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Straw utilization for biofuel production: A consequential assessment of greenhouse gas emissions from bioethanol and biomethane provision with a focus on the time dependency of emissions

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Abstract

The shift from straw incorporation to biofuel production entails emissions from production, changes in soil organic carbon (SOC) and through the provision of (co-)products and entailed displacement effects. This paper analyses changes in greenhouse gas (GHG) emissions arising from the shift from straw incorporation to biomethane and bioethanol production. The biomethane concept comprises comminution, anaerobic digestion and amine washing. It additionally provides an organic fertilizer. Bioethanol production comprises energetic use of lignin, steam explosion, enzymatic hydrolysis and co-fermentation. Additionally, feed is provided. A detailed consequential GHG balance with in-depth focus on the time dependency of emissions is conducted: (a) the change in the atmospheric load of emissions arising from the change in the temporal occurrence of emissions comparing two steady states (before the shift and once a new steady state has established); and (b) the annual change in overall emissions over time starting from the shift are assessed. The shift from straw incorporation to biomethane production results in net changes in GHG emissions of (a) −979 (−436 to −1,654) and (b) −955 (−220 to −1,623) kg CO_2-eq. per $t_{dry\ matter}$ straw converted to biomethane (minimum and maximum). The shift to bioethanol production results in net changes of (a) −409 (−107 to −610) and (b) −361 (57 to −603) kg CO_2-eq. per $t_{dry\ matter}$ straw converted to bioethanol. If the atmospheric load of emissions arising from different timing of emissions is neglected in case (a), the change in GHG emissions differs by up to 54%. Case (b) reveals carbon payback times of 0 (0–49) and 19 (1–100) years in case of biomethane and bioethanol production, respectively. These results demonstrate that the detailed inclusion of temporal aspects into GHG balances is required to get a comprehensive understanding of changes in GHG emissions induced by the introduction of advanced biofuels from agricultural residues.

KEYWORDS
2G biofuels, advanced biofuels, bioethanol, biomethane, consequential LCA, GHG emissions, lignocellulosic biofuels, second-generation biofuels, time dependency of emissions, wheat straw

1 | INTRODUCTION

At present, high efforts are undertaken to reduce the dependency on fossil fuels and thereby to lower anthropogenic greenhouse gas (GHG) emissions. For these purposes, biofuels were promoted as a substitute for fossil energy carriers in the transportation sector in the past decades. Today, the majority of biofuels are made from starch, sugar and oil crops that are also used to fulfill the needs for food and feed. This has led to substantial criticism on biofuels made from these kind of feedstocks (Escobar et al., 2009; Koizumi, 2015). Recently, policy instruments were implemented that facilitate the introduction of the so-called second-generation or advanced biofuels. Among potential feedstocks, cereal straw presents a mostly commercially unused resource that could be used to produce advanced biofuels such as bioethanol or biomethane (Monforti, Bódis, Scarlat, & Dallemand, 2013).

Recently, several consequential life cycle assessment (cLCA) studies on bioenergy, biomethane and bioethanol using wheat straw as feedstock were published. Of these, some do not specifically model soil organic carbon (SOC) changes as a consequence of lignocellulosic feedstock removal (cf. Yesufu et al., 2019; Zucaro, Forte, & Fierro, 2018), some studies use simplistic approaches and assumptions to account for changes in SOC (cf. Buchspies & Kaltschmitt, 2018; O'Keeffe, Franko, Oehmichen, Daniel-Gromke, & Thrän, 2019; Parajuli et al., 2017), while others model the (avoided) decay of lignocellulosic material or the influence of a change in organic matter (OM) supply (cf. Tonini, Hamelin, & Astrup, 2016; Witing et al., 2018). Among these studies, investigations conducted by O'Keeffe et al. (2019) and Witing et al. (2018) show that the production of biomethane and the subsequent application of organic fertilizer can help to increase SOC. None of the available literature addresses the impact of the temporal decay of SOC on GHG balances in the context of agricultural residues other than residues produced in perennial cultivation systems. In contrast, temporal aspects of carbon stock changes have been widely discussed with regard to forest biomass or other perennial cultivation systems. Norton et al. (2019), for example, questioned the capability of forest biomass to mitigate climate change in view of the carbon payback times of forest bioenergy and the time remaining to realize a substantial reduction in (anthropogenic) GHG emissions. While forest biomass and first-generation feedstocks (food, feed and oil crops) have received attention with regard to temporal changes in carbon stocks and the resulting time dependency of emissions (e.g., Eliasson, Svensson, Olsson, & Ågren, 2013; Guest, Bright, Cherubini, & Strømman, 2013; Hammar, Ortiz, Stendahl, Ahlgren, & Hansson, 2015), studies addressing other feedstocks are scarce. Few studies address this aspect for feedstocks other than forest biomass

(Almeida, Degerickx, Achten, & Muys, 2016; Brandão, Kirschbaum, Cowie, & Hjuler, 2019; Daystar, Venditti, & Kelley, 2017). Even fewer studies focus on agricultural residues, for example, straw, for biofuel production with consideration of the time dependency of emissions and the impact on the GHG balance (Pourhashem, Adler, & Spatari, 2016). The inclusion of temporal changes in carbon stocks and the impact on GHG balances is however of crucial importance regarding the present challenge of mitigating climate change in a timely manner. The aim of this paper is therefore to include temporal aspects into an evaluation of potential changes in GHG emissions arising from the shift from straw incorporation to bioenergy production. Two temporal aspects are evaluated:

A The effect of changes in the temporal occurrence of emissions: A shift from a present use of biomass, for example, straw incorporation into soil, to an alternative use, for example, bioenergy production, changes the quantity of C returned to the soil. Aside from the absolute change in C stocks, the timing of emissions changes: if straw is used for biomethane production and arising digestate is returned to the field, C contained in digestate follows different decomposition patterns than C contained in straw. Such a change in the temporal occurrence of GHG emissions changes their presence in the atmosphere. This is relevant with regard to GHG balances that most commonly use the metric global warming potential (GWP). The methodology presented by Petersen, Knudsen, Hermansen, and Halberg (2013) was chosen to account for these aspects. In this study, the methodology is extended to non-CO_2 GHG emissions and applied for the first time in a cLCA/ GHG balance of wheat-straw-based bioenergy provision that evaluates changes in SOC and the effects of returning OM to agricultural land.

B The change in absolute emissions through time: The shift to bioenergy production (or from bioenergy provision to other ways of biomass or land utilization) involves changes in C stocks that change over time. A shift from straw incorporation to bioenergy production changes C stocks over a period of time following the respective shift, for example, the former decomposition of straw ceases while OM returned to the agricultural field, for example, digestate, entails different decomposition rates. The shift to another system might lead to a new equilibrium state or remain changing throughout production cycles, for example, a forest plantation that is harvested and replanted. The establishing of the new equilibrium state takes up to several years and involves a decrease in SOC and thereby an increase in GHG emissions.

These aspects are addressed in a consequential GHG balance and the results are compared with results applying

the methodology provided by the RED Renewable Energy Directive II (RED II; Directive (EU) 2018/2001 of the European Parliament and of the Council of 11 December 2018 on the promotion of the use of energy from renewable sources, 2018). The scope is limited to GHG emissions due to the importance GHG emissions and their time-dependent release regarding climate change (mitigation).

The assessment evaluates the shift from straw incorporation to production of bioenergy by two near-future concepts with capacities of 150 GWh/a biofuel output located in Germany (Figure 1):

- Biomethane is produced by comminuting straw and subsequent anaerobic digestion. Biogas is thereafter upgraded to biomethane by amine washing. Biomethane is subsequently injected into the existing natural gas grid. The produced digestate is dewatered to yield a solid organic fertilizer. Apart from emissions arising from the concept, several displacement effects are assessed (e.g., substitution of natural gas). The organic fertilizer is sold as a product and is thus applied elsewhere to contribute to SOC formation and to provide nutrients. This alters the quantity and quality of OM and nutrients that are applied to the field as well as the temporal occurrence of emissions (in comparison to the previous incorporation of straw).
- Bioethanol is produced from straw via pre-treatment by steam explosion and subsequent fermentation. Lignin is energetically used to provide process energy. The bioethanol concept provides ethanol, a substitute for fossil gasoline, and molasses, a substitute for animal feed.

The RED II will promote the introduction of advanced biofuels and therefore, is likely to promote biofuel production similar to the concepts evaluated. These two concepts exemplarily represent two concepts with differing approaches to handle lignin (and other biomass components): a concept that returns lignin and other components to the agricultural system and another concept that uses lignin for energetic purposes. Anaerobic digestion and fermentative ethanol production with hydrothermal pretreatment recently reached the commercial stage of cellulosic biofuel production. Concepts using these principles and feedstock are either already in operation or under construction in Europe and North America. Compared to other pre-treatment options, mechanical comminution of straw was shown to be the most viable and cost competitive option in comparison to other pre-treatment options (Andersen, Parsin, Lüdtke, & Kaltschmitt, 2020). To account for the variety of environmental conditions and management practices, minimum and maximum reduction scenarios are defined that represent the boundary conditions of technical and biological parameters as well as decay patterns of OM found in literature.

At present, the major share (ca. 85%) of cereal straw in Europe is incorporated into soil while the remaining share is used for animal bedding, animal feed or horticulture (e.g., cover for plants or substrate for mushroom cultivation; Monforti et al., 2013; Thrän, Arendt, Adwiraah, & Kaltschmitt, 2016). Straw used for these purposes is likely to be returned to agricultural land and will thereby contribute to SOC formation. To be eligible to receive direct payments from the EU, farmers need to show that their SOC balance is balanced by applying specific methods (e.g., Ebertseder et al., 2014). According to Weiser et al. (2014), 44% of straw in Germany can be sustainably used for bioenergy production while complying with the SOC balance requirement. It is therefore assumed that (on average) 56% of straw are incorporated into soil to comply with legal requirements (to receive direct payments from the EU). The remainder is assumed to be available for bioenergy production. Any quantity of straw used for bioenergy purposes is not incorporated any longer. Thus, C contained in this share of straw does not follow the decomposition as it did before when it was incorporated into
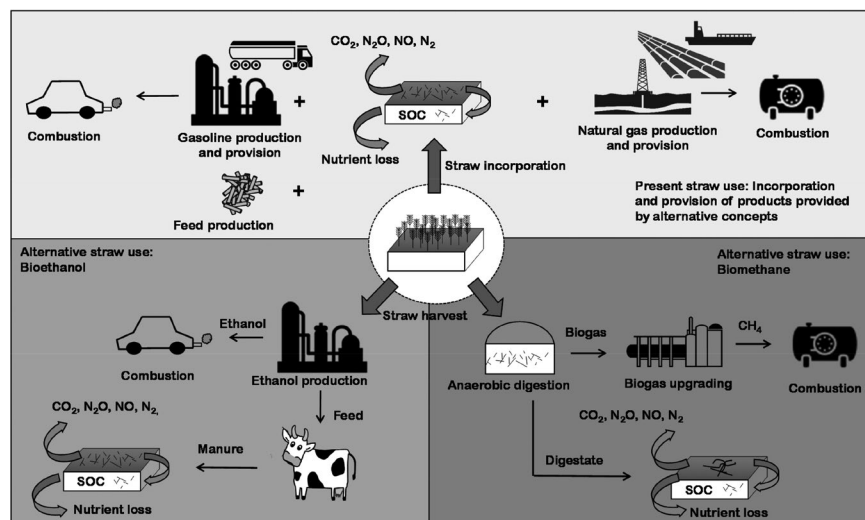
FIGURE 1 Simplified depiction of the analyzed shift from straw incorporation to biomethane and bioethanol production. SOC, soil organic carbon

soil. The remainder that is incorporated still contributes to SOC formation.

2 | METHODS

The change in GHG emissions entailed to the shift from present straw use to bioenergy production is assessed by means of a cLCA and the methodology defined by the RED II.

2.1 | Consequential GHG assessment

The methodological concept of cLCA provides a conceptual framework that seeks to evaluate the impact and implications of a change in production modalities, demand, behavior, etc. (Joint Research Centre [JRC], 2010). In the present paper, the framework is applied to evaluate potential changes in GHG emissions arising from the shift from straw incorporation to its use for biomethane and bioethanol production. In recent years, cLCA became a popular methodology to evaluate political decisions and options (McManus & Taylor, 2015). Yet, there is an ongoing debate on to the extend effects triggered by evaluated changes should and can be fully comprised in cLCA (cf. Brander, 2017; Dale & Kim, 2014; Hertwich, 2014). The present assessment provides an evaluation of potentially arising changes in GHG emissions caused by the fact that a share of straw is used for bioenergy production instead of being incorporated into agricultural soil. The evaluation comprises the following aspects (Figure 1):

- Wheat cultivation: Wheat is cultivated as before the shift to bioenergy production, but a share of straw is not incorporated any longer. A share of straw can be used for bioenergy production while complying with legal requirements regarding the SOC balance (Weiser et al., 2014). This share of straw does not contribute to SOC formation any longer. The change in SOC formation related to this share of straw is evaluated. Additionally, nutrients contained in this share of straw are harvested and need to be replaced. The production of mineral fertilizer, emissions from its application and avoided emissions from straw decomposition are included in the assessment. Additional expenditures to harvest and collect straw are included as well.
- Biofuel production: The production of biofuels, that is, biomethane and bioethanol, increase the demand for energy, auxiliary materials, transportation processes and emits GHG emissions. All emissions arising from the provision of products and services to the biorefinery as well as all GHG emissions emitted by the biorefinery are included.
- Provided products: A substitution approach is applied to account for potential displacement effects (JRC, 2010).

The marginal suppliers of the provided products and services are identified and changes in GHG emissions caused by a change in demand is determined (Section S3). The potential formation of SOC and nutrient release from organic fertilizer are also included.

By consideration of these aspects, the results reflect the potential changes in GHG emissions arising from the decision to use a share of available wheat straw for bioenergy production.

In this study, a time frame of 100 years is considered regarding changes in SOC and the chosen characterization factors (GWP$_{100}$; IPCC, 2013). This choice is justified by the long time (up to several decades) needed until soil reaches a new equilibrium state after changes in management practice. Commonly, the timing of emissions is neglected in most LCA studies and GHG balances assessing annual crops. However, the shift from straw incorporation to bioenergy production results in a change in the temporal occurrence of emissions and in emissions changing over time. For both aspects, the decomposition of OM plays an important role. Straw incorporated into the soil and organic fertilizer applied to soil decompose following a pattern that can be approximated by a two-component exponential decay function (Equation S1; Section S1.1). The decomposition rates of wheat straw and organic fertilizer were derived from field studies and laboratory experiments.

2.1.1 | Temporal aspect A: The change in the temporal occurrence in GHG emissions and the effect on the GHG balance

A certain quantity of C incorporated into the soil in a specific year decomposes over a certain period of time. If this straw is not incorporated but used for bioenergy production instead, C release follows different temporal patterns (e.g., biofuel combustion and differing decomposition patterns if organic fertilizer is returned to the field). This change in the temporal occurrence of emissions is accounted for by the methodology proposed by Petersen et al. (2013). The method is extended to other GHG emissions (Section S1.2). The methodology's rationale is that a GHG emitted remain in the atmosphere for a certain period of time during which it exerts radiative forcing. In the time following its emission, any GHG emitted might be absorbed by sinks (e.g., oceans and vegetation) or transform through chemical or solar-radiative reactions. The fraction of the GHG remaining in the atmosphere can be approximated by decay functions as well (Figure 2; Equations S2 and S5). In case of the instant release at $t = 0$, the time-integrated radiative forcing refers to the 100 years following the emission at $t = 0$. This corresponds to the GWP$_{100}$ of the GHG. The time-integrated radiative forcing of GHG
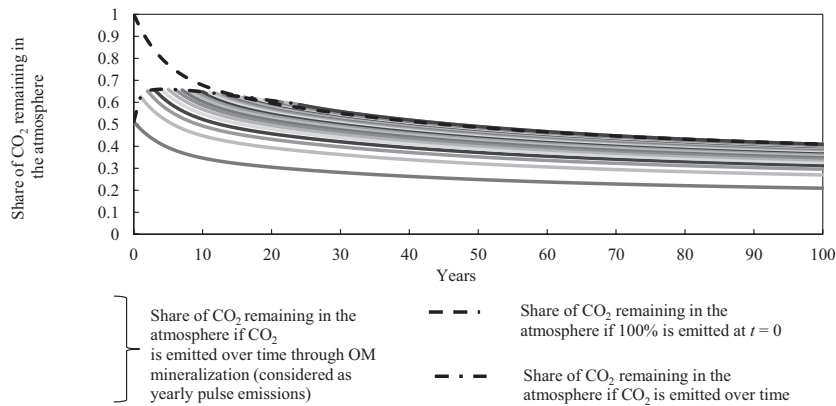
FIGURE 2 Share of CO_2 remaining in the atmosphere as a consequence of a pulse release of CO_2 at $t = 0$ (dashed line) and as a consequence of decomposition of the same quantity of C (only emissions of the first 24 years are shown, dot-dashed line). The dashed line depicts the share of CO_2 remaining in the atmosphere if 100% of CO_2 are emitted at $t = 0$ according to the Bern Carbon Model (Equation S2; Section S1.3). The time-integrated radiative forcing is determined by the integral from 0 to 100. The dot-dashed line presents the sum of CO_2 remaining in the atmosphere if the same quantity of CO_2 was emitted over a longer period of time (e.g., through decomposition, Equation S1). All yearly emissions follow Bern Carbon Model's decay curve. The time-integrated radiative forcing is determined by the sum of integrals of yearly emissions from the year of emission to year 100 (Figure S2). OM, organic matter

emissions emitted at a later point in time at $t = x$ as a consequence of OM application in year $t = 0$ corresponds to an atmospheric load of the emission from $t = x$ to $t = 100$ (Figure 2; Figure S2).

The method is applied to determine the difference between the incorporation of a specific amount of wheat straw and the production of bioenergy from an equal amount of straw. Thus, only the decomposition of that specific amount of straw and the changes in GHG emissions related to bioenergy production and the provision of (co-)products are considered.

2.1.2 | Temporal aspect B: The change in C stocks and GHG emissions over time
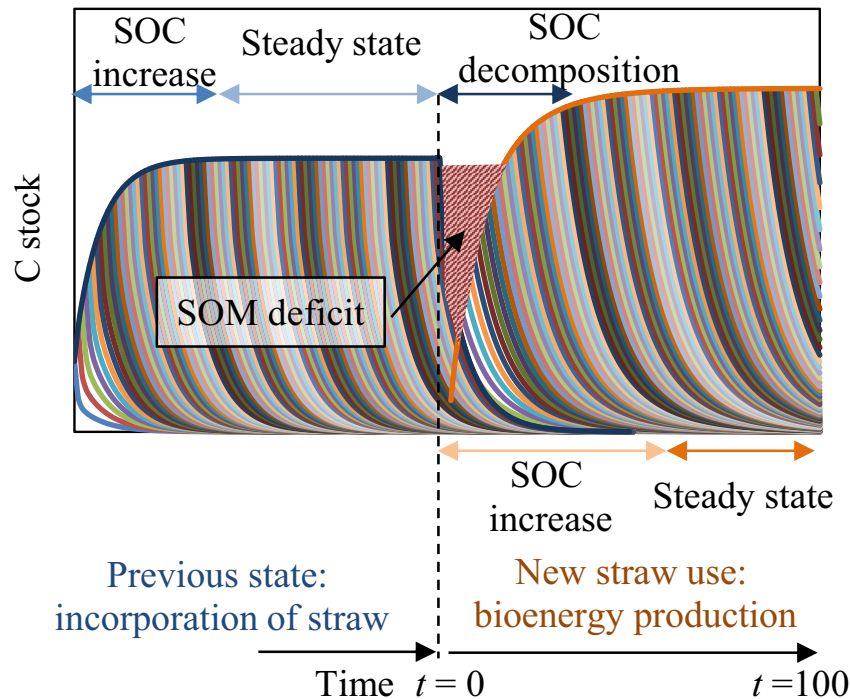
Before the shift to bioenergy production, all straw was incorporated into agricultural soil. The steady supply of OM has led to an equilibrium state in which OM decomposition and OM input were balanced (Figure 3). To produce bioenergy, a share of straw is removed. C contained in this share of straw does not contribute to SOC formation any longer. The decomposition of SOC remaining and SOC formation from organic fertilizer (biomethane concept only) change in time. This aspect is evaluated by calculating yearly GHG emissions following the year in which the shift is conducted. In contrast to temporal aspect A, all GHG emissions in a specific year are considered comprising GHG emissions from the decomposition of OM incorporated into the soil in the preceding years. A carbon payback time is calculated to estimate the time needed to compensate potentially arising decrease in C stocks (Section S1.4).

2.2 | Assessment according to RED II methodology

The methodology defined in the RED II follows a different approach serving the purpose of certification (Directive (EU) 2018/2001 of the European Parliament and of the Council of 11 December 2018 on the promotion of the use of energy from renewable sources, 2018). The methodology allows the estimation of GHG emissions related to the provision of biofuels but does not account for effects induced by the introduction of novel biofuel provision concepts such as displacement effects occurring through the supply of (co-)products, a detailed consideration of changes in SOC and the time dependency of emissions. According to the RED II methodology, average data (e.g., electricity mix instead of marginal electricity supply) are used and emissions are allocated to all products based on their energy content (lower heating value). The results are expressed in *GHG savings*:

$$\text{GHG savings} = \frac{E_f - E_b}{E_f}, \tag{1}$$

where E_f and E_b are the emissions entailed to the production and use of fossil fuel and the respective biofuel. E_f is set to 94 g CO_2 per MJ fuel. E_b comprises GHG emissions entailed to feedstock provision, fuel production, distribution, its use and several possible emission credits, for example, for surplus electricity fed to the public grid, surplus heat that is used elsewhere, improved agricultural management, CO_2 capture and subsequent storage or utilization. All emissions and credits are allocated to all products except those for which credits are given (i.e., captured CO_2, electricity and heat). According to the RED II

FIGURE 3 Schematic depiction of carbon (C) stocks associated with C contained in straw that is used for bioenergy production. The system is in a steady state before the shift to bioenergy production because C input from straw incorporation and C decomposition are balanced. Once, straw is used for bioenergy production, C contained in straw is not supplied any longer. In the given example, the bioenergy concept yields a source of C (i.e., organic fertilizer) that can be applied to agricultural land to contribute to soil organic carbon (SOC) formation. The confining blue and orange curves depict the sum of C remaining from a yearly addition of straw and organic fertilizer, respectively. The colored curves below the blue and orange curves depict decomposition curves of straw and organic fertilizer applied to agricultural lands before and after the evaluated shift, respectively. Once straw supply ceases, straw-related C decomposes while C stocks build up due to the application of organic fertilizer. During that time, a C deficit might arise (depending on organic matter decomposition and application rates). No other emissions (e.g., biofuel combustion, biofuel production, substitution effects, etc.) are shown

methodology, no emissions related to feedstock cultivation are allocated to residues up to the point of collection. The methodology uses GWP of 1, 298 and 25 kg CO_2-eq. per kg CO_2, N_2O and CH_4, respectively. The RED II defines minimum *GHG saving* requirements contained in the RED II. They are complemented by an obligation to monitor and address impacts on soil quality and SOC. For comparison, Equation (1) is applied to results of the consequential approach as well. In this case, E_f and E_b comprise the displaced fossil products (i.e., natural gas and mineral fertilizer as well as gasoline and feed in case of biomethane and bioethanol, respectively) and all changes in GHG emissions related to the biofuel concept, respectively. In this case, no allocation is applied.

2.2.1 | Credits for SOC increases

The RED II allows granting credits if an increase in SOC is proven by measurements conducted in regular intervals. If no other measurement than the first one is available, the change in SOC can be inferred by representative experiments and

soil models. In the present case, empirical data from incubation experiments are available in case of organic fertilizer made from digestate. Due to the lack of long-term field data, results including and excluding SOC credits are provided. Furthermore, results are provided, if the credit is allocated to grain and straw.

3 | SYSTEM DESCRIPTION

The use of straw as feedstock for bioenergy production requires additional operations to harvest, collect and handle straw before it is further processed. Additionally, extracted nutrients need to be replaced and former activities to incorporate straw are not required any longer. All other cultivation practices remain unchanged and are thus not considered by means of the consequential approach. The change in SOC evaluated refers to the SOC formation of the share of straw that is used for bioenergy production instead of being incorporated. The share that is still incorporated (56%; Section 1), still contributes to SOC formation. The composition of wheat

straw produced organic fertilizer (biomethane concept) and molasses (bioethanol concept) plays an important role concerning SOC formation and nutrient cycling (Table S6). For most important parameters and decay functions, average, minimum and maximum values are applied to a base case, a minimum and a maximum reduction scenario (Section S2). The minimum and maximum reduction scenarios provide best and worst-case scenarios providing an evaluation of the range of potential changes in GHG emissions and *GHG savings*. They therefore reflect a broad range of potential environmental conditions and management practices. The provision of products triggers displacement effect that are accounted for by evaluating the GHG intensities of marginal supply (Section S3).

3.1 | Alternative straw use: Biomethane production

Due to the fact that the major share of straw is presently incorporated into soil, it is assumed that the energetic use of straw displaces the incorporation of straw. At first, straw is harvested (besides grain) with a combine harvester (Figure 4a). Subsequently, straw is baled and transported by tractors to a storage facility where it is covered in foil and stored. Later on, straw is transported by heavy duty vehicles to a central storage facility located nearby the biomethane production facility. Heat and electricity are supplied by biogas combustion (thermal efficiency of 90%) and from the

public electricity grid, respectively. Before anaerobic digestion, straw is comminuted by a hammer mill. Thereafter, anaerobic digestion is accomplished in steel tanks, as currently applied in the only commercially operating facility producing biomethane from straw in Germany. The average biogas yield determined by a literature review is 9.1 MJ biomethane per kg_{DM} (kg dry matter) straw digested (Table S7). To upgrade biogas to be fed into the existing natural gas grid, biogas is first de-sulfurized using activated carbon and then purified by amine washing with monoethanolamine, reaching a purity of 99.9% (Miltner, Makaruk, & Harasek, 2017). An additional liquefaction of arising CO_2 is assessed. Arising digestate is dewatered with a screw-press to yield a solid fertilizer (22% DM). The fertilizer is stored in an enclosed facility. Due to the uncertainty regarding the location where the fertilizer is applied, similar transport distances as for straw are assumed. The detailed inventory can be found in Section S4.4.

3.2 | Alternative straw use: Bioethanol production

The assessed bioethanol production concept comprises straw harvest and transportation (as described above), pretreatment, ethanol production and distillation (Figure 4b). After the harvest, straw is pre-treated by steam explosion (210°C, 5 min) to yield a liquid stream that mainly consists of xylose and a solid stream that mainly consists of glucan and lignin (Table S8). Due
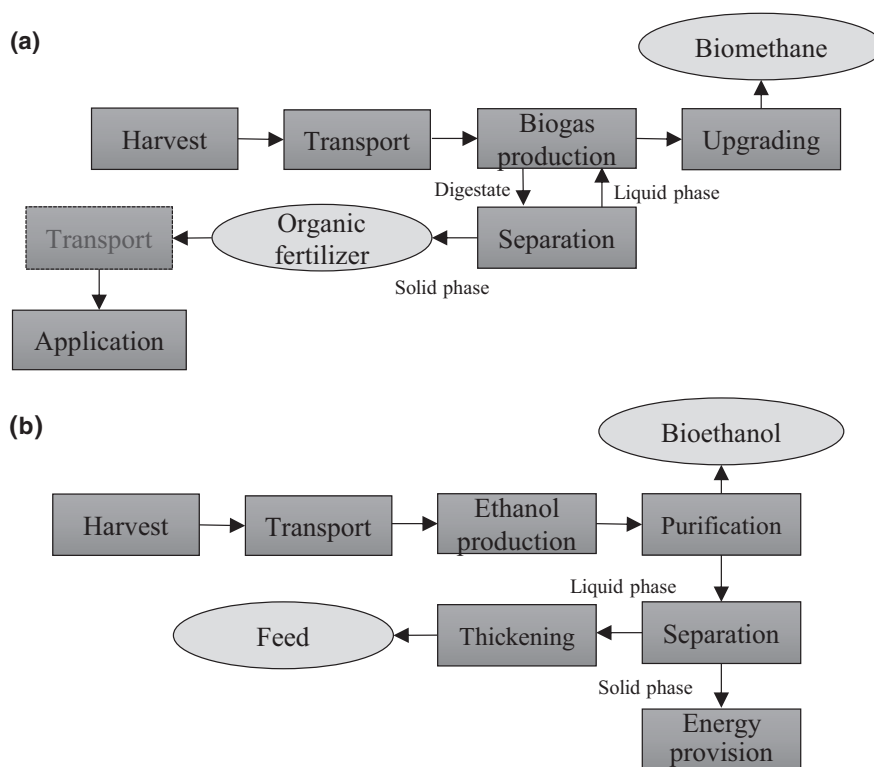
FIGURE 4 Schematic depiction of the analyzed biogas (a) and bioethanol (b) production chain. The organic fertilizer is sold as a product and is thus not (necessarily) applied to the same field

to the severe pre-treatment conditions inhibitors, such as furfural, 5-hydroxymethylfurfural, acetic acid and formic acid are formed (Almeida et al., 2007). The liquid and the solid stream are detoxified by resin-wafer electro-deionization and washing, respectively (Dutta, Dowe, Ibsen, Schell, & Aden, 2010; Gurram, Datta, Lin, Snyder, & Menkhaus, 2011; Rajan & Carrier, 2014). Subsequently, oligomeric sugars are enzymatically hydrolyzed with efficiencies of 81.8% and 63.3%, in case of glucan and xylan, respectively (Alvira, Negro, Ballesteros, González, & Ballesteros, 2016). In the subsequent fermentation process, glucose and xylose are fermented with efficiencies of 95% and 93% of the theoretical maximum, respectively (Tao et al., 2014; Vicari et al., 2012). After fermentation, ethanol is separated by distillation, rectification and molecular sieves. The remaining stream is separated into a solid phase mainly consisting of lignin and a liquid stream containing unfermented sugars. The liquid stream is dewatered to a DM content of 60% and can be used as animal feed (molasses). It is assumed that excreta are applied to agricultural fields (Section S4.5.1). The solid stream is combusted to provide process energy (heat and electricity) by a combined heat and power plant with a total energy conversion efficiency of 0.8. The power-to-heat ratio was determined in a way that the thermal energy demand is met by the combustion of lignin. All surplus capacity is used to generate electricity. As for biomethane production, an optional liquefaction of arising CO_2 is considered.

4 | RESULTS

All results reflect changes induced by the shift from straw incorporation to biofuel production. Only 44% of straw are available for bioenergy production. Thus, the remainder still contributes to SOC formation. Changes in SOC presented refer to these 44% of straw that are removed for bioenergy production. A sensitivity analysis addresses the influence of crucial assumptions and important parameters (Section S5.2). The minimum and maximum reduction scenarios reflect the potential range in changes in emissions due to different environmental conditions, management practices and technical parameters (Section S2).

4.1 | Temporal aspect A: Steady state and the impact of the temporal pattern of emissions

Converting straw to bioenergy instead of incorporating it into agricultural soil changes the temporal occurrence of emissions aside from the absolute change in GHG emissions (Section 2.1.1). Results are additionally presented without the inclusion of the temporal change in emissions by applying the GWP$_{100}$ to all emissions. Unless otherwise stated, all presented values refer to one t$_{DM}$ straw that is converted to

biomethane or bioethanol instead of being incorporated into the agricultural soil.

4.1.1 | Biomethane production

The shift from current straw use (incorporation) to biomethane production potentially reduces the GHG emissions by 979 kg CO_2-eq., including the temporal pattern of GHG release and the time-integrated atmospheric load of GHG emissions following 100 years after the emission (Figure 5). The decrease in GHG emissions amounts to 998 kg CO_2-eq. without consideration of the time-integrated atmospheric load of GHG emissions (net change in absolute emission quantities). The change in GHG emissions per t$_{DM}$ straw converted comprises:

- 30 kg CO_2-eq. emissions arising from the overall process of biomethane production from straw harvest to the delivery of the final products (i.e., biomethane and organic fertilizer; Figure 5b). Of these, straw harvest and transportation, biogas upgrading, biomethane production (including pretreatment) account for 37%, 36% and 27% of the GHG emissions, respectively.
- 18 CO_2-eq. emissions arising from the transportation of organic fertilizer.
- A net decrease of 458 kg CO_2-eq. (including the consideration of the time-integrated atmospheric load of emissions) arising from the change in carbon utilization (from straw mineralization to biomethane production and its use, fertilizer application and its decomposition). If the time-integrated atmospheric load of GHG emissions is neglected, the emission reduction due to changes in SOC amounts to 485 kg CO_2-eq. This value corresponds to a SOC increase of 261 and 277 kg C per t$_{DM}$ of fertilizer with consideration of the time-integrated atmospheric load and without consideration of it. The inclusion of the time-integrated atmospheric load of CO_2 results in a lower net GHG emissions decrease (−458 vs. −485 kg CO_2) because biogenic CO_2 emissions emitted during production (e.g., emitted in the upgrading step or from combustion of CH_4 for internal heat provision) and using biomethane (i.e., combustion) are released at the time of production or shortly thereafter. In contrast, C mineralizes slower if straw is incorporated. Thus, the time-integrated atmospheric load of the emissions emitted during production or shortly thereafter is higher than that of mineralizing C. This results in an increase in GHG emissions relative to straw incorporation if the atmospheric load is considered. This increase outweighs the effect of the time-integrated atmospheric load of emissions released by the slower decomposition of organic fertilizer in comparison to straw decomposition.

FIGURE 5 Change in greenhouse gas (GHG) emissions as a consequence of the shift from straw incorporation into soil to biomethane production (a). Results are presented per t_{DM} straw that is converted to biomethane. Bars include the time-integrated atmospheric load of GHG emissions. Only fossil GHG emissions and changes in biogenic GHG emissions arising from the shift are shown. Organic fertilizer (produced from digestate) includes the substitution of mineral fertilizer, emissions from fertilizer application (C mineralization and N-based emissions) as well as the transport and application of the fertilizer. Natural gas substitution comprises the provision and combustion of natural gas. Biogas provision in part (a) is the sum of all process steps presented in part (b)
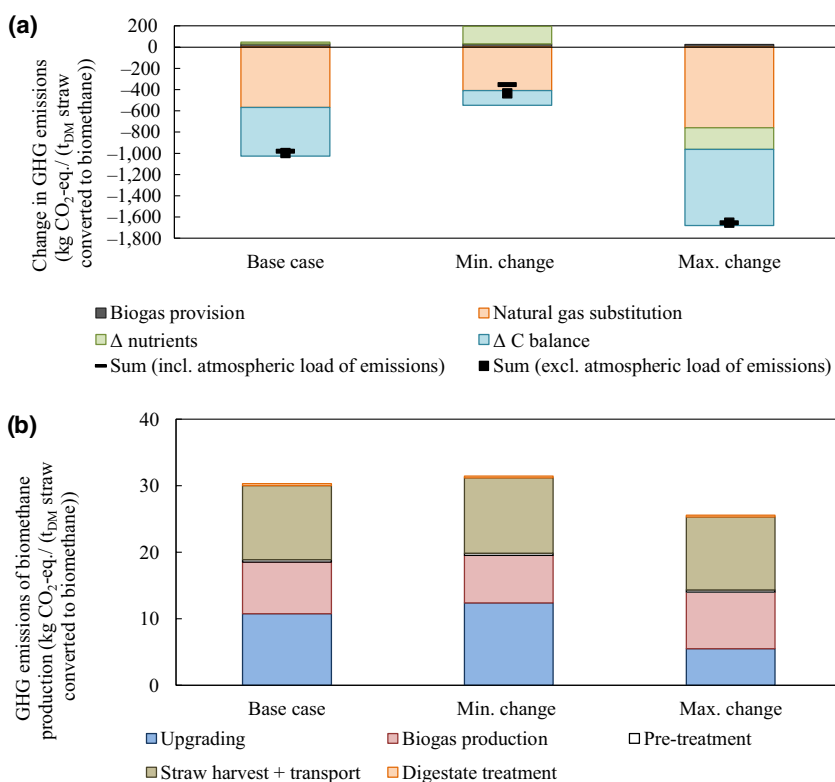
- A part of N in the organic fertilizer derived from digestate is contained in mineral form. The higher availability of mineral N in digestate compared to nutrients bound in wheat straw increases N-losses by shifting from straw incorporation to digestate application. Additionally, digestate exhibits a slower decomposition. Thus, less organically bound nutrients are released over the 100-year period from organic fertilizer than from incorporated straw. The overall required additional fertilizer sum up to 1.3 kg N. The provision of this mineral fertilizer and emissions arising from its application result in GHG emissions of 12 kg CO_2-eq. The lower release of N bound in organic compounds results in lower N-related GHG emissions (most importantly N_2O). This leads to a decrease in emissions of 14 kg CO_2-eq.
- The substitution of natural gas results in a net reduction in GHG emissions of 568 kg CO_2-eq. The combustion of natural gas accounts for 83% of this emission reduction. The remainder stems from the provision of natural gas.
- Optional: If captured CO_2 substitutes CO_2 originating from ammonia production (steam reforming of methane) an additional 225 kg CO_2-eq. can be avoided. The replacement of CO_2 only triggers a reduction in CO_2 capturing. The CO_2 that was formerly captured is still released because it is a waste product of steam reforming.

In the minimum reduction scenarios, the shift from straw incorporation to biomethane production results in a net decrease in GHG emissions of 353 and 436 kg CO_2-eq. with

consideration of the time-integrated atmospheric load of GHG and without consideration of it, respectively. In the maximum reduction scenario, the overall reduction in GHG emissions amounts to 1,654 kg CO_2-eq. A detailed discussion of results of these scenarios is provided in Section S5.1.1.

4.1.2 | Bioethanol production

The shift from straw incorporation to the provision of bioethanol results in net decreases in GHG emissions of 409 and 457 kg CO_2-eq. with consideration of the time-integrated atmospheric load of emissions and without consideration of it, respectively (Figure 6):

- The production of ethanol results in 73 kg CO_2-eq. Of these, 60% originate from the provision of auxiliary materials (ammonia 27%, corn steep liquor 9%, enzymes and diammonium phosphate 12% each). Because the electricity demand of the overall process cannot be fully covered by the energy cogeneration, electricity needs to be drawn from the grid accounting for 25% of the GHG emissions.
- The provision of straw results in GHG emissions of 17 kg CO_2-eq.
- The overall nutrient balance results in a net increase of about 3 kg CO_2-eq. This contains additional fertilizer production (12 kg CO_2-eq.), its application (13 kg CO_2-eq.) and avoided emissions (−22 kg CO_2-eq.). The avoided
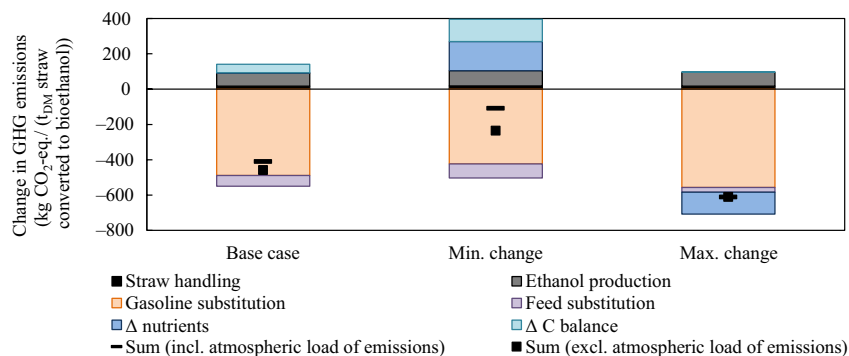
FIGURE 6 Change in greenhouse gas (GHG) emissions as a consequence of the shift from straw incorporation to bioethanol production. Results are presented per t_{DM} straw converted to bioethanol. Bars include the time-integrated atmospheric load of GHG emissions. Only fossil GHG emissions and changes in biogenic GHG emissions arising from the shift are shown. Gasoline substitution comprises the provision and combustion of gasoline

emissions are higher than the emissions from mineral fertilizer due to higher volatilization (NH_3) of nutrients contained in OM (Table S5).

- Ethanol potentially substitutes gasoline. This results in a reduction of 489 kg CO_2-eq.
- Molasses can be used as animal feed. It is assumed that molasses potentially substitutes feed (maize). Thereby, emissions are reduced by 61 kg CO_2-eq.
- Optional: The additional capture of CO_2 and replacement of fossil derived CO_2 reduces emissions by another 133 kg CO_2-eq.

In the minimum reduction scenario, the overall reductions in GHG emissions are 107 and 235 kg CO_2-eq. with consideration of the time-integrated atmospheric load of emissions and without consideration of it, respectively (Section S5.1.2). In the maximum reduction scenario, the reduction in GHG emissions is 610 kg CO_2-eq.

4.2 | Temporal aspect B: Changes in emissions following the shift to bioenergy production

Results presented in this section address the temporal change in GHG emissions per t_{DM} straw converted to biomethane or bioethanol over a 100-year period following the shift from straw incorporation to bioenergy production (Figure 3). The dependency of results on the duration of bioenergy production is evaluated in the sensitivity analysis (Section S5.2).

4.2.1 | Biomethane production

As a consequence of the shift from straw incorporation to biomethane production, the share of straw used for bioenergy production is not incorporated any longer and therefore does not contribute to SOC formation any longer (exemplarily

depicted in Figure 3). In addition, GHG emissions previously arising from straw decomposition are avoided. Due to different decomposition rates and application quantities of straw and organic fertilizer, a certain time passes until a new equilibrium state establishes. The production of biomethane entails carbon payback times ranging 0, 58 and 0 years if only SOC is considered in the base case, minimum and maximum reduction scenario, respectively (Figure 7). Once the payback time is reached, more C is sequestered than before the shift to bioenergy production. The overall carbon payback time (including other changes in emissions than changes in the SOC) ranges from 0 to 49 years: 0 years in the base case and maximum reduction scenario and 49 years in the minimum reduction scenario. The average change in GHG emissions over the 100-year period are −955, −220 and −1,623 kg CO_2-eq. per t_{DM} straw converted to biomethane in the base case, minimum and maximum reduction scenario, respectively. A main factor responsible for the difference in the net GHG reductions is the decomposition of OM. In the minimum reduction scenario, the slow decomposition of wheat straw and the fast decomposition of organic fertilizer (after less than 1 year, more than half of the organic fertilizer is decomposed, whereas it takes 6 years until half of the straw is decomposed) result in a slow increase in SOC through organic fertilizer application and high quantities of straw related C stocks that decompose. Due to these effects, the overall net reduction in GHG emissions depends on the duration of bioenergy production. In the minimum reduction scenario, a duration of less than 50 years could lead to an overall increase in emissions (Figure S5).

4.2.2 | Bioethanol production

In case of the analyzed bioethanol provision concept, only very small quantities of C are returned to the agricultural field (C contained in animal excreta originating from molasses). Thus, there is no significant formation of SOC from production residues. The overall carbon payback times range from 0 to no
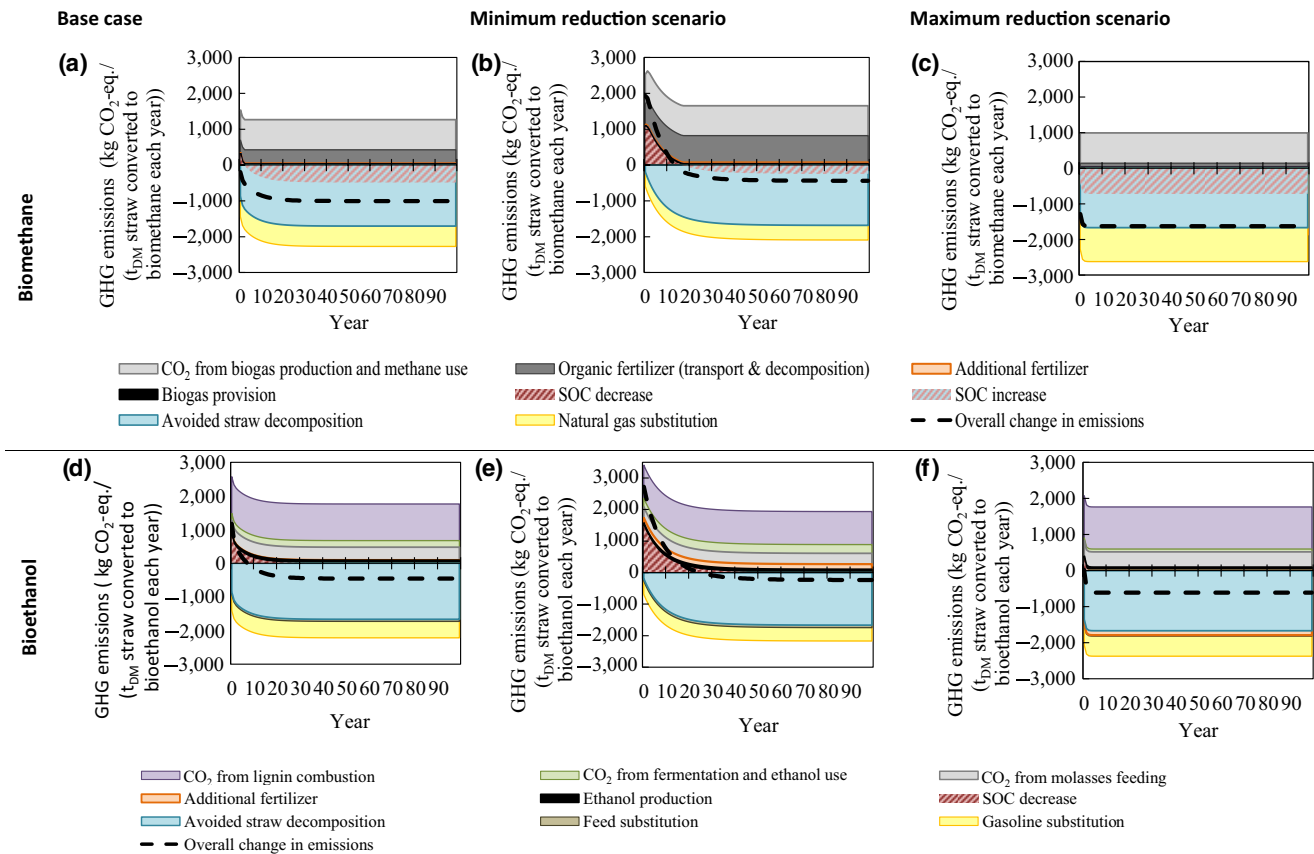
FIGURE 7 Change in greenhouse gas (GHG) emissions per t_{DM} of straw converted to biomethane (a: base case; b: minimum reduction scenario; c: maximum reduction scenario) and bioethanol (d: base case; e: minimum reduction scenario; f: maximum reduction scenario)

compensation of emissions arising from SOC decomposition: less than 1 year in the maximum reduction scenario, 19 years in the base case and no compensation of GHG emissions from SOC changes within the assessed 100-year period in the minimum reduction scenario (Figure 7). In the latter case, a slow decomposition of wheat straw before the shift results in a high accumulation of SOC. The SOC decomposes after the shift to biofuel production as a consequence of the cessation of wheat straw incorporation. The average changes in GHG emissions are −361, 57 and −603 kg CO_2-eq. per t_{DM} straw converted to bioethanol in the base case, the minimum reduction and the maximum reduction scenario, respectively. If bioenergy production is conducted for a shorter time, net emission reductions are lower in the base case and maximum reduction scenario (Figure S5). These results show that the shift to bioenergy production (without returning OM to the field) could potentially displace the storage of high quantities of C in SOC.

4.3 | *GHG savings* according to RED II methodology

The biomethane concept results in slightly higher *GHG savings* than the bioethanol concept (Table 1). In all cases, the

assessed concepts comply with legal minimum *GHG saving* requirements. The overall efficiency of the respective concept only has a minor impact on determined *GHG savings*. This is mainly caused by the change in allocation factor (less efficient concepts yield higher quantities of co-products. The consideration of CO_2, SOC formation and the consideration of the cultivation phase influence results:

- In the base case of the biomethane concept, the SOC credit (Section 2.2.1) amounts to 63.6 g CO_2-eq. per MJ (without allocation), corresponding to 0.28 kg C per kg DM organic fertilizer. The allocated SOC credit is 40.6 g CO_2-eq. per MJ biomethane. In case of ethanol production, there is no credit for SOC formation that can be given due to the use of lignin for energy provision. The granting of credits, as allowed by the RED II, is inconsistent with the rationale to exclude the production phase of agricultural co-products/residues. If no emissions are assigned to the production of straw, credits should not be assigned either.
- The *GHG savings* according to RED II of the minimum reduction scenario result in a higher *GHG saving* if CO_2 is captured than the base case and maximum reduction scenario due to lower conversion efficiency and thus more digestate as well a higher CO_2 concentration in the biogas.

	Biomethane			Bioethanol		
	Base case (%)	Min. change (%)	Max. change (%)	Base case (%)	Min. change (%)	Max. change (%)
RED II methodology						
Standard	93	93	93	89	88	87
Incl. CO$_2$ capture	120	131	111	116	122	131
Incl. SOC credit[a,b]	136	119	142			
Incl. allocated SOC credit[a,c]	107	97	112			
Consequential approach						
Temporal aspect A						
Incl. AL[d], incl. SOC	173	92	220	74	21	104
Excl. AL[d], incl. SOC	176	111	221	83	47	105
Incl. AL[d], excl. SOC	94	60	113			
Excl. AL[d], excl. SOC	93	55	113			
Temporal aspect B						
Excl. AL[d], incl. SOC	168	72	214	74	18	104

Abbreviations: RED II, Renewable Energy Directive II; SOC, soil organic carbon.

[a]The inclusion of the SOC credit is reported separately as SOC credits can only be granted according to RED II if long-term studies confirm an increase in SOC (Section 2.2.1). Results presented in this study rely on incubation experiments.

[b]100% of credit allocated to straw.

[c]Credit allocated to grain and straw (Section S5.3).

[d]AL: Time-integrated atmospheric load of emissions, time horizon 100 years.

- The difference between RED II and results obtained by applying the consequential approach in the minimum and maximum change scenarios stem from the inclusion of the cultivation phase and the nutrient balance. The nutrient balance results in changes in emissions ranging from −202 to 187 and −122 to 163 kg CO$_2$-eq. per t$_{DM}$ straw converted to biomethane and bioethanol, respectively. The RED II approach excludes the cultivation stage.

5 | DISCUSSION

5.1 | General discussion of results

The analysis provides insights into the potential ranges of changes in GHG emissions arising from the shift from straw incorporation to bioenergy production. The results are based on literature data and modeling results reflecting a broad range of environmental conditions and management practices that affect these changes. The results demonstrate that the shift from the presently common use of wheat straw (incorporation into soil) to the production of biomethane and bioethanol results in a net decrease in GHG emissions. The GHG emission reduction of the shift from straw incorporation to biomethane production is about twice the net reduction in

GHG emissions of the shift to bioethanol production. This is mainly due to the effect of SOC increase in the biomethane concept and the loss of C due to lignin combustion in the ethanol concept. The contribution of digestate to SOC formation confirms previous findings (O'Keeffe et al., 2019; Witing et al., 2018). Alternatively, lignin could be applied to soil as well. However, if lignin is used for other purposes than energy generation, the high energy demand of second-generation ethanol production could lead to a significant increase in GHG emissions from the production process if process energy is provided by fossil energy carriers. The low economic competitiveness of second-generation ethanol at present is likely to stimulate the search for alternative, high-value applications of lignin (Cao et al., 2018). In the light of these two aspects, the return of lignin to the agricultural field seems unlikely.

The benefits of both systems arise from the substitution of fossil energy carriers and, in case of biomethane production, the preservation of nutrients and SOC. The substitution of fossil energy carriers serves the political targets to reduce anthropogenic GHG emissions the dependency on (imported) fossil energy carriers. When straw is incorporated, as presently done, the largest part of contained C decomposes within a few years and is thus converted to CO$_2$ (and released to the atmosphere). The production of bioenergy therefore presents a cascade use of C contained in straw. In case of biomethane

production, an additional share of C otherwise mineralized can be returned to the soil in the form of organic fertilizer. This alters the temporal occurrence of GHG emissions (Section 2.1):

- The inclusion of the temporal occurrence of emissions (temporal aspect A) shows that the consideration of this effect changes the resulting net decrease in GHG emissions in the biomethane base case, minimum change and maximum change scenario by 2%, 19% and 0%, respectively. In case of the shift from straw incorporation to bioethanol production, the inclusion of the aspect into the GHG balance yields GHG reductions differing by 11%, 54% and 0% from the net change in GHG emission without consideration of the temporal aspect in the base case, minimum change and maximum change scenario, respectively. The largest contributor to this effect is the decomposition rate of wheat straw. If straw decomposes slowly, emissions are released over a long period of time. In contrast, a considerable share of this C previously emitted through decomposition is immediately released at bioenergy production or shortly thereafter by the use of the respective biofuel. Therefore, the difference in the time-integrated atmospheric load of these emissions is highest in the minimum reduction scenario in which the slowest decomposition of straw is assumed. In case of the shift to biomethane production, this effect is less pronounced due to a share of C contained in organic fertilizer that is subject to decomposition as well.
- The assessment of the change in emissions in the years following the shift to bioenergy production shows that a transition time occurs until a new steady state establishes (temporal aspect B). The transition time is characterized by a decrease in straw-related SOC and SOC formation from digestate-derived organic fertilizer (biomethane concept only). In the biomethane concept, it takes between 3 and 36 years until a new steady state is reached. The introduction of the bioethanol concept might ensue a transition time lasting up to 52 years. However, the overall net change in GHG emissions is negative even before the equilibrium state is reached. Depending on the decomposition of OM, considerable carbon payback times might occur. The average net changes in GHG emissions in the 100 years following the shift to biomethane production are 4%, 50% and 2% lower than those determined at the steady state in the base case and maximum reduction scenario, respectively. This is due to the higher emissions occurring within the first few years after the shift. In case of the shift to bioethanol production, the net decreases in emissions are 21% and 1% lower in the base case and maximum reduction scenario, respectively. In the minimum reduction scenario, the net decrease determined for the steady state turns into a net increase of 57 kg CO_2-eq. per t_{DM} straw converted to

bioethanol. This is due to the high quantity of C formerly stored in SOC that is released following the shift.

These findings show that temporal changes of emissions should also be considered in case of feedstock that is sourced from annual cropping systems. In most cases, the inclusion of temporal effects leads to a lower net GHG emission reduction than determined without the consideration of the temporal pattern of emissions. Nevertheless, the evaluated bioenergy systems lead to substantial net reductions in GHG emissions. The determined carbon payback times are lower than those of most forest biomass (Bentsen, 2017; Buchholz, Hurteau, Gunn, & Saah, 2016). The reported payback time of forest biomass has led to questioning the capability of forest bioenergy to contribute to the needed timely reduction in (anthropogenic) GHG emissions (Norton et al., 2019). The findings presented within this paper reveal that straw-based bioenergy could yield reductions in GHG emissions within acceptable time-scales. Only if very high quantities of C are stored in SOC, long carbon payback times arise. These effects are at present neglected by the RED II methodology because agricultural residues, including straw, are considered to entail no GHG emissions. The comparison presented within this paper therefore shows that the overall choice of handling lignin affects the overall GHG balance of novel fuel concepts and should therefore be considered in biofuel certification. With this regard, the results demonstrate that a return of lignin to agricultural soil can present a way of maintaining SOC levels, while the use of lignin for energetic purposes can result in substantial GHG emissions resulting from the shift from straw incorporation to alternative uses. The use of lignin in other products, for example, products derived from high purity lignin, presents an intermediate storage of lignin eventually resulting in the release of C contained in lignin when products containing lignin are disposed (and potentially incinerated).

Other studies confirm the positive effect of biogas systems and digestate application on SOC formation in general (Breunig, Amirebrahimi, Smith, & Scown, 2019; Franko & Witing, 2020; O'Keeffe et al., 2019; Prays & Franko, 2018; Witing et al., 2018). Studies comparing residue incorporation and biogas digestate application report a higher SOC formation in the latter case (Chen et al., 2012; Nielsen et al., 2020). The experiments conducted by Nielsen et al. (2020) show a slower decomposition of digestate in comparison to straw and determine a comparably high SOC formation potential of solid fraction of digestate. The authors conclude that SOC formation is negatively correlated with lignin content of the material. These results are in line with modeling results and assumptions made in this study. Similar to findings presented in this study, studies evaluating bioenergy concepts using straw as feedstock confirm that concepts returning lignin perform better than those using lignin for energetic purposes (Karlsson et al., 2017).

5.2 | Limitations

The results are connected to certain limitations. Most importantly, the following limitations regarding digestate decomposition and SOC formation apply:

- The decomposition pattern of the organic fertilizer is an important parameter governing the potential changes in GHG emissions. The empirical data of digestate decomposition indicate a stable fraction (Section S4.4.1). This stable fraction leads to higher quantities of C that are sequestered in SOC in comparison with wheat straw. The few available literature on the decomposition behavior and SOC formation potential of digestate report a similar behavior (Nielsen et al., 2020; Pantelopoulos, Magid, & Jensen, 2016; Prays, Dominik, Sänger, & Franko, 2018; Section S6). The sensitivity analysis reveals that if the organic fertilizer decomposed in the same manner as straw, the net reduction in GHG emissions would be lower in case of a shift to biomethane production compared to a shift to bioethanol production (Section S5.2).
- Yet, there are no long-term field studies on the real-world behavior of digestate produced from anaerobic digestion of wheat straw available. Due to the lack of long-term field trials evaluating SOC formation caused by straw-based digestate application, the study was conducted using data from incubation experiments and modeling assumptions. The result indicates a higher C fixation in the biogas concept compared to straw incorporation. Similar results were reported from field trials comparing digestate and wheat straw (Nielsen et al., 2020). SOC formation is a complex process that depends on other factors aside from the composition and quantity of OM applied to soil. For instance, local environmental conditions as well as crop rotation and management practice affect the formation of SOC. To account for a range of potential management practices and environmental factors, the minimum and maximum reduction scenarios were defined (presenting boundary conditions of parameters and decay functions found in literature reflecting a wide range of environmental conditions and management practices). Therefore, the results seek to provide ranges of potential changes in GHG emissions that could arise under various boundary conditions found in reality.
- It is assumed that nutrient release follows the same pattern as the decomposition of OM. This is a simplification that does not account for complex mechanisms and biological processes involved. If these nutrients are released within days, weeks or months as estimated by the decay curve, this assumption can be considered acceptable.

These limitations are inherently caused by the lack of knowledge regarding the SOC formation potential of digestate made from wheat straw. Further research and re-evaluation of results presented in this study are recommended once more data from long-term field studies are available. Aside from SOC formation, the following aspects should be considered:

- The change in the temporal patterns of C release as a consequence of corn substitution is not considered. With regard to the low quantities of corn substituted, the effect is considered of minor importance.
- Biogenic CO_2 emissions from anaerobic digestion and fermentation processes may relatively easily be captured, and possibly utilized (e.g., upgraded with H_2 to solar fuels) or stored underground. Both concepts could yield additional reductions in GHG emission.
- The promotion of advanced biofuels provides additional revenue to farmers. This might create incentives to change the varieties of wheat grown (e.g., varieties with a lower grain-to-crop ratio). Likewise, the promotion of bioenergy from non-food crops might motive some farmers to grow other non-food lignocellulosic feedstock for bioenergy instead of food crops. Such changes might trigger additional effects (e.g., increasing demand for land) that are not considered in this study.

5.3 | Recommendations for future research

Only few studies report experimental data on digestate decomposition. Studies show that the composition of digestate as well as environmental factors and management practices ultimately affects its decomposition behavior and its nutrient release (cf. Möller, 2015; Nielsen et al., 2020). Therefore, the need of further studies addressing these aspects is strongly emphasized with regard to the promotion of advanced biofuels by the RED II and the advent of commercial concepts using agricultural residues. Once data obtained from long-term field trials is available, modeling approaches should be applied that compare modeled crop cycles with results from long-term field trials. Furthermore, it should be noted that this study solely focuses on GHG emissions. The change in agricultural practice, the application of different types of organic fertilizers and the use of straw that was previously incorporated into soil as source for bioenergy results in various effects on soil and soil quality. The incorporation of straw contributes to SOC formation, prevents soil erosion, increases soil porosity, influences nutrient mobilization and immobilization and positively affects soil biodiversity. These effects need to be included to evaluate the overall sustainable potential of straw-based bioenergy.

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CONFLICT OF INTEREST
The authors declare that there is no conflict of interest.

DATA AVAILABILITY STATEMENT
The data that support the findings of this study are available
in the Supporting Information of this article.

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SUPPORTING INFORMATION

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