial environmental impacts (Schmidt, 2015). These effects and additional environmental impacts can be precluded under the present conditions as no replacement of agricultural production occurs.

4.2 Biomass processing and supporting activities

Impacts from biomass processing and supporting activities are mostly related to the combustion of residues (biogas and lignin) and to the refinery operation itself. Impacts from the latter are mainly relevant for GWP and LU and are due to upstream activities and wastewater treatment. Almost a quarter of the GWP impacts are due to the use of chemicals required for the refinery operation. The contribution analysis (Figure 7) and the endpoint results (Figures 4 and 5) show that the combustion of residues and associated emissions are the major source of impacts on the damage level. This applies to the indicators ecosystem quality (via terrestrial acidification) and human health (via fine particulate matter formation). For human health in particular, it should be emphasized that the emissions related to the refinery operations will occur in an area of low population density (Sisak-Moslavina: 32.95 inhabitants km$^{-2}$; Croatian average: 72.03 inhabitants km$^{-2}$; Croatian Bureau of Statistics, 2020) and emissions from residue combustion will be released through a tall flue gas stack. In combination, this may reduce the actual human exposure and corresponding human health impacts. In addition, it should be noted that the assumed amounts of the corresponding emissions, mainly SO$_2$, NO$_x$ and particulate matter, are based on the current European legal limits for gaseous emissions and can be considered worst-case scenarios. In practice, additional emission reduction could be achieved through technical solutions. SO$_2$ and NO$_x$ concentrations in the flue gas could be further reduced by the use of supplementary lime and ammonia. Although these supplements may come at the expense of a few additional impacts, they could improve the overall environmental performance of the ethanol production (see Figures S3 and S4). The additional emission reduction could be tackled by the refinery operator through the management and process design of the plant. It needs to be highlighted that commercialization of lignocellulosic ethanol production has just begun and improvements in biorefining technologies can be expected (Field et al., 2020). In previous sustainability assessments of bioethanol projects, environmental impacts of residue combustion have often been overseen. This is probably due to the fact that GWP is the most commonly investigated impact category (Morales et al., 2015). The results of this study however underline the importance of considering related emissions and impacts in order to optimize the entire design in terms of environmental performance.

4.3 CCS

The significance of the carbon capture and storage stage is underlined by the fact that it substantially reduces the climate change impacts and thus the associated damage to human health and ecosystem quality. For this reason, its feasibility—in technical and economic terms—needs to be considered. An annual refinery output of 55 kt ethanol corresponds to approximately 52 kt of CO$_2$ per year, which amounts to 1560 kt over an assumed refinery lifetime of 30 years. Clearly, this poses the question of whether CCS can be delivered at sufficient scale. The cavities provide adequate volume to store the annual CO$_2$ production for several hundred years (INA d.d., personal communication, March 2020). From an economic perspective, the question is what incentive there is to store CO$_2$ when it can easily be emitted into the air. In this specific case, the infrastructure for carbon storage is already in place and only a few additional expenses are required. A price of CO$_2$-emission certificates of approximately 25 € t$^{-1}$ CO$_2$ would suffice to cover these supplementary costs (INA d.d., personal communication, March 2020). Following this line of argument, CO$_2$ storage could be considered a by-product of the
ethanol production, which would require changes in the allocation approach. However, even assuming an economic allocation based on current prices of fuel ethanol and emission certificates, the ethanol production would achieve the European Union’s reduction targets.

4.4 Use phase

The use phase includes the contributors car production and maintenance as well as road construction and exhaust and non-exhaust emissions. Apart from land use, all relevant impact categories are substantially influenced by this life cycle stage. Major impacts derive from car production and maintenance, while exhaust and non-exhaust emissions are comparatively unimportant. The present study assumed a medium car size (1600 kg). During the previous decade, the average size of passenger cars in Europe has substantially increased (CCFA, 2020). The assumption of a larger car would marginally influence the results in all impact categories. However, it would not affect the comparison between ethanol and petrol, as the same impacts are assumed for the use phase (except for the biogenic and fossil origin of the exhaust emissions).

Overall, we conclude that the ethanol produced within this biorefinery project, which combines lignocellulosic ethanol production and CCS, can clearly achieve the European Union’s GWP reduction target for liquid biofuels. In addition, ethanol can reduce the risks related to resource availability. In order to prevent potential trade-offs with respect to human health and ecosystem quality, it will be imperative to monitor and manage in particular the emissions from residue combustion, which are a significant driver of the overall environmental impacts.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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